

# ***Planning for environmental restoration of radioactively contaminated sites in central and eastern Europe***

## ***Volume 1: Identification and characterization of contaminated sites***

*Proceedings of a workshop held  
within the Technical Co-operation Project on  
Environmental Restoration in Central and Eastern Europe  
in Budapest, Hungary, 4–8 October 1993*

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**PLANNING FOR ENVIRONMENTAL RESTORATION OF RADIOACTIVELY  
CONTAMINATED SITES IN CENTRAL AND EASTERN EUROPE: VOLUME 1  
IDENTIFICATION AND CHARACTERIZATION OF CONTAMINATED SITES**

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## FOREWORD

The radioactive contaminant materials resulting from diverse activities in relation to the nuclear fuel cycle, defence related operations, and various industries in addition to medical and research facilities represent perhaps the most severe and immense pollution left from a past era. The political changes in central and eastern Europe (CEE) not only brought some disclosure of the radioactively contaminated sites, but also resulted in a political condition in which this region became receptive to co-operation from a range of outside countries.

It is under these circumstances that the IAEA decided to launch a Technical Co-operation (TC) Project on Environmental Restoration in Central and Eastern Europe. The project was initiated in the latter part of 1992 and ended in 1994. The countries that were involved and represented in this forum are: Belarus, Bulgaria, Croatia, Czech Republic, Estonia, Hungary, Kazakhstan, Poland, Romania, Russian Federation, Slovakia, Slovenia and the Ukraine. Several experts from countries outside the region participated and offered their co-operation throughout the project.

The TC regional project consisted of three workshops that addressed different, but sequential, themes. The basic criterion consisted in matching the structure of the IAEA project with a real-scale environmental restoration project. The main focus was to identify radiological conditions in the region and remediation plans, if any.

The subject of the first workshop held in Budapest, 4-8 October 1993, was the identification and characterization of radioactively contaminated sites in the region. The second part of the project and the second workshop (Piestany, Slovak Republic, 12-16 April 1994 ) involved planning and preparing the identified sites for restoration. This included items such as the restoration objectives, dose and environmental assessment, cost analysis, strategy and prioritization. Eventually, the third part of the project covered technologies for, and the implementation of, environmental restoration. The third and final workshop was held in Rež, Czech Republic, 12-16 December 1994.

A great deal of technical and scientific information which was formerly classified or only available confidentially was disclosed under the auspices of the project. Information available only in national languages (mainly Russian) was made available in English. The three volumes of this TECDOC incorporate reports submitted by national experts and invited speakers at or following the three workshops. Volume 1 includes papers describing the identification and characterization of contaminated sites in the region. It also presents the objectives of the project, illustrates past and current IAEA activities on environmental restoration, provides a scientific framework for the project and the individual workshops and summarizes the results achieved. Volume 2 includes the papers that involve planning and preparing the sites for restoration. Volume 3 presents technologies for, and the implementation of, environmental restoration.

It should be noted that papers submitted by national experts are variable in length and content, as this reflects national conditions and approaches. Countries having one or two contaminated sites concentrate on technical details, countries with dozens of sites offer a general overview. Problems associated with contamination from the uranium mining and milling industry are intrinsically different from those related to accident generated contamination. By means of the papers contained in this TECDOC, the reader may get a general impression of the vastness of the problems in central and eastern Europe. The IAEA officer responsible for the workshops was M. Laraia, of the Division of Nuclear Fuel Cycle and Waste Management. Papers were compiled and edited by J. Wiley, of the same Division.

The IAEA wishes to express its thanks to all participants in the programme and would like to take this opportunity to acknowledge the excellent co-operation and hospitality of the institutions that hosted the project workshops.



## *EDITORIAL NOTE*

*In preparing this publication for press, staff of the IAEA have made up the pages from the original manuscripts as submitted by the authors. The views expressed do not necessarily reflect those of the governments of the nominating Member States or of the nominating organizations.*

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## **INTRODUCTION**

### **INTERNATIONAL CO-OPERATION ON ENVIRONMENTAL RESTORATION: THE ROLE OF THE IAEA**

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#### **1. THE ROLE OF THE IAEA**

Article II of the IAEA Statute states that the "IAEA shall seek to accelerate and enlarge the contribution of atomic energy to peace, health and prosperity throughout the world...." In fulfilling these objectives the IAEA encourages, sponsors and coordinates research work and the development of data and technology in promising areas. Technical assistance is provided to developing Member States in form of expert missions, design assistance and various technical co-operation programmes.

Radioactive waste management has a prominent status as an Agency activity and has to be dynamic in nature to keep pace with the changing needs of Member States. Meeting these needs is a challenge in itself considering the diverse nature of waste management activities that are planned or underway in our Member States. The main objective of the waste management programme is to ensure the safe management and disposal of radioactive waste in accordance with the IAEA's mandate to promote the safe and peaceful use of atomic energy.

#### **2. RADIOACTIVE CONTAMINATION--A WORLD WIDE PROBLEM**

Although this project is named the Technical Co-operation Regional Project on Environmental Restoration in Central and Eastern Europe (CEE), the need for environmental restoration is not confined to this region alone. There are sites all over the world that are in an unacceptable radiological state due to bad practices in the handling of radioactive waste, nuclear accidents or weapons testing activities. The wastes stem from a wide range of activities and the nature and character of the radioactive materials may differ from country to country.

Uranium mining and milling facilities may be a source of radioactive contamination and it is of interest to know to what extent there are radiological problems related to this industry. If properly managed, through adhering to national and international standards and guides, there are small chances that these facilities pose a threat to the surrounding environment. The operators of these centres have over the years realized that the protection of the environment in relation to uranium production is a part of the procedures. Safety precautions are normally taken and tailings are covered and prevented from spreading contamination.

The need for environmental restoration, especially in relation to uranium production facilities, is world wide. Each year approximately 55,000 tons of uranium are produced. Given the fact that uranium ores usually contain about 0.15% of uranium, roughly 26 million tons of mill tailings accumulate each year.<sup>1</sup> The residuals in most cases are piled up and stored near the production facilities potentially exposing the environment to radiation. This radiation is relatively low and short term exposure is generally not harmful. However, the long term effects of radon radiation have, through several studies, shown to have adverse effects. There are no exact data estimating how much uranium has been produced over the years. Given the fact that large scale

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<sup>1</sup>Since the share of in-situ leaching is estimated to be 30%, only 70% of the produced uranium produces waste in the form of tailings.

production, through conventional means, has gone on for at least 30 years, one can try to get a perspective of the amount of radioactive mill residues throughout the world.

Satisfactorily securing and containing of the uranium mill residues do not necessarily require a lot of resources. A recent IAEA Technical Co-operation mission to the Rössing Uranium Mine in Namibia disclosed that limited resources and management dedication has yielded very acceptable results regarding contamination and spreading of radioactive substances. IAEA sponsored assessments of the radiological conditions at mines and mills in several other countries have also shown that uranium production can be carried out in a safe and environmentally sound mode.

Due to a decline in demand, falling prices, exhaustion of deposits and changes in export agreements of uranium over the past few years, several uranium mines and mills have been closed. Thus, several of these facilities have been decommissioned. Closeout projects of uranium production facilities--in order to protect the environment from previous activities--have successfully been carried out in countries such as France, Sweden and Spain. Other countries, like Australia, China, Germany and the United States are also heavily involved with such undertakings.

These examples have shown that uranium production does not necessarily have to harm the environment in which the activities take place. Nevertheless, according to IAEA data, only seventeen of the world's uranium processing centres are defined as decommissioned. 119 centers are defined in the categories "In Operation, Stand By and Closed," meaning that **extensive** environmental restoration efforts have not yet taken place (**Table 1** ). Thus, several sites on all continents potentially are, or soon will be, in need for one or another form of remediation.

**Table 1**

**STATUS OF THE URANIUM MINING AND ORE  
PROCESSING CENTRES OF THE WORLD**

57	IN OPERATION
12	UNDER STUDY
5	UNDER CONSTRUCTION
1	AWAITING LICENSE
7	PLANNED
21	STAND BY
41	CLOSED
17	DECOMMISSIONED
1	CANCELED PROJECT
5	DEFERRED PROJECT
1	UNKNOWN

There are several factors that contribute to an increased risk of radioactive contamination and spreading.

- The longer the production has been carried out, the greater the risk for contamination
- Higher ore grade increases radiation dose rates from the residues
- Physical conditions like precipitation and heavy winds contribute to the spreading of the contaminant materials
- The less wealthy states tend to give environmental restoration marginal attention

Another form of radioactive contamination in relation to mill residues is the use of radioactive sand and gravel as building materials for civil engineering constructions. The mill tailings are specifically attractive for construction use because of the fine grain composition and the low cost of acquiring them. Over the years, several countries have investigated the use of mill residues and found it to be wide-spread. The IAEA therefore has reason to believe that areas close to uranium production facilities in several countries have one or another form of such contamination as well.

Poor radioactive waste management practices, mostly of low level waste, originating from factories, hospitals, research facilities, etc. have contaminated the environment in many countries throughout the world. The relatively high volume of such wastes has often made it inconvenient to store it in safe, designated and controlled facilities due to the high cost of construction of such repositories. The waste has in many cases been disposed of as "conventional" waste or at sites where the population can be exposed to it. From such locations radioactive contaminants can be spread through air or ground water. Dumped radwaste like this is often not registered and it is hard to find the sites at which it has been disposed of. Although the waste in many cases does not pose an immediate threat to the surrounding environment, the sites cannot be released in their present condition and should therefore be subject to one or another form of remedial action. If nothing else, the waste should be registered and secured. The spreading of contamination also should be stopped along with the making of plans for the lasting confinement or removal of the radiation sources.

Radioactive fallout or dispersion of radionuclides as a result of a nuclear accident or atomic weapon tests is a third source of radioactive contamination of the IAEA's concern. Radioactive fallout contamination is generally not intense but spread over a vast area. This makes the problem truly international because many countries can be affected by the same accident or event.

### 3. INTERNATIONAL ORGANIZATIONS AND ENVIRONMENTAL RESTORATION

In the new and somewhat transformed world, international co-operation seems to be the rule rather than the exception. New alliances are made in technological as well as in economical rounds. Countries with former political differences now work together to reach mutual goals. These changes and trends of collaboration should also be taken advantage of by parties working in the field of environmental restoration in order to draw from the benefits from such work.

Given the fact that the need for environmental restoration exists on all continents the clean up and restoration operations should have an international nature. There are three main modes of international co-operation that can be utilized in environmental restoration. The first is through bilateral arrangements between countries and/or organizations. The second is co-operation on a regional level and the third is through international organizations. These forms of co-operation, with emphasis on information and technology exchange, including joint research and development and demonstration projects, have been very successful. Co-operation of this nature has many benefits and is extremely practical for several reasons. First, it makes good economic sense to share and learn from each other's experiences and compare future strategies. The resulting benefit is that it prevents some duplication of efforts. International organizations like the Commission of the European Communities (CEC), the Organization for Economic Co-operation and Development/Nuclear Energy Agency (OECD/NEA) and the International Atomic Energy Agency (IAEA) play a major role in this area by facilitating the information and technology exchange and transfer. A second point worth mentioning is that projects initiated by any or all of these international organizations tend to be considered more credible and therefore generate more financial support. Third, joint projects create a support network and a system of formal and informal peer reviews. This external review process enhances and adds technical credibility and validity to national approaches and methodologies. And, finally, co-operation and exchange are required and used by countries as a means of checking their own progress--a sort of calibration.

International organizations can be useful in other areas than information and technology exchange as well. An international organization can also, for instance, set up standards and guides of how to perform environmental restoration projects in the most effective and safest way. There should be standardized procedures of how to dispose of the radioactive waste that an environmental restoration project may yield. Guidelines regarding how to secure large amounts of low level wastes in the best possible way could also be set up by an international organization based on the knowledge of the Member States.

#### 4. THE IAEA AND ENVIRONMENTAL RESTORATION

The IAEA has a central position in work relating to environmental restoration. The IAEA has recognized its responsibilities and has throughout the years kept its Member States as up to date as possible regarding the technical aspects of decontamination and decommissioning (D/D) of nuclear facilities and related activities. D/D activities were introduced in the IAEA's programme in 1973. At present, all D/D related activities of the IAEA are consolidated within the Waste Management programme. The IAEA's role in D/D is to assist its Member States in decommissioning their nuclear facilities, including uranium mining and milling, in a safe, timely and cost effective manner. In this role, the IAEA reviews current scientific, technological and regulatory information and disseminates them as internationally acceptable good practices, guidelines and standards.

In respect to environmental restoration the IAEA has focused on two different areas, namely 1) accident response (including cleanup of large areas as a result of a nuclear accident) and 2) cleanup in relation to uranium mining and milling. The involvement and encounter with the Chernobyl accident in 1986 is an example of response to a nuclear accident. The IAEA has gained and spread experience in the area of closeout of uranium production centers through many projects, e.g. two technical co-operation projects on environmental restoration of uranium mines and mills that recently took place in Argentina and Portugal.

Over the years several documents in the IAEA Technical Report and Safety Series have been written, published and revised on subjects relating to environmental restoration (Table 2). These contain, at any time, the most advanced knowledge and experience in the field of the presented subject.

The IAEA's efforts in publishing these documents have in general satisfied the current needs of the Member States; most states now have the technical know how required to perform cleanup projects. Therefore, as Member States become more involved with the actual performance of environmental restoration the IAEA's emphasis has been shifted to another area of interest: safety requirements and guidelines.

#### 5. THE RADWASS PROGRAMME

The IAEA has recognized that there is a growing need for regulations, standards and guides in the area of the management of radioactive wastes. In response to a request from Member States the IAEA in 1991 established the Radioactive Waste Safety Standards (RADWASS) programme to develop a special series of safety documents specifically directed at radioactive waste management. Although several publications in the IAEA Safety Series have been issued, covering most areas of importance, the documents were prepared during the phase when philosophies and technologies were still emerging and, as a result, some are considered as being interim documents. The RADWASS Programme is being developed to (1) document the existence of international consensus in the realm of radioactive waste management, (2) facilitate and create a mechanism to establish consensus where it does not exist, and (3) provide Member States with a comprehensive series of documents to complete national standards and criteria. The



Table 2

**LIST OF SELECTED IAEA TECHNICAL REPORT AND SAFETY SERIES IN  
THE AREA OF ENVIRONMENTAL RESTORATION**

**URANIUM MINING AND MILLING**

**Radiation Monitoring in the Mining and Milling of Radioactive Ores, Safety Series No. 95 of 1989.**

Over the past decade there have been significant developments in the techniques and methods of monitoring radon and radon daughters, thoron and thoron daughters, and radioactive dusts encountered in the mining and milling of radioactive ores. The development of new techniques and methods of measurement has received increased attention owing to the fact that the efficient control of radon and its daughters in the mining atmosphere, particularly in underground mines, is a difficult task. Epidemiological studies have clearly demonstrated that the incidence of lung cancers among underground uranium miners, and also among underground non-uranium miners who were exposed to radon and radon daughters in their occupations, is higher than that among the general public. This publication is a revision of IAEA Safety Series No. 43, originally issued in 1976. The revised version has been limited in coverage to radiation monitoring and medical surveillance.

**Safe Management of Wastes from the Mining and Milling of Uranium and Thorium Ores, Safety Series No. 85 of 1987.**

This publication consists of two parts: a Code of Practice and a Guide to the Code. The Code sets forth the requirements for the safe and responsible handling of the wastes, while the Guide gives assistance in the use of the Code together with some discussion of the technology and concepts included.

**The Application of the Principles for Limiting Releases of Radioactive Effluents in the Case of the Mining and Milling of Radioactive Ores, Safety Series No. 90 of 1989**

This publication provides general guidance on the application of Safety Series No. 77 (1986), Principles for Limiting Releases of Radioactive Effluents into the Environment, to the setting of limits for the release of radioactive substances during the normal operation of the mining and milling of radioactive ores, as well as general guidance on assessing the resulting individual and collective doses.

**The Environmental Behavior of Radium, Technical Report Series No. 310 of 1990**

This report provides an up-to-date review of the environmental properties of radium and deals with the sources, properties, environmental behaviour and the methods for the analysis, controls and assessment of radium-226.

**Measurement and Calculation of Radon Releases from Uranium Mill Tailings, Technical Report Series No. 333 of 1992**

The purpose of this report is to present a review of all the factors involved in estimating the release of radon from uranium tailings and the methods and techniques for measuring such releases. The report should be useful for those involved in the design, operation, decommissioning and regulation of uranium tailings impoundments, especially in developing countries or countries with small uranium mining programmes. In addition, it will be useful to those concerned with the measurement and calculation of radon releases from other sources.

## **Table 2 (cont.)**

### **Current Practices for the Management and Confinement of Uranium Mill Tailings,**

*Technical Reports Series No. 335 of 1992*

The report discusses the current practices in the design, siting, construction and closeout of impoundment facilities for uranium mill tailings. The objective is to present an integrated overview of the technological, safety and radiation protection aspects of these topics in order to ensure that the potential radiological and non-radiological risks associated with the management of uranium mill tailings are minimized now and in the future. The report should be useful to those involved in the design, operation, decommissioning and regulation of uranium tailings impoundment facilities, especially in developing countries or countries with small programmes.

### **Decommissioning of Facilities for Mining and Milling of Radioactive Ores and Closeout of Residues, Technical Report to be published in 1993**

The purpose of this report is to provide information to Member States to assist in planning and implementing the decommissioning or close-out of uranium mine/mill facilities, mines, tailings impoundments, mine debris piles and unprocessed ore stockpiles. The report presents an overview of the factors involved in planning and implementing the decommissioning/close-out of uranium mine/mill facilities. The information applies to situations when mines, mills, tailings piles, mining debris piles and leach residues already exist as either still operational, mothballed or abandoned projects, as well as to future mining and milling projects. The report also identifies major factors that need to be considered in the decommissioning/close-out activities.

## **ACCIDENT RESPONSE**

### **Cleanup of Large Areas Contaminated as a Result of a Nuclear Accident,**

*Technical Report Series No. 300 of 1989*

Experience at Chernobyl showed that the main long term radiological consequence to the population will probably be external exposure from radioactive fallout deposited on the ground. The present text provides an overview of the methodology and technology available for cleaning up large areas and gives preliminary guidance on the planning, implementation and management of such cleanups.

### **Planning for Cleanup of Large Areas Contaminated as a Result of a Nuclear Accident,**

*Technical Report Series No. 327 of 1991*

The present report is the second of three IAEA publications dealing with the cleanup of large areas contaminated as a result of a nuclear accident. It is mainly a planning and management document, outlining the broad strategic and tactical approach to cleanup, the management structure and other key requirements. The report also shows how the various subplans interface and interact to ensure that the cleanup can be performed safely, efficiently and as quickly as possible under adverse conditions.

### **Disposal of Material from the Cleanup of Large Areas Contaminated as a Result of a Nuclear Accident, Technical Report Series No. 330 of 1992**

The problem of ground contamination in the case of a severe nuclear accident is one of special concern. This report gives guidance on planning and management of safe transportation and disposal of large volumes of contaminated materials, with the objective of minimizing the consequences of such an accident.

### **Derived Intervention Levels for Application in Controlling Radiation Doses to the Public in the Event of a Nuclear Accident or Radiological Emergency: Principles, Procedures and Data,**

*Safety Series No. 81 of 1986*

This publication provides practical support to the guidance contained in IAEA Safety Series No. 72, Principles for Establishing Intervention Levels for the Protection of the Public in the Event of a Nuclear Accident or Radiological Emergency. In the event of a nuclear accident or radiological emergency there is a need to determine the levels of projected dose at which it may be necessary to

### **Table 2 (cont.)**

introduce relevant protective measures. However, more practical quantities are needed for the comparison of measurement results made in environmental materials and in foodstuffs. These derived intervention levels (DILs) need to be determined for the radionuclides of potential radiological importance. This report gives guidance in elaborating the principles, procedures and methodologies relevant to the evaluation of DILs.

#### **Principles and Techniques for Post-Accident Assessment and Recovery in a Contaminated Environment of a Nuclear facility, Safety Series No. 97 of 1989**

This safety guide presents: (a) information and practical guidance relevant to assessing the off-site consequences during the late phase of a serious accident in a nuclear facility; (b) guidance on recovery operations off the site and associated decision making process; and (c) proposals for consideration by national authorities regarding the organizational structure for the conduct of recovery operations.

#### **Response to a Radioactive Materials Release Having a Transboundary Impact,**

*Safety Series No. 94 of 1989*

This guide addresses the special problems in States affected by transboundary release of radioactive material from a site outside their national boundaries. No new or radically different principles are thought to be called for to deal with this type of event. However, the Chernobyl experience of many countries, even those with excellent emergency plans for their own or for nearby nuclear facilities, suggests that some additional guidance on emergency planning is necessary beyond the well documented, nuclear facility centre approach.

#### **Recovery Operations in the Event of a Nuclear Accident or Radiological Emergency,**

*Proceeding Series.*

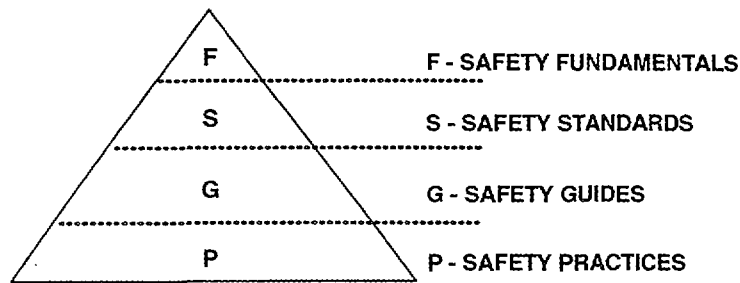
Proceedings of a symposium, Vienna, 6-10 November 1989. The purpose was to provide a forum for review of actual experience gained and lessons learned from recovery techniques and operations in response to serious accidents at nuclear facilities and accidents associated with radioactive materials, and also to consider the development of emergency planning and preparedness resources. A special feature of the symposium programme was a full session on an accident involving a chemical explosion in a high level fuel tank at a plutonium extraction plant in the Southern Urals in the USSR in 1957.

#### **Techniques and Decision Making in the Assessment of off-Site Consequences of an Accident in a Nuclear Facility, Safety Series No. 86 of 1987**

This guide is intended to complement the IAEA's existing technical documentation on emergency planning and preparedness by providing information and practical guidance related to the assessment of off-site consequences of an accident in a nuclear or radioactive materials installation and to the decision making process in implementing protective measures.

implementation of the RADWASS programme will impose a more logical structure on safety publications in the waste management area and improve the visibility and status of these safety related publications.

The RADWASS programme has been organized within the framework of a hierarchical structure of four levels of safety documents. The top level publication will be a Safety Fundamental document which will provide the basic safety objectives and fundamental principles or requirements that must be incorporated in a national waste management programme. The lower levels include Safety Standards, Safety Guides and Safety Practices documents (**Figure 1**).



***Waste Management Subject Areas***

1. Planning for a National Radioactive Waste Management System
2. Pre-Disposal Management
3. Near-Surface Disposal
4. Deep Geologic Disposal
5. Management of Radioactive Waste from Mining and Milling Ore
6. Decommissioning

**Figure 1**

**THE STRUCTURE OF THE RADIOACTIVE WASTE  
SAFETY STANDARDS (RADWASS) PUBLICATIONS**

Among the six standards in the RADWASS programme, two apply directly to environmental restoration. Standard 5 covers the management of wastes from mining and milling of ores containing uranium and thorium. This Standard will address the requirements for: the minimization; collection; treatment and discharge/disposition of the solid, liquid and airborne wastes from the development and operation of mines handling radioactive ores (including in-situ leaching) and the operation of the mills and the closeout of the mining and milling facilities. Emphasis will be given to siting, design, operational control, stabilization, environmental restoration and long time surveillance of mill tailings impoundments and waste rock piles (including controlling their use as construction material). The Standard will also address the mining and milling of other ores producing wastes containing significant radiological amounts of uranium, thorium and their decay products.

Standard 6 in the RADWASS Programme covers the complete decommissioning of facilities that handle radioactive material as well as environmental restoration. Also emphasized will be the need to design facilities for ease of decontamination and decommissioning. The Standard will cover decommissioning of all facilities except for those related to mining and milling. **Table 3** gives an overview of the proposed plan of RADWASS publications in the two standards relating to environmental restoration.

**6. THE TECHNICAL CO-OPERATION REGIONAL PROJECT ON ENVIRONMENTAL RESTORATION IN CENTRAL AND EASTERN EUROPE**

One result of the political changes in Central and Eastern Europe (CEE) was the revealing of these countries' environmental state. An extensive industrial build up and exhaustion of the area's natural resources were two of the means used to accomplish the quota based productivity goals. In many areas, the preservation and protection of the environment were often neglected.

Table 3

**PLAN OF RADWASS PUBLICATION IN THE  
AREA OF ENVIRONMENTAL RESTORATION**

<p><i>Uranium and Thorium Mining and Milling</i></p> <p><b><u>SAFETY STANDARD</u></b></p> <p><b>111-S-5.</b> Management of waste from mining and milling of U/Th ores</p> <p><b><u>SAFETY GUIDES</u></b></p> <p><b>111-G-5.1</b> Siting, design, construction and operation of facilities for the management of waste from mining and milling of U/Th ores</p> <p><b>111-G-5.2</b> Decommissioning of surface facilities and closeout of mines, waste rock and mill tailings from mining and milling of U/Th ores</p> <p><b>111-G-5.3</b> Safety assessment for the management of waste from mining and milling of U/Th ores</p>	<p><i>Decommissioning and Environmental Restoration</i></p> <p><b><u>SAFETY STANDARD</u></b></p> <p><b>111-S-6</b> Decommissioning of nuclear facilities (and environmental restoration)</p> <p><b><u>SAFETY GUIDES</u></b></p> <p><b>111-G-6.1</b> Recommendations for decommissioning of nuclear power and large research reactors</p> <p><b>111-G-6.2</b> Recommendations for decommissioning of medical, industrial and small research facilities</p> <p><b>111-G-6.3</b> Recommendations for the decommissioning of nuclear fuel cycle facilities</p> <p><b>111-G-6.4</b> Safety assessment for the decommissioning of nuclear facilities</p> <p><b>111-G-6.5</b> Environmental restoration of previously used or accidentally contaminated areas</p> <p><b>111-G-6.6</b> Recommended cleanup levels for contaminated land areas</p>
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**Table 3 (cont.)**

<p><i>Uranium and Thorium Mining and Milling</i></p> <p><b><u>SAFETY PRACTICES</u></b></p> <p><b>111-P-5.1</b> Procedures for closeout of mines, waste rock and mill tailings</p> <p><b>111-P-5.2</b> Operational and post operational monitoring, surveillance and maintenance of facilities for the management of waste from mining and milling of U/Th</p>	<p><i>Decommissioning and Environmental Restoration</i></p> <p><b><u>SAFETY PRACTICES</u></b></p> <p><b>111-P-6.1</b> Techniques to achieve and maintain safe storage of nuclear facilities</p> <p><b>111-P-6.2</b> Procedures and techniques for the decommissioning of nuclear facilities</p> <p><b>111-P-6.3</b> Methods for deriving cleanup levels for contaminated land areas</p> <p><b>111-P-6.4</b> Monitoring for compliance with cleanup levels</p>
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The radioactive contaminant materials resulting from diverse activities in relation to the nuclear fuel cycle, defense-related operations, various industries in addition to medical and research facilities are perhaps the most severe and immense pollution substances left from the cold war era. The waste resulting from some fifty years of activities has piled up--often at unregistered sites--leaving the public in the potential danger of long term radiation exposure.

The political changes not only brought forward a fragmentary disclosure of the nuclear contaminated sites, but also resulted in a political condition in which these countries became receptive to co-operation from a range of countries the region previously had been isolated from.

It is with these circumstances of political change that the IAEA decided to launch this Technical Co-operation Project on Environmental Restoration in Central and Eastern Europe. The project was initiated in the latter part of 1992 and is scheduled to end by 1994. The countries that are involved and represented in this forum here today are: Belarus; Bulgaria; Croatia; the Czech Republic; Estonia; Hungary; Poland; Romania; the Russian Federation; the Slovak Republic; Slovenia and Ukraine.

Several countries have over the years gained valuable experience in environmental restoration and the fields relating to such work. These countries have performed several successful restoration projects and most CEE countries can gain valuable data and information from them. As a result of this, papers from Canada, France, Germany, Italy, Spain, Sweden, the United Kingdom and the United States are included to share their experience with environmental restoration.

This report on the Technical Co-operation Regional Project on Environmental Restoration in Central and Eastern Europe is planned to be presented in the form of three volume that each will address different, but sequential, themes.

The subject of this first report will be the identification and characterization of radioactively contaminated sites in the region of CEE. Before any action in regards to environmental restoration can be taken, the involved countries and the IAEA need to get an overview of the environmental state in each of the countries. The Agency has therefore requested selected experts from the targeted Member States to present a thorough report identifying contaminated sites in their home countries. The experts have, as thoroughly as possible, surveyed and categorized each site according to location, volume and radioactive concentration. Also included is data regarding the sites' radioactive concentrations, source(s) of contamination, description of radiological hazard (e.g. proximity to populous areas), potential spreading of contamination, etc. Techniques and methods related to radiometric surveys, laboratory analyses, statistical evaluation etc, are an important part of this workshop as well. The works also, where possible, have identified which organization is responsible for the monitoring and cleanup of each of the contaminated sites. This information is desirable for one specific reason; environmental restoration work in other countries has shown that without any responsible party, there is little chance that any cleanup will be performed.

The IAEA's **primary** objective has not been to locate areas on the sites of nuclear facilities like nuclear power plants and research localities. Those are mostly known and, normally, subject to regulatory control. Rather, the IAEA aims at identifying and characterizing sites related to uranium mining and milling activities, in particular, disused mines, abandoned mill tailings and sites potentially needing environmental restoration now or in the future. Some of the mills in the CEE region are among the largest production centers in the world, many of which are located in rather populous areas. It has therefore been desirable for the IAEA to get assessments on whether these sites pose a threat to the surrounding locations.

If the uranium production facilities are found to be in a state that requires some form of remedial action, the size and location of the CEE production centres cause potential complications in the restoration work. For instance, in the United States most mines are smaller and located in less densely populated areas than are most CEE mines. The U.S, in the mountain states of Colorado, Utah and Wyoming, for instance, also enjoy the fact that there are large deserted areas to which the radioactive residues can be transported and disposed of in a safe manner. For two obvious reasons this cannot be done in the uranium industry areas of CEE. First, the volume of the accumulated radioactive waste material are far too high to be removed within the sphere of reasonable costs. Secondly, there are no safer and alternative sites to where the tailings can be removed.

Another factor that distinguishes the CEE uranium production centres from those in the US is that little or no money is made available to perform the proper decommissioning of the CEE production facilities. Some of the centres were closed down because of financial difficulties and no or little resources were (or are) put into securing the contaminated areas. These uranium production centres will therefore be given special attention from the IAEA in the environmental restoration program.

Sites contaminated as a result of nuclear accidents, defense-related activities, past waste dumping practices and deserted factories having used radioactive substances (e.g. radium sources) are also part of the IAEA project. However, sites in this second category are often not recorded or monitored. Several of these are therefore unknowingly exposing the environment and population to radioactivity. Radwaste, unlike many of the "conventional wastes," cannot be seen, as smog for instance, nor can the waste be smelled. The IAEA would therefore like scientists to perform radiological measurements of most areas believed to be contaminated in order to get a complete picture of the sources of contamination.

After the information obtained as a result of this first workshop is compiled, organized and evaluated, the second part of the TC project is due to commence. This part involves preparing the identified sites for restoration. This includes restoration objectives, dose and environmental

assessment, cost analysis and a plan prioritizing each registered site. The objective for the remediation of each of the sites will also asserted. Following remediation activities, there are three different levels at which a site can be classified:

1.       **Prohibited to human access**
2.       **Allowed for restricted release**
3.       **Allowed for unrestricted release**

In deciding what form of remediation should take place a cost-benefit or multi-attribute analysis will be carried out. The analysis will indicate whether it is most beneficial to secure the site rather than cleaning it up for release. If secured, the waste should be safely contained in order to prevent spreading of nuclear contamination. Although a total cleanup is in principle preferred, this may cost more than society benefits from the cost of consuming resources in the remediation. Previous experiences have shown that if a contaminated site lies in a remote area, the resources spent on this site will, in most cases, provide less benefit than resources spent on a site in close proximity to a populous area. However, the more contaminated an area is, independent of the risk of human exposure, the greater the chances are that action to clean up the site will have to be taken. Regardless of this, it is up to each of the participating countries to determine how many resources to put into the restoration and to what extent each individual site will be remediated.

At this stage of the project, it would also be beneficial to gather information about cleanup projects that are planned, being conducted, or complete. Thus, the IAEA can get a general picture of what problems are prevalent in such projects. This will also give the IAEA and the countries participating in the project the opportunity to meet with several of the companies performing environmental cleanup of radioactively contaminated sites.

The third volume of this three step TC project covers the technology for restoration of the contaminated sites, including removing, safeguarding and disposal of the radioactive waste. Another objective will be to review the available experience in the field of environmental restoration including current trends in research and development. The United States Department of Energy (US DOE) is, through (among other projects) its Remedial Action Program, an institution with tremendous experience in the field of environmental restoration in relation to uranium production wastes.

The restoration efforts at Germany's Wismut sites in Saxony and Thuringia, from which we will hear more of during this workshop, could be used as a model for potential restoration projects as the operation of these facilities took place in socio-political conditions similar to those in the other CEE countries. Although the Wismut operations were of a larger scale the problems at some CEE sites may be comparable. Canada, France, Spain, Sweden and UK are also countries having faced and are now facing environmental contamination issues. They are here to share their experience in identification and characterization of contaminated sites.

## 7.       CONCLUSION

The IAEA is pleased to present the results of this Technical Co-operation Regional Project on Environmental Restoration in Central and Eastern Europe. The IAEA is definitely eager to address the environmental situation in regional like this because there are several common features and similar problems. The participating Member States are in close geographic proximity and the socio-political conditions are comparable as well.



**IDENTIFICATION AND RADIOLOGICAL CHARACTERIZATION OF  
CONTAMINATED SITES: CRITERIA AND METHODS**

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**Abstract**

The purpose of a site characterization programme centres on the need to obtain specific radiological data concerning areas that have become radioactively contaminated as the result of accidents, uranium mining and milling or other nuclear activities. A site characterization requires a logical and planned approach in order to obtain the data necessary for planning an environmental restoration programme. The data is used to determine effective and appropriate techniques and sequencing to support the environmental restoration effort. This data is also needed for planning radioactive waste disposal, assessing accurately scheduling and estimating the cost of the environmental restoration programme. The first step in any characterization effort is the development of a plan. The basis for this plan is provided by definition of the final restoration objective(s). The characterization plan may require preliminary investigations to support existing information. Based on the objectives of the plan, data acquisition instruments and protocols are selected that best obtain required data in the most cost-effective manner. Radiological information is obtained by a combination of in situ measurements and laboratory sample analysis.

**1. BASIC INFORMATION ON THE AFFECTED AREA**

During preliminary planning for cleanup, the objectives of characterizing a contaminated area are as follows:

- a) To assist the cleanup team to get a good knowledge of the affected area, including such things as land types and usage, geology, hydrogeology and population distribution. The information should enable the team to assess how the affected area interrelates with surrounding regions and the implications to affected communities of actions taken.
- b) To assist in the selection of the best cleanup methods for individual zones.
- c) To select potential disposal sites and transport routes of waste arisings.
- d) To understand the hydrogeological systems and select area specific parameters for use in computer codes so that pathway analyses can be done and the impact of cleanup assessed.

The collection of data should start with those which are most readily accessible and then proceed to the least accessible data. The information should be reviewed periodically to ensure that it is relevant and up to date.

Good maps and regional atlases are invaluable sources of information; these include road and city maps, topographic maps and survey maps prepared by government agencies or industry. These maps show things such as road systems, urban and rural regions, land contours, surficial geology, natural resources, mineral deposits, land use, groundwater contours and surface water systems. Both large and small scale maps are very useful in analysing the area and its interconnection with other areas.

Other sources of information include aerial photographs, demographic data and projections, land use forecasts and agricultural, forestry and mining industry data. Other information comes from the siting, environmental assessment and licensing of nearby facilities such as power plants, dams, municipal and town waste disposal areas. In many countries, these data are readily available but they need to be collected and collated by those in charge of planning the cleanup.

However, more detailed analyses will generally have to be done to get the information required to model the affected area, predict the interaction of the contamination with the soil and determine the pathways to man. The type of information required and some of the means of obtaining the data are shown in Table I. Procurement of enough data to accurately characterize each zone in the affected area would be very costly. However by using available information and selective sampling, a fair understanding of the geology, hydrogeology and geotechnical aspects of the area can be determined.

If the whole area is fairly homogeneous and simple geologically then sample requirements and modelling will be much less onerous.

## 2. PLANNING AND IMPLEMENTATION OF ENVIRONMENTAL CHARACTERIZATION

The types of information that should be developed by the management team during preliminary planning for cleanup, and upgraded during final planning are summarized in Table II. Environmental and radiological characterization criteria and methods will be unavoidably affected by the overall planing for cleanup.

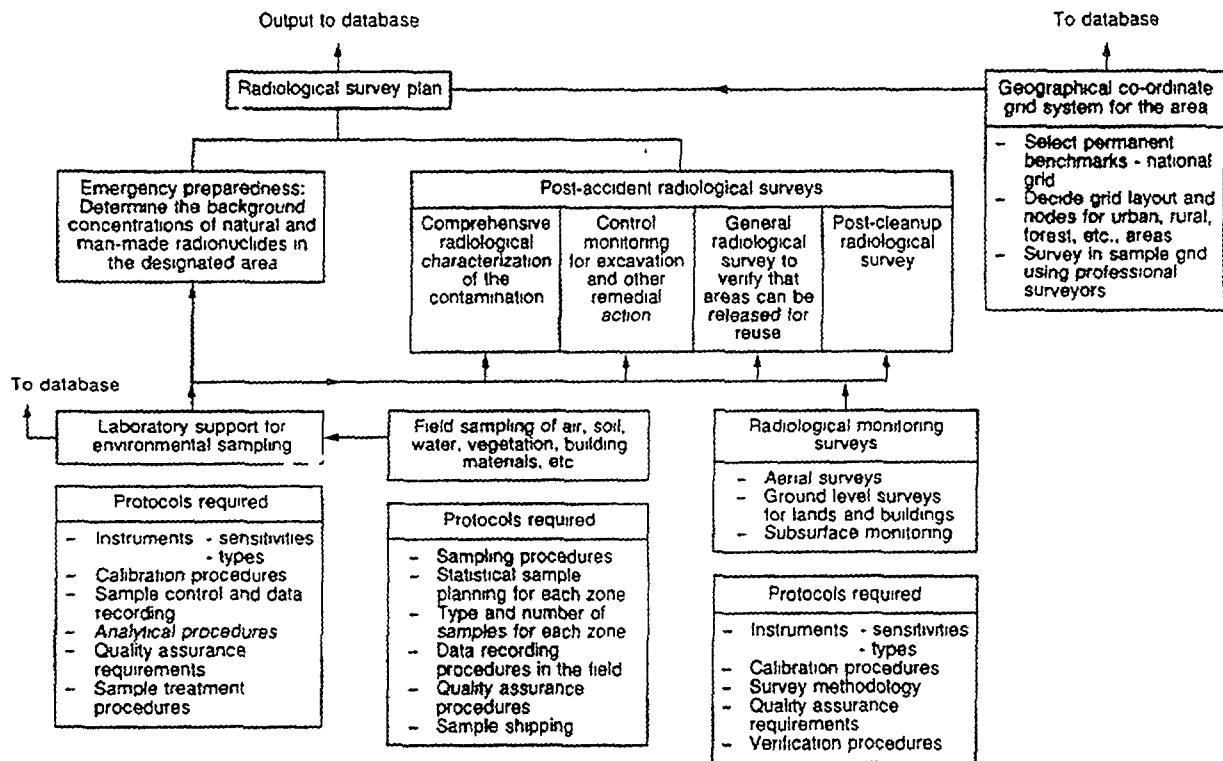
### 2.1. Objectives of a Radiological Survey Plan

A good radiological survey plan is an essential element in planning and implementing the cleanup of contaminated areas. This plan has the following objectives (Fig. 1):

- (i) To provide a comprehensive radiological characterization of the contamination and to characterize the different areas after various stages of cleanup;
- (ii) To determine how and to what extent radionuclides are moving as a result of runoff, resuspension, migration, plant uptake and so on;
- (iii) To do control monitoring during cleanup to decide on further actions;
- (iv) To have a general radiological survey at the end of cleanup operations to verify that the activity and dose rate levels in the affected area are within levels acceptable for the purpose for which the area will be used;
- (v) To ensure that the dose to the affected population remains within acceptable levels.

Figure 1 shows the various components of the radiological survey plan and indicates the interrelationships of the survey data acquired during such a process. The figure also lists some of the protocols that need to be developed for each component of the plan.

The majority of the data for these purposes would probably be obtained as a result of radiological monitoring surveys using non-destructive scanning and monitoring equipment either mounted in aircraft or ground based vehicles or hand held. Laboratory analysis of selected samples of air, soil, water, vegetation, building materials, etc., is also required to validate radionuclide ratios if they are likely to change.



**Figure 1**  
**Example of a radiological survey plan**

Each of these monitoring functions would produce a large amount of data which would provide the essential information required to characterize the radiological contamination and to plan and implement the cleanup. To ensure that these data are of maximum value and that results are readily available to the cleanup teams, it is important that:

- Protocols be defined, outlining step by step the methods to be used in taking and analysing samples, in taking monitor readings, in presenting data, for validation of analytical methods, comparison of different analytical methods, etc.
- The teams be well trained to follow the protocols and use the instruments. The provision of protocols and training is particularly important since inexperienced people may have to be trained and used because of the likely magnitude of the cleanup problem and the limited number of available experts.
- The location of the sample points be unambiguously referred to a standard grid system.
- The number and type of samples taken during the cleanup should not greatly exceed the throughput of available laboratories.

If the off-site dispersion of radionuclides from an accident at a nuclear facility occurs while wind speed and direction are uniform and there is no precipitation, plotting the distribution of radionuclides and dose isopleths may be relatively straightforward. On the other hand, if the dispersion occurs while there are localized heavy showers and/or gusty multidirectional wind currents, radioactive hot spots would occur and dispersion patterns would be complex and more difficult to define. If the release is over a longer period, a more complicated dispersion pattern may be expected in any case.

Whatever the deposition, the radiological survey plan should define the distribution and concentration of radionuclides accurately to determine hot spots and assist in setting cleanup priorities.

## **2.2. Protocol Development**

During the cleanup of large contaminated areas, many people having different abilities would have to be used as surveyors, samplers, laboratory assistants, etc. The level of training of these people would vary considerably. Also, a large number of measurements and samples would have to be taken to validate that the cleanup meets the set radiological criteria. Therefore, detailed protocols outlining step by step procedures for a variety of tasks associated with the cleanup should be prepared in advance to ensure that the tasks being undertaken by different operators are performed in as consistent and reproducible a manner as possible. In addition, statistical sampling protocols should be established to limit the number of samples to the minimum required to validate that the cleanup meets the established criteria.

Protocols for some of these functions have already been developed and these should be reviewed during preparation of the required protocols for large scale cleanup. For example, detailed survey and sampling protocols and procedures based on quality control and statistical analysis principles have been developed for remedial action and other programmes. These procedures are being modified continually to improve efficiency as a result of cleanup actions on relatively large areas ranging from 0.5 to 5 km<sup>2</sup> and the movement of large volumes of contaminated material.

Other good sources of certain protocols and procedures are the manufacturers of the instruments and equipment that would be used during the cleanup.

## **2.3. Sampling and Measurement**

The overall sampling protocol should be designed with speed and efficiency in mind but it must provide consistent and reproducible results. Sampling procedures should take into account the possibility of field sampling errors. These procedures should be developed as a matter of preparedness in parallel with laboratory analytical procedures.

Where possible, field monitoring techniques should be used rather than physical sampling and laboratory analyses of soil, water, flora and fauna. In fact, field monitoring would probably be the major data acquisition method, with samples only being used for key quality control measurements, definition of radionuclide ratios as required, analysis to determine contamination depth, and some final verification. The use of field monitoring techniques not only saves sample procurement and analysis costs but also reduces delays in getting results back to cleanup crews.

Information on the need for and location of samples to determine contamination depth can be gained if computerized pathway analysis programmes are available. For agricultural areas, with knowledge of the soil type, soil partition coefficients, stratigraphy, hydrogeology, meteorological conditions, etc., calculations can be done to estimate the depth of penetration and the migration rate and direction of radionuclides. For urban areas, understanding the drainage systems and building materials used would assist in determining where sampling should be carried out. For lakes, rivers and reservoirs, samples of the water and sediments should be taken during the cleanup and for long periods after termination of the cleanup, especially in the most seriously contaminated areas. In forested areas, sampling of the forest floor layer as well as leaves should be done periodically to determine whether the uptake of radionuclides by the trees is changing.

## **3. RADIOLOGICAL CHARACTERIZATION**

### **3.1. Introduction**

Before any cleanup action can be initiated, a detailed picture of the type, characteristics, mix, concentration and spatial distribution of the radionuclides must be determined. A preliminary assessment would be based on a knowledge of the source terms, operational and emergency monitoring

instrumentation, weather conditions and other conditions resulting in area contamination. However, this information may be inadequate to assess the actual distribution and concentration of radionuclides in the affected area to permit the cleanup operation to commence in an efficient manner.

Characterization efforts should be designed to provide information to determine which, if any, cleanup actions should be initiated in various zones. The precision and accuracy of the conclusions resulting from characterization data analyses should be in proportion to the cost sensitivities associated with each cleanup alternative. That is, as the cost of the remedy increases, the need for more precise knowledge of waste characteristics and volumes also increases. Conversely, data needs should be limited by some predetermined, acceptable cost risk. For low cost alternatives, as an example, it may be acceptable to incur a relatively large error in waste volume estimates; while higher cost alternatives will tend to require more precise estimates. General guidance on using an optimized phased approach is given below.

a) *Screening surveys*

Initial efforts should be directed to delineating areas of major concern. In situations involving widespread contamination, aerial surveys are a cost effective method for rapidly delineating and quantifying such areas. Assessment of aerial survey data may provide sufficient information to support design of interim measures necessary to mitigate the effects of high levels of contamination, or be used to locate areas requiring further definition. Aerial surveys are usually incapable of precisely locating contamination boundaries, or pinpointing hot spots. Ground based scanning systems mounted in vans or trucks are capable of better definition, but require more time, and are limited to areas which have relatively good access.

b) *Boundary definition surveys*

Precise delineation of the limits of the contamination can be accomplished using hand held instruments that are sufficiently sensitive to detect levels of concern. Mobile scanning systems may be used if access is sufficient to provide the desired level of definition. The outer boundaries of the contamination should be well marked and mapped to aid future characterization and/or remedial action efforts.

c) *Waste characterization*

Knowledge of characteristics and volumes of the waste arisings may be necessary for assessment of various remedial action alternatives. In such cases, laboratory analyses of the environmental media of concern should be obtained to correlate isotopic concentrations with field instrument readings and to help predict contaminant behaviour in the environment. Depending on the local surface geology and hydrology, subsurface soil samples, sediment samples and down-hole gamma logging data should be analysed to evaluate migration. Additional field readings may be required to better define waste volumes. In this case, an approach utilizing systematic measurements within an established grid may be applicable.

### **3.2. Monitoring Equipment and Techniques**

In this section the methods available to characterize the actual distribution and inventory of contaminated materials are briefly described.

The techniques include: airborne and vehicle borne monitors, semi-portable and hand held instruments, air and soil (surface and subsurface cores) sampling, etc. It is beyond the scope of this paper to review in detail the types of detectors and analytical techniques which are available for such purposes. However, examples of special pieces of equipment which should be very useful are briefly described.

For many years remote gamma sensing from aircraft has been an effective way of rapidly locating, monitoring and mapping gamma activity on the ground. Helicopters or fixed winged aircraft can be used as the platforms for sensitive NaI or germanium detectors to measure total gamma count rate. Helicopters are used for low level work where maximum sensitivity is required. Positioning of the aircraft during surveys is accomplished with microwave locating systems which feed indicators to guide the pilot accurately along preselected routes. Gamma signals, flight path, altitude and meteorological data are fed into an onboard data acquisition system for post-flight analysis. Gamma survey data overlaid on aerial photographs indicate the location of the contamination very accurately. Identification of the gamma emitting radioisotopes is done using a germanium detector, a multichannel pulse height analyser and computer. This information is immediately useful for planning purposes. Depending on the radionuclide mix, it may also serve as data with sufficient integrity to guide initial cleanup actions. These systems are also useful for a final survey to confirm that the cleanup meets the required standards. Figures 2 and 3 show examples of a helicopter mounted unit and calibration pads for such units, respectively.

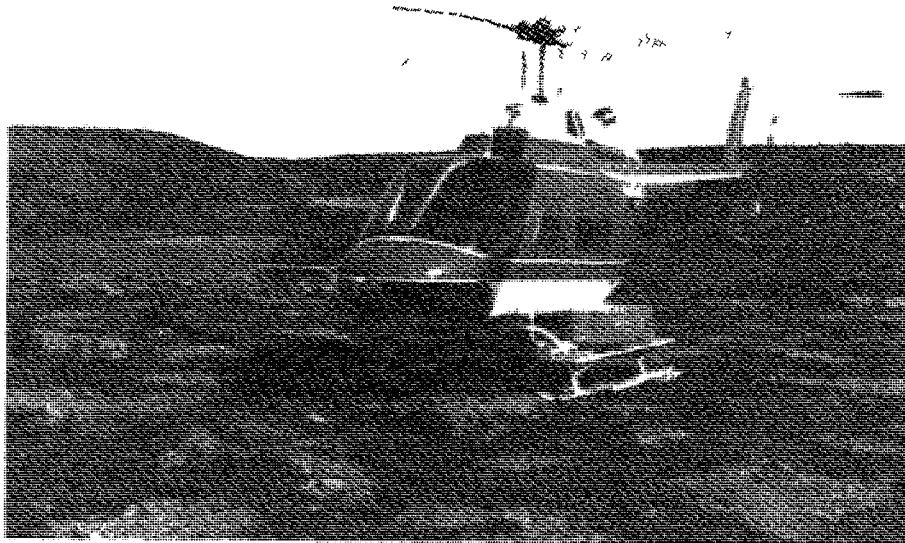
Vehicle borne monitoring systems can be equipped with more elaborate equipment including microwave and UHF ranging systems. Alternatives include satellite global positioning systems and possibly inertial navigation. Since vehicle borne systems can more easily monitor at one spot longer, traverse more slowly and get closer to the contamination, such systems generally provide improved detection sensitivities and greater resolution of changing contamination. The ability to resolve changing concentrations can be very important if it gives the opportunity to show that certain areas need no further cleanup. A variety of ground based systems are available. Figure 4 shows a system (called RTRAK) which is currently in use for large scale cleanup in the Uranium Mill Tailings Remedial Action Project in the USA. The tractor borne gamma scanning system is coupled with microwave telemetry to conduct rapid scanning, automatic recording of location, complete coverage of the area of interest and automatic data entry and plotting. Figures 5 and 6 show other representative types of such monitoring systems.

Very sensitive high volume Ge-Li detectors have been used on a variety of vehicles and platforms and in many geometries. They improve the ability to interpolate data between measurement points if large samples are either collected or measured in situ. Several workers have reported the advantages of mounting such detectors on extendable booms with repeatable fixed geometries. With this technique it is possible to scan over a large area and this improves the statistics and quality control of the analyses.

In developing the plan for monitoring for contamination distribution and inventory, the required laboratory analyses should not be overlooked. To completely characterize the contamination in an area, chemical analyses are required, especially if the contamination has penetrated well below the surface. Such samples are also required to determine the ratios of various radionuclides in the contamination. For example, if only the gamma activity is monitored after an accident at a nuclear power plant, the ratios of  $^{137}\text{Cs}$  to  $^{90}\text{Sr}$  must be determined from soil radiochemical analysis. Similarly, if the  $^{241}\text{Am}$  photon is used to map the associated  $^{239}\text{Pu}$ , the ratio of Am/Pu must be determined by radiochemical analyses. In cases where these ratios are not constant, significant soil analysis is required. Both fixed and mobile analytical laboratories could be considered, depending on needs.

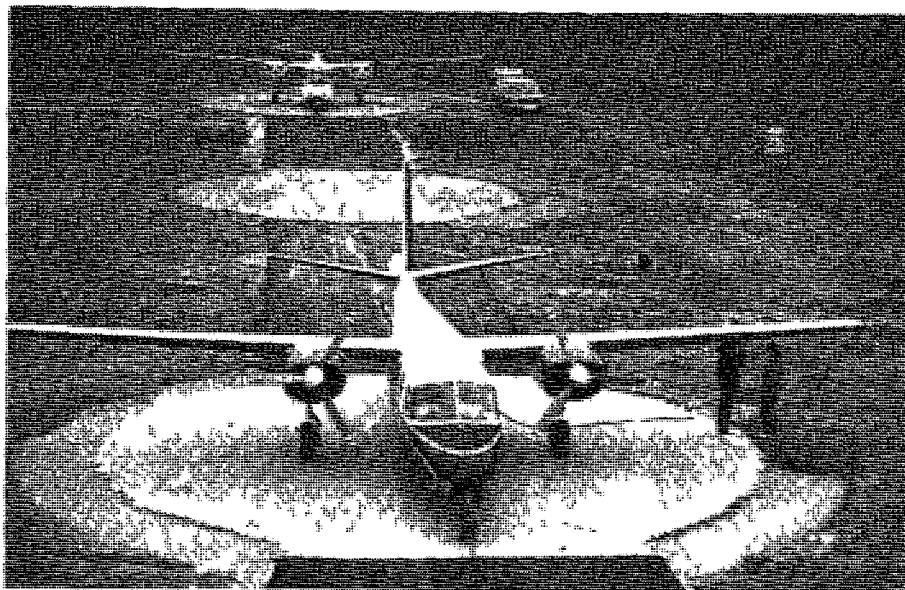
In addition to the sensitive airborne and vehicle borne gamma survey systems, a wide variety of hand held alpha, beta and gamma detectors are available for ground verification of the activity, especially on buildings and equipment. Portable high resolution germanium diode spectrometer systems have been developed which are capable of measuring transuranics, activation products and fission products at sensitivities below uncontrolled release criteria. Figure 7 shows one example of this type of instrument.

Experience in the United States Department of Energy (USDOE) remedial action programmes involving the cleanup of thousands of hectares indicates that a hand held sodium iodide detector, coupled with a ratemeter, is a most effective tool for locating surface depositions of gamma emitting



*Helicopter equipped with one NaI(Tl) gamma ray sensor on each side for use in rapid area surveys. The gamma signals, flight path, altitude and meteorological data are fed into an onboard data acquisition system for ground based studies (Credit Scintrex Ltd.)*

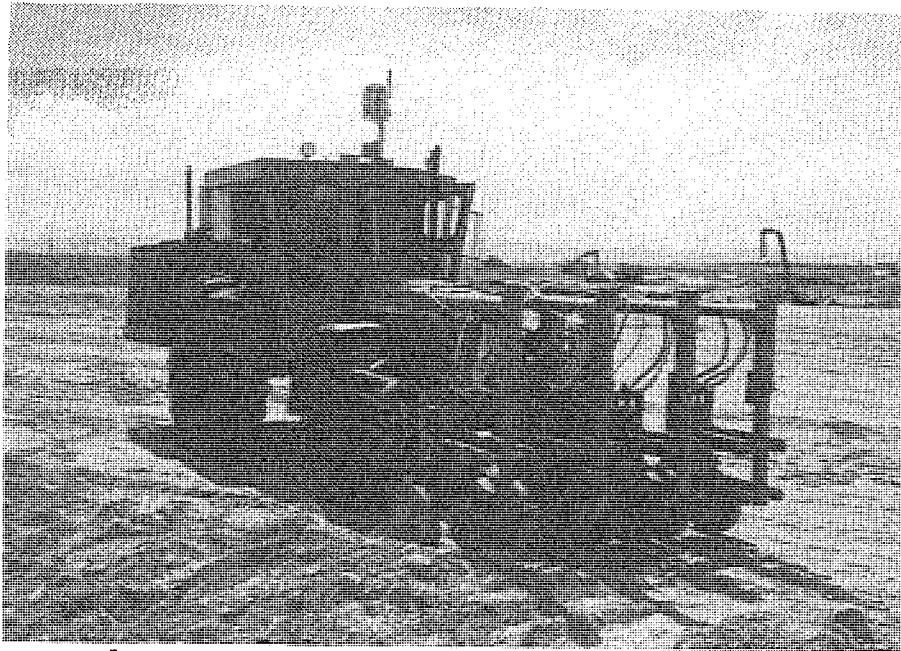
**Figure 2**



*Calibration pads for airborne and ground gamma spectrometers (Credit Swedish Geological)*

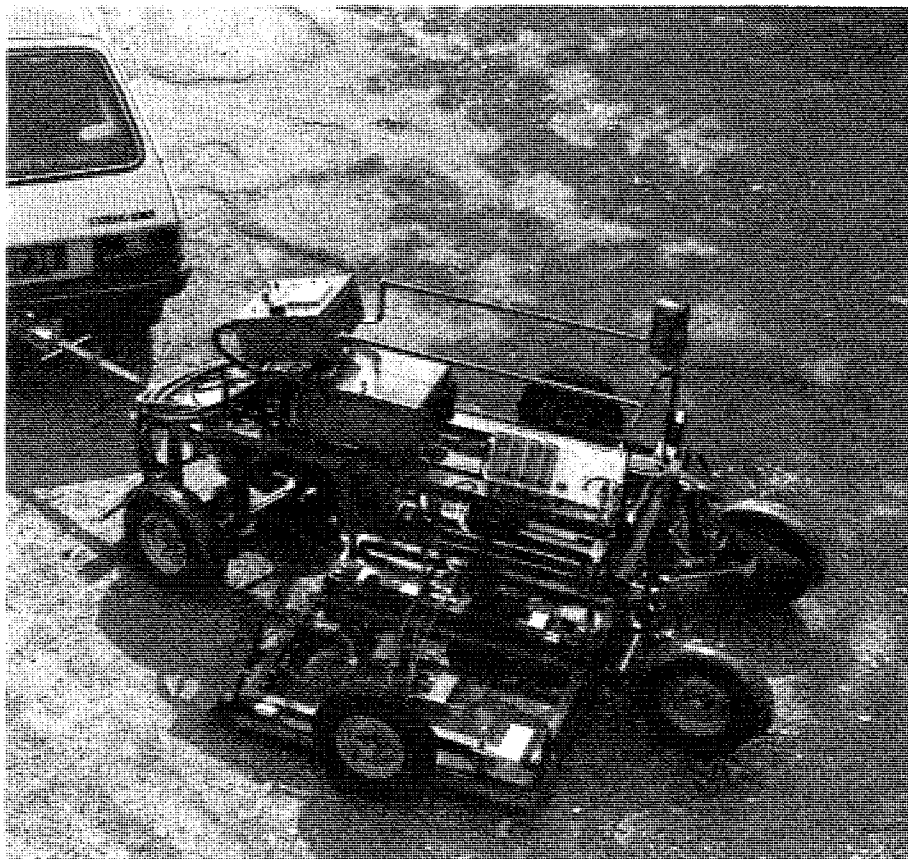
**Figure 3**





*Tractor borne surface contamination monitor for all types of terrain to scan a 2.5 m wide strip at  $3 \text{ km} \cdot \text{h}^{-1}$  using four NaI(Tl) gamma detectors mounted at the front. The system is coupled with microwave telemetry for rapid scanning, automatic data entry and plotting. Paint sprayers behind the detectors automatically mark hot spots for cleanup crews. (Credit: USDOE, M.K. Ferguson, Chem-Nuclear Systems.)*

**Figure 4**



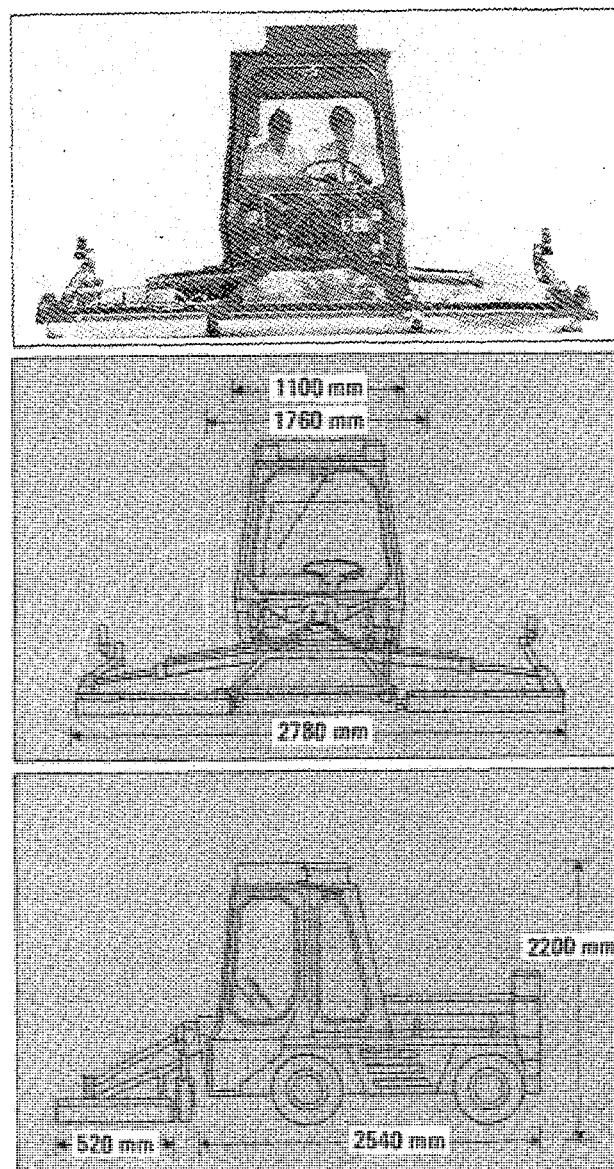
*Road contamination monitor consisting of a gas flow proportional counter ( $\text{Ar}-10\% \text{CO}_2$ ) mounted on a vehicle drawn trailer. The effective width of the monitor is 180 cm. For alpha/beta/gamma contamination. (Credit: NOVELEC CEA.)*

**Figure 5**



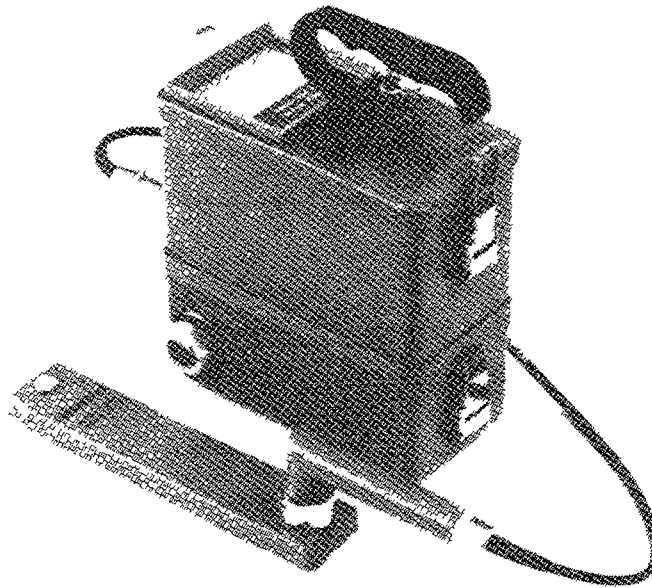
radionuclides. Sensitivities as low as  $0.1 \text{ Bq.g}^{-1}$  for  $^{226}\text{Ra}$  and  $0.1 \text{ Bq.g}^{-1}$  for depleted uranium can be achieved without the need for expensive spectrometer systems. This approach is also used in these programmes to identify hot spots after completion of the remedial action. A recent development couples this scanning technique with an ultrasonic location device to provide for automated data acquisition and computer produced isopleth mapping. Such hand held or back pack equipment is reported to offer substantial operational advantages such as low cost, ruggedness and mobility.

During preliminary planning, minimum performance standards should be specified, when possible, for the instruments and techniques used in the monitoring and sample analysis programmes, including: minimum sensitivity for radionuclides expected to be present, error and confidence levels.



*Road survey monitor consisting of 10 gamma compensating gas flow proportional counters (Ar-10% methane) mounted on the front of the vehicle. Sensitive area of each beta/gamma counter is  $600 \text{ cm}^2$ . Separate counters are provided for gamma and beta/gamma monitoring to allow detection of beta contamination in a background gamma field. (Credit: British Nuclear Fuels plc.)*

**Figure 6**



*A portable high resolution germanium diode spectrometer system capable of measuring transuranics, activation products and fission products at sensitivities below the uncontrolled release criteria limits for the USA. (Credit: Battelle Pacific Northwest Laboratories.)*

**Figure 7**

It is beyond the scope of this paper to specify required sensitivity, error and confidence levels and other related parameters since such factors are dependent on energy, radionuclide and time, and require specialized knowledge.

A large number of individuals and groups will be taking radiation measurements and samples for analysis during cleanup. Since it is important that measurements are made in a consistent and reproducible manner and appropriate instruments are used, protocols should be established during preliminary planning for instrument usage. These protocols should define parameters such as measurement time and distance of instrument from surface. Since personnel having little or no past experience in radiation monitoring may have to be involved, these protocols for instrument usage should be quite explicit and suitable training programmes should be established.

The specific locations of key radiation measurements should be well defined and recorded so that they can be subsequently checked after a regional cleanup operation or during final release of an area. These data points would also be useful for observing changes in environmental radiation readings which could result from the migration of radionuclides from elsewhere.

TABLE I  
TYPES OF DATA REQUIRED TO CHARACTERIZE, ANALYSE  
AND MODEL TERRESTRIAL SYSTEMS

### **Geological characterization**

*Stratigraphy* -- stratigraphic relations with surficial deposits, as well as rock underlying the site or having potential hydraulic connections with regional aquifers

*Lithology* -- including mineralogy, texture and fabric (grain size parameters) and classification

*Structure* and discontinuities in both soil and rock influencing groundwater flow

### **Hydrological/hydrogeological**

*Regional hydrogeology* -- including description of groundwater and surface water systems, recharge/discharge zones, natural surface drainage, limits of the hydrological system, flow directions, interrelation with surrounding water systems

*Parameters* -- for example for the soil: hydraulic conductivity, permeability, porosity, anisotropy, water holding parameters, hydraulic potential; for water systems: flow velocity and volume in summer and winter, lakes without surface inlet/outlets

**Geochemical/geotechnical parameters** -- for example: partition coefficient ( $K_d$ ), grain size distribution, clay mineralogy, ion exchange capacities, soil pH, surface and groundwater chemistry

**Techniques** -- including: laboratory studies, borehole logging, remote sensing, in situ tests and measurements, airborne and ground geophysical surveys, geological field mapping

**TABLE II**  
**SUMMARY OF ITEMS TO BE INCLUDED IN PRELIMINARY**  
**AND FINAL PLANS**

**Rationale and goal for cleanup**

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**Logistic support for cleanup**

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- provision of required personnel
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**Transportation plan**

**Waste disposal plan**

**Radiation protection and safety plan**

# RADIOECOLOGICAL PROBLEMS OF THE REPUBLIC OF BELARUS AFTER THE CHERNOBYL ACCIDENT AND THE WAYS OF THEIR SOLUTION

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## Abstract

The data on the scale of contamination of Belarus with radionuclides after the Chernobyl accident are given. The overall contaminated areas with radiation density of caesium-137 exceeding  $3.7 \cdot 10^4$  Bq/sq.km amount to 46,5 thousands of square kilometers. It is shown, that in 1993 the radioactivity at the territory of Belarus accounts for more than  $3.7 \cdot 10^{16}$  Bq. The paper presents the characteristics of the main contributors to the radiological situation. The characterization of the patterns of contamination of soil, water systems, forests is considered. The paper gives the organizing structure of control over the processes of elimination of the post-effects of the Chernobyl accident. The need for urgent development of the project for changing the contaminated territories into ecologically safe system is indicated. The suggestions on organizing the International Safety System against accidents at potentially hazardous industrial enterprises are given.

From 1991 Belarus is an independent republic. The area of the territory of Belarus amounts to 207600 square kilometers. In the East Belarus borders on Russia. In the North on Lithuania and Latvia. In the West- on Poland and in the South- on the Ukraine. The enormous plain areas are predominant in Belarus. Maximum altitude is 370 m. Certain rivers flow into the Baltic Sea, the rest of them go to the basin of the Black Sea. Climate is mild continental and temperate-humid. Size of the population totals 10.3 millions. 67 % of population live in towns. The largest cities are: Minsk - 1634, Gomel - 503, Mogilev - 363, Vitebsk - 362, Grodno - 285, Brest - 277 thousands of people.

In connection with above mentioned cities the Republic is divided into six Regions. 78% of peoples inhabiting Belarus are Byelorussians; 13 % - Russians; 4 % - Poles; 3 % -Ukrainians.

As a result of the Chernobyl accident more than 20 percent of the territory of Belarus with 3668 populated areas have been contaminated by radionuclides, with radiation density of caesium-137 exceeding  $3.7 \cdot 10^4$  Bq/sq.m. According to the data of 1993, the overall area of the contaminated territory totals 46,5 thousands of square kilometers. After the accident Belarus becomes the zone of ecological disaster. It is necessary to note the following characteristic properties of contamination.

1. Extensive experimental and theoretical work on the fuel and radionuclide balance of the Unit-IV of the Chernobyl NPP have shown that the emergency discharge of activity accounts for  $3.7 \cdot 10^{19}$  Bq ( instead of earlier determined  $1.85 \cdot 10^{18}$  Bq). The short-lived ra-

dionuclides ( iodines, neptunium, krypton, xenon and others) have been the main contributors to radioactivity. More than  $1.85 \cdot 10^{18}$  Bq have been discharged only with fragments of the core. Belarus experienced the main load of the initial period of the discharge of radioactivity. The results obtained nowadays on restoring the radioactive damage and exposure doses at the initial stage of the accident show that the latter are essential in estimation of radiological situation.

2. Contaminated territories show pronounced variation in density of radionuclides. During several days after the beginning of the accident, the uneven deposition of radionuclides on the large areas of Belarus occurred. Such pattern of contamination has been formed as a result of the peculiarity of the dynamics of the abnormal process in the reactor, measures taken for confining the accident and meteo-



Fig Contamination of the territory of Belarus with caesium-137

rological conditions. At present, there are places with density of contamination with caesium-137 in the range of  $3.7 \cdot 10^4$  Bq/sq.m. to  $3.7 \cdot 10^7$  Bq/sq.m. Even at small territories the density varies significantly. For example, in the village Kaliban of the Bragin area of the Gomel Region the density of contamination with caesium-137 varies in the range of  $18.5 \cdot 10^4$  -  $259 \cdot 10^4$  Bq/sq.m. In accordance with norms and regulations in force (Regulations for Radiation Safety RRS-76, SPORO-85), the parts of the territory because of their specific activity are the low-level and intermediate-level wastes.

Provided for the large gradient of density of contamination of the territories, there is the real treat of a new catastrophe at the expense of the secondary contamination with radionuclides by airborne, water, technologic transport especially in a case of natural disasters and unfavourable climatic conditions. Theoretical and experimental studies and the regrettable experience of the secondary catastrophe in Kyshtym in 10 year after the accident are the basis for these conclusions. The secondary contamination of large territories took place because of airborne transport of  $2.22 \cdot 10^{16}$  Bq of activity.

3. The large scale of contamination of the territory and long-duration of emergency state of the Republic . Presently, there are  $3.7 \cdot 10^{16}$  Bq of activity at the territory of Belarus with Cs-137, strontium-90 and plutonium- 239-240 as the main contributors to the radiological situation. Half-lives of the above radionuclides make-up 30 years, 29.1 years, 24390 years, 6537 years respectively. It defines the long-time emergency situation in Belarus. The analysis of the dynamics of behaviour of the overall activity in Belarus without reference to migration show that even in 2001 overall activity will total  $3.108 \cdot 10^{16}$  Bq. The increase of activity of americium will take place.

The Table 1 gives the dynamics of change of total activity in Belarus.

Table 1

Year	May 1986	December 1992	December 2001	December 2016
A	$4.4 \cdot 10^{18}$ Bq	$3.7 \cdot 10^{16}$ Bq	$3.108 \cdot 10^{16}$ Bq	$1.998 \cdot 10^{16}$ Bq

According to contamination with caesium-137, Belarus is characterized as follows:

From  $3.7 \cdot 10^4$  to  $18.5 \cdot 10^4$  Bq/sq.m. - the area of 29.9 thousands of square kilometers;

From  $18.5 \cdot 10^4$  to  $55 \cdot 10^4$  Bq/sq.m. - the area of 10.2 thousands of square kilometers;

From  $55 \cdot 10^4$  to  $148 \cdot 10^4$  Bq/sq.m. - the area of 4.2 thousands of square kilometers;

Beyond  $148 \cdot 10^4$  Bq/sq.m. - the area of 2.15 thousands of square kilometers.

The Gomel and Mogilev Regions are the most damaged. According to the data of the State Committee for Hydrometeorology, contamination with strontium and plutonium is of local character. The areas of contamination with strontium-90 with density more than  $3.7 \cdot 10^4$  Bq/sq.m. account for about 600 sq.km .

4. The effect of radioactive fallouts in Belarus combines with harmful chemical contaminations of individual regions. Thus, in particular, high content of NO and H S is observed in Mogilev.

2 2

Therefore, the combined effect of contaminants of the territories should be considered.

Let us consider in details the radioecological state of soils, water, forests and other contaminated subjects. The area of contaminated agricultural lands with caesium-137 density more than  $3.7 \cdot 10^4$  Bq/sq.m. totals 16 thousands of square kilometers, 2.6 thousands of square kilometers of them are completely excluded from the usage.

The total area of agricultural lands amounts to 94 thousands of square kilometers. About 12 thousands of square kilometers of agricultural land are subject to erosion and 5 thousands square kilometers are classified as the lands of severe erosion.

Water-marshy areas play an important role in the system of water resources, amounting to 10 thousands of square kilometers. On the whole, radionuclides are found in the upper layer of soil in the depth 5 - 30 cm, depending on the type of soil and its ploughing. Depending on composition, soils of Belarus are divided into 4 types: sandy-loam -49 %; clay-loam -27 %; sandy -19 %; peat -5 %.

Vertical migration in clay soils is slow. In sandy and peat soils migration is more rapid. Presently, the content of water-soluble caesium is insignificant and accounts for not more than several percent.

According to the data obtained in 1992 for the Vetka area of the Gomel Region, the water-soluble forms of caesium account for 3.8 - 5 %. The content of water-soluble strontium exceeds the content of water-soluble caesium. In analogous conditions the water-soluble strontium amounts to 11 - 26 %. The studies carried out in the same conditions have shown, that water-soluble forms of Pu-239 account for 2.8 - 9.5 %. The greatest effect of extraction, that is 50 %, has been reached with the help of solutions of nitric acid.

The contaminated territories of forests amount to 16.85 thousands of square kilometers. The overall areas occupied by forests in Belarus amount to 58 % of the whole forest territory. Fir-grove occupies 11%, birch grove - 17% and alderthicket - 8%. 60-year-old



Table 2.

Areas (thousands of hectares) of contaminational  
arable lands, pastures and meadows

with caesium:

The types of land	:Level of contamination in Bq/sq.m.*10 <sup>4</sup>		
	: 3.7-18.5 : 18.5 -55 : 55 - 148		
Arable lands and gardens	614	255	92
Pastures and meadows	431	140	49
Total	1045	395	141

with strontium :

The types of land	: Level of contamination in Bq/sq.m.*10 <sup>4</sup>		
	: 0.37 - 3.7 : 3.7 - 11.1 : more than 11.1		
Arable lands and gardens	200	58	8.6
Pastures and meadows	155	47	8.9
Total	355	105	17.5

forests account for only 13%. It should be noted, that presently there are large areas of unthinned out forests. The overall stockpile of wood totals 804 millions of sq.m .

The affected forest areas are divided into three types according to the density of contamination:

3.7\*10<sup>4</sup> - 55\*10<sup>4</sup> Bq/sq.m.- the area equals to 11 thousands of square kilometers - insignificant consequences for forestry;

55\*10<sup>4</sup> - 148\*10<sup>4</sup> Bq/sq.m. -the area equals to 1.7 thousands of square kilometers - restrictions in forestry;

More than 148\*10<sup>4</sup> Bq/sq.m. -the area equals to 0.3 thousands of square kilometers- all kinds of lumbering are forbidden.

In 1988 Polesse radiation forest reserve has been created which is the most contaminated region with total area 1.4 thousands of square kilometers.

The main fraction of radioactivity is found in a forest cover. According to the data of investigations, 60-80% of radionuclides are found in covers. It should be noted, that concentration of radionuclides in forests is significantly larger than in meadows, peats and other types of territories. In case of fires, forest covers, which are the most contaminated part of forest, burn up in the first place. In addition, the radioactive particles sorbed at the surfaces of peat and uptaken by plants turn into gaseous and aerosol state. The time of life of such radioactive cloud is about 7 days at the height 1,5 km, in the top troposphere - about a month, in stratosphere - 1-5 years, according to the data of High Engineering-Technical School for Firemen.

Everything indicates the danger of possible radiation catastrophes in case of large-scale fires at the territories contaminated with radionuclides.

Let us consider the radioecological state of water systems of Belarus on the basis of results obtained mainly by the State Committee for Hydrometeorology and the Institutes of the Academy of Sciences. The Republic of Belarus is characterized by large number of small rivers. 455 from them flow to the Baltic Sea and 555 flow to the Black Sea. The overall length of all rivers of Belarus totals more than 90000 km. The number of lakes exceeds 10000, but there are only 470 lakes with the area of water table more than 50 hectares. The majority of natural lakes are in the North of the country. Annual precipitation amounts to 650 mm. The main mass falls out from April till October. The river system of the Southern direction belongs to Dnepr. Pripyat', Sozh and Berezina are its tributaries. The Byelorussian rivers are characteristic of slight slopes, winding streams, braided slight watershed and availability of flood beds. At the time of spring flood, the flood beds are submersed for a certain period of 10 - 30 days.

In the contaminated areas there are the largest inflows: Braginka, Besed', Iput', Senna and others. Sod-podzol soils prevail in places of water parting. Alluvial soils are typical for valleys. They are formed under the effect of submergence at the period of flood.

Contamination of water reservoirs with radionuclides is typical for two regions. The first one, the South region includes the basin of Prypyat', Braginka, Vita rivers. In this region, the high dose rates have been observed at the initial stage of the accident at the expense of short-lived radionuclides. In the areas of the down current of Pripyat' and Braginka rivers the region is characterized by radiation density of Cs-137 from  $185 \cdot 10^4$  to  $740 \cdot 10^4$  Bq/sq.m., strontium-90 - to  $18.5 \cdot 10^4$  Bq/sq.m. and plutonium-239 - more than  $0.37 \cdot 10^4$  Bq/sq.m. The second, North region includes the basins of Dnepr, Sozh, Iput', Besed' rivers. Here, radiation situation has been formed at the account of atmospheric fallouts. Radiation density of Cs-137 reaches to  $555 \cdot 10^4$  Bq/sq.m. and Sr-90 - to  $3.7 \cdot 10^4$  Bq/sq.m.

Immediately after the accident, the concentration gradient of water of the lower reach has gone over allowable limit ( 13.6 Bq/l

for limited contingent of population). But already at the end of May 1986, the concentration decreased to 3.7 Bq/l. The annual average concentration of caesium-137 in Pripyat', Dnepr, Sozh, Iput', Besed' rivers is below the allowable limit and don't exceed the Republican control limits ( RCL amounts to 18.5 Bq/l). If at the initial stage of the accident the fallout of radioactive aerosols on the water surface has been the main factor of contamination of water, then the level of contamination has depended on the washing off the surface of the basin and the exchange with bottom sediments.

According to the data of the State Committee for Hydrometeorology, the total carrying out of caesium-137 by five above mentioned rivers amounted to:  $2.2 \cdot 10^{13}$  Bq in 1987,  $1.25 \cdot 10^{13}$  Bq in 1988,  $7.03 \cdot 10^{12}$  Bq in 1989,  $3.03 \cdot 10^{12}$  Bq in 1990,  $2.66 \cdot 10^{12}$  Bq in 1991. Sozh river is the main contributor:  $1.036 \cdot 10^{13}$  Bq in 1987,  $5.18 \cdot 10^{12}$  Bq in 1988,  $2.22 \cdot 10^{12}$  Bq in 1989,  $1.11 \cdot 10^{12}$  Bq in 1990,  $0.925 \cdot 10^{12}$  Bq in 1991.

The analysis of the character of flow shows, that in spring and in summer the main amount of caesium is carried out by flow of liquid. In autumn, the fraction of caesium carried on the dispersed particles increases. The main fraction of strontium falls at liquid flow. Such behaviour of strontium is connected with its high mobility and water-solubility. The analysis of contamination of the areas of the basin show that Cs-137 is the main factor of activity, but anomalies are possible. Thus, in Kulazhin village at the bank of the Nesvich river, the contamination with strontium exceeded the levels of contamination with caesium and accounted for  $112.85 \cdot 10^4$  Bq/sq.m. Ru-106 and Ce-144 largely contributed to radioecological situation of this place ( $22.57 \cdot 10^4$  Bq/sq.m. and  $5.18 \cdot 10^4$  Bq/sq.m., respectively).

It should be noted, that the increase in density of contamination has been observed here from  $566.1 \cdot 10^4$  Bq/sq.m. in March, 1991 to  $1927 \cdot 10^4$  Bq/sq.m. in October, 1991. The similar phenomenon has been observed in Yasen village on the bank of Braginka, where density of contamination with Cs-137 changed from  $1.85 \cdot 10^4$  Bq/sq.m. in March to  $21.46 \cdot 10^4$  Bq/sq.m. in October.

The main fraction of activity of water systems is found in bottom sediments. Their specific activities are several orders as high as in water samples. The measurements have been done on Nesvich, Slovechna, Braginka, Dnepr and Lina rivers. There were 11 places of sampling. In these places, specific activity of Cs-137 accounted for  $3.7 \cdot 10^4$ – $7.4 \cdot 10^2$  Bq/kg and of Cr-90–  $3.7$ – $11.1$  Bq/kg. The specific activities are of season character. Thus, in v. Kulanichi on the bank of the Nesvich river, specific activity of Caesium-137 was  $11.83 \cdot 10^3$  Bq/kg in March, 1991 and  $48.1 \cdot 10^3$  Bq/kg in October and in v. Gorodchan–  $1.48 \cdot 10^3$  Bq/kg and  $59.2 \cdot 10^3$  Bq/kg, respectively. At the same time, in v. Yaseni-Dublin on the bank of the Braginka river, the changes in specific activity are of the reverse character –  $4.44 \cdot 10^3$  Bq/kg in April and  $1.11 \cdot 10^3$  Bq/kg in October (for Cs-137). Transport of radionuclides by rivers and accumulation of them occur in certain places. For example, at the Iput' river ahead of the dam near the Dobrush town, the contaminated spot of silt has been formed with specific activity to  $10^5$  Bq/kg. According to the results of modelling, similar accumulations

can be found at other rivers, being of great danger for radiation situation.

In accord with the density of contamination, the territories are divided into 5 zones:

1. The evacuation zone ( alienation ) 30 km. zone

$$\text{Sr-90} > 11.1 \cdot 10^4 \text{ Bq/sq.m.},$$

$$\text{Pu-238-241} > 0.37 \cdot 10^4 \text{ Bq/sq.m.}$$

the regime of strict control;

2. The zone of immediate change of residence

$$\text{Cs-137} > 148 \cdot 10^4 \text{ Bq/sq.m.},$$

$$\text{Sr-90} > 11.1 \cdot 10^4 \text{ Bq/sq.m.},$$

$$\text{Pu-238-241} > 0.37 \cdot 10^4 \text{ Bq/sq.m.}$$

the regime of strict control;

3. The zone of subsequent change of residence

$$148 \cdot 10^4 \text{ Bq/sq.m.} > \text{Cs-137} > 55 \cdot 10^4 \text{ Bq/sq.m.}$$

$$11.1 \cdot 10^4 \text{ Bq/sq.m.} > \text{Sr-90} > 7.4 \cdot 10^4 \text{ Bq/sq.m.}$$

$$0.37 \cdot 10^4 \text{ Bq/sq.m.} > \text{Pu-238-241} > 0.185 \cdot 10^4 \text{ Bq/sq.m.}$$

or dose > 5 mSv ( 0.5 rem )/year

4. The zone with the right of changing the residence

$$148 \cdot 10^4 \text{ Bq/sq.m.} > \text{Cs-137} > 18.5 \cdot 10^4 \text{ Bq/sq.m.}$$

$$7.4 \cdot 10^4 \text{ Bq/sq.m.} > \text{Sr-90} > 1.85 \cdot 10^4 \text{ Bq/sq.m.}$$

$$0.185 \cdot 10^4 \text{ Bq/sq.m.} > \text{Pu-238-241} > 0.074 \cdot 10^4 \text{ Bq/sq.m.}$$

5. The residence zone with periodical radiation control

$$18.5 \cdot 10^4 \text{ Bq/sq.m.} > \text{Cs-137} > 3.7 \cdot 10^4 \text{ Bq/sq.m.}$$

$$1.85 \cdot 10^4 \text{ Bq/sq.m.} > \text{Sr-90} > 0.555 \cdot 10^4 \text{ Bq/sq.m.}$$

$$0.074 \cdot 10^4 \text{ Bq/sq.m.} > \text{Pu-238-241} > 0.037 \cdot 10^4 \text{ Bq/sq.m.}$$

doze < 1 mSv ( 0.1 rem )/year

The detailed picture of contamination of Belarus with Cs-137, Sr-90 and Pu-239-240 is given in the map. There are 1927 popula-

ted areas in the zone with the density of contamination  $3.7 \cdot 10^4$  -  $18.5 \cdot 10^4$  Bq/sq.m., inhabited by 1955 thousands of people; 942 populated areas inhabited by 270 thousands of people in the zone with the density of contamination  $18.5 \cdot 10^4$  -  $55 \cdot 10^4$  Bq/sq.m.; 330 populated areas inhabited by 46 thousands of people in the zone with the density of contamination  $55 \cdot 10^4$  -  $148 \cdot 10^4$  Bq/sq.m.; 48 populated areas inhabited by 6.5 thousands of people in the zone with the density of contamination  $148 \cdot 10^4$  -  $222 \cdot 10^4$  Bq/sq.m. and 22 populated areas inhabited by 3.2 thousands of people in the zone with density of contamination more than  $222 \cdot 10^4$  Bq/sq.m. The work on radiation survey has began in Belarus from the first days of the Chernobyl accident. The Institutions of the Academy of Sciences, State Committee for Hydrometeorology, Ministry of Agriculture, Ministry of Forestry have been engaged in this work. State Committee for Chernobyl of the Republic of Belarus controls the state of radioactive contamination of Belarus. State Committee for Hydrometeorology is charged with assessment and management of radiation situation in line with the law "The Legal Aspects for Territories Contaminated with Radionuclides as a Result of the Accident at the Chernobyl NPP". The map on radiation situation in the Republic of Belarus presented in the article is an official document compiled by the State Committee for Hydrometeorology. 54 fixed stations of the State Committee for Hydrometeorology control the radiation situation in the Republic. They are evenly located on the territory of Belarus. The map shows the places of location of these stations. In addition, the control over daily fallouts is carried out by 30 stations; filters and plotting boards are daily measured and analysed for the content of aerosols.

The Government of the Republic adopted the special program for elimination of the consequences of the Chernobyl accident. The expenditure for this program amounts to 4.9 milliards of roubles for 1991, 40 milliards of roubles for 1992, 154 milliards of roubles for 1993 year, accounting for 16.8 %, 12.6 % and 10.9 % of the State budget respectively. Until 1991, the expenditure has been allocated from the USSR's budget. In 1992 9 milliards of roubles came from Russia. Presently, Belarus, without a single NPP on its territory, faces these problems by itself. The main funds have been used for investments and compensation of privileges for population, year after year:

	1991	1992	1993
Investments	66%	53%	63%
Compensation of privileges	32%	43%	30%
Total	98%	96%	97%

As can be seen from the above data, the key funds have been invested in building of new settlements on the ecologically safe territory and spent on compensation of privileges for victims.

After disintegration of the USSR, the Byelorussian Government collects 18 % taxes from wages and salaires for financing the Chernobyl programs.

Such distribution of funds leads to the fact, that Belarus is unprepared for restoration of the contaminated territories, industrial contamination, radioactive waste management and ensurance of radiation safety of the territories with high density of contamination with radionuclides.

Inspite of the efforts directed to elimination of the consequences of the accident, the main problem, the problem of changing the contaminated territories of Belarus into ecologically safe system, hasn't been yet solved.

The Government of the Republic of Belarus, realizing its own responsibilities to Byelorussian people and to the people of other countries for possible transport of radionuclides from contaminated territories, created the International "Cleanup" Consortium, the aim of which is to unite efforts of firms and experts from all over the world directed to changing the territory of Belarus into ecologically safe system and development of the protection systems against repeated radiation and chemical damage in case of accidents at potentially hazardous industrial enterprises. The International Consortium is the nongovernmental organization, its activities extend from scientific investigations to presenting to the Government the ecologically safe territories with the special system of protection. Byelorussian and International firms are the members of the Consortium. The Byelorussian Scientific - Industrial Association and the Institute of Radioecological Problems are the main founders of the Consortium. The Institute of Radioecological Problems carries out the scientific guidance and coordinates the work in the field of ecology.

The basic conclusions from the above mentioned are as follows.

1. The problem of development of the project of changing Belarus into the ecologically safe system with allowance for the radiation and chemical contamination is considered to be the problem of paramount importance in 1994 - 1995.

2. Taking into account the aggravated ecological situation in Belarus, the Ukrain, Russia and in European countries, it is advisable to develop the International project on changing the contaminated territories into ecologically safe system and to create the system of protection from radiation and chemical damage.

3. It is necessary to work out the International Register of Hazardous Technologies as the base for ensuring safety and on the basis of the Register to develop the substantiation for safe residence of population with allowance for potential natural disasters and emergencies .

The International Register ought to contain technical characteristics of potential dangers and particular features of location of units. Complex criterion of danger should be the basis for entering the unit into the International Register.

4. Taking into consideration the transboundary after-effects of the accident at the potentially hazardous enterprises, it is advisable to create the International Agency for obligatory issuing licences and insuring the enterprises which come within the scope of the International Register for Hazardous Technologies.

It allows to prohibit hazardous technologies of poor quality and to compensate damaged countries for caused detriment.

## REFERENCES

1. Sharovarov G.A. The Problems of Restoration of the Territories of Belarus Contaminated as a Result of the Chernobyl Accident. Minsk, 1992 AS B , preprint, IREP., 15 p. ( In Russian).

2. The Chernobyl Catastrophe: Its Causes and Consequences. Edited by V.B.Nesterenko,D.S.Firsova, Minsk, 1993, 213 p. ( In Russian).

3. G. Sharovarov, J. Milligan, A. Buoni. Contamination of the Territory of Belarus as a Result of the Chernobyl Accident, NAEP, 18-th Annual Conference, Proceedings, 1993, North Carolina, pp.34 - 38, ( In English).

4. V.I. Gorbachev, G.A. Gotovchits, S.S. Ogorodnic, G.A. Sharovarov. The Problems and Prospects of Mitigation of the Consequences of the Chernobyl Accident, 1992, AS B, Minsk, 1992, 29 p. ( In Russian).

5. Zh. V. Davydova, S.V. Morozov, G.A. Sharovarov. The Methods of Mathematical Modelling of Distribution of Radioactive Discharges of Nuclear Power Plants, AS B, NPI, Minsk, 1990. (In Russian).

6. The Chernobyl Trace in Belarus, Minsk, 1992, 19 p. ( In Russian and in English).

7. The Materials of the Interdepartmental Conference (the Strategy in the Field of Environment of Belarus), Minsk, 1993. 211 p.,( In Russian).

# **RADIOACTIVELY CONTAMINATED SITES IN BULGARIA**

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## **Abstract**

The report contains a description of radioactively contaminated sites in Bulgaria due to uranium industry and polymetallic ore mining. A detailed picture of the most contaminated areas due to the two processing plants in Buhovo and Eleshnitsa is presented. The case of Buhovo is of special concern since due to continuous operation (1947 - 1958) without tailings pond large area is contaminated with radium. The source terms for some underground mines and in-situ-leaching are described and radiologically characterized.

A case of interest of large area radioactive contamination is the Bay of Vromos which is not related to uranium industry but to copper ore mining.

## **Introduction**

The major source for radioactive contamination of large areas in Bulgaria is the uranium industry. The coal burning power plants, the phosphate fertilizers, phosphogypsum, construction



materials, medicine and scientific research have much less radioecological impact and therefore they will not be discussed in the present report.

The total contaminated area by U-industry (mining, milling) is approximately  $2 \times 10^7 \text{ m}^2$ , including  $4 \times 10^7 \text{ m}^2$  forest. Part of the land is in mountains, in low productive regions, but the major part is agricultural land along the rivers Maritsa, Tundzha and Struma which had been lent for in-situ leaching.

The problems of occupational exposure will not be discussed in the present report since they are out of the scope of the meeting. The remediation measures already taken or planned are also not included in the report.

#### 1. Uranium industry in Bulgaria (U-industry).

The first uranium explorations were in the Buhovo region near Sofia and had been performed by German geologists before the II World War but no mining followed.

The U-industry in Bulgaria starts immediately after the end of the war. The first mines for U-ore are under peak Goten in Stara planina (The Balkan mountain), next to the Buhovo village in 1945 and the ore was directly sent through the railroad station of Yana to USSR (Yanska ruda - ore, later the name is used for ore with greater than 0.5% U-content). On a hill in the end of Buhovo the first processing facility was built in 1947 and the wastes were dumped in Manastirsko dere (Monastery gully) above Yana village. A tailings pond was built in 1958.

New sites were discovered, studied and put into operation in other regions - Smolyan, Dospat, Eleshnitsa, Sliven, Selishte, Kurilo, Beli Iskar, Partizanska polyana. The ore was transported to Buhovo for processing.

In 1966 a second plant was built in Eleshnitsa which processed ore from South Bulgaria. The plant was more modern but

it did not produce the final product (yellow cake) which was dried, grind and packed in Buhovo. The total number of workers for both milling plants was approximately 1000 and the total annual production of the final product is approximately 800 tones/annually or 35000 tones for the production period 1947-1992.

During this period other mines started operation - Melnik, Sarnitsa, Sborishte, Smolyanovci, Proboynitsa, Dolna Banya. Some of the sites were depleted and closed.

The experiments with the method of in-situ leaching of low grade ore started in 1968-69. This technology was applied for the sites "Orlov dol" in the Toplovgrad region, "Selishte" near Velingrad (the site before that had been a classical mine), in the Haskovo region, to the north of Plovdiv near the town of Rakovski, in the Struma valley near Simitli.

Later new sites for in-situ leaching near existing in-situ leaching mines started operation - "Mudren", "Vladimirovo", "Navusen", "Maritza", "Chukarovo", "Tenevo", "Okop", "Debar", "Troyan", "Ceretelevo", "Tsarimir", "Belozem", "Trilistnik", "Igralishte", "Gradeshnitsa". Some of the sites due to poor geological conditions were not put into operation after the initial studies - "Melnik", "Prepechene", "Maritza", "Zlatolist".

Approximately 15 sites for in-situ leaching for a period of 20 years were started. The sites were with rather poor (between 0.006 and 0.030%, or from 60 to 300 g/t) ore.

The leaching mines reduced the number of underground miners from 5000 in the period 1965-1970 to approximately 500 in 1988.

The in-situ leaching was experimentally applied also for leaching uranium from unused low-grade ores in classical underground mines (combined methods/systems). Such methods were applied for the mine "Selishte", experiments started in "Dospat" and "Byalata voda". Later the combined methods were applied in

"Kurilo", "Polyane" and "Druzhba-2" in Eleshnitsa, "Smolyan", "Deveti septemvri", "Zdravets".

Leaching also started in the waste heaps of the mine "Byalata voda" and "Sborishte". The method of the heap extraction was applied in Eleshnitsa and Smolyan.

Initially the combined extraction was with sulphuric acid solutions, later the "soda" scheme was applied with  $\text{Na}_2\text{CO}_3$ ,  $\text{NaHCO}_3$  and  $\text{NH}_4\text{CO}_3$ .

Near the end of the U-industry approximately 30% of the uranium concentrate was obtained by in-situ leaching with all its modifications.

During the whole period of U-industry only 4 open pit mines existed - "Dospat", "Eleshnitsa", "Deveti septemvri", "Senokos". Only the last one was functioning when U-industry began closing-down.

All U-mining sites are positioned in the south part of Bulgaria (south from the Balkan mountains) and only one site is in north Bulgaria (Smolyanovtsi).

In addition to the two processing plants, underground and open pit mines and in-situ leaching sites, there are auxiliary objects which have also a certain impact on the environment. Such auxiliary objects and sites are: loading and reloading stations, experimental sites, truck bases, geology investigation teams. These auxiliary objects due to the transport, loading and reloading of ore, or to large scale experiments have a certain environmental effect.

In Fig.1 the uranium sites in Bulgaria are shown [1], the numbers of the sites on the map correspond to the numbers in Table 1.

Table 1

## U-industry sites in Bulgaria

site, place	potential ecological impact		contaminated water		status
	close-by	required remediation	under, surface		
1 Smolyanovtsi Smolyanotsi			+		c
2 Proboynitsa Tserovo			+		c
3 Kurilo Kutina			+	+	io
4 Seslavtsi Seslavtsi	+		+	+	cl
Buhovo plant	+	+		+	c
Buhovo,Yana, Gorni Bogrov	+	+			
5 Simitli Simitli		+		+	c
6 Senokos Senokos			+	+	io
7 Igralishte Igralishte		+		+	cl
8 Gradeshnitsa Gradeshnitsa					cl
9 Melnik Melnik					cl
10 Gabra Gabra			+		cl
11 Beli Iskar Beli Iskar					cl
12 Partizanska Polyana Kirilova polyana					cl
13 Kostenets 2 Ochusha					io
14 Beslet Selishte		+	+	+	cl
15 Druzhba Eleshnitsa	+	+	+	+	io
Polyane (?) Eleshnitsa					cl
Zvezda Eleshnitsa plant				+	io
16 Dospat Barutin			+	+	io,cl
17 Zdravets Byala cherkva				+	cl

18	Narechen			cl
	Narechen			
19	Chatroka			cl
	Lucki			
20	Smolyan	+	+	io
	Gerzovitsa			
21	Sborishte	+	+	io
	Sborishte			
22	Sliven	+		cl
	Sliven			
23	Sarnitsa			cl
	Sarnitsa			
24	Planinets			cl
	Planinets			
25	Tsarimir/	+	+	io
	Ceretelevo			
	Tsarimir			
26	Momino/	+	+	io
	Rakovski			
	Momino			
27	Belozem/	+	+	io
	Trilistnik			
	Belozem			
28	Debar	+	+	io
	Parvomai			
29	Cheshmata	+	+	cl
	Haskovo			
30	Navusen/	+	+	cl,io
	Troyan			
	Navusen			
31	Mudrets/	+	+	cl
	Vladimirovo			
	Mudrets			
32	Orlov dol	+	+	cl
	Orlov dol			
33	Chukurovo	+	+	cl
	Chukurovo			
34	Tenevo/Okop	+	+	cl
	Okop			

c - preserved

cl - closed

io - in operation (in 1990)

## 2. General description of the environmental impact of the U-industry.

In the beginning of U-mining nothing had been done for restriction of the environmental contamination. The choice of a site was determined by the presence of U-ore. The rest of the conditions - near-by villages, national reserves, possible contamination of surface and ground waters - were very often neglected for the purpose of quick start of operation and low cost.

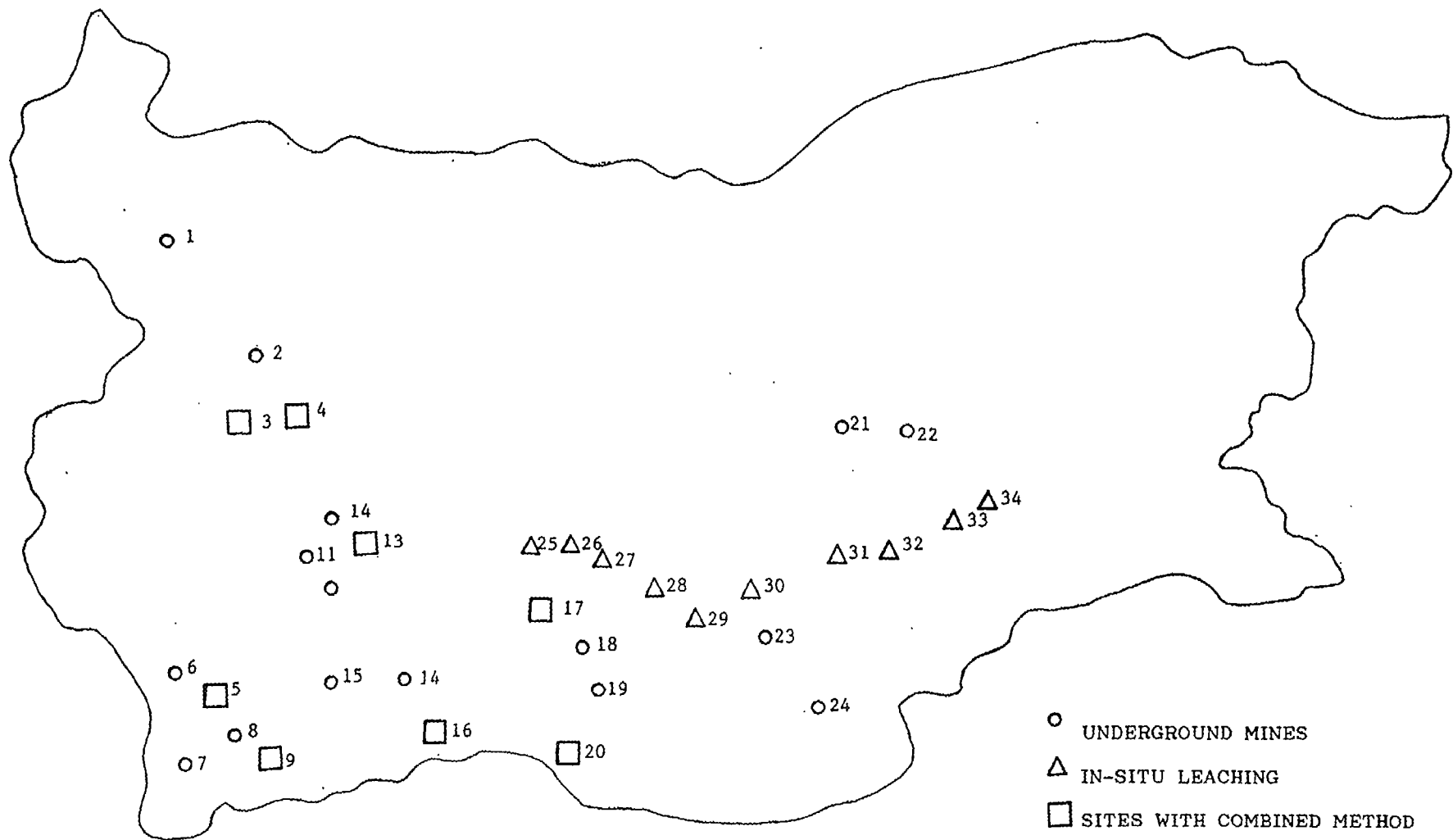


FIG. 1. URANIUM MINING SITES IN BULGARIA

The only mine which was closed since there had been a possibility for contamination of the drinking water for Sofia is "Beli Iskar" in the Rila mountain.

The secrecy of the U-industry had been another reason for the lack or inadequacy of the preventive measures and most probably that has been the case in many countries.

The contamination from the U-industry are both radioactive and chemical - acids, chemicals, oil. Most of the sites are sources of the common for the industry noise, vibrations, dust. We shall try to give a characteristic of a each group of the sites of U-industry in Bulgaria. A summary of the wastes from U-industry is presented in Table 2.

#### 2.1.Environmental conditions near processing (milling plants).

There are two uranium processing plants - Buhovo and Eleshnitsa. Both plants are situated close to living areas, but the situation of the Buhovo plant "Metalurg" and especially of its tailings ponds is very unfavorable. Due to continuous operation without tailings ponds large areas are contaminated.

Table 2  
Waste from U-mining sites

site	place	number of waste heaps	area $\times 10^3 \text{ m}^2$	volume $\times 10^3 \text{ m}^3$	quantity $\times 10^3 \text{ t}$
"Druzhba"	Eleshnitsa	43	153	1747	2900
"Vazhod"	Smolyan	12	71	743	1240
"Izgrev"	Barutin	13	14	118	200
"Septemvriitsi"	Smolyanovtsi	1	39	270	480
"9 Septemvri"	Seslavtsi	200	475	4800	7900
"Minyor"	Sliven/Sborishte	18	27	82	140
"Iskra"	Kutina	5	35	330	530
"Senokos"	Senokos	2	19	70	120
waste	Selishte	1	0.2	0.5	0.8
"Byalata voda"	Dolna banya	3	12	151	220
TOTAL		298	845	8311	13720

The Buhovo milling plant.

The tailing ponds have an area of approximately  $1.3 \times 10^6 \text{ m}^2$  with dumped waste of  $1 \times 10^7 \text{ m}^3$  and mass of approximately  $1.6 \times 10^7$  tones. The average specific activity is less than 5 kBq/kg U and more than 100 kBq/kg Ra. The specific activities show that the waste should be considered as radioactive waste (Table 3).

The potential radioactive hazard of the plants is:

-contamination of the air from the ventilation systems. The air contamination is restricted only in the site limits and it is negligible in the living zone. The different filters have almost 100% efficiency in reference to dust.

-drainage water and excess water which is released in emergency conditions when the the tailings pond is overfilled. When the plant operates normally the process is closed and there should be no release of water.

Nevertheless water is released occasionally, but the content of nuclides from the uranium-radium chain is low: U- 0.5 - 0.7 mg/l (limit 0.6mg/l) and the Ra activity is below the limit 0.15 Bq/l. The corresponding values for the water of rivers Yanestitsa, Lesnovska and Mesta are below the current limits. The drainage water however contains sulphates usually 5-6 g/l (upper limit 300

Table 3

Status of the tailings ponds of the uranium milling plants

site of the t. pond	status of the t. pond	Area		Volume		present present Bq	
		project/ real area	project/ real vol.	present $\text{m}^3$	present $\times 10^3 \text{ t}$	$\times 10^3$	$\times 10^{14}$
Metalurg	conserved	350/350	1300/1300	1300	2080	5.4	
Buhovo	operated	560/550	10000/8800	3900	6240	12.0	
Zvezda	operated	950/420	34000/10600	4800	7680	14.9	
Eleshnitsa							
total "Redki metali"		1860/1320	45300/20700	10000	16000	31.0	



mg/l) and also some metals - Cu, Zn, Fe, Mo, Cd, Mn. Only the concentration of the last one (Mn) is above the limit.

-resuspension of dust from filled and dried tailings ponds during strong winds. An irrigation system could prevent resuspension but such system does not exist for the Buhovo tailings pond. For the case of Buhovo due to broken fences cattle use the older tailings pond as a pasture ground.

-radon emanation from milled rock. The emanation does not increase the normal radon concentration in Yana village which has been proven by numerous measurements by different groups.

A special concern is the large contaminated with Ra area. At some points the specific Ra activity is 10 kBq/kg and the exposition rate reaches 10  $\mu$ Sv/h (1000  $\mu$ R/h).

The uranium processing plant in Buhovo is approximately 15 km northeast from the end of the city of Sofia. Due to operation for more than a decade without a tailings pond approximately 1.2E6 m<sup>2</sup> had been contaminated with radium. Most of the land is agricultural land but a certain fraction is within the limits of the neighboring villages.

In the beginning, immediately after the II World War, the ore was shipped by train through the Yana railroad station to the former USSR. In 1947 a small installation for chemical concentration was built which after increasing its capacity processed all the ore mined in Bulgaria until 1968 when a second plant was built. The ore is crushed and grind, oxidants are added to the slurry and after leaching with acid and selective extraction of uranium, the slime is neutralized and by hydrotransport is dumped. Until 1958 when a tailings pond was built the wastes were dumped in Manastirsko dere (Monastery gully) above Yana village. The insoluble component precipitated in the gully and the rest of the slime drifted freely through the

Yana village and through the old river beds of rivers Buhovchitsa and Yanestitsa went to the Lesnovska river. The result is contaminated village yards in the Yana village and contaminated land approximately  $1.2 \times 10^6 \text{ m}^2$  between the villages Yana, Gorni Bogrov and Dolni Bogrov (Upper Bogrov and Lower Bogrov), on both sides of the road to the small town of Elin Pelin.

From the most contaminated land  $1.183 \times 10^6 \text{ m}^2$  were paid and approximately  $2.8 \times 10^6 \text{ m}^2$  were afforrested. Later the fences were broken and at the present moment it is very hard to say their former places.

The measurements in the middle of the 50-ies identified some hot spots with exposure rate of  $5 \text{ } \mu\text{Sv/h}$  reaching at some places  $10 \text{ } \mu\text{Sv/h}$ . The long term monitoring has shown that due to migration the exposure rate has decreased roughly by 30%.

The map in Fig.2 shows the result of airborne gamma-spectroscopy with a 50 l NaI(Tl) (L.Kerbelov\*) in the Ra-window. Detailed information can be obtained from the authors

The local hot spots are marked with black dots and the absolute maximum with a dot and a cross. The main concern is the contaminated land within the limits of the Yana village and the land which is used for growing vegetables and other food plants for the people and the cattle. A certain portion of the contaminated land is overlapped by the tailings pond and the waste area of the nearby steel plant "Kremikovtsi".

The uranium content in soil is in average 10 times and for the most extreme case 40 times above the average content of uranium in soil. For the private yards the uranium content is comparable with the natural content in soil.

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Airborne Geophysics service,  
Geology and Geophysics service, tel.44-17-39  
Sitnyakovo str. No.23, Sofia

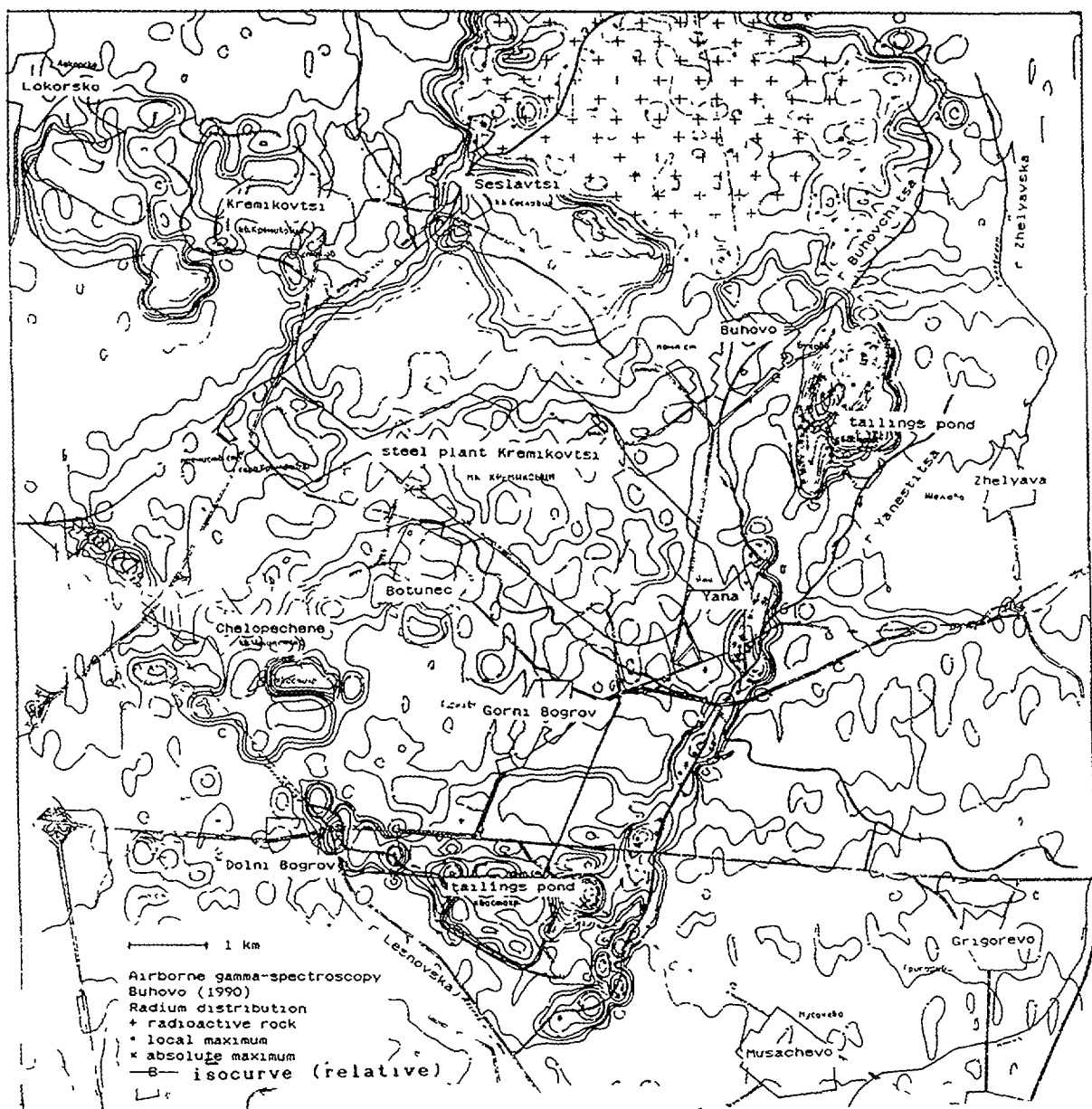


FIG. 2.

The uranium content of the yards is in average 20 times less than the content of uranium in contaminated soil, but the ratio for the radium activity is approximately 6 for sediments and 3 for soil (Table 4).

The main radiation hazard is the intake of radium with agricultural food and with water either directly food - man or indirectly fodder - animals - man. Both chains are of importance and the intake is comparable.

The measurements for uranium and radium in waters show that due to sorption and filtration the content and activities are very low and are close to the accepted limit for drinking water even close to the tailings pond wall. Deep sampling (up to 100 m) before a number of years and recently show that uranium varies within 1 - 5 micrograms per liter (limit 600  $\mu\text{g/l}$ ) while radium in 6 out of 8 deep sampling wells is above the limit of 150 mBq/l, but only in three of the closest wells the value is approximately 5 times that limit.

The conclusion for the water is that radium and uranium intake with water is not a dominating radiation hazard.

Another radiation hazard is the inhalation of radon and radon daughters. The inhalation of dust which is partially radioactive has much less importance since the specific activity of the soil and ore is low. A source term for dust is the dried coast line of

Table 4

Uranium and radium activity in soil, grass and vegetables

sample	number of samples U/Ra	Activity Bq/kg	
		U min-max average	Ra min-max average
agricultural land			
soil	94/72	20-23000 642	50-11000 2184
grass	124/119	1- 200 36	1- 3100 144
fodder	48/37	1- 960 29	1- 1850 42
grain	16/15	1- 230 21	4- 1100 34
private yards			
soil	10/10	20- 77 28	9- 107 68
grass	5/5	1- 2 1.3	9- 107 68
vegetables, fruits	48/36	0- 2 0.3	4- 60 26
grain			
river Lesnovska	106/106	1- 1980 70	0- 4500 373
grass	30/27	0- 175 2.7	0- 1520 21

the tailings pond when the wind blows towards Yana but the distance makes that possible hazard negligible.

The measured maximal potential alpha energy for the Yana village is 18-440 MeV/l the upper value of which corresponds to approximately 30% of the limit for the population. For the villages Gorni Bogrov and Dolni Bogrov the radon contamination of the air is not different from other places in Bulgaria.

The Eleshnitsa milling plant.

The main hazard for the milling plant in Eleshnitsa is the radon inhalation. In the vicinity of this village large areas are contaminated with wastes from uranium mining and milling. The village is situated in a valley, surrounded on all sides with hills, so the natural ventilation is poor and thermal inversions occur frequently. In addition the region as a whole is with enhanced natural radioactivity and uranium ores were recognized almost on the earth surfaces. One former open pit uranium mine (still not restored and rehabilitated) is placed in very short distance (200-300m) from the village and in the same valley. These factors are a serious reason to expect problems, associated with radon.

The values of EEC (equilibrium equivalent concentration) cover more than one order of magnitude: from background ( $< 10 \text{ Bq.m}^{-3}$ ) to about  $370 \text{ Bq.m}^{-3}$ . The highest values were found in the interval  $0-10^h$  (average EEC outdoors =  $107 \text{ Bq.m}^{-3}$ ). In the rest part of the day only about 10% of values are above  $37 \text{ Bq.m}^{-3}$  (average EEC outdoors =  $25 \text{ Bq.m}^{-3}$ ). The diurnal averaged EEC is  $59 \text{ Bq.m}^{-3}$ . The equilibrium factor F is on average  $F = 0.8$  in  $10-24^h$  and  $F = 0.92$  in  $0-10^h$  (the equilibrium factor was estimated assuming equilibrium between  $^{222}\text{Rn}$  and  $^{218}\text{Po}$ ). Especially in the subinterval  $6-10^h$  the average equilibrium is  $F = 1.0$ . In this subinterval sometimes states above equilibrium ( $F > 1$ ) were

observed - a phenomena still not fully understood. The concentrations of Rn progeny close to open mining adits were up to  $EEC = 5300 \text{ Bq.m}^{-3}$ . They decrease quickly in short distance (about 30 times in distance of 20m) and are not supposed to be a main source of outdoor radon. However the risk for persons which are being often in their vicinity could be significant (some of the adits are almost in the village). Therefore isolation of their outlets was recommended as a priority task.

About half of outdoor measurements were conducted when the central ventilation system and any mining activities were stopped for months. However no systematic difference in results has been observed. Hence, the early consideration of mines exhaust air as the main cause for higher concentrations of Rn outdoors has been rejected.

The risk estimations are based only on radon exposure outdoors and do not account for radon indoors.

The annual exposure was estimated to be 0.82 WLM. Hence the cumulative exposure for a 50 year old person is about 40 WLM - a value which approaches some occupational exposures of miners.

## 2.2. Environmental conditions near the underground mines.

As a rule the underground mines are in mountain or semi-mountain regions far from villages and towns. Exceptions are the mines "Druzhba 1,2" in Eleshnitsa and "9 Septemvri" near Seslavtsi which are close to the living areas.

The environmental impact of the underground mines is determined by:

- a) the water from the mines.

The water contains natural radionuclides, slurry, chemical agents and heavy metals in case combined methods (in situ-leaching) had been applied, oil also.

The main radiation hazard is due to radium and much less to uranium, thorium and polonium.

The average uranium content of the surface water is above the upper limit of 0.6 mg/l and the radium content is between 50 and 800 mBq/l (the accepted limit is 150 mBq/l). The radium content quickly decreases below the limit of 150 mBq/l in the tailings ponds.

-b)the mined waste rock deposited on the surface. The rock had been mined during investigations and also during excavations around the ore layer.

The mined rocks contain all daughter products of uranium in equilibrium. Some rocks are close to the accepted ore concentration limit -0.025 - 0.030 %. The presence of radium is a reason for the contamination of the air close to the rock heaps, but it normalizes at 0.5 - 1 km from it. The air and water erosion lead to spreading of natural radionuclides in the hydro net thus creating an areal around the mines.

At present the waste heaps are almost 300 spread over  $8.3 \times 10^5$  m<sup>2</sup>. The total waste is approximately  $13 \times 10^6$  m<sup>3</sup>, out of which the uranium containing waste is  $8 \times 10^6$  with average uranium content of 0.025% and total activity of  $8 \times 10^{13}$  Bq.

The specific mass activity is estimated to be 6.3 kBq/kg which is an argument to consider those wastes as non-radioactive but with increased level of natural radioactivity.

-c)the ventilation systems.

The radon content in the ventilated air is 30 - 800 Bq/m<sup>3</sup> at potential alpha energy of  $(10 - 50) \times 10^5$  MeV/l for the different mines. The maximal recorded values for the most close villages Seslavtsi and Eleshnitsa are 920 and 6000 MeV/l. The daily average value in Eleshnitsa is below the upper limit of 1330 MeV/l, but in certain periods of temperature inversions the value

of the potential alpha energy is close to the limit [3]. The ventilation system also releases inert and radioactive dust and toxic (explosive) gases which are diluted very quickly.

### 2.3. Environmental conditions near sites of in-situ leaching.

The ore is 120 - 200 m thick and is spread over several hundred millions of  $m^2$ . The paid land for in-situ leaching is  $1.6 \times 10^7 m^2$ ,  $2 \times 10^6 m^2$  from which are non-agricultural land and  $3 \times 10^6 m^2$  are forest. The land which has been actually used for in situ-leaching is  $6 \times 10^6 m^2$  which include the mining land and the sorption installations. The input and the output channels are spread in a net with 10 to 30 m between them, and hence all tubes and the pumping facilities total to some  $0.5-0.6 \times 10^6 m^2$  which could be eventually contaminated.

The main problem is that the greater part of that land is agricultural and since the land is expected to be returned to the owners the problem for remediation is very urgent. Not more than  $2 \times 10^6 m^2$  is restored.

For each site the quantity of solution which is processed daily is between 4 and  $30 \times 10^3 m^3$  with average content of uranium between 5 and 20 mg/l. The content of salts varies within 15-20 g/l from which 10-12 g/l are sulphates and the rest are other salts and micro quantities of heavy metals and rare earths.

There is a potential hazard for contamination of the surface of the land and also water - surface and underground. There is no air-contamination.

The ion exchange resins are enriched to 40-50 g/kg or 1000-1300 kBq/kg and that can be a radiation hazard since if they are dumped they will exceed the limit of  $7 \times 10^3 Bq/kg$  for alpha-emitters.



The radiation hazard for the sites of in-situ leaching is not the major problem since the extracted solution is enriched to uranium and the radium remains in the ore (more than 97%). The result is that the exposure rate very rarely exceeds the doubled natural background. At certain spots of accidental release of the solution the soil is enriched in reference to uranium approximately 10 times and in reference to radium 2- 3 times, or U - 600-800 Bq/kg, and Ra - only occasionally above 100 - 150 Bq/kg.

The most serious problem is the radioactive contamination of the deep underground water which reaches the region of the ore. The uranium content in this case reaches 20-30 mg/l and that of radium 1 - 2 Bq/l. The same problem exists for the shallow underground water in the case of accidental release and tube defects. For the shallow water the recorded maximal concentrations are 3-4 mg/l U and 0.5 Bq/l Ra in spite of the dilution.

In general during in-situ leaching due to evaporation and hydrotransport the level of the underground water decreases which leads to a certain restriction of the contamination. After the leaching is stopped the contaminated water migrates. The water is slightly acidic and may be if it reacts with the carbonates a certain hydrochemical barrier could be formed. That is not a clear problem and certain investigations are necessary. A special group of experts has been formed to study the problem.

Some of the sites (Bolyarovo, Tenevo, Okop) are close to places where water is used for drinking which could lead to decrease of the quality of the water.

After the closing of the U-mining a special order prescribed the continuation of the solution recycling without adding acid. For most of the sites the flow decreased and the salts started to deposit to the tubes and the filters of the sorption columns. The salts are with increased radioactivity and a possible

contamination hazard could be expected when the tubes will have to be disassembled. When the problem was identified it was ordered to stop the recirculation.

Only one site (Selishte) had not that problem.

The technology for remediation of the sites of in-situ leaching is not at present finished.

The radiation hazard is less important than the chemical and acid contamination (which should not happen), sulphates and some heavy metals. It could be concluded that the main ecological hazard is due to the sulphuric acid the quantity of which is estimated to approximately 2.5 millions tones ( $2.5 \times 10^6$ ). This problem is extremely important especially when the U-industry is closed.

A case of special interest are the areas in the Gorna Trakiiska nizina (Upper Tracian plain) near Plovdiv where the soil is very good for agriculture and the deep water is considered to be an important reserve.

### 3. The Bay of Vromos case.

The case of the Bay of Vromos is an example of radioactive contamination which is not connected to U-mining. The contamination is connected with mining of copper ore with increased content of U.

During the period of 1954 - 1977 in the beginning partially and later after 1968 the total waste from the polymetallic mine "Rosen" (Burgas copper mines), which is approximately 30 km east from the town of Burgas on the Black sea, was dumped in the sea. The coast line between Atia and the village of Chernomorets moved approximately 150 m in the sea.

The slime is predominantly rocks and 2-6% polymetallic ore. Uranium is also included in the content. The total waste is

estimated to be approximately 8 000 000 tones, from which on the coast the waste is approximately 1300 m long, width 120 m and thickness 2 - 3 m.

The exposure rate is 1.2-1.5  $\mu\text{Sv/h}$  in the central part and 0.3-0.4  $\mu\text{Sv/h}$  at the end of the contaminated area. The natural background in the nearby beaches is 0.08-0.2  $\mu\text{Sv/h}$ .

After the dumping was stopped (1978), the coast line slowly is returning to its previous position.

In 1985 an experimental project was performed - approximately 10 000 tones of ore were processed and the metal was extracted. The exposure rate over the processed areas reduced 4-5 times and is within the limits of 0.3-0.4  $\mu\text{Sv/h}$ .

The secondary processing proved to be economically profitable.

A interesting phenomenon has been observed in the last several years. The coast line which is sand is slowly enriched to metal, or the metal ore is slowly returned by the sea.

## Conclusion

The main problems of the contamination U-industry at present are due to its very sudden closure and the lack of needed funding for investigations and remediation procedures. Those problems should be solved as quickly as possible although the contaminated areas are not great and the number of population living close to them is not big.

The radionuclides can migrate through underground and surface water and therefore purification facilities where possible should be built.

The tailings ponds and waste heaps should be stabilized and rehabilitated which will reduce or prevent water migration and radon exhalation.

The intake of radionuclides from sites of in-situ leaching can be reduced by neutralizing and remediation of the sites.

All rehabilitation measures can be realized with the labor of the workers of the former U-industry which will reduce the social tension.

#### **REFERENCE**

[1] S.D.Simov, I.B.Bojkov, Case Histories and New Areas for Uranium Exploration in Bulgaria, in New Developments in Uranium Expolration, Resources, Production and Demand: Proc. of a Technical Committee Meeting Jointly Organized by the International Atomic Energy Agency and the Nuclear Energy Agency of the OECD, Vienna, 26-29 August, 1991.

# IDENTIFICATION AND RADIOLOGICAL CHARACTERIZATION OF CONTAMINATED SITES IN CANADA

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## Abstract

Several instances of localized site contamination as a consequence of past practices involving radioactive materials (in particular, the mining and processing of radioactive ores) have occurred in Canada. In general, these situations evolved over several decades, and originated with historical practices which predate the current climate of environmental consciousness and commensurate regulatory controls. Although such practices included storage, handling or disposal in accordance with methods and standards considered acceptable at the time, subsequent developments, including observed impacts and increased societal expectations, have rendered many of these standards and methods unacceptable to current generations. Consequently, since the early 1970's, Canadian government and industry have become increasingly active in initiatives to identify and remediate contaminated sites, and to prevent similar recurrences. This paper describes specific instances of contamination, past and present. Relevant details such as histories of occurrences, descriptions of contaminated sites and contaminants, and descriptions of pending or completed remedial programs are provided.

## 1. INTRODUCTION

Radioactive contamination of public and private lands within Canada has been primarily, but not exclusively, associated with early mining, processing, transport and disposal of radioactive ores and their by-products. In particular, events associated with radium mining, refining, and use in Canada resulted in contamination incidents dating back to the 1930's which continue to be addressed today. In addition, the initial boom-bust cycle of uranium mining and refining which followed the collapse of the radium industry in Canada contributed incidents of localized contamination as a consequence of inadequate waste storage and disposal practices, spill incidents, and abandonment without attention to site decommissioning, reclamation or post-closure safeguards and monitoring. With the resurgence in the 1970's of the domestic uranium mining and processing industry, new or re-activated sites and operations were subjected to increasing regulatory scrutiny and controls in the interests of health, safety and protection of the environment.

The birth and growth of the uranium industry in Canada paralleled the world wide development of nuclear weapons, the increased usage of nuclear materials and devices in industry, medicine and research, and the development and growth of the nuclear power program. Nuclear research activities in Canada led to the early establishment of a large complex near Chalk River, Ontario (currently, the Chalk River Laboratories), and the establishment later of the Whiteshell Nuclear Research Establishment (WNRE) near Pinawa, Manitoba. The CANDU reactor designs developed at Chalk River formed the basis for subsequent CANDU units which currently provide all of Canada's nuclear-generated power.

The health, safety and environmental impacts of the nuclear industry are currently regulated by the Atomic Energy Control Board (AECB), in co-operation with other federal and provincial jurisdictions. The AECB, a federal agency, regulates pursuant to the Atomic Energy Control Act and Regulations, with a mandate "to ensure that the use of nuclear energy in Canada does not pose undue risk to health, safety, security and the environment". This responsibility is discharged through a comprehensive licensing, inspection, and compliance system.

The role and activities of the AECB with respect to waste management and environmental protection are described in detail in reference (1) which is available from the AECB Office of Public Information: P.O. Box 1046, Ottawa, Ontario, Canada K1P 5S9.

## 2. HISTORICAL OVERVIEW

The dawning of the "radioactive age" in Canada corresponded to the mining, refining and use in medicine and industry of the radioactive element, radium-226.

In 1930, "pitchblende" was discovered at a remote location on the shores of Great Bear Lake in the Northwest Territories of Canada. In 1932, a private company, Eldorado Gold Mines, developed the Port Radium mine at Great Bear Lake, and established a complementary radium extraction refinery at Port Hope, Ontario. (The location of the refinery was chosen on economic grounds...each ton of pitchblende concentrate required several tons of chemicals for treatment, and consequently it was cheaper to transfer the ore to the manufacturing source of the required chemicals.) Port Radium concentrates were initially transported south by barges (and later by air) to a rail terminus near the present site of Fort MacMurray, Alberta, and forwarded to Port Hope via railroad.

The Port Hope refinery extracted radium from Port Radium mine and mill concentrates from 1932-1940. The radium obtained was used primarily for cancer treatment, and in the manufacture of luminous paint. With the exception of the use of small quantities of uranium oxide in ceramic glazes, the refinery by-products, consisting of the unwanted pitchblende and the associated uranium, nickel, cobalt and arsenic elements were discarded on company property throughout the town of Port Hope. Control standards for industrial wastes were virtually non-existent during these years, and no special precautions were taken to protect the wastes from subsequent dispersion by the elements, or to limit public access. In addition, radioactive effluents and wastes were periodically discharged to the Port Hope harbour of Lake Ontario. The radium industry declined steadily subsequent to 1937, and the Port Radium mine closed in 1940. The Port Hope refinery continued to operate by processing stockpiled feedstock.

In 1941, the Manhattan Project created demands for north-american sources of uranium, and revived interest in the Port Radium mine and the Port Hope refinery. The mine was re-opened in 1942, the Port Hope refinery was modified to permit extraction of uranium, and residues from earlier radium processing operations were recycled to recover uranium. In 1944, the Canadian Government expropriated Eldorado Gold Mines Ltd. and thus became sole owner of the Port Radium and Port Hope operations.

Following the second world war, demand for radium was supplanted by the production of radioisotopes in the uranium-fuelled nuclear reactors that were built thereafter. Consequently, subsequent to 1951, the uranium industry in Canada burgeoned, with numerous new uranium mining and milling operations springing up in Saskatchewan, the Northwest Territories, and Ontario. Through these years, Eldorado continued to operate as a federal Crown Corporation, concentrating increasingly on uranium exploration, mining, and milling at locations in the Lake Athabasca region of northern Saskatchewan and the Northwest Territories. With the subsequent construction of uranium-concentrate-producing mills near mining sites, similar operations ceased at Port Hope, but were supplanted by other uranium or nuclear-fuel processing activities. These continued to produce reduced volumes of on-going wastes requiring management.

The initial cycle of intense activity in the Canadian uranium industry diminished abruptly in the late 1950s-early 1960s. Many mines and mills closed down, and were permanently or temporarily abandoned during this period. A skeleton industry continued at Port Hope (refining) and Elliot Lake (mining and milling) in Ontario, and at Eldorado's Beaverlodge Operations (mining and milling) near Uranium City, Saskatchewan.

The development and growth of the nuclear power program in the late '60s - early '70s revived the demand for uranium fuel, and led to a resurgence in the uranium mining and processing industry. This demand has continued, to a greater or lesser extent, to present.

During the above evolution of, and fluctuations in, demand for radium and uranium products, the Eldorado refining operations in Port Hope evolved and changed accordingly. Wastes representative of numerous processes, as well as the periodic refittings of the plants, were produced. These were initially stored or buried at various locations in or around the Town of Port Hope. The first residues produced (of those not discharged to the Lake or harbour) were stored at the plant site from 1933 to 1939. From 1939 to 1944, wastes were deposited in the Lakeshore Residue Area near Lake Ontario, adjacent to the Eldorado refinery. Other wastes were placed in temporary storage at various locations within the town. The wastes stored at the plant site were reprocessed from 1945 to 1948, and during this period the tailings from the refinery were deposited at the Monkey Mountain Residue Area in Port Hope. In 1949, the Welcome Residue Area was opened at a location to the west of Port Hope, and received refinery wastes until 1954. From 1948 to 1974, the Pidgeon Hill Storage Area adjoining the municipal garbage dump was used for the surface storage of contaminated equipment and radium waste, and for occasional incineration of combustible waste. Over the period 1957 to 1959, radioactive wastes were recovered from the Lakeshore site, and sold to a United States company for recovery of heavy metals such as cobalt and nickel. Similarly, in 1959, residue from Monkey Mountain was sold to a Canadian company for the recovery of cobalt. The remaining wastes from Monkey Mountain were transferred to the Port Granby Residue Area which opened to the west of Port Hope in 1955 and operated until 1988 (at which time it was closed to further receipt of wastes in accordance with a directive from the Atomic Energy Control Board). Currently, most wastes from the Port Hope plant are recycled, and the small volume not recycled is being held in closed storage pending the establishment of a national low-level waste repository.

In addition to its application in medicine, radium refined at Port Hope was used domestically in the manufacture of luminous paints which were subsequently applied to instrument dials, such as those of aircraft, boats, and watches. These operations have been linked, directly and indirectly, to several contamination situations; most notably, large scale, low-level radioactive contamination of residential and industrial land in Scarborough, Ontario, part of Metropolitan Toronto.

Instances of site contamination, both historical and current, resulting from radium and uranium processing and use, nuclear research, and non-nuclear industrial activity are described in the following chapter. The locations of the sites are indicated on Figure 1.

### 3. CONTAMINATED SITES

#### 3.1. Town of Port Hope

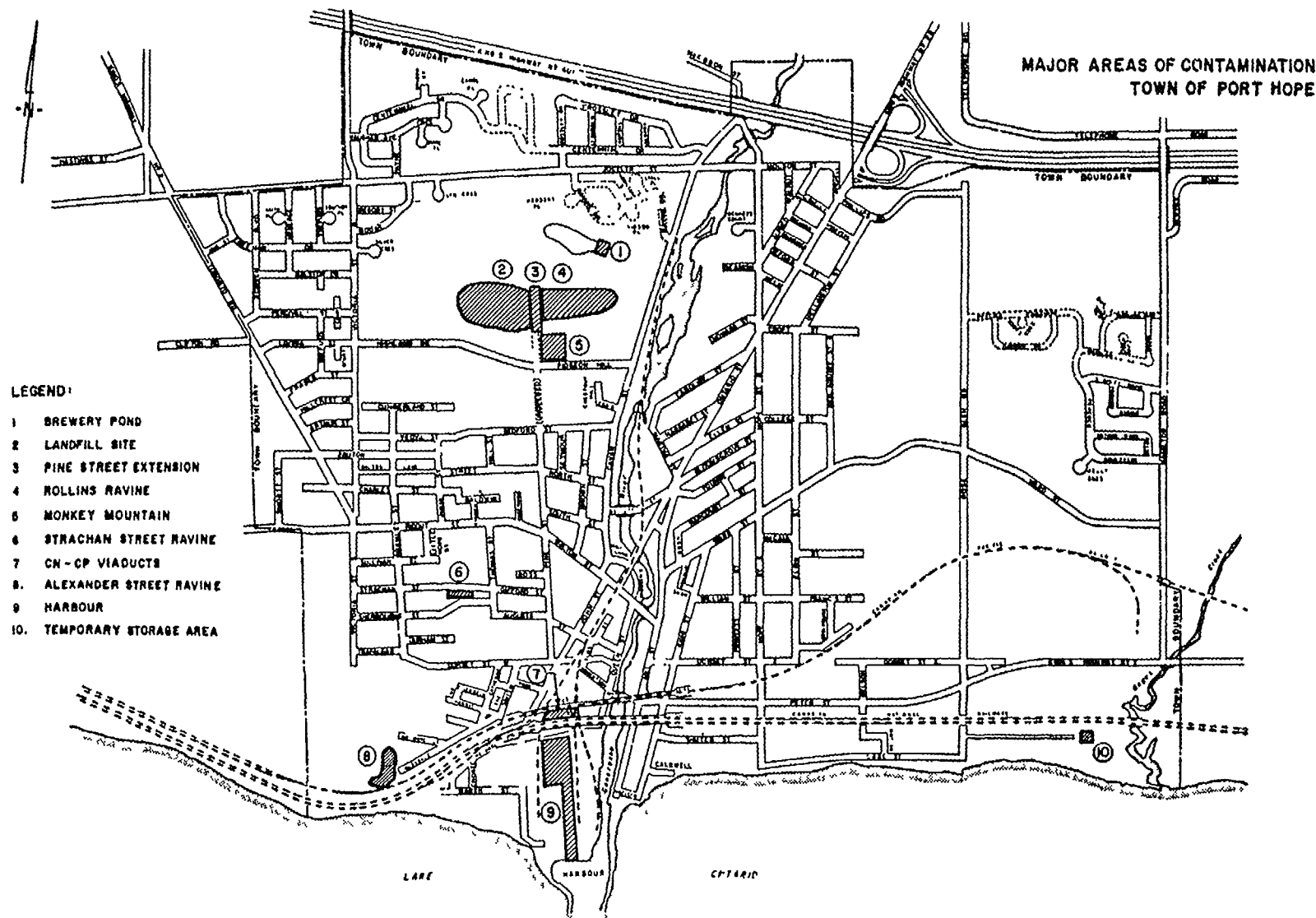
Contamination of large areas of the Town of Port Hope with low-level radioactivity associated with uranium-radium series nuclides occurred gradually and progressively over a number of years. These occurrences resulted from historical practices whereby wastes were handled and disposed of with little attention to provisions other than basic physical containment, and with no preventative measures to address potential leaching or transport by wind and water. The history of events that gave rise to widespread contamination within the Port Hope community is complex, and, in retrospect, a matter of some speculation. However, some of the contributing factors and practices are known:

- (a) Spillage of radioactive ore or residues during shipment by road to disposal areas, or during handling at railroad sidings;
- (b) Temporary and longer-term storage at various location without measures to prevent water infiltration, leaching of wastes and contaminants, or overland migration via wind or water; and



FIG. 1. IDENTIFICATION AND RADIOLOGICAL CHARACTERIZATION OF CONTAMINATED SITES IN CANADA





**FIG. 2 IDENTIFICATION AND RADIOLOGICAL CHARACTERIZATION  
OF CONTAMINATED SITES IN CANADA (cont'd)**

- (c) Removal by the public of large quantities of contaminated rubble, fill, and other materials originating from operation or refitting of the Port Hope plant(s).

It is reported that town residents burned contaminated scrap lumber in their woodburning stoves and furnaces of the day, and routinely discarded the resulting "radioactive" ashes on residential lots, vacant land, etc. Fill, rubble and contaminated building materials originating from the Eldorado plants were used in residential construction. When upsets in refinery process resulted during the early phases of process development, large quantities of wastes were often discharged directly to the harbour.

Over the history of the Port Hope refineries, a number of problems were recognized with the prevailing disposal practices, and some early improvements were implemented by the company. For example, in 1956, as a consequence of the contamination of a local creek and surrounding lands by surface runoff from the Welcome site, runoff retention ponds were added to the Welcome site. Various initiatives to relocate and consolidate wastes, and to improve waste storage practices, were undertaken. Exposed wastes at residue areas were covered to reduce wind and water erosion, and surface radiation fields.

In 1975, the Atomic Energy Control Board (AECB) assumed a pro-active role in licensing and regulation of the residue areas at Port Granby and Welcome, and this gave added impetus to improvement of the sites. In subsequent years, major improvements were implemented at the sites. These modifications and on-going maintenance have, to a large extent, arrested further migrations, or significant releases, of radiation or contaminants to the surrounding areas. Ground water interceptor systems, surface water collection and control works, and effluent treatment and discharge systems have been constructed and are maintained and operated in compliance with regulatory requirements. Waste burial areas have been covered with clean soil, recontoured, and vegetated. Inadvertent access to the sites is prevented by perimeter security fences. Use of the sites for additional disposals has been discontinued, and the sites are being actively maintained and monitored by the current owner pending eventual decommissioning. Decommissioning of the sites is dependent on the ongoing efforts of Task Force appointed by the federal government to locate a volunteer community within Ontario willing to host a low-level radioactive waste disposal facility.

In 1975, initial media coverage of the discovery in Port Hope of elevated radioactivity on local streets led to significant public and political concern over the potential impacts within the town and elsewhere. Eldorado Nuclear Limited responded to this concern by investigations which included searches of plant records, interviews of long-term employees, and solicitations of public assistance through the local media. The results exposed a significant number of cases of contamination, including that at a local school. The increasing number of apparently unrelated cases prompted the AECB and the Ontario Ministry of Health to implement a more systematic investigation.

In February, 1976, the Federal-Provincial Task Force on Radioactivity was formed, with the AECB as lead agency, to co-ordinate the investigation of private and public premises in Canada in which radioactive waste was known or suspected to exist and to expedite the necessary remedial actions. In co-operation with the provincial Ministry of Health, the AECB established an information office in Port Hope, and initiated further investigations.

A radiation survey of all roads in the community was carried out using a sensitive gamma detection instrument mounted on a vehicle. When abnormal radiation levels were encountered during the road survey, follow-up gamma and radon-radon daughter surveys of nearby buildings and properties were instituted. The results of these initial surveys confirmed the presence of elevated gamma or interior radon-radon daughter levels in several residences, suggesting a significant, wide-spread contamination problem.

The Task Force established clean-up criteria for radon-daughter levels inside buildings, and hired program managers to complete the required surveys and remedial work. Over 4000 Port Hope

properties were eventually surveyed. The clean-up proceeded on a priority basis, and the properties with the highest levels of radioactivity were dealt with first. Ultimately, about 400 properties were subjected to remedial work, and resurveyed to confirm the effectiveness of the actions taken. The remedial work completed varied according to site specific factors, and practical considerations. Where contaminated materials, fill, etc were identified as the source of the problem, and proved accessible at reasonable cost, the materials were removed and disposed of as radioactive waste. In other situations involving elevated in-door concentrations of radon-radon daughters, the buildings were subjected to various types of remedial activities to reduce levels. These actions were designed and implemented to ensure compliance with specific remedial criteria, rather than detection and removal of all known or suspected contamination.

The Federal-Provincial Task Force soon recognized that an additional repository to accommodate contaminated soil and building materials was required in order that the large-scale clean-up of vacant properties within the Town of Port Hope might proceed. Ultimately, access to such a facility was not obtained, and the clean-up of nine major areas of contamination within the town was deferred on anticipation of the timely siting and construction of a national low-level radioactive waste disposal facility. This postponement followed a successful legal challenge to the authority of the Atomic Energy Control Board (AECB) to act as program manager of clean-ups within the communities of Port Hope and Scarborough, Ontario.

As a consequence of the above challenge, the federal government subsequently formed the Low-Level Radioactive Waste Management Office (LLRWMO) in 1982 to discharge the federal government's responsibility for "historic wastes". "Historic wastes" are defined as certain radioactive wastes for which the original producer can no longer be considered responsible, and for which the federal government consequently recognizes a residual responsibility. Historic wastes thus include those scattered around the Town of Port Hope, as well as others linked with contamination incidents described in the following sections.

To date, the LLRWMO has been hampered in its efforts to finalize clean-up and disposal of historic wastes in general, and those in Port Hope in particular, by the obstacle encountered earlier by the AECB and the Federal-Provincial Task Force On Radioactivity - the absence of a national low level radioactive disposal facility. This lack has also hindered decommissioning (in accordance with a directive issued by the AECB in 1980) of the Port Granby and Welcome waste storage facilities. In 1986, efforts by Cameco (then owner and operator of the Port Hope plants and the Welcome and Port Granby waste storage areas) to site a repository to accommodate radioactive wastes from Port Granby, Welcome and the Town of Port Hope, encountered significant public opposition at the site selection stage. The related controversy received national media coverage, and the federal government cancelled environmental hearings for the project, and appointed a Task Force to develop "a non-confrontational" siting process for potential implementation in lieu of the traditional approach.

In its subsequent report to government, the Siting Process Task Force recommended adoption of a "voluntary" siting process. This process was accepted in principle by the federal government, and a Siting Task Force was appointed to identify "volunteer communities" within the province of Ontario which might be willing and able to "host" an engineered facility for disposal of some 900,000 cubic metres of low-level radioactive wastes from Port Granby, Welcome, and the Town of Port Hope. Fourteen communities expressed an initial interest in the voluntary process and its objectives. Currently (in 1993), all but 2 of these volunteer host communities have formally withdrawn from the process, and negotiations continue with those remaining.

Since its formation, the LLRWMO has undertaken several investigative or remedial measures to improve the interim management of those wastes remaining in Port Hope. Additional detailed field investigations were completed in 1986 in order to augment existing technical data on waste occurrences and their impacts. The results obtained were used as input to the design of interim waste clean-up and consolidation programs subsequently implemented at the Strachan Ravine and Monkey Mountain-Rollins Ravine-Brewery Pond sites. On the basis of investigations to date, it appears that

some 210,000 cubic metres of wastes within the town of Port Hope may require remediation. This includes about 85,000 cubic metres of contaminated sediments in the harbour, 50,000 cubic metres at the east end of the landfill, 30,000 cubic metres at hundreds of individual small-scale private properties, and about 45,000 cubic metres at other major sites.

The details and results of the 1986 investigations, as summarized in the SENES report, Environmental Assessment of the Port Hope Remedial Program (1987) are described in the following sections.

#### *3.1.1. Monkey Mountain Residue Area*

The original Monkey Mountain Residue Area was a fenced compound located at the corners of Highland Drive and Pine Street in Port Hope. Contaminated material, consisting of residues and sandy soil, extended over some 5000 square metres of the site. The depth of contamination ranged from near-surface to depths of 3.5 m. Contact gamma levels in the contaminated zones ranged from less than 10 microroentgens per hour (2.58 nanocoulombs per kilogram per hour) to approximately 400 microroentgens per hour (103 nanocoulombs per kilogram per hour), with an average of 14 microroentgens per hour (3.61 nanocoulombs per kilogram per hour). The closest residences were located within 10 to 20 metres of the site. Sub-surface investigations in 1986 included 3 geotechnical boreholes to depth, 19 shallow boreholes, and 6 test pits. Ground water samples were collected from the 3 geo-technical holes and from 3 wells installed during previous investigations. Four of the wells were located within site boundaries while the other 2 wells sampled were located in the ravine to the south of the site. Samples of surface water were collected in the ravine to the south of the site, and from the body and outlet of one of the ponds. Seventeen of the 19 boreholes drilled in 1986 indicated radium concentrations less than 0.3 becquerels per gram (Bq/g). Analyses of a composite sample taken from more heavily contaminated areas indicated radium concentrations of 5.6 Bq/g, a uranium concentration of 15 parts-per-million (ppm), and an arsenic level of 112 ppm. The average concentration of the material requiring removal was estimated to be 1.7 Bq/g. Elevated uranium levels (3 to 140 times the "Maximum Acceptable Concentration", or "MAC", of uranium in drinking water) and arsenic (5 to 130 times the MAC) were measured in groundwater collected from 4 of the 6 holes sampled. Surface water samples collected in the south ravine also exhibited elevated levels of uranium and arsenic. The volume of contamination requiring cleanup from the Monkey Mountain site was estimated to be 6240 cubic metres.

#### *3.1.2. Pine Street Extension*

The Pine Street Extension is a 250-metre-long section of loose-surfaced roadway, of which the more northerly section adjoining Monkey Mountain, Rollins Ravine, and the Municipal Landfill contains, or overlays, contamination. This contamination consists of a mixture of sand, gravel, bricks, glass, wood, ash, cinders and other types of refuse used in the construction of the roadbed. Contamination occurs near surface, and at depths up to 4.5 metres. Contact gamma levels range from 8 to 2000 microroentgens per hour (2.06 to 516 nanocoulombs per kilogram per hour), with an estimated average of 32 microroentgens per hour (8.26 nanocoulombs per kilogram per hour). Twenty-seven samples collected from the highest areas of contamination, and the results of gamma-logging of boreholes, indicated concentrations ranging from 0.9 Bq/g to 1040 Bq/g of radium-226. The volume of contaminated material requiring removal was estimated to be approximately 5000 cubic metres at an average radium concentration estimated at tens of becquerels per gram.

#### *3.1.3. Rollins Ravine Area*

Rollins Ravine originally comprised a wooded ravine and open fields (situated north of Monkey Mountain) of which approximately 16,500 square metres were contaminated. This contamination included sands, ashes, glasses, pieces of metal, ceramics, bricks and other identified materials. Although the depth of contamination ranged from near-surface to about 3.5 metres below grade, most of the contamination appeared to be located within the top 0.5 metres. Gamma levels

measured at contact varied from background to over 5000 microroentgens per hour (1.29 microcuries per kilogram per hour), with an average of 17 microroentgens per hour (4.39 nanocoulombs per kilogram per hour).

The 1986 investigations included the drilling of 2 geotechnical and 46 shallow boreholes. Groundwater samples were collected from a geo-technical hole located at the bottom of the ravine and from 2 boreholes installed during previous investigations. On the basis of gamma survey results, the average radium concentration of the material to be removed was estimated at 1.9 Bq/g. A composite sample was collected from the more heavily contaminated areas of the ravine, and from the earth-fill dam at the outlet of Brewery Pond. The corresponding analyses yielded results of 174 Bq/g radium, 1500 ppm uranium, and 200 ppm arsenic. Analyses of the ground water samples indicated elevated concentrations of uranium (16-225 times the federal MAC for drinking water) at locations corresponding to the maximum surface and sub-surface gamma levels.

The volume of contaminated material requiring removal was estimated to be 5900 cubic metres.

#### *3.1.4. Brewery Pond Area*

Brewery Pond is a small pond, approximately 6500 square metres in area, situated to the northeast of Rollins Ravine. It originally created as a water supply pond for a brewery by construction of an earthen-fill dam across a small stream. Contamination existed in a 3000-square-metre area bounded on the east by Cavan St, and on the west by the eastern edge of Brewery Pond. Contaminated material consisted of pieces of wood, bricks, cinders, metal, glass, ash, concrete, and municipal refuse utilized in the construction of the downstream portion of the dam and in the backfilling of the old creek channel. Contamination was identified at surface and at depths of up to 6 metres. Contact gamma levels ranged from near background to over 5000 microroentgens per hour (1.29 microcuries per kilogram per hour), with an estimated average field of 17 microroentgens per hour (4.39 nanocoulombs per kilogram per hour).

In 1986, 4 sediment samples and 8 benthos samples were collected from Brewery Pond, 5 geotechnical and 31 shallow boreholes were drilled, and 8 test pits were excavated. Groundwater samples were collected from 2 of the holes east (i.e., down gradient) of the pond. Surface water samples were collected at the outlet and downstream.

The sediment samples did not indicate any contamination of the pond bottom.

#### *3.1.5. Strachan Ravine Area*

The Strachan Ravine site was originally a section of heavily-wooded, steeply-sloped ravine located at the junction of Strachan and Thomas Streets. The bulk of the contamination was located along the north bank of the ravine, with smaller pockets of contamination occurring along the north face, and contamination extending under the adjacent roadbed. Contamination within the ravine existed over some 2400 square metres of surface area, occurred on surface and at depths up to 3 metres, and consisted of silty sand, gravel, organic material, ash material, chemical residues, and scrap metal, bricks, bottles, cinders, and crockery. Contact gamma levels ranged from a few microroentgens per hour (approximately 1 nanocoulomb per kilogram per hour) to more than 5000 microroentgens per hour (1.29 microcoulombs per kilogram per hour). The average gamma level associated with the material was about 40 microroentgens per hour (10.32 nanocoulombs per kilogram per hour).

The 1986 investigations by the LLRWMO included the construction of 6 geo-technical wells, 56 shallow groundwater wells, and 3 test-pits. Groundwater samples were collected from 3 of the geotechnical holes.

Analysis of a composite sample collected from more heavily contaminated areas produced a radium concentration of 81.5 Bq/g, a uranium level of 690 ppm, and an arsenic level slightly in excess of 100 ppm. Analyses of the ground water samples collected indicated uranium levels slightly above the federal MAC for drinking water.

The volume of material requiring removal was estimated at approximately 1800 cubic metres.

#### *3.1.6. Landfill Site*

Port Hope's municipal landfill is located within the town, adjacent to the Rollins Ravine site. It is underlain by radioactive materials which were probably placed there over 40 years ago. These materials include ashes, cinders, residues, and refuse mixed with sand.

Gamma levels at the surface of the landfill are indistinguishable from natural background as a consequence of the shielding afforded by the conventional garbage overlying radioactive materials. In 1986, 30 geo-technical boreholes and 14 shallow boreholes were drilled. Down-hole gamma logging was used to generate estimates of the volumes of contaminated material to be removed to achieve compliance with various clean-up criteria. Groundwater samples were collected from 8 boreholes, and surface water samples were collected from springs north and south of the site.

Composite soil samples collected from the more heavily contaminated zones of the landfill were analyzed, and found to contain 12 Bq/g radium, 38 ppm uranium, and 94 ppm arsenic. Ground water samples indicated contamination (chlorides, dissolved organic carbon) from landfill leachate, and elevated radionuclide levels. Analyses of surface water indicated leachate contamination south of the site, and elevated uranium levels at the sample locations above and below the site.

Clean-up of the landfill to the most stringent of 4 criteria being considered (i.e, to 0.3 pCi/g radium-226) may require the removal of 125,000 cubic metres of contaminated material, and clean-up to the least stringent criterion may require the removal of 5000 cubic metres.

#### *3.1.7. Port Hope Harbour*

Sediments within the turning basin of Port Hope Harbour contain elevated levels of several radioactive and non-radioactive contaminants, primarily linked to the earlier years of operation of the Port hope refineries.

The 1986 investigation of the extent of contamination in harbour sediments included 16 geotechnical boreholes. Sediment/soil samples were collected from each hole, and subsequently analyzed to determine concentrations of radionuclides and other elements.

Radioactive contamination within the turning basin of the harbour was determined to extend from the surface of the bottom to depths of from 0.5 to 3 metres. Contamination exists within the approach channel in a lens 1 to 2 metres thick, overlain by from 0.3 to 2 metres of uncontaminated sediment.

Average radium levels in the contaminated sediments of the turning basin and approach channel are approximately 20 Bq/g radium-226 and 1.5 Bq/g radium-226, respectively.

The estimated volume of contaminated sediments in the approach and turning basin is 88,840 cubic metres.

Monitoring of the water quality within Port Hope harbour and the adjoining waters of Lake Ontario indicates only minor impacts on water quality within the harbour, and no measurable impacts on Lake Ontario at large. However, the accumulation of sediments within the harbour prevents access

by other than shallow-draft vessels. Dredging, and subsequent open-water disposal of dredged material, is currently prohibited by regulations due to elevated concentrations of certain (non-radioactive) elements.

#### *3.1.8. CN/CP Viaducts*

Radioactive contamination occurs over 8500 square metres of an open area occupied by the Canadian National (CN) and Canadian Pacific (CP) railway viaducts at the corner of Queen and Hayward Streets in Port Hope. The depth of the contaminated zone ranges from near surface to 1.2 metres, comprising an estimated volume of 7220 cubic metres at an average radium-226 concentration of approximately 4 Bq/g. Contact gamma readings over the contaminated area varies from near background to a maximum of 1600 microroentgens per hour (413 nanocoulombs per kilogram per hour) at one localized spot. Average gamma levels over the area are approximately 22 microroentgens per hour (5.7 nanocoulombs per kilogram per hour).

Because the contamination exists at shallow depth, the 1986 investigations were limited to 10 shallow boreholes. A composite soil sample collected from the areas of greatest contamination had radium, uranium and arsenic concentrations of 96 Bq/g, 2290 ppm, and 2940 ppm, respectively,

#### *3.1.9. Alexander Street Ravine*

The Alexander Street Ravine is a heavily-wooded, steeply-sloped ravine at the west end of Alexander Street. Radioactive contamination exists over an area of 3500 square metres, extends from near surface to over 2 metres in depth, and comprises an estimated volume of 2500 cubic metres of contaminated material at an average radium concentration of 5 Bq/g. Radioactive material consists of ash, municipal garbage, cinders, cobbles, bricks, process crockery, and chemical process residues. Contact gamma readings in the contaminated area range from 8 to 4200 microroentgens per hour (2.06 to 1084 nanocoulombs per kilogram per hour), with the average field over the area estimated to be 50 microroentgens per hour (12.9 nanocoulombs per kilogram per hour).

The 1986 investigations of Alexander Ravine consisted of 5 geotechnical and 48 shallow boreholes, and excavation of a test pit. Groundwater samples were collected from 3 shallow boreholes installed at the bottom of the ravine. Surface water samples were collected from a small stream which flows through the ravine.

Analysis of a composite soil sample representing the zones of greatest contamination yielded radium, uranium, and arsenic concentrations of 45 Bq/g, 1067 ppm, and 134 ppm, respectively.

The surface and subsurface water samples collected were analyzed for various radioactive and non-radioactive parameters. Only uranium exceeded the federal Maximum Acceptable Concentration (MAC), established as a measure of the suitability of drinking water supplies for long-term consumption.

#### *3.1.10. Water Treatment Plant*

The Port Hope Water Treatment Plant is located on Marsh Street, near the shore of Lake Ontario and immediately west of the Port Hope refinery. Radioactive material, in the form of contaminated soil, extends over 14,000 square metres, and generally exists at depths of 0.15 to 0.30 metres, with occasional pockets at depths of a metre. The estimated volume of material requiring removal is 6520 cubic metres. Contact gamma levels over the contaminated area range from near background to a high of 260 microroentgens per hour (67 nanocoulombs per kilogram per hour), and average approximately 12 microroentgens per hour (3.1 nanocoulombs per kilogram per hour). The average radium concentration of the contaminated soil is estimated to be approximately 0.7 Bq/g.

The 1986 investigations of the area consisted of the drilling of 16 shallow holes.

#### *3.1.11. Interim Consolidation Of Port Hope Wastes*

Over recent years, the LLRWMO has completed interim measures to consolidate, and prevent further migration of contaminants from several of the waste accumulations described above. In particular, radioactively contaminated soil from the Strachan Ravine and Rollins Ravine-Monkey Mountain-Brewery Pond areas have been excavated and placed in separate engineered storage mounds at the respective sites. The exposed boundaries of these mounds are capped with impervious synthetic membranes (HDPE) sandwiched between layers of clean fill and topsoil.

The Strachan Street Consolidation Site comprises some 1700 cubic metres of soil contaminated with low levels of uranium, radium and arsenic. This soil was excavated from the formerly contaminated areas of the ravine outside the boundaries of the storage site, and does not include the contaminated materials remaining under adjacent roadways.

Clean-up criteria of 0.3 Bq/g radium-226, 35 ppm uranium and 50 ppm arsenic were applied to the Strachan Ravine project, and post-remedial compliance sampling of the cleaned areas indicated that residual levels of these were well below the respective criteria.

The Pine Street Extension Consolidation Site was constructed within a section of Rollins Ravine, and contains approximately 30,000 cubic metres of soil averaging 2.9 Bq/g radium-226, 26 ppm uranium, and 14 ppm arsenic. Prior to the consolidation of these wastes, which were removed from the locale collectively designated Rollins Ravine-Monkey Mountain-Brewery Pond, the total area affected by contamination was approximately 3.3 hectares. The associated contamination was variable with respect to concentration, areal extent and depth, ranging from larger areas of slight surface contamination (as a result of wind and water transport over the last 30 to 40 years) to more concentrated pockets of contamination at depth (e.g., a few metres below existing grade as a consequence of historical construction activities on abutting residential properties and the Brewery Pond outlet dam). Areas were decontaminated in accordance with criteria similar to those applied to the Strachan Ravine project.

The Strachan and Rollins Ravine Consolidation Sites are currently licensed and inspected by the AECB as interim storage facilities, pending eventual relocation of the associated wastes to a permanent disposal facility. Approved maintenance and inspection plans, radiation protection, and environmental monitoring programs are in effect for both sites.

#### *3.1.12. The Port Hope Construction Monitoring Program*

The LLRWMO, in co-operation with the Town of Port Hope, has put in place within the town, a program referred to as the Port Hope Construction Monitoring Program. This program is designed to detect, and remove from properties scheduled for additional construction, development or modification, small quantities of radioactive materials such as contaminated building materials, soil, fill, etc. Such radioactive materials are received by the LLRWMO at an engineered radioactive waste storage facility (the Pine St. Extension Temporary Storage Facility) near Rollins Ravine, constructed and operated by the LLRWMO for that purpose. The Port Hope Construction Monitoring Program, and the complementary operation of the Pine St. Extension Temporary Storage Facility, collectively facilitate minor development or modification of contaminated sites within Port Hope to proceed relatively unhindered, while encouraging the detection and removal of radioactive materials in a cost-effective manner.

The local by-laws of Port Hope require that persons wishing to make significant modifications to their property obtain authorization permits from the Town. Upon receipt of such an application for properties known, or likely to be contaminated, the Town of Port Hope advises the LLRWMO of the application received. The LLRWMO subsequently provides various services to the property



owner/developer, such as radiation monitoring services or field supervision of waste removal operations, and also accepts for storage at the Temporary Storage Facility referred to above the radioactive wastes encountered.

The Port Hope Construction Monitoring Program is defined by formal administrative arrangements between the Town and the LLRWMO. Execution of the Program is assured through the technical capabilities, support and funding provided by the LLRWMO. The Program is not intended to accommodate developments requiring large-scale removals of contaminated materials because of the limited capacity of the Temporary Storage Facility.

The Temporary Storage Facility is licensed and inspected by the AECB.

### *3.1.13. On-going Initiatives*

Satisfactory resolution of the Port Hope waste problems require that related socio-economic problems be resolved. Ideally, wastes currently existing in-situ or within engineered facilities within the Town should be removed to a permanent disposal facility. However, since no such facility currently exists in Canada, disposal must await siting and construction of such a facility. The possibility of an appropriate disposal facility being available in the near-term appears dependent on the success of the "voluntary siting process" currently being pursued by a federally appointed Task Force. After several years of related developments, this Task Force is continuing discussions with two potential "host communities", as well as the "source community" of Port Hope. Assuming that one of these communities eventually opts (and is selected) to host the required disposal facility, other obstacles such as achieving public acceptance of the proposed facility and associated transport operations, and obtaining the government funding required for the project, may remain.

The Town of Port Hope, and Community Liaison Groups composed of local residents, are intimately involved in the "voluntary process" referred to above, and/or in related discussions, primarily because of Port Hope's status as a "source community". In addition, the LLRWMO, as a consequence of its mandate for historic wastes, including those in Port Hope; its operations and presence within Port Hope; and its role as advisor to the Siting Task Force, is also a key player.

An environmental lobby group, Great Lakes United, identified the Port Hope Harbour as an "area of concern", due, in part, to the radioactive sediments contained therein. As a consequence of this designation, the federal Environment department, in consultation with the public, other agencies, and interest groups, has for some time been involved in the corresponding development of possible Remedial Action Plans for the Port Hope Harbour. However, the Siting Task Force, through the "voluntary process," has assumed a lead role in discussions related to disposal of Port Hope area radioactive wastes, including those in the harbour.

Ultimately, proposals to remove, store or dispose of the Port Hope wastes will require approvals from the Atomic Energy Control Board prior to implementation.

The Town of Port Hope has recently requested an Interim Waste Management (IWM) Plan, to be executed by the LLRWMO, for the 210,000 cubic metres of waste/contaminated material remaining within the Town. "Interim Waste Management" has been defined to include any remedial work activities that could be carried out by the LLRWMO prior to the availability of a permanent disposal facility for the Port Hope area wastes.

The IWM Plan proposed by the LLRWMO recognizes that remedial actions have been completed at all high-priority sites within the Town of Port Hope. The Plan proposes that programs for small scale sites be addressed through on-going programs; in particular, the Construction Monitoring program, a Property Compliance Program, a Property Resurvey Program, and a possible Groundwater Remediation Program. Remediation of major sites, including the landfill, the harbour, and seven major on-land sites will be addressed through Remedial Action Plans currently being

developed by the Siting Task Force Technical Working Group on Remedial Action Plans (TWG-RAP). The range of options being considered by the TWG-RAP include: "do nothing", in-situ management, consolidation on site, and relocation to interim storage.

### **3.2. Welcome "Off-Site" Clean-up**

The Welcome Waste Management Facility occupies the site, and reflects the evolution, of the former Welcome Residue Area. It is currently owned and operated by Cameco, a uranium exploration, mining, and refining company and is maintained in a care and maintenance mode, in accordance with a licence issued by the AECB. The facility has been improved substantially since its early establishment as the Welcome Residue Area, and currently includes runoff diversion systems and contaminated seepage interception, storage and treatment facilities. No new wastes are being received at the site and the former waste burial area has been capped with clean material, contoured, and vegetated. On-going environmental and effluent monitoring results indicate that the facility is being maintained safely, with minimal impacts over the short term pending eventual decommissioning.

The current status of the Welcome facility notwithstanding, the historical operation of the site resulted in substantial low-level contamination of the surrounding environment. These impacts have since been remediated by recent clean-up programs. The early waste burial operations were conducted without the benefit of adequate engineering or operating measures to prevent erosion and overland transport of contaminants by wind and water. Consequently, these processes distributed uranium, radium and arsenic contamination over adjoining farm land and in low-lying areas such as drainage channels and stream beds.

Over 1984 to 1987, the owner of the Welcome Waste Management Facility completed an "off-site" clean-up of the lands affected by the historical operations of the facility. Extensive investigations were undertaken to delineate and characterize the contamination. These included surface gamma surveys, borehole drilling, down-hole gamma logging, soil sampling and analyses, and collection and analyses of surface water samples.

Surface gamma measurements were initially carried out on a 100 metre x 100 metre grid which was later extended in all directions from the waste burial area until levels approximating background were encountered. Three types of data were recorded at each survey point: contact gamma readings obtained at the ground surface, gamma readings obtained at a height of 1 metre above surface, and the "delta" value corresponding to the difference between contact readings with and without a 3 millimetre-thick lead shield. As a result of the survey, potential areas of contamination surrounding and/or extending out from the Welcome site were identified.

A number of boreholes drilled over areas of potential contamination were gamma-logged to determine contamination at depth. The initial drilling program consisted of over 400 shallow (1-3 metres deep) and 50 deep (up to 8 metres deep) boreholes. Soil samples were collected from boreholes, and analyzed for uranium, arsenic, and radium.

In addition to the above investigations, supplementary investigations of various areas were also completed prior to initiation of cleanup activities.

The results of the initial, field investigations indicated that the upper limits of background for the areas in question were 3 pCi/g radium, 2 ppm uranium, and 5 ppm arsenic. Measured concentrations and depths of off-site contamination were highly variable, ranging from near background to orders-of-magnitude higher. The occurrence and extent of contamination were influenced strongly by local topography, the direction of prevailing winds, natural water courses, etc. For purposes of the planned clean-up, areas of contamination were mapped according to common characteristics (e.g., "flat-lands", "wet-lands", relative contamination, etc.) For example, the area to the north of the site was contaminated to a depth of up to 0.5 metres with radium concentrations ranging from 0.4 to 11 Bq/g and uranium up to 50 ppm. Contamination was found to extend along

the east property fence at concentrations of 1.0 to 5.6 Bq/g radium and 10 to 50 ppm uranium at depths less than 0.5 metres. At adjoining locations, radium and uranium concentrations up to 74 Bq/g and 500 ppm, respectively, were recorded. With the exception of a 100 metre of low-level contamination adjoining the fence-line (10 ppm uranium, 1.1 Bq/g radium, at depths up to 0.3 metres), and low-lying areas (drainage channels, water courses) extending out from the facilities, the area to the south of the facility was generally clean. The bed of Brand's Creek and its tributaries contained radium and uranium contamination extending to a depth of 0.5 to 1 metre in concentrations ranging up to 1.9 Bq/g and 100 ppm, respectively. Similar concentrations of contaminants were detected at several other locations. In a few locations, uranium concentrations ranged up to 5000 ppm and radium up to 3.2 Bq/g.

Clean-up criteria were developed on the basis of pathways analyses, which considered a number of factors, including existing and potential land use. The possibility of future development of flat-lands for residential housing necessitated particular considerations of the implications of residual radium concentrations which could give rise to exposure of occupants as a consequence of the interior radon-radon daughter pathway. Adherence to the ALARA philosophy of dose limitation, and considerations related to the chemical and phyto-toxicities of radioactive and non-radioactive elements, resulted in the promulgation of separate criteria for the clean-up of "flat-lands" and "wet-lands" (e.g., creek channels). The selected "flat-lands" criteria were 0.3 Bq/g radium-226 (0.2 Bq/g above natural background), 35 ppm uranium, and 50 ppm arsenic. The corresponding "wet-lands" criteria were 0.8 Bq/g, 100 ppm, and 150 ppm, respectively.

The Welcome Off-Site Cleanup included the excavation of 42,500 cubic metres of native and/or waste materials from contaminated zones, post-remedial compliance sampling, and backfilling of excavated areas with clean fill.

Excavation operations were directed in part by continuous radiation monitoring using portable gamma survey instruments, influenced by previous surveys, and augmented by supplementary soil sampling and analyses. Compliance with clean-up criteria was determined on the basis of soil samples collected, processed, and analyzed in accordance with procedures previously agreed to by the company and regulatory agencies. These procedures involved the collection of soil samples at agreed-upon intervals, homogenization of individual samples into composite samples, splitting of composite samples in support of replicate analyses, and laboratory analyses of "split" soil samples.

In practice, remedial activities necessary to ensure compliance with the clean-up criterion for a particular element (radium, uranium, or arsenic) often ensured that the clean-up criteria for the other elements of concern were met, and, in general, bettered. Compliance sampling of all cleaned areas indicated that the residual levels of contaminants were generally substantially less than the agreed-upon criteria, and, in many cases, approaching, or at, natural background levels.

Routine monitoring of the ground water table in the vicinity of the Welcome Waste Management Facility indicates that radioactive contamination is primarily limited to contamination of the shallow zone underlying the licensed site. Off-site domestic water supply wells in the vicinity of the facility are also monitored regularly, and exhibit no contamination. Contamination existing over the licensed site is considered to be part of the associated waste inventory, and will be dealt with during eventually decommissioning, or by post-closure controls.

### **3.3. Scarborough, Ontario**

In 1980, low-level radioactive contamination was discovered in a residential area (McClure Crescent) of Scarborough, Ontario, near the western edge of Metropolitan Toronto. This contamination originated with activities involving radium-226 which occurred in the 1940's on a 4 hectare farm that occupied the site prior to its commercial development in the 1970's. These early activities reportedly involved the receipt, storage, incineration, and dispersion of various materials (contaminated scrap, luminous paint, radium-impregnated tubing) containing radium-226. Subsequent

development and soil moving activities have spread low levels of radium contamination over two areas: McClure Crescent-Burrows Hall Boulevard, and McLevin Avenue.

Prior to 1981, a partial cleanup of the McClure Crescent area was undertaken, concurrent with a radiation survey of various properties occupying, and adjoining, the site of the original farm. Discrete pieces of radioactive materials were located and removed. The interior of all structures within the study area were sampled to determine if the air contained radon or radon daughter concentrations exceeding the criteria adopted by the Federal-Provincial Task Force on Radioactivity for the clean-up of Port Hope. No radon-daughter levels in excess of the criteria were identified. Extensive surface gamma surveys were carried out in 1980, 1981, and 1983, and collectively identified some 40 properties with contaminated soil. Six hundred boreholes drilled in 1980-81 identified some 2200 cubic metres of contaminated soil. This soil was estimated to have an average radium concentration of 1.4 Bq/g, with a 'hot spot' of 12 Bq/g detected.

The health risks and socio-economic factors affecting the Malvern property owners were assessed. On the basis of socio-economic considerations, removal of the soil was recommended. As lead agency for the Task Force, the AECB attempted to find a site to accommodate the contaminated soil. Successive efforts to relocate the material to Bancroft, the Canadian Forces Base at Borden, and the Beare Road landfill met with local opposition and were unsuccessful, leading indirectly to the demise of the Federal-Provincial Task Force on Radioactivity. The federal Low-Level Radioactive Waste Management Office (LLRWMO) was formed in 1982 to discharge the federal government's responsibility for historic wastes.

A number of actions for damages related to the McClure Crescent contamination were subsequently brought against agents of the federal and Ontario governments, and various decisions were rendered by the Courts. In 1983, the federal and Ontario governments entered into an agreement whereby the LLRWMO would excavate and transport contaminated soil from Scarborough to a temporary storage site named by Ontario, and the federal government would retain responsibility for eventual, permanent, disposal. The parties to this agreement subsequently announced plans to move the soil to the Reesor Road site in Scarborough. In 1984, an opposition group obtained an injunction in federal court against the Reesor Road initiative pending a trial of the issues. In the interim, Ontario purchased the contaminated properties in the McClure Crescent area. The ensuing trial resulted in a 1987 federal court ruling against the injunction. In 1988, the agreement referred to above was automatically renewed for a five year term. The Ontario government declined to exercise its option on the Reesor Road site, and the option was allowed to expire.

In 1989, a resurvey of the Scarborough properties identified elevated indoor-radon concentrations in 15 properties. Remedial works, consisting of the installation of sub-floor ventilation systems, successfully reduced radon levels in the properties.

A pre-development survey in 1990 identified additional radium contamination at a location north of McLevin Avenue in Scarborough. The screening-level gamma survey of a 20 hectare property bounded by Neilson and Tapscott Roads located discrete pieces of plastic tubing coated with radium-226. Detailed gamma surveys of the property were commissioned by the LLRWMO. Measurements were taken on contact with the ground and at a height of 1 metre, at 100 points corresponding to the intersections of a 50-metre grid. The lines connecting grid points were also scanned for radiation anomalies. The results obtained indicated that general gamma levels were elevated slightly above natural background. Four piles of radioactive material were noted. These were scanned using a 2.5 metre grid. Measured exposure rates ranged from 6 to 1500 microroentgens per hour (1.55 to 55 nanocoulombs per kilogram per hour) on contact, averaging about 10-30 microroentgens per hour (2.58 to 7.74 nanocoulombs per kilogram per hour) at 1 metre. Gamma surveys of the property and piles located some 55 pieces of radioactive tubing, which subsequent analyses showed to be impregnated with radium. The tubing segments were about 0.6 cm in diameter and 1.5 to 10 cm in length, and each contained up to 4.4 microcuries (163 kBq) of radium-226.

The above discovery prompted the use of a close-to-the-ground gamma scanning technique to locate additional areas or spots of contamination. This technique involved the use of very sensitive gamma survey instruments, suspended in close proximity to the ground and swept back and forth over the ground in a manner similar to that employed with portable metal detectors. The areal extent and limits of radioactive contamination were determined by this technique.

Boreholes were hand augured into the piles of contaminated material, and the holes were gamma-logged at 5 centimetres intervals. The results indicated that the sources of activity (tubing) were randomly distributed throughout the piles. Composite soil samples were collected from the boreholes, and subjected to spectrometric analyses which indicated radium concentrations similar to natural background. Radioactive material was also identified at depth, extending under the sidewalk to the north of McLevin Avenue.

Various activities associated with the excavation and processing of the contaminated soils at the McLevin Avenue site occurred from July to December, 1990, in accordance with a license issued by the AECB. Some 2500 cubic metres of potentially contaminated soil were stripped from the McLevin property, processed to remove discrete pieces of radioactive tubing, and segregated according to residual radium concentrations. Sorting operations consisted of a combination of manual and automated methods.

The segregation of radioactive tubing from the McLevin Ave. soil was accomplished in two stages. The soil was initially fed to a soil-screening plant to separate it into three fractions on the basis of particle size. Screen spacing was chosen so as to concentrate the pieces of tubing in the "medium" fraction. The screened soil was then placed on a conveyor belt which carried it past fixed radiation detectors. The detectors were interfaced with a computer-microprocessor unit which applied a mathematical algorithm to discriminate between bulk soil and discrete sources (tubing or "hot" particles). When radioactive "hits" in excess of a pre-set level were encountered, the computer would halt the feed conveyor and alert the operator. The offending source would then be located and removed manually. This operation often required additional scanning with a sensitive, hand-held scintillometer in order to locate the source which might be masked by the soil matrix. In practice, this automated scanning system proved slow and labour intensive, but relatively effective in locating the small pieces of randomly distributed radioactive tubing which had escaped the earlier screening process, or in identifying fractions of more highly contaminated soil.

During scanning operations, computer algorithms were varied to compensate for differences in the activity of the soil fractions. This resulted in variations in detector sensitivity and response. It was also discovered that the radium decay products in the pieces of tubing were out of equilibrium more than originally believed. This suggested a possibly significant, corresponding loss of sensitivity of detection. Consequently, a retrospective analysis of the efficiency of the applied algorithms was carried out, and about 5% of the scanned soil was re-examined to assess the accuracy of the original analysis. In support of the latter, the detector system was modified to increase its sensitivity as much as possible.

Approximately 20,000 pieces of radioactive tubing, with an average radium content of 2.2 MBq, were removed from the McLevin Ave. soils, and transferred to the Chalk River Laboratory of AECL Research for interim storage.

The processed soil has been sorted into two fractions: 1800 cubic metres of soil with radium concentrations ( $< 0.07$  Bq/g, average of 0.05 Bq/g) similar to those of uncontaminated Ontario soil, and 1000 cubic metres of soil with elevated radium concentrations (0.07-0.5 Bq/g, average of 0.15 Bq/g). The intent of the sorting completed to date is to permit unrestricted disposal of the normal soil and controlled usage of the elevated soil. Discussions and negotiations related to possible options continue.

Some contaminated soil/fill remains in situ at the McLevin Ave. site. Excavation was halted north of the road surface of McLevin Ave. At that point, the roadway and the adjacent boulevards and sidewalks appear to be built on contaminated fill located at the level of the original grade. The results of borehole testing indicate that contamination continues across the full width of the road allowance for a length of 30 metres. The thickness of the zone averages about a metre, and approximately 700-800 cubic metres of contaminated soil are estimated to remain beneath the road allowance. An estimated 1,200 cubic metres of overburden requires removed in order to access the contamination.

Following removal of the "contaminated" soil from a portion of the McLevin Ave. site, the excavated area was subjected to a post-remedial gamma scan, and a soil sampling campaign to verify the effectiveness of the cleanup. The results of the analyses completed on the soil samples indicated that residual radium concentrations in bulk soil over the cleaned area were indistinguishable from natural background concentrations. A specially developed survey vehicle was utilized for the post-remedial survey. This vehicle consisted of three sodium iodide gamma detectors mounted on mobile pull cart, along with an associated computer-microprocessor unit. The detector units were mounted on a support frame, and fixed at significant distances from each other in a triangular configuration. Signals from the detectors are fed to the computer processor which compares the signals received with each other and with those received previously, and applies statistical/vector analyses to detect and signal significant anomalies. The effectiveness of the equipment was confirmed by the results obtained, whereby a few additional pieces of radioactive tubing not located in the earlier surveys were detected, and removed.

Further survey and remedial work remains to be completed at, and in the vicinity of, the McLevin Ave. site. As a result of recent developments, it appears that these activities may be completed within the context of an overall survey and cleanup of the "McClure" and "McLevin" districts. The federal and Ontario governments have recently (1993) resumed co-operative efforts to relocate radium contaminated soil from in-situ locations in Scarborough to an interim storage site, pending eventual disposal.

In June, 1993, the government of Ontario announced that it had selected an industrial site on Tapscott Road, Scarborough as a potential location for processing and interim storage of contaminated soil to be excavated from Scarborough properties. The announcement noted that the project would be subjected to the federal Environmental Assessment and Review Process Guidelines Order.

It is proposed that, under the supervision of the federal LLRWMO, soil be removed from 48 residential properties in the McClure Crescent area and transported to the Tapscott Road site. There, it would be sorted to remove radium-coated plastic tubing and particles, the cleaner soil would be temporarily stored on the site, and the contaminated tubing would be forwarded to Chalk River Laboratories. Previously-treated soil from McLevin Avenue would also be moved to the Tapscott site for temporary storage. The project is scheduled for 1994.

The Environmental Assessment and Review Process (EARP) Guidelines Order is a federal legislative requirement that projects undertaken, funded or regulated by federal agencies be subjected to environmental screening (assessment) before proceeding in order to determine their potential impacts. For such projects, the Order includes specific requirements related to public consultation, and assessment and addressing of related concerns.

The proposed Scarborough clean-up project is subject to the EARP Guidelines Order, and thus a draft Environmental Screening Report for the project has been prepared and was released for public comment in September, 1993. Comments are due by November, 1993, following which decisions as to whether, and under what (if any) conditions, the project may proceed will be taken. In the interim, a group comprised primarily of Tapscott Road landholders and business interests has made representations opposing selection of the Tapscott site. Contrary representations have also been made by McClure residents in favour of the project.

### 3.4. "Abandoned" Uranium Mines

Prior to the early 1960's, uranium tailings management practices in Canada and elsewhere were primarily determined by operational requirements. Chief amongst these were water balance considerations and the desire to dispose or contain the large volumes of tailings generated as conveniently and economically as possible. In Canada, the latter requirements were most easily satisfied by the use of nearby lakes or natural depressions for tailings deposition. Topographical features were often improved upon for purposes of tailings confinement by the addition of retention berms or dams. These structures were usually constructed with locally-available materials, most commonly waste rock from mining operations. Their generally permeable nature permitted relatively significant seepage losses.

The severe recession in the Canadian uranium mining industry in the 1960's led to the closure of most mines and mills. These ceased operation without any consideration, or implementation, of special decommissioning or reclamation measures. (For purposes of this paper, such operations are referred to as "abandoned", although the sites and any remaining assets of such former operations may remain under the control of government, individuals, or corporations.) In the absence of regulatory imperatives, any decommissioning activities completed coincident with, or subsequent to, shutdown were implemented voluntarily, primarily for practical or economic considerations. The potentially adverse effects of un-contained uranium tailings on the environment were largely undocumented at the time. However, subsequent studies and observations (and, in particular, those related to pyritic uranium tailings) identified specific examples of environmental degradation, and implied the existence of others. Public concern over the potentially adverse environmental effects of uranium mining activities increased, reflecting in part escalating public awareness of, and concern over, environmental issues.

In the early 1960's, Tsivoglu and O'Connell of the Robert A. Taft Sanitary Engineering Center in Cleveland, Ohio researched radium-226 levels in the Animas River, and subsequently (1962) published "A Waste Guide for the Uranium Milling Industry". Concern in Ontario regarding the increasing concentrations of radium in the Serpent River Basin led to establishment of a government committee to assess the problem. The resultant report, issued in 1965, gave impetus to the introduction by Canadian operations of such measures as water conservation in uranium mills, barium-chloride treatment of mill effluent to precipitate radium, and environmental monitoring programs. The Canadian operations that survived the economic downturn of the 1960's-early 1970's, and those which were re-opened or developed later, adopted tailings neutralization, effluent treatment, and improved containment of mill tailings and liquid wastes.

Since the mid-1970's, all uranium mines and mills operating in Canada have been subjected to increasing, and extensive, government regulation, both federal and provincial. These regulations includes requirements for public hearings, as part of environmental review processes for proposed operations, as well as radiation dose limits, effluent discharge limits for radioactive and non-radioactive parametres, monitoring requirements, decommissioning requirements, prohibitions against non-approved abandonments, and various other conditions of licence or regulation related to health, safety, and protection of the environment.

Canadian uranium mines and mills are subject to the federal Atomic Energy Control Act and Regulations, and consequently are regulated by the Atomic Energy Control Board (AECB), in co-operation with other federal and provincial agencies. The policy of the Atomic Energy Control Board with respect to the long-term management of radioactive wastes, including those resulting from uranium mining operations, is described in Regulatory Document, R-104, "Regulatory Objectives, Requirements and Guidelines For The Disposal Of Radioactive Wastes". This document establishes as a primary objective the post-operational decommissioning/reclamation of nuclear facilities so as to permit eventual unrestricted access, and use, of the associated site. However, this policy also recognizes that this objective may be unrealistic for certain situations involving large volumes of long-lived wastes such as uranium tailings. Consequently, for these situations, the imposition of, and

reliance upon, long-term institutional controls may be necessary. Thus, the current Canadian position recognizes that the pursuit of uranium mining and processing, like other mineral exploitation, cannot occur without some impact on the environment. Societal and regulatory expectations are not that these impacts be "zero", but rather that they be minimized to the extent practical, and that the associated activities be conducted safely. To this end, a number of uranium mining and milling operations have undergone approved decommissioning actions, as described elsewhere.

The following sections describe uranium mines which were abandoned years ago, and consequently were not subject to environmental controls over their operating lifetimes or upon shut-down. Negotiations between the federal government and the provinces in which these mines are located regarding cost-sharing for decommissioning of these abandoned mines are underway. In the interim, related studies have indicated that these sites do not pose significant risks to public health and safety over the near-term. However, impacts over the longer remain a concern. Abandoned sites are located in areas which are currently very sparsely populated and/or remote from permanent settlements with no convenient transportation links to major centers.

#### *3.4.1. The Gunnar and Lorado Sites*

Over the first decade of its uranium mining history, the Beaverlodge area of Canada, located along the north shore of Lake Athabasca in the north west corner of the province of Saskatchewan, had 10 mines producing uranium ore. These mines fed 3 mills: Eldorado Mining and Refining (2000 tonnes/day), Gunnar Mining Ltd. (2000 tonnes/day), and Lorado Uranium Mines Ltd. (700 tonnes/day). The Gunnar and Lorado operations closed down without reclamation by 1964. The Eldorado operations near Eldorado (Uranium City), Saskatchewan continued until 1982, and were decommissioned over the ensuing 3 years.

##### *3.4.1.1. Gunnar*

The Gunnar uranium mine and mill are located on Crackingstone Point on the north shore of Lake Athabasca near Uranium City, Saskatchewan. The mill began production in September, 1955 at a nominal capacity of 1000 tonnes/ day, underwent a number of subsequent expansions, and operated until closure and abandonment in 1964.

The Gunnar mill generated some 4,000,000 tonnes of tailings over its lifetime. These were initially discharged into Mudford Lake, 500 m. to the northwest. When the Mudford Lake basin approached capacity, its outlet was lowered by blasting, allowing tailings to spill into a down gradient basin, and subsequently into Langley Bay of Lake Athabasca.

The Gunnar Mine consisted of an open pit originally constructed immediately adjacent to (i.e., to the north of) St. Mary's channel. The narrow section of bedrock separating the pit and channel was breached in 1964, permitting water from Lake Athabasca to flood the pit. Waters from the pit and Lake mingled via this channel until 1966, at which time the breach was backfilled with crushed rock. Waste rock from the mining operations was placed in two piles to the southeast of the pit.

A 1989 study of the Gunnar site by Beak Consultants identified environmental and/or health and safety concerns with respect to the tailings (radiation exposure, water contamination), waste rock (radiation exposure and contaminated runoff), mine pit (water contamination), and site buildings (structural hazard). The corresponding report identified 3 reclamation options for the Gunnar site. These ranged in estimated cost from \$225,000 to \$5,000,000.

The Gunnar tailings are only slightly acidic, at pH 6-7. Although the surface zones of the upper reaches are susceptible to wind erosion, the substrate moisture is high and consequently the surface is generally stable. The coarser fractions of the tailings are generally non-saline, and the finer material moderately- to strongly-saline. With the exception of elevated concentrations of lead (200-



300 ppm), preliminary data indicates that the concentrations of heavy metals in the tailings are generally within the ranges reported for normal soils.

Vegetation of the Gunnar tailings appears to be occurring naturally, and relatively quickly, over most of the exposed surfaces. However, two areas of 17 hectares and 1 hectare remain bare of vegetation, primarily due to the impacts of wind erosion and the low fertility of the tailings. Thus, stabilization of these sections of the Gunnar tailings ("Mudford Lake") may be required.

The primary water quality concern at the Gunnar site relates to the results of tailings encroachment into Langley Bay of Lake Athabasca. Although these tailings are not acid-producing, they are contributing to enhanced suspended and dissolved solids, and elevated radionuclide concentrations. Preliminary results suggest that fish in Langley Bay may be concentrating radionuclides. Additional study to determine if this is indeed the case, and if so, whether fish migrate into other areas of Lake Athabasca, appears warranted. If it appears that sediment contamination from Langley Bay is spreading, and water quality or fish outside of the Bay are being impacted significantly, measures to contain the contamination, such as a dam across the mouth of the Bay, may be required.

Discussions are currently underway between the federal and Saskatchewan governments regarding decommissioning of the Gunnar site, and cost-sharing of the related costs.

#### 3.4.1.2. Lorado

The Lorado site is located on the west shore of Nero Lake, approximately 10 km west, by road, of the town of Uranium City, Saskatchewan. Nero Lake is a small lake which drains into Beaverlodge Lake, which in turn discharges to Lake Athabasca via the Crackingstone River.

The Lorado mill began production of uranium concentrate in May, 1957 at a design tonnage of 650 tonnes/day, and operated until April, 1960. During that period, approximately 350,000 tonnes of ore were milled, generating a similar volume of tailings. The tailings were discharged untreated at a pH of 2.0 into a small "lake" at the west end of Nero Lake. This depression filled the first year of operation, and more than 300,000 tonnes of tailings were subsequently discharged to Nero Lake, which is separated from Beaverlodge by bridge of land. Over the years to present, seepage from Nero Lake has occurred through this formation, resulting in the formation of a precipitate flume extending a short distance into Beaverlodge Lake.

A number of studies and investigations of the Lorado site have identified environmental and/or public safety concerns related to the tailings (radiation exposure and water contamination) and mill building (structural hazard). A 1989 study recommended three potential levels of remedial action, ranging in cost from \$220,000 to \$1,200,000.

In 1990, the company which owns the land which the mill and a small portion of the tailings occupy demolished the mill and cleaned up the associated area.

Residual environmental concerns at the Lorado site are associated with the tailings. The exposed portion of the Lorado tailings covers an area of 14 hectares, with the remainder of the tailings covered by water. Access to exposed tailings is unrestricted by fencing or other barriers. The tailings appear to be pyritic with a pH ranging from 2.4 to 3.9, and strongly saline. Wind erosion is apparent on the drier, coarse-textured sections of the tailings, and indications of water erosion are apparent in the lower regions. Natural revegetation of the tailings is sparse, and limited to areas where the depth of tailings is shallow. Gamma fields over the exposed tailings are typically several hundred microrentgens per hour (77 nano-coulombs per kilogram per hour).

Nero Lake is highly acidic and strongly saline. Radionuclide and heavy metal concentrations are significantly higher than those of surrounding lakes. The high acidity of Nero Lake prevents fish survival and hence the potential for significant aquatic food chain transfer.

Currently, the province of Saskatchewan and the federal government are involved in discussions regarding close-out criteria, decommissioning and reclamation, and associated cost-sharing.

#### *3.4.2. The Port Radium Site*

The community of Port Radium and the Port Radium mine and mill, were established in the 1930's on the shores of Great Bear Lake in the North West Territories as a source of radium and silver. The initial phase of operation extended until shutdown in 1940. The mine was reopened by the Canadian government in 1942 to provide uranium for the Manhattan Project. This second phase continued until 1964. During these consecutive phases, the Port Radium mine produced approximately 1,000,000 tonnes of ore, and radium and uranium concentrates for further refinement at Port Hope, Ontario. Tailings were discharged (directly, and indirectly) to an adjacent, deep bay of Great Bear Lake, and also deposited in depressions (e.g., Radium Lake, Murphy Lake ) and surface pits on land. From 1952 to 1960, most of the submerged "radium" tailings (over 300,000 tonnes) were recovered by dredge, and processed to recover their uranium content.

In 1974, the Port Radium mine was reopened as a silver mine, and operated until 1982. In 1985, Echo Bay Mines completed various decommissioning activities at the site. In addition to the closure of the mine openings and removal of surface structures, and cleanup of resultant debris, exposed tailings were covered with waste rock to reduce radiation fields and discourage further surface erosion.

A number of investigations and studies of the impacts of the Port Radium wastes were completed prior to the above decommissioning. These included assessments of the impacts of the submerged tailings on the water quality of Great Bear Lake, and on fish in the Lake. Given the great depth (up to 100 metres) of the water cover over the submerged tailings, impacts on the water column appear limited to slightly elevated levels of radium and uranium in the aqueous zone near the tailings surface. The results of fish monitoring studies indicated that fish from Lake Athabasca do not appear to exhibit evidence of bio-accumulation of radionuclides as a consequence of the Port Radium tailings. Thus, although 1.5 million tonnes of silver and uranium tailings remain in close contact with Great Bear Lake, only localized impacts have been observed to date. It has been suggested that the current conditions of the submerged Port Radium tailings and the associated aquatic environment support arguments favouring deep water disposal of tailings.

#### *3.4.3. The Rayrock Uranium Mine and Mill Site*

Like the Gunnar, Lorado, and Port Radium sites, the Rayrock site is also isolated in the Canadian wilderness, accessible only by helicopter or overland via winter road. It is situated approximately 145 kilometres northwest of the city of Yellowknife, capital of the North West Territories, and 56 kilometres north of Marian Lake. The Rayrock mine and mill operated from 1957-1959, producing about 60,000 tonnes of tailings which were originally deposited in two piles immediately south of the mine site. These piles covered areas of about 6 hectares and 8 hectares were weakly acidic and bare of significant vegetation, extended into the vicinity of small ponds or marsh, and gave rise to surface gamma fields representative of similar uranium tailings.

Several studies of the Rayrock site have been carried out since the late 1970's. These suggest that the smaller water bodies immediately adjacent to the tailings may be contaminated, but that Sherman Lake and its tributaries do not exhibit evidence of alteration of water quality.

### **3.5. Surrey, British Columbia**

In the early 1970's, ore concentrates originating in South America were imported from the United States by Fundy Chemical Company of 7800 Anvil Way, Surrey, B.C., and smelted for the extraction of rare earth elements intended for use in the subsequent production of ferro-alloys. Approximately 290 tonnes of slag generated by the smelting process were left on company property. This slag contained about 1% by weight of natural thorium and 0.026% natural uranium. The property and the accompanying radioactive slag were transferred to a new owner, who, believing the slag to be inert, mixed some of it with sand and utilized the resulting mixture as fill on another property he owned. An investigation in 1981 of the latter property at Anvil Way revealed that use of the mixture had contaminated a significant portion of the property to a depth of approximately a metre, and estimated that some 4000 cubic metres of fill might require removal in order to clean the site to its former state. The owner of the Anvil Way property subsequently initiated legal actions against the vendor, and the federal and provincial governments.

In 1984, the LLRWMO was given responsibility, on behalf of the federal government, for disposal of the "Surrey" wastes referred to above. Subsequently (1985), the LLRWMO excavated 2700 cubic metres from under and around buildings and pavement at 7800 Anvil Way, and placed it in an interlocking concrete block structure, or "bunker", which was constructed in a corner of the property as interim storage of the Anvil Way wastes, pending its final disposal along with the 90 tonnes of undiluted slag stored at another Surrey property. The latter inventory is contained in steel drums stored inside a secured building. The Anvil Way storage bunker is located over a paved section of the property, which is still underlain by about 1000 cubic metres of radioactive fill. This fill will be excavated and disposed of along with disposal of the waste stored in the bunker.

In October, 1989, the federal Minister of Energy, Mines and Resources (EMR) announced the appointment of a 2-member Siting Task Force with a mandate to seek out volunteer communities willing to accept a facility for disposal of the Surrey wastes. Since its appointment, this Task Force, supported by the LLRWMO and EMR, have been actively pursuing a disposal solution,

### **3.6. Chalk River Laboratories**

The Chalk River Laboratories (CRL) of Atomic Energy Of Canada Limited (AECL) comprise a large, long-established nuclear research and commercial services complex, located on the southern shore of the Ottawa River, 160 km northwest of the city of Ottawa (Figure 1). The facility occupies a restricted area of 37 square km, extending 6 km back from the Ottawa river. The restricted area is bordered on the southwest by the Petawawa Military Reserve which extends 20 km downstream. The land surrounding CRL is primarily covered with forests of small commercial value, and is not utilized for significant agriculture. The immediate area is sparsely populated, and the nearest settlement is Chalk River, with a population of 870.

For several years, AECL has operated commercial waste storage and treatment services at CRL. In addition, solid and liquid wastes generated by various AECL activities and facilities over several decades were routinely disposed of at the Chalk River site by a variety of methods. These historical operations created several "waste management areas" which require on-going care and monitoring, and possible remediation in the future. The sophistication of these facilities vary from unlined burial pits and trenches to modern, engineered storage structures. Current waste management practices at CRL have evolved over several decades. Consequently, the active waste management areas at CRL also include a blend of waste management approaches and structures representative of this evolution.

The earlier waste disposal operations at CRL did not include features to prevent infiltration of surface water and precipitation, or to prevent the release or migration of liquid wastes and

contaminants. At best, the accompanying waste management philosophy encouraged "controlled dispersion" rather than "containment" of potential contaminants, and some of the disposal areas incorporated corresponding features such as ion-exchange beds, etc. Consequently, the shallow groundwater underlying some of the older disposal areas has become increasingly contaminated, and the associated contamination plumes have spread beyond the immediate boundaries of the disposal areas, and continue to spread, albeit slowly.

Reference (2), an AECL submission to the Atomic Energy Control Board, describes the results of investigations to-date into ground water contamination at CRL, as well as the associated remedial activities implemented or contemplated.

## REFERENCES

- [1] Atomic Energy Control Board, Controlling Low-Level Radioactive Wastes, Information Series.
- [2] Atomic Energy Research - Chalk River Laboratories, "Groundwater Plume Summary for Chalk River Laboratories", Submission to Atomic Energy Control Board, February 26, 1993.
- [3] Barnard, J.W., Deverall, R., Safe Handling and Disposal of the Surrey Wastes, 1990.
- [4] Barsi, R.G., Ashbrook, A.W., A Brief Historical Review of the Beaverlodge Mining Area of Northern Saskatchewan, 1992.
- [5] Kalin, M., Port Radium, Northwest Territories - An Evaluation of Environmental Effects of the Uranium and Silver Tailings, University of Toronto, 1984.
- [6] Knight, G.B., Regulatory Concerns Arising from the Port Hope Situation, (Canadian Nuclear Association Conference, Toronto, Canada, June 13 - 16, 1976).
- [7] Griffith, J.W., The Uranium Industry - Its History, Technology, and Prospects, Mineral Report 12, Mineral Resources Division, Department of Energy, Mines and Resources, Ottawa, 1967.
- [8] Low-Level Radioactive Waste Management Office, Annual Report to the Atomic Energy Control Board for Prescribed Substance Licence 197/92, 1991.
- [9] Low-Level Radioactive Waste Management Office, Low-Level Radioactive Contamination in the Scarborough East Area - Current Status of Remedial Work and Future Considerations, Malvern Remedial Action Committee, 1991.
- [10] MacLaren-Lavalin, Radiation Surveys of the McClure Crescent Area of Scarborough, Ontario - Interim Report, Atomic Energy Control Board, 1981.
- [11] MacLaren Inc., Port Hope 1988 Remedial Program-Project Summary Report, Atomic Energy Of Canada Limited - Low-Level Radioactive Waste Management Office, 1991.
- [12] MacLaren Engineers - Golder Associates, Port Hope Remedial Program - Radiological Assessment and Conceptual Engineering Design for the Cleanup of Major On-Land Areas of Contamination, Low-Level Radioactive Waste Management Office, 1987.
- [13] Main, D.E., 1991 Annual Report to the Atomic Energy Control Board for Prescribed Substance Licence 197/92, Low-Level Radioactive Waste Management Office, 1992.
- [14] Senes Consultants Limited, Development of Clean-Up Criteria for the Port hope Remedial Program, Low-Level Radioactive Waste Management Office, 1987.

- [15] Senes Consultants Limited, Environmental Assessment Of The Port Hope Remedial Program, Low-Level Radioactive Waste Management Office, 1987.
- [16] Senes Consultants Limited, Environmental Screening For McLevin Avenue Remedial Action Program, Atomic Energy Of Canada Limited - Low-Level Radioactive Waste Management Office, Ottawa, Ontario, 1990.
- [17] Senes Consultants Limited, Engineering Summary Of 1990 McLevin Avenue Remedial Action Program, Atomic Energy Of Canada Limited - Low-Level Radioactive Waste Management Office, Ottawa, Ontario, 1991.
- [18] Senes Consultants Limited, Retrospective Analyses Of McLevin Avenue Detection System, Low-Level Radioactive Waste management Office, 1991.
- [19] Senes Consultants Limited, 1992 Annual Licence Report To The AECB - (AECB-WFOL-344-1) - Port Hope Waste Management Facility, Low-Level Radioactive Waste Management Office, 1993 .
- [20] Swanson, S., Abouguendia, Z., The Problem Of Abandoned Uranium Tailings In Northern Saskatchewan - An Overview, SRC Publication No. C-805-48-C-81, Saskatchewan Research Council, 1991.
- [21] Surrey Siting Task Force, Interim Report, 1990.
- [22] Whitehead, W., Decommissioning Of Uranium Mines And Mills - Canadian Regulatory Approach And Experience, Atomic Energy Control Board, 1986.
- [23] Zelmer, R.L., Main, D.E., Recent Developments In Historic Waste Program, Atomic Energy Of Canada Limited - Low-Level Radioactive Waste Management Office, Ottawa, Ontario, 1992.

# RESTORATION OF RADIOACTIVELY CONTAMINATED SITES IN THE REPUBLIC OF CROATIA

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## Abstract

In March 1993 Croatia has joined to the IAEA Technical Co-operation Project on Environmental Restoration in Central and Eastern Europe. The proposed project structure was fully accepted, i.e. the project implementation in Croatia is supposed to be carried out through three stages: (1) identification and characterization of radioactively contaminated sites; (2) preparation for restoration, and (3) restoring radioactively contaminated sites. The Hazardous Waste Management Agency from Zagreb has been appointed to be the project co-ordinator in Croatia. Through contacts with experts from the IAEA who are responsible for the international project implementation, it was defined the general structure of the project performance in Croatia in respect to specific situation in our country i.e. regarding the types of radioactively contaminated sites. There are four groups of radioactively contaminated sites in Croatia: (1) sites containing coal slag/ash piles; (2) sites containing phosphates and phospho-gypsum from fertilizers industry; (3) geothermal springs and gas/oil drillings; and (4) sites containing natural radioactive materials. The highest priorities in the clean-up action should be given to the INA-VINIL plant in Kaštel Sućurac and the coal-fired power plant PLOMIN (group 1), as well as the fertilizers factory INA-PETROKEMIJA in Kutina (group 2). The project performance is controlled by the Ministry of Health, the Ministry of Civil Engineering and Environmental Protection and the Ministry of Economy - Department of Energy. Main co-ordinating institutions in the project implementation are the institute "Ruđer Bošković", the Institute for Medical Researches and Occupational Medicine, and "ECOTEC" - the company dealing with ecological engineering and ionizing radiation protection. Unfortunately, there are few considerable obstacles aggravating the regular project performance. Firstly, there are some difficulties referring to institutional and capacity building, as well as the infrastructure set up in Croatia as a newly independent country. Nevertheless, the problems connected with war running in some parts of our country, lack of financial means, repulsiveness of the public to issues close to radioactive waste, and - finally - the absence of convenient temporary storage capacities or final repository for radioactive materials which should be restored through the project implementation, deserve a special attention in order the project to be regularly realized.

## 1. INTRODUCTION

On the basis of the International Atomic Energy Agency (IAEA) initiative, Croatia joined the Technical Co-operation Project on Environmental Restoration in Central and Eastern Europe in March 1993. According to the IAEA suggestions referring to multi-phase project performance, the identification and characterization of radioactively contaminated sites are the main objective to be

achieved in first phase of the project. The Hazardous Waste Management Agency from Zagreb<sup>1</sup> has been appointed as the project co-ordinator for Croatia. The main co-operating institutions in the project implementation are the Institute "Ruđer Bošković", the Institute for Medical Research and Occupational Medicine, and the ECOTEC company, authorized to handle radioactive materials. The project is supported also by some governmental bodies like the Ministry of Health, the Ministry of Economy and the Ministry of Civil Engineering and Environmental Protection.

There are two issues to be considered during the project implementation in Croatia: (1) completion of measurement of natural (background) radioactivity; and (2) identification and characterization of sites suspected to be radioactively contaminated.

With regard to the item (1) it is worth saying that at few sites (eg geothermal springs) have an increased radiation level, but they can not be considered as radioactively contaminated or polluted spots due to natural origin of their radioactivity. Nevertheless, they should be examined thoroughly in order to protect people coming close to them for professional or any other reason. Similar or even more attention should be paid to gas- and oil exploitation fields (ie boreholes), where considerable activities have been measured. Radiation protection of people working at sites where increased radiation level could be expected, but also for the reason of providing comparable "zero" levels for assessment of artificially generated radiation, it is necessary to set up the radiometric map of Croatia as soon as possible. Since not more than preparatory actions on accomplishment of the map have been done so far, it is realistic to expect in some cases certain difficulties in identification and characterization of radioactively contaminated sites. Fortunately, at some areas fairly detailed surveys of background radiation have been carried out.

Furthermore, there are four groups of sites to be identified and characterized as locations suspected to be radioactively contaminated, and therefore involved into the project performance: (a) sites containing slag- and ash piles from coals having increased radioactivity levels; (b) sites containing phosphates, ie waste gypsum and fertilizers; (c) geothermal springs, gas- and oil drillings; and (d) exploitation sites of the materials having increased background radiation. Common facilities to which are referring sites classified in groups (a) and (b) are coal- and oil-fired plants, coking plants, plants producing plastic materials, light metal factories, iron works, cement works, fertilizer factories, oil refineries, old railway slag- and ash piles etc.

All these sites are foreseen to be examined in order to get valuable information on their geographical distribution, history of contamination (at sites where institutional inspection and monitoring have been carried out), identification of routes used for transportation of radioactively contaminated materials, proximity of sites to population centres, radiation concentrations in considering materials and contamination spreading directions, as well as methods planned to be applied in sampling and radiation measurements. It is true that all the issues mentioned above, can not be fully described in the present report, but they are supposed to be elaborated - according to our realistic abilities - during following six months. An international support to the project will be welcome, in particular for those issues which have not been well-experienced in Croatia so far. It is referring in some extent also to preparation of new Croatian legislation in the field of radiation protection, expected to be conceived in harmony with the highest international standards.

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<sup>1</sup> The Agency was established in 1991 by the Croatian Government as the national agency responsible for management of radioactive waste coming from nuclear power plant Krško - sited in Slovenia, but being a joint venture facility of both Slovenia and Croatia - and radioactive waste generating in Croatia (hence, former name of the Agency was Croatian Radioactive Waste Management Agency). However, it has meanwhile widened its competencies also to the field of management of other hazardous wastes, and - consequently - changed its name. The Agency is subordinate to both the Ministry of Economy and the Ministry of Civil Engineering and Environmental Protection. The main objective of the Agency's activities is to organize and perform actions resulting in improved management of both the radioactive and other hazardous wastes, and contributing thereby to improvement of the human health safety and environmental preservation.

Additional activities supposed to be involved in the project are identification, separation and removal of building materials, radioactively contaminated in the war due to induced radioactivity from damaged radiation sources, like radioactive lightning rods, ionizing smoke detectors, radiation sources used in industry and medicine etc. (Fig. 1). The removal of damaged/destroyed radiation sources from the areas of the country which have been affected by the war, is the objective of another on-going project, being also considerably supported by the IAEA.

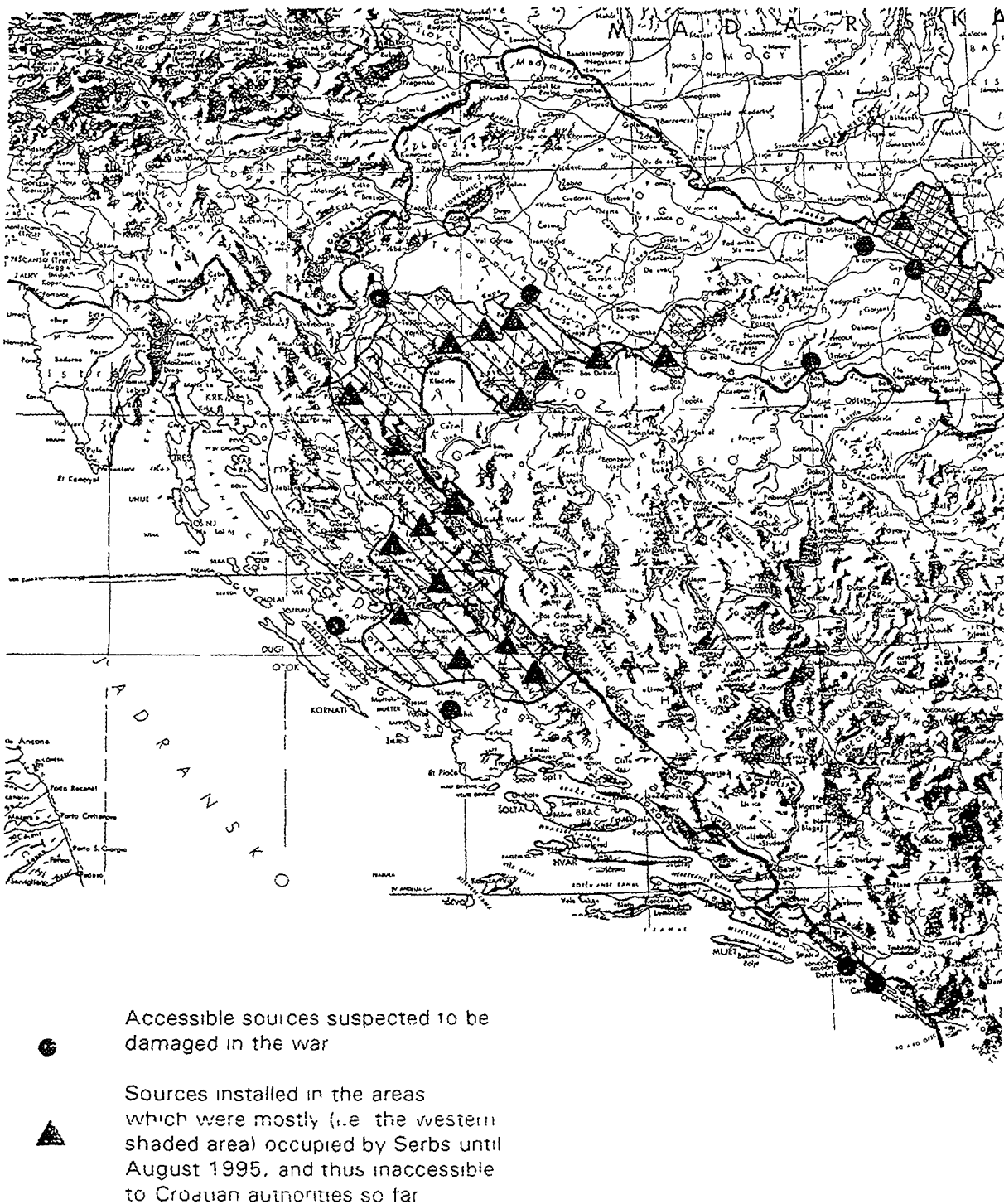


Fig 1 Sites in the areas of Croatia where radiation sources (radioactive lightning rods, ionizing smoke detectors, sources in medicine and industry) have been installed



From the viewpoint of the project scale, ie with regard to objectives supposed to be reached during the project implementation, it should be emphasized that there are neither uranium mining and milling industry, nor areas contaminated by nuclear accidents or past waste dumping practices in Croatia. In addition, no abandoned factories where radioactive substances were used, and areas contaminated by defence-related activities have been reported in our country so far.

Nevertheless, there are several considerable obstacles aggravating the regular project performance in Croatia. First, there are difficulties related to institutional and capacity building, as well as infrastructure set up in Croatia as a newly independent country. However, acute problems connected with the war running in our country, lack of financial means, repulsiveness of the public to issues close to radioactive waste, and - finally - absence of any convenient temporary storage or final repository for disused radioactive materials planned to be restored through the project implementation, should be also considered.

It must be mentioned that environmental state in Croatia is not yet satisfactory. Therefore, we have recently started some actions in the field of institutional conceptualization, including set up of organizational responsibilities needed for efficient performance of programmes expected to contribute to improvement of environmental preservation in the country. Ministries of environmental protection, economy (including industry and energy), and health, as well as some subordinate agencies, have prepared actions in the field of waste management strategy, improvement in technology and corresponding more stringent regulations aiming to minimize releases in soils, water and atmosphere, and decrease waste production. Some efforts on protection of human health due to present deterioration of environmental status have been conducted as well, but it is difficult to forecast their efficiency due to actual financial troubles in entire Croatia, and problems related to the implementation of institutional control in some of its regions which have been occupied.

What can be expected of this paper is to make clear the present status on, at least, majority of sites supposed to be radioactively contaminated, give priorities in the clean-up programme, describe structure of responsibilities, and put some light to the project on-going actions in Croatia. Of course, some additional data on the country and its geography, geology, population and economy could be useful for better understanding of the project implementation in Croatia.

## 2. GENERAL INFORMATION ABOUT CROATIA

Republic of Croatia (56,538 km<sup>2</sup>) is the country sited in southern part of Central Europe, participating in three major physical-geographical regions: Pannonian basin, Dinaride mountains and the Adriatic coast. As a part of former Yugoslavia, it proclaimed independence in June 1991. There are 4,800,000 inhabitants living in Croatia, and population density in the country is 85 inhabitants per km<sup>2</sup>. Due to its unusual shape, the total length of Croatian borders is remarkable - 2,100 km. The neighbouring countries are Slovenia, Hungary, Serbia, Bosnia & Herzegovina and Montenegro (the border with Italy is running along the Adriatic sea). The major Croatian cities are Zagreb (the capital, 800,000 inhabitants), Split (200,000), Rijeka (180,000), Osijek (110,000), Zadar (80,000) and Pula (65,000). Farming, forestry, fishery, industry, shipbuilding and tourism are the main economic activities practised in Croatia. Industry (including mining and energy production) gives 44 % of national gross income.

The country has been suffering *a lack of energy* for a long time, and it cannot be expected any significant improvement in energy supply until the war in both Croatia and Bosnia & Herzegovina will be stopped. The most important sources of energy in Croatia are oil and gas, as well as hydro-energy. Exploitation of most coal-mines were not economic, and they (containing mainly low-energy coals) have been shut down. The only exception is the stone-coal mine at Raša, supplying the nearby power-plant Plomin (Istria).

It is true that there is a number of gas (Molve, Stružec, Lipovljani etc.) and oil fields (Beničanci, Stružec, Žutica, Gojlo, Šandrovac, Deletovci etc.) being exploited in Croatia, but their production (eg ca. 2.5 million tons of oil per year) can not satisfy actual needs. Almost all of oil and gas fields are sited in the interior of the country (in the Pannonian basin), but there are also some gas investigating drillings organized offshore, in the north Adriatic. Domestic oil and gas participate with some 60% of total energy consumption in Croatia.

There are two oil refineries in Croatia, in Rijeka and Sisak (annual refining capacities of each refinery are 7-8 million tons), and the oil pipeline connects the oil terminal at Omišalj (the island Krk) with the Sisak refinery, running further in two directions - eastward (along Sava river) and northward (Hungary, Slovakia). Unfortunately, the pipeline is due to actual war operations out of operation.

Hydro-energetic potential of Croatia is estimated to be some 11,200 GWh of electricity per year, and there have been operating hydro-power plants able to produce 6,500 GWh (ie ca. 60 % of the estimated potential). The share of electricity produced in hydro-power plants in Croatia is about 35 % of total electricity consumption. Additional sources of energy are coal-, oil-, and gas-fired power plants (35 %) as well as nuclear power plant Krško (Slovenia), producing some 15 % of total electric consumption in Croatia. The rest of energy necessary to satisfy total needs is imported from other countries.

It should be emphasized that the mentioned data on energy production are referring to pre-war situation, and actual figures are remarkably deteriorated. Since a third of Croatian territory - including some oil- and gas fields - has been occupied (Fig. 1), and electric network of the country has not been established as a completed common system<sup>2</sup>, some parts of the country (above all Dalmatia) are suffering severe lack of energy, accompanied by periodical cut-offs in energy supply.

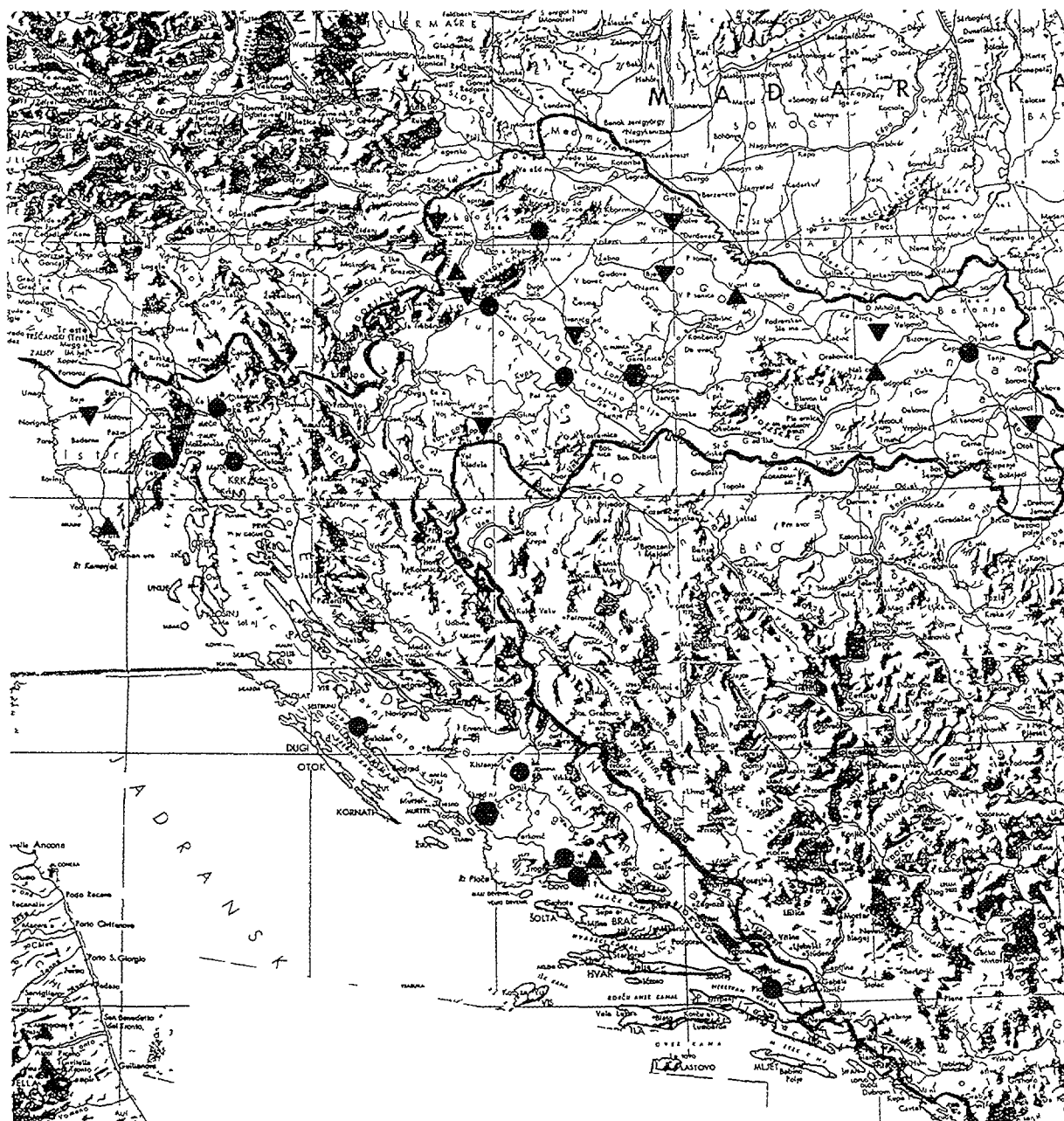
From the viewpoint of both natural radiation and hydrogeology, it seems useful to say that sedimentary rocks prevails in geologic structure of Croatia, occupying even 95 % of *surface lithology*. In addition, it should be pointed out the distribution of Mesozoic carbonate rocks (limestone, dolomite), prevailing throughout the southern part of the country (southward from Karlovac): morphology of this area is characterized by karst (Fig. 2), occurring in two types - shallow karst covered by soils, in interior parts of karst area, and deep, barren karst along the coast. Due to irregular hydrogeology and very sensitive mechanisms influencing the quality of surface- and groundwater, and controlling some specific bio-lithologic processes (eg generation of travertine), the area has been threatened by a number of human activities leading eventually to considerable environmental pollution. It must not be neglected the fact that a lot of hazardous industries and energy generating facilities have been located just in this area. Furthermore, it has been observed that soils ("terra rossa"), developed in places throughout the karst area are, owing to containing uranium, radium and thorium, characterized by considerably increased natural (background) radiation. The coal, mined in carbonate rocks (eg in Raša and Promina regions), has been also reported to have considerably increased natural radiation.

In the northern, Pannonian part of the country, fluvial, derasional and aeolian clastic sediments are dominant (in the lowland areas along the rivers Sava and Drava, and East Croatian Plain, in particular). Central Slavonic massifs (in north Croatia) are mainly composed by old, Palaeozoic igneous (granite, basalt etc.) and metamorphic rocks (crystalline schists, gneiss etc.) (Fig. 3).

The territory of Croatia is unstable with regard to both *tectonics and seismicity*. Few major longitudinal faults run north-west - south-east along both parts of the country, and places where they intersect transversal faults are often characterized by geothermal springs. Earthquakes as strong as

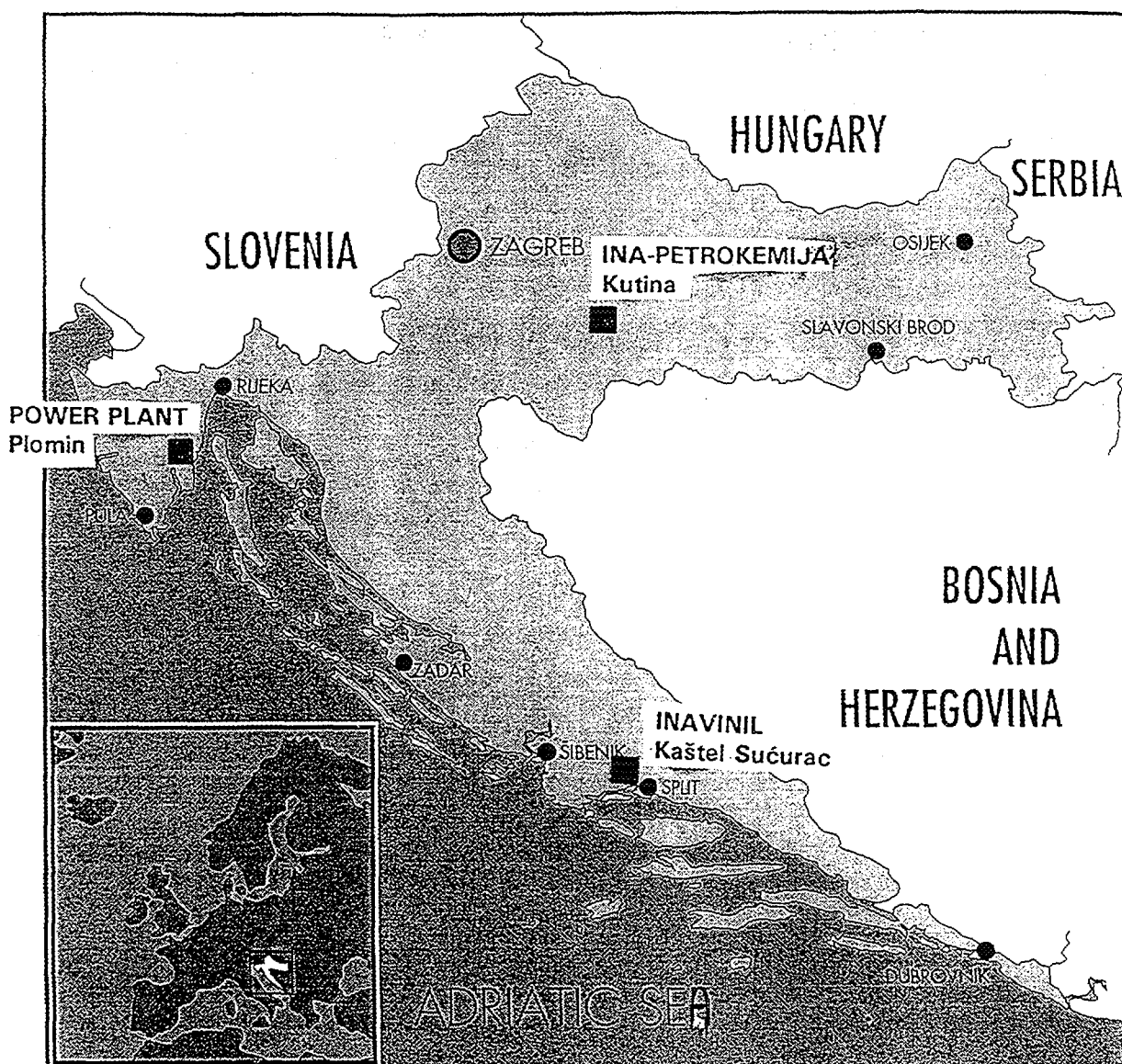
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<sup>2</sup> South parts of Croatia have been connected with the network in Bosnia & Herzegovina as a part of former Yugoslavia, but not directly with the northern part of Croatia itself.



- coal (mines, slag ash)
- ◆ phosphates (waste gypsum and fertilizers)
- ▲ cement factories and naturally radioactive materials
- ▼ geothermal springs oil and gas wells

Fig 2 Restoration of radioactively contaminated sites in Croatia



### GENERAL DATA ON THE REPUBLIC OF CROATIA

Population (the 1991 census)	4,760,000
Surface area	56,538 sq. km
Surface area of the territorial sea	31,000 sq. km
Length of the coastline, including islands	5,740 km
Number of islands (66 inhabited)	1,185

Fig. 3. Geographical position of high-priority sites

VIII-X MCS could be expected in major parts of Adriatic coast (in south Dalmatia, in particular), but they can also occur in some regions of north-western Croatia.

From the viewpoint of *natural radiation* it is worth mentioning that systematic measurement of background doses in Croatia was initiated by both the Institute "Ruđer Bošković" and Croatian Hydro-meteorologic Institute in 1986, immediately after the Chernobyl accident. The measurements have been performed at numerous meteorologic stations throughout Croatia by using thermoluminescent dosimeters (TLD), kept 1.5-2 m above the ground. The results of measurements, which were carried out during three months in 1988, are given in the following table (results are corrected for the values of corresponding transport doses):

<u>L o c a t i o n</u>	<u>Radiation dose (uSv)</u>
(A) PANNONIAN REGION	
Bilogora	888
Bjelovar	1,023
Daruvar	1,062
Gradište (at Županja)	1,071
Karlovac	876
Osijek	999
Puntijarka (Medvednica mount.)	816
Sisak	981
Slavonski Brod	987
Varaždin	1,074
Vinkovci	1,023
Zagreb (Grič)	1,038
(B) INTERIOR MOUNTAIN REGION (karst prevails)	
Gospić	1,071
Knin	810
Ogulin	1,116
Parg	1,029
Plitvice	1,239
Zavižan (Velebit mountain)	975
(C) COASTLAND (karst prevails)	
Dubrovnik	759
Lastovo (island)	636
Mali Lošinj (island)	756
Pazin (interior of Istria)	900
Ploče	771
Pula	918
Rab (island)	720
Rijeka	1,017
Senj	747
Split	729
Šibenik	729
Zadar	984

### 3. SCOPE OF WORK

In order to achieve all proposed goals by the IAEA, ie (a) to get a good knowledge of the affected areas; (b) to select best clean-up methods; (c) to select potential sites and transport routes; and (d) to understand the hydrogeological systems and select specific parameters of the area, it is necessary to have not only enough time, money and high-skilled experts but also well-harmonized facility building in the field of institutional responsibilities. As the first step in the project implementation in Croatia, it seems realistic to get reliable data on contaminated sites with regard to their position, history of contamination, quantities and origin of contaminated materials, their transportation routes, proximity to population centres, radioactive concentrations and spreading of contamination, methods to be used in further investigation and institutional responsibilities for cleanup and monitoring.

With regard to the above mentioned, we have completed the list of actions or tasks needed to be carried out in *the first phase* of the project implementation, ie "Identification and Characterization of Radioactively Contaminated Sites":

- |        |  |
|--------|--|
| TASK 1 | Completion of the list of all sites suspected to be radioactively contaminated, disregarding the kind of sites, ie material containing radioactive substances;   |
| TASK 2 | Preliminary estimation of radiological hazards at identified sites to the environment and human health, and prioritization of the restoration programme;   |
| TASK 3 | Identification of contamination history at selected sites, ie to find out origin, quantities and transportation routes of contaminated materials (and their activities, if possible from available data);                                      |
| TASK 4 | Preliminary classification of contaminated sites from the viewpoint of their proximity to population centres, radioactive concentrations and spreading of contamination (in extent allowed by available data);                                 |
| TASK 5 | Selection of techniques and methods related to sampling, radiometric surveys, laboratory analyses, statistical evaluation etc. to be implemented;  |
| TASK 6 | Conduction of radiometric surveying at high-priority sites and their radiologic characterization;  |
| TASK 7 | In accordance with results obtained by characterization, decision making on further remedial action (in forthcoming project phases) at each of involved sites (eg identification and securing the waste; removal the waste to safer site etc.) |
| TASK 8 | Evaluation and interpretation of results relevant for the project continuation and their issuing in the Progress Report, which is expected to be assessed by the experts of the IAEA.  |

It is supposed that the project can not be carried out without reliable data on the background radiation. These data are expected to be available in complete, simultaneously with the project performance.

The project implementation should be supported by institutional efforts to recognize bodies which will be responsible for both the economic and regulatory aspects of its continuation, as well as a monitoring programme of the follow-up process after the restoration of radioactively contaminated sites will be completed.

There has been identified a majority of contaminated sites in Croatia, which have been classified according the kind of contaminated materials. Distances to major population centres and transport routes of contaminated materials are in most cases also known. In addition, some aspect of preliminary prioritization of the involved sites has been carried out as well. However, available data are extensive and have to be properly evaluated by detailed field and laboratory surveying. Anyhow, some aspects of tasks (1), (2), (3) and (4) have been elaborated so far.

#### 4. PRESENT STATUS AT THE IDENTIFIED SITES

##### 4.1. Types of radioactively contaminated sites

There are four main groups of sites suspected to be radioactively contaminated in Croatia: (1) sites containing coal slag/ash piles; (2) sites containing phosphates and waste gypsum (ie phospho-gypsum) from fertilizers industry; (3) geothermal springs and oil/gas drillings; and (4) sites containing natural radioactive materials used in human activities (Fig. 4).

The following sites should be involved in the project:

##### A. Sites containing coal slag/ash piles

1. Coal-fired power plant Plomin
2. "INA-VINIL", the PVC synthesis and treatment plant, Kaštel Sućurac
3. Power-plant Zagreb (old slag piles)
4. Coal-fired power plant Jertovec
5. Coking coal storage at the port of Bakar

It should be also mentioned that some additional quantities of coal slag and ash are present at numerous firerooms of individual buildings, institutions (hospitals, schools) etc. Furthermore, some attention should be also payed to the old slag/ash piles remaining from use in railway transportation.

##### B. Sites containing phosphates and phospho-gypsum remaining from fertilizer industry

1. INA-PETROKEMIJA, fertilizers plant, Kutina
2. Port of Šibenik - import of phosphates

##### C. Geothermal springs and gas/oil drillings

1. Istarske Toplice
2. Topusko
3. Velika Ciglena

These *geothermal springs* (Velika Ciglena in particular) show that considerable attention should be payed also to *gas and oil drillings* where contamination of pipelines and separators by occasionally high-radioactive scale is possible (the problem is evaluated in more details in the chapter 4.1.3.). Therefore, it seems reasonable to accomplish preliminary measurements not only at the mentioned geothermal springs, but also at gas- and oil exploitation fields.

##### D. Sites containing natural radioactive materials

There have been measured increased natural radiation at some substances used in building materials industry (cement, bricks), as well as at substances composing ceramics (eg zircon-silicate, silicon sand etc.). Special attention in cement industry should be payed to cement admixtures (eg coal slag/ash, dross remained from coking etc.), which were reported to be considerably radioactive.

Hence, the performance of periodically repeating measurements in ceramics industries (INKER-Zaprešić), brickworks (eg. ZAGORKA-Krapina, Virovitica, Sladojevci etc.), but -above all- at cement plants (eg at Našice, Solin, Kaštel Sućurac, Pula, Koromačno, Umag and Omiš), is suggested.

Due to the supposed content of thorium in sludge generating in aluminium industry during the treatment of hydrated alumina (water is especially affected by contamination of radon), attention should be paid also to the Light Metals Plant at Šibenik.

#### 4.1.1. Sites containing coal slag/ash piles

There are several coal mines in Croatia sited in carbonate (karst) area of Dinarides. At almost all of them an increased natural radiation - due to considerable content of radionuclides as  $^{238}\text{U}$  and  $^{226}\text{Ra}$  - have been identified (above all at Raša/Labin mines in Istria, but also in mines situated in the interior of Dalmatia, like Širitovci and Dubravice). A lot of firerooms operating at numerous large buildings, schools and hospitals along the Adriatic coast, on the islands and in the interior of Dalmatia have been reported. Majority of them has been fuelled by coal. It is assumed that coal originates from nearby mines like Labin, Raša, Dubravice, Širitovci, Sinj and few mines in Herzegovina. Due to fairly high content of radionuclides participating in the uranium series, which are typical for coals sited in carbonates (eg Istria, "Promina" deposits, Herzegovina etc.), it is reasonable to expect considerably increased radioactivity in slag and ashes remained after use of the coal. That is why it seems necessary to identify at least the largest consumers of this coal, as well as to find out where and in what manner the remaining slag and ashes have been stored. Sampling and measurement of slag/ash radioactivity should be accomplished obligatorily. Further measures will depend on results of the analysis. The control of recent and old colliery tips at operating or closed coal mines does not seem necessary at the moment. Slag and ashes of the coal originating from mines in northern Croatia and northern Bosnia could be investigated later on, since no increased content of natural radionuclides is expected there. After available data, increased radioactivity has been reported in Slovenian coals (being in use also in Croatia) and they would have to be included in radiation measurements after the restoration of the above mentioned slag/ash piles will be completed.

After *our legislation* [1] there is a total activity threshold  $10^6 \text{ Bq/m}^3$ , separating radioactive materials and substances considered as non-radioactive.

Limit activities for materials used in construction of houses and other buildings where people live, are 5,000 Bq/kg for  $^{40}\text{K}$ , 300 Bq/kg for  $^{228}\text{Ra}$  ( $^{220}\text{Rn}$ - or "thoron" series), and 400 Bq/kg for  $^{226}\text{Ra}$  ( $^{222}\text{Rn}$ -series), but total activity from artificial sources is not allowed to be higher than 4,000 Bq/kg. In other words, the following relation should be respected (A - activity):

$$A(^{40}\text{K})/5,000 + A(\text{artificial sources}/4,000) + A(^{228}\text{Ra}) + A(^{226}\text{Ra}) < 1$$

##### 4.1.1.1. Kaštel Sućurac

There are two sites containing slag/ash piles with increased radioactivity identified at Kaštel Sućurac, 5 km northward from Split (200,000 inhabitants). The slag/ash has remained after the burning of coal used as energy source for the PVC synthesis and treatment plant INA-VINIL in Kaštel Sućurac. This radioactively "contaminated" coal originates from the Raša mine in Istria. It has been transported by the sea.

##### Case 1: INA-VINIL 1

The site, having dimensions 100 x 100 x 1 m, i.e. 10,000 m<sup>3</sup> of the stored coal slag, has been only partly organized. It is located indoor the INA-VINIL facility (former JUGOVINIL). The material has been covered by plastic sheets, and gravel- and soil layers above them. Slag and ashes have remained after the coal had been used as fuel during the 1950s. The material partly originates from



fireroom of the facility energy-plant, but a part of it has been shipped by the sea from Rijeka and Boka Kotorska. After available data it is realistic to suppose that coal from which the slag deposits have been derived, was mined mostly at minor mines in the karst area (carbonates!) of the Šibenik hinterland (Dubravice, Širitovci). Deposits shipped from Rijeka probably represent residue remained from combustion of coal mined at Labin and Raša. There is no reliable information on origin of deposits delivered from Boka Kotorska. Results of measurements of radionuclide content in three samples of this material, which were carried out in 1991, are shown in the Table 1.

Table 1

Coal slag and ashes stored at the "ina-vinil" facility  
(stored in the 1950s; in bq/kg)

S a m p l e	<sup>40</sup> K	<sup>228</sup> Ra	<sup>226</sup> Ra	<sup>238</sup> U	<sup>235</sup> U
Slag 1	184	24	799	2,830	130
Slag 2	105	26	6,195	18,640	858
Slag 3	148	18	166	4,690	216

Note: According to measurements of these slag samples the total specific alpha-radioactivity was estimated to be 35,000 Bq/kg, and total beta-radioactivity 29,000 Bq/kg (following the regulations of former Yugoslavia [1] being temporary applied in Croatia, the analyzed material is classified as low-active solid waste).

Some calculations show that there are some 7.5 tons <sup>238</sup>U, and about 55 kg <sup>235</sup>U contained in the dumped slag pile at the INA-VINIL plant.

Recommendations for further actions:

- (1) Due to insufficient data and a great variability of available results, it is necessary to carry out further sampling at the site (at grid density not lower than 20 x 20 m) in order to get reliable estimate of total radioactivity containing in the material.
- (2) Aiming to measure radiation doses and to compare them with the background radiation, it would be useful to set several dosimeters at the site itself, as well as somewhere in its immediate vicinity.
- (3) In accordance with results of dose measurements and more precise estimation of total radioactivity, and bearing in mind the fact of considerable repulsiveness of the public against the landfill sites, removal of the material could be assessed as the best possible solution (but it is not possible before the organized interim storage or final repository will be completed).

## Case 2: INA-VINIL 2

The site is located indoor the ex-JUGOVINIL facility area. It is completely non-organized, and partly attached to the above mentioned site (INA-VINIL 1). It is worth mentioning that a part of deposits (ie coal slag) has been dumped into the sea. The slag originates from the facility's energy-producing plant, where the coal from Dubravice was used during the 1980s, but the coal originating in Herzegovinian mines (probably the mine of Tušnica near Livno) has been preferred in last few years. The results of measurements, performed during late 1980s, are given in the Table 2. There is no information on radioactivity of slag and ash from coals used more recently.

Table 2 Coal slag and ashes stored at the "ina-vinil" plant /former jugovinil/ (samples have been taken in the period 1988-1990 from both the energy-producing facility and non-controlled pile; in Bq/kg)

S a m p l e	<sup>40</sup> K	<sup>228</sup> Ra	<sup>226</sup> Ra	<sup>238</sup> U	<sup>235</sup> U
1	275	57	1,942	3,493	161
2	259	61	2,035	3,804	175
3	213	73	1,991	3,590	165
4	230	63	2,157	3,902	180
5	456	71	2,080	3,445	159
6	308	75	1,752	3,211	148
7	331	47	2,102	3,591	165
8	291	76	1,782	2,847	131
9	218	62	1,799	2,952	136
10	358	92	2,316	NM	NM
11	275	53	2,203	NM	NM
12	118	23	NM	610	28
13	277	39	NM	1,088	50
14	241	41	NM	884	41
15	332	69	NM	1,442	66
16	197	30	NM	489	23
17	256	43	NM	1,171	54
Kaštel Lukšić*	147	16	233	513	24

NM - not measured

\* measured in the material accumulated beneath the floor, ie in the foundation of a family house

It seems that some quantities of the slag and ash were used in cement industry as admixture, in order to improve matrix quality of cement. The content of some radionuclides in cement and concrete, produced in Solin, is given in the Table 3 (measurements were accomplished during late 1980s). There were no additional measurements carried out during last few years.

Table 3

Content of radionuclides in cement and concrete produced at dalmacijacement industry (Bq/kg)

S a m p l e	<sup>40</sup> K	<sup>228</sup> Ra	<sup>226</sup> Ra	<sup>238</sup> U	<sup>235</sup> U
CONCRETE 1	51	5	40	NM	NM
CONCRETE 2	47	4	53	NM	NM
CONCRETE 3	59	4	72	NM	NM
CONCRETE 4	56	4	69	NM	NM
CONCRETE 5	50	4	78	NM	NM
CONCRETE 6	56	4	56	NM	NM
CEMENT 1	244	19	122	NM	NM
CEMENT 2	213	16	84	NM	NM
CEMENT 3	246	21	21	NM	NM

NM - not measured

Note: Concrete samples represent a mixture of cement with different shares of coal slag and ash, as well as limestone.

Cement samples contain various percentage of coal slag and ash.

Recommendations for further actions:

- (1) It is necessary to perform sampling and radiation measurements of coal being used at the moment, as well as at the slag and ashes landfills. Further measures will depend on obtained results.
- (2) Further sea-dumping of slag and ashes or their rejection anywhere into the environment (as it has been usually practised so far) must be immediately stopped, if no radiation measurements of the material are not previously performed.
- (3) Obligatory permanent radiation control of slag and ashes, used as admixture in cement industry, should be introduced.
- (4) There are some signs that slag and ashes from the site INA-VINIL 2 (i.e. former JUGOVINIL) were used for filling up the foundations of family houses in the area of Kaštela (see the site Kaštel Lukšić in the Table 2).

#### 4.1.1.2. Coal-fired power plant Plomin

Coal slag and ash, stored at the coal-fired power plant unit Plomin 1 come freely by wind and streams into environment (Table 4). There are not precise data on dimensions of the area covered by these materials at the power plant Plomin. Radioactivity of locally mined coals varies considerably, and mean content of uranium in ashes and slag might be somewhere around 2,000 Bq/kg (after bibliographic sources). Some results of natural radiation measurements (in marine and fluvial sediments) in vicinity of Raša coal mine and Plomin power-plant, as well as in ashes generating in the power-plant Plomin 1, are presented in the Table 4. It should be added that besides the stored slag and ash piles, there is another source of pollution acting in the power plant: release of gas and contaminated smoke into the atmosphere.

Recommendations for further actions:

- (1) Precise dimensions of storing area and quantities of stored material should be identified.
- (2) Due to great variations of radionuclide content, it is necessary to take sufficient number of samples and estimate the total activity of analyzed materials.
- (3) Dosimeters should be set up at the suspected areas in the same way as it is suggested in the Kaštela region (INA-VINIL).
- (4) Radioactive contamination, influenced by radionuclides released in the atmosphere by the power-plant Plomin, should be evaluated through surveying of  $^{226}\text{Ra}$  concentrations in moss.
- (5) In accordance with some recent radiation contamination surveys, it seems to be useful to carry out radiation measurements of marine sediments in the bay of Plomin (they are reasonably assumed to be radioactively contaminated).
- (6) In perspective, carry out the restoration of the storing area.

#### 4.1.2. *Sites containing phosphates and waste gypsum remaining from fertilizers industry*

##### 4.1.2.1. Fertilizer factory INA-PETROKEMIJA in Kutina

The storage of waste gypsum ("phospho-gypsum"), remained after production of fertilizers in INA-PETROKEMIJA facility, is organized some 5 km southward of the facility site. Phospho-gypsum has been transported to the storage by special pipeline. There are some indicators that

Table 4

Radioactivity measured in stream sediments, ash and slag in istria (in bq/kg)

S i t e	<sup>40</sup> K	<sup>228</sup> Ra	<sup>226</sup> Ra	<sup>238</sup> U	<sup>137</sup> Cs
FRACTION < 0.063 mm					
(a) <u>stream sediments</u>					
Raša (bridge)	680	28	26	18	27
Potpican (bridge)	471	38	46	30	8
Glogorički potok	516	27	31	35	HD
Floričiči (cascade)	497	21	22	24	14
Raša 1	571	49	39	32	HD
Raša 2	674	47	39	36	HD
(b) <u>marine sediments</u> <u>of Raša bay</u>					
Trget (- 12 m)	464	23	22	38	25
Sv. Mikula (-36 m)	470	21	19	45	14
Sočaja (-40 m)		534	26	33	43 12
Plomin Power Plant (ash)	206	51	1,020	NM	NM
Plomin ("soil")	30	93	683	748	3
Tupljak (coal)	58	6	86	77	HD
FRACTION < 0.5 mm					
(a) <u>stream sediments</u>					
Rabac (tunnel)	60	15	272	181	2
Plominski potok	239	22	428	313	11
Raša potok (Raša)	325	31	110	102	62
Raša (Pićan), bridge	309	28	55	46	6
Rušanski potok I	42	5	10	10	6
Rušanski potok II	328	20	15	13	34
Karbunski potok	301	19	18	21	14
Pedrovica potok	255	15	14	33	23
Raša (mouth), bridge	318	24	27	21	16
Vlaški potok	310	26	22	37	8
Rušanski p. (mouth)	234	16	16	17	5
Boljunčica (Šušnj.)	339	26	21	15	23
Studena (mouth)	114	8	17	10	4
Rušanski p. (Boljun)	347	22	28	31	8

HD - hardly detectible (i.e. the value is nearly 0)

NM - not measured

measurements of radioisotopes in surrounding groundwater were carried out, but no data are available at the moment. Radioactivity of particular components (ie radioisotopes) in all stages of fertilizer production, from raw materials until final products, is given in the tables 5-8.

Table 5

Radioactivity of raw materials (in bq/kg); sampled in 1988

Raw material (origin)	<sup>40</sup> K	<sup>228</sup> Ra	<sup>226</sup> Ra	<sup>238</sup> U	<sup>235</sup> U
Potassium salt 1 (ex-USSR)	15,780	NM	NM	NM	NM
Potassium salt 2 (ex-USSR)	15,890	NM	NM	NM	NM
Potassium salt 3 (ex-USSR)	15,320	NM	NM	NM	NM
Potassium salt 4 (ex-USSR)	15,090	NM	NM	NM	NM
Potassium salt 5 (ex-GDR)	16,400	NM	NM	NM	NM
Potassium salt 6 (ex-GDR)	16,380	NM	NM	NM	NM
Dolomite filler	< 28	< 4	15	26	1
Phosphate 1 (Morocco)	42	12	1,359	2,642	122
Phosphate 2 (Morocco)	43	11	1,359	2,638	122
Phosphate 3 (Morocco)	31	15	1,254	2,446	113
Phosphate 4 (Senegal)	51	10	1,093	1,919	89
Phosphate 5 (Senegal)	51	9	1,086	1,956	91
Phosphate 6 (Senegal)	53	12	1,129	1,996	92

NM - not measured

Table 6

Radioactivity of the phosphate acid and mono-ammonium phosphate /map/ (in bq/kg); sampled in 1988

S a m p l e	<sup>40</sup> K	<sup>228</sup> Ra	<sup>226</sup> Ra	<sup>238</sup> U	<sup>235</sup> U
Phosphate acid 1	24	3	6	3,020	140
Phosphate acid 2	23	3	< 2	2,856	132
M A P 1	103	10	23	3,215	149
M A P 2	58	3	8	3,217	149
M A P 3	34	3	12	3,146	146
M A P 4	56	11	26	3,208	149
M A P 5	25	3	19	3,144	146

Table 7

Radioactivity of waste gypsum (phospho-gypsum) (in bq/kg); sampled in 1988

S a m p l e	<sup>40</sup> K	<sup>228</sup> Ra	<sup>226</sup> Ra	<sup>238</sup> U	<sup>235</sup> U
Gypsum 1	122	19	708	576	27
Gypsum 2	90	25	624	545	25
Gypsum 3	148	17	733	566	26

Table 8

Radioactivity of phosphate fertilizers (final products)  
(in bq/kg); sampled in 1988

S a m p l e / t y p e	<sup>40</sup> K	<sup>228</sup> Ra	<sup>226</sup> Ra	<sup>238</sup> U	<sup>235</sup> U
TRIPLEX 1	49	23	212	1,070	50
TRIPLEX 2	52	22	218	1,059	49
N-P-K (10-30-20) 1	5,530	3	6	1,710	79
N-P-K (10-30-20) 2	6,816	5	5	1,549	72
N-P-K (10-30-20) 3	6,880	5	5	1,486	69
N-P-K (8-26-26) 1	5,648	7	9	1,502	70
N-P-K (8-26-26) 2	7,081	5	4	1,162	54
N-P-K (8-26-26) 3	7,088	5	6	1,130	54
N-P-K (10-20-30) 1	7,106	7	9	1,163	54
N-P-K (16-16-16) 1	3,973	6	83	842	39
N-P-K (16-16-16) 2	3,840	10	84	805	37
N-P-K (16-16-16) 3	3,334	4	105	764	35
N-P-K (14-14-14) 1	3,683	4	5	874	40
N-P-K (14-14-14) 2	3,971	9	7	884	41
N-P-K (14-14-14) 3	3,741	6	6	805	37
N-P-K (13-10-12) 1	3,083	4	226	774	36
N-P-K (13-10-12) 2	2,952	5	207	816	8

After the study of radioactivity in the phosphate fertilizers and waters of the Kanovci area in eastern Slavonia, north Croatia [2], there were measured the following values for annual deposition of uranium and radium in soils: 4.5 Bq/m<sup>2</sup> for <sup>226</sup>Ra, 0.5 Bq/m<sup>2</sup> for <sup>228</sup>Ra, 3.1 Bq/m<sup>2</sup> for <sup>235</sup>U, and 67 Bq/m<sup>2</sup> for <sup>238</sup>U. The greatest concentrations of both radium isotopes were measured in water from drainage channels, having mean value of 120 Bq/m<sup>3</sup> for <sup>238</sup>U, and 5.5 Bq/m<sup>3</sup> for <sup>235</sup>U.

Recommendations for further actions:

- (1) It is known that minor shares of uranium and almost all <sup>226</sup>Ra participating in phosphates, have been separated during process of fertilizer production, into waste gypsum. On the other hand, most of uranium remains in fertilizers and comes in this way into environment, polluting waters in particular. For this reason, it would have introduce modifications in the process of production, resulting in separation of uranium itself from the fertilizers (eg in the processing stage of phosphate acid production).
- (2) Radioactivity of both the water in the vicinity of gypsum landfill, and raw phosphates, should be periodically controlled.
- (3) Attention should be payed also to the separation vessel in the fertilizer plant, since the material accumulating there could be contaminated by uranium, radium and radon daughters.

#### 4.1.2.2. The port of Šibenik

The port of Šibenik as the site where imported phosphates have been reloaded in railway to be transported to Kutina, should be carefully inspected. Due to war in the region of Šibenik (and along broader area in hinterland) the import of phosphates has been stopped, and the port of Rijeka was chosen as reloading point of phosphates. There are no data on possible pollution by phosphates in Rijeka so far.

#### 4.1.3. Geothermal springs & oil/gas drillings

It is known that separation of uranium and thorium occurs during crystallization of magma. Therefore, considerable concentrations of these radionuclides and their decaying series are found in acid igneous rocks and hydrothermal formations. In addition, it was found an increased level of natural radiation in geothermal waters and sediments accumulated by them (in fact, these materials are not contaminated, since they are naturally radioactive). The results of radionuclide content in some natural geothermal waters are presented in the Table 9, and composition of radionuclides contained in geothermal waters (often high-mineralized) at some of investigating drillings, is given in the Table 10.

The radioactivity of oil and gas themselves has not been determined so far. Nevertheless, since radon is released in the atmosphere during combustion of gas, it is reasonable to screen radioactivity at these drillings.

Table 9

Radionuclide content in natural geothermal water (bq/m<sup>3</sup>)

Site (borehole)	<sup>40</sup> K	<sup>228</sup> Ra	<sup>226</sup> Ra	<sup>238</sup> U	<sup>235</sup> U
Istarske toplice (spa)	418	43	1,750	1,340	62
Istarske toplice ("grotlo")	12	69	3,180	2,610	120
Topusko (spa)	454	42	360	550	25
Thermal spring (Požega)	278	46	75	170	8
Varaždinske toplice (spa)	829	4	NM	22	1

NM - not measured

Table 10

Radionuclide content in geothermal water of boreholes (bq/m<sup>3</sup>)

Site (borehole)	<sup>40</sup> K	<sup>228</sup> Ra	<sup>226</sup> Ra	<sup>238</sup> U	<sup>235</sup> U
Zagreb ("Mladost")	1,060	305	400	1,340	64
Podsused (INA)	131	9	54	40	2
V. Ciglana 1A (INA)	11,260	515	7,080	6,550	302
Istarske toplice (borehole)	19	100	2,700	1,670	77
Podsused (INA)	492	51	89	273	13
Borehole 7215 (INA)	368	16	115	193	9
Kumrovec (INA)	196	18	31	77	3
KBCNZ-1B (INA)	1,226	272	NM	1,281	59
SU-3 (INA)	342	42	NM	107	1
Scale (INA)*	511	530	878	2,090	96

NM - not measured

\* scale in separator (Bq/kg)

However, another type of radioactive contamination, possible to occur at gas- and oil drillings should be also discussed. During gas- and oil exploitation, the parameters (pH, pressure, temperature, etc.) of the "in situ" stable stratum-water could be disturbed, generating "scale" which is reported to be occasionally high-radioactive. The scale, including often corrosion products, paraffins and silicates, is collected in pipelines, drilling pipes and separators at the oil- and gas fields. As an illustration,

there have been measured dose rates higher than 0.1 Gy/h (Smith A.L., 1987) [3] on the surface of pipelines in cases of the high-radioactive scale (i.e.  $10^6$  Gy/h higher than natural gamma dose rates). There are no data on this phenomenon, measured in Croatia so far. Nevertheless, in order to find out the situation at gas- and oil drillings, and - if needed - to improve protection measures of operating workers, the project team is going to organize and start the scale sampling at the boreholes in Croatia.

Recommendations for further actions:

- (1) Sampling and measuring of drilling scale at gas- and oil exploitation fields should be started, and a long-term co-operation with people from the INA-NAFTAPLIN oil company should be set up in order to carry out occasional or periodical sampling of materials at oil- and gas drillings, suspected to be considerably radioactive.
- (2) As the most serious problem in radioactive scale stands the content of  $^{226}\text{Ra}$  (in less extent also  $^{228}\text{Ra}$ ) and its decaying products. Further actions will depend on results of radium measurements.
- (3) As the highest concentrations of uranium are expected in heavy oil distillation fractions (ie asphalt and bitumen), they should be sampled in order to determine the uranium activity. Further actions will depend on results of the analyses.

#### 4.1.4. Natural radioactive materials used in human activities

Some materials, used in industry, contain high-level natural radiation. Therefore, their use is limited by legislation. In the Table 11 there are presented some of them, being used by producers of ceramics (natural radioactivity of quartz sands and bricks are given for comparison). Materials used as admixtures in cement industry are given in the Table 12.

Table 11

Radioactivity of some natural materials used in industry (Bq/kg)

Site / material	$^{40}\text{K}$	$^{228}\text{Ra}$	$^{226}\text{Ra}$	$^{238}\text{U}$	$^{235}\text{U}$
Zirconium silicate (INKER)	32	513	3,360	NM	NM
Kreutzonit (INKER)	39	480	3,375	NM	NM
Macino (INKER)	147	473	2,660	NM	NM
Silicon sand (Virovitica)	88	12	16	29	1
Brick (Zagorka)	570	52	52	92	4

NM - not measured

Table 12

Radioactivity of some waste materials used in industry (in Bq/kg)

Site / material	$^{40}\text{K}$	$^{228}\text{Ra}$	$^{226}\text{Ra}$	$^{238}\text{U}$	$^{235}\text{U}$
Dross (import from Austria)	437	50	85	145	7
White slag (JUCEMA)	14	20	62	87	4
Dross (import from Italy)	133	33	100	NM	NM
Red mud (Mostar)	11	290	116	273	13
Slag (JUCEMA)	81	28	20	26	1

NM - not measured



Comments and recommendations for further actions:

- (1) There are not expected cases of considerable environmental pollution in the industry of building materials, which would be caused by increased natural radioactivity of these materials.
- (2) The control of final products seems to be satisfying at present (in terms of check whether their radioactivity does not surpass the limits defined by the law).
- (3) The present legislation in the field (taken from the regulations of former Yugoslavia) seems to be too mild. Some modifications resulting in more stringent law are expected to be more convenient.

## 5. RESOURCE MANAGEMENT

### 5.1. Project personnel

Due to lack of experience in the field of restoration of radioactively contaminated sites, Croatia appreciates this possibility to take advantage of the hard-won experience gained in other countries, participating in the Technical Co-operation Regional Project for Central and Eastern Europe on Environmental Restoration. From its own side, Croatia is going to make possible necessary technical assistance through both the training and direct assistance in the field, within the frames of its realistic possibilities. Anyhow, we expect to get an expert consultancy in the project implementation from the IAEA, as well as from the other international organizations close to the project like the Commission of the European Communities (CEC) and the Organization for Economic Co-operation and Development/Nuclear Energy Agency (OECD/NEA).

Besides institutions authorized to participate in the project implementation in Croatia, there are few governmental institutions (i.e. ministries) supporting the project. In short, the organizational scheme of the institutions taking part in the project performance in Croatia is as follows:

- (1) Project Co-ordinator:

Hazardous Waste Management Agency

- (2) Authorized Expert Institutions:

Institute "Ruđer Bošković"  
Institute of Medical Research and Occupational Health  
ECOTEC - Company authorized for handling radioactive materials  
Mining Institute  
Institute of Electric Power Industry and Energy  
Water Economy Board

- (3) Governmental Institutions:

Ministry of Civil Engineering and Environmental Protection  
Ministry of Economy - Department of Energy and Industry  
Ministry of Health  
Ministry of Finance  
Ministry of the Interior  
Ministry of Defence  
Mining Inspectorate

Project Leading Team is composed of representatives of Hazardous Waste Management Agency (D. Subašić, A. Šaler) and the Institute "Ruđer Bošković" (D. Barišić, B. Vekić, S. Lulić).

The list of the project participants is allowed to be modified during the project performance in order to make its implementation as successful as possible.

## **5.2. Methods to be applied and equipment needed for project performance**

### **5.2.1. Methods**

It is recommended to evaluate the preliminary status of radioactively suspected sites by performance of reliable measuring of dose rates at each selected site. The most expeditious collecting of reliable data can be performed by dose measurement using calibrated instrument which satisfies secondary standard category, or by instrument, calibrated according to the secondary standard, being able to measure low doses of natural radiation. Sites at which increased radiation doses are expected to be evaluated, should be additionally surveyed by: (a) reliable long-term measurement of radiation doses by thermo-luminescent dosimeters (TLDs); (b) repeated sampling at same sites and measurement of samples activity by using reliable laboratory methods, as eg gamma spectrometry (Ge/Li- and/or HP Ge semi-conductor detector). The radiation dose estimate could be also performed by the method described in (b), and then compared with measurements given in (a), as well as with preliminary dose rate measurements.

Preliminary measurements, sampling and set up of TLDs at the sites suspected to be radioactively contaminated, are supposed to be carried out by the same team, following the accepted strict criteria. It is assumed the complete equipment to be calibrated, and correlation of measurements is needed if the analyses will be carried out in different laboratories.

### **5.2.2. Equipment**

In order to accomplish all the mentioned measurements in a proper way, the following missing equipment should be available:

- (1) Satisfactory calibrated portable instrument (provided also with battery unit) for measuring low radiation doses within the secondary standard range (eg device H7907-1, produced in Halle, Germany) - 1 unit.
- (2) Satisfactory calibrated portable instrument (provided also with battery unit) for measuring low radiation doses within the secondary standard range (eg NaJ(Tl) scintillation detector) - 2 units.
- (3) TLD-700 - 1,000 units  
CaF<sub>2</sub>Mn TLD - 1,000 units
- (4) Multi-purpose gamma-monitor MFM-202, measuring radiation doses in the range 50 nSv/h - 100 mSv/h; measures continuously, identifies and print out the measured radiation doses - 3 units.
- (5) Portable detector (monitor) for measuring of radon - 2 units
- (6) Equipment for laboratory measuring of radon - 1 set.

## **6. PROJECT SCHEDULING**

Croatia is going to follow the time-schedule, proposed by the project leading co-ordinator, ie to carry out three phases as three annual sub-projects in the period 1993-1995. However, it does not seem realistic the project to be fully completed in this period; not only for the reason of later incorporation of Croatia into the project, but much more due to difficulties facing our country in both fields of state economy and war running in some regions of the country for last two years.

Hence, the preliminary plan regarding the more realistic deadlines (in accordance with our possibilities) for the project activities in the first phase, is as follows:

TASK 1	October	1993
TASK 2	November	1993
TASK 3	December	1993
TASK 4	December	1993
TASK 5	December	1993
TASK 6	April	1994
TASK 7	May	1994
TASK 8	June	1994

## 7. PROJECT CONTROL

From the Croatian side, the project supervision and verification of interim (phase) results will be carried out by both the Ministry of Civil Engineering and Environmental Protection, and the Ministry of Health. *An overall project control body is expected to be the IAEA*, along with co-operating international institutions (CEC, OECD/NEA), which figure as the main expert and financial supporters of the project. Along with this, Croatian Co-ordinating Team (CCT) is going to stay in permanent contact with the IAEA, informing it continuously on the project realization.

## REFERENCES

- [1] THE LEGISLATION OF THE REPUBLIC OF CROATIA, "The Law on Ionizing Radiation Protection and Special Safety Measures Related to Use of Atomic Energy", Official Gazette of Yugoslavia 62/1984 (accepted as temporary applied law in the Republic of Croatia until the issuing of the new Croatian legislation in the field), Zagreb (1991).
- [2] BARIŠIĆ, D., LULIĆ, S., MILETIĆ, P., "Radium and Uranium in Phosphate Fertilizers and their Impact on the Radioactivity of Waters", Water Resources, Vol. 26, United Kingdom (1992), 697-706.
- [3] SMITH, A.L., "Radioactive Scale Formation", Journal of Petroleum Technology 39 (1987) 607-706.

# ENVIRONMENTAL RESTORATION OF URANIUM MINES AND MILLS IN THE CZECH REPUBLIC

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## Abstract

A great number of uranium mines and mills, a long-termed history of uranium industry, a lack of environmental concern in the socialist period and application of both mining and in situ leaching method are the most important causes of great environmental restoration problems. The most complicated situation is on the ore deposit Stráž pod Ralskem - Hamr in Northern Bohemia. The paper describes principal results of the complex investigation performed as a basis for the environmental restoration programme in this area.

## 1. INTRODUCTION

Problems of environmental restoration of uranium mines and mills are complicated for several reasons:

1. The uranium mining has taken place in several parts of our country. The most important regions were: (Fig. 1)

- Western Bohemia (Jáchymov, Horní Slavkov, Zadní Chodov, Vítkov)
- Central Bohemia (Příbram)
- Western Moravia (Dolní Rožínka, Rožná, Jasenice, Pucov)
- Northern Bohemia (Stráž pod Ralskem - Hamr)

The number of uranium mines was more than one hundred, including small shallow mines for investigation.

2. There are three uranium mills:

- MAPE (Southern Bohemia, near České Budějovice)
- DIAMO (Western Moravia, Dolní Rožínka)
- Stráž pod Ralskem (Northern Bohemia).

The uranium mill MAPE is already out of operation. There are several tailings next to every uranium mill.

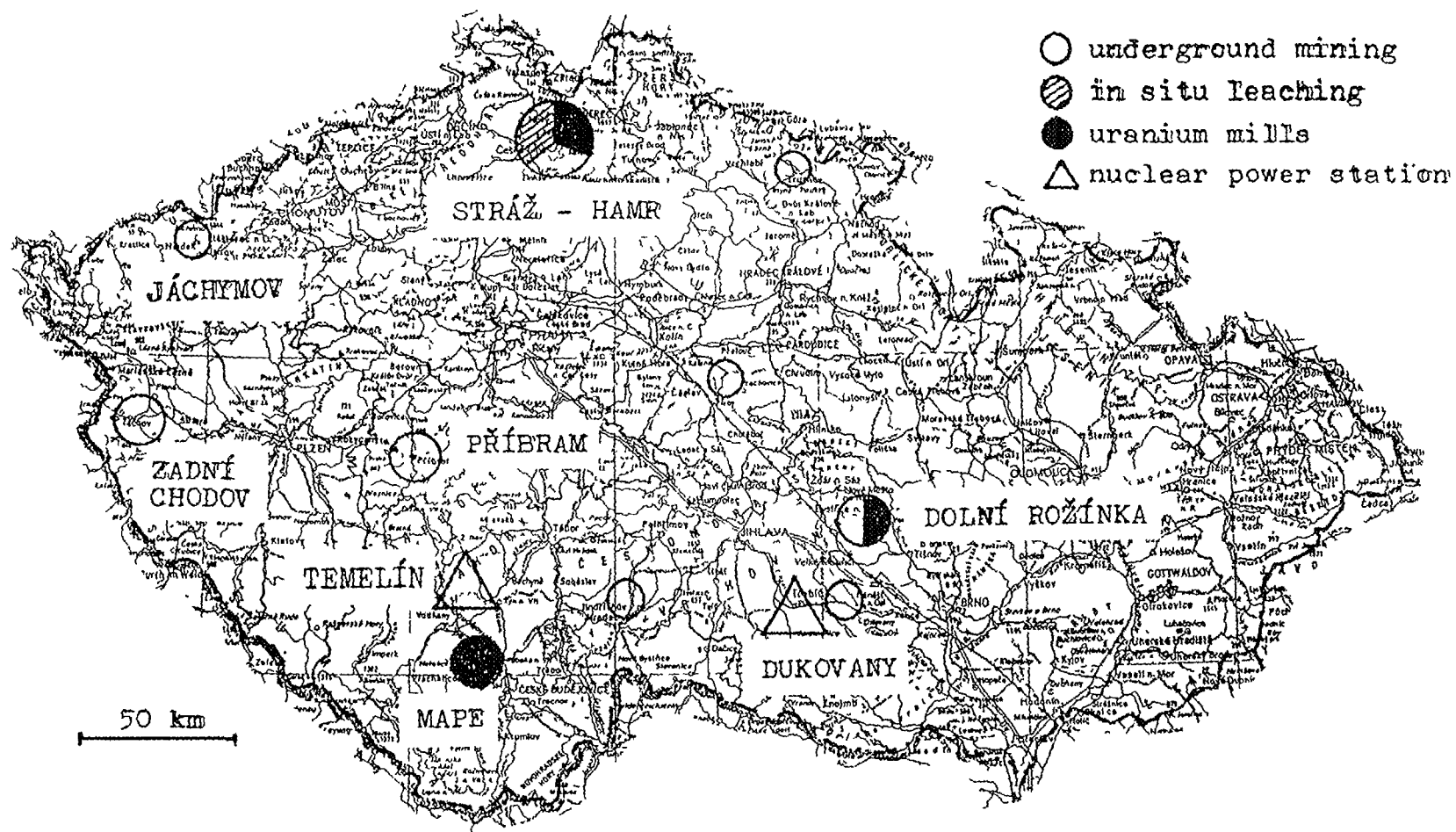


Fig. 1: Localities of the uranium industry

3. Two methods for uranium production were used:

- the mining and milling
- in situ leaching

Both the methods have different influences on environment.

4. The production of uranium has a long history in our country. The began after World War II and typical for the socialist economy mainly in the 50-ies and 60-ies was a lack of environmental concern. The serious ecological problems risen have to be solved now.

The in situ leaching is in operation on deposit Stráž pod Ralskem. It is the largest open ore deposit in the Czech Republic and both methods are used simultaneously. The region is characterized by complicated and for mining unfavourable hydrogeological and hydrobiological conditions and is situated in a drinking water reservoir area. The problems of environmental protection in this ore deposit are much more complicated and relevant than those in other localities of the Czech uranium industry. Therefore this paper will deal with deposit Stráž pod Ralskem - Hamr.

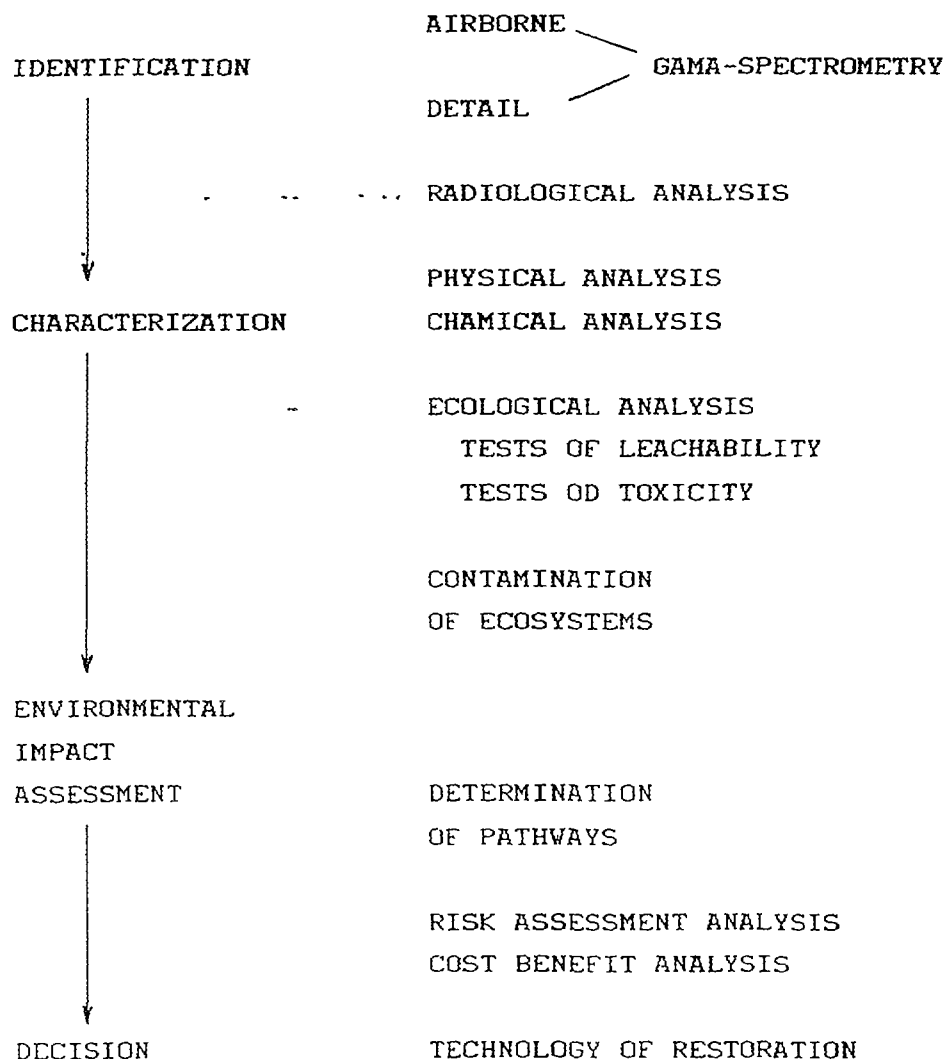
The investigation of the deposit Stráž p. R. - Hamr is very good example of the procedure of identification and radiological characterization of contaminated sites. (Fig. 2, 3).

The decisive part of the results presented are from the great at investigation programm which was compiled in MEGA - Institute for Research and Development at the request of Federal Committee for Environment in 1991. /1/

## 2. CHARACTERISTICS OF THE REGION

From the standpoint of geology the region forms the northwestern part of the Czech Cretaceous Plateau. The ore mineralization is located in sedimentary sandy rocks of the Cenomanian Age (about 200 metres below surface). The association of the element is unusual: U - Zr - P - Ti.

The hydrogeology of the region is very complicated. There exist two basic aquifers: the Cenomanian, the Turonian. The Cenomanian aquifer is tied up with sandstones of sea and freshwater sediments and in the ore deposit there occur artesian confined waters. The general direction of the flow is from northeast to southwest. The Turonian aquifer lies above the Cenomanian one separated from it by a stratum of poorly permeable clayey and limey silt - stones up to 100 metres thick - and represents a significant reservoir of high quality drinking water (integral part of a protected region of natural groundwater accumulation North Bohemia cretaceous plateau, pursuant to Regulation Nr 85/1981 LG of the Czech Government).



**Fig. 2: Identification and radiological characterization of contaminated sites**

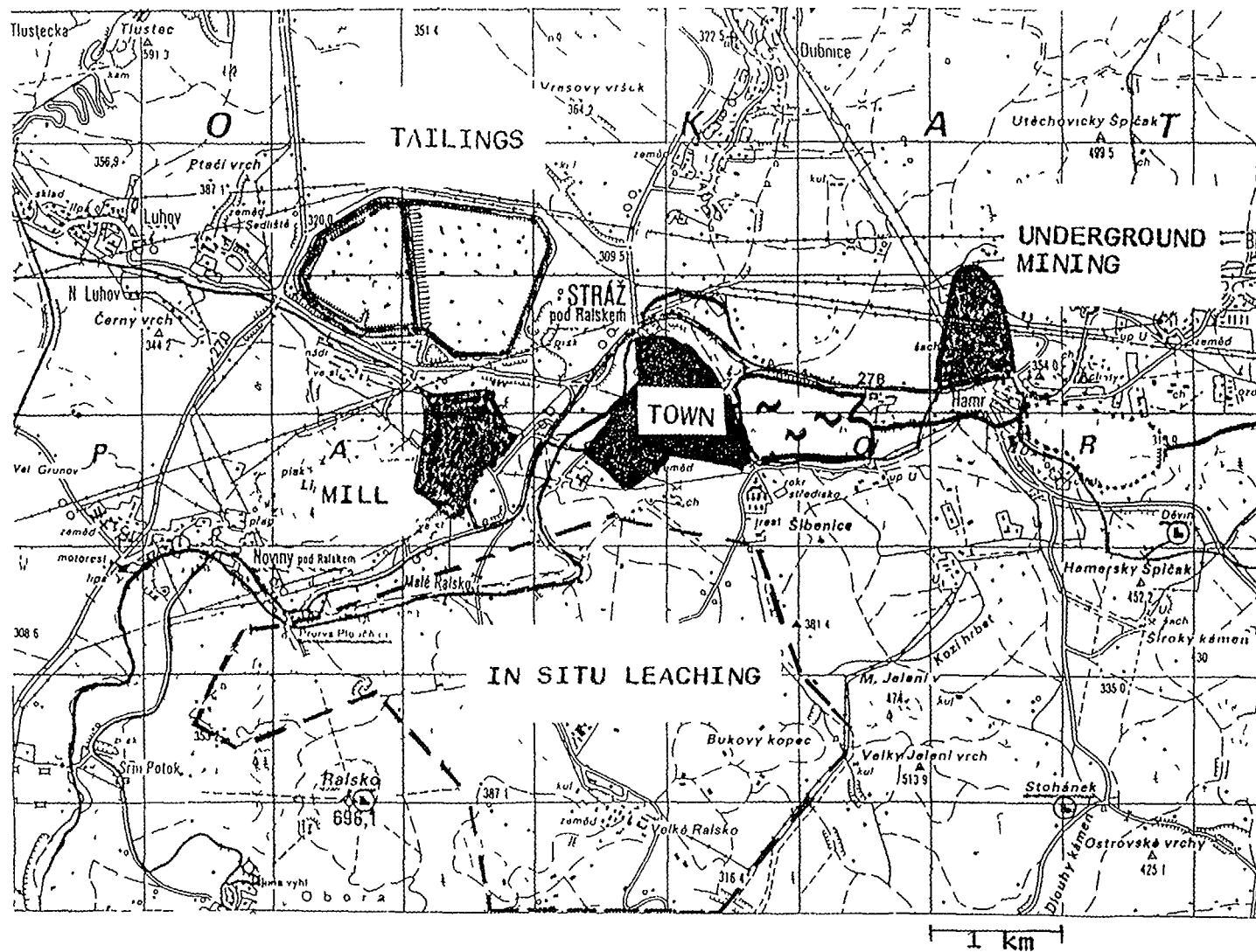


Fig. 3: Deposit Stráž pod Ralskem - Hamr



### 3. MINING AND MILLING

Mining in the concept of environmental protection comprises all activities beginning with the extraction of ore to the final product manufacture (ammonium diuranate), ore deposition, pit airing, pit drainage, filling, transport route, chemical ore processing and leached ore deposition. (Fig. 4)

Deads are deposited in dump-piles within a total area of about 130 thous. square metres and a total volume of 1 300 thous cubic metres. Basic environment influencing factors are: negative aesthetic impact on landscape, potential source of radioactive contamination due to dust and Rn emissions, danger of underground and surface water contamination.

Pit airing is the source of Rn emissions (about 30 per cent of all technological emissions of Rn) and of exhaust gases of pit machinery.

#### underground mining → environment

1 t U

= 1 100 t ore in the tailings

= 130 m<sup>3</sup> deads in dump-piles

= 11 GBq Ra in the tailings

= 20 000 m<sup>3</sup> waste mining water

= 520 m<sup>3</sup> contaminated groundwater

= 34 m<sup>3</sup> contaminated soils and sediments

= 120 GBq Rn released in the air

= 1,8 t SO<sub>2</sub>- released in the air

= 0,9 t NO<sub>x</sub> released in the air

Fig. 4: Environmental impact of underground mining

The mining is being made in aquiferous sandstones. To drain the pit about 15 million cubic metres of pit water per year is pumped and after processing discharged in the river Ploučnice, which is about 40 per cent of the average flow of Ploučnice in this area.

The worked-out pit space is filled with solidifying cement mix, so that no downthrow faults occur.

The transport route comprises all ore handling, from the extraction to the surface to the input in the chemical processing plant. This is a significant source of environmental contamination with the ore, mainly with its fine particles which contain the greatest concentration of radionuclides and heavy metals. The ore is transported over public highways in open lorries with no additional treatment.

The chemical ore processing in the chemical processing plant influences the environment with emissions, mainly of  $\text{SO}_2$ , aerosols of  $\text{H}_2\text{SO}_4$ , and Rn.

After neutralization the leached ore is deposited in a tailing, the area of which is in the 1st phase 165.6 hectares, in the 2nd phase 166.0 hectares. Water infiltration of the tailing estimated at 28 litres per second (i.e. 884 thous. cubic metres per year, which is 53 per cent of the input balanced) has caused an extensive contamination of groundwater in the accumulation, reaching 5.3 million cubic metres with sulphate content of 5 800 tons. The head of the contamination proceeds toward the river Ploučnice at a speed of about 40 metres per year and according to a hydrogeological model the infiltration in the river Ploučnice will occur around the year 2 011 with the result that the concentration of sulphates would grow by more than 200 mg per litre if no (correstive) technological measures are taken.

The attitudes towards and projects to deal with the tailing which do not warrant long-term stability and harmlessness for the surroundings have to be revised and the practice of western countries should be adopted.

Mining influences both the landscape and its environmental stability - the abolition of the Sedlišť Lake, temporary draining of Hamry Lake, canalization of the Ploučnie River (which caused the liquidation of a unique landscape whole), calling into existence ruderal areas around buildings and plants, dump and refuse piles.

#### 4. IN SITU LEACHING

The "in situ leaching" (ISL) technology of the uranium recovery began in the ore deposit Stráž pod Ralskem in 1968 and lasts till today. (Fig. 5)

Foreign literature of the ecology of underground leaching shows:

- departure from the use of sulphuric acid and ammonium hydrogencarbonate as leaching agents, mainly for reasons of the difficult regeneration of the underground after leaching

#### in situ leaching → environment

1 t U

= 7 260 m<sup>3</sup> contamin. water in cenoman

= 1 500 m<sup>3</sup> contamin. water in turon

= 274 t SO<sub>4</sub><sup>2-</sup> injected into groundwater

= 7,9 t NH<sub>4</sub><sup>+</sup> injected into groundwater

= 53 GBq Rn released in the air

= 0,95 t SO<sub>4</sub><sup>2-</sup> released in the air

= 1,18 t NO<sub>x</sub> released in the air

Fig. 5: Environmental impact of the in situ leaching

- the use mainly of sodium hydrogencarbonate or only a solution of  $\text{CO}_2$ . The regeneration of the underground is oriented towards achieving such a composition of the water after leaching that would most closely resemble the original composition, and sometimes lower values than established by the drinking water standard

- the concentration of leaching agents:

.  $\text{H}_2\text{SO}_4$ ....2-10 g/l,  $\text{NH}_4\text{HCO}_3$ ....1-5 g/l,  $\text{NaHCO}_3$ ....1-10 g/l.

The most preferred leaching agent in western countries today is a solution of several tablets of sodium bicarbonate per litre of water

- the leaching period to achieve 60 - 80 per cent yield is 2 to 5 years

- disposal of liquid waste:

. vaporization in natural setting (lakes, ponds)

. RO concentration or electrodialytic concentration (not so common) and injection of concentrated solutions into deep horizons.

Parametres of the technology used in the ore deposit Stráž pod Ralskem are basially different:

-  $\text{H}_2\text{SO}_4$  concentration .....50 - 70 g/l

- leaching period (for the 60 - 80 % yield) .....15 - 20 years.

This is the result of the natural conditions of the ore deposit, namely because the ore is not easily leachable and because of the uranium occurrence in impermeable locations. Generally such conditions are considered inappropriate for underground leaching.

Description of the underground leaching technology:

A sulphuric acid solution with a given content of nitrates - the leaching agent is injected into the productive horizon by means of injection bores. The flow over the ore leaches the uranium content and forms a solution. A system of mining bores allows to pump uranium solution to the surface. The uranium in solution is

sorbed on ionex and at the same time nitrates from ionex pass into the solution. The ionex sorbed uranium is then eluted by a solution of nitric acid. Out of the eluate the uranium is precipitated by gaseous ammonium to ammonium diuranate. The separation of the precipitate results in the s. c. yellow cake and mother liquor. The whole area of leaching fields is about 630 hectares, total amount of technological bores is approximately 9 340. (Fig. 6, 7)

Due to the simultaneous application of mining and hydrochemical extraction within one productive horizon in a close vicinity of each other not even the second general condition of underground leaching is fulfilled, namely an independent hydraulic system. The depression cone which is the result of drying of the horizon for mining influences also the chemical extraction and technological solution of chemical extraction runs towards the mine. The dispersion of technological solution in void water within the mining area results in s. c. dispersion solutions.

The vertical flow of technological solutions caused mainly by the fact that the underground leaching does not operate in a state of slight deficit resulted in a dispersion of technological solutions into fucoidal sandstones situated over the permeable layers of the ore mineral.

CHEMICAL CONSTITUENTS			
MAJOR (g/l)		MINOR (ppm)	
H <sub>2</sub> SO <sub>4</sub>	15 - 38	Ca	200 - 300
SO <sub>4</sub> <sup>2-</sup>	40 - 65	Si	100 - 200
NH <sub>4</sub> <sup>+</sup>	1 - 2	P	50 - 150
Al	4 - 6	K	40 - 70
Fe	0.5 - 1.5	Zn	30 - 50
NO <sub>3</sub> <sup>-</sup>	0.2 - 0.8	Mg	20 - 30
F <sup>-</sup>	0.1 - 0.3	Ni	20 - 30

Fig. 6: Chemical composition of the technological solution of the in situ leaching

	Technological solution	Solution of dispersion
$^{234}\text{Th}$	1 870	270
$^{230}\text{Th}$	1 640	100
$^{226}\text{Ra}$	3	20
$^{210}\text{Pb}$	45	20
$^{210}\text{Pb}$	0.8	0.5
$^{231}\text{Pa}$	< 3	< 0.7
$^{227}\text{Ac}$	cca 15	cca 13
$^{227}\text{Th}$	15	13
$^{223}\text{Ra}$	16	12
$^{232}\text{Th}$	58	< 4
$^{228}\text{Ra}$	3	0.7
$^{228}\text{Th}$	19	0.9

Fig. 7: Concentration of radionuclides in solutions of the in situ leaching ( $\text{Bq.l}^{-1}$ )

From the point of view of ecology the most relevant facts about the technology of underground leaching in the ore deposit Stráž pod Ralskem are as follows:

- the passage of technological bores through the overlying bed of the Turonian horizon which contains an accumulation of quality water. Due to technological deficiencies these Turonian waters were contaminated by technological solutions. The contaminated Turonian waters spread over 245 hectares, i. e. 43 per cent of leaching fields. While there is not a complete balance of these waters, estimates vary in the range of 2 to 20 million cubic metres
- the disposal of waste effluent of the sorption-elution uranium process (the excess part of mother liquors) by means of pressure injection into the underground together with the leaching solution. This method up to now introduced about 100

thous. tons of ammonium into ground waters. The average amount of injected  $\text{NH}_3$  in 1990 was about 220 kg per hour

- the coexistence of mining and chemical extraction causing big volumes of dispersion solutions.

The amount of chemicals injected into the underground from the beginning of extraction till the end of 1990:

$\text{H}_2\text{SO}_4$ .....	3 734 704 tons	$\text{NH}_3$ .....	102 913 tons
$\text{HNO}_3$ .....	270 054 tons	$\text{HF}$ .....	25 093 tons

The yearly amount of injected sulphuric acid (about 150 thous.tons) was approximately 13 per cent of the total production of ČSFR.

The total volume of contaminated water is very large : about 200 million cubic metres. The affected rock volume is about 720 million cubic metres, the affected area is about 2 800 hectares.

We can divide contaminated water into two main groups:

- a) technological solutions. They are highly mineralized waters with sulphate content higher than 20 g per litre, total dissolved substances about 35 -70 g per litre. Their volume is about 50 per cent of the total contaminated water ( $100 \cdot 10^6 \text{ m}^3$ )
- b) dispersion solutions. They are mineralized waters with the content of total dissolved substances about 4.5 g per litre.

The environmental relevancy of solutions for chemical extractions is considerable due to the possible influence on the quality in large surroundings. As a measure of relevancy the research chose the multiple of exceeding the value of the drinking water standard, laid down by the Czechoslovak Standard (hereinafter only: ČSN) Nr. 75 7111 or of the value laid down in Regulation Nr. 25/1975 LG., for water supply flows. While the original Cenomanian water has been contaminated by only one contaminant -  $^{226}\text{Ra}$  in average values by 100 times, the contaminants of

chemical extraction solutions exceed these standards as follows (in multiples of the standard):

Technolog. solutions		Dispersion solutions		
1	$\Sigma$ alfa	17 580	$\Sigma$ alfa	1 520
2	Be	4 640	$\Sigma$ beta	305
3	$\text{NH}_4^+$	2 540	$\text{NH}_4^+$	300
4	$\Sigma \beta$	1 930	Be	160
5	As	271	Ra	48
6	Ni	234	Ni	33
7	Cr	216	Cr	19
8	$\text{F}^-$	158	As	18
9	V	121	Cd	18
10	Mn	100	V	16

The environmental relevancy of underground leaching in the ore deposit Stráž pod Ralskem is evident from:

- a) the environmental relevancy of components of chemical extraction solutions
- b) large volumes of chemical extraction solutions
- c) the pressure level of Cenomanian aquifer being above the level of the free Turonian level; such was the state before the beginning of the extraction and will return after the end of the extraction: at present in locations of leaching solution injection the value of pressure difference is higher than the initial one
- d) the interlinking of the Cenomanian and Turonian aquifer is by all means the result of technological boring errors: there exists also - with high probability - a natural interlinking and communication of Cenomanian with Turonian



- e) the penetration of technological solutions into the Turonian aquifer and its contamination is the result of the above mentioned facts
- f) the linkage of the Turonian aquifer to the surface water of the Ploučnice River adds weight to the relevancy of the contamination frontier shift of the Turonian water towards west and southwest because the contaminated water approach the region where there exist a direct link between the Turonian aquifer and surface water, namely in the region of Stráž pod Ralskem - Noviny pod Ralskem. In this region the Ploučnice River gains about 40 litres per second from the Turonian aquifer
- g) the significance of the Turonian aquifer of the North Bohemia Chalk a protected area of natural water accumulation for water management. The proceedings of the Resource Categorization Commission of June 30 1982 estimated natural water resources in the category C 2 of the Ploučnice River catchment at 5 115 litres per second, out of which usable resources at 1 750 litres per second. At present within the Stráž block taking facilities with the capacity of 1 500 litres per second are in operation or are being prepared. The frontier of the contaminated Cenomanian water in southwest approached the zone of the lind stage of Mimoň water source sanitary protection zone (70 litres/s). The southeast frontier of dispersion solutions is within 1.2 - 1.5 km distance from the lind stage of Dolánky water source sanitary protection zone (200 litres/s - taken for the city of Liberec)
- h) contamination of the environment during the clean up of the chemical extraction impact. Any up to now designed technology for the disposal of chemical extraction solutions results in the discharge of waste into the environment:
- into water flow: processed residual solutions - where the main problem poses the capacity of the Ploučnice River for  $\text{SO}_4^{2-}$ ..... $\text{NH}_4^+$  and RL, which at present is practically used up
  - into tailings the solid waste deposition and with them all environmentally relevant components of chemical extraction solutions.

Liquidation of chemical extraction effects. The main problem is the disposal of chemical extraction residual solutions. It is necessary to reckon with a further contaminants release from the affected rock so that the pumping out and processing of one pore water volume removes only about 70 per cent of harmful matter and to achieve 90 per cent decontamination requires the replacement and processing of 5 pore water volumes. i. e. about 940 million of cubic metres of water. The relevant literature does not mention any other deposit in the world with the same big volumes of contaminated water containing such amount of sulphuric acid and dissolved salts occurring in the liquidation process of underground leaching.

While the western countries require that before the authorization to underground leaching is granted the operator proves his ability to return the underground waters at the termination of the extraction to a required (usually + - the original) state, on the ore deposit Stráž pod Ralskem the solutions disposal technology has not been approved until today.

A technological method is being prepared in uranium industry (state enterprise DIAMO, where uranium production is organized and carried out). This technology is based on pretreatment, reverse osmosis, evaporation, crystalization and makes an appropriate effort to utilize some components:  $H_2SO_4$ ,  $Al_2O_3$ ,  $NH_3$ , gypsum etc. A number of technological problems remains unsolved, for example the removal of radium and other toxic metals which might spoil the products.

The estimation of the costs for restoration of this ISL deposit is very difficult. It depends on:

- environmental demands for the state of the underground after the liquidation
- costs for technology of cleaning water and possibilities of utilizing by-products.

It is clear that costs for restoration will be huge and will reach tens of billions Czech Crowns.

Thus it is necessary to seek a reasonable compromise between environmental demands and economic feasibility. In no case the

problem should be further aggravated by enlarging the ISL technology.

As an objective for the state of the underground after the liquidation has been terminated we recommend:

For the Turonian aquifer - to aim at the original composition of the Turonian water as much as possible.

Cenomanian aquifer - it seems evident that in the space of leaching to achieve the original composition of Cenomanian water at reasonable cost is impossible. The composition of the Cenomanian water after the liquidation of underground leaching has been terminated should be such as to warrant that possible infiltration of the Cenomanian water into the Turonian horizon and further into surface water will produce a mixture of an appropriate quality, i. e. the Turonian - drinking water, and the surface water - water not suitable for water supply systems, pursuant Regulatorio Nr. 25/1975 LG.

## 5. THE IDENTIFICATION AND CHARACTERIZATION OF CONTAMINATED SITES

A lot of data about the state of the environment are necessary for preparing a restoration programme. The basic survey is in the following part.

Geofyzika Brno made in the framework of this research aerial photographs for gamma radiation over an area of 185 square km. This method showed a very high indicative ability and proved that in the region in question the radioactive contamination concentrates in areas of mining extraction including the route of ore transportation. Outside this area a widespread contamination was proved with respect to sediments and flood-plain soils of the Ploučnice River. (Fig. 8, 9)

A surface measurement of gamma batch input in an area of 42.5 sq.km show that 78.9 per cent of the area lies within the limits of  $0.005 - 0.125 \mu\text{Gy.h}^{-1}$ , 20.9 per cent within the limits of  $0.125 - 0.250 \mu\text{Gy.h}^{-1}$  and 0.2 per cent over  $0.5 \mu\text{Gy.h}^{-1}$ . For the background the value of  $0.1 \mu\text{Gy.h}^{-1}$  is recommended. No disas-

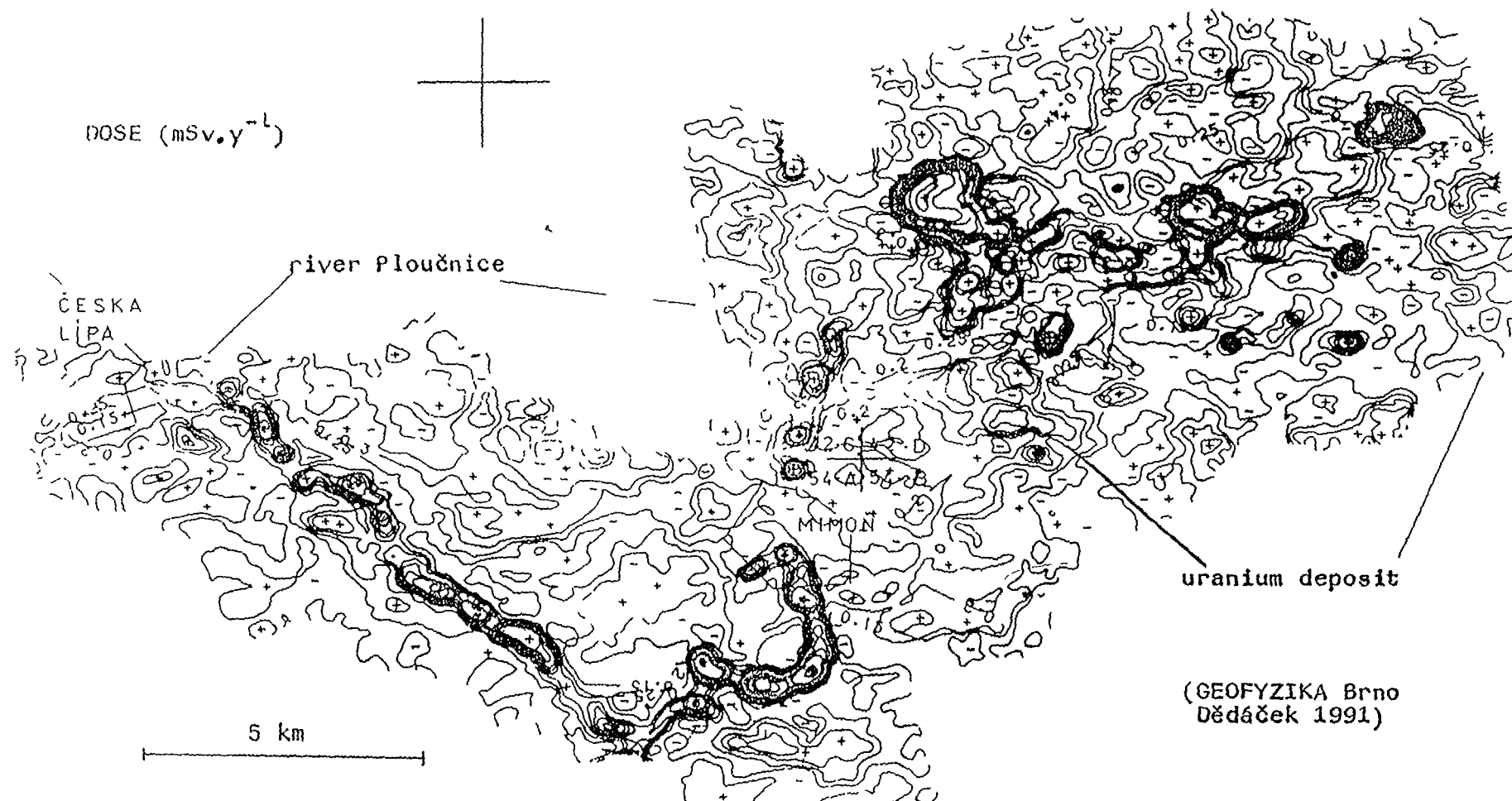


Fig. 8: Airborne gama-spectrometry of the region Stráž p. R. - Hamr

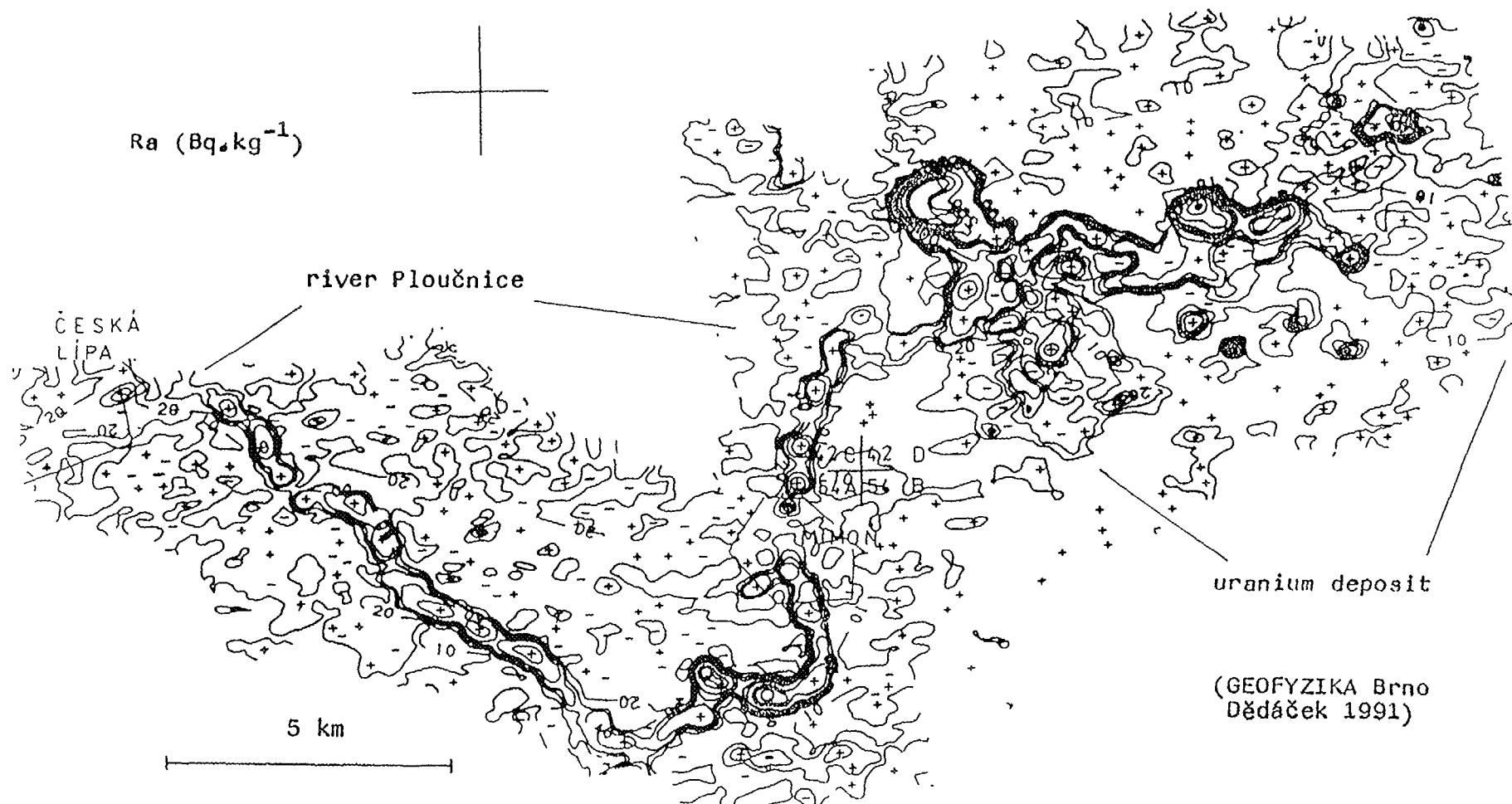


Fig. 9: Airborne gama-spectrometry of the region Stráž p. R. - Hamr

ter locality was found. Due to the activities of the uranium industry the level of contamination of the whole region under consideration is slightly and consistently growing.

In the production of common exhalations the uranium industry belongs to sources of medium significance ( $\text{SO}_2$  emissions per year about 2 000 tons,  $\text{NO}_x$  about 900 tons). Decisive for the emission situation is the influence of long distance (transborder) sources, in the first place from Germany and Poland (65.8 per cent). The estimate of average yearly concentrations for  $\text{SO}_2$  is 40 - 50  $\text{mg.m}^{-3}$ , for  $\text{NO}_x$  35 - 45  $\text{mg.m}^{-3}$ , suspended particles 50 - 70  $\text{mg.m}^{-3}$ .

Rn emissions due to technology are estimated at 126.2 TBq per year. According to a mathematical study of emission situation Rn the highest short-term voluminous activities are achieved in the community of Útěchovice, where the activity of 20  $\text{Bq.m}^{-3}$  is being exceeded in 11 days per year. The mean yearly activity at Stráž pod Ralskem is 2.5  $\text{Bq.m}^{-3}$ . An estimation on the radiation load on the population for radon due to technology is 3 - 7 per cent of the yearly limit pursuant to the Ministry of Health ČR Regulation Nr. 59/1972 LG. i. e. 15 - 21 per cent of the yearly limit pursuant to ICRP recommendation.

Solid particles deposition is the important source of which ore handling is a relevant factor of environmental influence since it not only contains a higher amount of radionuclides of all their decay chains but also heavy metals. Clark concentrations of an average dust deposition within the region are being exceeded 47 times for As, 18 times for U, 17 times for Zn, 9 times for Sb etc. The sanitary standard of solid particles deposition 150  $\text{t.km}^{-2}$  per year has been exceeded along the route of transportation. In the centre of Stráž pod Ralskem mean annual values vary in the range of 50 - 80  $\text{t.km}^{-2}$  per year.

The groundwater is contaminated mainly with the chemical extraction solutions, by infiltration from tailings, dump-piles and locations where the ore is manipulated. Over the area of about 30  $\text{sq.km}$  today about 200 million of cubic metres of water are contaminated.

Two basic kinds of pit water are pumped and treated in the region, namely neutral pit water from mining and acid pit water due to dispersion solutions between mining and chemical extraction. Total volume of pit water discharged annually is about 15 million cubic metres. The treatment of neutral pit water was done from the beginning of extraction till 1989 in temporary, provisional decontamination plants of an inadequate efficiency. The result of it is the contamination of sediments and the Ploučnice River flood-plain. Since the Central decontamination plant was put in operation (1989) the treatment of neutral pit water has been highly efficient. This corresponds with the lowering of radium concentrations in the Ploučnice River. (Fig. 10)

Acid pit waters are processed in neutralizing decontamination plants. Here the removal of ammonium ions by chlorination is problematic which is a typical example of the difficult removal of effects when the cause is neglected (ammonium ions are still injected into the underground). For the future the main problem will be the content of sulphates and ammonium ions in the Ploučnice River and the potential risk of by-products of chlorination (chloramines, chlorine, derivatives of organic materials).

The result of the inadequate treatment of uranium industry pit water is a widespread contamination of the sediments and flood-plain soils of the Ploučnice River in the length of about 30 kilometres of the flow from Stráž pod Ralskem towards Česká Lípa. About 340 hectares of soil and about 340 thous. cubic metres of it are contaminated over and above the input  $\text{gama} > 0.2 \mu\text{Gy.h}^{-1}$  (which is about the double of the natural background or the region).

The total land claim of agricultural and forest land is 1 669.4 hectares, out of which the claim of chemical extraction is 1 022.5 hectares, and that of mining 646.8 hectares.

There are three basic soil types in the region, i. e. podzolic, brown agricultural soil and flood-plain soil. The main source of contamination are solid particles arising from ore handling.

The area of the contaminated zone with a concentration higher than  $5 \mu\text{g U.g}^{-1}$  is about 500 hectares.

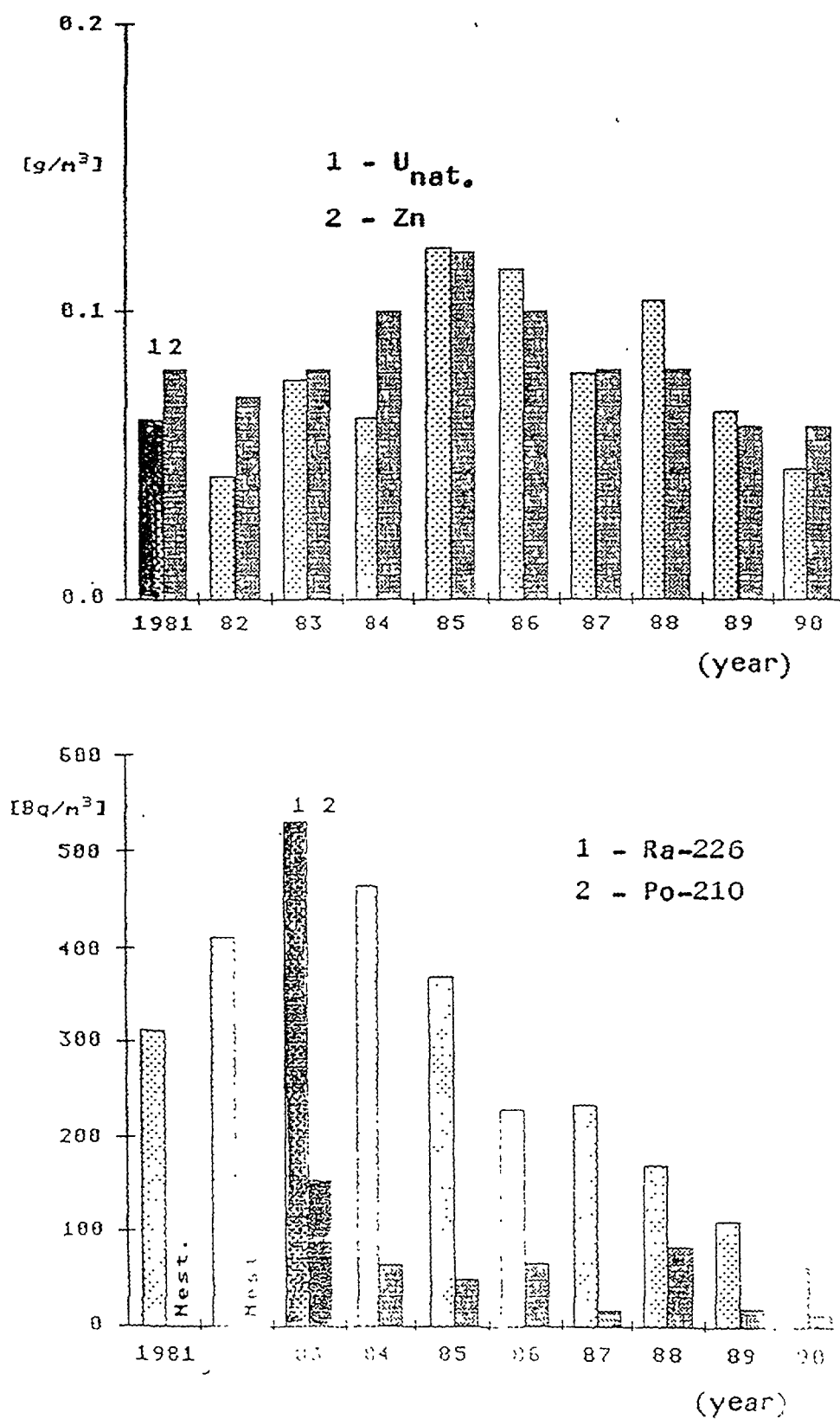


Fig. 10: Concentration of U, Zn, Ra, Po in the river Ploučnice



Chemical extraction, mainly in older fields causes mechanical and chemical soil degradation characterized by the growth of alkalization and salification of the soil. In the leaching fields on the slopes of Ralsko an extensive water erosion takes place which is mostly only belatedly stabilized. To improve the situation biotechnical measures are taken (grass mats, hydro sowing).

The main problem of vegetable production contamination is the solid particles deposition of ore handling. It is therefore necessary to procure protective biological barriers and in the sowing plan to prefer plants with low level transfer of radionuclides into alimentary chains (cereals).

The extraction and processing of uranium does not directly endanger the inhabitants. The most hazardous technology in this respect is the chlorination of acidic pit water including transport and handling chlorine.

## 6. CONCLUSION

Present problems of environmental protection are in the first place the result of erroneous decision making of the past (e. g. the opening of an ore deposit simultaneously for mining and in situ leaching). That makes present decisions on the future concept of uranium industry in our country more important. The government of the Czech Republic is preparing a complex programme for reducing the uranium production. The general energetic concept of the state, environmental demands and market situation of uranium in the whole world must be taken into consideration.

At the same time the programme of the restoration of all uranium mines and mills is being prepared. It is clear that the most complicated and most expensive will be the solution of the problems in deposit in North Bohemia. This target will last several tens (or hundreds) years and international experience will be very valuable.

## REFERENCES

1. ANDĚL, P., PŘIBÁŇ, V. et al: Vliv uranového průmyslu na životní prostředí oblasti Stráž pod Ralskem [Environmental impact of the uranium industry in the region Stráž pod Ralskem]. - MEGA, Stráž pod Ralskem, 1991
2. kolektiv: Československá ložiska uranu. - [Uranium deposits in Czechoslovakia]. - Československý uranový průmysl, Praha 1984
3. BENEŠ, V.: In-situ leaching of uranium in north Bohemia. - IAEA, Technical committee Meeting on Uranium In-situ Leaching. Vienna, October 1992
4. FIEDLER, J. e SLEZÁK, J.: The experience with coexistence of classical deep mining and in-situ leaching of uranium in northern Bohemia. - IAEA, Technical committee Meeting on Uranium In-situ Leaching. Vienna, October 1992

# IDENTIFICATION AND RADIOLOGICAL CHARACTERIZATION OF CONTAMINATED SITES IN ESTONIA

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## Abstract

Three sites are considered radioactively contaminated areas by the Estonian public. This report presents the available information concerning the environmental consequences of the radioactive waste depository at Saku, the Russian Navy training center with two submarine reactors at Paldiski, and the uranium mill tailings at Sillamäe. On the basis of the presented information only the Sillamäe area can be considered a radioactively contaminated site in Estonia which needs clean-up and immediate remedial actions. The other two sites should be considered potential sources of radioactive contamination. Mill tailings of the Sillamäe plant are located at the coast of the Gulf of Finland (59°24.9' N, 27°43.5 E). According to the data from the plant tailings contain about 9 million tons of waste which consists of some 1200 tons of uranium, 800 tons of thorium and about  $3 \times 10^{14}$  Bq (7000 Ci) of  $^{226}\text{Ra}$ . The dose rate on the depository varies between 0.5 and 35  $\mu\text{Sv/h}$ . The international working group of Estonian, Finnish, Norwegian and Swedish experts is currently preparing the final report of the environmental impact assessment study of the Sillamäe depository. This report presents the preliminary results of this study.

## 1. INTRODUCTION

The Republic of Estonia can be considered a non-nuclear country as there are currently neither nuclear power plants nor research reactors under our responsibility, but concern and worry about radiation problems is widely spread among the population of Estonia. This is primarily based on lack of knowledge about the problems connected with radiation, and distrust towards the relevant authorities which is mainly caused by the former classification of information.

Thus, there are three problem areas of major public concern in the field of radiation and nuclear safety in Estonia:

- shallow-land depository for low-level radioactive waste at Saku (inland, near Tallinn),
- two submarine reactors and radioactive waste storage at Paldiski (Russian Navy training center at the NW coast), and
- mill tailings of the Sillamäe plant (North coast).

## 2. SAKU RADIOACTIVE WASTE DEPOSITORY

The shallow-land depository for low-level radioactive waste in Saku, 12 km outside Tallinn, is only radioactive waste handling facility in Estonia. The site has been exploited since 1964 and has been designed for deposition capacity below 7.5 TBq/y (200 Ci/y).

The area of the depository is 1 km<sup>2</sup>. In the disposal site itself (200 m<sup>2</sup>) there are two separate tanks in the ground, one for solid and the other for liquid waste. The disposal site for solid waste is a concrete box with a volume of 200 m<sup>3</sup> which is divided into sections by walls. When a section is full, it is covered with concrete. By now some 55% of the volume have been filled up. For storage of liquid waste there is a 200 m<sup>3</sup> tank made of stainless steel surrounded with concrete which has nearly not been used in recent years since no liquid radioactive waste has been produced in Estonia.

Since the taking into exploitation of the radioactive waste depository in Saku, about 220 TBq (6000 Ci) of low and medium level radioactive waste such as spent radiation sources and radioactive substances from industrial radiography equipment, hospitals and scientific laboratories have been

stored there. Currently the remaining activity of the waste is some 75 TBq (2000 Ci). High-activity sources have not been disposed in Estonia, these have been sent to Russia for special handling.

The radioactive waste depository in Saku does not meet modern radiation and environmental protection requirements and planning of remedial actions is necessary in order to bring the depository to an acceptable condition. However, according to data from the Radio-Hygiene Department of the Estonian Center of Health Protection, which is responsible for environmental supervision of the site, no surface or ground water contamination have been identified.

### 3. RUSSIAN NAVY TRAINING CENTER IN PALDISKI

The second problem argued as a potential hazard for the population and the environment are two nuclear reactors in Paldiski, 47 km NWW from Tallinn, which belong to the Russian Navy (the former Soviet Navy). Both of these reactors were built into cylindrical submarine sections used for training of submarine crews and were closed down in 1989. The older reactor has been refueled once, in 1981. On the basis of available information the reactors are being kept in safe shut-down conditions and do not present any significant risk to safety.

There are two interim storage facilities for radioactive waste in the territory of the Training Center - one for liquids and the other for solid waste. According to information from the Training Center the amount of liquid and solid waste is 473 tons and 30 tons respectively, the estimated total radioactivity being about 370 and 3700 TBq respectively at present. Because of the limited information available and uncertainties concerning the quality of the storage facilities, the activity of the waste, the quality of the storage tanks, arrangements inside the storage facilities, etc., it has been impossible to carry out technical evaluation of the waste storage facilities.

At the radiation safety survey of the Training Center it was confirmed that the environmental radioactivity in the vicinity of Training Center is continuously monitored. However, data of these investigations have not been presented to the Estonian authorities.

### 4. SILLAMÄE MILL TAILINGS

The Sillamäe Plant was built in 1948 as a top secret facility for uranium production for military and civil use, processing originally alum-shale (containing about 0.03% U) from Estonia. Later, uranium ore from Eastern Europe (up to 1% U) was processed. In total, more than 4 million tons of uranium ore were processed in Sillamäe. Until 1970, only uranium ore and alum-shale were processed at the plant. In the beginning of the 1970s the plant switched to processing loparite - a mineral from Kola Peninsula rich in niobium, tantalum, titanium and other rare earth metals. Loparite also contains uranium (about 0.03%) and, in particular, thorium (about 0.6%). The waste from uranium processing and fly ash from the local power plant were transported to the first marine terrace of the Gulf of Finland and stored at the surface until 1959, when the site was surrounded by a dike and the waste depository was established. The depository has been reconstructed a couple of times in the last decades. In 1969-70 it was expanded to its present size. Today the depot is an oval retention impoundment at the coastline of the Gulf of Finland with an overall area of about 330 000 m<sup>2</sup>. The bottom of the depository consists of permeable coarse-grained material (sand and pebbles) lying on a 2-10 m thick layer of watertight Cambrian clay, at about 4 m above sea level, the closest point of the embankment is about 30 m from the shoreline and the top of the dam is about 25 meters above sea level. The dose rates measured on the top of embankment dam and on its slopes varied between 0.5 and 35  $\mu$ Sv/h. About 30% of the depository's surface is covered by a sedimentary pond containing some 150 000 m<sup>3</sup> of waste water with a depth of 0-3 meters.

Together with the former storage site for uranium ore, the radioactively polluted area covers some 100 ha in total. The clean-up activities (the contaminated surface material is being removed and transferred to the top of the embankment wall) of this storage area is currently under way.

In 1992 an international co-operation project for environmental impact assessment of the Sillamäe mill tailings was started. The tasks of the project group were defined as follows:

- estimation of the amounts of radioactive and chemical pollutants in the depository,
- estimation of the amounts of the radioactive and chemical pollutants leaking from the depository to the Gulf of Finland,
- estimation of the environmental impact of the Sillamäe waste pond to the Gulf of Finland, to the Sillamäe town, and to the groundwater in the area.

The working group consisting of Estonian, Finnish, Norwegian and Swedish experts will complete the final report of the project by the end of May, 1994.

The preliminary results of investigations suggest that about 6.3 million tons of uranium mining residues and about 6.1 million tons of oil-shale ash together with wastes from rare earth elements production is stored in the depository. This material contains about 1830 tons of uranium and 850 tons of thorium. The production estimates given by the plant are 1200 tons of U and 800 tons of Th. Thus, the thorium values are in good agreement, whereas the uranium content is higher than the estimation of the plant.

#### *4.1. Solid samples from the depository*

Concentrations of  $^{238}\text{U}$  and  $^{226}\text{Ra}$  in uranium processing wastes varied from 1500 to 23 000 Bq/kg and from 10 000 to 220 000 Bq/kg dry weight, respectively. Concentration of  $^{232}\text{Th}$  in uranium mining residues varied between 25 and 330 Bq/kg. Recently deposited waste of the factory from loparite processing contains between 180 and 3600 Bq/kg uranium, between 130 and 2300 Bq/kg radium and from 200 to 4000 Bq/kg thorium.

#### *4.2. Outdoor radon measurements*

The waste depository exhales great amounts of radon into ambient air. At the height of 0.3 m above the surface of the depository the airborne concentration, measured by a portable radon monitor in September 1992, varied from 60 to 500 Bq/m<sup>3</sup>. The highest mean concentration of radon in outdoor air measured near the depository in period from September 1992 to May 1993 was 310 Bq/m<sup>3</sup>.

#### *4.3. Groundwater samples*

Large variations of the elemental composition of the water samples collected in the monitoring wells between the depository and the Gulf of Finland were observed. The concentration of some elements varies with more than two orders of magnitude, for example U concentration varies from 6 to 37 200 µg/l, Cu from <25 to 13 000 µg/l, and Zn from <20 to 4 410 µg/l.

On the basis of the obtained results and estimated water flow rate under the depository (some 300 to 700 thousand m<sup>3</sup>/year) the annual release of some elements into the Gulf of Finland was calculated (Table 1).

Comparison between background values of groundwater and pond water shows that the concentration of many elements is over 100 times higher in the pond water. These elements are Zn, Cu, Al, Cr, Fe, La, Sr, Co, P, Ni and V in decreasing order.

Concentrations of  $^{238}\text{U}$  in tap water samples vary from below 0.01 to 0.15 Bq/l, and  $^{226}\text{Ra}$  concentrations from 0.02 to 0.15 Bq/l. These concentrations are quite low and they can be expected to be natural levels in ground waters of this area. These results indicate that the waste depository does not contaminate the groundwater layer which is being used for local water supply in Sillamäe.

Table 1

Element	Conc. in the tailings pond	Avrg. conc. in the well	Content in tailings pond	Release from the depository	Content in the interstitial water
	$\mu\text{g/l}$	$\mu\text{g/l}$	tons	tons/year	tons
U	2800	7500	0.5	3.5	15
Th	<5	<5	<0.0009	<0.0024	0.001
Ra(Bq)	0.1	0.1	1.8E7	4.7E7	2.0E8
Cu	2800	3250	0.5	1.5	6.5
Zn	10400	730	1.9	0.4	1.5
F	1450	11200	0.3	5.3	22

#### 4.4. Samples from Gulf of Finland

The results of sea water sampling show that the impacts of the depository on the marine environment are clearly detectable in the waters close to the shore-line but the concentrations of radioactive substances decrease rapidly when going further away from the site. On the shore-line, straight below the pond, the highest uranium concentration was 190  $\mu\text{g/l}$  (2.3 Bq/l of  $^{238}\text{U}$ ).  $^{226}\text{Ra}$  concentration at that site was only 0.03 Bq/l. The most far-away measurements of elevated radionuclide concentrations were made at a distance of 300 m from the shore-line. The uranium concentration there was 3  $\mu\text{g/l}$  (about 0.04 Bq/l of  $^{238}\text{U}$ ).  $^{226}\text{Ra}$  concentration at that distance was 0.01 Bq/l. The normal (background) level of  $^{238}\text{U}$  in the water of the Gulf of Finland is 0.01-0.02 Bq/l.

In bottom sediments, elevated radionuclide concentrations were detected in a shallow cove alongside the pond, where the sediment was typical organic "gyttja-clay". The concentrations of  $^{238}\text{U}$  and  $^{226}\text{Ra}$  were 450 and 470 Bq/kg dry weight. At all the other sampling sites the concentrations were at the level typical for the Gulf of Finland. The concentrations of artificial cesium isotopes,  $^{137}\text{Cs}$  and  $^{134}\text{Cs}$ , were in all samples at the same level as in the Gulf of Finland in general.

Due to rapid mixing and dilution of the leaking waters with the sea water, elevated radionuclide concentrations can only be seen in a very narrow coastal area.

#### 4.5. Stability Analyses

On the basis of a preliminary study of the shore-line in the vicinity of the depository it was concluded that the area is affected by the active erosion process. The development and taking of effective protection measures is urgently necessary.

Analysis of the available information and the results of recent geotechnical investigations indicates that the stability factor of the depository embankment wall is currently some 1.2, although the safety standards in the former USSR for similar constructions required the stability factor of at least 1.5.

### 5. CONCLUSIONS

On the basis of currently available information only the Sillamäe area can be considered a radioactively contaminated site in Estonia. The other two sites should be considered as potential sources of radioactive contamination.

According to preliminary results of our project group, the present condition of the depository does not meet modern requirements of safety and environmental protection, and remedial actions are

necessary at the site. In order to perform remedial actions, the present pattern of waste management should be changed and deposition of the waste should be localized to some other area in order not to stop the operation of the plant.

Although the present impact of the waste pile to the Gulf of Finland is quite limited, remedial actions are necessary to guarantee the long-term safety of the depository. These actions should aim at reduction of the external dose-rate to the level typical for the surrounding area and prevent:

- leakage of radioactive and non-radioactive effluent from the depository to groundwater and to the Gulf of Finland,
- radon emanation into ambient air,
- wind erosion of the dry tailings,
- erosion of the shoreline.

These objectives will be achieved by strengthening the protection embankment of the pile by establishing a combined wave breaker and counterload along the most exposed shoreline, reducing the slope of the embankment wall, and covering the wastes with an engineered cover or liner system.

Taking into account that the depository is located at the shore-line of the Gulf of Finland, any activity in the area which may result in activation of the erosion processes must be avoided in the future.

# **IDENTIFICATION AND RADIOLOGICAL CHARACTERIZATION OF SITES CONTAMINATED DUE TO MINING AND MILLING: METHODS AND CRITERIA APPLIED IN GERMANY**

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## **Abstract**

Conventional mining and milling has been in place in certain parts of Germany for centuries. As the mined ores are frequently accompanied by uranium minerals, radiation protection problems can be associated with such conventional waste. After World War II intensive uranium mining and milling was started resulting in significant environmental contamination and radiation exposure of the public.

Decisions on restoration and remediation of radioactive contaminated sites require systematic investigation and objective evaluation of the existing radiation exposure. On behalf of the Federal Ministry for the Environment, Nature Conservation and Reactor Safety the project is guided by the Federal Office for Radiation Protection (BfS). The primary objective of the project is the evaluation of the radiological relevance of contaminated sites and their contribution to radiation exposure. The economic development of the regions concerned is to be taken into account and the public will be informed. In view of the large number and scattering of contaminated sites, a gradual procedure for radiological characterization had to be developed.

## **1. SITUATION IN MINING DISTRICTS**

### **1.1. Consequences of traditional mining**

Since the Middle Ages ores of tin, silver, copper, nickel, bismuth and others but also hard coal have been mined and processed in certain districts of Saxony, Thuringia and Saxony-Anhalt. The mined ores, and a special type of coal, too, are frequently accompanied by uranium minerals. Problems of radiological protection can therefore be associated with the waste materials of the traditional mining and the areas affected by them.

Many waste rock piles and other left properties cannot be found any longer since they were covered, dumped materials were removed and used for different purposes. Former mining sites were frequently used as building sites.

In particular, the utilization of former mining sites and material can result in a radiation exposure of the public higher than the natural (terrestrial) level.

Radioactive contamination and radiation exposure owing to waste products of mining and processing cannot only be found in the mining districts themselves because materials (e.g. slag and slag bricks produced from copper slag, coal) were transported and used all over the country.

### **1.2. Consequences of uranium mining and milling**

After World War II intensive uranium mining was started in some of the traditional mining districts of Saxony, later on extended to other districts of Saxony and Thuringia occupying much land and changing the landscape. Numerous mines, shafts, mills and other facilities were in operation and waste rock piles and tailings ponds of partly enormous dimensions were set up.



Radioactive-contaminated substances (e.g. pit waters, waste waters from mills and seepage waters from waste rock piles and tailings ponds, exhaust air from mines and mills) were released into the environment and resulted in environmental contamination and radiation exposure of the public.

In the 1950s and early 1960s many mines and the small mills were closed down and the waste was handled, dumped and disposed of with little attention to provisions other than mining safety and with simple measures to prevent potential leaching or transport by water taking into account the standards of that time. Only simple clean-up measures were made in buildings, at sites, grounds etc. which were not used for uranium production any longer and were passed on to other enterprises, communities and citizens.

Materials from the traditional mining and materials from uranium mining (in particular waste rocks) were used on a large scale for land filling, road construction and other purposes. This can result in radiation exposure of the public if, provision of radiological protection were not made.

The production of uranium was abandoned for commercial and other reasons in 1991. Mines, mills and other facilities used for uranium production are being decommissioned. Adequate practices for waste storage and disposal as well as for clean-up of contaminated lands are developed and restoration and remediation measures for radioactive contaminated sites are included in the process of decommissioning. Critical situations with regard to radiation protection occur only exceptionally but in many cases remedial measures are required to decrease the radiation exposure to an acceptable level.

Relics of the uranium industry are found in other countries, too. But apart from the quantity of left properties and their dimensions there are other differences since the Eastern German mining districts are densely populated and intensively used for industrial and agricultural activities and for recreation. Therefore restoration and remediation of radioactive-contaminated sites are of special importance in the mining districts of Saxony, Thuringia and Saxony-Anhalt.

## **2. IDENTIFICATION AND RADIOLOGICAL CHARACTERIZATION OF CONTAMINATED SITES**

### **2.1. Programmes and methods**

Decisions on restoration and remediation of radioactive contaminated sites and their environment require systematic investigation and objective evaluation of the existing radiation exposure.

Differentiated considerations have to be made since in the mining districts the situation regarding to radiation exposure is much more complex and both natural and man-made components contribute to the level of radiation exposure.

A comprehensive Federal project has been launched to study the contributions of the geological conditions and the residues of mining activities to the radiation exposure of the public. On behalf of the Federal Minister for the Environment, Nature Conservation and Reactor Safety the project is guided by the Federal Office for Radiation Protection (BfS).

Investigations and methods which have to be implemented and applied are prescribed by the Federal Office according to its legal obligations. The investigations are carried out by the BfS itself or on its behalf by contractors for sites of traditional ore mining and processing and for sites of early uranium mining and milling. Sites, piles, ponds etc. still in possession of the uranium industry (Wismut Company) are investigated by the company itself in the course of decommissioning.

The primary objective of the project is the evaluation of the radiological relevance of objects and their contribution to the radiation exposure. At the same time the territorial and economic development of the regions concerned shall be supported and the public informed reliably. In view of

the large number and the scattering of the relics under consideration, a gradual procedure had to be developed and applied in order to solve the tasks efficiently in an appropriate time.

## **2.2. Implementation of investigations**

In a first step "suspicious areas" have been spatially defined taking into account the data, available and information about relevant mining activities (uranium mining and milling, mining and processing of uranium bearing ores and coal) and about areas where the terrestrial gamma radiation is increased in comparison with the average level. Thus, 34 "areas of suspicion" were defined, their total area amounts to about 1 500 km<sup>2</sup>.

Figure 1 gives an impression of the widespread distribution of the areas for which all existing data assumed to be relevant to the radiological evaluation were compiled. In this way about 8 000 mining-related objects of different kind have been identified and registered in the data bank, most of them being waste rock piles. The total area covered by the objects amounts to about 73 km<sup>2</sup>, the total area of above-average gamma radiation ("gamma anomalies") to about 170 km<sup>2</sup> [1]. In many cases the existing data were incomplete and not sufficient for a radiological classification of the relics. Therefore, additional investigation was required.

The registered objects are verified by field inspections (checking and revising their state and related data, e.g. size, volume, covered area etc.) including screening measurements (generally local gamma dose rate, in specific cases estimations of radionuclide concentrations in soil samples). Other relevant information such as demographic data and projections, land use forecasts and agricultural, forestry and industrial data as well as information about goods and other interests to be protected are collected.

Based on the revised and completed data a first radiological evaluation is made applying generic criteria (see chapter 2.3). Objects which almost certainly do not cause radiation exposure of the public significantly above the natural level are separated from further investigation. The remaining objects are subjected to measuring programmes ("basic programmes") to provide detailed information about

- dimensions of affected areas
- thickness of contaminated layers
- concentrations of radionuclides of the uranium decay chain in the materials
- radioactivity released and spread
- relevant pathways and radiation exposure of the public.

More specific but still generic criteria (see chapter 2.3) are applied to define the objects which can result in significant radiation exposure of the public. Only for this category of objects additional site-specific investigation and evaluation are carried out to decide whether remedial actions are required. In this decision procedure "realistic but sufficiently conservative" parameters are applied for the estimation of the radiation dose to man. If the specific estimations result in a radiation dose to man above the primary reference level (see chapter 2.3) remedial measures should be considered taking into account all aspects (e.g. future utilization, costs, public concern etc.). Local authorities are included in the process of decision.

## **2.3. Radiological assessment and characterization**

The German Commission on Radiological Protection arrived at the conclusion that the situation associated with radiation exposure of the public owing to mining, milling and ore processing is a "pre-existing" situation which cannot be assessed and handled with the usual legal tools of radiological protection for planned practices [2].

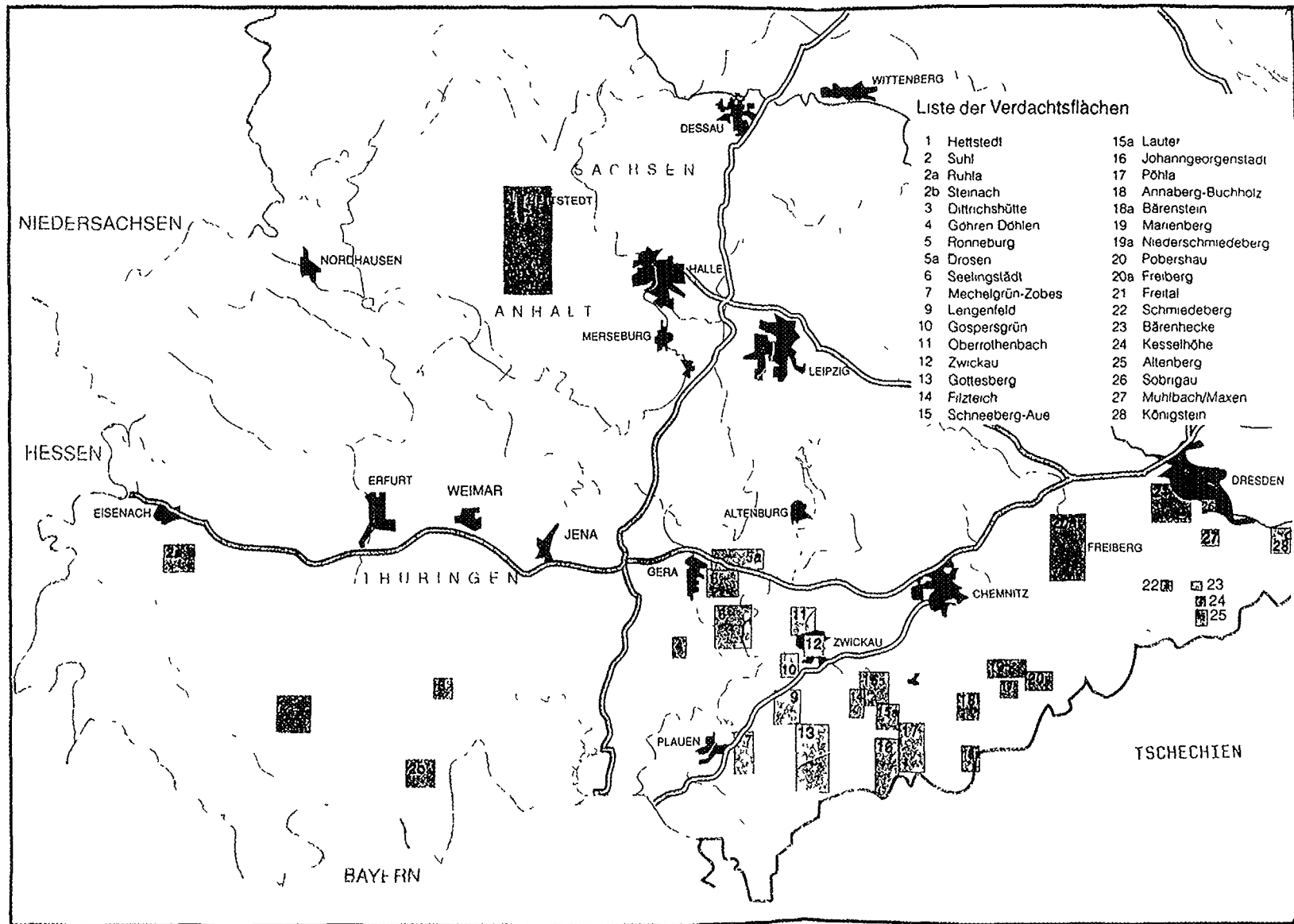


FIGURE 1

The Commission recommended that the radiation dose to man due to the residues of mining activities should not exceed the reference level of 1 mSv/year (primary reference level of effective dose). This primary level means a decision criterion for the radiological significance: if no person will be exposed to more than 1 mSv/year owing to an object (waste rock site, contaminated ground etc.) it can be classified as insignificant and there is no need to consider remedial measures. On the other hand the Commission made clear that this reference level must never be understood as prescribed objective which has to be achieved by remedial measures. The goals of which have to be set up as a result of optimization procedures taking into account not only radiological aspects but also aspects of mining safety, contamination by chemical and other noxious substances, public concern, costs etc.

In addition to the primary dose level the Commission recommended measurable quantities (specific radioactivity in soil, local gamma dose rate, volume of dumped materials, area covered by residues, concentrations of radionuclides in waters affected by uranium mining used for drinking purposes) as release criteria for unrestricted or restricted use (cf. Tab 1). These values are used as insignificance criteria for the classification following the verification.

Table 1

### Criteria and Reference Levels Recommended by the German Commission on Radiological Protection for the Use of Areas Contaminated by Uranium Ore Mining

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* Dose criterion: Reference level for the effective dose, mining-related in addition to the natural regional level: 1 mSv/a	
* Radioactivity criteria: Measurable quantities (e.g. specific activity $C_A$ of soil; dose rate), natural level included	
— $C_A \leq 0.2$ Bq (Ra-226)/g	————→ unrestricted use
— $C_A < 0.2 \dots 1$ Bq (Ra-226)/g (e.g. for industrial sites with dose rate $\leq 0.3$ $\mu$ Sv/h)	————→ restricted use
— $C_A > 1$ Bq (Ra-226)/g	————→ site-specific investigation calculated for volumes and areas of dumped material of $\leq 10^5$ m <sup>3</sup> and $10^4$ m <sup>2</sup> , resp.

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Additional generic criteria (concentration of radionuclides in seepage waters etc.) which are applied for classification following the basic measuring programmes can be derived from the primary dose level. According to the Commissions recommendations "realistic but sufficiently conservative" assumptions are made in setting up such criteria.

The flowcharts developed for the definition of relevant objects regarding radiation exposure are shown in figure 2 and 3 [3, 4].

### 3. GENERAL CONCLUSIONS

The left properties of uranium mining and milling can be considerable sources of radioactive environmental contamination and radiation exposure of the public. Causes for such situations may have been created during a period of time when principles of radiological protection were unknown,

appropriate regulations were not yet at the latest standard or regulations were not observed. The situation is characterized by the fact that there are already sources of radiation, exposure pathways and individuals exposed are already present. Decisions have to be made to reduce the radiation exposure by intervention, if required.

In accordance with the ICRP recommendation No. 60 these situations should be assessed as "pre-existing" ones which cannot be handled with the usual legal tools (e.g. dose limitation system) of radiological protection for planned practices. But basic principles of radiological protection (justification, optimization) are applicable in such situations, too. They have to be referred to the intervention (remedial measures) and require an estimation of radiation dose to man "as realistic as possible".

The extent of radioactive contamination as well as the radiation exposure of the public owing to the residues and left properties of uranium mining and milling is unknown in many cases. Systematic investigation of the situation is required to define and characterize the objects which can cause significant radiation exposure of the public.

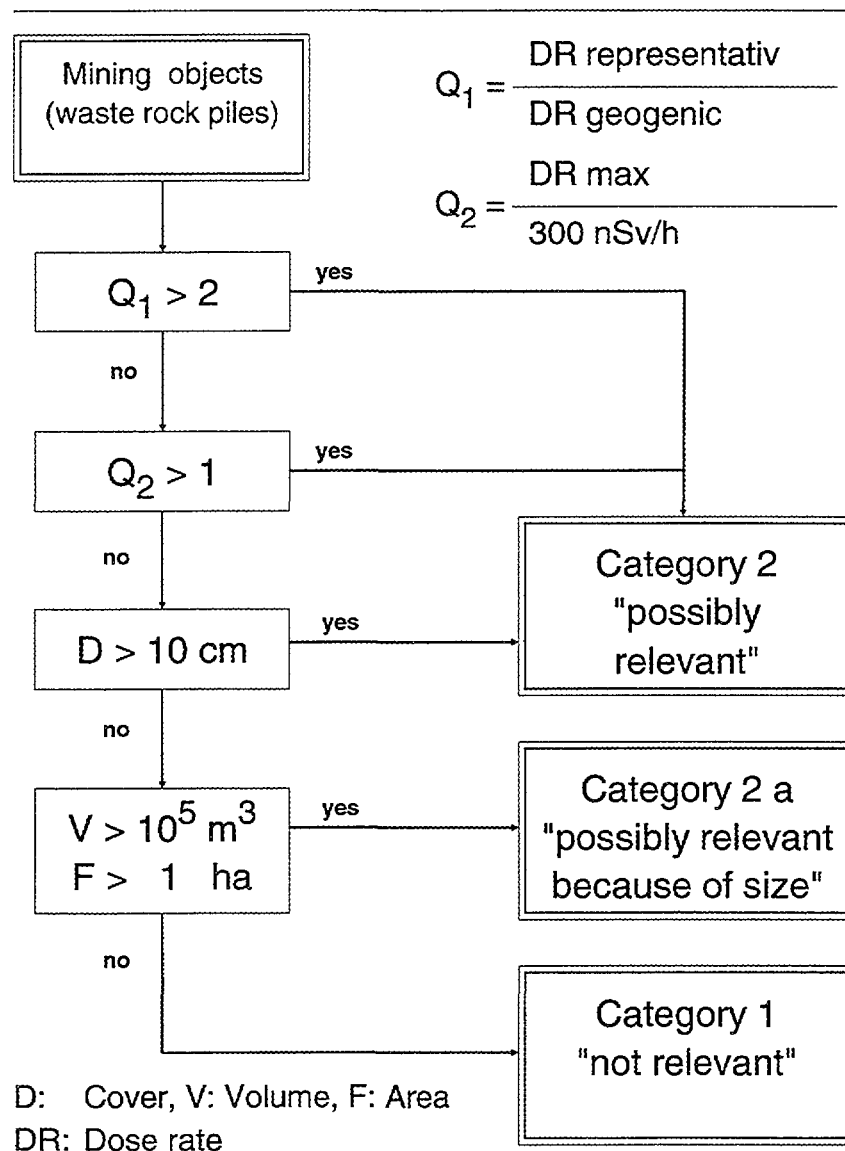


FIG. 2. Flowchart for classification following verification.

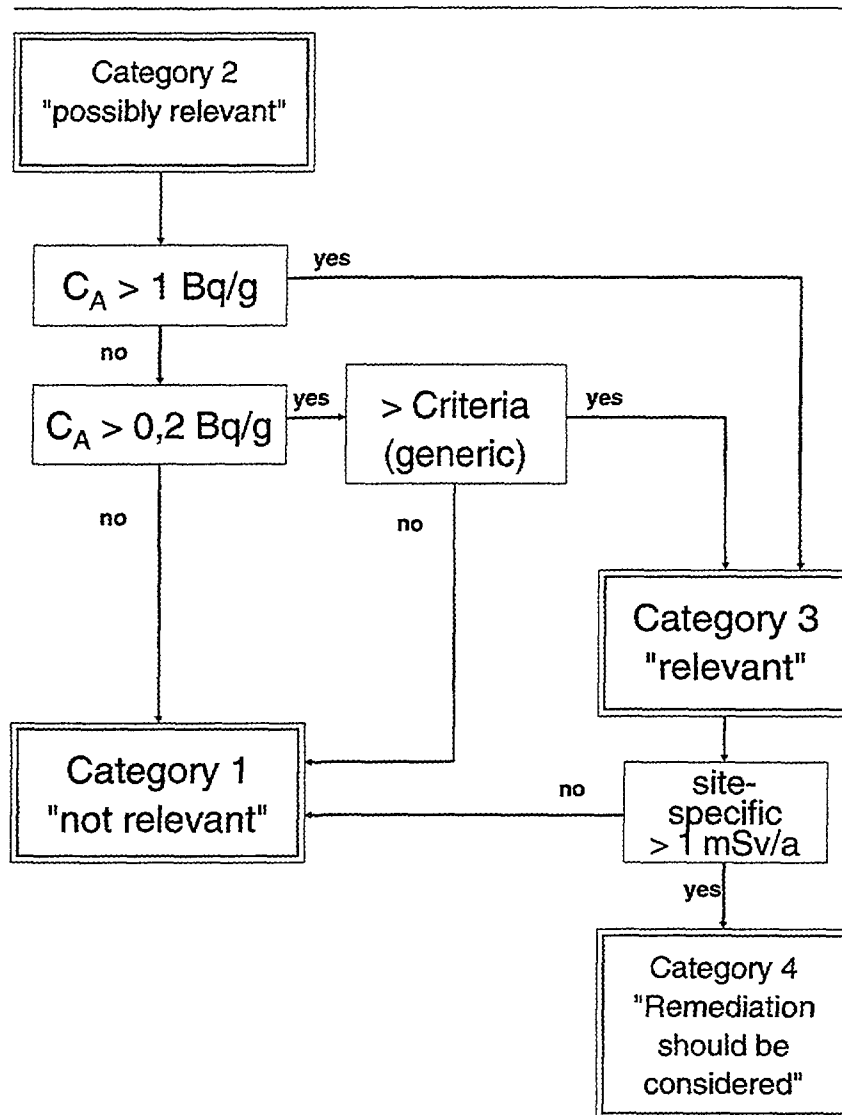


FIG. 3. Flowchart for classification following measuring programmes.

In planning programmes appropriate for approaching the task the following issues should be considered:

- In districts where uranium ore is mined the natural (terrestrial) radiation exposure is very often higher than compared with the average. A good estimation of the terrestrial component of radiation exposure is required to differentiate the respective sources.
- Residues of uranium mining and milling can have been used for different purposes and can be widespread.
- The exposure pathways to be investigated have to be defined taking site-specific features into account.
- If other than uranium ores were mined and processed in the same district their left properties should be included in the investigation since problems of radiological protection can be associated with them, too.

Depending on the quantity and type of objects which have to be subjected to investigations, or on the extent of contamination, it is advisable to plan a gradual procedure:

- define the areas concerned
- collection and compilation of data available and relevant for radiological assessment and characterization of the objects
- verification and revision or completion of the data, if required. Screening measurements should be included in the investigation at this stage
- measuring programmes to get more detailed information, if required.

Criteria for the steps of investigations (primary dose level, measurable quantities) have to be set up in an appropriate manner to separate from further (more detailed) investigations such objects which are insignificant for the radiation exposure of the public. For the remaining objects (objects of relevance) "site-specific" assessments have to be made for the estimation of the radiation dose to man.

If the estimated radiation dose to man exceeds the primary dose level, remedial measures should be considered.

Setting up fixed objectives (standards) for remedial actions is not advisable. The objectives for remedial actions should rather be set up "as low as reasonably achievable" for each site taking into account all relevant aspects.

The objectives for remedial measures and the measures, too, should be approved by the competent authority.

## **REFERENCES**

- [1] Bundesamt für Strahlenschutz  
Radiologische Erfassung, Untersuchung und Bewertung bergbaulicher Altlasten - Abschlußbericht zum ersten Teilprojekt.  
BfS Schriften 8/92 (1992)
- [2] Radiological protection principles concerning the safeguard, use or release of contaminated materials, buildings, areas or dumps from uranium mining.  
Recommendations of the Commission on Radiological Protection with explanations  
Veröffentlichungen der Strahlenschutzkommission, Band 23, Gustav Fischer Verlag, Stuttgart-Jena-New York
- [3] W. Röhnsch, E. Ettenhuber  
Relics of Mining and Milling in Eastern Germany and their Radiological Consequences  
International Symposium on Remediation and Restoration of Radioactive - contaminated Sites in Europe  
(Symposium held from 11 - 15 October 1993, Proceeding in preparation)
- [4] W. Röhnsch, E. Ettenhuber, H. Biesold  
Verifizierung und radiologische Erstbewertung bergbaulicher Altlasten in den neuen Bundesländern  
Veröffentlicht in der Publikationsreihe "Fortschritte im Strahlenschutz; FS-93-67-T"  
Berichte der 25. Jahrestagung des Fachverbandes für Strahlenschutz, Band I, S. 1

# CONSEQUENCES OF THE HUNGARIAN URANIUM MINING AND MILLING

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## Abstract

In the south part of Hungary the uranium is produced from a low grade underground sandstone ore (0.07-0.12 % U). The mining operation in two mines was started in 1958, nowadays three new mines gives the uranium production. Till 1962 the ores only had been ground and sorted, later the milling was completed with chemical processes for uranium extraction (acid and alkaline leach technology). The total amount of the waste rocks disposed of near the mines is  $1.8 \cdot 10^7$  tons. The mill tailings has been emplaced in two retention ponds built with ring dike. The ponds contain  $1.9 \cdot 10^7$  tons of solid particles and  $7 \cdot 10^6$  tons of solution. The amount of the ore heaps for alkaline leach process is  $7 \cdot 10^6$  tons. The radiation levels on the mining and milling area change from place to place in the range of from the background level to 10-100 times the background.

## 1. INTRODUCTION, HISTORICAL BACKGROUND

Hungary has only one uranium mine site in the south part of the country nearby the city Pécs (Figure 1.). The uranium ore could be found in surfacial soil, however uranium mostly occurred in underground sandstone with a low grade. In 1953 the soviet experts started to explore uranium ore by using radiation measurements in this area. The measurements included the determination of gamma dose rate and radon emanation. The exploration measurements can be really considered to be a background measurements, but unfortunately exploration data were not available, because the data were handled secretly.

After the exploration work, the mine operation started in 1956-57, but till 1962 the ores only had been broken, sorted and ground, and afterwards the pre-milling ores were transported to the USSR for uranium extraction. In 1962 the mill operation was completed with acid leaching process and, later in 1965 with alkaline heap leaching technology. Now the company operating the mining and milling activities could produce yellow cake, which has also been exported to the Russia. [1].

## 2. BRIEF SUMMARY OF MAIN SITE CHARACTERISTICS

Regarding the geology and geography, the mining site is located on a hilly countryside and the main part of the milling site is a flat area. The geological formation of the mining site mainly consists of limestone, sandstone and marl. In connection with hydrology, it might be noted that this territory has a complex water movement in underground and a relatively lot of surface water, like streams, canals, fishing ponds. Small lakes can also be found around mining and milling site. Altogether 9 streams and 2 canals flow close to or across this area. Concerning the meteorology, the territory has a moderate continental climate and the monthly precipitation ranges from 37 to 83 mm. It's still remarked that the site is located in a densely populated area, for example the city of Pécs is in distance of 5 km from the mill site and 12 km from the mine site, and it has 170.000 habitants.



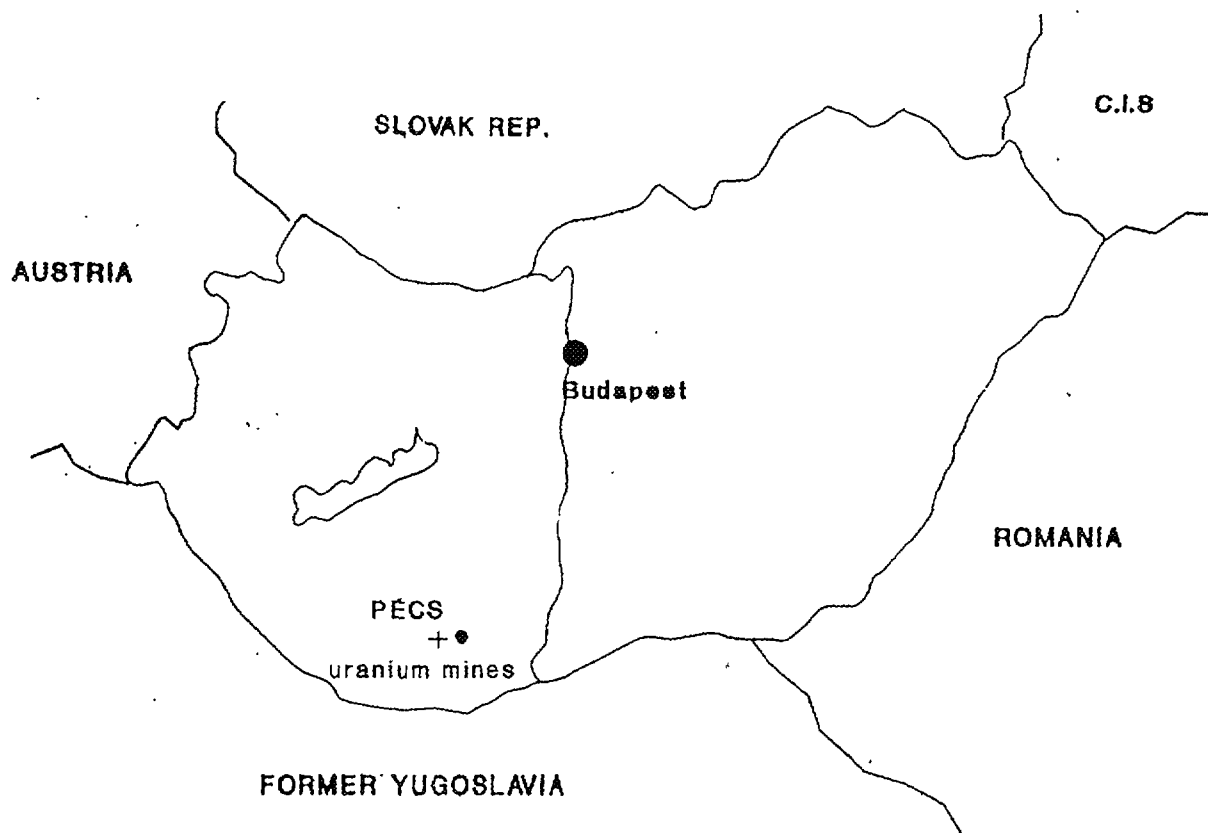


Figure 1.

### 3. ENVIRONMENTAL IMPACT

Around the mine and mill sites, the environmental contamination has originated from mining activity, waste rock piles, transportation and storage of ores, milling activity, tailings pond and heaps for leaching. Figure 2. gives a general view of mining and milling site and of settlements in surroundings of the site [2, 3, 4].

#### 3.1 Mining activity

The 1st and 2nd mines were opened during 1955-56, and the 3rd mine in 1961, the 4th mine in 1971 and at the end the 5th mine in 1988 were in the operation. In the course of mining the uranium content in the ore has quite changed, so at the very beginning the grade was close up to the value of 0.2% and now is around or below 0.1%. Because there was a lack of uranium ore, the 1st mine was closed in 1971, and the 2nd mine in 1988.

The annual ore production was 700-800 thousand tons in the close past, but presently production is 400-600 thousand tons. In the mines the air content of the radon activity is 1800-2000 Bq/m<sup>3</sup> and the uranium dust concentration is 1.-1.5 Bq/m<sup>3</sup>. According to the activity the radon release from the mines through the ventilation system is 10-20 TBq and uranium dust release is 3-3.5 GBq in one year. Regarding the grade, the ore is sorted out at the mines below the value of 100 g uranium/t ores, and until now the accumulated amount of waste rocks is 18 million tons. The rocks have been emplaced in spoil banks covering an area of 450 thousand m<sup>2</sup> at 7 different sites around the mines. The average uranium content in the rock is about 50g/t and the average specific activity including all isotopes of uranium series

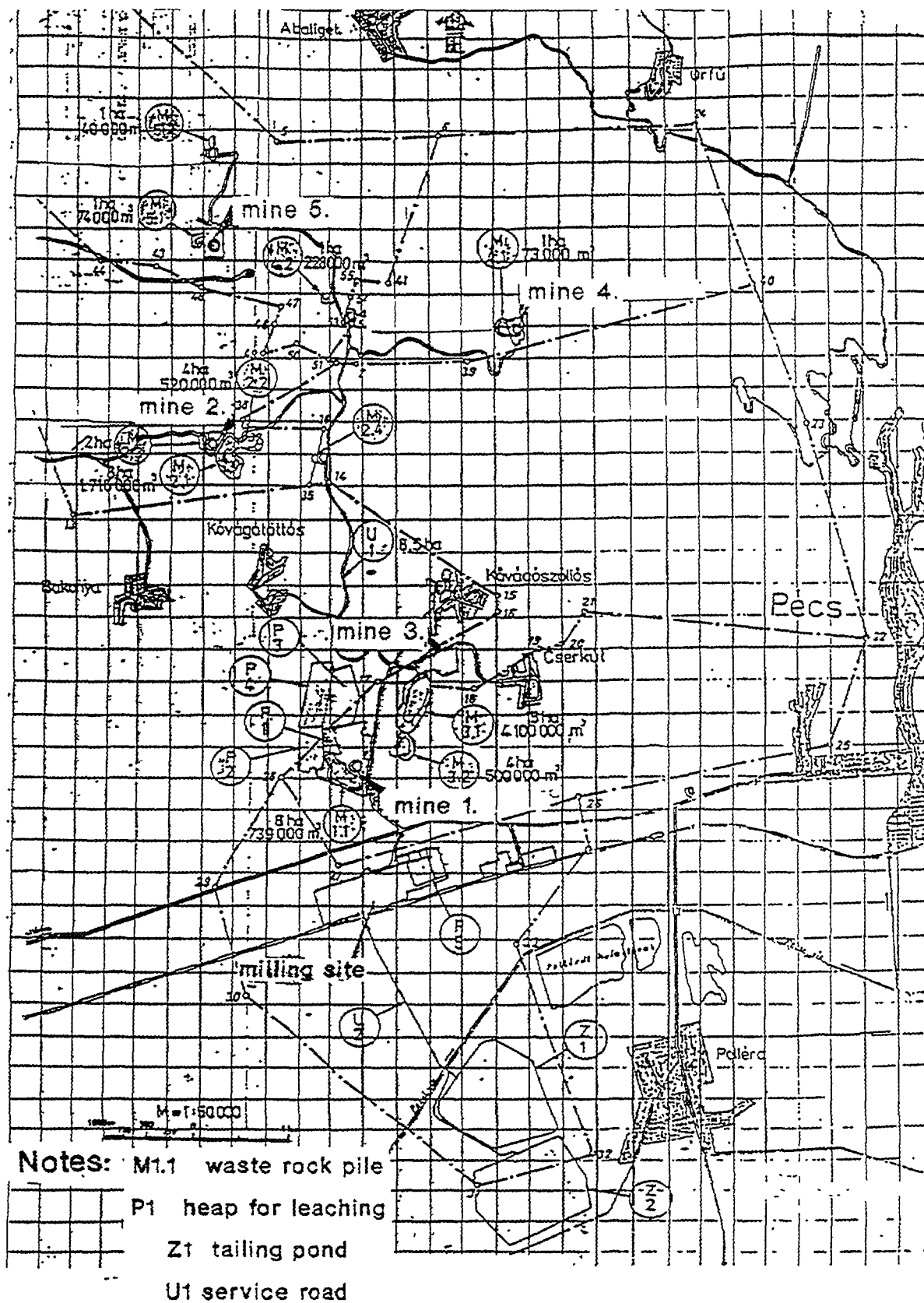


Figure 2.

is around 8 Bq/g. The radon concentration above piles is 5-40 Bq/m<sup>3</sup> and the associated dose rate is 400-1200 nGy/h. The data are the following:

Annual ore production in the past:	7-8 10 <sup>5</sup> ton
at present:	4-6 10 <sup>5</sup> ton
Radon activity concentration in the air of mines:	1800-2200 Bq/m <sup>3</sup>
Uranium dust concentration in the air of mines:	1.-1.5 Bq/m <sup>3</sup>
Annual radon release from the mines:	1.-2. 10 <sup>13</sup> Bq
Annual uranium dust release from the mines:	3.-3.5 10 <sup>9</sup> Bq
Average uranium content in the rock:	50 g/t
Average specific activity of the rock:*	8.2 Bq/g
Dose rate above piles:	0.4-1.2 μGy/h
Radon concentration above piles:	5 -40 Bq/m <sup>3</sup>

\*including all isotopes from U-series

### 3.2 Transportation, storage

While the ores are transported from mine to mill the territory found along both side of the private road has been contaminated with dust containing uranium. The service road is length of about 2 km, and the value of contamination is 0.1-1. kBq/m<sup>2</sup>, and hence the value of the dose rate is 0.2-1. μGy/h.

Before milling the ore is stored and it gives the other possibility for release, like a radon emanation and dust dispersion. In the past the amount of stored ore was 8-10 thousand tons, now a few thousand tons, and the value of radon flux above ores is 3-4 Bq/m<sup>2</sup>/sec.

### 3.3 Milling activity

The milling process begins with breakage, the amount of 3400 t/day was broken in the past, but nowadays half of this is performed. The next step is the radiometric sorting where the ore is sorted out according to the uranium content. The amount of reject ores is about 500-1000 t/day. After sorting process, a medium and fine grinding is carried out, and then the chain of leaching technology is performed. The amount of tailings discharged into pond is 4500-6500 t/day, it includes the recycled water from the pond. Table I shows the detailed data of discharged tailings. The amount of the annual radon release from all process of milling is 2-3 TBq, and uranium dust release is 1.0-1.5 GBq in one year. The value of the dose rate at the boundary of mill is 0.2-0.8 μGy/h.

### 3.4 Tailings ponds

There are two ponds for retention of tailings. On this area the deep geological basement is granite, above this sand and clay formation can be found in depth of 20-200m, and loess appears in depth of 10-15 m. The ponds have been built with ring dike and drainage system. The bottom of these ponds was prepared with lime. The area of each pond is 1 million m<sup>2</sup>. One of these ponds is not functional (it's full) and this contains 12 million tons of solid particles and 5 million tons of solution. The other pond contains 7 million tons solid and 2 million tons solution phase. Data of the impoundments are the following:

Average uranium content of solid phase:	70 g/t
Average uranium content of solution phase:	0.1 mg/l
Average <sup>226</sup> Ra specific activity of solid phase:	12.6 Bq/g
Average <sup>226</sup> Ra activity concentration of solution:	5.2 Bq/l
Dose rate inside impoundment:	0.4-3 µGy/h
Radon concentration inside impoundment:	40-1100 Bq/m <sup>3</sup>
Radon concentration outside impoundment:	8-10 Bq/m <sup>3</sup>
Gross alpha activity of aerosol outside impoundment:	0.3-0.5 mBq/m <sup>3</sup>

### 3.5 Heap leaching

The other uranium extraction method is the alkaline leaching technology used from 1965 to 1990. The total amount of residues is 7 million tons, and the residues cover an area of 600 thousand m<sup>2</sup>. The base of heaps was lined by double plastic materials. Data of residues are the following:

Average uranium content:	80 g/t
Average specific activity:*	19.5 Bq/g
Dose rate above heaps:	0.4-1.5 µGy/h
Radon concentration above heaps:	5-370 Bq/m <sup>3</sup>

\*including all isotopes from U-series

Table 1

#### Discharged tailings data

Material	Quantity	Isotope (portion of U series)	Activity (Bq/day)
Solid phase (slurry)	920 t/day	U-238 U-235 65 g/t U-234 Th-230 70% of U-series Ra-226 99% of U-series	1.6 10 <sup>9</sup> 3.3 10 <sup>10</sup> 2.7 10 <sup>10</sup>
Sorption phase	5300m <sup>3</sup> /day	U-238 U-235 2g/m <sup>3</sup> * U-234 R a-226 1% of U-series	2.5 10 <sup>8</sup> 2.8 10 <sup>8</sup>
Filtration phase	380m <sup>3</sup> /day	U-238 U-235 3g/m <sup>3</sup> U-234	2.8 10 <sup>7</sup>
Spent resin	20m <sup>3</sup> /year	U-238 U-235 500g/m <sup>3</sup> U-234	8.3 10 <sup>5</sup>

\* after neutralization less than 0.1 g/m<sup>3</sup> in liquid phase

#### 4. CONCLUSION

This territory has complicated hydrological features and to date the examination of site has not been carried out completely. Especially, the water table connection across strata should be investigated in more details, because wells for supplying drinking water to the city of Pécs can be also found at this area.

The lack of the background measurements also involves difficulty for the restoration works, because there is no real radiation level, which will have to be restored.

One of the main problems is that there is a lack of national regulations for uranium mining and milling activities and for environmental restoration, nevertheless the company is working with standards recommended by the international organizations.

#### REFERENCES

- [1] Gy. Szomolanyi: The 30-year period of uranium mining at Mecsek Mountain, Mining No.10, 1986 (in Hungarian)
- [2] The report on the environmental impact of the Mecsek Ore Mines, made by National Research Institute of Radiobiology and Radiohygiene, 1991 (in Hungarian)
- [3] I. Barany, I. Vados, A. Varhegyi: The radiological impact of Mecsek Ore Mines to the natural environment, 17th Workshop on Radiation Protection, 6-8, May 1992, Balatonkenese (in Hungarian)
- [4] L. Juhasz, B. Kanyar, N. Fulop, A. Kerekes, I. Vados: Radiohygienic study of the uranium mining and milling for the promotion of the environmental remedial action, 18th Workshop on Radiation Protection, 12-14 May, 1993, Balatonkenese (in Hungarian)

# **A PROJECT CARRIED OUT IN ITALY TO SECURE A SITE CONTAMINATED BY CS-137 OF UNKNOWN ORIGIN**

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## **Abstract**

This paper describes the main phases of a work carried out to secure a site consisting in an industrial waste disposal contaminated by Cs137 of unknown origin. A campaign survey to evaluate the amount of radioactivity on the surface of the storage waste plant was undertaken; then, a complex process was set up and carried out, taking into consideration the environmental conditions, the characteristics of the waste involved and of the materials added to it, the waste transport conditions, the structure and management of the waste storage plant, in order to realize Cs137 ions mobility and diffusion in that peculiar situation. A first sealing of the surface was carried out to secure the site at least for some months, in order to let technicians and vehicles go freely and safely on the surface of the plant taking samples for laboratory analysis. A special drilling system, capable of picking up contaminated samples from the depth of the mass of waste without any (or almost any) contact with the environment, was then performed. As conclusion of that investigation, an "on site" security action was decided, in consideration of the great amount of waste involved and in order to avoid that some contaminated waste could come in contact with the environment. The radiological survey, the interim actions for a temporary securing of the site, the drilling campaign and the system used to pick up samples, as well as the results of laboratory analysis are described in the lecture.

## **1. INTRODUCTION**

During the latter half of 1989, just near the nuclear Power Plant of Caorso (Piacenza, Italy), an increase of Cs137 in the water of Po river was detected during routine controls. The level recorded was about five times the background value, but anyway far below the safety limit. For safety reasons, the presence of that radioisotope in the power plant site was monitored, even if, by Cs137/Cs134 ratio determination, it had already been pointed out that the radiocontamination was not due to any fission product correlated with any nuclear power plant operations or accidents, Chernobyl included.

As the phenomena had been still continuing, in April 1990 a survey on the Po river sediments and some of its affluents started, in order to individuate, by means of a well known geochemical survey method, the origin of that contamination and to locate it. A first Cs137 contamination source was then localized near Saronno, about fifty kilometers north-west of Milan, in two scrap aluminum refineries. By a heavy control of many scrap refineries all over Italy, and through administrative investigations on foreign suppliers, it was then pointed out that two more firms near Brescia, about 90 Km east of Milan, were contaminated (fig.1).

As melting salts from the drum ladles of the latter two refineries were discharged into a specific dump facility, i.e. into a disposal for industrial waste, that facility was controlled and it was found to be contaminated by Cs137 as well.

Our task from Lombardy Region Administration was to put up a project and to carry out all the actions necessary to decontaminate the four refineries, to estimate Cs137 quantity in the dump facility and to secure the site of the same facility. This lecture mentions only the actions concerning the waste facility.

First of all, a survey for the radiological characterization of the site, i.e. the waste disposal itself and the surrounding areas, was undertaken: a complex process was then set up and carried out, taking into account the environmental conditions, chiefly from the geologic and hydrogeologic point of view, the characteristics of the waste involved and of the materials added to it, the waste transport



Fig. 1

conditions, the structure and management of the waste storage plant, in order to realize Cs137 ions mobility and spreading in that peculiar situation.

A first temporary sealing on the surface was then carried out to secure the site at least for some months, in order to let technicians and vehicles go freely and safely on the surface of the plant, taking samples from the depth of the mass of waste for laboratory analysis. A special drilling system, capable of picking up contaminated samples from the depth without any (or almost any) contact with the environment, was then performed.

As conclusion of that investigation, an "on site" action was decided to secure the site, in consideration of the great amount of waste involved and in order to avoid that some contaminated waste could come in contact with the environment.

The surface radiological survey, the interim secure action, the drilling campaign and the system used to pick up samples, as well as the results of laboratory analysis are described in the lecture.

The further part, concerning remediation, will be focused in the lectures to be presented in the subsequent workshops scheduled in 1994.

## 2. THE REFERENCE FRAME

Before any securing and restoration action could be undertaken, it was necessary to collect as many data as possible about the storage plant and the way of managing it, about the characteristics of the wastes there dumped and the materials added to them, as well as of the discharging schedules during the previous months, in order to realize the behaviour and diffusion of Cs137 in that peculiar situation.

What the essential data: the dump facility was a waste storage plant consisting of 7 basins, about 50.000 m<sup>3</sup> each and a total area of about 30.000 m<sup>2</sup>, with an average depth of the waste themselves of about 14 m: in it, about 280.000 m<sup>3</sup> of the mentioned industrial waste had already been discharged at that time (fig. 2, 3).

At the bottom of every basin there was a clay layer, 40 cm depth, covered with a geomembrane, i.e. a polyethylene sheet (fig.4-7) to retain the rain water in case percolating in the waste mass during the discharge operations.

Over and under the polyethylene sheet, in contact with it, there was two distinct drainage networks: the one under the sheet had the task of revealing any leakage of the sheet itself; the other over, the task of collecting the percolating water, conveying it into a water tank.

The waste materials discharged in that plant were the melting salts from the two mentioned aluminum refineries, the most of them consisting of a fine powder ( fig. 8, 9) to which a 10% of compact blocks were added: to the powder, to prevent powder spreading in case of heavy wind, had continuously been added somewhat of terrain reach in clay.

All materials were discharged in every basin going up layer after layer in steps of about 2-3m, for a total height of about 13-14 m.

In this situation, we realized that Cs137 had still to be inside the facility, and could be monitored inside the waste disposal itself, chiefly in the drain network, which is in the drainage tanks, as well as on the surface of the disposal and in the body of the waste.



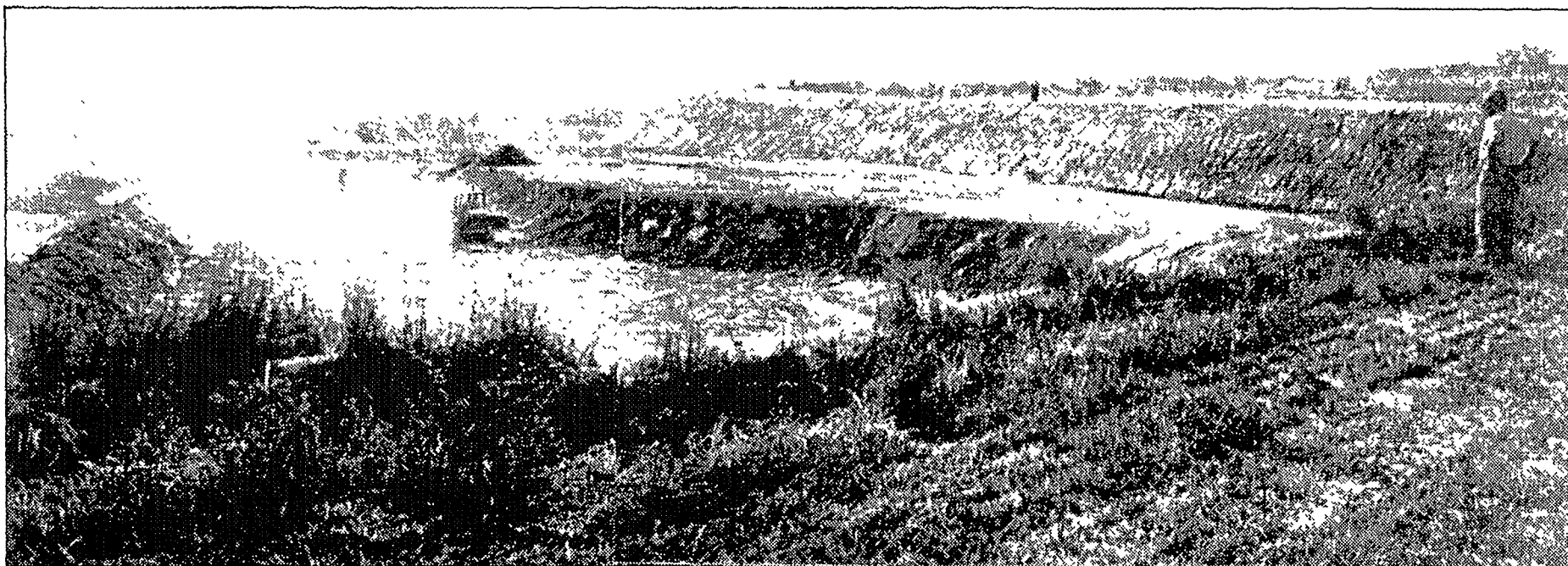


Fig. 2 - Waste disposal of Capriano del Colle (BS) - Comprehensive view.

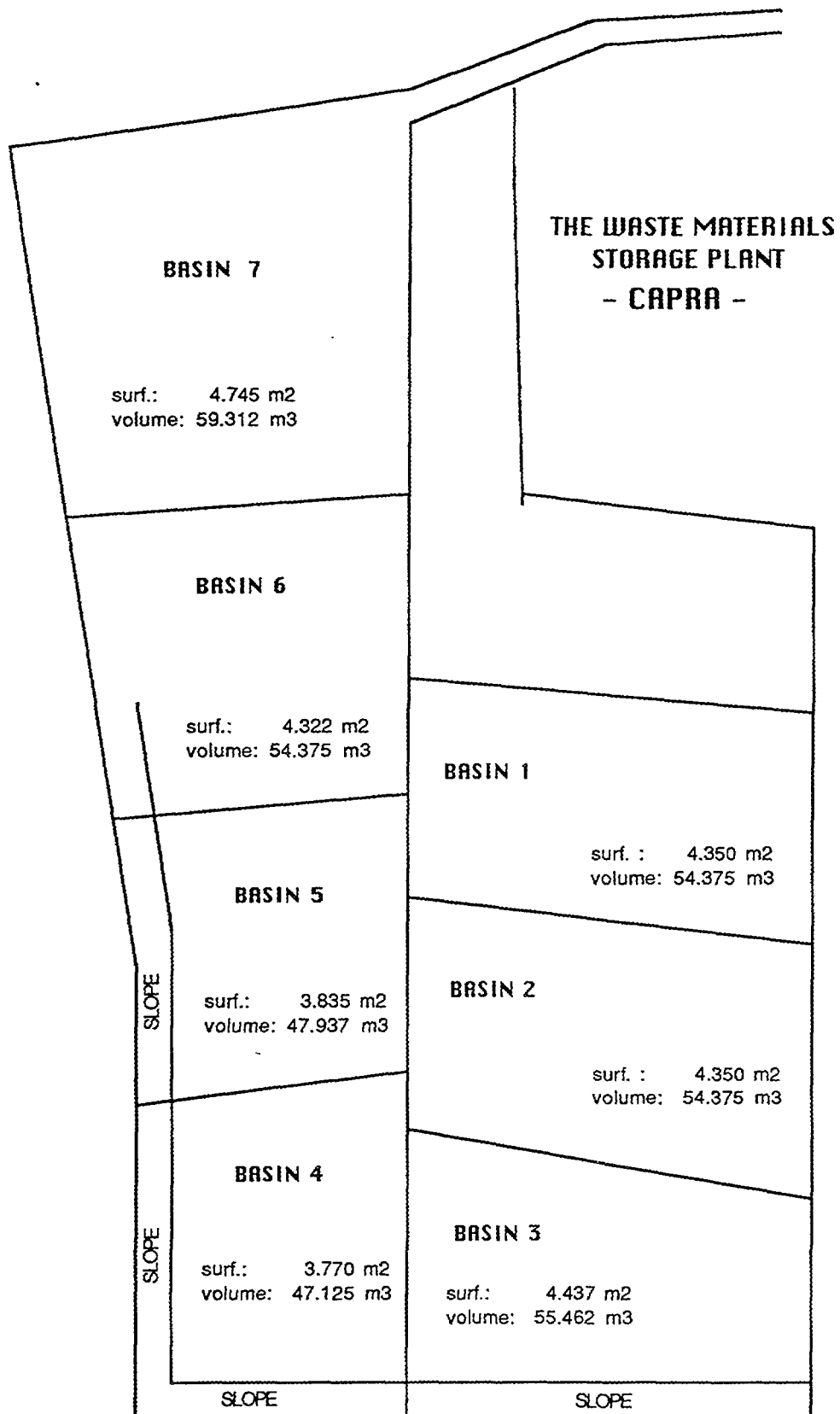


Fig. 3: Geometric Figures

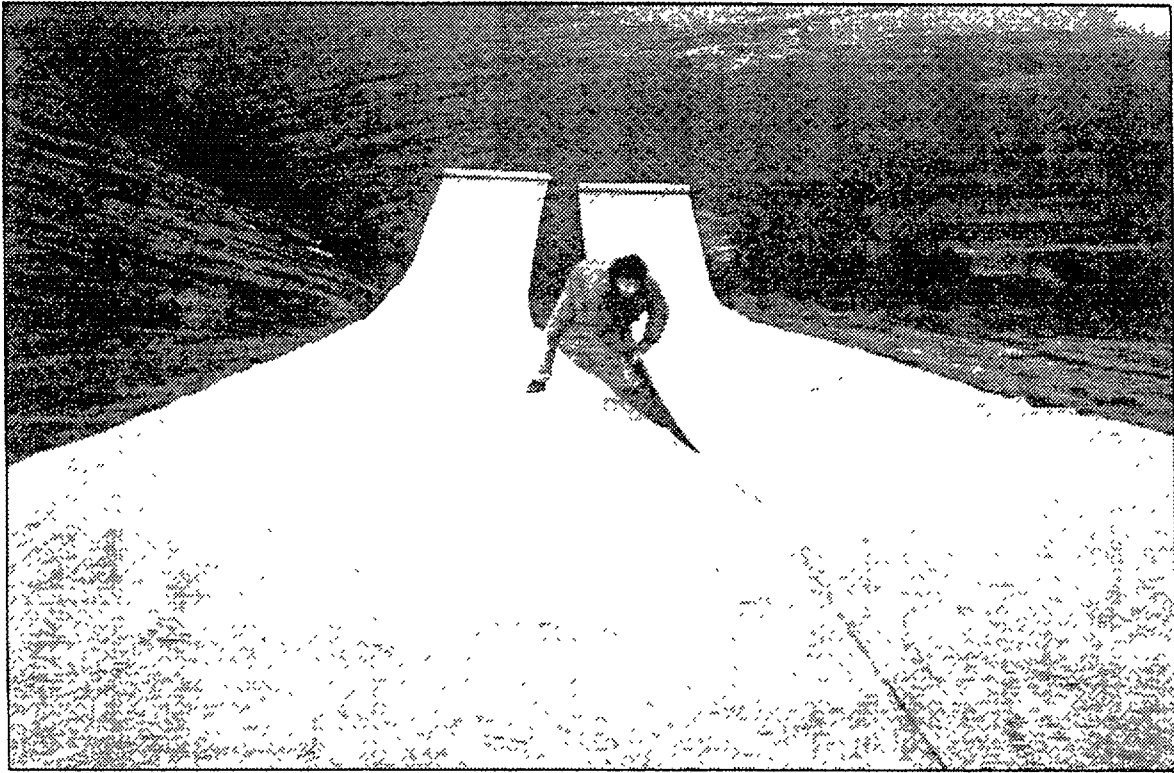


Fig. 4 - The laying of the geomembrane on the slope.



Fig. 5 - The geomembrane on the bottom of a basin.



Fig. 6 - The geomembrane on the slope of basin 7.

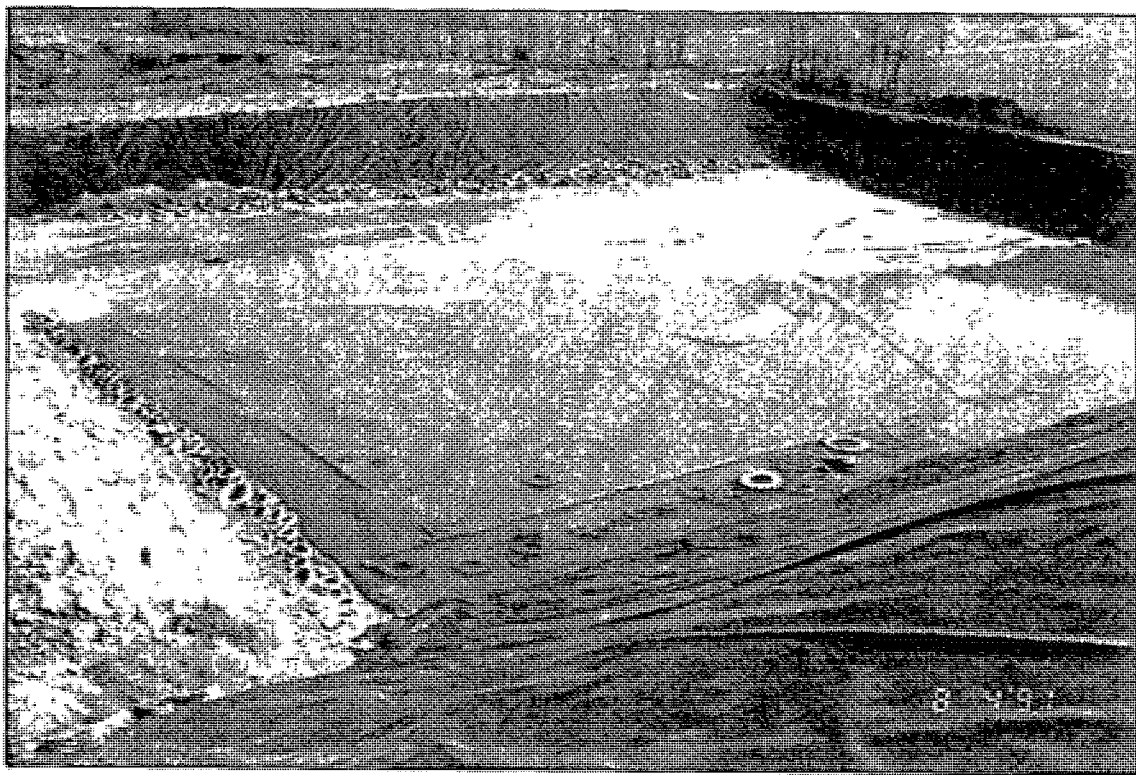


Fig.7 - Basin 7, still empty at the time of the securing intervention.



Fig. 8 - Waste discharging.

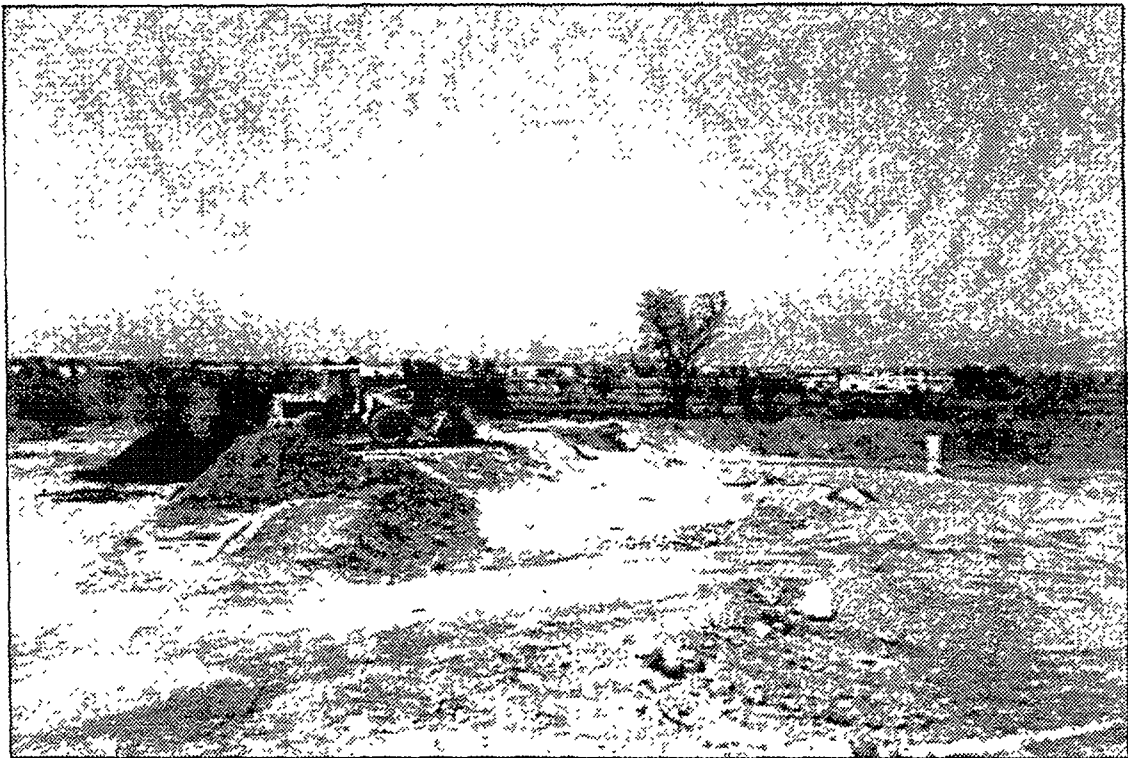


Fig. 9 - Waste discharging.

### 3. THE RADIOLOGICAL SURVEY

Before any securing action could be initiated, a radiological characterization of the dump facility had to be performed, i.e a detailed picture of the concentration and spatial distribution of the radionuclide. Thus, on behalf of an ENEA's team, a first radiological survey on the surface of the waste plant was undertaken.

The initial effort was directed to delineate areas of major surficial contamination by an aerial screening survey undertaken using hand held instruments: then, radioactivity was measured on surficial and shallow samples; the results are reported in tables 1 and 2.

TAB. 1

#### $\gamma$ ACTIVITY FROM FIELD MONITORING

POINT	NUCLIDE	ACTIVITY (Bq/kg)
1	Cs 137	-
2	Cs 137	9,01 E+01
3	Cs 137	1,23 E+01
4	Cs 137	-
5	Cs 137	-
6	Cs 137	2,82 E+01
7	Cs 137	1,03 E+01
8	Cs 137	1,81 E+01
9	Cs 137	1,33 E+01
10	Cs 137	1,34 E+01
11	Cs 137	1,05 E+01
12	Cs 137	3,18 E+01
13	Cs 137	9,10 E+00
14	Cs 137	2,30 E+01
15	Cs 137	2,30 E+01
16	Cs 137	2,01 E+01
17	Cs 137	5,48 E+01
18	Cs 137	1,66 E+01
19	Cs 137	5,71 E+01
20	Cs 137	3,91 E+00
21	Cs 137	8,16 E+00
22	Cs 137	5,53 E+01

TAB 2  
 $\gamma$  ACTIVITY OF PHISICAL SAMPLES .

	DEPTH (cm)	ACTIVITY (Bq/kg)	DEPTH (cm)	ACTIVITY (Bq/kg)
A	0÷10	2,2 E+00	10÷20	1,6 E+01
C	0÷20	2,0 E+00		
D	0÷10	9,9 E+00	10÷30	7,9 E+02
E	0÷50	6,0 E+00		
F				
G	0÷10	3,1 E+01		
H	0÷10	1,2 E+01		
I	0÷20	1,2 E+02		
L				
M			20÷40	4,0 E+01
N	0÷30	5,9 E+01		
O	0÷10	2,7 E+02		
P	saline block	2,8 E+04		
Q	saline block	1,8 E+04		
R	0÷10	1,2 E+01		
S	0÷30	6,5 E+00		
T	0÷10	1,9 E+01		
U	0÷100	2,0 E+01		
V	0÷10	3,0 E+01		
Z	0÷10	2,2 E+00		

A precise delineation of the boundary of the surficial contamination was obtained, showing that only in correspondence of basin n°3 the radioactivity was significant, with values as high as 28 Bq/g in samples picked up from some saline compact blocks along the slope of the dump.

In other words, from the first results, radioactivity seemed to be present mostly in the blocks of the melting salts directly coming from the drum ladles of one of the two refineries, in a buried layer some meters below the surface.

#### 4. THE INTERIM ACTIONS

In the face of such a huge mass of waste to be investigated and secured, our immediate care was to plan actions capable of avoiding any spreading of the radiocontaminant and, successively, actions capable of monitoring Cs137 inside the body of the waste and to find out the distribution of that radioisotope in it.

An interim action was then carried out, based on a surface clay covering, in order to make the surface of the site safe from the radiological point of view, and to let people and vehicles go freely all around taking measurements and samples, obviously with some precautions.

Clay was chosen because of the well known behaviour of Caesium in that material, where it is captured by adsorption and ion exchange mechanisms. A special clay, with a very good permeability coefficient (  $k=10^{-8}$ -  $10^{-9}$  cm/s ) was chosen and supplied from a controlled quarry.

On the horizontal top plane of the disposal, a 10 cm sand layer in contact with the waste itself was laid, for aeration reasons; then, on both the horizontal plain and the slopes, a further 20 cm layer of clay was laid. The clay layer was compacted to the right density, following specific prescriptions previously obtained by setting up the rolling process procedures on a specific embankment. In fig. 10-11 it is possible to see some steps of the process undertaken.

After the rolling of the clay, on the surface of the slopes a special covering fabric to prevent any meteorological erosion or incidental landslides was laid. At the foot and in the middle of the slopes, some suitable grips were built in order to prevent any water erosion, by conveying rain water directly out of the facility. After all these actions, the facility could be considered safe, from a meteorological point of view, say for a period of time of at least six months, enough to carry out deep drilling operations and define all the actions necessary for the final sealing.

## 5. THE DRILLING CAMPAIGN

The aim of the drilling campaign was to pick up deep samples from the mass of the waste in order to determine Cs137 distribution and quantity. Anyway, it was necessary to save two antithetical requirements: 1) having as many samples as possible for a precise result and 2) avoiding any new contact of the contaminated material, from the depth of the mass of the waste, with the environment.

At the same time, it was necessary to carry out all the actions in a relatively short time, say few months, to avoid that the interim cover could be dismantled by meteorological agents, also considering that we were going toward winter.

By the examination of the discharging schedule, it had been forecasted that the basin n°3 could contain the most of the radioactivity. Anyway, a drilling on all the basins was decided, 3 drillings per basin, as showed in fig. 12, except basin n°4, where samples from the depth had already been picked up some months before for control reasons.

The drillings were carried out for almost all the depth of the mass of waste and confirmed the theoretical model: as a matter of fact, the most of the radioactivity was found in basin n°3, where four more drillings were therefore undertaken, exactly drillings 3.4, 3.5, 3.8, 3.9 (fig. 12).

The last three drillings were not vertical, but were carried out with an inclination of about 30°, in that way following the inclination of the slope itself.

## 6. THE PHYSICAL SAMPLING METHOD

The system used for picking up deep samples is worthy to be mentioned, because it was planned in order to avoid any contact of the samples themselves with the environment.

Some PVC tubes were prepared, well fitting the inside core barrel dimensions of the drilling rig, and were located in the core barrel itself during the drilling operations, in such a way that the drilled contaminated material could go directly in them. The PVC tubes became, in such a way, the samples containers themselves.

Fig. 13-17 show the drilling rig and the mentioned PVC tubes; fig. 16 shows how the contaminated material was sealed by terminal plugs; Fig. 18 shows how the samples, inside the PVC tubes, were protected by plastic bags and located inside a sample-box.



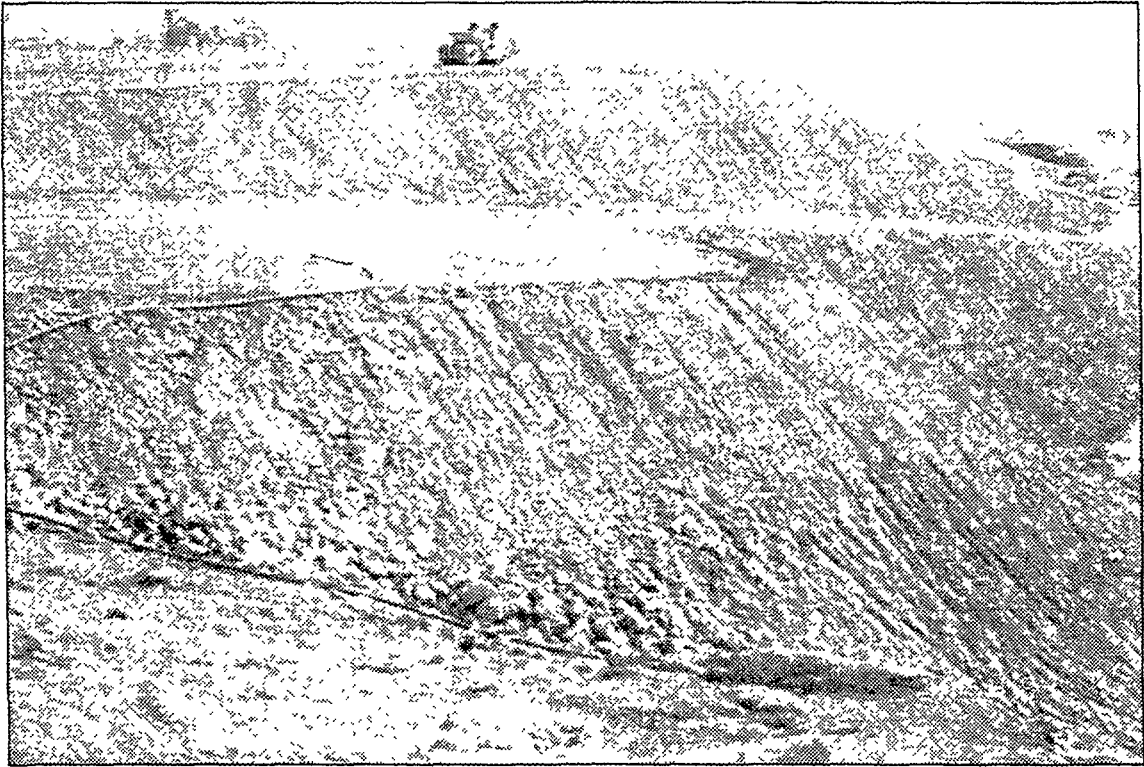


Fig. 10 - The interim clay covering of basins 5 and 6.

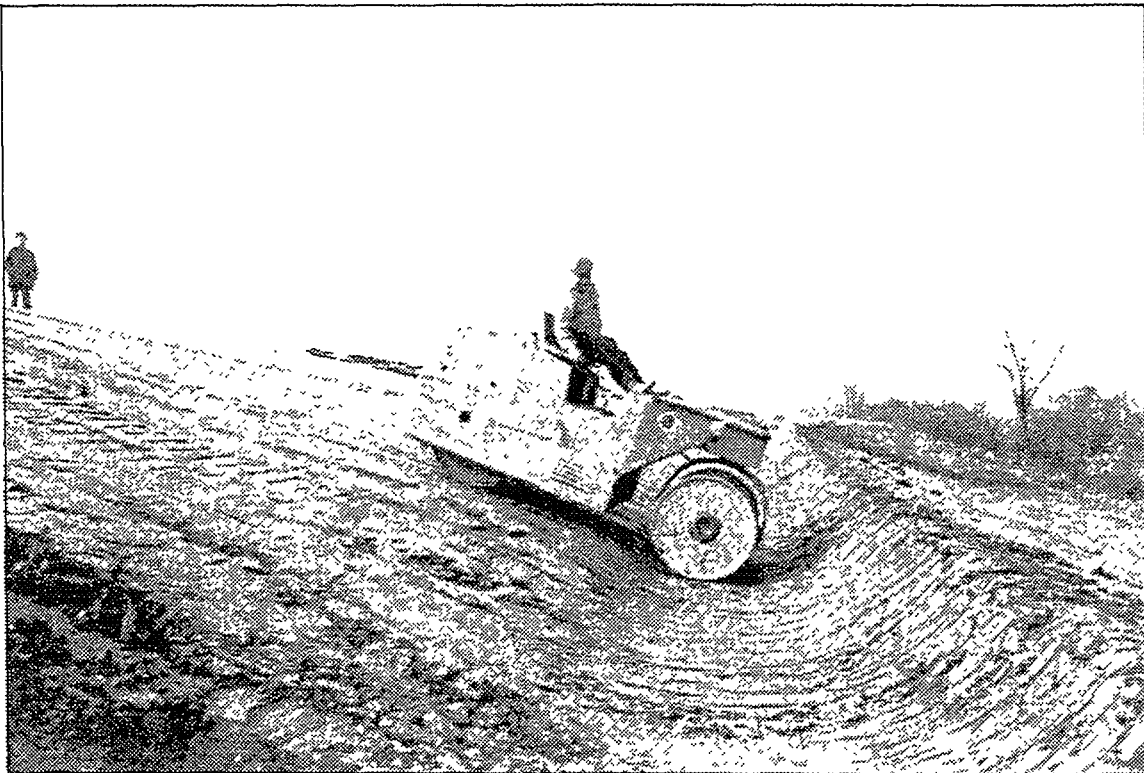


Fig. 11 - A temporary grip for rain water conveying.

*Discarica Montenetto  
di Capriano del Colle (Bs)*

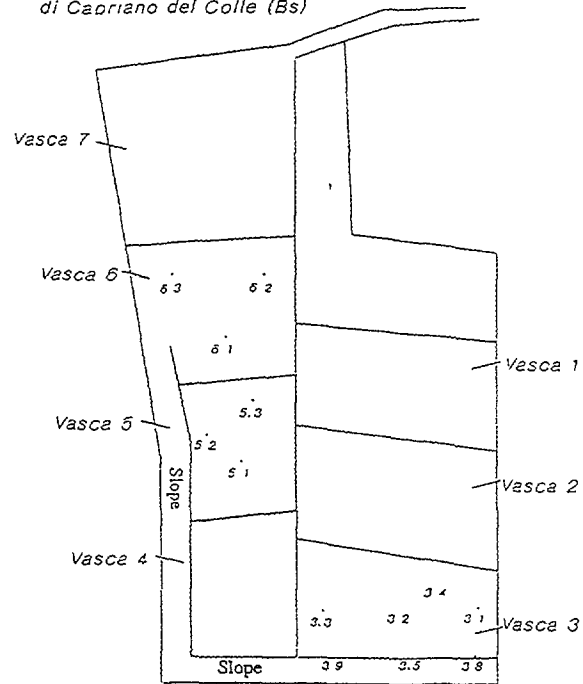


Fig. 12 - The drilling scheme

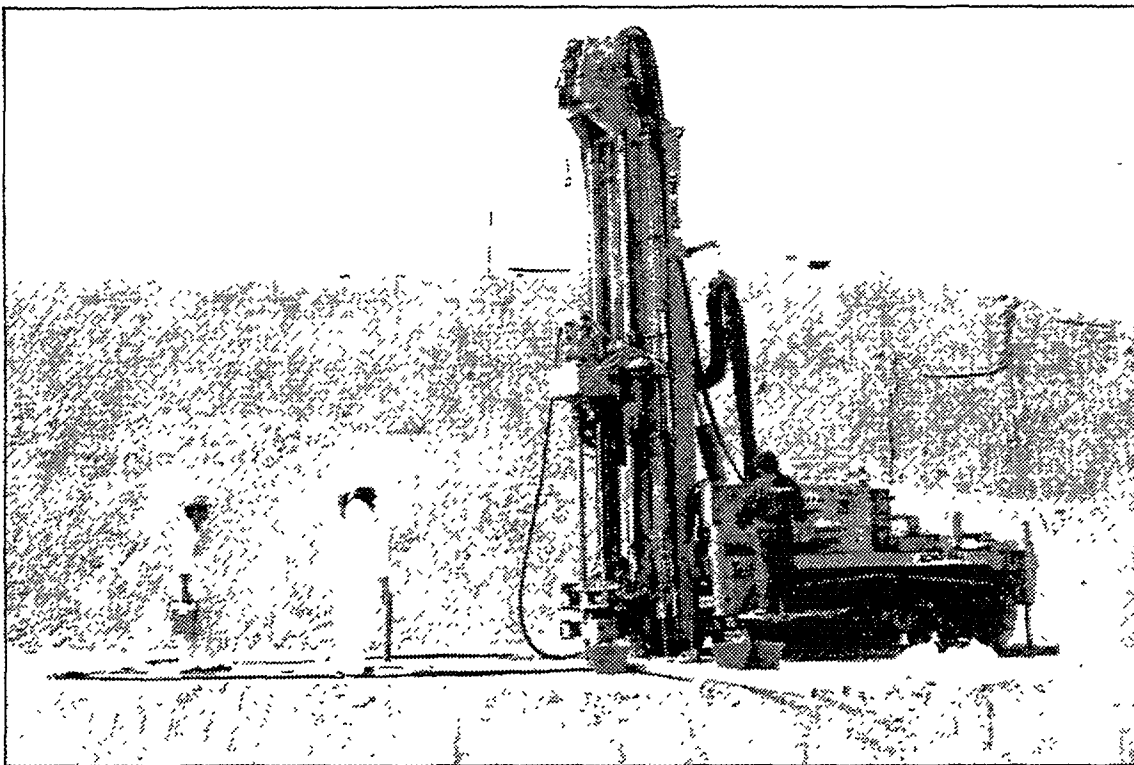


Fig. 13 - The drilling rig for picking up samples.

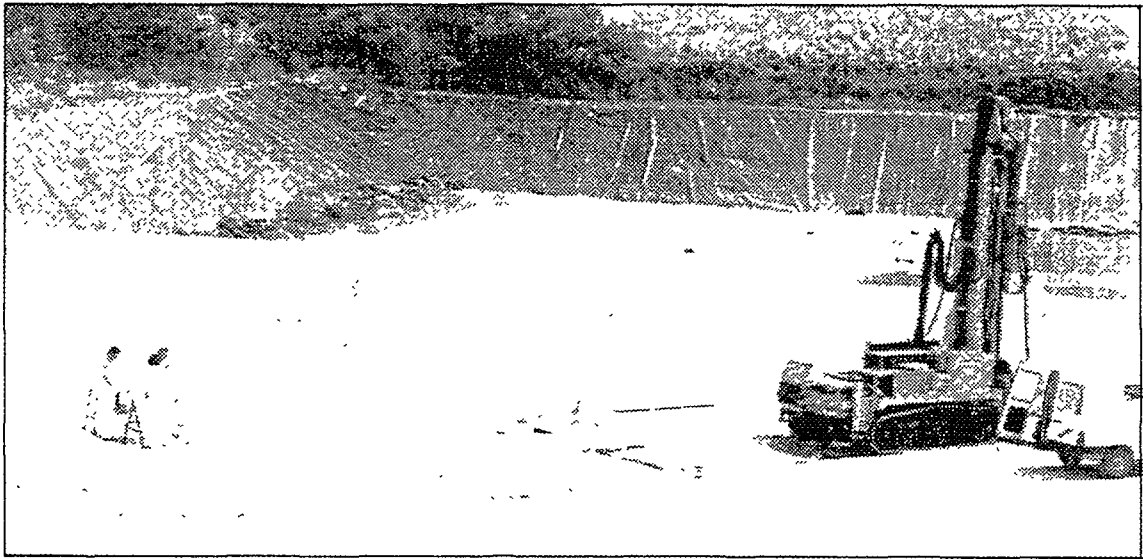


Fig. 14 - The drilling rig for picking up samples.

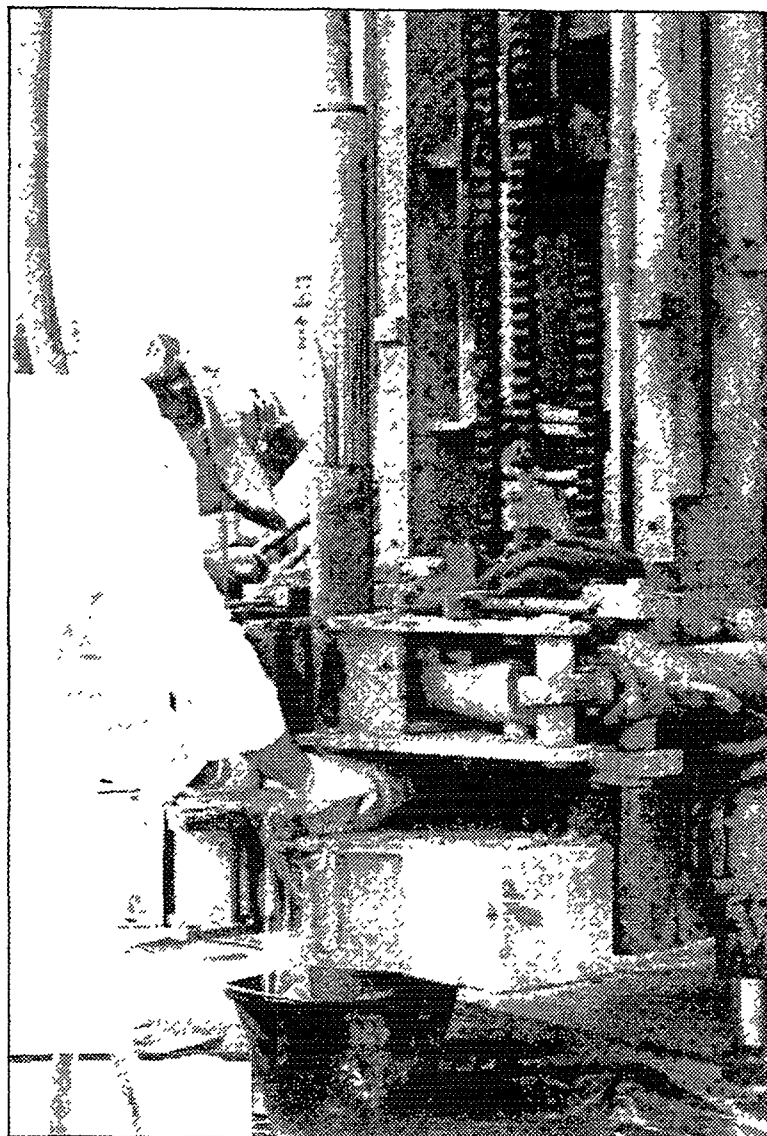


Fig. 15 -The PVC tube while extracting

## 7. THE RESULTS OF THE DRILLING CAMPAIGN

The results of the radioactivity measures of the samples of all the drillings are shown in fig. 19-25, which show the Cs137 profiles along the depth of the waste dump.

In short, the most important results are the following:

- Significant radioactivity has been found only in the basin 3, some meters under the surface
- The total activity of Cs137 was about  $1.1 \times 10^{12}$  Bq (~ 29 Ci)
- In the basins 5 and 6, the average concentration of Cs137 was respectively about 125 Bq/Kg and 6 Bq/Kg.
- The most active samples were the ones from the salt blocks, located in the intermediate layers of waste, according to the information on the discharging schedule and according to the provisional model.
- The most of the radioactivity concentration was found at the edge of the basin 3, in correspondence with the slope, where the concentration of the salt blocks was maximum.

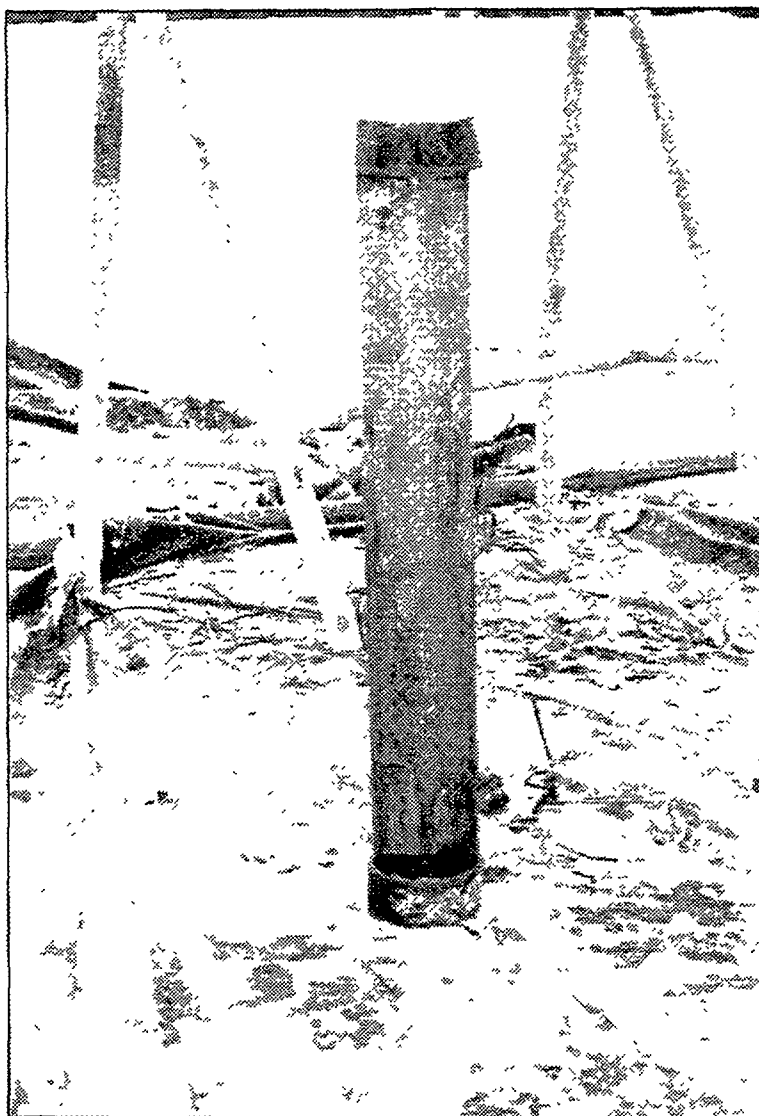


Fig. 16 - The sample container

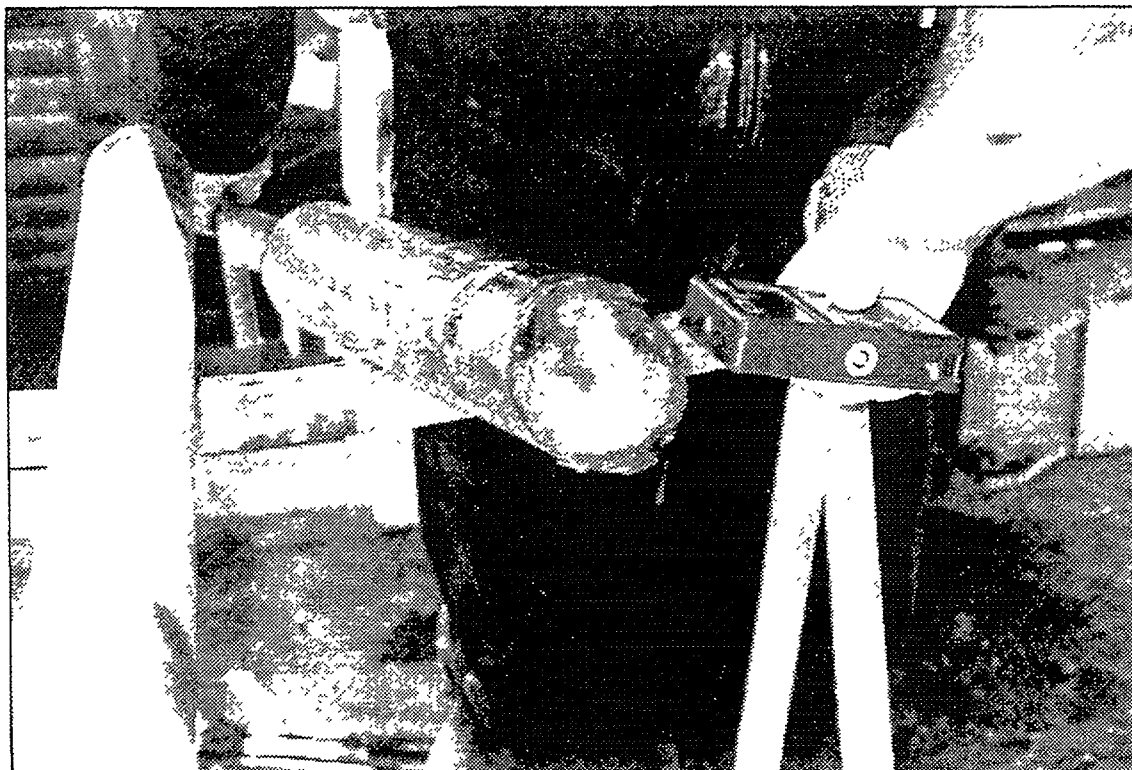


Fig. 17 - View of a sample inside the container.

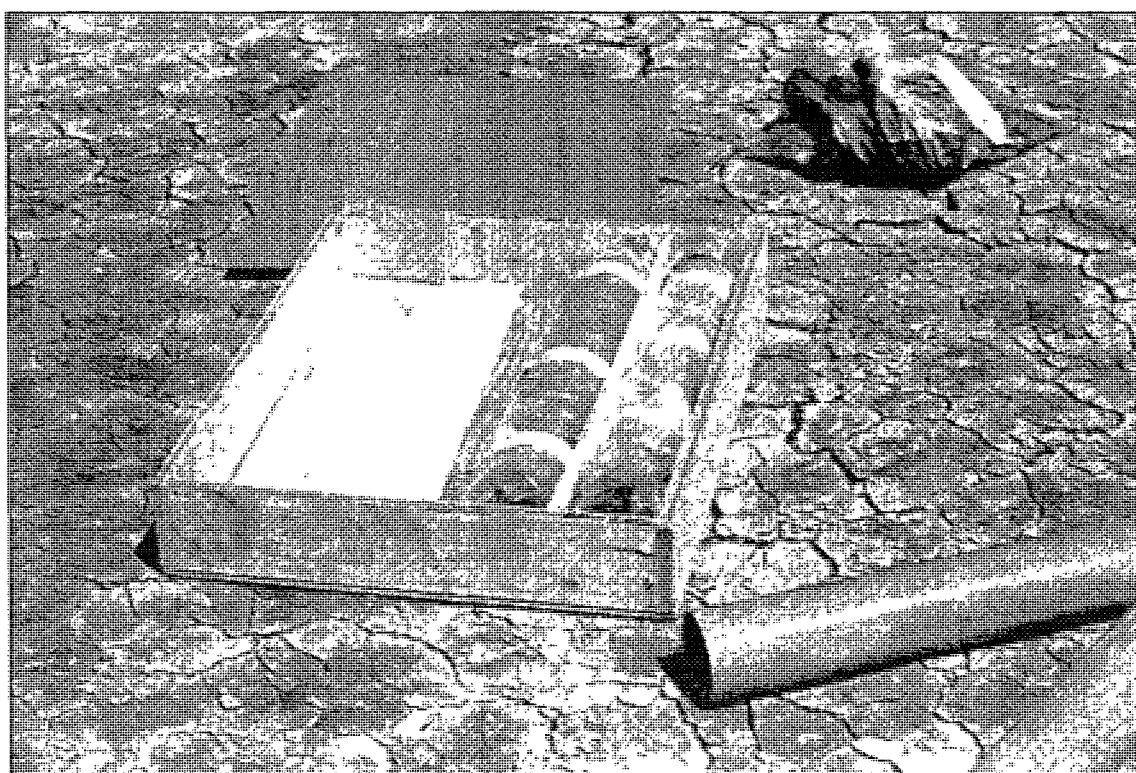
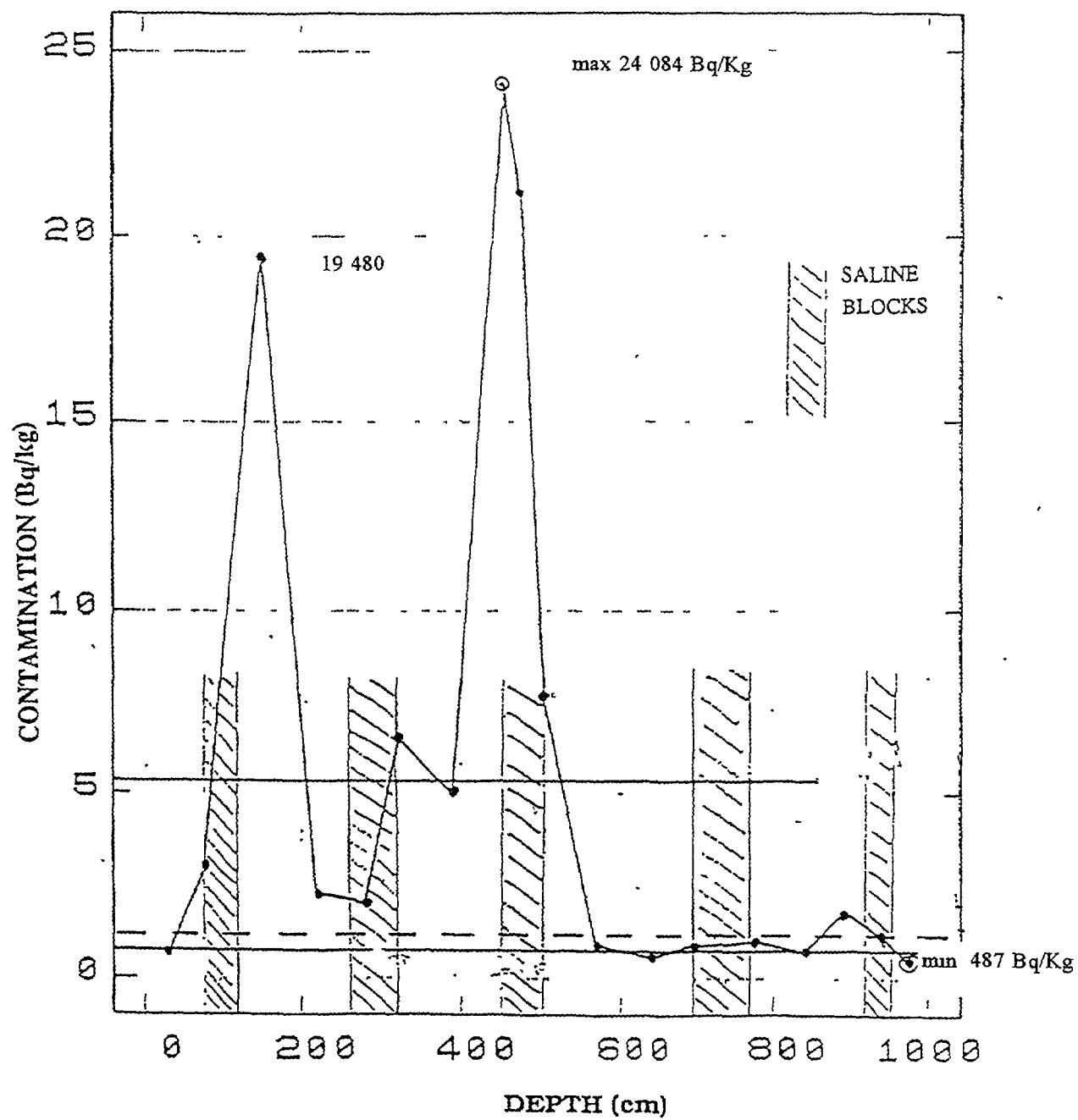


Fig. 18 - Samples container.

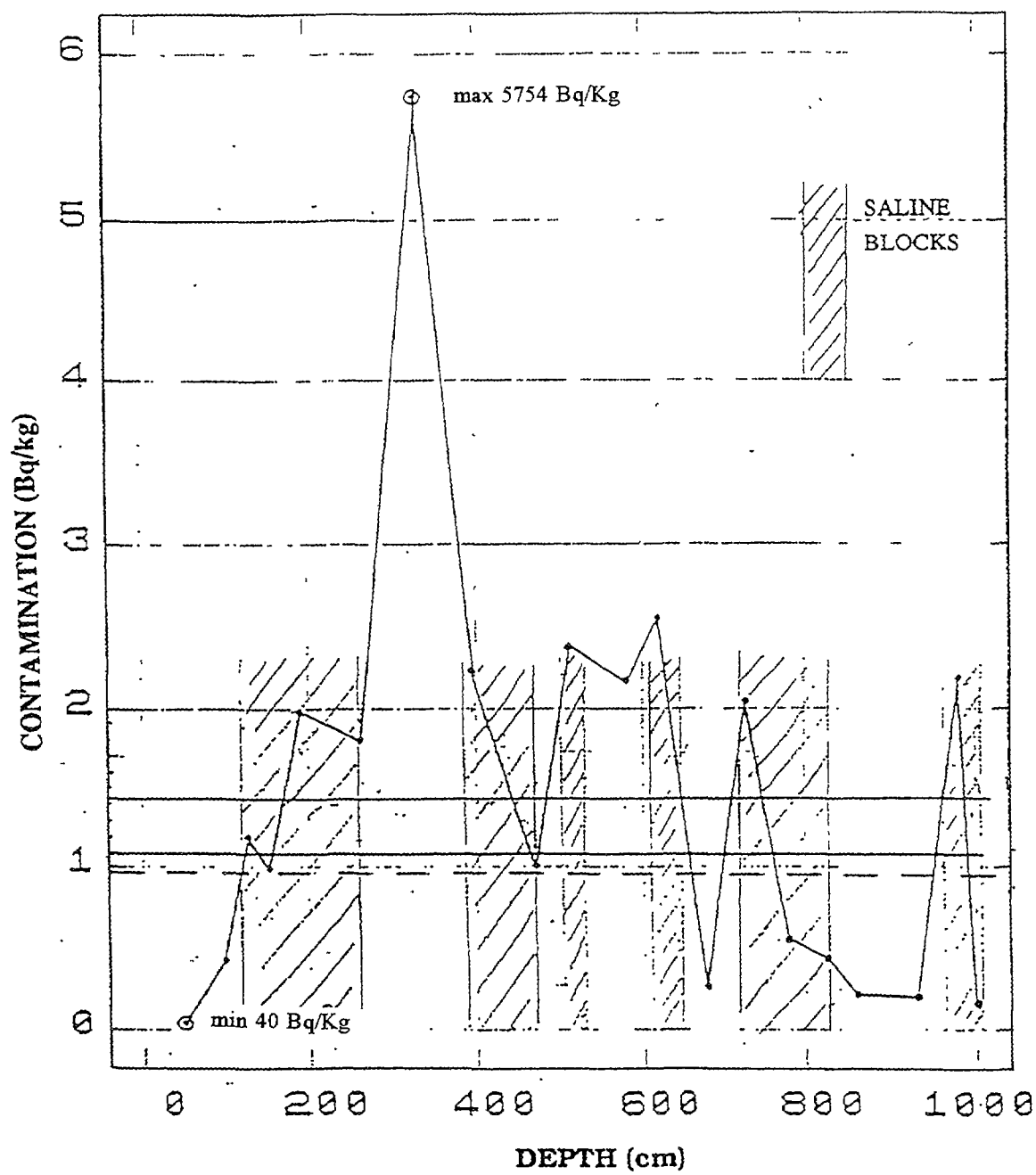
# Cs 137 PROFILE



RANGE 23 597 Bq/Kg  
N. CAMPIONI 18

FIG. 19.

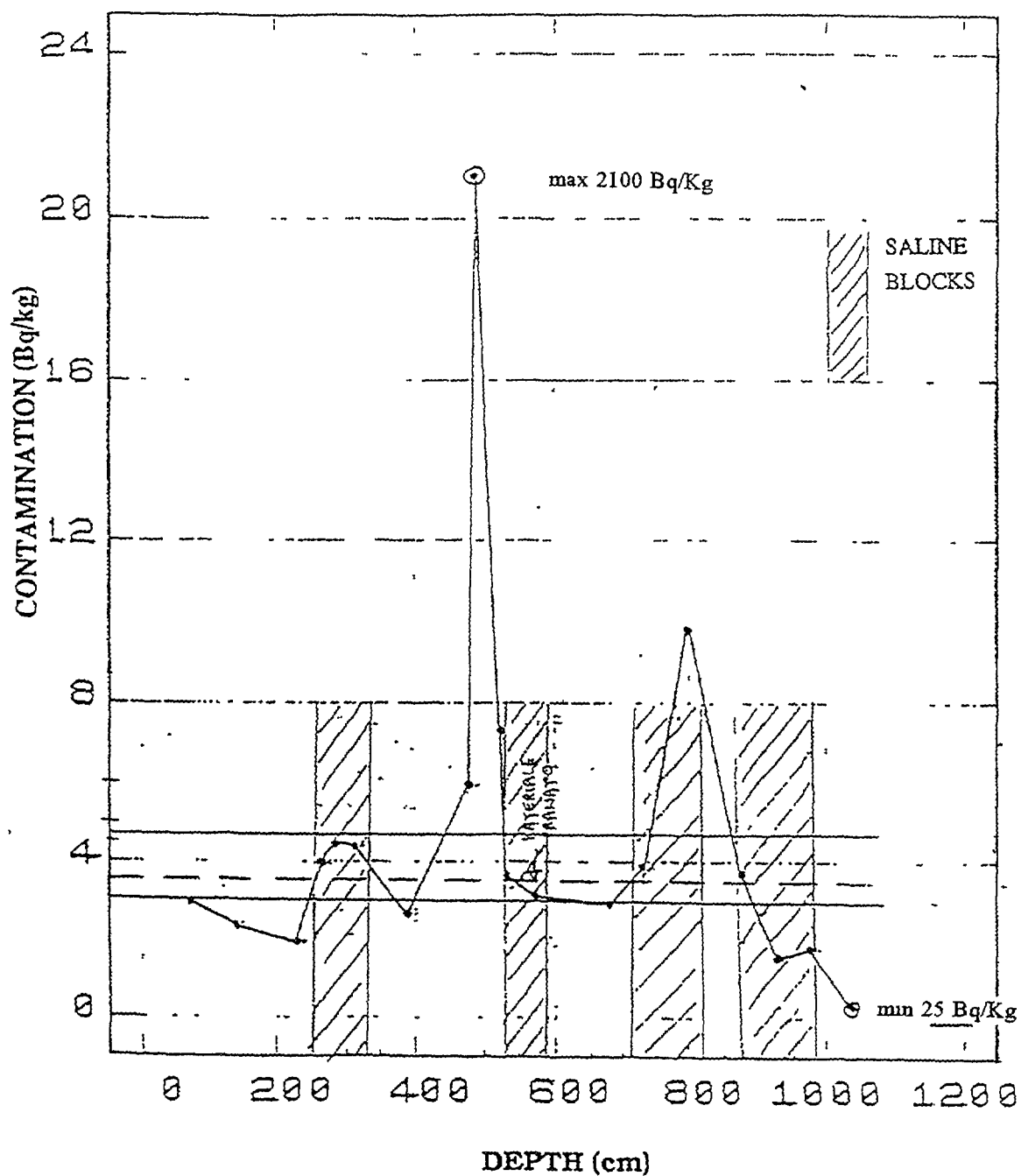
# Cs 137 PROFILE



RANGE: 5714 Bq/Kg  
N. CAMPIONI: 20

FIG. 20.

# Cs 137 PROFILE

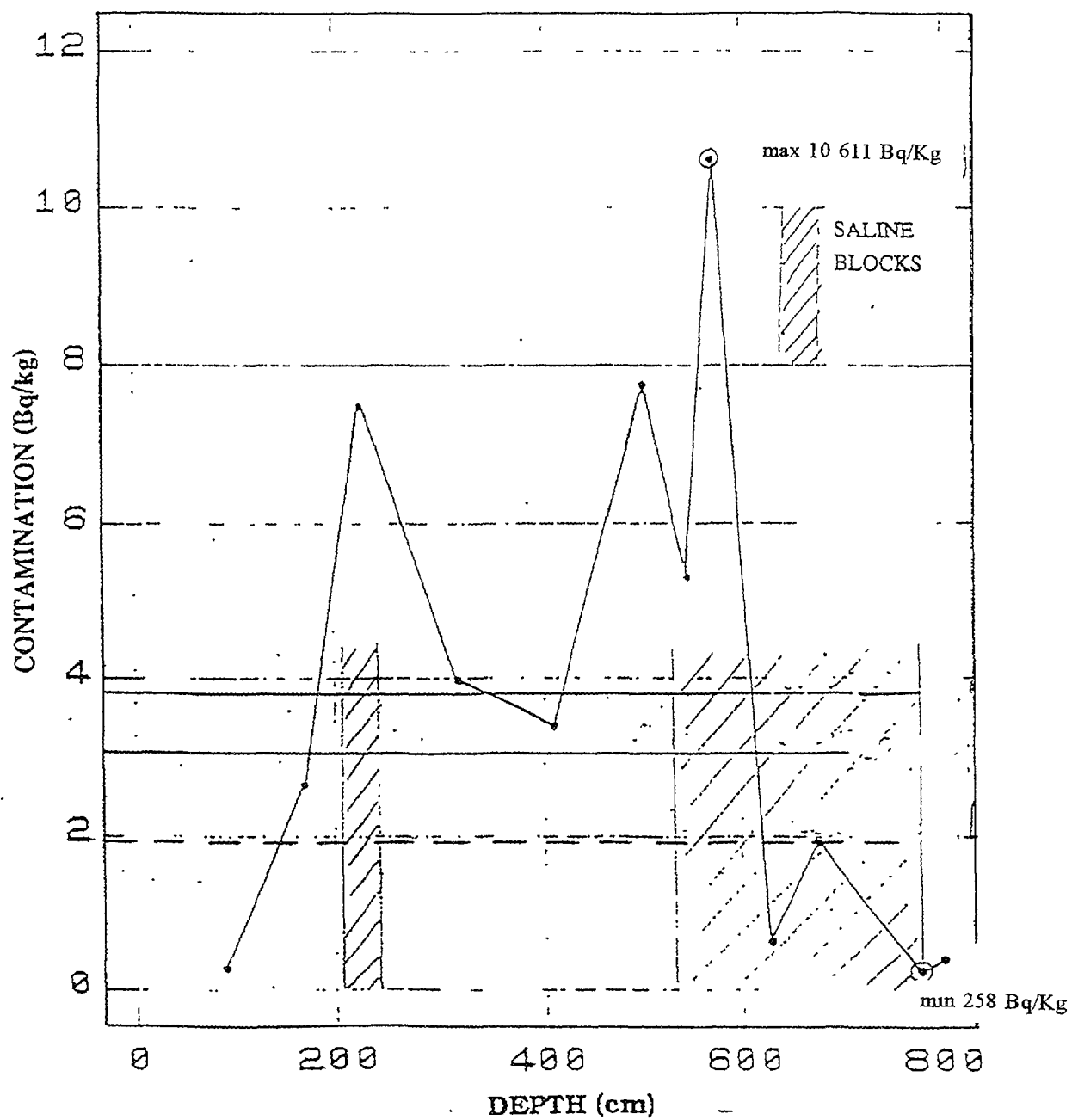


RANGE 2075 Bq/Kg  
N CAMPIONI 19

FIG 21.



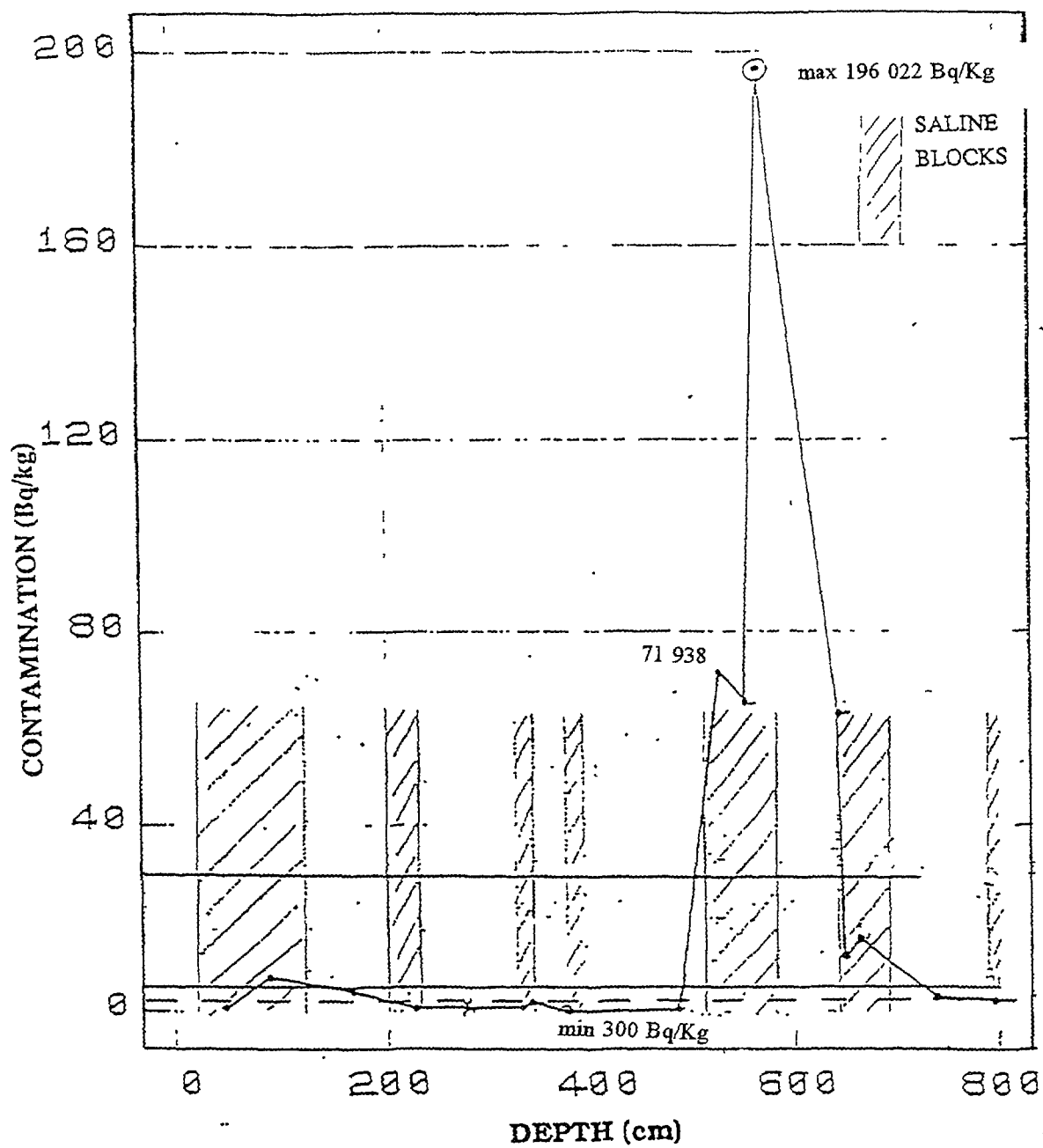
# Cs 137 PROFILE



RANGE 10 353 Bq/Kg  
N CAMPIONI 12

FIG. 22.

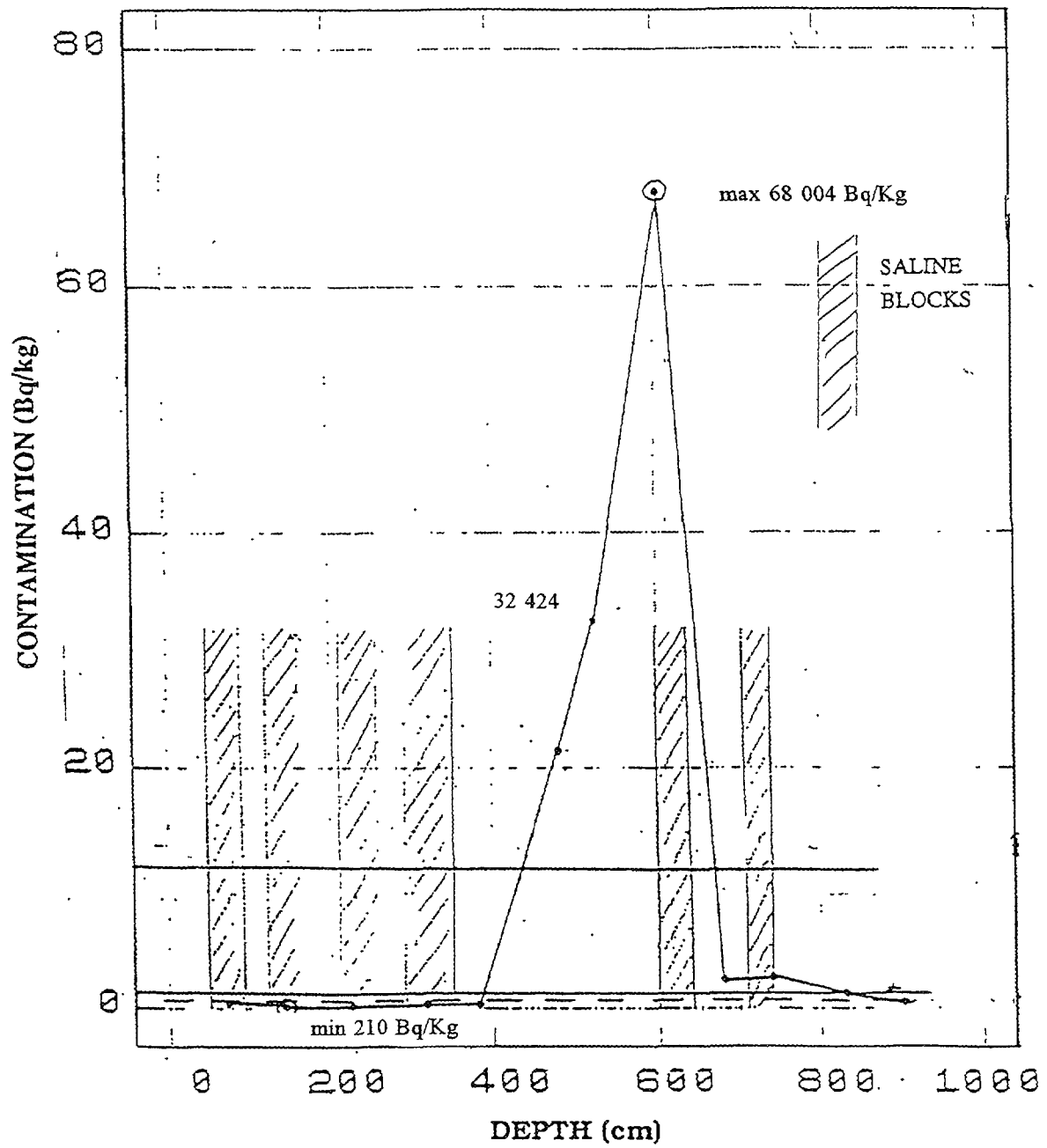
# Cs 137 PROFILE



RANGE: 195 733 Bq/Kg  
N. CAMPIONI: 16

FIG. 23.

# Cs 137 PROFILE



RANGE: 67794 Bq/Kg  
N. CAMPIONI: 12

FIG. 24.

# Cs 137 PROFILE

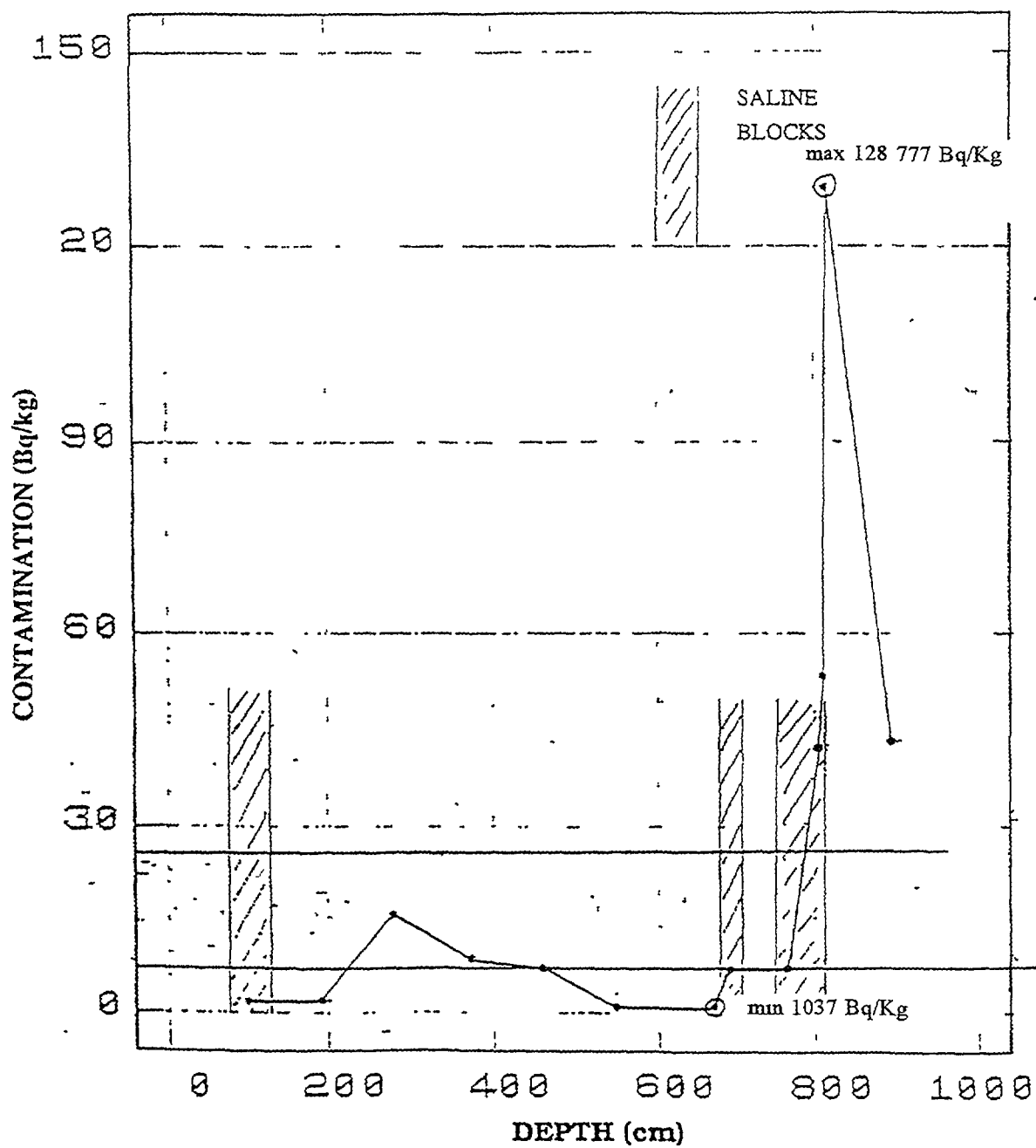


FIG. 25.

## 8. CONCLUSIONS

So far, the waste materials contaminated by Cs137 are well confined inside the mentioned waste disposal, well protected by suitable physical and geochemical barriers, that are capable of avoiding any diffusion of that radioisotope in the environment for a very long time.

The methodology used has reached the aim of both localizing and measuring Cs137 inside a great quantity of material; moreover, all the actions have been carried out in safe conditions and without any contact of contaminated samples with the environment.

The waste to be confined may now be considered as a well defined "body" of contaminated waste, located inside the basin n° 3.

The interim action was very well proof against the bad weather conditions of winter and spring 1991, very stormy and rainy seasons after a four-year period of dry conditions.

## REFERENCES

- [1] Antonioli F., W. Bocola (1983): *Esperienze sulla migrazione di Cs, Sr e I in argille prelevate in alcuni bacini italiani* - ENEA/RT/PROT(83)4.
- [2] Antonioli F. et al.(1985): *Element diffusion and oxidation phenomena along permeable fractures in clay formations at Monterotondo, Italy* - Proceedings of the International Clay Conference, Denver, 1985 - The Clay Minerals Society, Bloomington, Indiana, 415-421(1987).
- [3] Beone G., Carbone A. I., Zagaroli M.: *La bonifica di aree contaminate* - ENEA/RT/PAS/88/31.
- [4] Cochi C. et al. (1989): *Programma di intervento per la messa in sicurezza della discarica di Capriano del Colle (BS) - Messa in sicurezza meteorica - Specifica tecnica* - ENEA- AMB-RIF-CRIA
- [5] Cochi C. et al. (1990): *Programma di intervento per la messa in sicurezza della discarica di Capriano del Colle (BS) - Piano di prelevamento di campioni nel corpo della discarica - Prescrizioni tecniche* - ENEA- AMB-RIF-CRIA.
- [6] Cochi C. et al. (1990): *Programma di intervento per la messa in sicurezza della discarica di Capriano del Colle (BS) - FaseIII - Copertura finale - Prescrizioni Tecniche* - ENEA- AMB-RIF-CRIA.
- [7] Daniel D. et al. (1986): *Field permeability test for earthen liners* - Proceedings of a Specialty Conference on Use of in Situ Tests in Geotechnical Engineering - Geotechnical Special Publication - Samuel P. Clemence Editor - pp. 146-160.
- [8] Jones et al. (1986): *Development of waste containment structure for Niagara Falls storage site* - Canadian Nuclear Society: Conference Proceedings of 2nd International Conference on Radioactive Waste Management - Sept. 7,11,1986.
- [9] Paris P. (1989): *Una valutazione di limiti di concentrazione non pericolose per lo smaltimento dei rifiuti radioattivi in una discarica superficiale* - ENEA DISP/SER/NOR-RT(89)6.

- [10] Saltelli A., Antonioli F. (1985): *Radioactive waste disposal in clay formations: a systematic approach to the problem of fractures and faults permeability* - Radioactive Waste Management and Nuclear Fuel Cycle - Volume 6(2), June 1985, pp. 101-120.
- [11] Amm.ne Prov.le di Brescia (1989): *Indagine idrogeologica dell'area interessata da inquinamento di cloruri e di potassio nei comuni di Capriano del Colle e Poncarale - rapporto conclusivo.*
- [12] Amm.ne Prov.le di Brescia (1990), Settore Ecologia - *Comunicazione personale e foto di sopralluoghi.*
- [13] Ditta Metalli Capra (1990): *Comunicazione personale e foto di stati di avanzamento dei lavori.*
- [14] U.O. Fisica e Tutela Ambiente - P.M.I.P. di Milano - U.S.S.L. 75/III - Sezione di Radioprotezione - *Relazione Tecnica: Risultati delle analisi di radiocontaminazione sui campioni di residuo prelevati presso la discarica Montenetto di Capriano del Colle (BS)* - Milano, 8 Novembre 1991.

# **RADIATION SITUATION IN THE REPUBLIC OF KAZAKSTAN, ITS CONTROL AND MEASURES ON LIQUIDATION OF RADIOACTIVE POLLUTION**

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## **Abstract**

The report describes procedures of supervision and control of radiation situation in the Republic of Kazakhstan, its general conditions and some methods used on practice of recultivation of polluted territories. In the report are described the main sources of radioactive pollution and conducted measures, which permit to prevent the distribution of radioactive contamination. It is shown that the main part of contaminated territories is supervised and if it is necessary deactivated. The work on revealing of unknown pollution is regularly carried out. Also in the report are given radiation safety regulations on liquidation of uranium enterprises and some problems connected with waste handling in the Republic.

## **1. INTRODUCTION**

Natural radiation background of majority of territories of the Republic of Kazakhstan is rather low and depends upon discharge availability of rocks on surface and does not present the threat to health of the population.

Radiation pollution of Kazakhstan is spotlike and it is stipulated by:

- execution of nuclear tests on territory of the Republic;
- presence of uranium mining and processing plants;
- presence of enterprises, raw materials or waste which are characterised by increased contents of natural radionuclides.

Moreover, the Republic has significant quantity of enterprises using sealed sources (annually about 100 thousand pieces), one power nuclear reactor and four research nuclear reactors.

## **2. SUPERVISION OF RADIATION SITUATION**

### **2.1. Supervision of radiation background**

For supervision of global precipitation and background radiation significance is used the existing network of hydrometeorological stations. Daily on 115 meteorological stations located on the whole territory of the Republic are carried out tool measurements of capacity of exposition dozes [1]. There are 40 points in which conducted daily sample selection of radioactive precipitation and selection of air sample on radioactive contamination. Concentration measurement of nuclides (Caesium-137, Strontium-90, Potassium-40, Radium-226, Thorium-232), density of their precipitation and also measurement of the capacities of

exposition doses are executed on techniques of State Hydrometeorology Committee of the former USSR. The results of measurements are published in a special year-book.

In the 1993 year-book were made the following conclusions:

- parameters of radioactive pollution of atmosphere in 1993 were very low;
- average annual dose power of external gamma-irradiation of the population in Kazakstan had been 11-17  $\mu\text{R/h}$  and it was stipulated by natural background emanation
- there was not found anomalies in isotopic structure of air samples.

## **2.2. The supervision on objects**

The control services of enterprises carried out the supervision of situation on radiation dangerous objects. For these purposes used devices which is planned to include in uniform system of communication and data processing. The primary measurement of fields of gamma-radiation is considerable shortage of such systems. Radionuclides of Plutonium, Strontium, and other as a rule remain uncontrolled.

The quality of this work is supervised by special services, which will be described later on. For supervision of condition of water layers of underground horizons around of radiation dangerous objects drilled monitoring bore holes.

For reception of operative information about radiation situation in the whole Republic and increase its reliability is planned to create national system of radiation monitoring. The methodical base of measurements will be improved and to be used the modern means of processing and transmissions of data.

## **2.3. Revealing of pollution**

For revealing spots of radiation contamination carried out aero-, auto- and pedestrian gamma-spectroscopic surveys.

As a result of these surveys in seven regions, 6 industrial cities and mining places, two resorts were registered more than 100 anomalies. Basically, it is low-active solid industrial waste which contaminate small sites of surface. Mainly, they formed as a result of loses in mining and processing places of uranium ore, straits of petroleum and other leakage of materials containing radioactive substance.

According to current rules the revealed contaminated territories have to be fenced in and deactivated. Collected radioactive waste (RW) should be disposed.

The air gamma-spectrometer survey of territory of Kazakstan is carried out now. This program includes measuring flights of all territory of the Republic along isolines oriented from North to South. The distance between isolines is 10 km.

Also other measures on detection of radioactive pollution are carried out. For example, in the past 3 years have been checked 400 educational institutes and in 139 were found 732 unaccounted sealed sources, including 605 ones in Almaty [2].

It is conducted the check of quality of water and food and human environment. There are known the cases of use of building materials with high radiation for house construction, which is inadmissible.



### **3. CONTROL**

On the territory of the Republic act the rules of nuclear and radiation safety of the former USSR.

#### **3.1 Main supervising bodies**

- (1) The bodies of sanepidemiological control (BSC) - BSC part of the structure of Ministry of Health. It was created in the former USSR for protection of life and health of citizens including protection from harmful effect of radiation. It controls observance of rules and radiation safety norms and safety of ionising source (IS ).
- (2) Atomic Energy Agency (AEA). - AEA executes control of nuclear safety, accounting for and control of nuclear materials, licensing of all kinds of nuclear activity and provides safety handling with RW.
- (3) Ministry of Ecology and Bioresources was created as the body of management and control of environment, and it issues permit on disposal of RW.

#### **3.2. Controls of nuclear safety**

The observance of rules and norms of nuclear safety are obligatory for all enterprises, which possess nuclear dangerous objects. AEA controls their observance. This control consists of three levels:

- (1) The service of nuclear safety of enterprise.
- (2) Inspectors of AEA on enterprises.
- (3) Inspection of enterprises by AEA.

The main task of this control is the prevention of nuclear incidents and discharges on nuclear objects.

#### **3.3. Control of radiation safety**

Rules and norms of radiation safety are necessary for radiation safety of the population and for the staff during their work. BSC controls observance of these rules and norms. The special services of radiation safety of enterprises provide their observance. The enterprises where the radiation and nuclear dangerous objects are present or conducted radiation dangerous works are obliged to create such services.

According to the acting order:

- All enterprises, using sealed sources or producing RW are obliged to be registered in BSC and Ministry of Internal Affairs (MIA) and to observe norms and rules of radiation safety. The enterprises are completely responsible for safety of all sealed sources and account them.
- Allowance on transmission and transportation of all types of IS is given by BSC and MIA. The way and route of transportation of IS should be agreed with BSC and MIA.

- Enterprises account RW and dispose them. All disposals or storages of radioactive wastes should be registered and controlled.
- Designing of radioactive objects and giving special places for radioactive objects should be agreed with supervising bodies.
- Liquidation of radiation dangerous object requires the execution of whole complex of recreation work in accordance with specially developed project, which should be agreed with supervising bodies. After completion of project work, the special commission determining restriction on use of land, order of supervision and control them.

### **3.4. Legal basis**

In the republic which has the advanced monitoring system for observance of radiation and nuclear safety, there is no any legislative basis determining order of use of nuclear energy, handling with RW, radiation protection of environment and health of the population. It is not developed general order of management and supervision in these fields.

Before the acceptance of appropriate laws in the Republic act " Temporary regulation about use of nuclear energy, nuclear activity, handling with radioactive waste and spent nuclear materials, providing of radiation safety of the population of the Republic of Kazakhstan", which is approved on April 1994 by special decree of Cabinet of Ministers of the Republic of Kazakhstan. In this document were taking into consideration the notes of SKI (Sweden) and Ministry of Atomic Energy of Russia.

Now the projects of laws "About maintenance of radiation safety of the population" and "About handling with radioactive waste" are practically ready. These projects are developed in view of world experience. Project of law "About handling with radioactive waste" took into account the remarks of Nuclear Energy Agency at Organization of Economic Co-operation and Development (OECD/NEA).

It is planned the creation of law about "Peaceful use of nuclear energy". All prepared projects of laws harmonize with projects of appropriate documents of CIS countries.

It was created the new Ministry of Ecology and Biorecources (MEB) for protection of environment. Now MEB is on the stage of creating the regulation system of nature resources use and regulation of discharge restriction. Side by side with it MEB are solving tasks of tracking for its conditions and prevention of irreversible industrial effects. The necessity of ecological examination of all economic objects is entered recently.

## **4. CHARACTERISTIC OF POLLUTION SOURCES**

### **4.1. Nuclear explosions**

According to the present data on the whole territory of Kazakhstan were made about 500 nuclear explosions. The main part of explosions - 472 (124 from them were surface and air explosions) were conducted on the former Semipalatinsk testing area (STA) [3,4]. The large part of other explosions were produced in region of settlement Azgir which is located on the west of Kazakhstan.

The volumes of produced RW in a result of nuclear explosions evaluated in 12 million of tons with general activity of 12.9 million Curie, out of which 6.5 million of tons attribute to underground explosions. It seems that waste of underground explosions is isolated in melted mass of rocks. Thus, wastes in cavities of explosions classified as medium active and waste on earth surface as low-active.

For organizing of nuclear tests were used the uninhabited spaces of Kazakstan, that has allowed to lower the risks of irradiation of the population. Besides, administration of polygons are obliged to observe all norms and rules of radiation and nuclear safety and now under control of MBR and AEA and before it were supervised by appropriate services of former USSR.

At the same time, during the execution of nuclear tests there was not always present the opportunity of total check of conditions. First of all it concerned the execution of the first nuclear explosions, when it was not accumulated of sufficient experience for acceptance of necessary measures of radiation protection of people and protection of environment.

It is trying to do approaches of reconstruction of radiation situation during execution of tests for valuations possible irradiation of inhabitants of close settlements. It was begun the work on maintenance of social protection of suffered citizens of the Republic.

The planning of restoration of environment in regions where the nuclear explosions were conducted, mostly complicated by the fact that the many archival materials on nuclear explosions (including data about radiation situation and polluted sites of territory in locations of execution of explosions) are in Russia.

#### 4.1.1. Radiation situation in region near STA.

In accordance with the present data radioactive clouds of some air and ground explosions left the limits of STA. Just they have caused radiation pollution of the part of the close territories.

For determination of scales of radioactive contamination in the region it is carried out the surveys of all territories close to the polygon. The researches with selections of soil tests, vegetation and products of meal are conducted.

At the request of government of the Republic of Kazakstan two IAEA missions with participation of experts from different countries conducted the expert valuation of radiation situation in the region. In some populated points of the territory they have selected the tests of soils, water, food, bones, vegetation and conducted radiation measurement.

According to the preliminary data of these missions the general radiation situation on the greater part of surface at the present time is normal and suitable for residing of people. These data are in good conformity with preliminary results of already started researches of radiation situation of the region, which by the order of the Ministry of ecology and bioresources was carried out the by specialised enterprises of the Republic. Underground conditions were not examined.

#### 4.1.2. Radiation situation in region of Azgir settlement

In salt deposits in region of Azgir settlement were conducted seventeen underground nuclear explosions. Energy liberation of nuclear explosions changed from 0.5 to 100 kt trotyl (TNT) equivalent [5]. The cavities formed as a result of

these explosions were opened by boreholes. In series of cases for executions of explosions were used the cavities of the previous explosions. In the whole there were formed nine underground cavities with total volumes of 1.2 billion litres on 160 to 1500 meters depth. Some of cavities are at present filled with radioactive solutions. The detailed research of conditions of cavities is required for determination of reliability of isolation of nuclear explosion waste and probability of pollution of underground waters by radionuclides.

The known pollution of sites of surface of this object have the local nature and basically were formed during the opening of underground cavities by boreholes. Complex survey of radiation situation in the region was not conducted yet by supervising bodies of the Republic of Kazakhstan. The main information on radiation situation in the region of Azgir settlement is given by Radium institute of name Chlopin V.G. (Russia, St.-Petersburg)

At the present it is conducting the work on recultivation of contaminated technological platforms of boreholes. Collected polluted ground and equipment are excluded in one of dry underground cavities.

#### **4.2. Pollution from uranium mining and processing enterprises**

In Kazakhstan is prospected and long time (more than 40 years) were developed more than 20 uranium deposits, some of which are large and unique on stocks. RW formed when prospecting, mining and processing processes the most significant on volume ( 93 % from the total volumes of RW of the Republic ) are widespread on the territory and make approximately 218 million of tons at general activity of 220 thousand Curie. They are low-active and only less than 1 % from total volume are with medium activity. In the whole 127 locations of storage and disposal of radioactive waste of this branch are known.

RW of uranium enterprises so named tails of production are stored on special storehouses of low-active wastes (tail storage) of enterprises which can be disposed after filling it.

Tail storages are designed in general project of the whole technological complex of uranium mining and processing enterprises and are intended for storage of radioactive wastes from technological processes of designed enterprise. The project of such storehouse usually does not provide the building of special engineering barriers and isolation of stored radionuclides is provided by choosing of location (location with low level waters) availability of waterproofing protection. The surface of disposal bottom covered by layer of waterproof ground of capacity not less than 0.5 m or by special waterproofing cover.

These tail storages can cover the large areas (to 1000 hectare) and make dust. Radioactive dust from tail storage spread over the close territories. The existing methods of struggle with formations of dust on such storehouses not are enough effective.

#### **4.3. Pollution from mining enterprises, raw materials or wastes of which are characterized by increased contents of natural radionuclides**

RW from mining enterprises, raw materials and wastes of which are characterized by increased contents of radionuclides mainly, enterprise, which working with deposit of coal, petroleum and gas are evaluated in 1,57 million. tons with activity 519 Curie.

The pollution of petroleum and gas deposits are basically concentrated in locations of mining and along ways of transportation and are displayed in kind of straits of products and accompanying waters, pipes and other bore equipment. So, in some regions of intensive mining and processing of petroleum revealed the plenty of low-active waste in kinds of oil-waste and soils polluted by radionuclides of volume about 10 million litres and pipes of weight of 540 tons on enterprises of oil extracting.

The low-active waste of this group as a rule are stored on special - allocated platforms under supervision of enterprises.

## **5. LIQUIDATION OF URANIUM ENTERPRISES**

The process of liquidation of uranium mining and processing enterprises requires much time and means. It consists of the following stages:

- (1) The acceptance of decision about enterprise liquidation.
- (2) The development of project of liquidation of enterprise.
- (3) The execution of complex of work on projects.
- (4) The reception of land by special commission.
- (5) The supervision for land.

The acceptance of decision about liquidation of enterprise and the project of liquidation require the co-ordination of supervising bodies, and in accordance with new order it is compulsory passing of ecological examination.

The project of liquidation of uranium enterprises is obliged to contain the complex of measures on maintenance of radiation safety on recultivated lands.

After completion of work on projects the special commission, determine restriction on use of land, the order of supervision and control of them, responsible person for observance of order on these land. The necessary documents made out, including close plans with drawing of disposal of RW, tails, other sites of limited use, also a plan with radiation characteristics of the land and other necessary information.

On practice the process of liquidation of uranium enterprise can begin earlier (works on disposal of filled tail storages and recultivation of waste), before closing-up of enterprise because of lack of means. There are some cases when no any opportunity to begin the restoration work on prepared projects.

The order of accumulation of necessary means enterprises is now not yet fulfilled. The considerable complexities of this questions are connected with transition from monopoly totalitarian state in economy to more free forms of managing and with processes of hyper inflation which follow this transition.

Further let's stop on some means on maintenance of radiation safety, execution of which require the special instructions on liquidation of uranium enterprises [7]. It will give the ideas about used methods for deactivation of territories, disposal of low-active wastes, recultivation of tails etc.

### **5.1. Measures of radiation safety of uranium enterprises liquidation**

There are present the special norms and rules of radiation safety of liquidation of uranium mining and processing enterprises. They require the execution of following measures on maintenance of radiation safety:

- disposal or deactivation of contaminated buildings and equipment;
- deactivation of polluted territories;
- disposal of tail storages;
- recultivation of rock tails.

If necessary the measure on reduction of allocation of Radon should be accepted.

Polluted soils are removed and disposed on waste storage for deactivation of territories.

The equipment and premises removed from radioactive dirt. Water used for it is cleared, solid RW collected and disposed on tail storages.

Repeatedly used premises and equipment should satisfy to requirements of radiation safety rules.

All RW and non-deactivated equipment disposed on tale storages.

Disposal of tail storage requires reliable isolation from surface waters and drainage system which should completely exclude storm and other surface flow on surface of disposed tail storage. The lay-out of disposal should exclude the opportunity of formation of pools and holes on the surface of tail storages.

The surface of tail storage covered by road metal with bitumen or other waterproofing. The thickness of cover layer not less than 0.5 m. After this surface covered by pure ground, thickness of which should be sufficient for development of root system of vegetation(not less than 0.5 m for grass, to 1 m - for bushes, about 2 m - for trees) and it is defined by particular geographical conditions of region, where tail storage is located.

The territory of disposed tail storage fencing in or designated in dependence on distance to populated points and it is under supervision. It is prohibited to use for any economic purposes.

At recultivation of tails they should be made even and planned, then covered by binding material (clay etc.), road metal layer and layer of ground then seed by grass or bushes. In zone of 300 m from them prohibits the housing construction. Distanced waste from populated places (not closer 5 km) can be not made even.

All un-recultivated waste are obligatory protected. The use of materials of waste for economic needs is prohibited.

The territories of recultivated waste fencing in or designated in dependence on distance from populated points and are under supervision.

## **6. HANDLING WITH RW**

It is specially necessary to note that in the Republic doesn't exist the uniform system of transportation and disposal of RW. The Republic do not have the regional points of disposal, there are luck of special transports and other special equipment. Before it, collecting and disposal of RW of the Republic of Kazakstan were engaged the special organizations of Russia. Now, by decree of president of Russian federation is prohibited disposal of radioactive waste on the territory of Russia.

On this reason the execution of work on utilising of RW is complicated. The order of disposal or storage of waste is defined in each particular case. The part of waste accept departmental and object disposal. The other waste are stored in locations of their formations where difficult to create necessary conditions of their safe storage.

The creation of four regional disposals of wastes of low and medium activity is planned. It is prepared the project of "Concept of disposal of radioactive waste of the Republic of Kazakhstan ". This document is developed with use of international experience and takes into account remarks of AEA Technology (Great Britain). It states the main principles of safe disposal of RW in the Republic. Document provides expert valuation of all existing locations of disposal (storage) of RW and, if necessary acceptance of measures on maintenance of all requirements which are produced in the Concept.

Annually it is required to dispose till 10000 sealed sources. For maintenance of their safe long-term storage there are carried out works on preparations of departmental storehouses in Kurchatov. It will accept sealed sources from all the Republic till the enter in operation regional disposal.

## **7. CONCLUSION**

The report can not pretend on completeness of descriptions. For representation of more detailed descriptions are required specialised reports on designated directions which possibly will be interesting issue, taking into account the experience accumulated in the Republic on investigation and operation of uranium deposits and availability of points of execution of nuclear explosions

At the same time it is required the total research of locations of development of uranium deposits and execution of nuclear tests for valuations of sufficiency of conducted measures of radiation safety. In view of new approaches about radiation protection of environment and the population which will be worked out during creation of legal basis, it is necessary to do also expert valuation of the existing pollution and ways of their liquidation.

Besides that there are series of considerable problems which were not discussed in this report. For example, the problem of population irradiation were not discussed which could take place during the tests of nuclear explosions.

Moreover, it was not discussed the problem of possible radiation pollution from Chinese nuclear polygon (Lobnor) which very disturb the public of all close to this polygon countries.

The problems of protection of environment not connected with radioactive pollution also were not discussed in this report, though the availability of the advanced complex on mining and processing of raw material induces the serious ecological problems.

## **REFERENCES**

- [1] RADIATION SITUATION ON THE TERRITORY OF THE REPUBLIC OF KAZAKSTAN ON YEAR 1993, Annual Report of the General Office on Hydrometeorology at the Cabinet of Ministers of the Republic of Kazakstan, Almaty (1994).
- [2] SHKOLNIK, V. S., BAYADILOV, E. M., et al., "Radioactive waste in Republic of Kazakstan and ways of it utilization", Remediation and Restoration of Radioactive-Contaminated Sites in Europe, (Proc. Int. Symp. Antwerp, 1993), Vol. 2, European Commission, Doc. XI-5027/94, 713-726.

- [3] GORIN, V. V., KRASILOV, G. A., et al., "Semipalatinsk test site: Chronology of underground nuclear explosions and their primary radiation effects (1961-1989)", Bulletin of the Public Information Centre on Atomic Energy, No. 9, Moscow (1993) 21-32.
- [4] DUBASOV, Yu. V., DUMIC, V. P., et al., "The chronology of nuclear tests carried out in atmosphere, space and under water (1949-1962)", Bulletin of the Public Information Centre on Atomic Energy, No. 2, Moscow (1994) 36-43.
- [5] DUBASOV, Yu. V., KEDROVSKIY, O. L., et al., "Underground explosions of nuclear devices for industrial purposes at the territory of the USSR in 1965-1988: Chronology and radiation effects", of the Public Information Centre on Atomic Energy, No. 1, Moscow (1994) 18-29.
- [6] KRIVOKHATSKIY, A. S., DUBASOV, Yu. V., et al., "Radiation effects of underground nuclear explosions for peaceful purposes at salt deposits Bolshoi Azgir", of the Public Information Centre on Atomic Energy, No. 9, Moscow (1993) 49-59.
- [7] SANITARY RULES ON LIQUIDATION, PRESERVATION AND PROFILE MODIFICATION OF THE MINING AND MILLING FACILITIES FOR RADIOACTIVE ORES (SP LKP-91), Ministry of Health of USSR, Moscow (1991).



# EFFECTS OF URANIUM MINING ON THE CONTAMINATION OF SOME AREAS IN POLAND

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Poland

## Abstract

In Poland there are two areas which are contaminated by natural radioactive materials. The first area is in Lower Silesia and the second in Upper Silesia. In Lower Silesia extensive uranium exploration and mining activities were carried out in the early 1950's under the direction of Soviet Union experts. Uranium exploration and mining took place at Kowary Podgórze (eastern Sudety Mts), Radoniów and Kopaniec villages and in the vicinity of Kletno. Extensive uranium-bearing mine dumps resulted from the exploration and mining. The contamination has become dispersed and the local people have not been informed regarding the health risk that this represents. Field  $\gamma$ -radiation measurements made first in 1971, showed that the highest levels of radioactivity were at the dump in Radoniów (up to 2000  $\mu\text{R/h}$ ) and in Kowary Podgórze (up to 1500  $\mu\text{R/h}$ ). Also, the dump at Radoniów has partly been reexploited for local use in construction. In the Upper Silesian region radium ( $^{226}\text{Ra}$ ,  $^{228}\text{Ra}$ ) contamination is present as suspended particulate matter and in solution in mine ground waters released during black coal mining. Near mine water discharge sites, as well as in active sediments of small rivers, up to 460 Bq/kg  $^{228}\text{Ra}$  and up to 1430 Bq/kg of  $^{226}\text{Ra}$  are present.

## 1. INTRODUCTION

In Poland two areas are known to be contaminated by natural radioactive materials: Lower Silesia and Upper Silesia (Fig. 1). In Lower Silesia extensive uranium exploration and mining activities were carried out in the early 1950's under the direction of Soviet experts. Uranium exploration and mining took place at Kowary-Podgórze (the eastern Sudety Mts), Radoniów and Kopaniec (the Iżera Mts) and in the vicinity of Kletno (the Śnieżnik Mt area). Uranium ores mined there were polymetallic and contained: pitchblende, uraninite, autunite, metaautunite, uranocircite, torbernite, metatorbernite, uranophane, sklodowskite, gummite, fourmarierite and libegite. Both the exploration and mining operations produced large uranium-bearing dumps. The largest dump is located in Kowary Podgórze and is 250x100x40m in size. The contamination has become dispersed and the local population has not been informed on the resulting health risk. Field  $\gamma$ -radiation measurements made first in 1971, showed that the highest radiation dose was acquired at the dump in Radoniów (up to 2000  $\mu\text{R/h}$ ) and in Kowary-Podgórze (up to 1500  $\mu\text{R/h}$ ). The radiation dose of fine-grained, red-colored sediments just in the foothill of the dump at Kowary Podgórze reached 2000  $\mu\text{R/h}$ . In these sediments 0.11 wt% U was indicated. During last decades these sediments have been eroded but measurements made in 1992 showed that a new anomaly of up to 2000  $\mu\text{R/h}$  has appeared closer to the closed mine adit. This anomaly is probably caused by radon and its daughter isotopes carried out by mine water. Recent  $\gamma$ -measurements also confirmed the presence of high radiation levels in the mine dumps at Kowary Podgórze, Kletno and Radoniów. However, the dump at Radoniów has partly been reexploited for local use in construction.



Fig.1 Localization of radioactive sites in Poland.

US- Upper Silesia, HCM- Holy-Cross Mts., FSB- Fore-Sudetic Block, SM- Sudety Mts., CM- Carpathians, 1- Kowary, 2- Radoniów, 3- Kopaniec, 4- Kletno, 5- Rudki.

In the Upper Silesian region radium ( $^{226}\text{Ra}$ ,  $^{228}\text{Ra}$ ) contamination results from its presence as suspended particulate matter and in solution in mine waters dumped during black-coal mining. Near mine water discharge sites, as well as in the recent sediments of small rivers, up to 460 Bq/kg  $^{228}\text{Ra}$  and up to 1430 Bq/kg of  $^{226}\text{Ra}$  were measured. Locally, in the sediments of both the Lesznica and Sztokówka Rivers radium pollution has also been observed.

## 2. EXPERIMENTAL AND FIELD TECHNIQUES

The field investigations have been done in the years 1971 and 1991-1993. Investigations aimed to determine the degree of uranium pollution. In 1971 the  $\gamma$ -measurements have been made using UR-4M portable radiometer of Soviet Union production. In both 1991 and 1992 RKP-1 portable radiometer (Polish) and in 1993 pocket radiometer RATON-901 (Russian) have been used. Radioactivity was measured directly on the surface of mine dumps and in their vicinity using 10x10m grid. The counting time at each point was calculated to be one minute. The background values have been measured 1m above the ground level, and was established to be 10  $\mu\text{R/h}$  in 1971 and 25  $\mu\text{R/h}$  in 1991. The increased background detected in 1991 has probably been caused by Tchernobyl catastrophe. The results are drawn on topographic background maps as isolines of  $\gamma$ -radiation dose rates (Fig. 2, 3, 4, 5, 6, 7, 9 and 10)

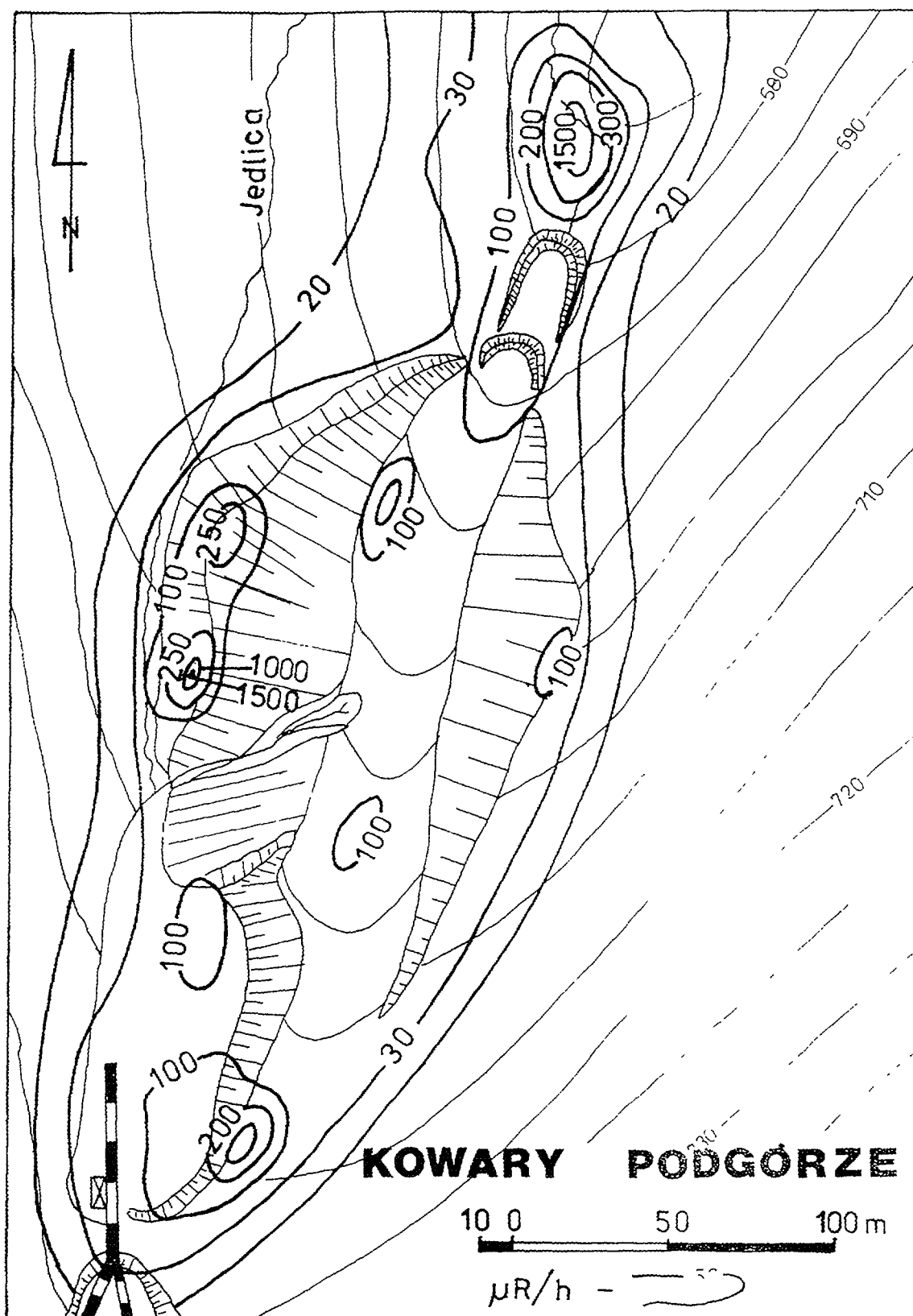


Fig.2  $\gamma$ -dose rates at Kowary-Podgórze, dump of the adits No. 19 and 19a. Measurements carried out in 1971.

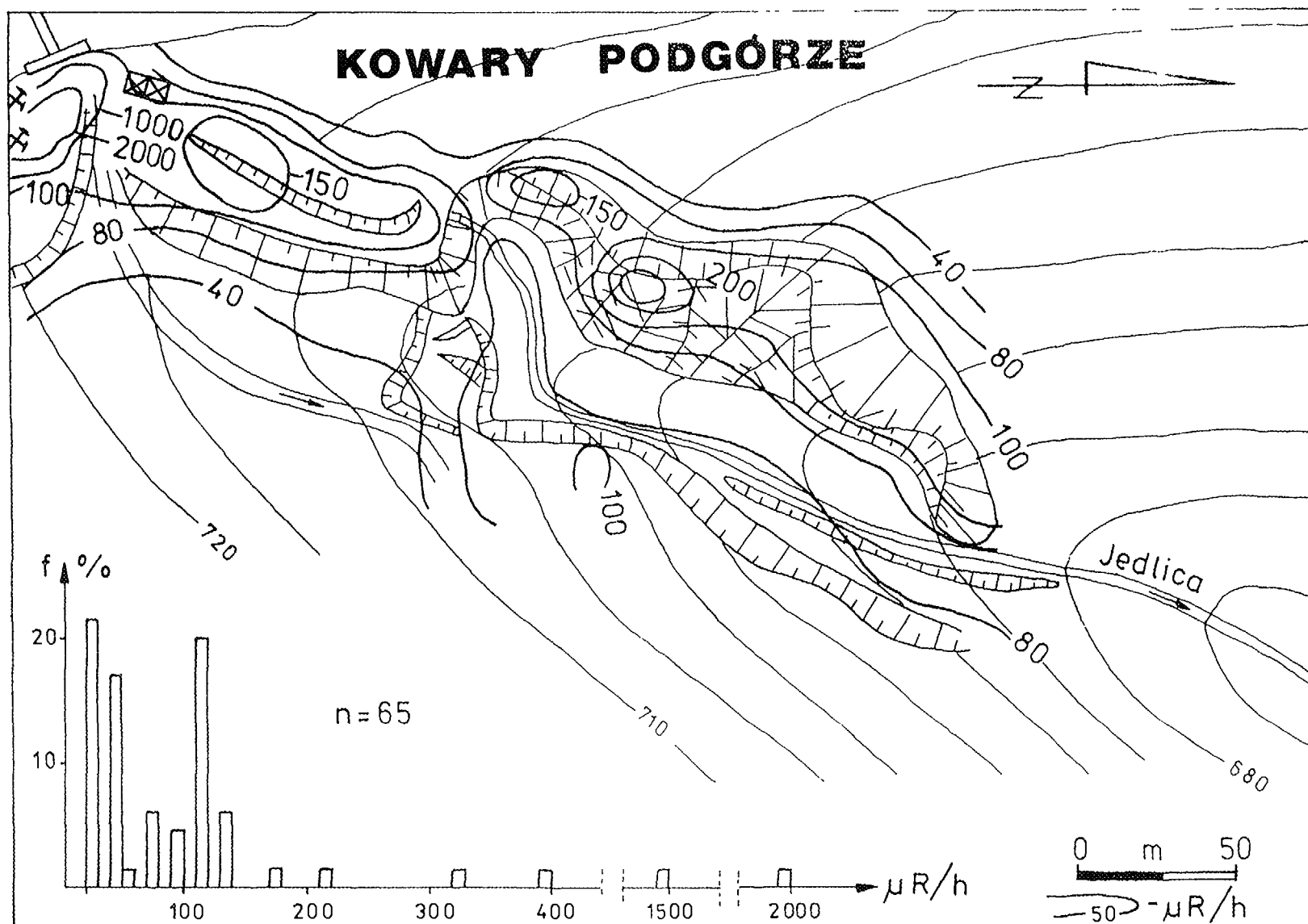


Fig.3  $\gamma$ -dose rates at Kowary-Podgórze, dump of the adits No. 19 and 19a. Measurements carried out in 1991.

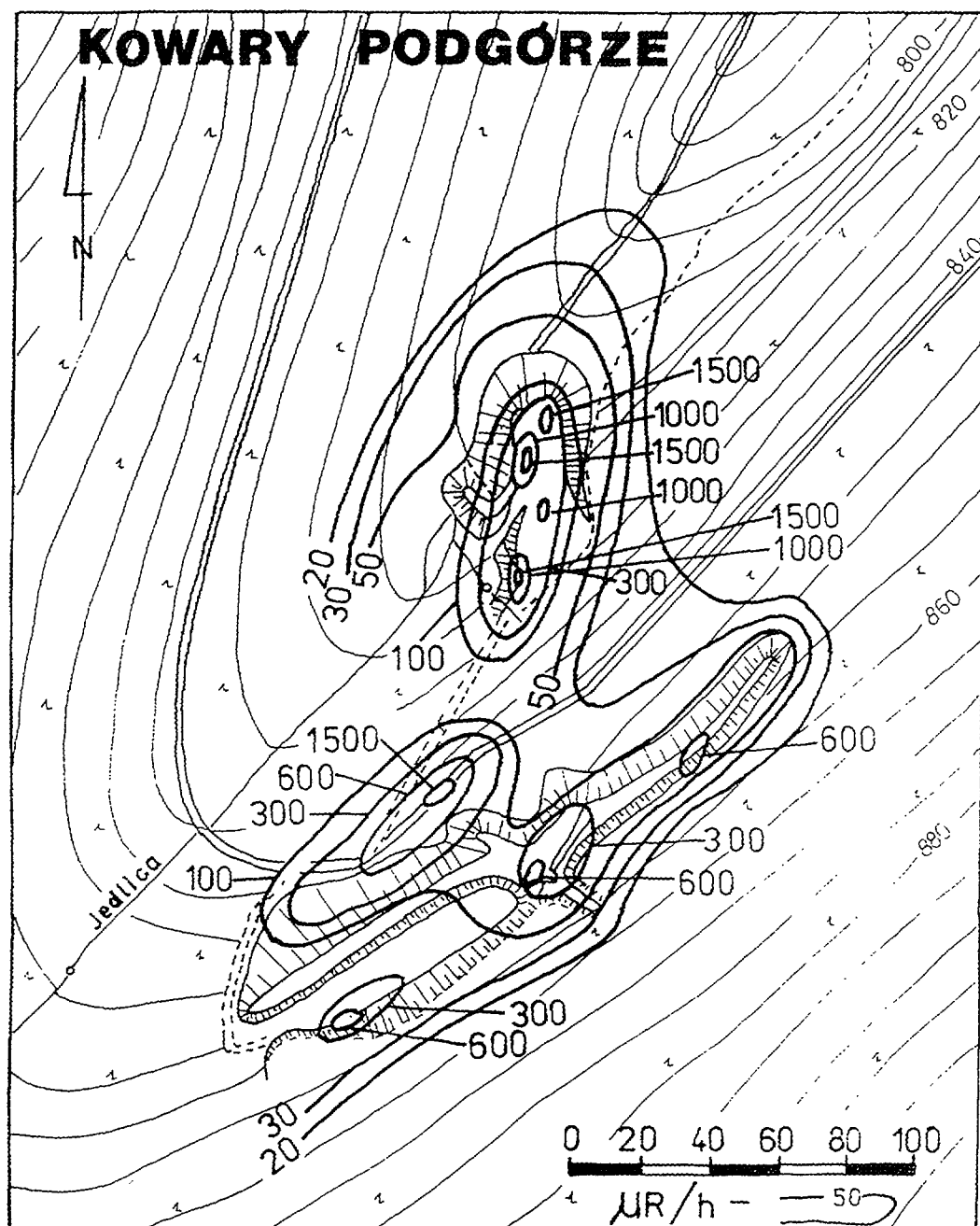


Fig.4  $\gamma$ -dose rates at Kowary-Podgórze, dumps of adits No. 16 and 17.

Quantitative U, Th and K analyses have been made using AZAR gamma spectrometer (modified) equipped with scintillator sonde with NGK-315 TESLA detector. The measurements applied the following energy channels: 1.25 - 1.65 MeV for K, 1.65 - 2.30 MeV for U and 2.3 - 2.85 MeV for Th. Standards containing 13.76 % K, 69.56 ppm U and 69.56 ppm Th have been used for calibration. The mass of the standards was 46 g each. All analyzed samples have been crushed, put into small (50 g) closed containers and left for three weeks to achieve nuclear balance before measurements. Optimum counting time was established experimentally to be 10 hours for both samples and standards. Analyses have been carried out at the Institute of Geophysics (University of Mining and Metallurgy) in 1992 using special procedure [1]. In 1971 uranium analyses have been carried out at the Institute of Physics and Nuclear Techniques (University of Mining and Metallurgy)

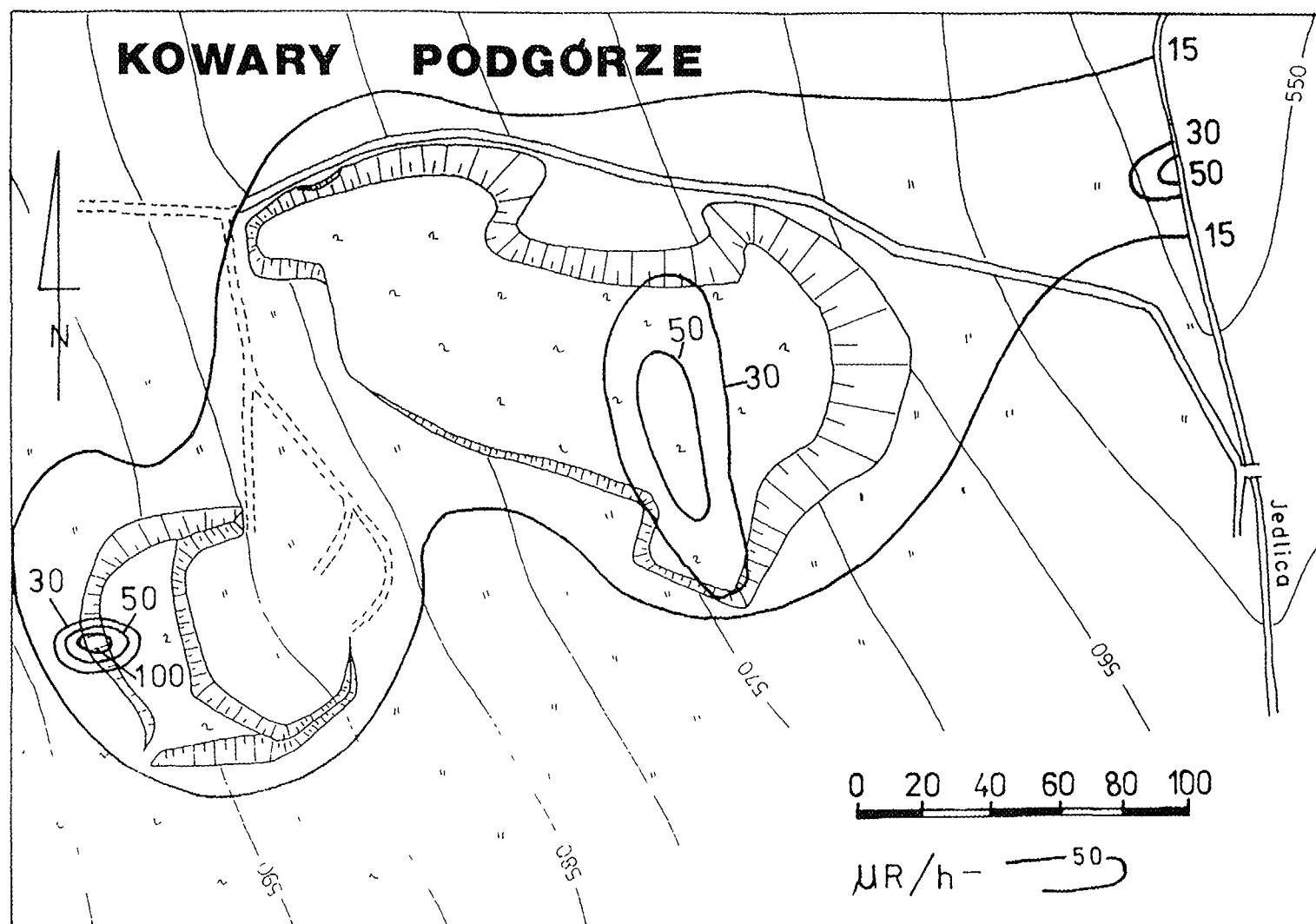


Fig.5  $\gamma$ -dose rates at Kowary-Podgórze, dump of the „Wolność” mine. Measurements carried out in 1971.

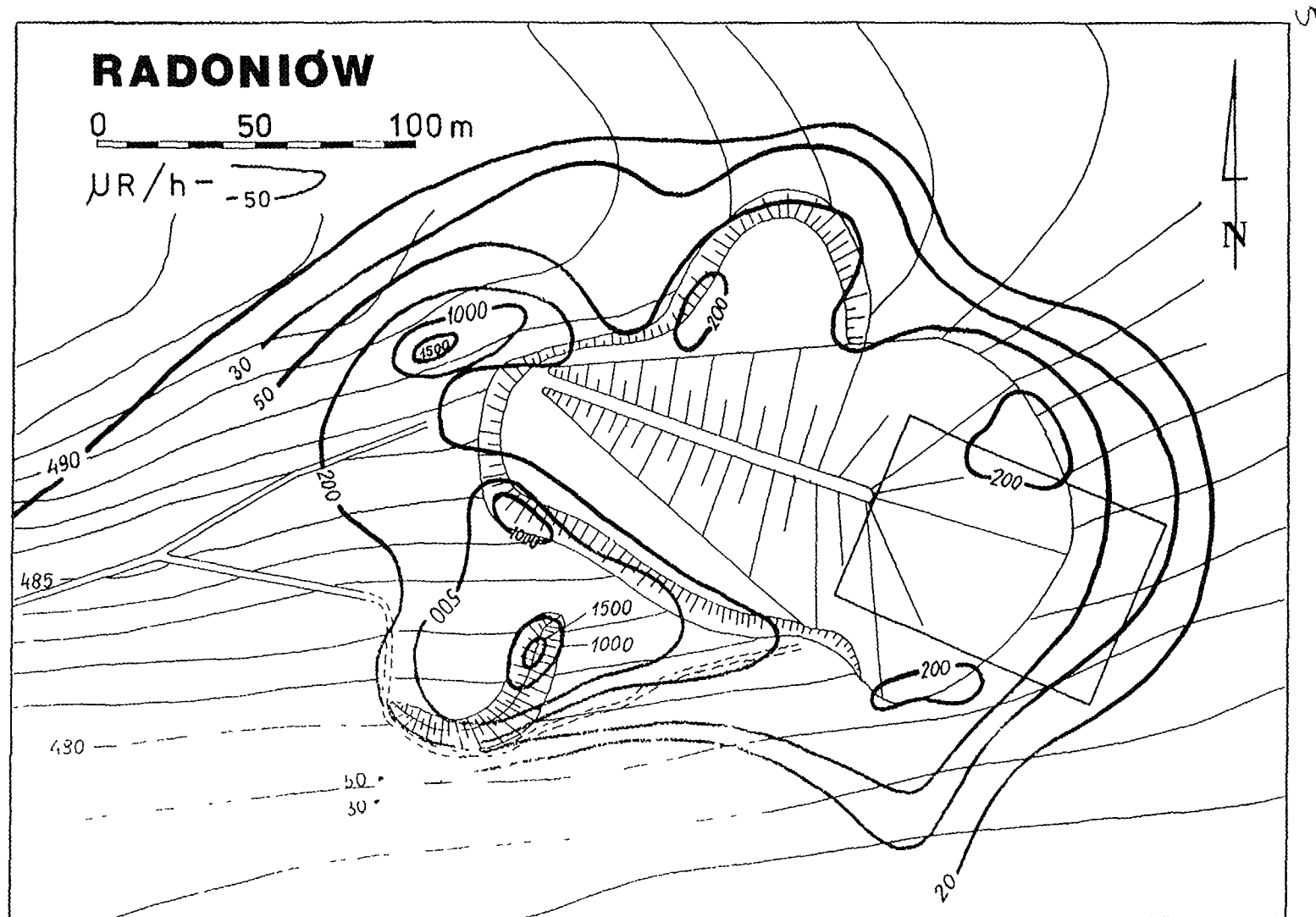


Fig.6  $\gamma$ -dose rates at Radoniów. Measurements carried out in 1971.

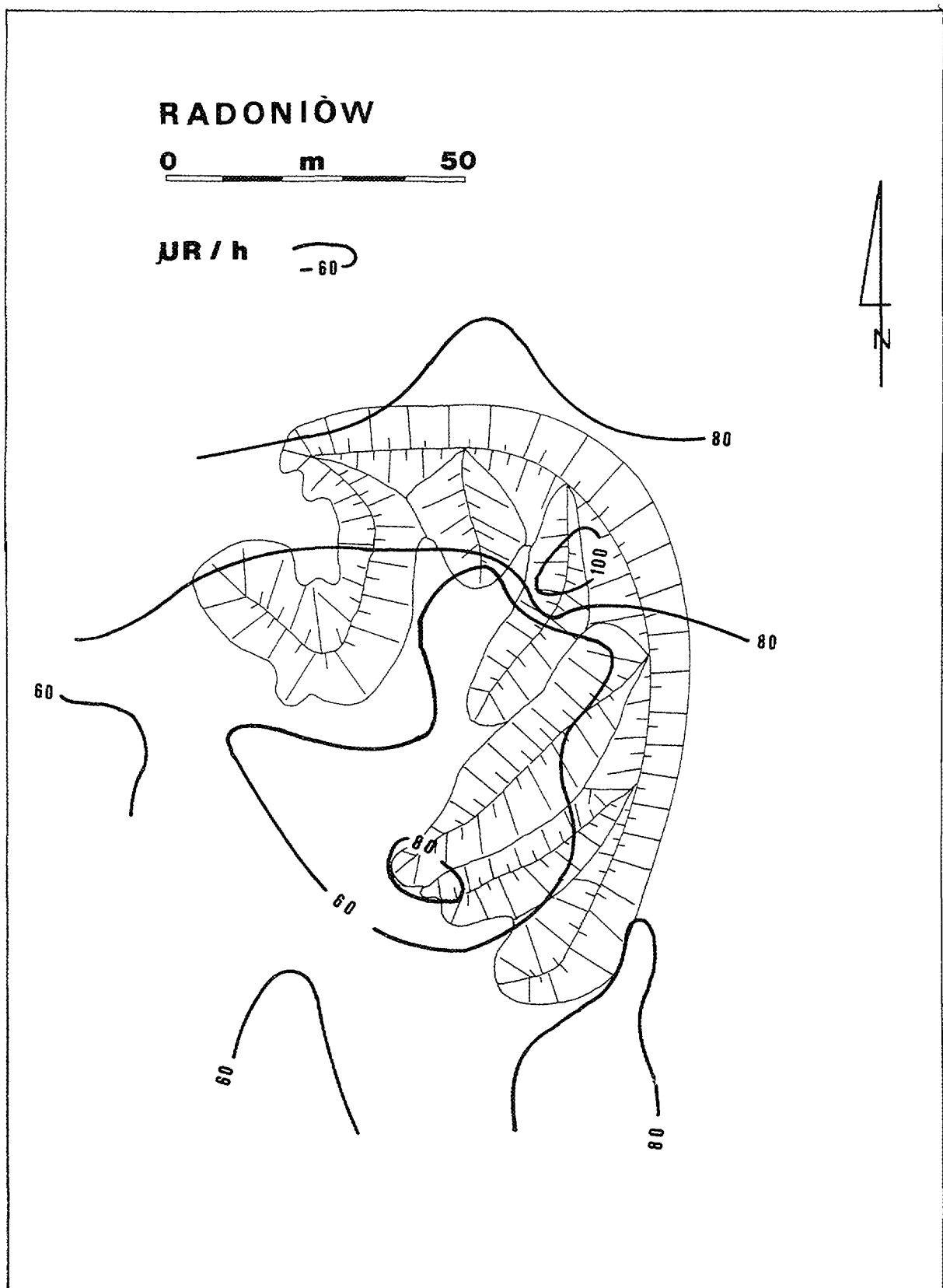


Fig.7  $\gamma$ -dose rates at Radoniów. Measurements carried out in 1994.



using the TESLA Ge-Li semiconductor detector and 400-channels Victoreen analyzer. Uranium ore containing 2.03 % U has been used as a standard. Counting times: one hour for sample and two minutes for standard were applied, the latter due to its high uranium content. Similar investigations have been carried out in 1993 by Prof. E. Chruściel and Dr. P. Jodłowski using a new generation equipment.

Uranium in water has been analyzed in 0.5 l water samples using colourimetric technique with „arsenaso I” indicator.

Concentration of radon in water samples from Kletno has been determined using alpha radioactivity of daughter products ( $^{214}\text{Po}$  and  $^{218}\text{Po}$ ). The detection limit has been calculated as 0.2 Bq/l for one hour counting time. Analyses have been carried out by Dr. H. Mikołajczyk at the Faculty of Physics and Nuclear Techniques of the University of Mining and Metallurgy.

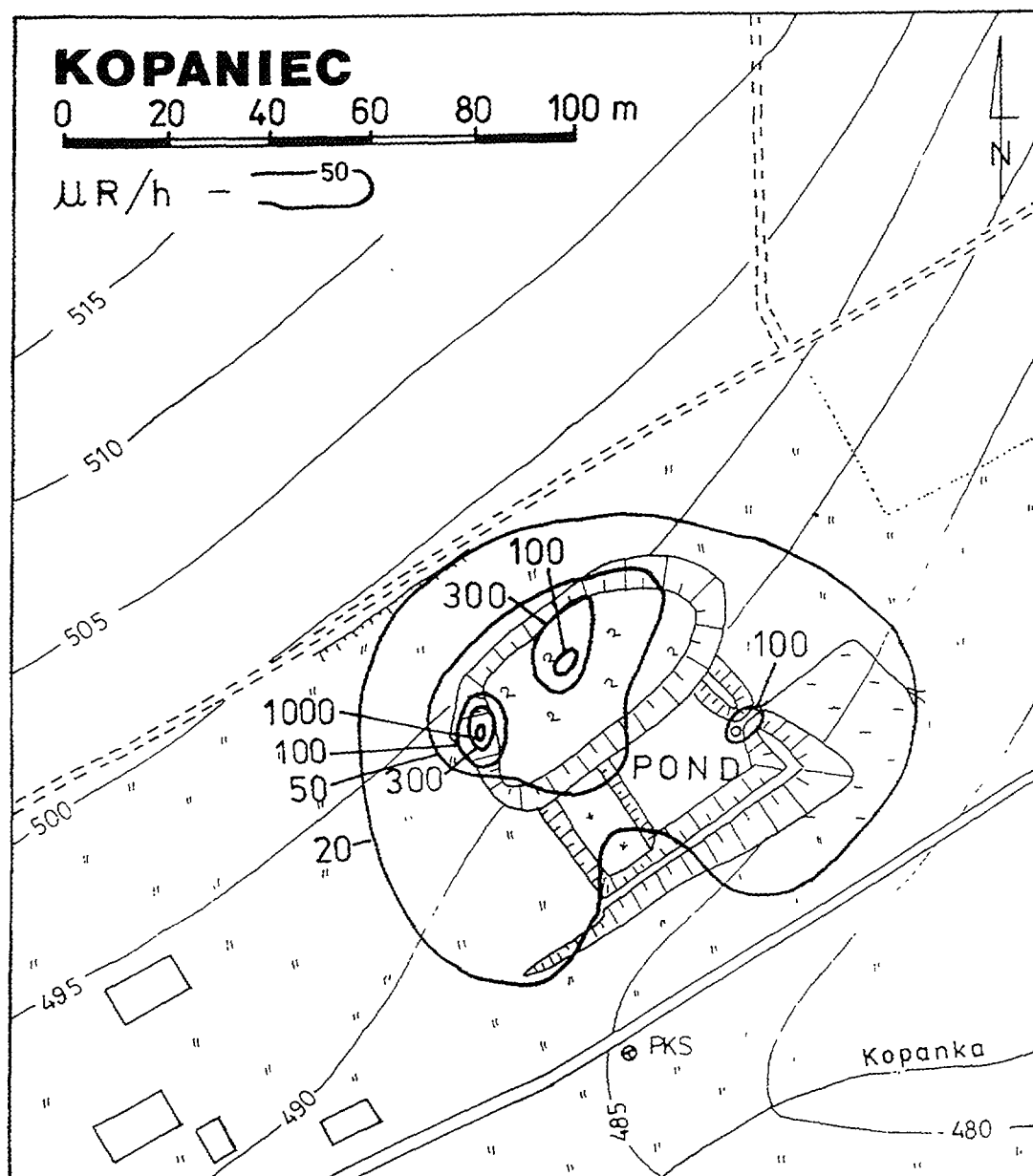


Fig.8  $\gamma$ -dose rates at Kopaniec. Measurements carried out in 1971.

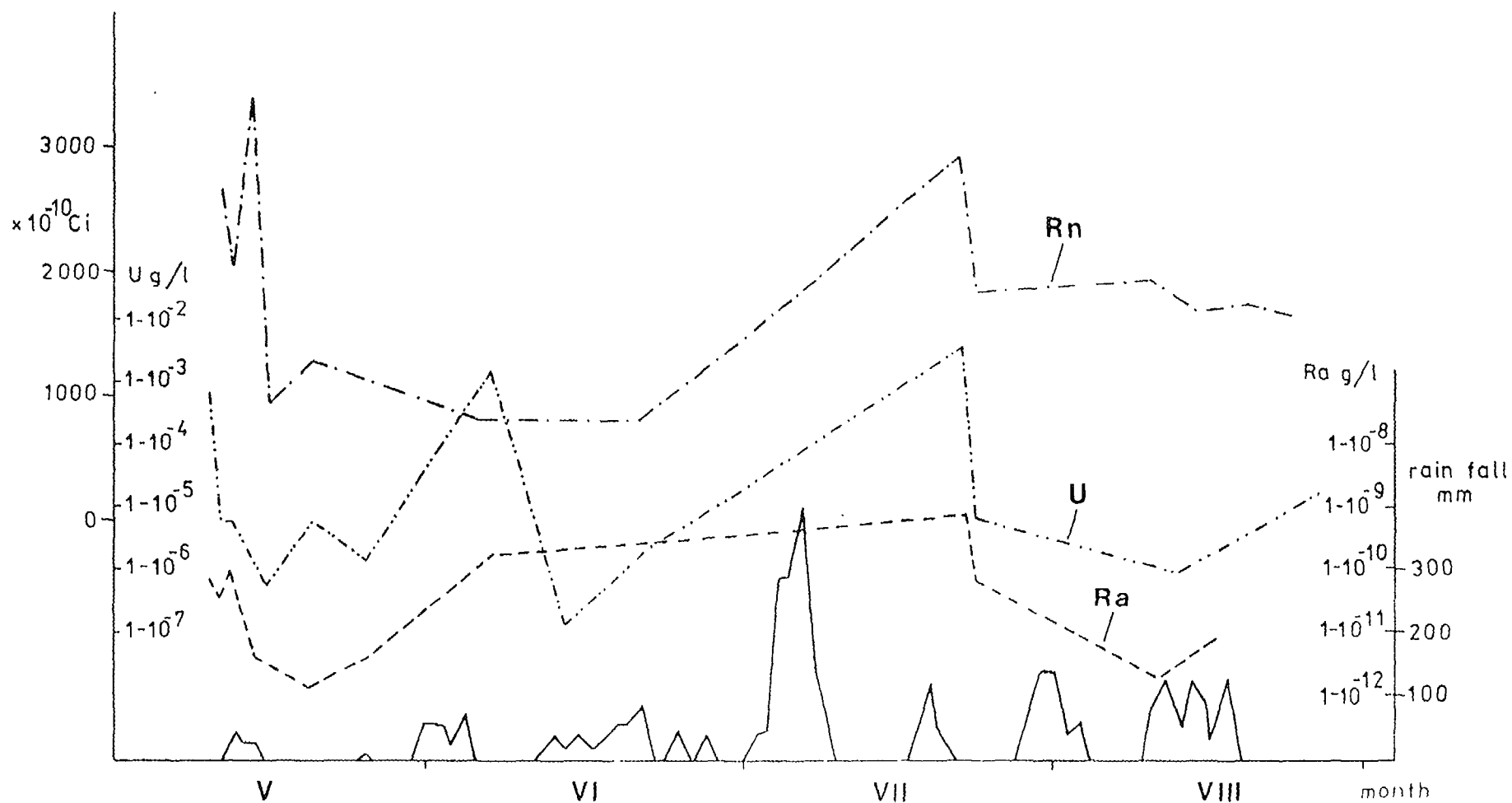


Fig.9 Concentration of radionuclides in ground water at Kopaniec (after ZPR-1 Company). Observations carried out in 1962.

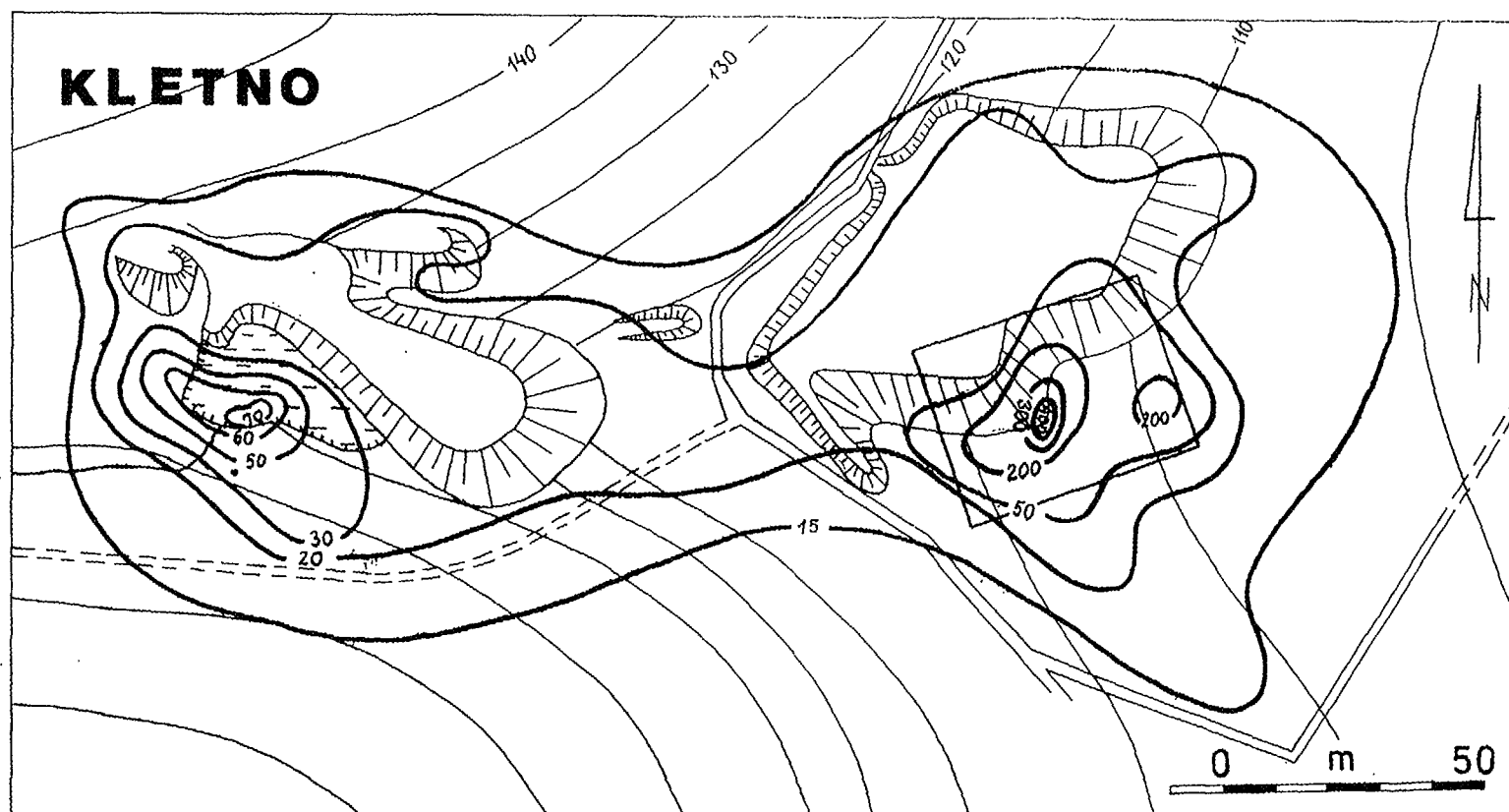


Fig.10  $\gamma$ -dose rates at Kletno, dumps of adits No.12 and 9. Measurements carried out in 1971.

### 3. URANIUM DEPOSITS

In Poland, about 100 localities of uranium mineralization have already been found. Only three of them contained sufficiently high reserves to be exploited. The uranium mining ceased in 1960's.

#### 3.1. Kowary, Kowary-Podgórze

The largest uranium mining fields - Kowary and Kowary-Podgórze are located in southern part of the metamorphic cover of the Karkonosze Granite. Mining activity in this area has started in 1148. Initially, magnetite ore has been mined. Uranium as well as polymetallic mineralization have been found and explored for the first time in 1910. Uranium ores had been exploited between 1950's and 1960's. So called „Ore Formation” is composed of marbles, erlans, various types of mica schists, pyroxene, garnet and vesuvianite skarns and magnetite [2]. Ore formation occurs as huge lens, up to 1.2km long and 0.2km wide, enclosed within the Kowary Gneiss Group. Two types of uranium occurrences are known: breccia pipes and veins/veinlets. Mineralogical composition includes mainly pitchblende accompanied by coffinite, liebigite, gummite, sklodowskite, uranophane, rutherfordite, schroëckingerite, autunite and sooty pitchblende [3]. Uranium mineralization is associated with polymetallic ores composed of arsenopyrite, löllingite, tiemannite, clausthalite, sphalerite, chalcopryrite, pyrite, cinnabar, bornite, covellite, native silver, emplectite, native bismuth, tetrahedrite, smaltite, rammelsbergite, niccolite, galena, hematite, stromeyerite, malachite and calcite [2, 3, 4]. No information is available in the published materials about uranium contents, reserves and total tonnage of the extracted uranium ores. In the mine dumps at Kowary Podgórze ore fragments containing up to 0.15% of uranium can still be found. Polymetallic mineralization is genetically related to the Karkonosze Granite [3].

#### 3.2. Radoniów deposit

The Radoniów deposit is located in the northern part of the Izera Metamorphic Complex. This unit is composed of granitic gneisses, quartz-biotite schists, mylonites and breccias. Deposit is related to the two faults striking 240-245° and dipping 80-65° to the north. Uranium mineralization with trace amounts of sulphides occurs in the breccia zone enclosed between the faults [5]. Primary and secondary uranium minerals have been recognized. Primary assemblage comprises blue fluorite, pitchblende, uraninite and hematite. Secondary assemblage includes several phases: metauranocircite, autunite, torbernite, uranopilite and elacherite [5, 3]. Additionally, metatorbernite, metauranocircite, metaautunite and uranophane have been found in the wall-rocks. Uranium mineralization has been investigated down to the depth 170m. Ore mineralization has been discovered by Soviet geologist S.W. Potapow in a zone of radon surface anomaly containing 880 eman ( $880 \times 10^{-10}$  Ci). Uranium mining has started in 1954 and lasted 6 years.

#### 3.3. Kletno deposit

The Kletno deposit is located in the Śnieżnik Metamorphic Complex. This Complex is composed of supracrustal series of Stronie Śląskie and infracrustal series of Śnieżnik Kłodzki. Between these two series ore-bearing zone composed of magnetite-bearing skarns, hornfelses and quartzites occurs. This zone is a host of polymetallic mineralization which includes containing pitchblende, magnetite, pyrite, pyrrhotite, arsenopyrite, sphalerite, chalcopryrite, bornite, chalcocite, galena, tetrahedrite, cosalite, stromeyerite, miargyrite, wittichenite, cinnabar, clausthalite, umangite, klockmannite, bohdanowiczite, tiemannite. These minerals formed veins, nests and veinlets filled up by quartz, barite, fluorite and calcite. Weathering zone contains sooty pitchblende, fourmarierite, uranophane, metatorbernite, native selenium, covellite, malachite, azurite, cerusite and anglesite. Geological and mineralogical features suggest a late Variscan origin of the hydrothermal mineralization [3]. Polymetallic ores have been discovered in 1948. During exploitation of the polymetallic ores, the uranium mineralization has not been considered as economic-grade. The Kletno mine has finished the production in 1957. Unknown mass of uranium ore has been reexploited from the dumps of No. 9 and No. 12 adits.

### 3.4. Kopaniec - Mała Kamienica area

Apart from the Radoniów deposit several localities of uranium mineralization have been found within the Izera Metamorphic Complex. The most important are Kopaniec and Wojcieszycze. In early 1950's mining-type exploration has been carried out at Kopaniec village and its vicinity. Uranium mineralization has been found in a tectonic zone containing breccia-type material composed of surrounding rock fragments: gneisses and mica schists. Uranium ores contained autunite, metaautunite, torbernite, metatorbernite, uranocircite, pitchblende, zeunerite, uranothorite and uranophan. Hematite and fluorite were main constituents of ore veins and nests with [6]. Hydrothermal origin has been attributed to this locality [6].

### 3.5. The Holy-Cross Mountains area

Only one locality with uranium mineralization has been recognized in the Holy Cross Mts. Pitchblende, sphalerite, galena and chalcopryrite have been reported from the Rudki pyrite deposit [7]. Ore mineralization of hydrothermal origin occurred in a tectonic zone cutting Silurian and Devonian sediments. Fragments of ore showing elevated radioactivity have been hand-separated and transported to the Kowary processing plant. The mass of uranium ore produced in this way is unknown.

## 4. URANIUM CONTAMINATION

### 4.1. Lower Silesia region

Several dumps have been left in the Kowary Podgórze vicinity as remnants of uranium mining. Four of them reveal relatively high radioactivity (Fig. 2, 3, 4, 5). The dump of the No. 19 and 19a adits at Kowary Podgórze is located in the Jedlica river valley. This dump is the largest in this area and is about 200 m long, 100 m wide and 50 m high (from the bottom of the valley). Surface  $\gamma$ -measurements made in 1971 and 1992 revealed high variations in dose rate (Fig.2 and 3 respectively). In 1971 the highest dose rate has been observed just below the dump, where meteoric water has accumulated fine-grained material with the dose above 1500  $\mu\text{R/h}$ . Spectrometric analysis of clay fraction indicated 0.11wt.% of uranium. Recently, these sediments have been eroded away. Chemical analyses of water from the No. 19 adit as well as from the river below the dump have shown  $1.0 \times 10^{-4}$  g/l of uranium. Water from No. 19 adit is still discharged by the pipe directly to the Jedlica River (Phot.1). In 1992 the highest dose rate (2000  $\mu\text{R/h}$ ) has been observed in the vicinity of No. 19 and 19a adits (Fig. 3). This anomaly has been probably caused by radon dissolved in mine water. Recent observations showed that the  $\gamma$ -values within the anomaly are changing rapidly in time. It can be connected with the changes in water flow which, in turn, depends on precipitation. Material from the dump still contains elevated amounts of uranium with a maximum up to 1500 ppm (Table 1, Phot.2). In 1971, high radioactivity has been also observed in the dumps of both No. 16 and 17 adits (Fig.4). The maximum of dose rate (1500  $\mu\text{R/h}$ ) was reported from the dumps as well as from the local forest road below the dump of No. 16 adit. This anomaly has been caused by meteoric water washing down contaminations from the dump above. Recently (Summer 1993), this anomaly does not exist any more. Yellow-colored rock fragments from both dumps revealed up to 2000  $\mu\text{R/h}$ . The dump located in front of the „Wolność” mine has been reexploited for local use in 1972-1980 probably as aggregate for construction purposes, but dose rate measured in 1971 (Fig.5) was relatively low.

Generally, all dumps at Kowary-Podgórze are well-preserved (Phot.3, 4, 5). Only the dump of No. 19 and 19a adits has been reexploited for the local use and partly eroded by the Jedlica River as illustrated by photographs taken in 1971 and 1991 (Phot. 4, 5). Erosion caused dispersion of still dangerous radioactive materials from this dump. The range of the dispersion is unknown, however, elevated radioactivity was observed up to 20 km downstream of the Jedlica River [9].

The dump at Radoniów is located amidst the agricultural land, close to the Radoniów village. It covers an area of about 25000 m<sup>2</sup>.  $\gamma$ -measurements have been made only in 1971, because till 1992

Table 1

Spectrometric analyses of different type of material from the dump and its vicinity at Kowary Podgórze No. 19 and 19a adits (after [8]).

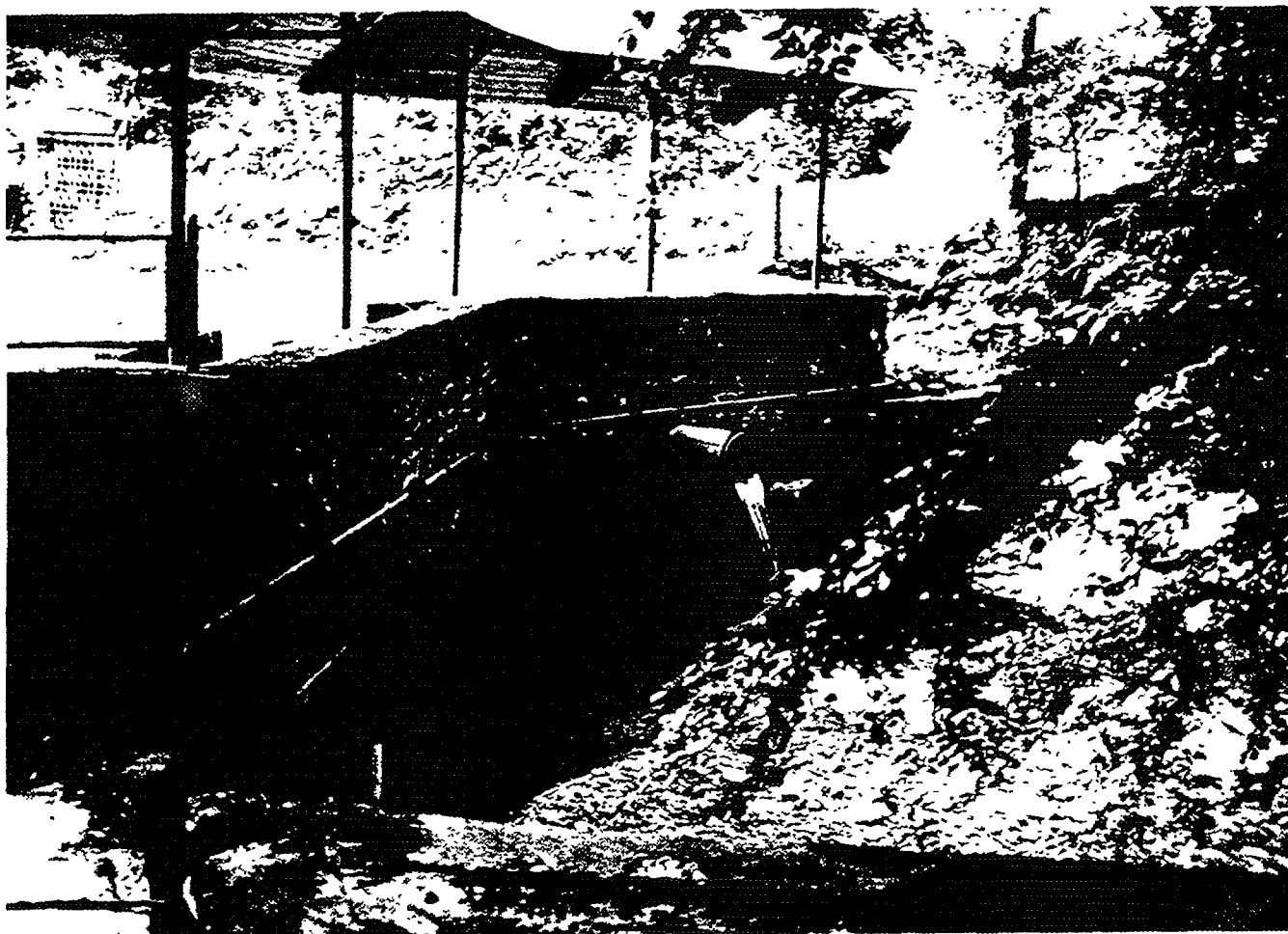
Sample	U [ppm]	Th [ppm]	$\mu\text{R/h}$	Type of material
K-1	83.744	12.521	80	sediment from the river
K-2a	4.897	4.438	2000	rock fragments, No 19 adit
K-2	4.987	5.597	2000	rock fragments, No 19 adit
K-3	3.669	1.979	140	humus material, No 19 adit
K-4	22.680	9.594	80	rock fragments, No 19 adit
K-5	108.831	9.594	120	rock fragments, dump
K-6	14.814	8.321	40	rock fragments, dump
K-7	137.587	8.231	40	rock fragments, dump
K-8	44.541	7.762	60	clastic material dump
K-10	796.512	4.219	nm	sediments from the river
K-11	0.672	b.d.l.	40	rock fragments, dump
K-12	0.714	0.800	20	rock fragments, dump
K-13	76.435	8.294	70	rock fragments, dump
K-14	69.233	6.499	60	rock fragments, dump
K-15	3.236	11.519	50	rock fragments, dump
K-16	11.676	11.409	40	rock fragments, dump
K-17	123.765	7.025	80	rock fragments, dump
K-18	6.317	8.762	30	rock fragments, dump
K-19	18.143	9.553	30	rock fragments, dump
K-19a	6.510	5.389	30	rock fragments, dump
K-20	107.686	7.304	80	rock fragments, dump
K-21	92.410	6.773	100	rock fragments, dump
K-22	130.313	8.934	80	rock fragments, dump
K-23	207.053	8.934	100	rock fragments, dump
K-24	224.268	9.943	80	rock fragments, dump
K-25	148.513	8.615	120	rock fragments, dump
K-26	1500.351	2.654	30	rock fragments, dump

\*- not measured

the dump has been almost completely reexploited for local purposes (probably for construction, compare Phot. 6 and 7). In 1971  $\gamma$ -radiation survey (Fig. 6) showed elevated radioactivity of all the collected material.

The radiation dose rate was 5 times higher than the background. Part of the dump revealed more than 500  $\mu\text{R/h}$  (Fig.6). The maximum dose rate measured on the dump surface reached 2000  $\mu\text{R/h}$ .

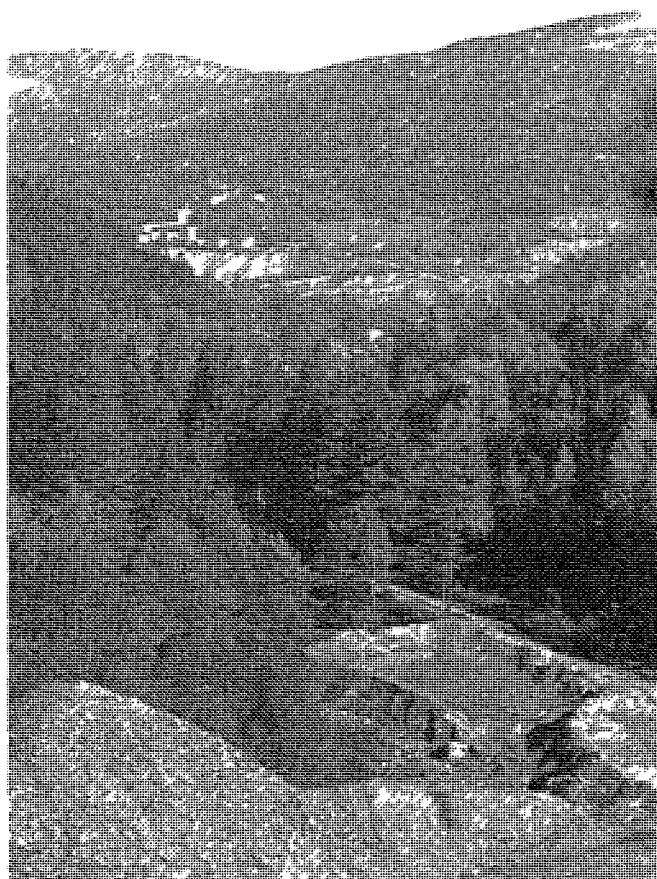
The dump has been strongly eroded (Phot.6) which caused local contamination, e.g. ash prepared from a larch, spruce and potatoes (collected 50 m from the dump) showed 260 ppm, 460 ppm and 330 ppm of uranium, respectively. Recently, the dump is exploited for local purposes (Phot.7), probably as aggregate for construction. However, rock fragments containing mineral assemblage from the ore zone are often found. Recent exploitation lacks any control and the access to the dump is free. The second time surface  $\gamma$ -measurements were made in 1994 (summer season) (Fig.7). The investigated area showed elevated radioactivity. The average of the radiation dose measured on the surface level was about 80  $\mu\text{R/h}$ , and the maximum reached 105  $\mu\text{R/h}$ . The radioactivity of selected, red colored rocks was 2-3 times higher. Spectrometric analyses of such rocks showed 490 ppm of



Phot.1 Discharge pipe of mine water from adit No. 19 flowing to the Jedlica river, Kowary-Podgórze, 1993.



Phot.2 Radioactive rock material from the dump of the adits No. 19 and 19a, Kowary-Podgórze, 1993. The radiometer indicates  $280\mu\text{R/h}$ .



Phot. 3 General view of the dump of adits No 19 and 19a  
 (upper figure, 1971).  
 Lower figure - lowermost part of the same dump with  
 waste disposal pond that has been eroded away by Jedlica  
 river (1971).



uranium. Recently (5-10 years ago), the dump was exploited for the local purpose. Crushed sand, prepared as building material reveals 30 ppm U, and soil sample which has been taken at a distance of about 50 m from the dump: 69 ppm U (see table 2). It shows that the uranium migrates from the dump to the soil. Traces of pyrite and chalcopyrite have been reported in the dump. Oxidation of sulphides can form favorable conditions of extraction of uranium from the solid rocks. All, these data show that the rocks collected on the dump reveal a significant amount of uranium. The available data are not sufficient to present a quantitative figures of dispersion of uranium around the dump.

In the vicinity of Kopaniec village (the Iżera Mts region) three dumps are known. Only one showed elevated radiation in 1971 (Fig. 8). As uranium has never been mined in this locality, the dumps are results of exploration works carried out in 1950's. The dump at Kopaniec is relatively small (Fig. 8). Field measurements showed only local but relatively high (up to 2000  $\mu\text{R/h}$ ) uranium contamination (Fig. 8). The problem of uranium pollution in Kopaniec is rather connected with ground waters. Chemical analyses made in 1962 by the ZPR-1 Company (responsible for uranium exploration) revealed a significant anomaly in local ground waters with uranium concentration reaching  $10^{-3}$  g/l, and an average content of radon up to 1000 eman (Fig. 9).



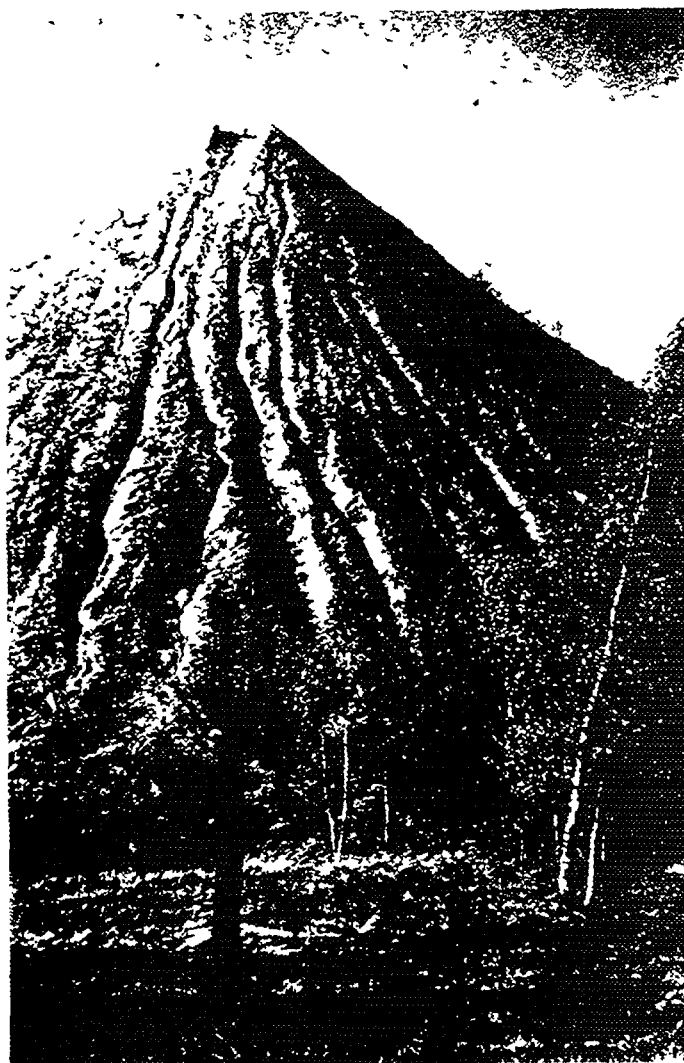
Phot.4 Initial erosion of the dump at Kowary-Podgórze, adits No. 19 and 19a, 1971.



Phot.5 Deep erosion of the dump at Kowary-Podgórze in 1993. (compare with Phot. No 4).

Water analyses made during a period of a few months in 1962 have shown quite strong correlation of physico-chemical properties of water rather with concentration of radionuclides than the volume of precipitation. The authors have no recent information about the preservation of the uranium-contaminated dump and about pollution around the Kopaniec village.

Twenty four dumps are known from the vicinity of Kletno village (the Śnieżnik Mt region). Only two of them revealed elevated dose rates (Fig. 10, 11).  $\gamma$ -measurements made in 1971 and 1991 (Fig. 10 and 11, respectively) have shown two anomalies [10]. The first anomaly is located exactly within a small swamp above the dump of the adit No. 9. The second anomaly is restricted to the edge of the dump of the adit No. 12 and to the nearest meadow. This anomaly is probably the relict of original uranium ore pile. Spectrometric analyses showed that the dump contains rock fragments low in uranium (Table 3). Single sample, taken from the dump of the adit No. 12 contained 9370 ppm U (11650 Bq/kg of Ra) (Table 3, ch-21). Recent observations reveal that the influence of uranium exploitation on the local environment in the Kletno area is relatively low. Increasing radioactivity was observed in water samples taken from Kleśnica River in front of the uranium dump (Table 4). This may suggest that rain waters are still washing out uranium from the dumps. Recently, one dump in this area has been partly covered with uranium-free waste from the adjacent limestone quarry.  $\gamma$ -measurements have shown that this carbonate coating was sufficient to blocked radiation from underlying radioactive waste.



Phot.6 Initial erosion of the dump at Radoniów by meteoric waters, 1971.

#### 4.2. The Holy-Cross Mts. area

Only one locality of uranium ores exploited in the past is known. Recently it was found that the dump around the „Staszic” pyrite mine does not exist. The dump has been subjected to land restoration and covered with fresh soil. Recent field  $\gamma$ -measurements at the sites of both dump and processing plant revealed a small anomaly containing maximum  $46\mu\text{R/h}$  of dose rate.

#### 4.3. Upper Silesia region

(Written by Irena Pluta Central Mining Institute, Katowice)

The problem of radioactive pollution of the Upper Silesia region is relatively well known from the literature [11, 12]

In the Upper Silesia region a large radiohydrological anomaly occurs in the Upper Silesian Coal Basin (USCB). Deep parts of Carboniferous coal formations contain brines with very



Phot.7 The same dump (Phot 6) after uncontrolled reexploitation for local purposes, 1992.

Table 2

Spectrometric analyses of different type material from the dump and its vicinity, the Radoniów mine. (analyses were made by Dr A. Ossowski in the Institute of Geophysics, Kraków in 1994)

Sample	U [ppm]	Th [ppm]	Type of material
R-1	42.43	4.8	debris from dewatering
	0.47	0.77	ditch
R-2	69.25	10.19	soil, 50 m from the dump
	0.68	1.12	
R-3	490.28	18.25	rock fragments from the
	3.69	5.88	dump
R-4	30.90	9.50	crushed sand
	0.48	0.84	

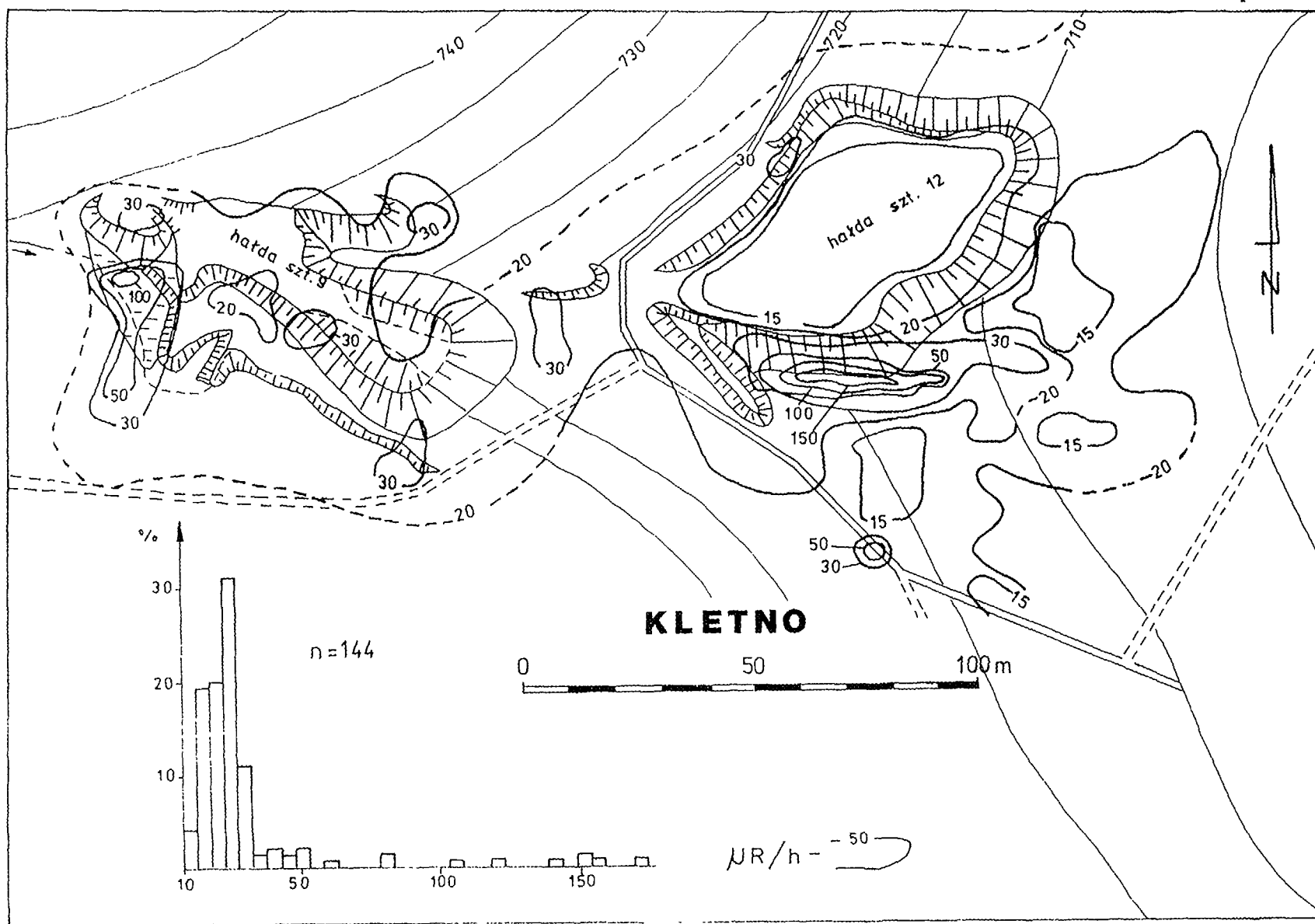


Fig.11  $\gamma$ -dose rates at Kletno, dumps of adits No. 12 and 9. Measurements carried out in 1991.

Table 3

Spectrometric analyses of various materials from the dumps at Kletno, adits No. 9 and 12.

Sample	$^{226}\text{Ra}$ [Bq/kg]	$^{228}\text{Th}$ [Bq/kg]	Type of material
ch-3	24.4	33.4	rock fragments
ch-5	40.0	30.0	rock fragments
ch-7	76.8	20.2	rock fragments
ch-8	90.6	54.5	rock fragments
ch-9	63.8	20.1	rock fragments
ch-10	45.7	24.2	rock fragments
ch-11	47.0	26.3	rock fragments
ch-12	34.4	37.6	rock fragments
ch-13	57.8	34.0	rock fragments
ch-14	80.0	45.8	rock fragments
ch-15	126.0	10.0	rock fragments
ch-17	123.0	17.0	rock fragments
ch-21	11,650.0	11.3	fragments of U ore, dump
ch-22	1,170.0	18.3	clays from the dump No 9
ch-23	50.1	12.4	rock fragments
ch-25	46.2	21.5	dump No. 14
ch-26	56.6	11.9	dump No. 15
ch-28	136.0	43.4	soil, dump No 12

Table 4

Spectrometric analyses of waters from Kleśnica river, Kletno (made by H. Mikołajczyk, Faculty of Physics and Nuclear Techniques)

Sample	Ra [Bq/l]	Ra [Bq/l]
No 3 Kleśnica river, above U dumps	0.38 + 0.34	0.409 + 0.004
No 5 Kleśnica river, above U dumps	0.34 + 0.31	0.468 + 0.004
No 9 Kleśnica river, in of U dumps	1.22 + 0.42	0.216 + 0.003
No 10 Kleśnica river, below U dumps	0.57 + 0.34	0.298 + 0.004

high concentrations of radium reaching  $400 \text{ kBq/m}^3$  of  $^{226}\text{Ra}$ . Similar phenomena have been noted in Germany (in pit- water from coal mines in the Ruhr and Saar districts) and in Iran. However, the problem in Poland is much more pronounced as the area of the USCB covers about  $4500 \text{ km}^2$  with 66 active coal mines. The mines pump out saline waters containing natural radioactive elements and discharge them to the local surface flows. Radium-bearing saline waters flow through a system of pipelines, canals and small rivers into the two largest rivers in Poland: Vistula and Oder.

About 1970's radioactivity of saline waters have been discovered [13]. These waters contained up to  $200 \text{ kg/m}^3$  of salt. Some of them contained barium ions, as well. From these brines radium is precipitated and forms sediments containing barium and radium sulphates ( $\text{BaSO}_4$  and  $\text{RaSO}_4$ ). The radioactivity of such deposits reaches  $1 \text{ MBq/kg}$  [12].

About 50% of total amount of radioactive material precipitates in the underground water system. The rest is pumped out to the surface waste disposal and to the river system. Precipitation of barium and radium sulphates causes radioactive pollution of the natural environment in mining areas and results in serious technical problems because they produce thick coatings of hard sulphates in pipelines and pumps used in dewatering systems. Radioactive deposits give high  $\gamma$ -radiation and some elevation of radon daughter concentrations in the air. However, in natural environment this contamination is limited to small areas where radium-bearing waters flow. The richest radium pollution is observed in two small rivers: Lesznica and Sztokówka. The size of pollution as well as its influence on the environment are actually under examination. Locally, concentrations of  $^{226}\text{Ra}$  in soil samples up to  $1 \text{ kBq/kg}$  have also been found.

In the USCB radium-bearing saline waters are one of the most important environmental problem because about 50% of the operating coal mines discharge polluted mine waters with Ra concentration exceeding  $10 \text{ kBq/m}^3$  [6]. Total volume of radium-polluted water is calculated to be  $184,000 \text{ m}^3$  per day and the radium dissolved in the mine water to be pumped out is estimated to be about  $252 \text{ GBq/year}$  [13].

## 5. CONCLUSIONS

Investigations of the influence of mining activity on the natural environment in Poland carried out during last 23 years revealed the local-scale radioactive contamination at four localities in the Lower Silesia. Uranium pollution is limited to the dumps and their nearest vicinity at Kowary-Podgórze, Radoniów, Kopaniec and Kleino. Radon anomaly has recently been found at Kowary-Podgórze, at the entrance to No. 19 and 19a adits and in both adits themselves, which are poorly protected against unauthorized visitors. All dumps not protected and are accessible for unauthorized visitors. Rock material from some of the dumps was locally utilized as aggregate for construction purposes. It must be emphasized that it is still possible to find fragments of uranium ores containing even up to 0.15 wt.% of U in the dump material.

The radium bearing mine waters in the USCB seem to have a significant influence on local environment as well as on stream sediments, waste disposal and mining dewatering system. In the streams, canals, rivers and pipelines recent precipitation takes place, together with sorption and coprecipitation of radon and barium sulphates. Locally, such sediments reveal high radioactivity up to  $1 \text{ MBq/kg}$ .

Sorption on clays seems to be a natural barrier in pollution mechanism. It causes removal of radioactive elements, but also local contamination of recent sediments. Such phenomena have been observed in both the Lower and Upper Silesia areas. It can be concluded that the clay minerals can be used as one of possible protection screen of radioactive wastes.

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## REFERENCES

- [1] T. Zorski, A. Ossowski, The rules of interpretation of a natural-radiation measurements. Unpublished manuscript Univ. Min. Metall., Kraków (1988), (in Polish).
- [2] K. Mochacka, Ore minerals of the polymetallic deposits at Kowary (Lower Silesia), *Prace Miner.* 4, (1966), 71 pp (in Polish).
- [3] M. Banaś K. Mochacka, Genetic types of uranium mineralization in Poland and their metallogenic relationships. In: *Metallogenesis of Uranium Deposits, Proceedings*, Vienna (1987), 187-200.
- [4] K. Mochacka, The geology of the polymetallic deposits at Kowary (Lower Silesia), *Prace Geol.* 40, (1967), 73 pp (in Polish).
- [5] S. Jaskólski, Polymetallic oxide and sulphide mineralization in gneissose granites of the Izera Mts (Lower Silesia) and its origin, *Prace Geol.* 43, (1967), 1-77 pp (in Polish).
- [6] M. Banaś Preliminary information about uranium mineralization at Kopaniec. (*Proceedings of PAN Kraków*), 241-242.
- [7] M. Nieć, Iron sulphide and siderite deposit in the Holy - Cross Mts. *Biul. Inst. Geol.*, Warszawa, (1968), 237, 85-98.
- [8] M. Bała, J. Jarzyna, N. Górecka, A. Ossowski, A. Piestrzyński, T. Zorski, Application of geophysical methods in environmental protection. *Int. rep. of the Institute of Geophysics*, Kraków (1992) 50pp.
- [9] J. Peńsko, J. Jagielak, M. Biernacka, A. Zak, Investigations of radioactivity at Kowary valley. *Nukleonika* v. 16, (1971), 5-6, (in Polish).
- [10] M. Banś, A. Piestrzyński, W. Mielniczuk, The secondary haloes of uranium dissemination round mine dumps. *Nukleonika* v.21, (1976), 639-645.
- [11] J. Lebecka, K. Skubacz, S. Chałupnik, I. Tomza, I. Pluta, J. Skowronek, Influence of mining activity on distribution of radium in the natural environment. *Proceedings of the 4<sup>th</sup> Working Meeting on Isotopes in Nature*, Leipzig, Germany, (1986), 149-153.
- [12] K. Skubacz, J. Lebecka, S. Chałupnik, M. Wysocka, Possible changes in the background radiation of the natural environment caused by coal mining activity. *Proceedings of Intern. Symp. on Nuclear Techniques*, Vienna 1990, (1991), 425-429.
- [13] K. Skubacz, J. Lebecka, S. Chałupnik, M. Wysocka, Possible changes in radiation background of the natural environment caused by coal mine activity. *Energy Source*, v.14, (1992) 149-153.



# CONSIDERATIONS ON SOME RADIOACTIVE AREAS IN ROMANIA POTENTIAL SUBJECTS FOR ENVIRONMENTAL RESTORATION

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## Abstract

Radioactive substances have started to be used in Romania practically at the same time with most of Eastern European countries, at first in the medical education fields, as well as minor technical applications during the fifties; radium being the main radioactive element that was used. After the year 1950, artificial radioactive substances have been imported; it also began the exploitation of radioactive ores. By 1957, the radioactive substances national production completed the existing radioactive sources. Subsequently to the nuclear field development, various nuclear regulations and appropriate authorizing and control organisms have been established, as well as the environmental radioactivity supervision by national networks. The radioactive waste question has been relatively well controlled, as far as medical and industrial waste is concerned. For some time its collection and dumping was not made in optimal conditions. This problem got solved by decommissioning the improvised dumping site, treating the waste in a special installation and depositing the confined and concentrated waste in depth geological formation. As a generic remark, in Romania are not any areas of uncontrolled accumulation containing radioactive wastes, which could be taken as a starting points for environmental restoration programs. Some incidents whose results were the contamination of small areas have been solved, no environmental restoration being required today. The main characteristics of the radioactive areas that arising as a consequence of radioactive ore exploration and exploitation as well as from the concentration of natural radioactive substances in fertilizer facilities or coal thermocentrales indicate that these could be potential subjects to be included in the environmental restoration programme. One of the conclusions, is that the environmental restoration programme initiated by the IAEA as well as international co-operation for its achievement is necessary and opportune.

## 1. Generalities, historical

Radioactive substances have started to be used in Romania practically at the same time with most of Eastern European countries, at first in the medical education fields, as well as minor technical applications during the fifties; radium being the main radioactive element that was used.

The radioactive substance usages in Romania has been initiated by physicists, teachers and physicians, most of these scientists completed their studies in Paris some of them has

participated and co-operated to the first world-wide radioactivity researches.

Proper radioactive sources, as the current word is used were imported for medical treatments; pins for cancer tumors and even balneary salts; for demonstrative works in the university labs; for luminescent dials painting and even in some electrical pillows used in heating, that have been imported; radium being the main radioactive element that was used. Obviously the amounts were small but not exactly negligible approximately in the range of a few radium curie before the fifties.

Most of these sources were procured and retrieved without any major complications, but some of these have had certain leaks; but the work was performant even in those days.

By the fifties scientists started to do some researches and applications in chemistry, physics, medicine, biology with radioactive substances, imported artificial products; the results have been presented at the first international conferences in Geneva, on Peaceful Application of Atomic Power.

Concerning artificial radioactive sources, usage have got a relatively strong impulse as soon as in July 1957 at IFA - Bucharest, a VVR Reactor become operational. Its purpose was the radioisotope production - the reactor is still operational and serves the same purpose.

Besides the medical applications such as radiodiagnose and radiotherapy, that became a common method in almost all the romanian hospitals, there have also appeared applications in the oil industry, including tracers, in the iron and steel works, blast furnaces, radiobiology, etc.

The legislation, authorization, control and supervision developed simultanously to the applications, so presently they require some improvements, completion and coordination, stipulated by the international programs.

The first legislation on this subject has been adopted by governmental decision in 1961. Unfortunately it did not include references to the uranium mines and medical X - rays fields. On the bases of this decision there has been established a committee to lead, control and authorize all nuclear activities, at the Institute of Atomic Physics. As a result of the instructions drawn up by the committee, its control and relationships with many researchers, that were attending preparatorial classes at the IFA in order to obtain their license, the radioprotection and nuclear security matters for the given domain to be properly handled, according to the level and requirements of that time.

During the same stage in Romania have been organized national networks for environmental radioactive supervision, for supervising the food stocks, etc. The networks were continuously developed and they are presently covering in a satisfactory way the romanian territory. The results that were obtained are reliable to be used for ecological restoration. The organisatorial and methodological capacity as well as their adaptability of these networks recommend them as partners for any institution that is interested in ecological restoration. [1, 2, 3, 4, 7, 8, 9, 14, 15, 16, 17, 18, 19, 20, 21]

In 1974 there has been adopted a new national regulation having the meaning of a law, together with many technical practices concerning radioprotection, transportation, ore exploitation, etc.

Almost all the guides are still in force today, but they are to be improved in the way the RAPAT, WAMAP missions of IAEA [1, 2]

recommended, at the time of their visits to Romania (1990 - 1991).

Nowadays, the authorization and control are exercised through a national committee by the subsecretary of state in the Waters Forests and Environmental Protection Ministry. This institution does not depend on utilisers or nuclear energy promotion organisms.

The radioactive activities were rather low, consequently the radioactive contamination of some areas was out of question.

The radioactive wastes from the radioactivity applications, the research, the education, the medicine, the industry, and from other related activities distinct from the uranium mines, were collected between 1957 - 1977 in Institute of Atomic Physics empowered from 1961 and afterwards by the 1974 Law as a national institution in this respect.

In the case of short life - time radionuclides some exceptions were accepted, in sense of the local collection through on sites repositories. In this way, these low activity local repositories were entirely restored, no problem being announced.

Unfortunately the former centralization tendency has conducted to the storage of the radioactive wastes in a fort built before the first World War in the proximity of the present IFA site.

Of course, it was an improvisation determined by an urgent situation, but in fact it was a better solution than an uncontrolled scattering of radioactive waste in the country in the practical situation of entirely lack of repositories and special radioactive waste treatment facilities.

One of the fewer advantages of this solution was the possibility of an invariably temperature during the year, consequently the unconfined and biological effects were practically excluded.

During 1977 - 1978 this storage was decommissioned the radioactive waste being tranfered in IFA - Radioactive Waste Treatment Plant ( its commission took place in 1975), the wastes were treated, confined in 220 l metallical drums [16].

This activity continued through the establishment of a national repository for radioactive waste in a depth geological formation, in fact a former richest uranium ore in Romania. In this way, this national repository for low and medium radioactive waste was subject of IAEA - WAMAP and RAPAT expert missions in Romania.

Incidents or accidents inherent in the development of nuclear field took also place in Romania on small controlated areas. Thanks to the administrative system, these incidents have been reported, confined and eventually controlled by collecting the resulting radioactive waste. Some examples are worth to be verified, such as the radioactive contamination of some fields and trees during experiments performed at the Forest Research Center. Their results led to an environmental restoration programme, by the year of 1975, [17].

A quantity in the range of a few tenths of soil cubical meters, as well as tenths of Cs-137 and Co-60 from contaminated trees has been collected. One of the major methodological impediments in dealing with this question was the lack of national or international regulations regarding the acceptable residual concentrations in the soil. At present the supervision networks do not have specific, quantitative criteria to declare

an artificially contaminated field or even an area where the concentration is too high. In Romania medium values of the soil activity or comparisons to a reference field there are used.

We regard the solving of the maximal admitted values or the reference values as being a major problem in finding the areas that required environmental restoration as well as the subsequent determination of restoration effects.

Other incidents were those concerning the utilization of the radioactive tracers for oil wells, that led to small floods and to the contamination of the surrounding areas. These were promptly solved by the personnel involved. A little more difficult problems were those involving some contaminations of the refractory materials that resulted from blast furnaces demolition. The furnaces were marked by inappropriate sources containing radioactive powders for wear checking.

Consequently, one could be confident that Romania has no radioactive contaminated areas resulted from incidents or accidents that would require the inclusion in environmental restoration programs.

Uranium exploration and exploitation in Romania started in 1950, and in their evolution there are three distinct stages depending on the way of organizing [18, 19, 20].

In the year 1950, the exploration and exploitation of uranium ores has begun, having three main stages of development according to the administrative system.

The founding of the Romanian - Russian joint venture SOVROM -CUARTIT marks the beginning of the first period which will be ended in 1961 when the soviet consultant leave the country.

The second stage is when the activities in this field gathered in the Rare Metals and Enterprise between 1961 - 1989, a state organization with industrial profile.

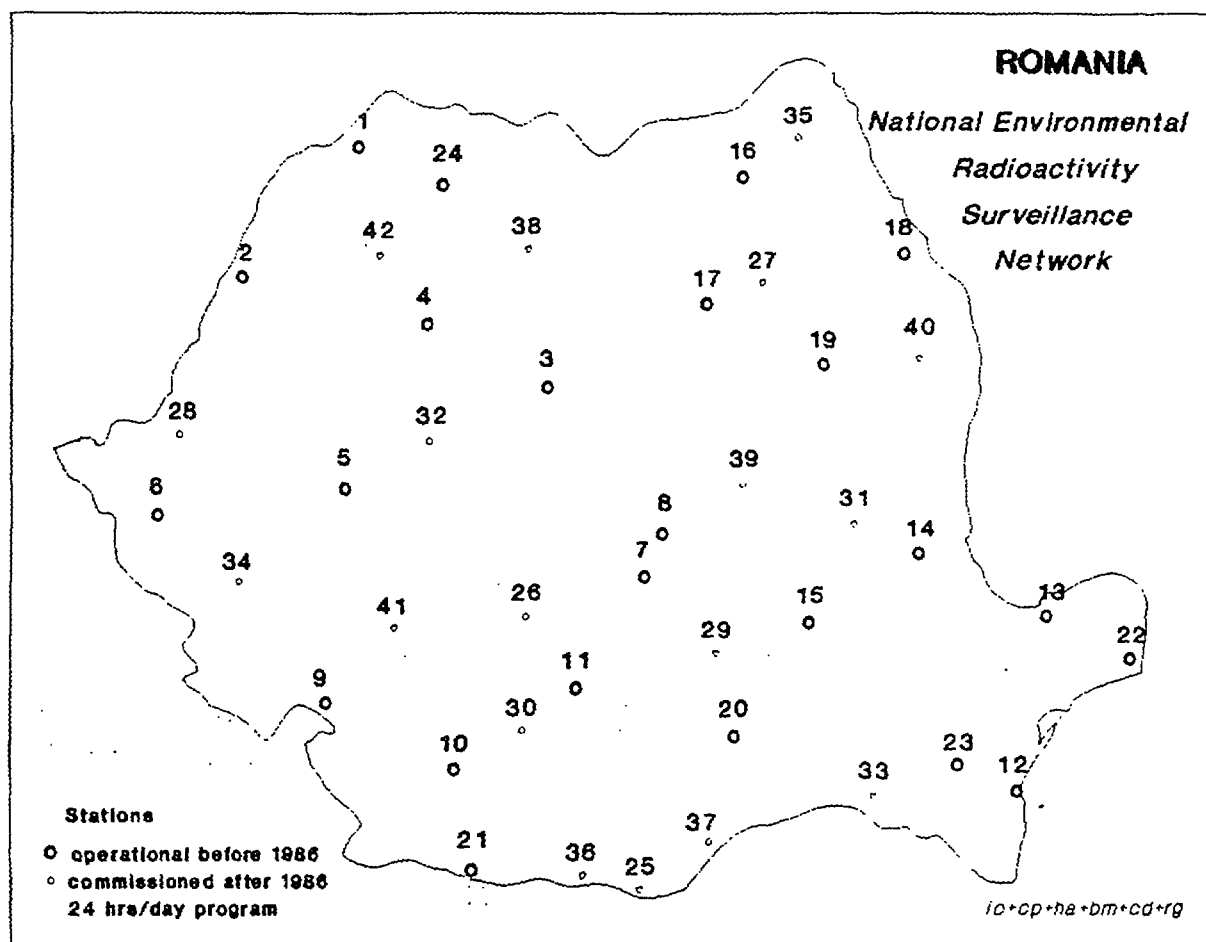
The third stage started in 1990, in the condition of a free economy, the entire uranium exploration and production activity is organized within the Rare Metals Autonomous Regia.

The first period is characterized by the development of ample geological surveys, air and land radiometric works, research studies in the underground mines and drillings executed for the exploration of the other metals. Thus two uranium metalogenesis provinces have been outlined in the western part of the country in the Apuseni Mountains and Banat Mountains respectively. Even during the first period within these areas, four medium size uranium deposits, (Ciudanovita, Natra, Dobreiul de Sud and Avram Iancu), and a large one Baita - Bihor, known at that time as the richest deposit in the world, were discovered. From exploration and exploitation activities, result that there are potential areas for environmental restoration.

## **2. Radioactive areas in exploration and exploitation of the radioactive ores; related environmental problems**

The activities of the nuclear raw materials ore exploration and exploitation are developed in Romania since 40 years ago and the dressing of these materials since 15 years ago.

Until the year 1965 the nuclear raw materials ore with a high grade of concentration was exported and after this period the ore was stocked at mining site until 1978 when the processing plant, named "Uzina R", became operational. This plant process nuclear raw material resulted from present mining activity and from stocks accumulated in the period 1965 - 1978.



National Environmental Radioactivity Surveillance Network Stations

- |                        |                   |
|------------------------|-------------------|
| 1 - Satu-Mare          | 22 - Sf.Gheorghe  |
| 2 - Oradea             | 23 - Cernavoda    |
| 3 - Tg.Mures           | 24 - Baia-Mare    |
| 4 - Cluj-Napoca        | 25 - Zimnicea     |
| 5 - Deva               | 26 - Rm.Vilcea    |
| 6 - Timisoara          | 27 - Piatra-Neamt |
| 7 - Babele             | 28 - Arad         |
| 8 - Brasov             | 29 - Ploiesti     |
| 9 - Drobeta Tr.Severin | 30 - Slatina      |
| 10 - Craiova           | 31 - Focsani      |
| 11 - Pitesti           | 32 - Alba-Iulia   |
| 12 - Constanta         | 33 - Calarasi     |
| 13 - Tulcea            | 34 - Resita       |
| 14 - Galati            | 35 - Botosani     |
| 15 - Buzau             | 36 - Tr.Magurele  |
| 16 - Suceava           | 37 - Giurgiu      |
| 17 - Toaca             | 38 - Bistrita     |
| 18 - Iasi              | 39 - Sf.Gheorghe  |
| 19 - Bacau             | 40 - Vaslui       |
| 20 - Bucuresti         | 41 - Tg.Jiu       |
| 21 - Bechet            | 42 - Zalau        |

FIGURE 1

The waste resulted by radioactive ore exploration, exploitation and dressing are:

- sterile rocks resulted from mining exploitation and exploration activity; a total of 5 350 000 tonnes.
- waters from mining exploitation with uranium worth above the normal background; activity/content 1-7 pCi Ra/l; 10-40 pCi alpha/l.
- waters from dressing plants; a total of 3 000 000 tonnes; 0.02 - 0.04 % U.
- uranium ores with concentrations between 0.02 % - 0.05 % which are deposited at mine site.
- ion exchange resin wastes from processing plant total of 1500 tonnes.

Present low-grade ore make the potential radon releases from mill residues relatively low.

The action to decrease environment contamination were applied from 1974 since " The Law 61 " about the activities in the nuclear fields.

A situation of the radioactive wastes, and environmental problems, is giving detailed on the departments of the Rare Metals Autonomous Administration.

#### 1. BIHOR MINING EXPLOITATION (IN OPERATION)

- Pile of ore: 16 600 m<sup>2</sup>;
- Piles with low level radioactivity: 529 300 m<sup>2</sup>.
- No wastes treatment at present.
- Forecasted measures for wastes processing:
  - \* the construction of water conditioning plant.
  - \*\* recovery of U by heap leaching
- Environmental restoration:
  - \* after conditioning waters will flow into Baita River.
  - \*\* the sterile will be covered with a vegetal strata.

#### 2. BANAT MINING EXPLOITATION (IN OPERATION)

- Piles with low level radioactivity + pile of sterile from the radioactive mining exploitation: 209 500 m<sup>2</sup>.
- Two waters conditioning stations (50 % efficiency)

- Forecasted measures for wastes processing:
  - \* modernization of the existing stations
  - \*\* recovery of U by leaching present stored low grade ores heaps
- Environmental restoration:
  - \* after conditioning the waters will flow into Ciuanovita and Natra river.
  - \*\* the sterile will be covered with a vegetal strata.

### 3. CRUCEA MINING EXPLOITATION (IN OPERATION)

- Piles with low level radioactivity: 364 000 m2.
- One depollution station (80 % efficiency)
- Forecasted measures for wastes processing:
  - \* modernization of the existing stations
  - \*\* recovery of U by leaching present stored low grade ores heaps.
- Environmental restoration:
  - \* after depollution the waters will flow both into Greaca and Botusana rivers.
  - \*\* the sterile will be covered with a vegetal strata.

### 4. SECTION II OF GEOLOGICAL WORKS TULGHES (IN OPERATION)

- Piles with ore: 1 400 m2;
- Piles with low level radioactivity + pile of sterile from radioactive mining exploitation: 156 800 m2.
- No treatment at present
- Forecasted measures for wastes processing
  - \* construct a station for mine wastes conditioning.
  - \*\* recovery of U by heap leaching at dumping sites.
- Environmental restoration:
  - \* after decontamination waters flow into Barsan river.
  - \*\* the sterile will be covered by a vegetal soil strata.

## 5. SECTION V OF GEOLOGICAL WORKS ALBA-IULIA

- Piles with ore: 500 m<sup>2</sup>;
- Piles with low level radioactivity + pile of sterile from radioactive mining exploitation: 122 300 m<sup>2</sup>.
- No treatment at present
- Forecasted measures for wastes processing - recovery of U by heap leaching.
- Environmental restoration:
  - \* the sterile will be covered by a vegetal soil strata.

### TOTAL ON PILES CATEGORIES

- Piles with radioactive ores: 18 500 m<sup>2</sup>;
- Piles with low level radioactivity + pile of sterile from radioactive mining exploitation and exploration: 1 381 900 m<sup>2</sup>.

TOTAL: 1 400 400 m<sup>2</sup>.

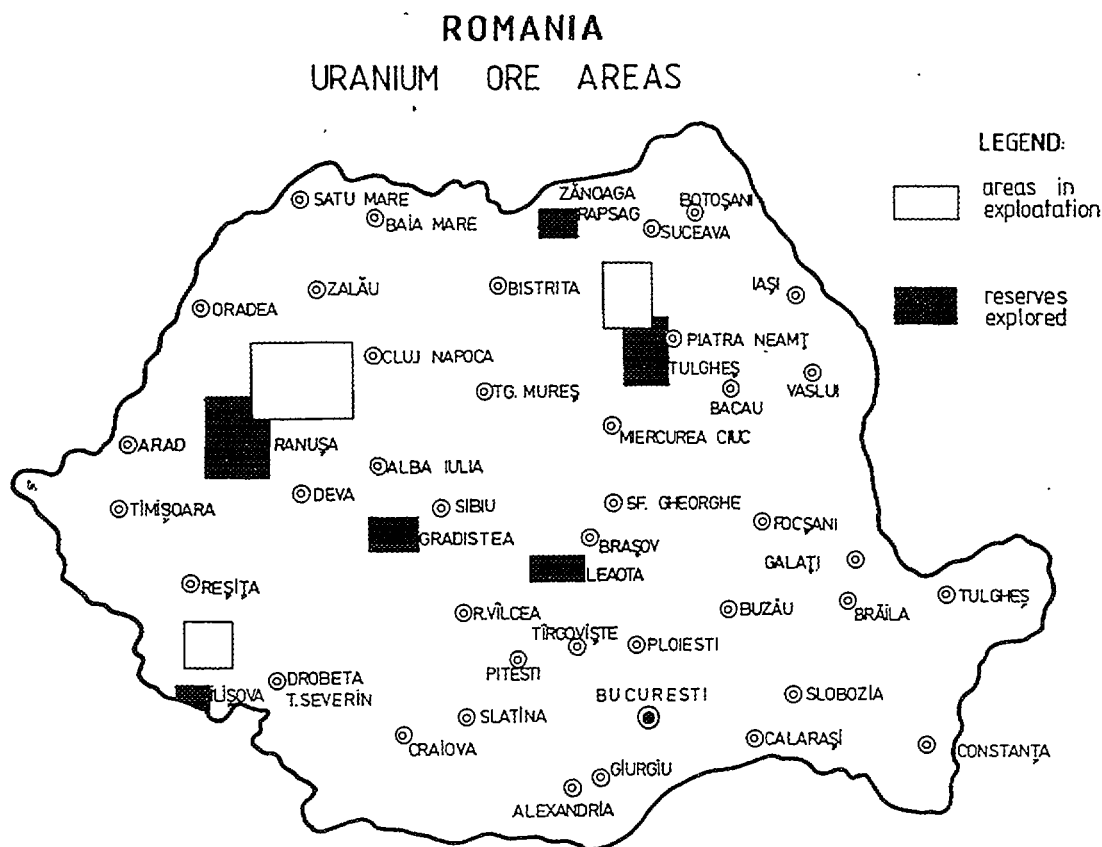


FIGURE 2



### 3. Radioactive areas in fertilizer industry

Romania is an important producer of phosphate fertilizer obtained from imported phosphates.

About 85 % of these phosphates are of sedimentary origin therefore are radioactive, containing 0.008 - 0.015 % uranium, usually in secular equilibrium with its decay products.

The phosphate fertilizer industry is located in eight large industrial plants, each having a processing capacity of 330 000 t/yr phosphates. Almost 3 million t/yr phosphates are processed, in Romania an eventual elimination, and recovery, of uranium could lead to 200 t/yr uranium. Half of the phosphate capacity mentioned are treated in 4 plants by nitric acid attack, obtaining finally a complex fertilizer of NP or NPK type containing all radioactivity of initial phosphates (both in U and Ra). The calcium carbonate resulting has waste as nonsignificant radioactivity. In the case of Kola volcanic phosphate the radioactivity of fertilizer is lower than 150 Bq/kg.

The result of approximative 1.5 million t/yr phosphates are processed in 4 plants by sulphuric acid attack. In this process the phosphate rock is dissolved as phosphoric acid resulting at the same time calcium sulphate (dihydrate) as  $\text{CaSO}_4 \cdot 2\text{H}_2\text{O}$  containing also some insoluble phosphate i.e. is named phosphogypsum which is considered a waste (one tonne of phosphate rock gives 1.5 tonnes phosphogypsum).

During the phosphate fertilizer using sulphuric acid attack process almost all uranium is dissolved in the intermediate product phosphoric acid and will be found in the fertilizer obtained. No uranium is carried by phosphogypsum obtain in the process as waste. Therefore  $^{226}\text{Ra}$  is found in this waste.

Depending on the phosphate rock used, the radioactivity for phosphates of sedimentary origin is 400 - 1 000 Bq/kg.

In the case of Kola volcanic phosphate, the radioactivity of phosphogypsum is 1 - 100 Bq/kg.

In this paper we must insist on phosphogypsum which is a very undesirable waste for each fertilizer plant due to large quantities accumulated each year. Such a plant working at normal capacity of 330 000 t/yr phosphate, apart of chemical fertilizer obtained also produces a phosphogypsum waste. The quantity of phosphogypsum involved is almost 500 000 t/yr as waste, which has to be deposited around the plant.

Until now, some quantities were used as soil amendment, or in the cement industry, but most of it accumulates in this deposit giving a head ache to producer. The main problem of phosphogypsum waste is its radioactivity, which makes it worthless for any other industry, including its used in agriculture. All the sulphur of sulphuric acid is bound by phosphogypsum.

Sulphur recovery may be an incentive for an eventual processing of phosphogypsum, but until now, no economic process was envisaged.

At present, to these problems, the radium radioactivity is also added involving dramatic restrictions for phosphogypsum removal from its deposits. Dissolution in water is also forbidden having in view radium accumulation in various plants and marine life. The chemists are now trying to eliminate  $^{226}\text{Ra}$  from phosphogypsum, but this problem is extremely difficult, due to its similarity with calcium.

This kind of preoccupation exists in Romania, but phosphogypsum used in construction materials is now forbidden.

Because no significant use of phosphogypsum was found, its accumulation in the deposits continues. The problem is that the phosphogypsum deposits are within 1 - 2 km range of human habitat. It must be remembered, that large accumulations of this material is also a hazard to human life, due especially to radon involved and dust (containing  $^{226}\text{Ra}$ ) spread by winds. The only solution envisaged at present seems to be a suitable protective layer.

Therefore, in Romania are four fertilizer plants processing phosphate by sulphuric acid attack leading to phosphogypsum waste. All these plants are located near human agglomerations, as the phosphogypsum deposits.

The four plants mentioned are located at:

1. Valea Calugareasca - 60 km North of Bucharest having phosphogypsum deposits of roughly 5 million tonnes, site on the two large areas of 27 and 29 ha (hectares).

2. Navodari - 250 km East of Bucharest, on the Black Sea Coast, with approx. 4 - 5 million tonnes, sited on three deposits with total 40 ha.

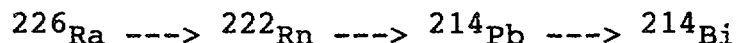
3. Bacau - 300 km North-East of Bucharest having phosphogypsum deposits of roughly 5 million tonnes, sited near plant on 17 ha areas.

4. Turnu-Magurele, on the Danube - 100 km South-West of Bucharest, with 5 million tonnes, sited on 20 ha. All these deposits have a nonuniform radioactivity, because no distinction among various types of phosphogypsum of different origin are given bellow:

- Iordan	- 800 Bq/kg;
- Marocco	- 1 100 Bq/kg
- Florida (USA)	- 1 000 Bq/kg
- Kola	- 100 Bq/kg
- Siria	- 700 Bq/kg

All Romanian plants are processing different types of phosphate rock, therefore phosphogypsum radioactivity has an average value of 500 - 1 000 Bq/kg but variations are possible as dependent on sampling place.

Phosphogypsum radioactivity depends mainly on the decay chain disintegration of:



Alpha radioactivity of  $^{222}\text{Rn}$  gas can be measured by special devices and methods.

Gamma radioactivity of phosphogypsum can be measured by the agency of Ge gamma - ray spectrometry system including Ge detector, electronics and computers [21].

#### 4. Radioactive areas resulted from coal fired power plants

Coal, as well as all natural materials contains traces of natural radionuclides. So, the coal - fired could imply the atmospherical emission of radioactive products, in this sense enhancing the public exposure.

So far, in Romania the measurement have been performed in the proximity of Mintia Thermal Power Plant (Mintia TPP).

Presently, these measurements are initiated in a frame of a research contract with the main goal: the radiological impact of Mintia TPP as main goal. Some preliminary results are given in the table no. 4.1.

Table 4.1

Natural Decay Products at Mintia - Deva

Radionuclide	Local samples pCi/m <sup>2</sup> xday	Reference samples pCi/m <sup>2</sup> xday	In Depth soil pCi/g	Superficial soil pCi/g
U-235	0.7+/-0.4	< 0.01	0.10+0.001	0.20+0.02
Ra-226	8+/-6	0.2 - 1.7	1.5+/-0.2	3.0+/-0.3
Pb-214	4.1+/-1.0	0.1 - 1.0	1.1+/-0.3	2.0+/-0.05
Bi-214	3.2+/-1.0	0.1 - 1.0	0.9+/-0.1	1.7+/-0.06
Ac-228	4.5+/-2.5	0.1 - 0.4	1.0+/-0.05	1.4+/-0.1
Pb-212	3.9+/-0.7	0.1 - 0.4	0.9+/-0.3	1.5+/-0.04
Bi-212	< 6.9	0.1 - 0.4	1.1+/-0.2	2.1+/-0.4
Tl-208	1.0+/-0.5	0.03- 0.1	0.3+/-0.1	0.53+0.03
K-40	37.9+15.8	2.0 - 4.6	11.5+/-0.3	14.1+/-0.6

In this respect, the exposure doses in the proximity of Mintia exceed enough the values determined from reference samples (being collected from other sides in Romania). This conclusion remain valid for the all natural radionuclides from depository samples. In order to determine the influence of this TPP on soil, both superficial and in depth soil samples were considered. It is worth to underline that the differences between the two kind of samples consist in a greater radioactivity in the superficial soil.

The measurements show the persistence of the radioactivity in volatile ash. The collective effective dose equivalent commitments owed to irradiation due to deposited activity resulting from a atmospheric release from old and modern coal-field power plants is given in the table no. 4.2 [22].

Further results will give us a comprehensive image on these aspects, in this respect the samples collected by the agency of National Network for Environmental Survey of Radioactivity will be considered [23].

Table 4.2

Coal-fired Power Plants	Internal Inhalation Due To Inhalation manSv per GW	External Inhalation Due To Inhalation manSv per GW
modern		
Romania	0.17- 2.31	0.009- 0.101
World- wide [24]	0.26	0.015
old		
Romania	7.2- 31.0	2.4- 10.5
World- wide [24]	1.35	0.30

## ROMANIA

LOCALITIES WITH RADIOACTIVE AREAS DUE TO FERTILIZER INDUSTRY AND COAL THERMOCENTRALS



FIGURE 3

## Conclusions

1. In Romania the specialists from monitoring networks, from Ministry of Health, from Ministry of Environment and from other different laboratories have carried out a number of studies related to the persistence and the effects of accumulation of the natural decay products due to human activities. These informations are of quite importance and provide a good background for an effective environmental characterization.

2. For a comprehensive evaluation, ecological studies, risk assessment, and cost-benefit analysis are required.

3. A good radiological survey plan is essential element in implementing the cleanup actions, for this purpose national experts should perform studies according to RER-IAEA programme.

4. The problems raised in the field of environmental restoration should be split into two classes, the first relates to the low specific activity, widely dispersed and the second relates to much smaller areas of contamination which are often of much higher specific activity.

5. So far, the national standards for soil contamination levels, clean-up levels or criteria for deciding such levels were not elaborated.

6. A first phase in these effort, focused on reviewing the accumulated experience may well conclude that not all the data on the record are truly reliable. While one should certainly bear in mind this aspects, the next phases must however concentrate on a proper preparation of the personnel, equipment and logistics as required by the technical dimensions of environmental restoration.

7. The IAEA programme on environmental restoration, should rightfully be considered as the most advanced, as far as strategy, technical dimension and international cooperative framework. Consequently, it is believed that the side programme may set terms of references for other industrial sectors, much as the chemistry. It is another instance when the Nuclear and its steering International Agency take the initiative and serve sound, lucrative examples of good comprehensive management.

8. In the following stages of the environmental restoration programme, the government institutions and entrepreneurs are the main pawns. The entire activity should be co-ordinated by adequate bodies; media publishing must gain public acceptance.

## REFERENCES

- [1] WAMAP Mission , IAEA International Report, Vienna(1990).
- [2] RAPAT Mission , IAEA International Report, Vienna (1991).
- [3] Laboratorul National de Poluare a Aerului; "Studiu Asupra Poluarii Radioactive in Vecinatatea Minelor", Bucuresti) (1988).
- [4] Sandru, P., Norme de Management al Deseurilor Radioactive, Raport Intern CSEN 1980- 1982.
- [5] Petcu, P., Sandru, P., Radioactivitate Aplicata in Industria Petroliera, Bucuresti (1971).
- [6] Sonoc, S., National Conference on Radioprotection, Oradea (1993).
- [7] Sonoc, S., Environmental Survey of Radioactivity Factors, Bucharest (1990).
- [8] Dinca, G., et. al., Natural Irradiation and its Influence Upon Health in Mining Industry, Review Paper, Bucharest (1991).

- [9] Nilesco, L., et. al., The Radioactivity of the Environment in the Area of the "R" Nuclear Industrial Unit of Feldioara, Bucharest (1991).
- [10] Herdea, N., et. al., Results of a Program in Subsurface for Determination on Concentrations of Natural Radionuclides in Non-Uranium Mines, Bucharest (1991).
- [11] Noditi, M., et. al., Possible Influences of Low Levels Background Radioactivity on Mortality by Digestive Cancer in the West Part of Romania, Bucharest (1991).
- [12] Modoran, N., et. al., Modifications of Natural Radioactivity by Coal Thermocentrals, Bucharest (1991).
- [13] Botezatu, E., et. al., Validation of Population Doses Depending on Filtering Efficiency in Coal - Fired Coal Power Plants, Bucharest (1991).
- [14] Dinca, G., et. al., Methodology for Monitoring the Internal Contamination with Natural Uranium in Mining of Uraniferous Ores, Bucharest (1991).
- [15] Toro, L., Modelling the Environmental Contamination by Wastes from Uranium Milling Activities, ( 1991).
- [16] Sandru, P., Decontaminarea Depozitului Intermediar de Materiale Radioactive Fort-Magurele IFA, Raport Intern, Bucuresti (1978).
- [17] Sandru, P., Decontaminarea Zonei Impadurite Scrovistea, IFA Raport Intern, Bucuresti (1975).
- [18] Bejenaru, C., Cioloboc, D., Uranium Exploration and Production in Romania, Bucharest (1992).
- [19] Bejenaru, C., Caplan, M., Areas Containing Ore and Sterile Uranium, Bucharest (1993).
- [20] Ghilea, S., Coroianu, A., Fekete, F., Environmental Radioactive Contamination Caused by Uranium Ore Mining; International Symposium on Remediation and Restoration of Radioactive Contaminated Sites in Europe, Antwerpen (1993).
- [21] Bunus, F., Dumitrescu, R., Radioactive Areas Due to Fertilizing Industry, Bucharest (1993).
- [22] Botezatu, E., et. al., Coal Fired Power Plants As Source of Population Exposure; National Conference on Radioprotection, Oradea (1993).
- [23] Sonoc, S., et. al. , The Radiological Impact of Mintia Coal - Fired Power Plant on the Mintia - Deva Zone, Bucharest (1993).
- [24] UNSCEAR Report/1988 Annexa A.

# IDENTIFICATION AND CHARACTERIZATION OF RADIOACTIVELY CONTAMINATED SITES IN RUSSIA

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## Abstract

*The present material is devoted to description of nuclear and radiation legacy of Russian Federation. The paper contains classification of radioactively contaminated sites in Russia and their characterization, including history, sources of contamination, surface areas, proximity to population centers, levels of radioactivity and spreading of contamination. It is noted that corresponding information is rather limited, and there is much scope for further evaluation and detailed elaboration of quantitative data.*

## 1. INTRODUCTION

The main purpose of this paper is to present a photographic picture of radioactively contaminated territories of Russian Federation. Naturally, some important but small details remain in shade. Some data require additional evaluation and some information is still unknown or even concealed.

Nevertheless, the data presented give enough complete notion about the general radiological situation in the country, and it is unlikely that additional details can incert radical changes in the picture described.

Analysis of the information available allows to distinguish a number of categories of sites which have been radioactively contaminated due to the following reasons:

- (1) In consequence of nuclear accidents.
- (2) By past operation of defence-related, fuel cycle and research facilities.
- (3) By bad practice of radioactive waste management and dumping.
- (4) In consequence of military nuclear testing and peaceful nuclear explosions.
- (5) By other reasons.

Although methodical perfection and comprehensivity of this classification might be a subject for discussion, in practice it covers almost all possible cases of contamination. The present material is organized in accordance with the above classification.

History, sources of contamination, contamination boundaries, surface areas, proximity to population centers, radioactive concentrations and spreading of contaminations are reflected in the text and/or in illustrative materials as completely and correctly as it was possible to do on the basis of present knowledge.

## 2. CONSEQUENCES OF NUCLEAR ACCIDENTS

Only the major nuclear accidents - Chernobyl (1986) and Ural (1957, 1967) - are discussed in this paper. Such approach is accepted because the consequences of these tragic events cannot be compared with contaminations due to the other known accidents (e.g., Tomsk-7, Leningrad NPP, etc.). The last conclusion is made, of course, with full understanding of the fact that any nuclear accident, regardless of its scale, requires the most close attention and prompt adequate actions.

### 2.1. CHERNOBYL ACCIDENT

According to the recent estimations [1], the following summarize the releases from the Chernobyl reactor during the accident (more detailed data are presented in Table I):

- the entire inventory of noble gases was released
- 50% to 60% of the  $^{131}\text{I}$  inventory was released, which corresponds to 1,5 to 1,9 EBq (40 to 50 MCi)
- $(33 \pm 10)\%$  of the  $^{137}\text{Cs}$  inventory was released, or  $85 \pm 26$  PBq ( $2,3 \pm 0,7$  MCi)
- approximately 4% of the  $^{90}\text{Sr}+^{90}\text{Y}$  inventory was released, or 7 PBq (0,2 MCi)
- approximately  $(3,5 \pm 0,5)\%$  of the fuel was released outside the immediate vicinity of the plant.



TABLE 1. ESTIMATES OF RADIONUCLIDE ACTIVITIES RELEASED DURING THE CHERNOBYL ACCIDENT (THE ACTIVITIES ARE DECAY-CORRECTED TO APRIL 26, 1986) [ 1 ]

Radionuclide	Activity, MCi	% of the inventory of some important nuclides released*
Xe - 133	170,0	100
Kr - 85	0,9	100
I - 131	45,0	50 - 60
Te - 132	11,0	
Cs - 134	1,2	
Cs - 137	2,3	$33 \pm 10$
Zr - 95	4,5	
Ru - 103	4,6	
Ru - 106	0,8	
Ba - 140	4,6	
Ce - 141	5,3	
Ce - 144	3,7	
Sr - 89	2,2	
Sr - 90	0,22	4
Np - 239	45,0	
Pu - 238	$8 \times 10^{-4}$	
Pu - 239	$8 \times 10^{-4}$	
Pu - 240	$1,2 \times 10^{-3}$	
Pu - 241	0,16	
Pu - 242	$2,3 \times 10^{-6}$	
Cm - 242	0,025	
Total	301,5	

\* Approximately  $(3,5 \pm 0,5)$  % of the fuel was released outside immediate vicinity of the plant.

The composition of the radionuclides released differed significantly from the composition of the reactor core because of the varying volatilities of individual elements and their compounds. The release composition also varied considerably over time due to the variations in the temperature, meteorological conditions and some other parameters during the post-accident period.

The largest radioactive particles were deposited in the "near zone", i.e. within 100 km of the reactor. Most of  $^{90}\text{Sr}$  released was also deposited in the near zone of the accident. In fact, areas with  $^{90}\text{Sr}$  surface activity levels on soil exceeding  $0,1 \text{ MBq/m}^2$  were almost entirely within the 30-km zone, and areas with  $^{90}\text{Sr}$  levels exceeding  $0,04 \text{ MBq/m}^2$  were almost entirely within the 100 km zone\*. Only a few separate sites with  $^{90}\text{Sr}$  levels in the range  $0,04$  to  $0,1 \text{ MBq/m}^2$  were found in the Bryansk hot spot, i.e. in the far zone.

Information on area contamination with plutonium isotopes is not as extensive because of difficulties in detecting these radionuclides. The only hot spot with  $^{239,240}\text{Pu}$  surface activity on soil exceeding  $4 \text{ kBq/m}^2$  was located completely within 30 km zone. In contaminated regions of Russia  $a_{239,240}$  levels ranged from  $0,07$  to  $0,7 \text{ kBq/m}^2$  and from  $0,07$  to  $0,3 \text{ kBq/m}^2$  in Bryansk and Kaluga - Tula - Orel hot spots respectively. Moreover, while  $a_{137}$  and  $a_{90}$  levels were well-correlated in the far zone, there was no correlation between  $a_{239,240}$  and  $a_{137}$  or  $a_{90}$ .

In the far zone the most important sources of radiological danger were located in so called "caesium spots". Overall, the

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\* General information about radioactive depositions is tabulated in Table II. The data presented in the form of the ratio of the surface activity of the  $i$ -th radionuclide on soil ( $a_i$ ) to the surface activity of  $^{137}\text{Cs}$  ( $a_{137}$ ). The list of radionuclides can be separated into three groups: (1) volatile (I, Te, Cs, Sb, Ag); (2) refractory nonvolatile (Y, Zr, Mo, Ce, Np, Pu, Cm), and (3) intermediate (Ru, Ba, Sr). This classification (see in details [2,3]) in large measure is consistent with the boiling temperatures of the elements or their oxides, and to some extent it gives a key to the understanding of the post-Chernobyl contamination in the far zone, including some regions of the Russian Federation.

TABLE II. GENERAL COMPOSITION OF RADIONUCLIDE DEPOSITIONS IN VARIOUS REGIONS OF THE FORMER USSR ( $a_1/a_{137}$  ON 1986-04-26) [2,3]

Radio-nuclide	Entire release	Near zone <100 km			Far zone (>100 km)		
		North	South	West	"Caesium" spots in Russia and Belorus	South of Kiev region	Northern Caucasus
Sr-89	1,0	0,7	12,0	4	0,14	0,3	1,0
Y-90	-	2,7	8,0	5	0,06	0,17	0,6
Zr-95	2,0	3,0	10,0	5	0,06	0,3	1,0
Mo-99	-	3,0	25,0	8	0,11	0,5	1,5
Ru-103	2,0	2,7	12,0	4	1,9	2,7	6,0
Ru-106	0,4	1,0	5,0	1,5	0,7	1,0	2,3
Ag-110m	-	0,01	0,01	0,005	0,008	0,01	0,01
Sb-125	-	0,02	0,1	0,05	0,05	0,1	0,1
J-131	20,0	17,0	30,0	15	10,0	1,0	3,0
Te-132	5,0	17,0	13,0	18	13,0	1,0	3,0
Cs-134	0,5	0,5	0,5	0,5	0,5	0,5	0,5
Ba-140	2,0	3,0	20,0	7,0	0,7	0,5	1,5
Ce-141	2,3	4,0	10,0	5,0	0,11	0,5	1,8
Ce-144	1,6	2,3	6,0	3,0	0,07	0,3	1,2
Np-239	20,0	7,0	140,0	25	0,6	3,0	10,0

territory of the former USSR initially contained approximately 3100 km<sup>2</sup> contaminated by <sup>137</sup>Cs with surface activity levels exceeding 1,5 MBq/m<sup>2</sup> ; 7200 km<sup>2</sup> with levels of 0,6 to 1,5 MBq/m<sup>2</sup>; 17600 km<sup>2</sup> with levels of 0,2 to 0,6 MBq/m<sup>2</sup> ; and 103000 km<sup>2</sup> with levels of 0,04 to 0,2 MBq/m<sup>2</sup> [4]\*. In Russian Federation it was contaminated approximately 55112 km<sup>2</sup> what is equal to 38,9% of contaminated territory of the former USSR (Tables III, IV and Fig.2).

\* Figure 1 presents the scheme of organization of the radiation monitoring system.

TABLE III. DIMENSIONS OF ADMINISTRATIVE TERRITORIES OF RUSSIAN FEDERATION CONTAMINATED WITH Cs-137 DUE TO CHERNOBYL ACCIDENT (MARCH 1992) [4]

Region, republic	Contaminated area, 1000 km <sup>2</sup>				% of the whole area
	1-5 Ci/km <sup>2</sup>	5-15 Ci/km <sup>2</sup>	15-40 Ci/km <sup>2</sup>	>40 Ci/km <sup>2</sup>	
Tula region	10,9	1,3	-	-	47
Orel region	10,0	0,1	-	-	40
Bryansk region	6,8	2,6	2,1	0,3	34
Kaluga region	3,6	1,4	-	-	17
Ryazan region	5,9	-	-	-	15
Belgorod region	2,2	-	-	-	8
Lipetsk region	1,8	-	-	-	8
Kursk region	1,3	-	-	-	4,4
Penza region	1,3	-	-	-	3,0
Rep. Mordovia	0,5	-	-	-	2,0
Tambov region	0,6	-	-	-	1,7
Voronezh region	0,8	-	-	-	1,5
Leningrad region	1,0	-	-	-	1,1
Ul'yanovsk region	0,2	-	-	-	0,6
Smolensk region	0,2	-	-	-	0,5
Total	47,22	5,45	2,13	0,31	~9,7

Remarks:

- (1) Some spots with Cs-137 level ~1 Ci/km<sup>2</sup> have been detected in Republic Tatarstan, Republic Mari El, Saratov region and Krasnodar region.
- (2) In other regions and republics of Russia the following levels of <sup>137</sup>Cs contamination were detected (Ci/km<sup>2</sup>): Kaliningrad reg. - up to 0,2; Rep. Kalmykiya - up to 0,2; Rep. Karelia - up to 0,3; Kostroma reg. - up to 0,2; Nizny Novgorod reg. - up to 0,8; Pskov reg. - up to 0,2; Tver reg. - up to 0,4; Stavropol reg. - up to 0,3; Rep. Udmurtiya - up to 0,4; Rep. Tchuvashiya - up to 0,2.

TABLE IV. POPULATION AND THE NUMBER OF SETTLEMENTS IN THE AREAS OF RIGID CONTROL [5]

State	Region	Number of settlements	Population, 10 <sup>3</sup> pers.		% of total
			Total	In area of rigid control	
Russian Federation	Bryansk region	247	153,6	111,8	72,7
Ukraine	Kiev reg.	28	35,8	20,8	58,0
	Zhitomir region	45	107,8	31,2	28,9
Belarus	Mogilev region	193	135,4	23,3	17,2
	Gomel region	246	339,5	85,7	25,2

Composition of radionuclides in "caesium spots" differed significantly from the composition of radioactive depositions in the near zone (Table V). The most contaminated areas in Russia are the Bryansk spot and Kaluga - Tula - Orel region.

The Bryansk spot centered 200 km<sup>2</sup> to the north-north-east of the Chernobyl (Fig.2) was formed on April 28-29 1986, as a result of rainfall. The soil surface activities of <sup>137</sup>Cs in the most highly contaminated areas in this spot were comparable to the levels in the Chernobyl region and reached 4 MBq/m<sup>2</sup> in the village Zaborye. The initial dose rates in air ranged from 3 μGy/h to 300 μGy/h; in 1991 the dose rates had fallen from 0,05 to 5 μGy/h. The predominant soil types in the Bryansk spot are turf-podzol of varying composition.

The Kaluga - Tula - Orel spot in Russia, centered approximately 500 km<sup>2</sup> north-east of the Chernobyl reactor was formed from the same radioactive cloud, that produced the Bryansk spot, as a result of rainfall on April 28-29. However, the levels of depositions in this area were lower - a<sub>137</sub> values were less than 0,6 MBq/m<sup>2</sup> (Fig.2, Table III). The initial dose rates in air ranged from 3 to 30 μGy/h. In 1991 the dose rates had fallen from 0,05 to 0,5 μGy/h. Turf-podzol sandy and sandy-le-

TABLE V. RADIONUCLIDE COMPOSITION OF DEPOSITIONS IN "CAESIUM" SPOTS IN THE FAR ZONE ( $a_i/a_{137}$  ON 1986-04-26) [ 1,2 ]

Radio-nuclide	Bryansk - Belarus spot			Tula region, Plavsk
	Bryansk region, west	Mogilev region, east	Gomel region, east	
Sr-89	0,2	0,16	0,3	0,2
Sr-90	0,02	0,01	0,03	0,03
Zr-95	0,03	0,05	0,11	0,07
Ru-103	1,7	1,7	2,0	-
Ru-106	0,5	0,5	1,4	0,5
Ag-110m	-	0,005	0,012	0,014
Sb-125	-	0,03	0,07	0,07
I-131	11,0	10,0	14,0	-
Cs-137	1,0	1,0	1,0	1,0
Ba-140	0,8	0,7	1,1	-
Ce-141	-	0,07	0,16	0,11
Ce-144	-	0,04	0,15	0,07

am soils predominate in the Kaluga region, and chernozem soils predominate in the Tula and Orel regions.

Outside these two main spots in Russia there were many areas of radioactive contamination with  $a_{137}$  levels mainly in the range 0,04 to 0,2 MBq/m<sup>2</sup> (Tables III,IV). Unfortunately, maps with isolines of  $a_{137}$  levels below 0,04 MBq/m<sup>2</sup> are not yet available. Evaluation of these areas is continuing.

At present, the most crucial sources of internal exposure of population in contaminated regions of Russia are <sup>137</sup>Cs and <sup>90</sup>Sr radionuclides (more general model is presented in Table VII). Injection of these isotopes in organism strongly depends from the properties of soil. Exposure from <sup>137</sup>Cs nuclides ranges from <5% in chernozem zone up to >50% in the regions with turf-podzol and sandy-leaf soils. Contribution of <sup>90</sup>Sr in the

far zone does not exceed 5%, and contribution of transuranium elements is assessed as approximately 1% or less.

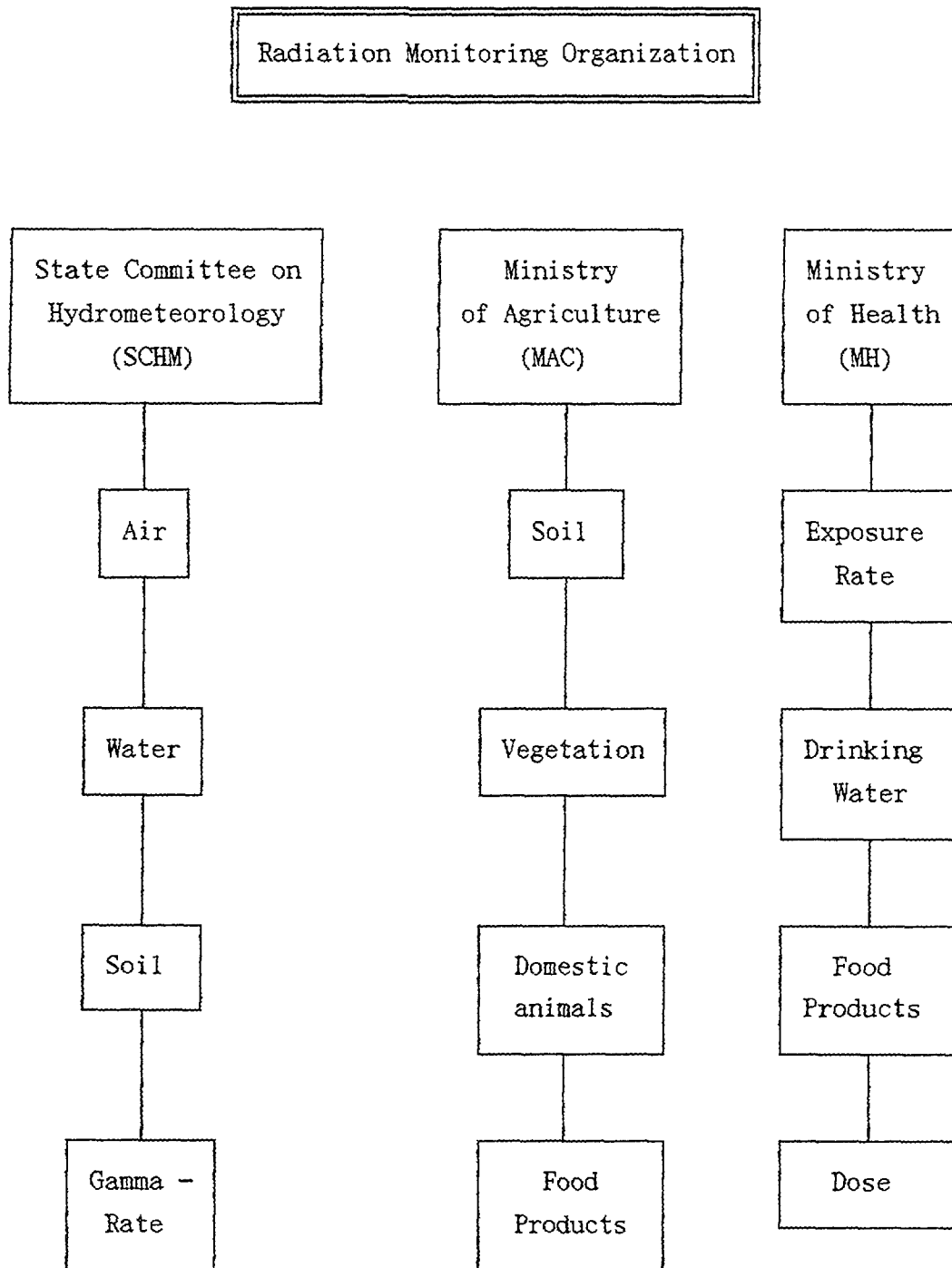


Fig. 1. Organization of the radiation monitoring system in the area contaminated following the Chernobyl accident.

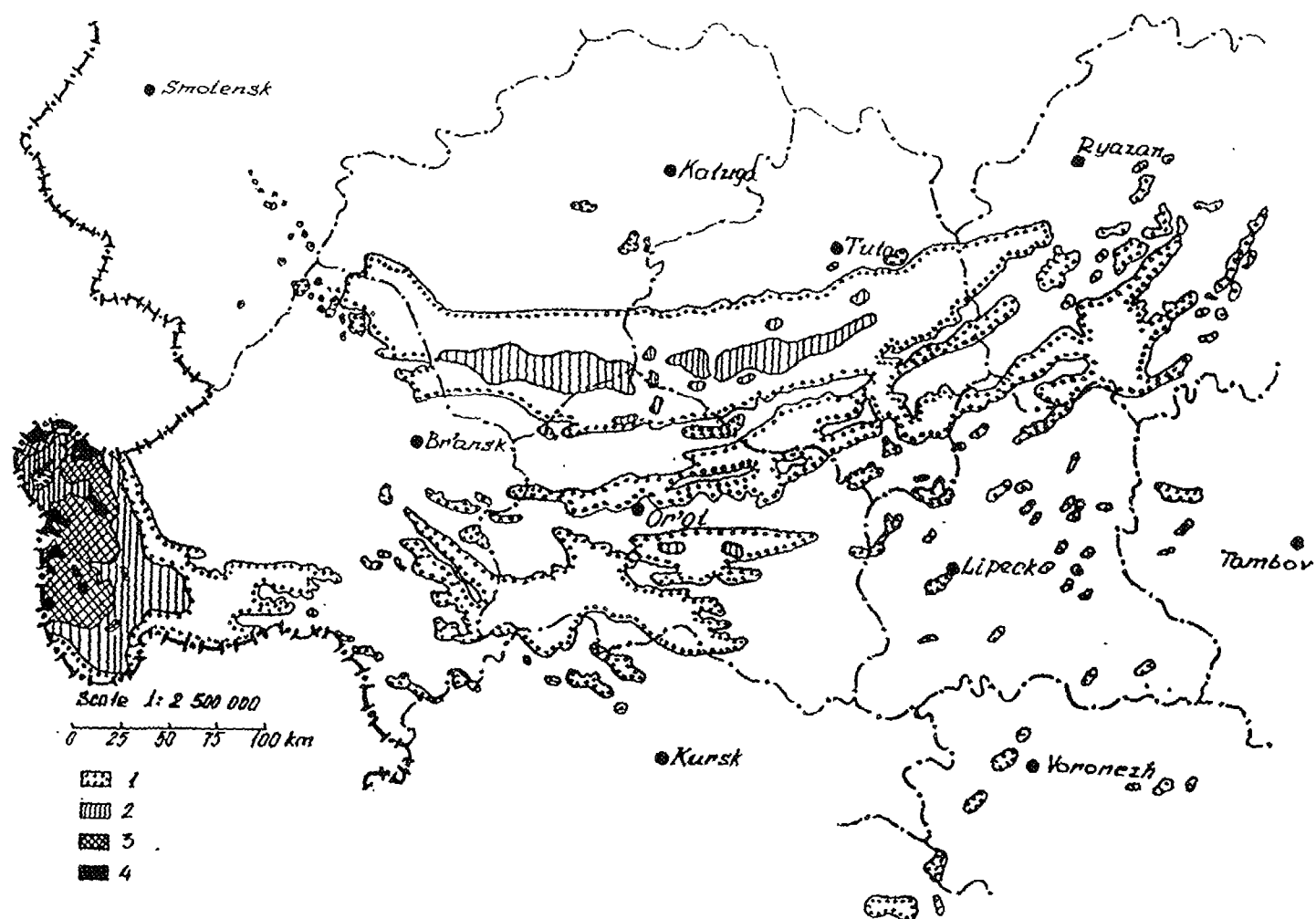


Fig.2. Distribution of surface ground radioactive contamination in the Bryansk and Kaluga- Tula- Orel regions: (1) - 1-5 Ci/km<sup>2</sup>; (2) - 5-15 Ci/km<sup>2</sup>; (3) - 15-40 Ci/km<sup>2</sup>; (4) - > 40 Ci/km<sup>2</sup>.



According to the recent estimations [7], dose of external exposure in the western regions of the Bryansk spot in 2015 will not exceed 20 mSv in average, and 40-50 mSv in the most contaminated settlements. Incidentally, it should be stressed that up to now approximately 112000 people reside in a 2400-km<sup>2</sup> controlled area of the Bryansk region with <sup>137</sup>Cs surface activity levels exceeding 0,6 MBq/m<sup>2</sup> (Table IV).

TABLE VI. DYNAMICS OF Cs-137 CONCENTRATION IN ECOSYSTEMS OF THE FAR ZONE (IN THE VICINITY OF SOSNOVYI BOR) [ 6 ]

Ecosystem component	1985	1986	1987	1988	1989	1990	Unit
Surface air	5	240	67	37	23	18	Bq/m <sup>3</sup>
Atmospheric fall-out	5	9300	166	33	32		Bq/(m <sup>3</sup> a)
Soil	43	340	124	160	110	110	Bq/kg (wet weight)
Vegetation	0,6	160 6700**	16	10	8	7	—"
Needles	5	3500 9700**	240	190	170		—"
Moss	—	5800	2170	1400	1700		—"
Fungi	40	1200 4700**	270	600	220	130	—"
Berries	5	90 220**	—	37	90		—"
Milk	0,15	17	3,9	0,7	2,0		Bq/l
Fresh water	5	60 6000**	58	54	30	10	—"
Sea water	10	1050	230	120	56		—"
Bottom sediments	1,2	41 185**	19	10	10	5	Bq/kg (wet weight)
Algae	3,9	175 2770**	30	30	24	14	—"
Sprat	1,4	54	60	25	26	36	—"
Perch	3,5	22	120	120	130		—"

TABLE VII. PRIMARY RADIONUCLIDES AND EXPOSURE MODELS AS A RESULT OF CHERNOBYL ACCIDENT [1,2]

Category of exposed persons	Time after accident, days	External exposure		Internal exposure	
		beta	gamma	Inhalation	Ingestion
Plant staff individuals who mitigated the accident	1 - 10	$^{106}\text{Ru/Rh}$ $^{144}\text{Ce/Pr}$ $^{132}\text{Te/I}$	$^{132}\text{Te/I}$ $^{131}\text{I}$ IRG*	$^{131}/^{133}\text{I}$ $^{132}\text{Te/I}$ TUE**	
	11 - 100	$^{106}\text{Ru/Rh}$ $^{144}\text{Ce/Pr}$	$^{95}\text{Zr,Nb}$ $^{106}\text{Ru/Rh}$ $^{134}, ^{137}\text{Cs}$	TUE $^{144}\text{Ce/Pr}$	
	> 100	$^{106}\text{Ru/Rh}$ $^{144}\text{Ce/Pr}$ $^{132}\text{Te/I}$	$^{132}\text{Te/I}$ $^{131}\text{I}$ IRG	$^{131}, ^{133}\text{I}$ $^{132}\text{Te/I}$ TUE	$^{131}\text{I}$ $^{132}\text{Te/I}$ $^{134}, ^{137}\text{Cs}$
Evacuated population	1 - 10	$^{106}\text{Ru/Rh}$ $^{144}\text{Ce/Pr}$ $^{132}\text{Te/I}$	$^{132}\text{Te/I}$ $^{131}\text{I}$ IRG	$^{131}, ^{133}\text{I}$ $^{132}\text{Te/I}$ TUE	$^{131}\text{I}$ $^{132}\text{Te/I}$ $^{134}, ^{137}\text{Cs}$
Remainders of population	<100	$^{106}\text{Ru/Rh}$ $^{132}\text{Te/I}$	$^{132}\text{Te/I}$ $^{131}\text{I}$ $^{134}, ^{137}\text{Cs}$	$^{131}\text{I}$ TUE	$^{131}\text{I}$ $^{134}, ^{137}\text{Cs}$ $^{90}\text{Sr}$
	>100		$^{134}, ^{137}\text{Cs}$ $^{106}\text{Ru/Rh}$	TUE $^{106}\text{Ru/Rh}$ $^{144}\text{Ce/Pr}$	$^{134}, ^{137}\text{Cs}$ $^{90}\text{Sr/Y}$

\* IRG denotes Inert Radioactive Gases.

\*\* TUE denotes Radionuclides of Transuranium Elements (Pu, Am, Cm).

## 2.2. URAL ACCIDENT

Due to a breakdown in the cooling system of a 300 m<sup>3</sup> storage facility in Kyshtym near Chelyabinsk, concrete containment, containing 70-80 tonnes of high level nitrate-acetate waste, exploded on 29 September 1957 with a force estimated at between 70 and 100 tonnes TNT. As a result of this accident approximately 2 MCi ( $7,4 \times 10^{16}$  Bq) of fission products (Table VIII) was released into the atmosphere to a height around 1000 m, forming a radioactive cloud. Fallout from this cloud, blown in a north-eastern direction from the plant, caused radioactive contamination along the path of the cloud in the Chelyabinsk, Sverdlovsk and Tyumen regions (Fig.3). This area was later referred to as the Eastern Ural Radioactive Trail (EURT). Initial distribution of contamination, contamination boundaries and surface area are reflected in Table IX, Fig.3 and Table X, respectively. During the initial period, the dose rate was around 150  $\mu$ R/h for 1 Ci ( $^{90}\text{Sr}$ )/km<sup>2</sup> with maximum values of about 0,6 R/h in the front part of the EURT.

After the accident the following measurements were undertaken:

- evacuation of the population (within 7 to 10 days 1054 peoples were evacuated, and later more than 9000 inhabitants were moved in noncontaminated regions)
- decontamination of part of territory (altogether 350000 m<sup>3</sup> of contaminated soil were removed)
- control of contamination levels of food products
- re-organization and refitting of agriculture and forest industry through creation of specialized enterprises, operating on special recommendations.

In 1967, the EURT once again was subject to accidental radioactive contamination. One of the waste pond, known as Reservoir 9 (natural lake Karachai), dried out, and around 600 Ci of radioactive dust was dispersed into environment by tornado-like wind as far as 75 kilometers off the lake [12].

TABLE VIII. CHARACTERISTICS OF RADIONUCLIDE MIXTURE COMING FROM ACCIDENTAL RELEASE [4,8,9,11]

Radio-nuclide	Part of mixture activity %	Radioactive period ( $T_{1/2}$ )	Radiation type	Nature of radiological risk
$^{89}\text{Sr}$	Traces	51 days	beta, gamma	Intern.expos.
$^{90}\text{Sr}+^{90}\text{Y}$	5,4	28,6 years	beta	(skeleton)
$^{95}\text{Zr}+^{95}\text{Nb}$	24,9	65 days	beta, gamma	gamma extern. exposure
$^{106}\text{Ru}+^{106}\text{Rh}$	3,7	1 year	beta, gamma	gamma extern. exposure
$^{137}\text{Cs}$	0,036	30 years	beta, gamma	gamma extern. & int. expos.
$^{144}\text{Ce}+^{144}\text{Pr}$	66	284 days	beta, gamma	external exposure
$^{147}\text{Pm}$	Traces	2,6 years	beta	
$^{155}\text{Eu}$	Traces	5 years	beta, gamma	
$^{239}\text{Pu}$	Traces	24380 years	alfa	

TABLE IX. INITIAL DISTRIBUTION OF RADIOACTIVE CONTAMINATION [9,11]

Average contamination, Ci/km <sup>2</sup>	Contaminated area, km <sup>2</sup>
1000 - 4000	17
100 - 1000	100
20 - 100	280
2 - 20	600
0,1 - 0,2	(15-23) x 10 <sup>3</sup>

TABLE X. DIMENSIONS OF SO CALLED EAST-URAL RADIOACTIVE TRACE (EURT) WITH MAXIMUM RADIOACTIVITY ON THE AXIS OF EURT [4,9,12]

Maximum radioactivity of soil, Ci/km <sup>2</sup>	Dimensions of contaminated territory, km <sup>2</sup>	
	length	width
140 000	1 - 2	0,5 - 1
14 000	12	1,5
1 400	10	3,5
280	65	?
28	75	?

At present radiological situation in EURT changed essentially (Table XI). Also it is useful to note that huge experience was accumulated in environmental restoration and some other related fields during extensive (but, unfortunately, secret) work in contaminated area of EURT. Apparently, this experience would be a good basis for successful implementation of the IAEA TC Regional project.

### 3. OPERATION OF NUCLEAR FACILITIES

In this chapter attention will be paid to the consequences of "bad practice" during normal (i.e. non-accidental) operation of defence-related and nuclear fuel cycle facilities.

#### 3.1. RADIO-CHEMICAL COMBINAT "MAYAK"

From 1949 until 1951, slightly radioactive water from nuclear weapon production reactors as well as high-level liquid radioactive waste, containing radionuclides of <sup>90</sup>Sr, <sup>137</sup>Cs, Nb,

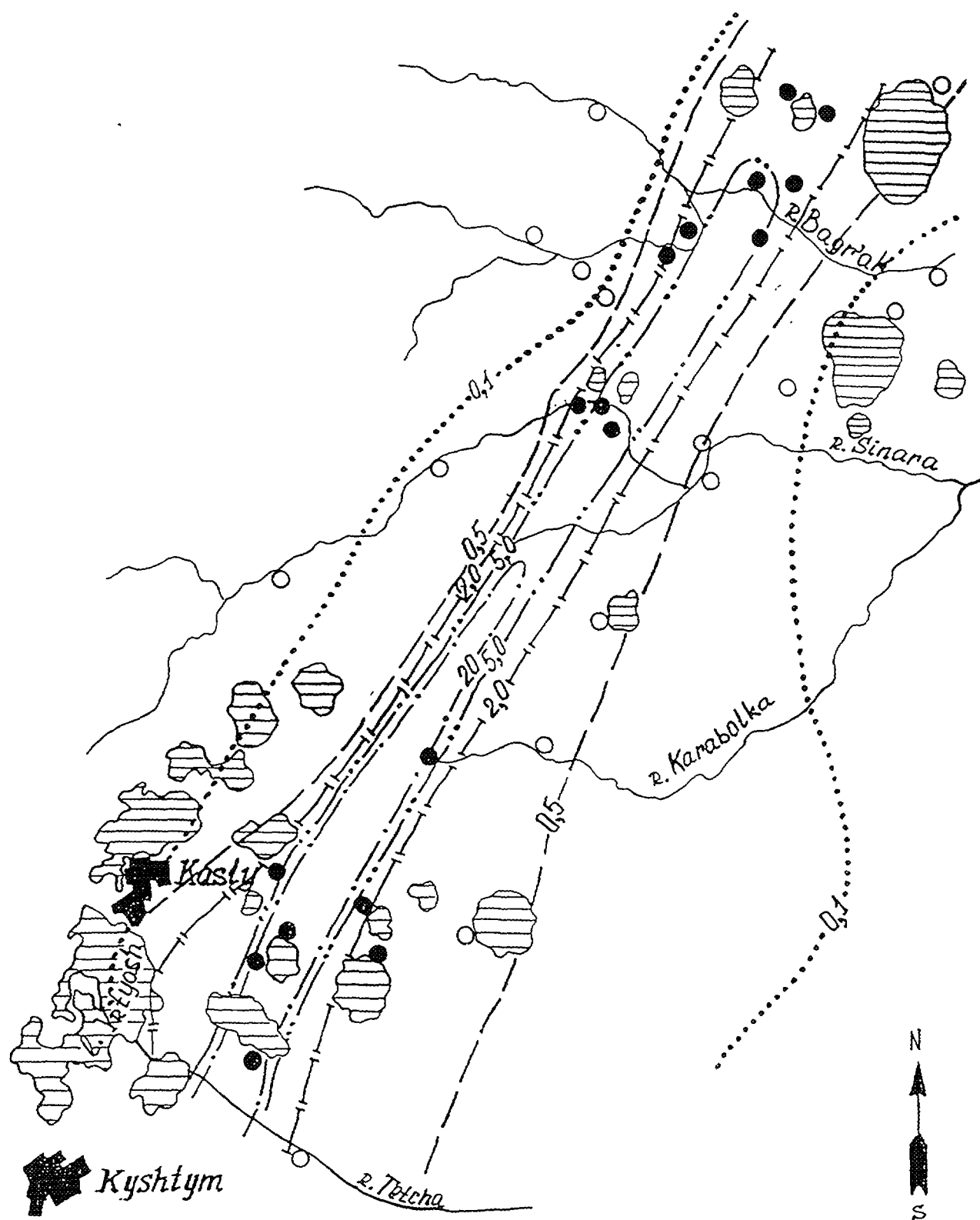


Fig.3. Schematic map of EURT with isolines of contamination

TABLE XI. ALTERATION OF RADIOLOGICAL SITUATION AFTER ACCIDENT  
[4,9,12]

Time after accident, years	Contamination, %		Conditional dose rate of gamma radiation $\mu\text{R/g}$ $\text{Ci/km}^2$	Concentration of radio-isotope, %		
	Total beta-activity	$^{90}\text{Sr}$		grass	grain	water
0	100	100	150	100	100	100
1	34	96	8,7	10	20	3
5	5,7	89	0,33	1	1	1
10	4,3	78	0,15	0,4	0,8	0,75
25	3	52	0,053	0,05	0,3	0,1
75-forecast	0,88	16	0,017	0,01	0,1	0,05

Ru, etc., were pumped directly into the lake Kyzyltash and river Tetcha. It should be stressed that at yearly days of "atomic era" such type of waste management was considered as a normal practice (and not only in the USSR). It is also important to mention that the weapon production complexes in the USSR could be characterized by the priority of production over safety and a single-minded way of reaching the set goal as quickly as possible. The total volume of liquid waste pumped in Tetcha was estimated as 75 millions  $\text{m}^3$  with total beta-activity around 2,75 MCi [12,13]. Length of the river Tetcha is 240 km<sup>2</sup>, and the whole length of the river system (Tetcha+Iset+Tobol) is equal to more than 1000 km<sup>2</sup>.

Maximum concentrations of radionuclides were accumulated in the upper Tetcha (Fig.4). In some sites the dose rate of gamma-radiation reached 5 R/h, and concentration of  $^{90}\text{Sr}$  in water was 2000-3000 times higher than permissible level.

The discharges affected 28000 local residents living in the riverside region of Tetcha (Table XII) and around 124000 people living down-stream who used the water from connected rivers (Iset, Tobol, Irtish and Ob).

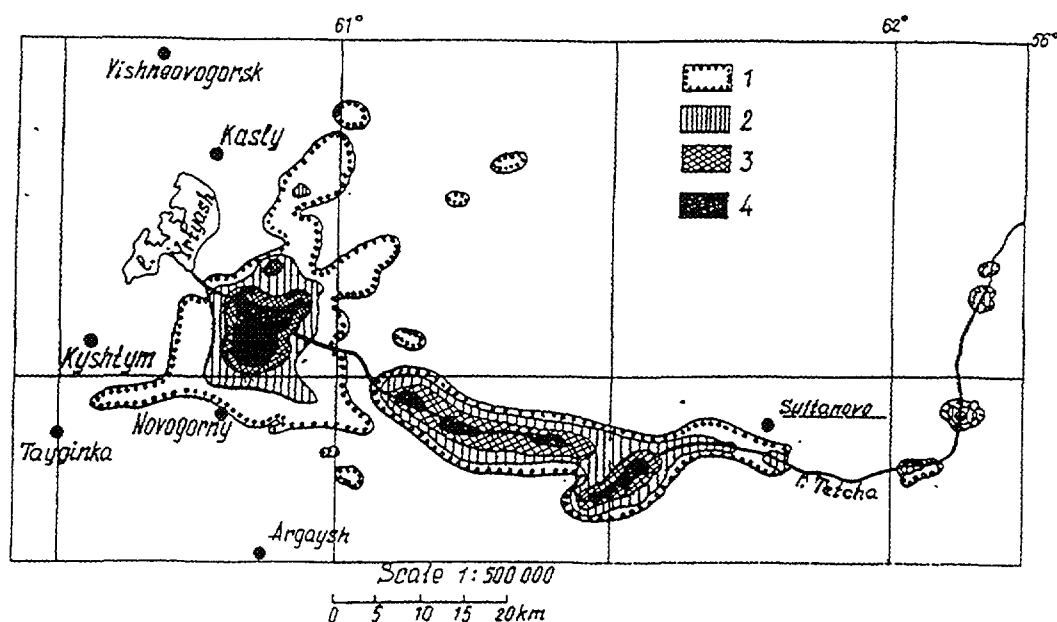


Fig.4. Contamination by Cs-137 of r.Tetcha region (1) - 1-5 Ci/km<sup>2</sup>; (2) - 5-15 Ci/km<sup>2</sup>; (3) - 15-40 Ci/km<sup>2</sup>; (4) - > 40 Ci/km<sup>2</sup>.

TABLE XII. AVERAGE EQUIVALENT DOSES AND EFFECTIVE DOSES OF IR- RADIATION IN SOME SETTLEMENTS ALONG THE r.TETCHA

Settlements	Distance from "Mayak", km	Equivalent dose of irradiation, cSv			Effective equivalent dose, cSv
		red bone marrow	bone surface	remainder	
Metlino	7	164	226	127	140
Tetcha-Brod	18	127	148	115	119
Asanovo	27	127	190	90	100
Nadyrovo	48	95	180	44	56
Muslimovo	78	61	143	12	24
Brodkalmak	109	14	31	3,3	5,8
Russian Tetcha	138	22	53	3,7	8,2
Novopetrovskoe	152	28	68	4,3	10
Shutikha	202	8	18	2,2	3,6
Zatechenskoe	237	17	40	3,2	6,6



Evacuation of population from Metlino (the nearest village to "Mayak") started at the end of 1951; towards 1957 practically all inhabitants, living in the upper Tetcha, were moved in non-contaminated regions. At present along the Tetcha there exist 19 settlements with a total population about 22000 people. However, even now, in 1990-th, radiological situation in this area can be assessed as a very intense: the dose rate in some villages reaches 0,2-0,9 mR/h.

After the year 1951 radioactive waste from "Mayak" were accumulated in special facilities and in a system of natural and constructed ponds located in a sanitary-protective zone of the Combinat (Fig.5). Concentrations of radionuclides in a storage ponds are presented in Table XIII. Today contamination in the lake Karachai is around 120 MCi, almost 2,5 times as much as the total contamination from the Chernobyl fallout. There is serious concern that the radioactive waste in the lake Karachai could lead to contamination of a regional aquifer that is a major source of drinking water. In the longer term, substantial contamination could migrate into the ocean and global environment.

### 3.2. ANOTHER NUCLEAR FACILITIES

At present the total activity of radwaste, accumulated as a result of the past operation of plutonium production plants (Chelyabinsk, Tomsk, Krasnoyarsk), is assessed as the following:

- 500 MCi in special storage tanks
- 500 MCi in open special ponds
- 200 MCi in a storage reservoirs
- 12 MCi of solid waste in shallow repositories.

Only relatively small fraction of high level waste is vitrified and stored in accordance with the modern requirements: according to different sources the quantity of vitrified HLW varying from 4 MCi to 106 MCi.

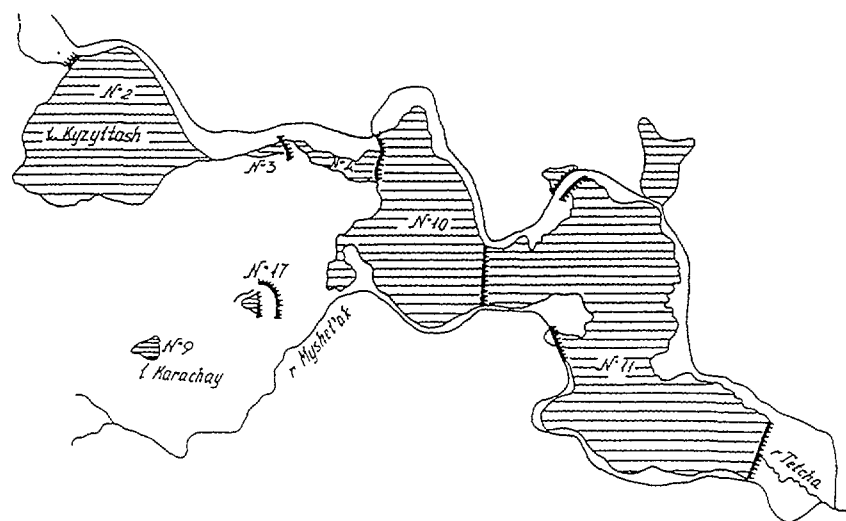


Fig.5. Scheme of storage reservoirs in sanitary-protective zone of radiochemical complex "Mayak"

TABLE XIII. CONCENTRATION OF RADIONUCLIDES IN THE PONDS OF SANITARY-PROTECTIVE ZONE OF RADIOCHEMICAL COMBINAT "MAYAK" (TOTAL AREA 30 - 40 km<sup>2</sup>) [4,12,13]

Pond			Concentration of nuclids, Ci/l				Total accumulated activity, Ci
number	area, km <sup>2</sup>	vol., 10 <sup>6</sup> m <sup>3</sup>	in water		in silt		
			Sr-90	Cs-137	Sr-90	Cs-137	
N2	19,0	83,0	1,1×10 <sup>-8</sup>	4,5×10 <sup>-9</sup>	1,3×10 <sup>-6</sup>	5,0×10 <sup>-5</sup>	2,0×10 <sup>4</sup>
N3	0,5	0,75	1,6×10 <sup>-6</sup>	2,0×10 <sup>-7</sup>	1,4×10 <sup>-4</sup>	1,0×10 <sup>-3</sup>	1,8×10 <sup>4</sup>
N4	1,3	4,1	1,7×10 <sup>-7</sup>	1,3×10 <sup>-8</sup>	4,0×10 <sup>-6</sup>	6,0×10 <sup>-5</sup>	5,9×10 <sup>3</sup>
N6	3,6	17,5	3,7×10 <sup>-10</sup>	2,0×10 <sup>-11</sup>	3,0×10 <sup>-7</sup>	3,9×10 <sup>-5</sup>	n.a.
N9	0,25	0,4	1,7×10 <sup>-3</sup>	1,2×10 <sup>-2</sup>	0,3	1,4	1,2×10 <sup>8</sup>
N10	16,6	76,0	3,5×10 <sup>-7</sup>	8,6×10 <sup>-9</sup>	3,5×10 <sup>-6</sup>	1,5×10 <sup>-1</sup>	1,1×10 <sup>5</sup>
N11	44,0	217,0	5,1×10 <sup>-8</sup>	2,0×10 <sup>-11</sup>	1,3×10 <sup>-6</sup>	1,3×10 <sup>-7</sup>	3,9×10 <sup>4</sup>
N17	0,17	0,3	7,0×10 <sup>-4</sup>	4,0×10 <sup>-6</sup>	0,12	3,3×10 <sup>-2</sup>	2,0×10 <sup>6</sup>

Uranium and thorium mining and milling facilities located in various regions of Russia (Ukhta in Komi Republic, near Norilsk, Chelyabinsk, Ekaterinenburg, near lake Baykal in Siberia, etc.) are another sources of radiological danger. Around 280000 Ci of low-level radioactive materials are accumulated at these active or obsolete sites which may pose a significant radiation health hazard to the public if mine waste and mill tailings are misused or dispersed by natural forces.

In addition, around 600000 Ci of radioactive wastes are accumulated at the factories for reactor fuel production.

In total surface areas contaminated in consequence of nuclear weapons production and the front-end nuclear fuel cycle activities are estimated as 1060 km<sup>2</sup>.

Also it should be taken into account 70000 m<sup>3</sup> of solid waste, 5350 tonnes of RBMK spent fuel, 940 tonnes of VVER-1000 spent fuel stored at nuclear power plants, and 1100 tonnes of VVER-1000 spent reactor fuel stored in centralized storage facility in Krasnoyarsk.

These sites (NPPs and storage facilities) remain a sources of potential radiation risk even after the nuclear reactors are shut down.

On the territory of Russian Federation there exist also 16 special centers for processing, conditioning and disposal of radwaste from research institutes, laboratories, hospitals, etc. Although there are no reasons to consider these centers as a sources of radiological hazard, they should be included in a special data base.

All the above quantitative information on radioactive waste arising and distribution requires careful inspection. With these purposes the "State Programme of the Russian Federation for Management of Radwaste and Spent Nuclear Materials, their Utilization and Disposal for the Period of 1992-1995 and in Perspective until 2005" (SPUD) was approved and started. This Programme stimulates, apart from other actions, creation of

centralized data base on quantity, characteristics, sources of radioactive waste; evaluation of prospective technologies for conservation of reservoirs, ponds and testing sites; rehabilitation of contaminated areas and some other measurements for the control of the situation and necessary improvements. The first results are expected already next year.

#### 4. RADIOACTIVE WASTE DUMPING/MANAGEMENT

According to the information available, (materials of President Administration published in "Izvestiya", 1993-04-01) from 1959 around 20,6 kCi of liquid radwaste and 2,3 MCi of solid waste, including 16 nuclear reactors from submarines and an icebreaker "Lenin", were dumped into the shallow waters of the Karsk Sea, and approximately 18 kCi - in the Far-East Seas. Besides, the nuclear submarine accident in the Sea of Japan on August 10, 1985 resulted in the release of 5 MCi of radioactive substances, and a radionuclide power source of 350 kCi activity was lost during transportation near Sakhalin. In the same document it was mentioned, that in 1989 a leakage of the liquid radioactive waste from spent fuel storage facility has taken place (without any official notification) in the Motovsky Bay and Litza-fiord.

It should be stressed that an average depth of the Karsk Sea, for example, does not exceed a few hundreds meters, so that, according to the London Convention, a dumping of radwaste in this sea is forbidden. However Fig.6 shows that this requirement was not met by responsible organizations (or, at least, not fully met).

The radiation situation in the Arctic and Far-East Seas due to radioactive waste dumping has not been examined in details. Today it is unclear what the long-term radiological effects will be as the corrosion by sea water may release fission products into the environment.

To clarify real situation and to evaluate radiological consequences of the radwaste dumping in the Arctic and Far-East

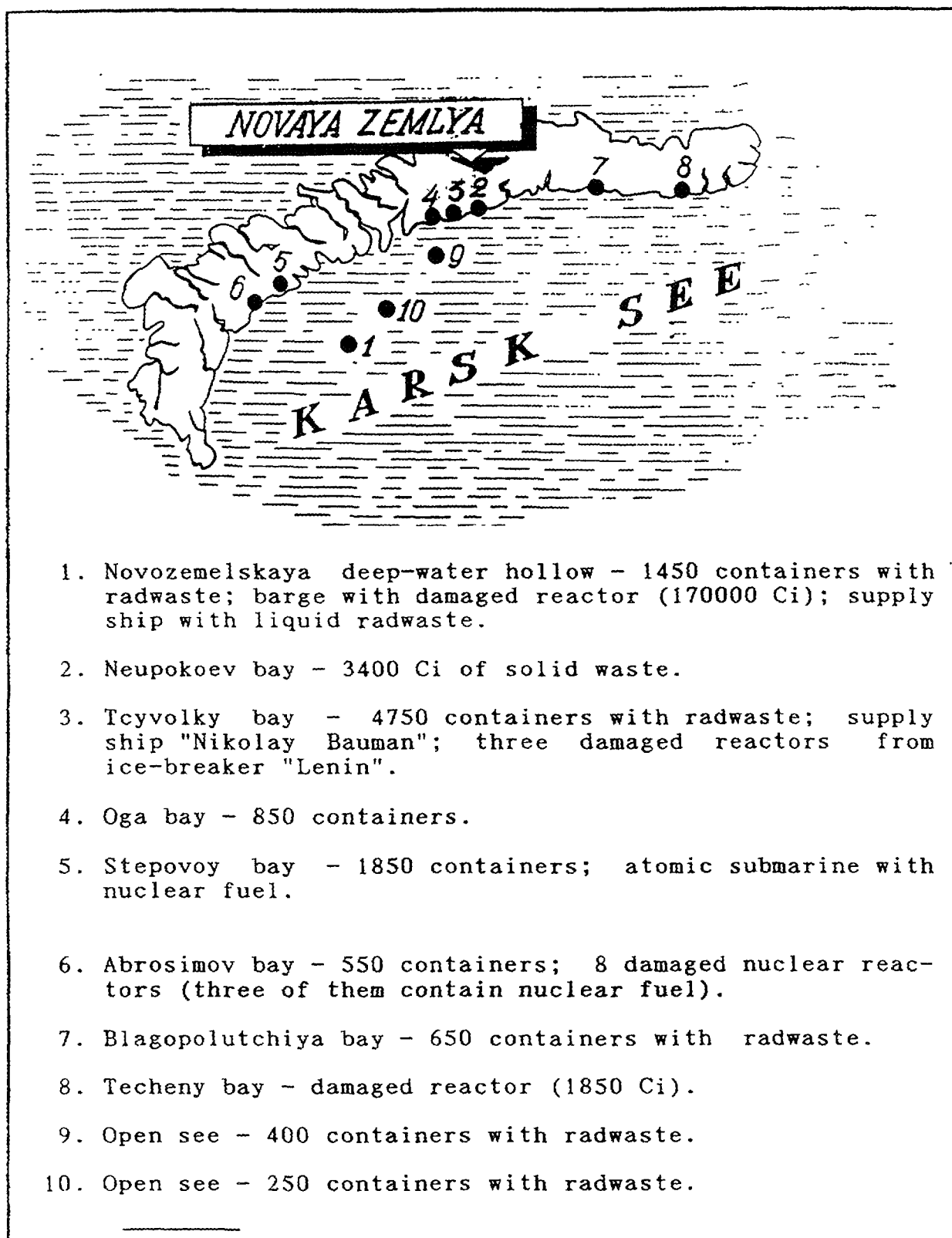


Fig. 6. Preliminary information about radwaste dumping  
in Karsk See

Seas, President of the Russian Federation set up in October 1992 special Governmental Commission, headed by Dr. Yablokov. Recently, the report of Governmental Commission, containing statistical data, was presented to the world nuclear community. However, there is much scope for further investigations on the subject and for the adequate administrative, legislative and technological measurements.

Serious problems of radiological character are also connected with the old, abandoned factories, laboratories, etc., where the modern requirements to the waste processing and conditioning have not been taken into account. As a result of this "bad practice" there are a number of sites contaminated with radionuclides in a large cities or in other populous regions. Some of such sites, unfortunately, are unknown up to now. As a example, one can mention the so called "Shkiperskaya protoka" (8000 m<sup>2</sup> almost in the down-town of St.Petersburg) contaminated with <sup>90</sup>Sr; settlement "Privetinskoye" in the resort zone of St.Petersburg contaminated with <sup>90</sup>Sr, <sup>137</sup>Cs and alfa-nuclides; territory of sports ground of Moscow Engineering and Physical Institute (27000 m<sup>2</sup> contaminated with long-lived radionuclides); scuttled ship "Kit" with radioactive substances in the lake Ladoga (was lifted and decontaminated only in 1991), etc. Discovery, careful evaluation and restoration of such contaminated sites is an item of a topmost importance.

## 5. NUCLEAR EXPLOSIONS

For the time being in Russia there remained only one experimental range for the military nuclear testing - archipelago Novaya Zemlya. From 1955 there were performed 42 underground explosions, 87 explosions in the atmosphere and 3 underwater explosions with a total power around 273 MT TNT (according to another source of information [14] - 320 MT). It represents about 80% of the cumulative power released from all soviet nuclear tests.

The whole surface area of Novaya Zemlya is equal to 81300 km<sup>2</sup>. By the gamma-spectrometry there were detected a few sco-

res of relatively small contaminated spots with the dose rates from 1 to 2 mR/h. However, nuclear tests at Novaya Zemlya did not influence seriously radiological situation in the continental regions of Russia (see for details [15,16])\* .

Some contaminated sites have been found in Loctev, Talmen and Tret'yakov regions of Altay (south-eastern part of the Western Siberia) Surface activity of  $^{90}\text{Sr}$  and  $^{137}\text{Cs}$  in these regions exceed the "natural" level of radioactivity by factors 3 to 7 [17]. This is a consequence of the nuclear explosions in the atmosphere performed in 1949-1963 at Semipalatinsk experimental range, situated in head-waters of Ishym and Irtysh rivers at the territory of Kazakhstan. Evaluation of contaminated regions in Altay is continuing.

It should also be mentioned that from 1963 until 1988 around 130 nuclear appliance have been detonated at the territory of the former USSR for so-called peaceful purposes such as construction of underground capacities, geophysical investigations, creation of underground pressure in oil and gas deposits, etc. 79 "peaceful" nuclear explosions took place in Russian Federation, including 12 explosion in Yakutia (near the Arctic Circle) one of which caused a release of radioactive materials to the atmosphere; 3 explosions in an apatite mine around 20 km east of Kirovsk on the Kola Peninsula; few explosions in the Northern Ural which left an artificial lake 400 meters wide and 600 meters long with the dose rates of 1,5 R/h on the surface and 5 R/h at the depth of 12 meters, etc. Radioactivity created by these nuclear explosions could be assessed as much as a few millions Ci. Predominant fraction of these radionuclides is incorporated in the molten rocks and disposed at a depth of 600 to 2000 meters. However, to investigate the radiation legacy

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\* The last conclusion is correct for the present time. In 1961, after the large nuclear explosion in the atmosphere, the daily radioactive precipitation in some nearest to Novaya Zemlya regions (i.e., Anderma) was 10000 times higher than that in 1988, the last year of underground testing.

that has been left by "peaceful" nuclear explosions is a matter of high priority. Careful evaluation of potentially contaminated sites is planned for the period of 1993-1995 in the framework of SPUD (see section 3.2). As a result of this work, concrete recommendations on environmental restoration have to be elaborated and realized in practice (if necessary and possibly).

## 6. OTHER CASES OF CONTAMINATION

Unfortunately, more than 70 years of practical experience in the field of radioactive materials management did not lead to the general understanding of such a simple fact that maximum attention, maximum thoroughness, maximum responsibility and rigid discipline are the only guarantees of radiological safety.

From 1979 until 1992 only in Moscow there were detected 1160 radioactively contaminated sites or radioactive devices. For instance, metallic source of radiation with a dose rate about 0,6 mR/h was discovered at the plant "Serp i Molot"; molten substance with the dose rate 1,3 R/h was detected near the garage almost in the center of Moscow; in a living house in Elektrostal it was found a source with the dose rate 2,5 R/h, etc.

Apparently, it is clear that for individuals and for the local community such incidents are, may be, more dangerous than the consequences of Chernobyl catastrophe.

It would also be irrational to overlook potential problems created by military conversion and disarmament in Russia. Specifically, the activities of  $^{90}\text{Sr}$  and  $^{137}\text{Cs}$  produced by nuclear power installations on the submarines are estimated as 1300 MCi. Today a few scores of nuclear submarines tied up at sea awaiting final disposal. Many of them still have nuclear fuel in reactors. These highly radioactive materials should be removed and safely stored/disposed. However, because of the lack of corresponding facilities, these operations are postponed with all that this implies for the environment and human media.



This is, unfortunately, not only problem created by conversion and disarmament.

Another side of the problem is connected with non-nuclear activities. Only over the last few years attention is paid to the radioactive contamination of such conventional industries as extraction of oil and gas, mining phosphates, etc. Simple analysis shows that in some cases the dose rates caused by the release of attendant natural radionuclides (mostly, by radon and radium) are compared with those at the territories contaminated in consequence of nuclear activities. For example, in Stavropol region (Oil-Gas Production Association "Stavropolneftegas") the dose rates at collector pipes reach 3 mR/hr, and levels of radiation near the bore-holes range from 240 to 600  $\mu$ R/hr. The high concentration of radium and thorium in underground water is at the bottom of this phenomenon: natural radionuclides are co-precipitated with  $\text{BaSO}_4$  on the internal surfaces of the pipes and partly on the soil around the boring wells. Keeping in mind that extractive industry is one of the most developed branch of the national economy in Russia, the necessity of radiological evaluation of all production sites is absolutely evident.

## 7. CONCLUSION

Undoubtedly, radiation situation in the Russian Federation is complicated and intense. Considerable part of the territory was contaminated with relatively long-lived radionuclides owing to unprecedented nuclear accidents; serious impact on radiological situation was exerted by past operation of defence-related and nuclear fuel cycle facilities as well as by "bad practice" of radioactive waste dumping. More than that, in many instances dumping of radioactive waste has been done without any registered information regarding the sites, quantity and characteristics of the wastes. The atmosphere of secrecy and relatively low level of "radiation culture" were also important factors influencing radiological situation in the country.

At the same time, it is evident that in Russia there is sufficient scientific and technological potential to solve these problems. Political changes provided (or better to say - are able to provide) necessary conditions for the open discussions on the subject, for the proper information of population and for the public control. A number of the projects on environmental restoration are started already or planned to be started in 1994-1995. Today the most severe problems are the lack of financial resources and, perhaps, uncertainties in organizational structure of responsible governmental bodies, the lack of co-ordination and imperfection of legislative basis.

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#### REFERENCES

- [1] Buzulukov Yu.P., Dobrynin Y.L., Release of radionuclides during the Chernobyl accident.- In: The Chernobyl Papers (Ed. Merwin S.E., Balonov M.I.), Richland: RE, 1993, Vol.1, p. 3-21.
- [2] Balonov M.I., Overview of doses to the Soviet population from the Chernobyl accident and the protective actions applied. *ibid*, p. 23-45.
- [3] Balonov M.I. Radiation factors of Chernobyl accident and man.- Proc.Conf. on Actual Problems of Retrospective, Routine and Prognostic Dosimetry of Exposure due to the Chernobyl Accident (27- -29 Oct. 1992).- Kiev: UNCRM, 1992, p. 12-22. (in Russian)

- [4] Izrael Yu.A., Nazarov I.M., Fridman Sh.D., et.al. Radiological situation on the territory of European part of CIS and Ural in 1991.- Meteorology and Hydrology, 1992, N11, p. 5-14. (in Russian)
- [5] Il'in L.A., Balonov M.I., Buldakov L.A., et.al. Ecological peculiarities and medico-biological consequences of the Chernobyl accident.- Medical radiology, 1989, N11, p. 59-81. (in Russian)
- [6] Kryshev I.I. Radioactive contamination and radioecological consequences of the Chernobyl accident.- Proc.Int.Conf. on Nuclear Accidents and the Future of Energy (April 15-17, 1991, Paris).- Paris: SFEN, 1991, p. 167-178.
- [7] Erkin V.G., Lebedev O.V. Thermoluminescent dosimeter measurements of external dose to population of the Bryansk region after the Chernobyl accident.- In: The Chernobyl Papers (Ed. Merwin S.E., Balonov M.I.), Richland: RE, 1993, Vol.1, p. 289-311.
- [8] Buldakov L.A., Demin S.N., Diboles I.K., et.al. Radiological and medical falldowns of Kyshtym accident on September 29th, 1957.- Proc.Int.Conf. on Nuclear Accidents and Future of Energy (April 15-17, 1991, Paris).- Paris: SFEN, 1991, p. 227-243.
- [9] Nikipelov B.V., Romanov G.N., Buldakov L.A., et.al. Ural nuclear accident in 1957.- Atomic Energy, 1989, Vol.67, N2, p.74-80. (in Russian)
- [10] Burnazyan A.I. Results of the Study and Acquired Experience for the Liquidation of Consequences of an Accidental Contamination of a Territory by Fission Products.- Moscow: USSR Publ.Health Dept., 1974.- 143 pp. (in Russian)
- [11] Romanov G.N., Nikipelov B.V., Drozhko E.G. The Kyshtym accident: causes, scale and characteristics.- Proc.Int.Seminar on Comparative Assessment of the Environmental Impact

of Radionuclides Released During Three Major Nuclear Accidents. Kyshtym - Windscale - Chernobyl. (CEC-IUR, IUR - 13574).- Luxemburg, 1990, p.25-40.

- [12] Kruglov A.K., Smirnov Yu.V. Nuclear Accidents, Their Consequences, and Prospects for Nuclear Power Development.- Moscow: CNIAtominform, 1992.- 116 pp. (in Russian)
- [13] Nazarov A.G., Burlakova E.B., Osanov D.P., et.al. Resonans: Conclusions of Expert Group.- Chelyabinsk: SUE, 1991.- 55 pp. (in Russian)
- [14] Polykarpov G.G., Aarkrod A. Problems of radioecology in Euroasia. Sources of radioactive contamination of environment in the former USSR.- Radiobiology, 1993, Vol.33, N1, p.15-24. (in Russian)
- [15] Chelucanov V.V., Saveliev V.A. On the radiation situation at experimental range for nuclear testing at Novaya Zemlya.- Metrology and Hydrology, 1992, N2, p.107-109. (in Russian)
- [16] Nikitin A.I., Katrich I.Yu., Kabanov A.I., et.al. Radioactive contamination of the Arctic Ocean.- Atomic Energy, 1991, Vol.71, N2, p.162-172. (in Russian)
- [17] Prokofiev O.N., Kovalenko V.I., Kolotvin V.A. Estimation of effective dose from the short-live radionuclides in some regions of Altay.- Proc.Conf. on Actual Problems of Retrospective, Routine and Prognostic Dosimetry of Exposure due to the of Chernobyl Accident (October 27-29, 1992).- Kiev, 1992, p.139-140. (in Russian)

# IDENTIFICATION AND RADIOLOGICAL CHARACTERIZATION OF CONTAMINATED SITES IN SLOVAK REPUBLIC

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## Abstract

This paper mainly deals with the methods and results of a detailed radiological survey in the vicinity of NPP Bohunice.

In the Slovak Republic, only one site has been positively identified as being contaminated with  $^{137}\text{Cs}$ . This is namely the smaller rated water canal- river system taking out the waste water from the shut down and decommissioning A1- Nuclear Power Plant of Bohunice NPP complex.

A detailed radiological survey showed that practically along the entire length of this 18 km long water way the lower parts of the grassy banks are contaminated mainly by  $^{137}\text{Cs}$  and  $^{90}\text{Sr}$  (but the  $^{90}\text{Sr}$  activity in contaminated soils is lower by two orders of magnitude).

The overall contaminated area along the polluted shores is app. 55,000 m<sup>2</sup> and the average  $^{137}\text{Cs}$  activity in the top soil layer (10 cm) is 6,100 Bq/kg. On the canal banks contaminated hot-spots were identified with a maximum  $^{137}\text{Cs}$  activity about 250,000 Bq/kg. The dose rates on the contaminated banks does not exceed the value of 1.5  $\mu\text{Sv/h}$ .

The  $^{137}\text{Cs}$  surface and depth distribution analysis results, as well as the radiological characteristics of the site, are presented and discussed in the paper.

## Introduction

There is no uranium mining in the Slovak Republic. Also there should be no problems connected with the disposal of sources from research and medical facilities. Disposal of the above mentioned sources has occurred in territory of the Czech Republic until 1993 (the former CSFR). In the presence the possibility of disposal these sources is the regional radioactive waste repository, which is under construction near the site of NPP Mochovce.

Chernobyl deposition did not represent a significant increase of contamination that special remedial actions should be taken. During cloud transition over the Slovak territory (except of the

southern districts) there was a dry fallout of the nuclides on the surface. The average activity of deposited  $^{137}\text{Cs}$  was  $3.1 \text{ kBq m}^{-2}$  as it was found after an all state monitoring in June 1986 [1]. On the limited area where heavy showers had occurred during the cloud transition the wet deposited  $^{137}\text{Cs}$  was found to be approximately ten times higher.

The systematic monitoring in forestry or on the places with higher altitude has not been carried out up to now.

Contamination by nuclear substances is topical namely in connection with the NPP Jaslovské Bohunice site surroundings. Here is located also the NPP which is called A-1 (the first Czechoslovak HWGCR NPP,  $150\text{MW}_e$ ) which was closed after two accidents in 1977. It is being decommissioned now.

In Slovakia only one site was positively identified as being contaminated by radionuclides and it is the above mentioned NPP A-1 surroundings. Contamination creates here a narrow and long spot which is localized at the banks of the Manivier Canal and the Dudváh River. Up to the end of the last year this low flow-rate surface water system transported all waste water being discharged from the NPP A-1 and from the NPP V-1 (other units on Jaslovské Bohunice site), into the larger Váh River ( $70 \text{ m}^3\text{s}^{-1}$ ).

The detailed survey which was carried out in NPP Jaslovské Bohunice surroundings has shown that this eighteen-kilometer-long water way is contaminated namely by Cs-137. Accompanying abundance of Sr-90 and Co-60 is approximately lower by two order of magnitude.

The aim of this paper is to describe the contaminated site as well as the procedure and methods used for detailed radiological survey and to summarize the results obtained concerning the identification and radiological characteristics of this contaminated site.

### The history of contamination

All the water way contamination was caused by drainage water from the contaminated NPP-A1 during heavy rains and floods in 1980. Eroded soil was contaminated by rinsing of the contaminated plant's drainage system (common special and rain drainage system) during the flood. A lot of these highly contaminated particles (high absorption of the  $^{137}\text{Cs}$  due to clay content) have been spread from this open drainage into the surface water (the Manivier Canal and the Dudváh River) having caused this water way contamination.

The parts of banks which were under water during the flood were contaminated by natural way via the settling of radioactive suspended matters from water. The contamination of the banks had been intensified later when the settled contaminated bottom sediments were transposed from the flow-bed to the flow banks during an after-flood cleanup of the canal-bed (the bottom paving) and the Dudvák River mouth.

Similar displacement of radioactivity to the banks and surrounding fields was executed when an irrigation station was built (and maintained) at the Dudvák River near the village of Bučany.

Up to 1990 the range of contamination was not assessed in the above mentioned localities. Such requirements arose after designing an engineering and construction adaptation and maintenance program of the unregulated parts of the Dudvák River. This part of the river flows through or next to the three villages. In 1991, firstly, the radiation survey of this part of the river (5-kilometer-long) was carried out as a part of this bank reconstruction project.

Later, in frame of a government supported research project [2], these works were extended to the overall 18-kilometer-long influenced water way aiming at also:

assessment of radiological impacts of the contaminated site to surrounding population, proposal of the solution concerning the released contaminated soils during the planned reconstruction, and other remedial measures concerning the overall influenced water way.

### The site characterization

The Bohunice Nuclear power station has three types of reactors. Two PWR reactor- VVER 440 V-230 models (the V1-NPP) have been operating since 1979 and 1980. Two VVER 440 V-213 models with more developed nuclear safety functions (the V2-NPP) have operating since 1982 and 1984. All four units each produce 440 MWe. There is also one HVGCR reactor with a power of 150 MWe (the A1-NPP) which was shut down in 1977 after two accidents in 1976 and 1977. It is currently being decommissioned.

At the A1-NPP liquid radioactive effluents have been considerable since above mentioned accidents due to the ongoing decommissioning, liquid radioactive waste processing, and also due to the old technological equipment. This later caused those

extraordinary water discharges with radioactivity exceeded the authorized concentration limit (37 Bq/l in environmental surface water) which were relatively frequent up to 1990. These discharges also periodically increased considerably the transportable bottom sediment's radioactivity in the canal and Dudvák River.

Since 1991 the liquid waste processing system is out of operation and together with the contaminated plant's drainage system it is under reconstruction. From the point of contamination it is important that since the end of 1992 all liquid discharges from the Bohunice-NPP complex have been leading out directly to the Váh River via a special 15-kilometer-long pipeline- see Fig.1.

The Manivier Canal is an artificial 5-kilometer-long paved canal which was built up for transporting the dumped waste water from the NPPs A1 and V1 into the Dudvák River. It starts next to the V1- NPP's fence and before merging with the Dudvák River it flows through the village Žlkovce (about 1000 inhabitants).

The affected Dudvák River section is approximately 13-kilometers-long and flows through or next to four villages with about 5000 inhabitants. The unregulated Dudvák (Di), is a natural river with no considerable engineering modification upstream of the Bučany Irrigation station.

Past the Irrigation station, the Dudvák has been modified to flood control and hydrological purposes. Here the river was straightened and widened up to the Váh River. This regulated part of river (DRi) is about 8-km-long. Levees contain both the regulated and unregulated Dudvák River. The scheme of this water system together with width profiles in the typical sections of canal (Ki) and river are shown in Fig.1a,b.

The water from this river is used for watering and in the influenced river section there are three irrigation station. The grassed banks are used by inhabitants from surrounding villages for swimming , fishing, animal fodder, etc.

#### Radiological survey, procedure and methods used

Detailed radiation monitoring within some established lines (inner and outer side of the levees) on the banks was carried out using a hand held shielded gamma detector with plastic scintillator  $\varnothing 75 \times 75$  mm (see Fig 5). The lower detectable activity by this detector has been about 1000 Bq/kg of Cs-137 with a used time constant  $\tau=10$  s.



The measurements were done with  $\tau=10$  s along a line with a step from 20 to 50 m. On the canal banks where hot spots of Cs-137 were recognized, more frequent measurements were necessary (each 10 m). Between these measurement points hand held continual monitoring with  $\tau=1$  s was applied. By this way it was possible to reveal most of the isolated  $^{137}\text{Cs}$  hot spots on the bank.

Firstly, more precise hand held scanning measurements within an established thick planary grid (2x1m) were used to determine the shape and location of the hot spots on the canal bank. The computer produced isoplethes of the measured quantity are shown in Fig.2.

As it can be seen, the various size of spots did not allow quantitatively to evaluate the detector responses. The effective mass activity of the Cs-137 in the typically contaminated zones of canal banks were estimated semiquantitatively according to the detector response data and laboratory analysis of soil samples taken from the most typical places and including the most intensive hot spots.

On the Dudváh river banks, where no hot spots were identified, more reliable evaluation of surface and mass activities of the  $^{137}\text{Cs}$  was possible. The shielded detector response data and in situ calibration measurements included the competent sampling of soils were used for this purposes. The calibration constants were finally obtained using the results of the laboratory semiconductor gammaspectrometry of sampled soils (10 cm thick top soil).

The uncertainty of the evaluation of  $^{137}\text{Cs}$  effective mass activities on the banks have been found to be about  $\pm 50\%$  due to various unhomogenieties of the source on the banks (the width of contamination, the density of soil, the depth distribution) [2].

The range of pollution on the contaminated field (near Bučany) was determined on the basis of a continual scanning using a shielded vehicle mounted NaI(Tl) detector (see Fig.5, down), coupled with a rate meter. The NaI(Tl) detector's data (total gamma counting for choosen measuring time period) was coupled with a linear positioning device to provide for automated data acquisition ( the EPSON HX-20 microcomputer in the vehicle) and computer produced isopleth mapping.

On the basis of in situ calibration measurements and competent sampling of contaminated soils the isoplethes of Cs-137 mass activity in the arable soil was evaluated for the scanned field (see Fig.1c). For used measuring time period of 5 s and existing conditions on the field ( $^{40}\text{K}$ , U, Th) the lower detectable limit of

this scanning equipment was better than 300 Bq/kg Cs-137 in arable soil.

Later, the same scanning equipment was mounted on a special tractor with a 7-meter-long hydraulic arm (a tractor for grass cutting), which have allowed the scanning (see Fig.6) and isopleth mapping (Fig 2a), also, on the steep Manivier Canal banks with bad access.

The above mentioned mobile scanning equipment was sensitive enough and is planned to be used to control the remaining contamination during and after completion of the planned remedial actions in this site ( removing of contaminated soil from the flow banks and field).

Dose rates in the air 1 m above the ground were evaluated according to the data from detailed monitoring of banks (shielded detector 30 centimeters above the ground) using the correlation coefficient between the shielded and unshielded detector response.

**Sampling and laboratory analyses.** The soils on the banks were sampled from some typically contaminated zones accordingly to the gamma detector response data. Individual soil layers, with thicknesses of 5 to 10 cm, were sampled down to the depth of 20 to 30 centimeters . On the banks, where higher activities of the soil were identified, the grass was regularly sampled for the duration of the two years of research project.

After standard laboratory processing the samples were analyzed using a semiconductor gammaspectrometer. Radiochemical determination of  $^{90}\text{Sr}$  and  $^{239,240}\text{Pu}$  was done in chosen samples, as well. Inventory of Sr-90 and Pu-239 in contaminated soils have been estimated on the basis of determined  $^{137}\text{Cs}$ -to- $^{90}\text{Sr}$ (or  $^{239,240}\text{Pu}$ ) ratio in most typical samples (see Tab.1).

To test the analytical quality of the above mentioned radionuclide determination methods, an international intercomparison measurements of real contaminated soil samples were organized. No significant systematic errors were found [4] in methods used by participants. The interlaboratory mean values of measured samples, named SOIL-1 and SOIL-2, obtained by averaging of 18 participant laboratorie results, are introduced in Tab.1.

## **Results of radiological survey and discussion**

During a detailed radiological survey, it has been recognized, that practically along the entire length of canal and influenced part of Dudváh the lower parts of the grassy banks are

contaminated mainly by  $^{137}\text{Cs}$  and  $^{90}\text{Sr}$ . But the  $^{90}\text{Sr}$  concentration in soils has on average been by about two orders of magnitude lower than that of  $^{137}\text{Cs}$ .  $^{60}\text{Co}$  and  $^{239}\text{Pu}$  were found in contaminated soils, but in not significant quantities. The  $^{137}\text{Cs}$  activities and competent  $^{137}\text{Cs}$ -to-radionuclide ratios, in sampled and analysed samples, are shown in Tab.1.

Two typical ranges of these ratios can be seen in Tab.1, the greater one with the mean value of  $^{137}\text{Cs}/^{90}\text{Sr}$  ratio around of 120

Tab.1  $^{137}\text{Cs}$  activities and  $^{137}\text{Cs}$ -to-radionuclide ratios in analyzed samples of soil and sediment, arranged by the  $^{137}\text{Cs}/^{90}\text{Sr}$  ratios

SAMPLE SITE	SECTION see Fig.1	DEPTH [cm]	Cs-137 [Bq/kg]	Cs/Co [1]	Cs/Sr [1]	Cs/Pu [1]
Brestovany-3	DR2	0-10	9594	400	178	2086
Siladice-1	DR3	0-5	11785	380	176	4533
Brestovany-2	DR2	0-10	11062	381	168	1651
Kanál-39.2b	K2	0-5	138487	386	165	8935
Siladice-1	DR3	5-10	25612	316	159	13480
Brestovany-1	DR2	0-10	15014	417	153	2195
Siladice-1	DR3	0-5	17160	301	143	8580
Siladice-1	DR3	5-10	25612	316	136	4341
Brestovany-1	DR2	10-20	14634	385	133	2076
Trakovice-5	D1	10-20	3934	151	123	2314
Kanál-25b	K2	0-5	243333	342	119	6176
Siladice-2	DR3	0-5	8151	326	115	13585
Kanál-95+10m	K2	0-10	82331	317	114	3333
Siladice-1	DR3	10-15	12842	273	109	9879
Trakovice-5	D1	0-10	2910	132	104	1265
Dudvák-4.7-70m	D3	0-5	31633	218	126	7189
Dudvák-4.8+50m	D3	0-5	13426	298	83	4795
Kanál-39.2b	K2	5-10	36734	319	80	6803
Kanál-127-39m	K2	0-10	34636	315	79	6185
Dudvák-bazant.	D3	0-5	3963	330	68	5661
SOIL-1(Field)	D3-Fi	0-10	3000	160	63	6000
AVERAGE 1				307.7	123.5	5764.9
STD.DEVIATION				81.3	35.5	3594.9
Trakovice-3	D1	10-20	12356	45	13	402
Kanál-62+22mb	K1	0-10	6122	40	34	1093
Kanál-62+22mb	K1	10-20	31112	40	54	1027
Kanál-61+30m	K1	0-10	11340	39	55	840
Kanál-62+22ma	K1	10-20	15879	39	40	760
Trakovice-3	D1	0-10	530	38	48	185
SOIL-2(Sedim.)	D3	0-10	1284	38	22	1427
Kanál-62+22ma	K1	0-10	3368	35	46	267
Bot.sedim.1991	D-aver.	0-5	711	51	-	-
Bot.sedim.1992	D-aver.	0-5	335	48	-	-
AVERAGE 2				39.3	42.7	799.9
STD.DEVIATION				2.8	11.8	446.5

note: the SOIL-1 and SOIL-2 samples data were obtained as the mean values in frame of an interlaboratory intercomparison measurements

being prevailing and being typical for the more contaminated places in the canal and in the river banks. The contamination with these ratios comes from (the old) post accidental releases from the NPP-A1. It can be also seen in Tab.1 that the  $^{137}\text{Cs}/^{90}\text{Sr}$  ratios correlate with the easier determinable  $^{137}\text{Cs}/^{60}\text{Co}$  ratios which can be used for the  $^{90}\text{Sr}$  inventory estimation, as well. The mean value of  $^{137}\text{Cs}/^{90}\text{Sr}$  ratio was conservatively assumed to be 45 for the radiological assessment carried out.

The surface contamination on the banks is not uniform. The  $^{137}\text{Cs}$  activity distribution, along the canal and Dudváh River, obtained on basis of the gamma scanning readings is shown in Fig.3,4. As it can be seen, the maximum  $^{137}\text{Cs}$  activity was found at about 20 000 Bq/kg wet w. on the river bank.

On the canal banks, except of that, small (about 1 m<sup>2</sup>) and irregularly located hot-spots have been identified and their maximum activity found in the soil by the sampling, has been about 250 000 Bq/kg dry w. of  $^{137}\text{Cs}$ . The isolated hot-spots are located, typically, about 1 to 2.5 m above the normal water level. Their density, in the higher contaminated 1.5 km-long parts of banks (K2, see Fig.1), is about 1 hot-spot to 10 m of bank length, in other 1.5 km-long parts (K1) it is about 1 spot to about 100 m.

On average, the width of contamination along the polluted canal and Dudváh banks is about 2.2 m. At one site, near the irrigation station Bučany, the levee-body and the nearby lying arable soils, a 100 m length was also found as contaminated. The  $^{137}\text{Cs}$  activity in this soil, covering about 2000 m<sup>2</sup>, and down to the depth of about 35 cm, reaches an average level of about 2500 Bq/kg.

Tab.2 Distribution of  $^{137}\text{Cs}$  activity in 10cm thick top soil layer on canal and Dudváh River banks in site of NPP Bohunice

Range of A [kBq/kg]	$l_{Ki}$ [km]	$S_{Ki}$ [ha]	Percents of cont.area
0.01- 1.0	12.7	2.92	34.9
1.0 - 5.0	11.8	2.76	31.7
5.0 -10.0	5.2	1.21	14.5
10.0-15.0	4.0	1.00	12.0
> 15.0	2.3(*)	0.58	6.9
1.0- >15.0	23.3	5.55	65.1

A - averaged activities inside of zones with a typical approximately even contamination

\* - length of hot-spots area on canal banks

Thus, the area of the contaminated soils along the polluted banks is approximately 55,000 m<sup>2</sup> and through out the entire length, the average <sup>137</sup>Cs activity in the top soil (10 cm thick) was found to be 6,100 Bq/kg (see Tab.3).

Tab.3 Data about the contaminated sections of canal's (K<sub>i</sub>) and the Dudvák River's (unregulated - D<sub>i</sub>, regulated - DR<sub>i</sub>) banks

	d <sub>i</sub> [km]	l <sub>i</sub> [km]	l <sub>Ki</sub> [km]	S <sub>Ki</sub> [ha]	A <sub>i</sub> ,Cs kBq/kg	P <sub>i</sub> [%]	D <sub>i</sub> μSv/h	D <sub>i</sub> max μSv/h
K1	0	4.0	4.0	1.00	6.7	76	0.19	0.8
K2	2.0	2.3	2.3	0.57	16.2	76	0.28	1.5
D3	9.5	2.1	1.9	0.37	4.7	76	0.21	0.5
D-Fi	10.5	0.1	0.1	0.15	2.5	30	0.28	0.4
DR3	15.0	4.6	4.2	1.05	9.6	60	0.53	1.2
Another contaminated sections (K3, D1, D2, DR1, DR2)								
	-	23.0	12.0	2.3	2.3	70	0.11	0.4
Σ		36.1	24.5	5.5	6.1	-	0.22	1.5

d<sub>i</sub> - distance from release point

l<sub>i</sub> - length of banks on the both side of flow

l<sub>Ki</sub>, S<sub>Ki</sub> - length and area (ha=10<sup>4</sup>m<sup>2</sup>) of the contaminated banks on the both side of flow where A<sub>i</sub> > 1.0 Bq/g <sup>137</sup>Cs

A<sub>i</sub> - effective area averaged <sup>137</sup>Cs activities in the top (0-10 cm) layer of soil.

P<sub>i</sub> - percent of activity in 10cm thick top soil layer from the activity in full depth profil

D<sub>i</sub>, D<sub>imax</sub> - average and maximum dose rate 1m above the surface

The top soil <sup>137</sup>Cs activity distribution analysis, based on the length averaging of gamma scanning readings (averaging for typical, approximately evenly contaminated zones), showed that about 65 % of the contaminated area (23 km of banks on both side) has a <sup>137</sup>Cs activity higher than 1,000 Bq/kg, and 19 % (6.3 km) higher than 10,000 Bq/kg (see Tab.2).

The overall radioactivity along the contaminated banks was estimated at about 110 GBq of <sup>137</sup>Cs and 1,8 GBq of <sup>90</sup>Sr. Their localization along the water flow are as follow:

53 GBq <sup>137</sup>Cs and 1,2 GBq <sup>90</sup>Sr along the Manivier Canal

15 GBq <sup>137</sup>Cs and 0.3 GBq <sup>90</sup>Sr along the unregulated Dudvák

33 GBq <sup>137</sup>Cs and 0.3 GBq <sup>90</sup>Sr along the regulated Dudvák.

Another data characterized the individual contaminated sections of banks (K<sub>i</sub>, D<sub>i</sub>, DR<sub>i</sub>- see Fig.1) and field (D-Fi) are collected in Tab.3.

## Depth distribution

The depth distribution found for the  $^{137}\text{Cs}$  rather differs depending on the location and nature of contamination. As typical for unregulated parts of Dudváh River and canal banks, including hot-spots area along the canal, the following depth distribution was found:

depth	$^{137}\text{Cs}$	depth	$^{137}\text{Cs}$
0-10 cm	76 %	15-20 cm	4 %
10-15 cm	13 %	> 20 cm	7 %

Nearer to the water level, and at not too highly contaminated sections of the regulated part of river (see Fig.3), some inverse depth distribution with maximum of  $^{137}\text{Cs}$  at the depth of 10 to 20 cm, has been identified by sampling. Probably the older and higher contaminated sediments (from the period of after the flood releases are covered by the fresh sediments settled during higher water levels).

For the higher contaminated regulated part of river, that is nearer to the estuary, a uniform depth distribution of  $^{137}\text{Cs}$ , down to the depths of 25 to 30 cm, has been found as typical.

The latter type of contamination on the bank arose, as it was mentioned in the introduction, as a result of a bad waste practice, in the past. Years ago, the contaminated bottom sediments were moved from the river-bed to the bank's lower terraces during a river cleanup and hydrology maintenance work.

The hot-spots on banks of the artificial canal arose in similar way only with higher activities, because it was nearer to the release point.

## 2.2 Radiometric and radiological data

Radiometric studies and insitu measurements showed that the gamma dose rates and competent radiological impacts are not too high. On average, the dose rates on the contaminated banks have been found about 3.2 times higher (0.22  $\mu\text{Sv/h}$  over the background) than those from the background radiation. The maximum values on individual contaminated sections have not been higher than 1.5  $\mu\text{Sv/h}$ , as it can be seen from the Tab.3.

The maximum activity of the grass on contaminated banks during the two year sampling (1991/92) was about 400 to 900 Bq/kg dry weight for  $^{137}\text{Cs}$  and about 50 Bq/kg d.w. for  $^{90}\text{Sr}$ . In case of  $^{137}\text{Cs}$ , it was recognized that these maximum levels had been connected with the wet resuspension of contaminated soil particles.

The water flowing in the canal and river, included their flow-bed, were not found significantly contaminated according to the completed survey. The  $^{137}\text{Cs}$  activity of bottom sediments had been found low enough, on the level around of 500 Bq/kg. It is permanently decreasing with the time, because of, since last year the waste waters from the NPP had been dumped through the pipe line (see Fig.1a) directly to the Váh River.

Based on a general scenario, the maximum estimated values of individual dose equivalents for a critical group of individuals who stay on the contaminated banks 400 h/y and consume of 30 kg meat and 100 l milk from small farmed live-stock fed by contaminated grass, using for calculation the IAEA recommended transfer factors [3], do not reach the values above 700  $\mu\text{Sv}/\text{year}$ .

The present radiation stage on the contaminated banks is not too harmful, but, it is not acceptable to the public and hygiene authorities. Strong public opinion has also been recognized to the disposal of contaminated soils from these sites to somewhere else in the environment.

The unregulated part of river is understood as a potential source of increased radiation risk due to the possible translocation of the contaminated soils (nearer to people) during some necessary hydrology maintenance or flow adaption work. In fact, such a type of work, in the case of the Dudváh River, is necessary, and planned for the immediate future.

As a result of these tendencies, a project of the restoration of these area, aiming at decreasing considerably the contamination on the banks and secure disposal of removed soils, has been started by the NPP in 1992. The restoration, as a part of more complex project of the adaptation of influenced unregulated Dudváh River, is considered as a multiproposal measure justified by benefits mainly in the field of hydrology utilization of this river.

The interim clean up level for contaminated soils has been set up by authorities on the level of 500 to 1000 Bq/kg  $^{137}\text{Cs}$ , the competent dose limit for these purposes being considered on very low level of 25  $\mu\text{Sv}/\text{y}$  as a fraction of the source-specific dose

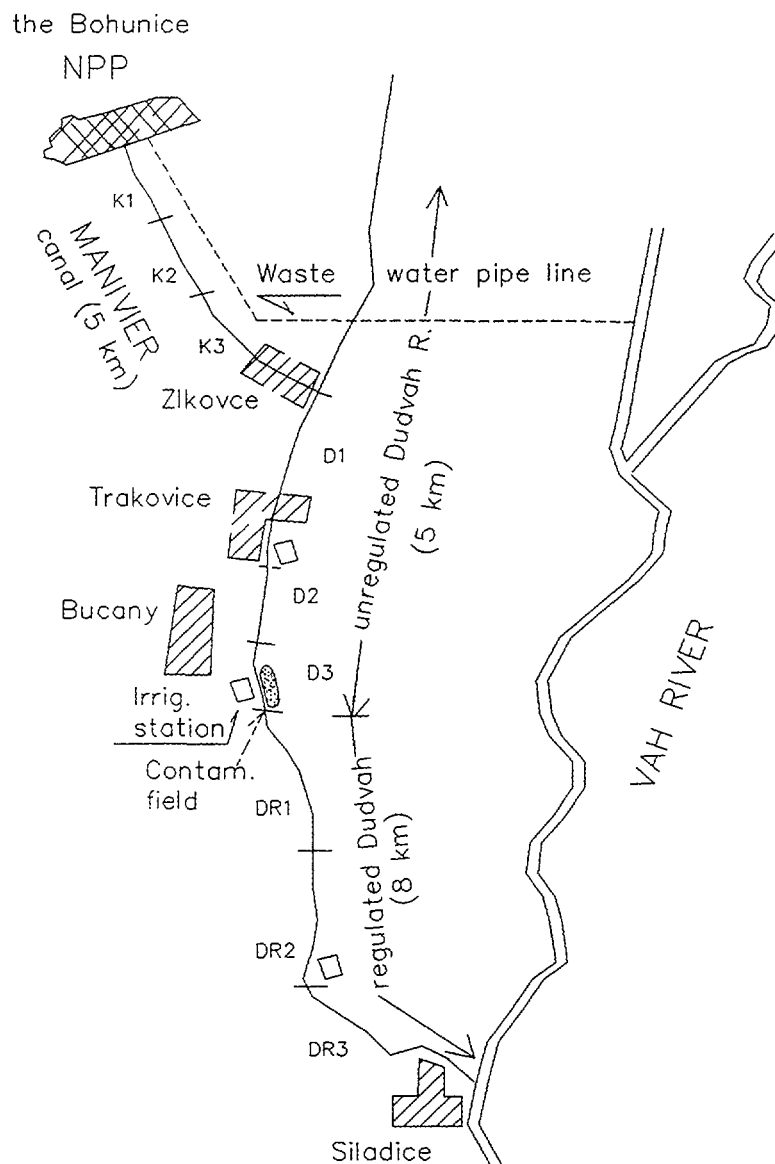


Fig. 1.a. Scheme of the water system taking out the waste water from the Bohunice NPP to the Vah River

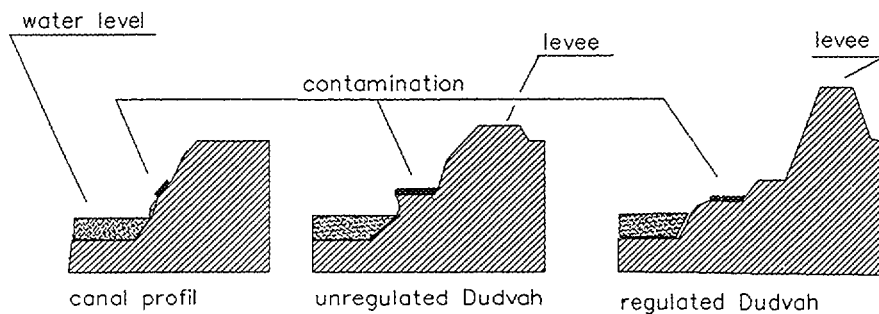
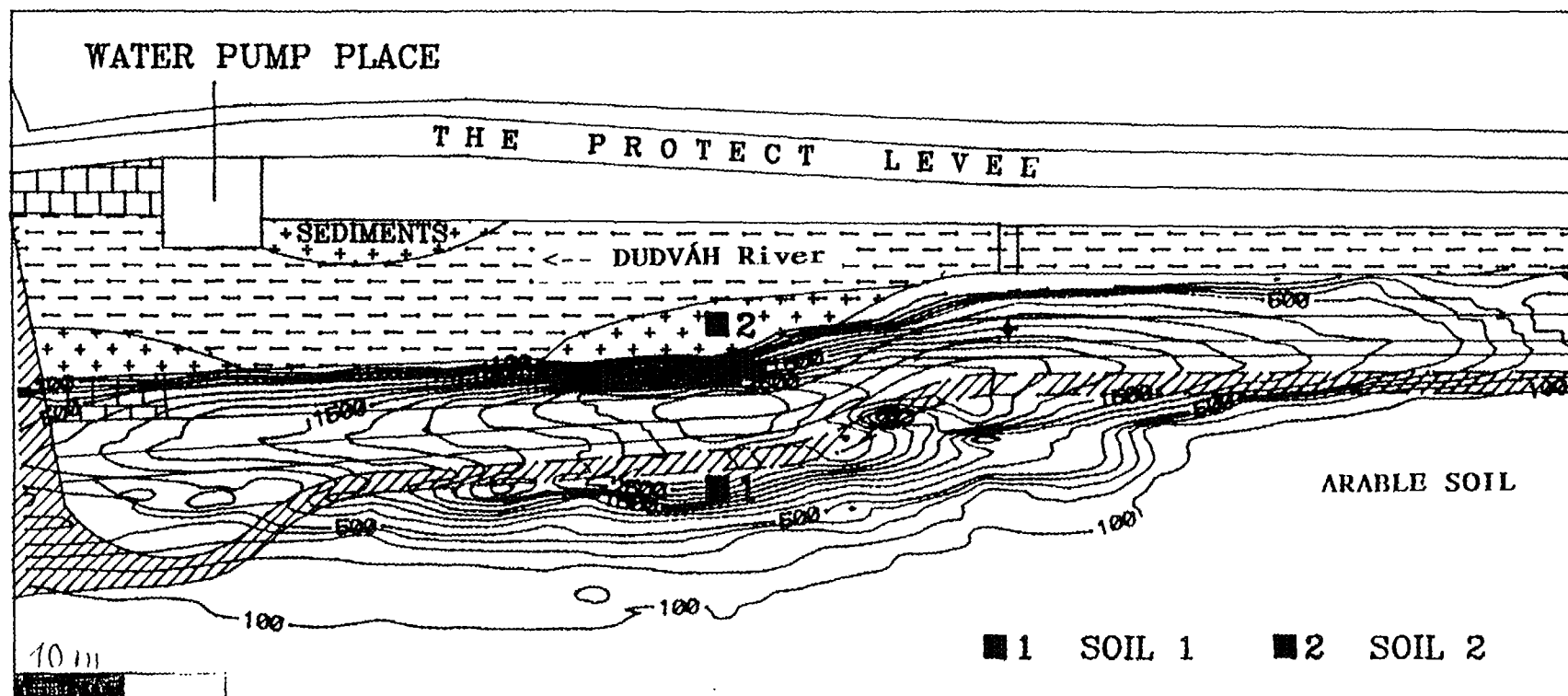


Fig. 1.b. Width profiles and the location of contamination in typical sections of flow





OBTAINED BY A VEHICEL MOUNTED NAI(TI) DETECTOR

Fig.1c Detailed top view of situation near the irrigation st. Bučany with isolines of Cs 137 mass activity in the soil on the contaminated field

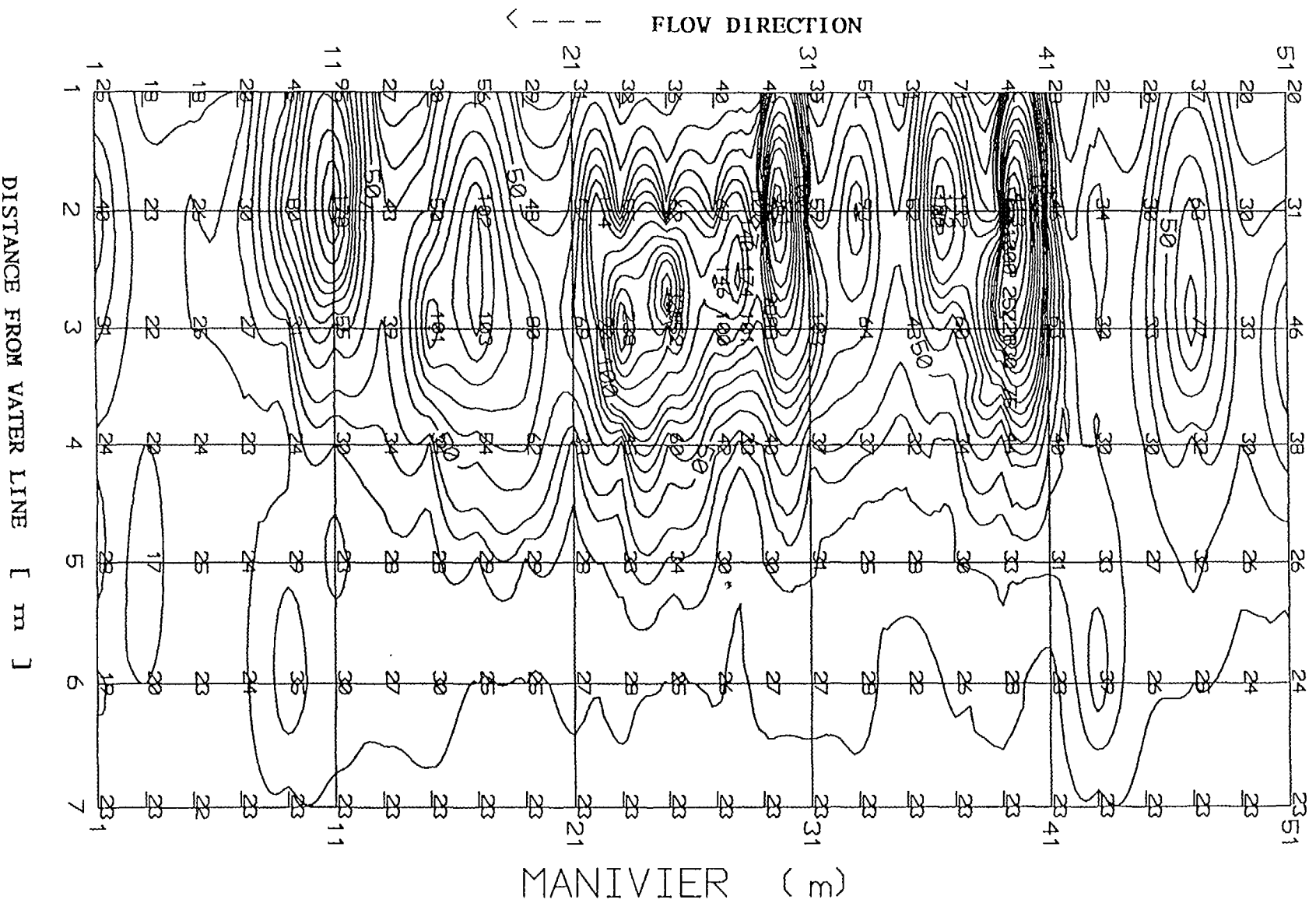


Fig.2 Distribution of Cs 137 (hot spots) in a 50 m long section of canal bank- hand held shielded gamma monitoring izolines in relative units, (2.5 km from releas point, numbers in grid means the actual monitored values)

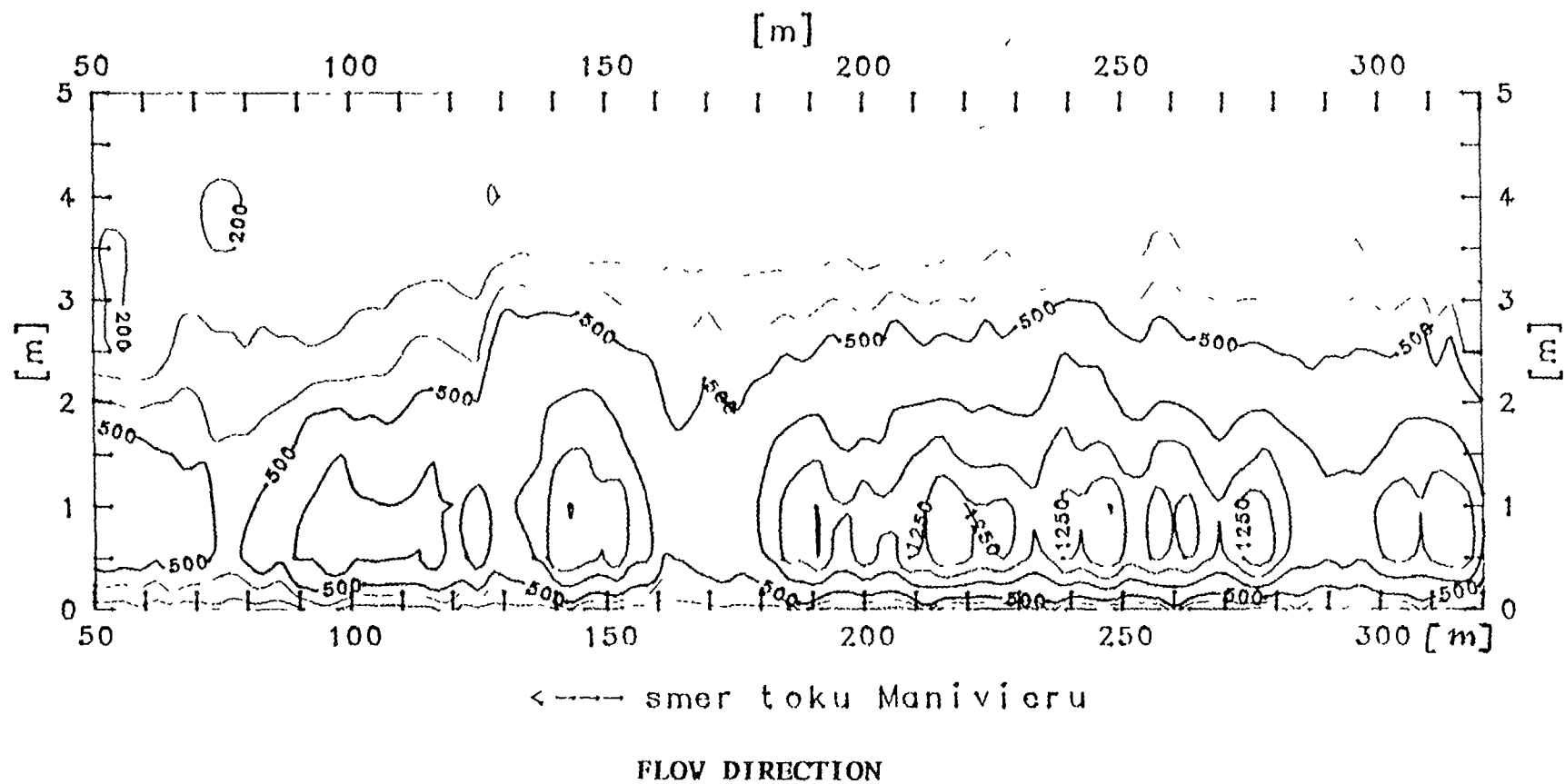


Fig.2a Distribution of Cs 137 on a section of canal bank - special tractor arm mounted NaI(Tl) monitor izolines in rel. units, 5 km from release point, right side near village Žlkovce (Fig.1)

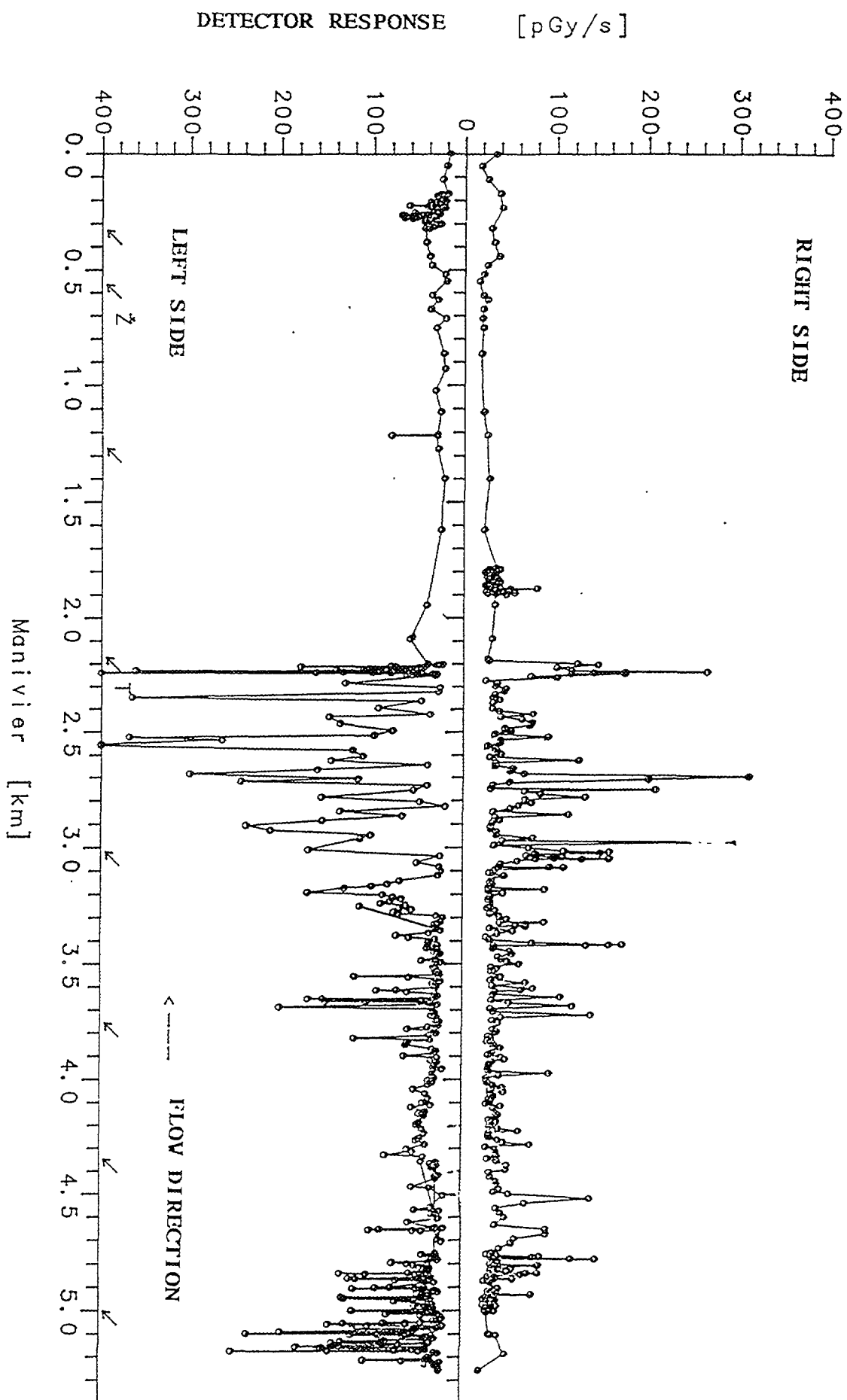


Fig.3 Canal Manivier - response distribution of a hand held shielded gamma detector along the bank of flow (1 to 2 m from water line),  
 "0 km" signifies when the Manivier joins the Dudvák River

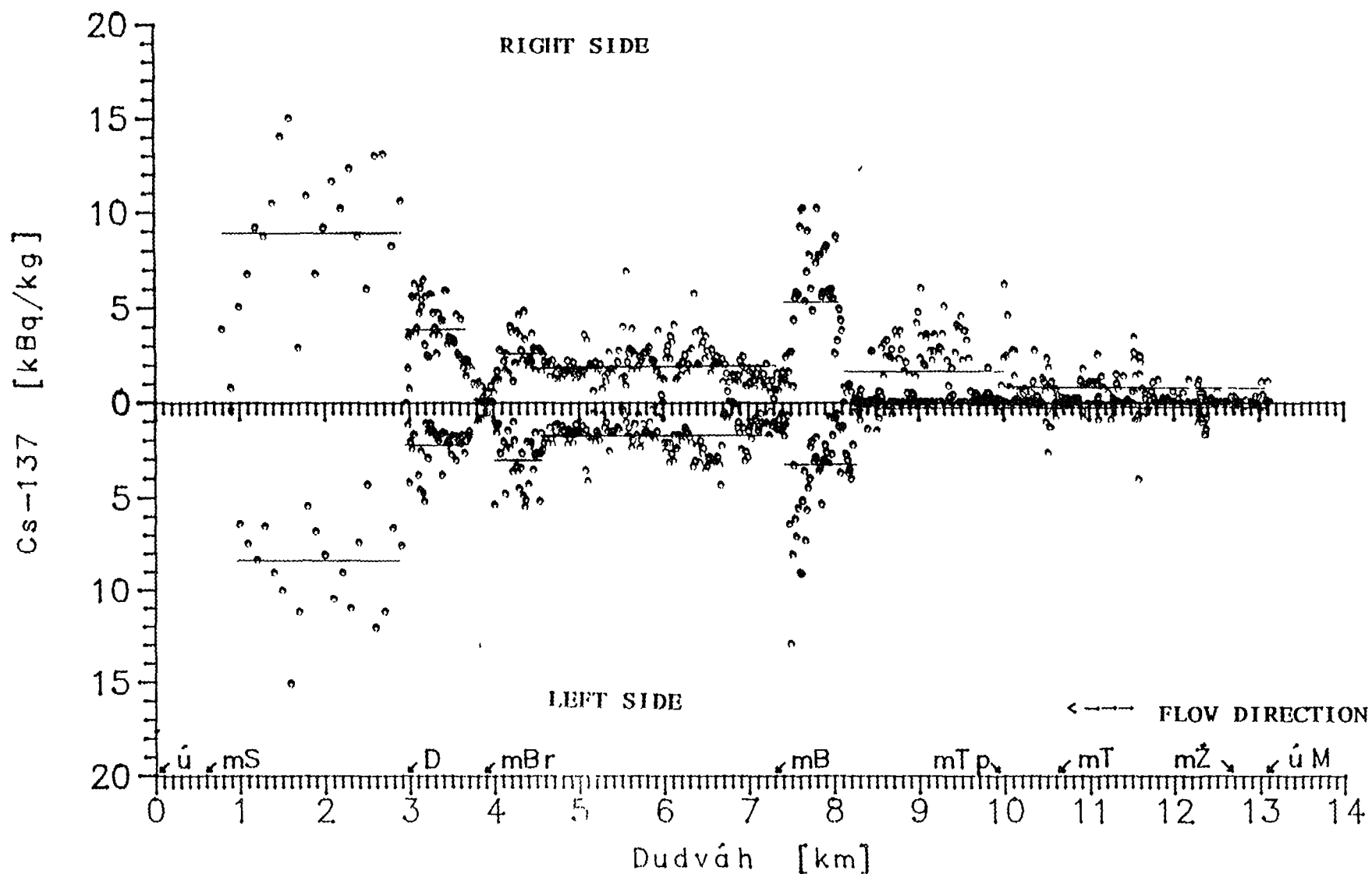


Fig.4 Dudvák-river -mass activities of Cs 137 in soils on the banks,  
obtained on the basis of hand held gamma monitoring,  
0 km- the mouth to the Váh-river

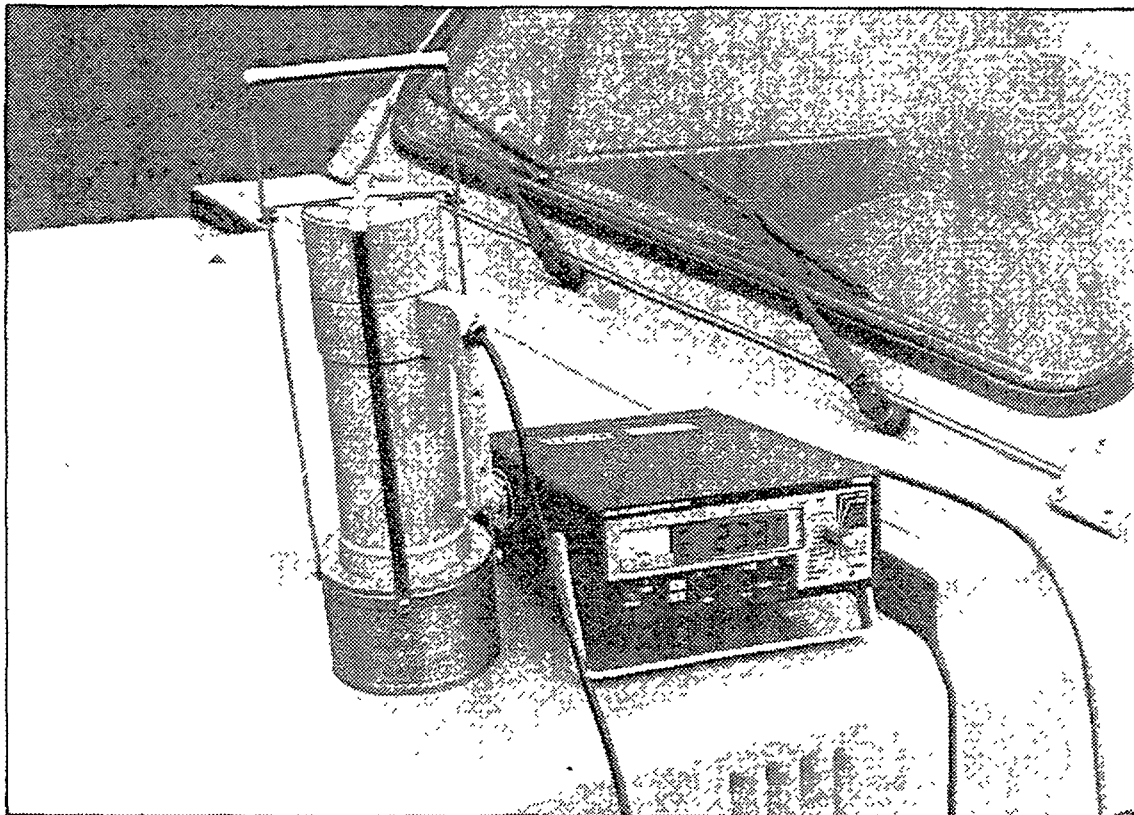


Fig.5 The hand held shielded (1 cm Pb) gamma detector (up) and  
vehicel mounted shielded (5 cm Pb) NaI(Tl) monitor (down)

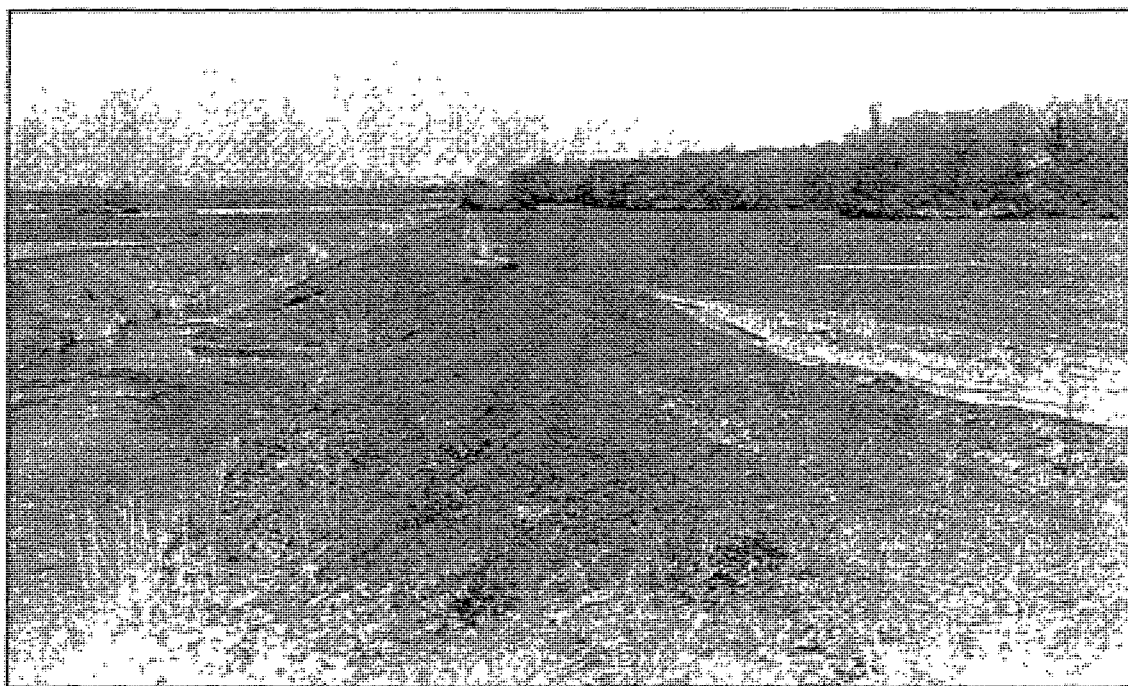


Fig.6 Hand held monitoring on canal bank (up) and the site of Cs137 contaminated field nearby the Dudváh-river bank (Bučany)

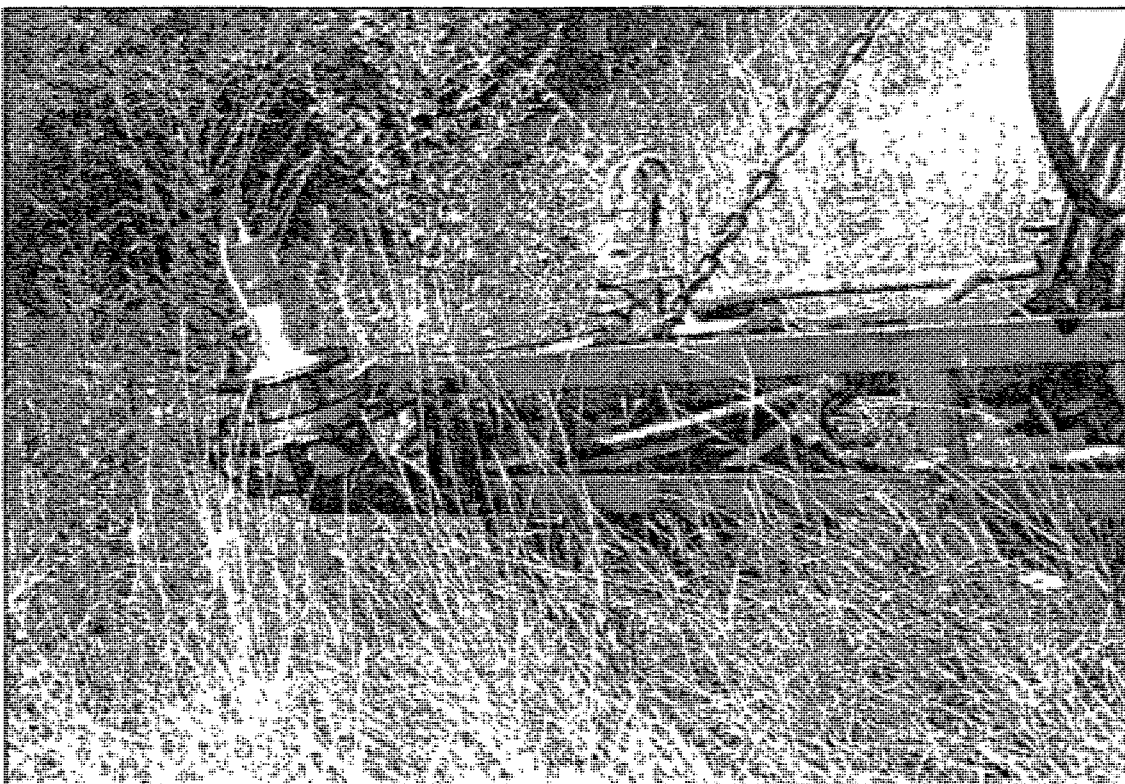
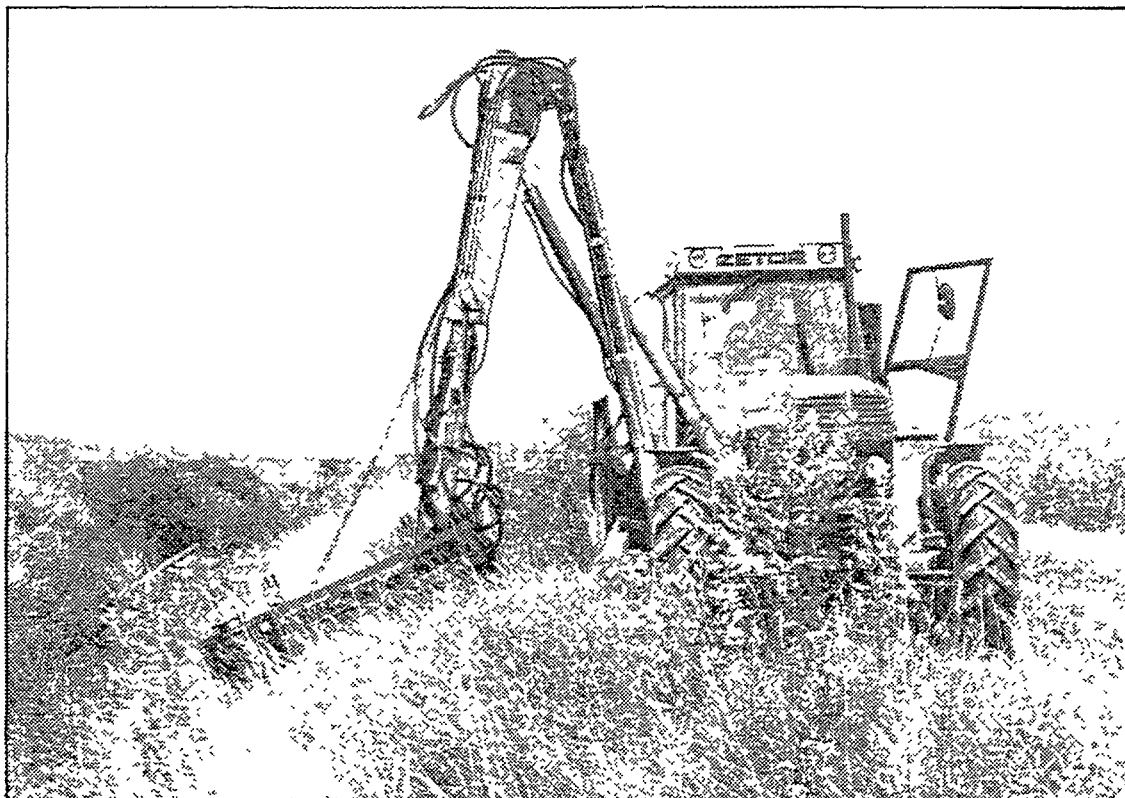


Fig.7 The special tractor arm mounted shielded (5 cm Pb) 100X100 mm NaI(Tl) monitor (up) and detailed sight to the detector during monitoring the canal bank (see the result in Fig.2a)



limit (50(liquid) + 200 (gaseous)  $\mu\text{Sv/y}$ ) for the site of the Bohunice NPP.

About 5 000  $\text{m}^3$  of contaminated soils with a inventory of about 60 GBq of  $^{137}\text{Cs}$  and 1.3 GBq of  $^{90}\text{Sr}$ , would be removed from canal and unregulated part of river banks (11 km) in the initial stage of this restoration. The removed soils will be stored in the Bohunice NPP area into a subsurface concrete basin, the building project of which being just now under final authorization.

Less expensive disposal of the contaminated soils (e.g. directly on the banks or without using concrete structure) was not accepted by the nearby population and authorities. The expenses of the complete restoration of this part of contaminated banks are covered by the NPP, and are supposed to be up to about 10,000,000 crowns (300,000 US \$).

## Conclusions

In the Slovak republic, only one site has been positively identified as being contaminated mainly with  $^{137}\text{Cs}$ . This is namely the smaller rated canal-river system which had taken out the waste water from the shut down nuclear power plant A1 of Bohunice NPP complex to the larger Váh River.

The contamination is located on the banks along the 18 km long affected water way. The contaminated area is app. 55,000  $\text{m}^2$  and the averaged  $^{137}\text{Cs}$  activity in the top 10 cm thick soil layer was found as 6100 Bq/kg. On the canal banks contaminated hot-spots were identified with maximum  $^{137}\text{Cs}$  activity about of 250,000 Bq/kg. 90 % of the activity was found in the top soil layer down to the depth of about 15 to 20 cm. The dose rate on the banks does not exceed 1.5  $\mu\text{Sv/h}$ .

About 5,000  $\text{m}^3$  of contaminated soils, which has to be removed and released during a hydrology adaptation of these banks, poses the main radiological problems. One solution, a complex restoration project for canal and unregulated parts of Dudvák banks is now going through local and central authorization.

## REFERENCES

1. Report on Radiation Situation in CSSR after Chernobyl Accident, Institute of Hygiene and Epidemiology CRH Prague, Revised edition of report to UNSCEAR, 1986

- 2.J.Morávek,O.Slávík, Evaluation of Radiological impacts of Bohunice Locality contamination, Final report No.163/92, NPP Research Inst. (VÚJE), Okružná 5, Trnava, 1992 (in Slovak)
- 3.Generic models and parameters for assesing the Environmental Transfer of Radionuclides from Routine Releases, Safety Series No.57, IAEA, Vienna,1982
- 4.J.Morávek, O.Slávík, R.Burcl, Intercomparison Measurement of Environmental Samples SOIL-1 and SOIL-2, Final Report of NPP Research Institute Trnava, February 1993 measurements of soil

# **IDENTIFICATION AND RADIOLOGICAL CHARACTERIZATION OF CONTAMINATED SITES IN SLOVENIA**

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## **Abstract**

Known radioactive contaminated sites and radioactive waste storage areas in Republic of Slovenia are identified.

The only significant site is closed Zirovski vrh Uranium Mine. Three main radioactive contamination sources are: mine waste disposal (1,5 million tons), mill tailing disposal (0,7 million tons) and run off mine water (60 - 90 m<sup>3</sup>/h).

The influence of the uranium and radium contaminated run off mine water, mine and mill tailing drainage waters on nearby water streams are shown. Affects have been monitored regularly from start up of the mine operations up to the present. Two small rivers collect all eventual radioactive and/or chemical contaminants. From radioactive contaminants point of view both streams are potable referring to the Slovene maximal permissible concentrations. Even these will be minimized in future.

There will be minor problems with the contamination in future due to small scale and short operating time of Zirovski vrh Uranium production centre. Mill waste tailing site represents serious reclamation problem because of site area earth slide. The moving mass is 7 million tons.

An effective equivalent dose contribution for population in year 1991 and 1992 was estimated to be 0,34 mSv/a, effective equivalent dose from natural sources is estimated to be 5,5 mSv/a in this area.

## **INTRODUCTION**

Due to peaceful and responsible use of radioactive materials and careful design of nuclear facilities no highly polluted areas exist in Republic of Slovenia. Known areas affected by radioactive contaminants and radiation sources are strictly monitored and controlled. Operating and active areas are monitored and controlled by operators and governmental authorized agencies. Other areas are controlled by governmental authorized agency.

Autorised agencies in Republic Slovenia are Ministry of Health - Republic Sanitary Inspectorate and Ministry of Environment and Regional Planing - Slovenian Nuclear Safety Administration.

This paper deals with all man - made radioactive sites except Nuclear power station Krsko. Locations of other very low radioactive wastes such as some coal ashes, different mercury ore wastes, phosphate fertilizer production wastes, etc. are also excluded. Even these might be considered as potential radioactive contamination sources in future.

The topic of the paper stresses closed Zirovski vrh uranium production site as the biggest radioactive materials contaminated location in the country. The decommission has started already and will last a few years.

## RADIOACTIVE MATERIALS STORAGE SITES

To the public known radioactive waste storage sites in Slovenia are (Figure 1):

### Zavratec

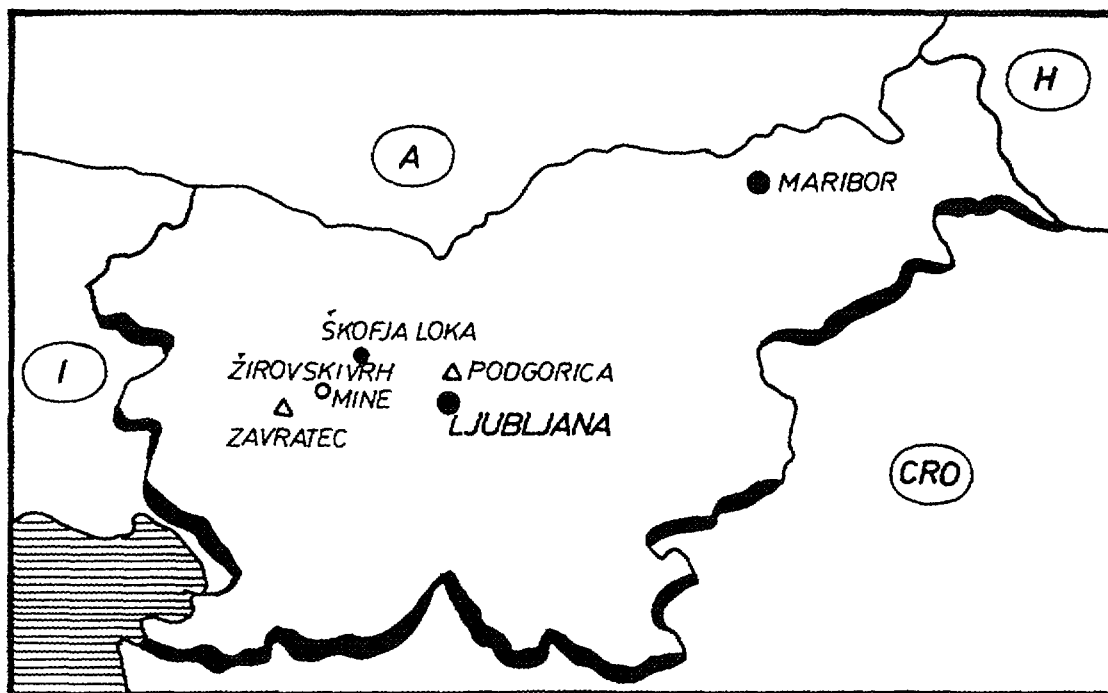
location	Zavratec, West from Ljubljana
coordinates	Y 5432800 X 5094000
altitude	700 m
land owner	private land
principal	Ministry of Environment and Regional Planing
radioactive material	by radium sulfate contaminated furniture, wood, flasks etc.
quantity	10 mg of RaSO <sub>4</sub>
gamma dose rate	0,1 - 0,2 $\mu$ Sv/h
activated	since 1961
river basin	Idrijca, Soca river (Adriatic sea)
status	closed to be relocated in near future (9)

Short description: Radioactive contaminated materials (radium) collected in metal drums from hospital accident are stored in an old previous military shelter, built of reinforced concrete (walls 0,4 m) with reinforced concrete roof plate covered over with soil material. Access to the contaminated material is not possible, the place is not protected by guard.(8)

### Reaktorski center Podgorica

location	Podgorica, North from Ljubljana
coordinates	5469250 X 5105500
altitude	280 m
owner	Institute Jozef Stefan, Ljubljana
principal	Institute Jozef Stefan, Ljubljana
radioactive material	different (lab.wastes, closed indust. sources)
quantity	30 m <sup>3</sup>
gamma dose rate(local)	0.28 - 0.4 $\mu$ Sv/h
activated	1987
river basin	Sava (Black sea)
status	operate

Short description: Different low and medium radioactive materials and sources are stored in reinforced concrete partially underground construction, covered by earth. Access is limited to the authorized personnel and well protected. This low and medium radioactive materials storage site is considered as temporary.(14)



### LEGEND

- △ RADIOACTIVE WASTE STORAGE SITE
- URANIUM MINE ŽIROVSKI VRH

*Figure 1 : RADIOLOGICAL CONTAMINATED SITES IN SLOVENIA*

### Rudnik Zirovski vrh

location	Gorenja vas, Northwest from Ljubljana
latitude	46 deg 05 min N Y 5435500
longitude	14 deg 10 min E X 5105500
owner	RS, Ministry of Environment and Regional Planing
principal	Rudnik Zirovski vrh, p.o.
river basin	Sora, Sava river (Black sea)

Figure 2. shows the whole area, where the Uranium Mine Zirovski vrh activities have been performed.

During the mine development building approximately 380.000 tons of mine waste was used for plateaus and road construction. It is estimated that approximately 30.000 tons of low grade mine waste has been used outside the mine property for the same purposes. Uranium concentrations were around 40 g U per ton (1).

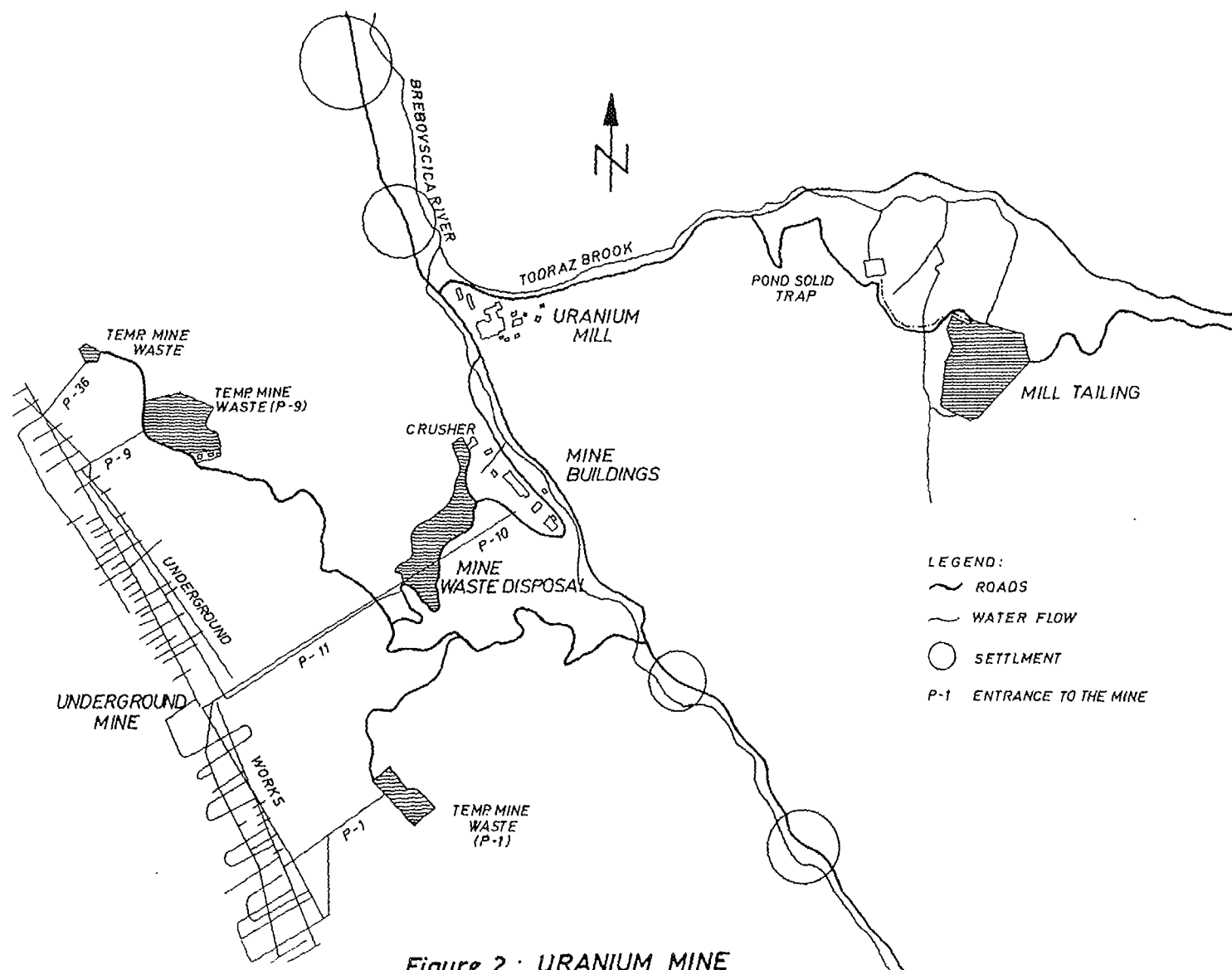


Figure 2 : URANIUM MINE  
ZIROVSKI VRH  
PRODUCTION SITE

**Mine waste disposal area**

land owner	Rudnik Zirovski vrh
altitude	450 m
radioactive material	natural uranium ore wastes, mine debris
quantity	1.5 million tons
gamma dose rate	0.4 $\mu\text{Gy/h}$
$^{222}\text{Rn}$ exhalat.rate	3 -5 $\text{Bq/m}^2\text{s}$
area	4.5 ha
activated	1980
status	closed, not decommissioned permanent

Short description: Located above ground in preengineered ravine in side valley of Brebovsica river at 450 m above sea level. Small brook flows in concrete duct under the waste disposal.

Two smaller mine waste disposals are located close to this central disposal. It is planned to relocate them in the near future.(1, 2, 3)

**Mill tailing disposal area**

land owner	Rudnik Zirovski vrh
altitude	550 m
quantity	700.000 tons
gamma dose rate	3.5 - 4 $\mu\text{Gy/h}$
$^{222}\text{Rn}$ exhalat.rate	3 -5 $\text{Bq/m}^2\text{s}$
area	3,5 ha
activated	1985
status	closed 1990, not decommissioned

Short description: Dry type, above ground, located on the hill side slope of Todrascica river at 550 m above sea level. Mill tailing sits on low permeable material and drainage rubber belts to consolidate the pore water pressure.The hole area is geomechanical unstable due to earth slide.(2, 4).

**URANIUM MINE ZIROVSKI VRH SITE RADIOLOGICAL CHARACTERIZATION****SITE AND VICINITY DESCRIPTION**

The uranium production center is located in the valley of the Brebovsica stream by the road from Gorenja vas to Lucine, 20 km Southwest of Skofja Loka. Deposit was discovered in 1979, the ore was mined from 1982 to 1990. Milling operations began in 1984 and ceased in 1990. During the operation of the mine 620.000 tons of ore were excavated and 452 tons  $\text{U}_3\text{O}_8$  were produced.

The site is situated in well inhabited farming area and surrounded by forests and meadows. Adjacent to the site (0,5 km circle) there live 200 inhabitants , 1 km away is a small community with 300 inhabitants and 2 km away is Gorenja vas with over 1000 inhabitants (10).

## URANIUM MINE

Uranium ore deposit and mine is situated in the massive of Zirovski vrh and belongs to the Ziri - Idrija tectonic region, on the west side of the Sava folds. The ore deposit lies in middle permian sediments in sheets of imbricated structure of an overthrust that overlies triassic autochthonous rocks.

Epigenetically the uranium ore is connected with clastic fluvial sediments and it is found most frequently in dark gray, medium to coarse grained sandstone. The ore deposit thickness is 150 m and lies in gray, lower Val Gardena Groeden formation. The ore bodies are of elongated lens shaped and grouped in ore belts. The known extent of ore body stretches to 15 km around the mine.

The closed uranium mine works are developed in a block approximately 2000 m long, 180 m deep and 200 m wide. The average run off mine ore grade was 0.84 kg  $U_3O_8$  per ton. Average ore body grade was 1.2 kg  $U_3O_8$  per ton and the highest values were up to 15 kg  $U_3O_8$  per ton. Table 1. shows chemical and mineralogical composition of the ore, which is the most significant to the environment after ceasing of the mining activities. Because of low sulfate (sulphide) content the acid generation potential is low.

Run off mine water general composition is given in Table 2. Uranium and radium values are elevated and especially uranium influence and contribution to Brebovscica river is noticeable. Natural water flow is from 36 to 60 m<sup>3</sup>/h. Surface water is added to the mine water through bore hole prior to the sampling and flow measuring point. Dependence of contamination from mine water flow is lured due to this fact. The differences in radioactive contaminants concentrations are distinctive in single flows inside the mine (7).

## MINE WASTE DISPOSAL AREA

Mine waste is composed mainly of mine waste material under 300 ppm  $U_3O_8$ , mill raffinate neutralisation filter cake (red mud) and some other building construction material.

Uranium mine waste chemical composition is shown in Table 3. and represents cca 1.5 million tons, average grade is estimated to be 75 ppm  $U_3O_8$ . Raffinate neutralisation filter cake (red mud) composition is shown in Table 4. and represents cca 40.000 tons (1). This material is relatively well soluble in water. Washing out must be prevented in future. Table 5. shows the influence of mine waste disposal to the mine waste disposal drainage water. This is combined from incoming brook, through waste body leakage rainfall water and some natural underground water. Uranium and radium contribution to the Brebovscica river is similar as in the case of mine water.



Detailed studies on uranium concentrations in single flows under the mine waste disposal have been done so far. The differences in uranium concentrations are very high between waste drainage flows and underground sources drainage flows (15).

**Table 1.: ZIROVSKI VRH URANIUM ORE CHEMICAL COMPOSITION**

Element	Symbol	Unit	Value
Aluminium	Al <sub>2</sub> O <sub>3</sub>	%	8.58
Calcium	CaO	%	3.93
Calium	K <sub>2</sub> O	%	1.73
Iron	Fe <sub>2</sub> O <sub>3</sub>	%	1.76
Magnesium	MgO	%	0.48
Manganeseium	MnO <sub>2</sub>	%	0.056
Sodium	Na <sub>2</sub> O	%	1.17
Quartz	SiO <sub>2</sub>	%	77.30
Sulfate	SO <sub>3</sub>	%	1.07
Titanium	TiO <sub>2</sub>	%	0.22
Carbonates	CO <sub>2</sub>	%	2.89
Water	H <sub>2</sub> O	%	0.28
Carbon org.	C	%	0.21
Arsenic	As	ppm	21
Barium	Ba	ppm	397
Bismuth	Bi	ppm	<10
Cadmium	Cd	ppm	1.8
Cobalt	Co	ppm	3.9
Copper	Cu	ppm	356
Chromium	Cr	ppm	194
Lead	Pb	ppm	1276
Mercury	Hg	ppm	0.6
Molybdenum	Mo	ppm	3.3
Nickel	Ni	ppm	52
Selene	Se	ppm	47
Silver	Ag	ppm	0.5
Strontium	Sr	ppm	103
Thorium	Th	ppm	15
Uranium	U	pm	1685
Zinc	Zn	ppm	1594
Vanadium	V	ppm	28

**Table 1 a.: MINERALS PRESENT IN THE MINE(11, 12, 13)**

**Ore minerals:**

Coffinite and other uranil silicates  
Pitchblende  
Uranil sulphate  
Uranil phosphate  
Uranil vanadate

**Other minerals:**

Quartzite  
Feldspars  
Muscovite  
Biotite  
Chlorite  
Dolomite  
Calcite  
Magnesite

**Other minerals (less than 1%):**

Pyrites  
Gallinite  
Halkopyrite  
Tenatite  
Sphalerite  
Arsenopyrite

**Table 2.: RUN OFF MINE WATER GENERAL COMPOSITION**  
**Monthly composite samples 1992**

	Flow	Ammon	Calcium	Magnesi	Iron	Chloride	Sulphat	Uranium	Radium
Month	m3/h	mg/l	mg/l	mg/l	mg/l	mg/l	mg/l	mg U/m3	Bq/m3
January	80.00	<0.1	25.00	7.50	<0.1	1.80	87.00	272.00	64.20
February	76.70	0.14	23.00	4.00	<0.1	6.20	25.60	227.00	64.00
March	70.10	<0.1	21.00	7.50	<0.1	6.20	23.10	310.00	63.20
April	90.90	<0.1	23.70	4.40	<0.1	7.00	136.00	221.00	88.50
May	97.10	<0.1	25.00	5.60	<0.1	5.30	32.00	251.00	104.40
Junij	79.30	0.14	28.00	6.40	<0.1	15.00	40.50	198.00	83.20
July	69.90	<0.1	30.00	7.60	<0.1	10.60	33.00	276.00	69.40
August	60.30	0.16	31.00	6.90	0.10	12.20	29.00	266.00	60.50
September	57.20	<0.1	27.00	7.30	<0.1	4.20	25.50	422.00	52.50
October	69.20	<0.1	24.00	5.20	<0.1	1.70	103.00	282.00	82.20
November	103.70	<0.1	26.00	4.20	<0.1	1.40	27.00	249.00	65.70
December	118.20	<0.1	24.00	4.20	<0.1	2.80	24.20	283.00	70.50
Average	81.05	<0.1	25.64	5.90	<0.1	6.20	48.83	258.55	72.36

**Table 3.: MINE WASTE SAMPLE ANALYZE**

Element	Symbol	Unit	Value
Aluminum	Al	%	6.8
Calcium	Ca	%	2.2
Magnesium	Mg	%	0.65
Iron	Fe	%	1.5
Quartz	SiO <sub>2</sub>	%	78.3
Sulfate	SO <sub>4</sub>	%	0.06
Sulfate tot.	SO <sub>4</sub>	%	0.26
Lime	CaCO <sub>3</sub>	%	5.3
Magnesium carb.	MgCO <sub>3</sub>	%	0.3
Carbonates	CO <sub>2</sub>	%	2.5
Cadmium	Cd	ppm	<5
Copper	Cu	ppm	44
Lead	Pb	ppm	105
Manganese	Mn	ppm	393
Uranium	U <sub>3</sub> O <sub>8</sub>	ppm	44
Zinc	Zn	ppm	84

**Table 4.: RED MUD CHEMICAL COMPOSITION**

Weekly composite (12.04.1990)

Element	Symbol	Unit	Value
Aluminium	Al	%	2.3
Calcium	Ca	%	17.6
Calium	K	%	0.14
Magnesium	Mg	%	2.00
Sodium	Na	%	0.47
Iron	Fe	%	5.5
Quartz	SiO <sub>2</sub>	%	5.4
Sulfate	SO <sub>4</sub>	%	48.0
Lime	CaCO <sub>3</sub>	%	6.3
Magnesium carb.	MgCO <sub>3</sub>	%	0.5
Carbonates	CO <sub>2</sub>	%	3.0
Arsenic	As	ppm	1010
Cadmium	Cd	ppm	10
Cobalt	Co	ppm	200
Copper	Cu	ppm	910
Chromium	Cr	ppm	210
Lead	Pb	ppm	470
Manganese	Mn	ppm	6110
Molybdenum	Mo	ppm	<10
Nickel	Ni	ppm	117
Phosphate	P <sub>2</sub> O <sub>5</sub>	ppm	8340
Strontium	Sr	ppm	517
Uranium	U <sub>3</sub> O <sub>8</sub>	ppm	25
Zinc	Zn	ppm	1430
Vanadium	V	ppm	71

**Table 5.: MINE WASTE DISPOSAL DRAINAGE WATER  
GENERAL COMPOSITION  
Monthly composite samples 1992**

	Flow	Ammon	Calcium	Magnesi	Iron	Chloride	Sulphat	Uranium	Radium
Month	m3/h	mg/l	mg/l	mg/l	mg/l	mg/l	mg/l	mg U/m3	Bq/m3
January	11.00	<0.1	85.00	31.00	<0.1	38.00	162.00	374.00	20.30
February	19.60	0.16	61.00	20.00	<0.1	35.00	146.00	262.00	13.30
March	35.50	<0.1	48.00	22.00	<0.1	29.00	134.00	262.00	20.90
April	78.50	<0.1	67.00	29.00	<0.1	26.00	187.00	256.00	31.60
May	37.00	<0.1	75.00	29.00	<0.1	43.00	194.00	328.00	57.10
Junij	26.20	0.10	100.00	33.00	<0.1	36.90	220.00	412.00	30.30
July	12.60	<0.1	116.00	42.00	<0.1	55.00	297.00	589.00	29.20
August	20.00	0.23	171.00	61.00	<0.1	84.00	446.00	970.00	44.90
September	6.80	<0.1	162.00	55.00	<0.1	76.70	404.00	1111.00	34.20
October	311.90	<0.1	111.00	40.00	<0.1	39.20	330.00	656.00	89.50
November	99.60	<0.1	65.00	24.00	<0.1	24.00	208.00	315.00	22.60
December	69.60	<0.1	77.00	28.00	<0.1	21.90	216.00	285.00	23.40
Average	60.69	<0.1	94.83	34.50	<0.1	42.39	245.33	485.00	34.78

## MILL TAILING DISPOSAL AREA

Mill tailing is located on the hill slope. The site has been prepared by earth works, natural underground water drainage piping and felt - rubber drainage belts. The bottom is sealed by low permeable autochthonous earth. Figure 3. shows schematically the mill tailing site crossection.

Mill tailing chemical composition is given in Table 6, average uranium content is estimated to be app. 80 ppm. Some chemical compounds are well soluble (sodium, ammonia and calcium, magnesium compounds) other less. Wash out by rainfall is evident because the tailing surface is not covered yet. Part of these is as surface water, which will not be important in the future. The chemicals and soluble materials are being leached very fast from the surface now. Second part penetrates through tailing body and affects mill tailing underground drainage water as it is evident from Table 7. Mill tailing drainage water has no important influence on Brebovsica river from uranium point of view. But chemical pollutants concentrations are high. Radium contributions are more important.

## CONCLUSIONS

The contributions of different previously mentioned radioactivity sources to the Brebovsica river are shown in Table 8. Influence of mill tailing drainage water is negligible from uranium contamination point of view. Other two sources seem equal.

Radium contribution is bigger by mill tailing drainage water than by other sources. Radioactive pollutants contribution is given in Table 9.

It is clearly evident from Figure 4. that mine water is not chemically contaminated. Other two, mine waste drainage water and particularly mill tailing drainage water contain calcium and magnesium sulphates.

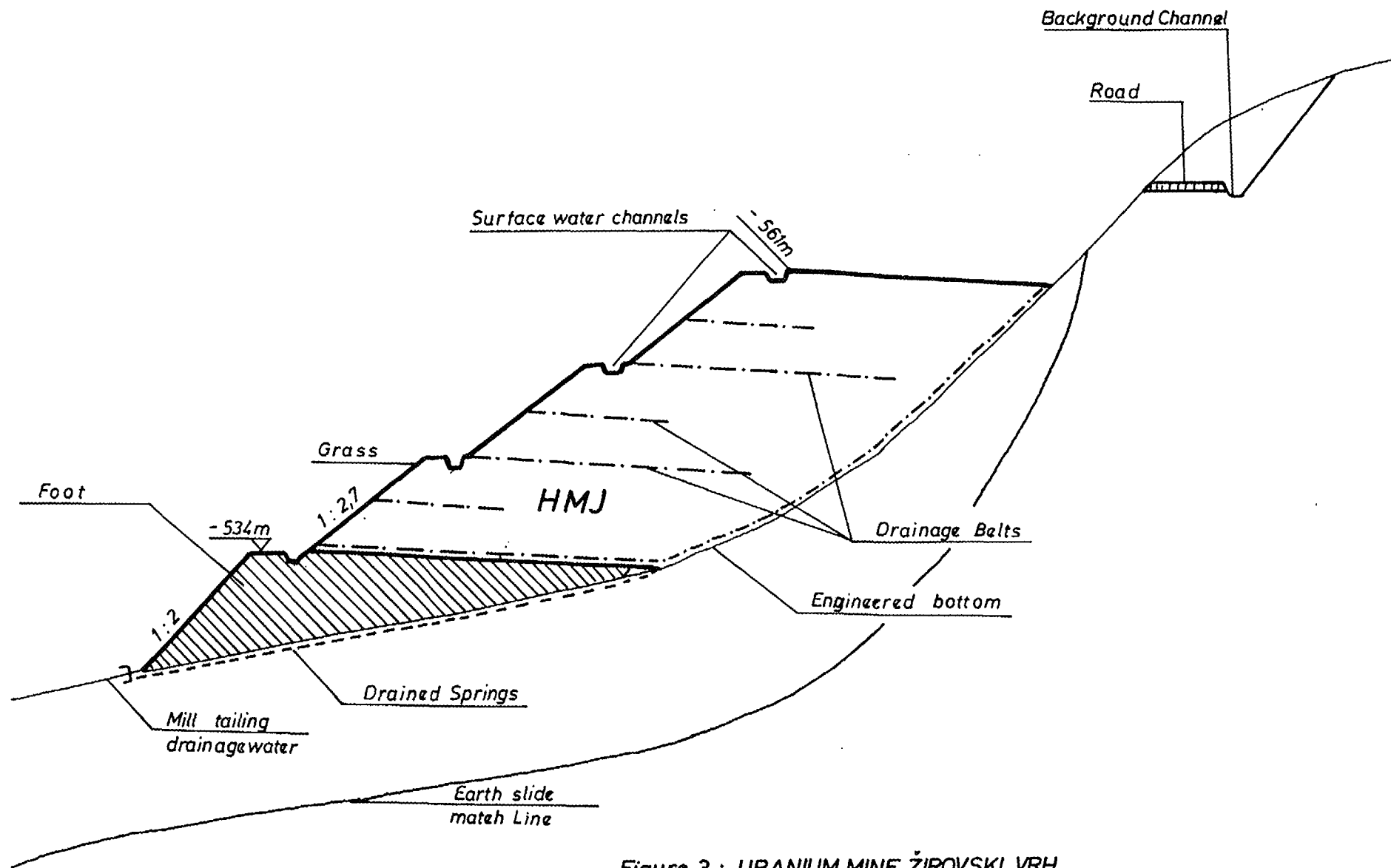


Figure 3 : URANIUM MINE ŽIROVSKI VRH  
MILL TAILING SCHEMATICAL  
CROSSECTION

**Table 6.: MILL TAILING CHEMICAL COMPOSITION**

Element	Symbol	Unit	Value
Aluminium	Al	%	5.5
Calcium	Ca	%	3.6
Calium	K	%	0.6
Magnesium	Mg	%	0.41
Sodium	Na	%	1.2
Iron	Fe	%	1.2
Quartz	SiO <sub>2</sub>	%	62.50
Sulfate	SO <sub>4</sub>	%	5.7
Arsenic	As	ppm	153
Cadmium	Cd	ppm	19
Cobalt	Co	ppm	30
Copper	Cu	ppm	98
Chromium	Cr	ppm	22
Lead	Pb	ppm	970
Manganese	Mn	ppm	842
Molybdenum	Mo	ppm	<10
Nickel	Ni	ppm	11
Phosphate	P <sub>2</sub> O <sub>5</sub>	ppm	130
Strontium	Sr	ppm	87
Thorium	Th	ppm	24
Uranium	U <sub>3</sub> O <sub>8</sub>	ppm	120
Zinc	Zn	ppm	164
Vanadium	V	ppm	110

**Table 7.: MILL TAILING DRAINAGE WATER GENERAL COMPOSITION**  
Monthly composite samples 1992

	Flow	Ammon	Calcium	Magnesi	Iron	Chloride	Sulphat	Uranium	Radium
Month	m3/h	mg/l	mg/l	mg/l	mg/l	mg/l	mg/l	mg U/m3	Bq/m3
January	1.80	27.80	171.00	47.00	<0.1	227.00	575.00	294.00	102.40
February	3.00	18.90	111.00	27.00	<0.1	151.00	347.00	156.00	303.00
March	3.50	20.60	86.00	25.00	<0.1	139.00	383.00	191.00	85.60
April	5.80	8.20	65.00	23.00	<0.1	83.00	227.00	60.00	49.90
May	3.50	17.90	100.00	33.00	<0.1	137.00	374.00	105.00	73.90
Junij	1.10	16.50	95.00	26.00	0.10	121.00	306.00	221.00	73.80
July	1.40	30.00	174.00	46.00	<0.1	211.00	561.00	324.00	115.00
August	1.10	37.00	218.00	58.00	<0.1	310.00	756.00	435.00	141.70
September	0.90	34.50	215.00	57.00	<0.1	303.00	725.00	496.00	133.90
October	7.60	13.70	108.00	32.00	<0.1	139.00	345.00	185.00	79.30
November	8.00	10.30	125.00	22.00	<0.1	83.00	360.00	240.00	310.50
December	5.40	13.60	86.00	25.00	<0.1	104.00	228.00	148.00	97.20
Average	3.59	20.75	129.50	35.08	<0.1	167.33	432.25	237.92	130.52

**Table 8.: URANIUM MONTHLY QUANTITIES IN EFFLUENTS AND BREBOVSCICA AND RECENT OF CONTRIDUTION IN BREBOVSCICA RIVER**

**Monthly composite samples 1992**

Month	MINE WATER		MINE WASTE D		MILL TAIL.DR		BREBOVSCICA	
	KgU/mo	% contr.	KgU/mo	% contr.	KgU/	% contr.	KgU/mo	*KgU/mo
January	16.20	79.80	3.10	15.30	0.40	2.00	20.30	19.70
February	11.70	48.60	3.50	14.50	0.30	1.20	24.10	15.50
March	16.20	31.80	6.90	28.50	0.50	1.00	50.90	23.60
April	14.50	26.10	14.50	26.10	0.30	0.50	55.60	29.30
May	18.10	59.30	9.00	29.50	0.30	1.00	30.50	27.40
Junij	11.30	26.00	7.80	17.90	0.20	0.50	43.50	19.30
July	14.40	71.60	5.50	27.40	0.30	1.50	20.10	20.20
August	11.90	89.50	14.40	108.30	0.40	3.00	13.30	26.70
September	17.40	104.20	5.40	32.30	0.30	1.80	16.70	23.10
October	14.50	15.30	152.20	160.70	1.10	1.10	94.70	167.80
November	18.60	34.50	22.30	41.40	1.40	2.60	53.90	42.30
December	24.90	36.40	14.80	21.60	0.60	0.90	68.40	40.30
Total/year	189.70	38.60	259.40	52.70	6.10	1.20	492.00	455.20
Montaver	15.80		21.62		0.51		41.00	37.93

\* KgU/mo is suma of single effluent contributions

**Table 9.: RADIOACT. CONST. IN BREBOVSCICA RIVER AND EFFLUENTS  
TABLE COMPARISONS  
Monthly composite samples 1992**

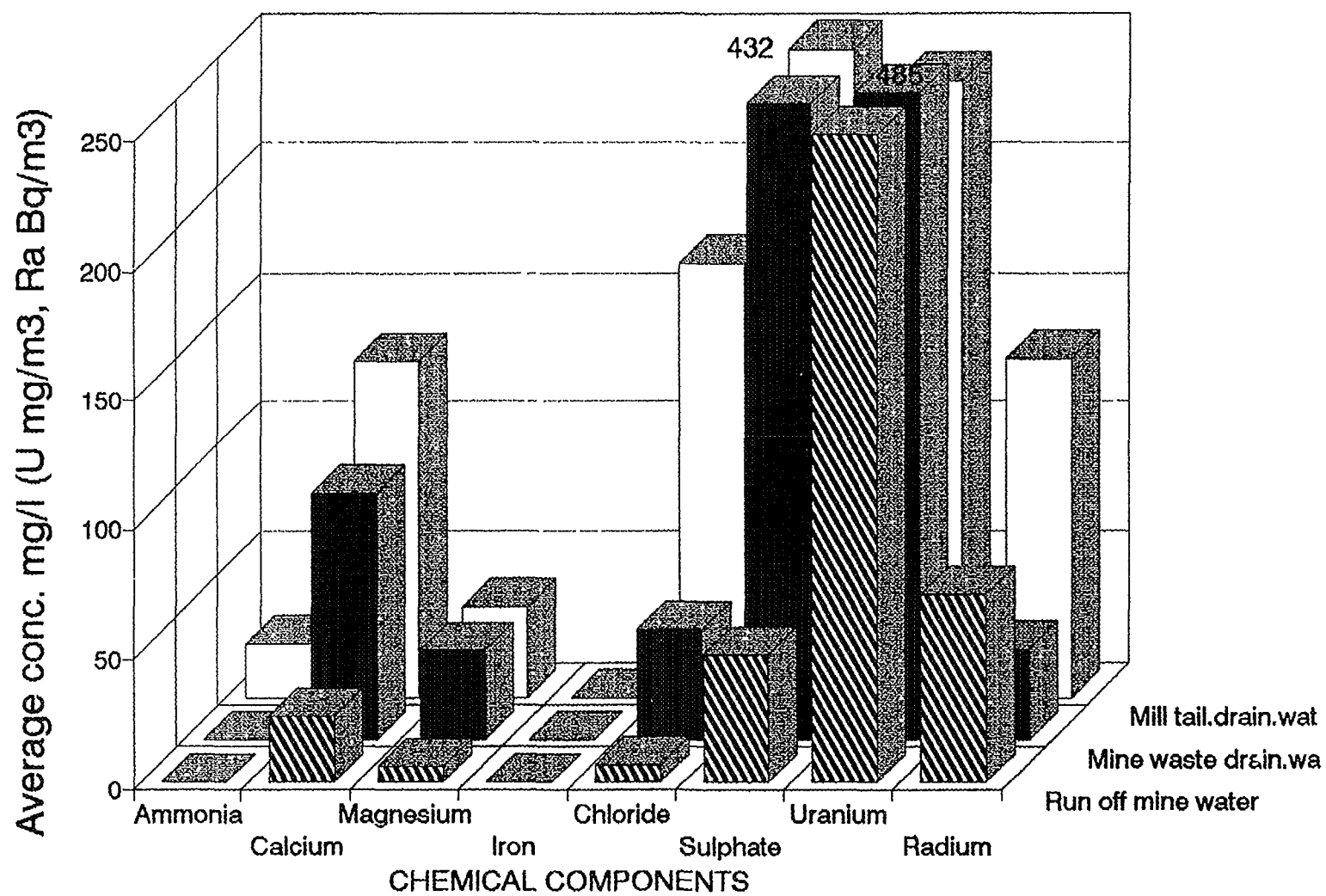
Month	BREBOVSCICA RIVER			MINE WATER			MINE WASTE DRAINAG			MILL TAILING DRAINAG		
	Flow	Uranju	Radium	Flow	Uranju	Radium	Flow	Uranium	Radium	Flow	Uranium	Radium
	m3/h	mg U/m	Bq Ra/	m3/h	mg U/m	Bq Ra/m	m3/h	mg U/m3	Bq Ra/	m3/h	mg U/m3	Bq Ra/m
January	720.00	37.80	10.70	80.00	272.00	64.20	11.00	374.00	20.30	1.80	294.00	102.40
February	1332.00	26.90	9.70	76.70	227.00	64.00	19.60	262.00	13.30	3.00	156.00	303.00
March	2160.00	31.70	9.00	70.10	310.00	63.20	35.50	262.00	20.90	3.50	191.00	85.60
April	4572.00	16.90	9.50	90.90	221.00	88.50	78.50	256.00	31.60	5.80	60.00	49.90
May	1620.00	25.30	28.00	97.10	251.00	104.40	37.00	328.00	57.10	3.50	105.00	73.90
Junij	1944.00	31.10	11.50	79.30	198.00	83.20	26.20	412.00	30.30	1.10	221.00	73.80
July	648.00	41.70	13.60	69.90	276.00	69.40	12.60	589.00	29.20	1.40	324.00	115.00
August	324.00	55.20	13.30	60.30	266.00	60.50	20.00	970.00	44.90	1.10	435.00	141.70
September	396.00	58.70	14.80	57.20	422.00	52.50	6.80	1111.00	34.20	0.90	496.00	133.90
October	5976.00	21.30	10.20	69.20	282.00	82.20	311.90	656.00	89.50	7.60	185.00	79.30
November	5544.00	13.50	5.90	103.70	249.00	65.70	99.60	315.00	22.60	8.00	240.00	310.50
December	4889.00	18.80		118.20	283.00	70.50	69.60	285.00	23.40	5.40	148.00	97.20
Average	2510.42	31.58	12.38	81.05	183.50	72.36	60.69	485.00	34.78	3.59	257.92	130.32

NOTE: Max.permis.conc.for potable water are:

Uranium - 50 mg U/m3  
Radium 226 1000 Bq/m3

From data, reference (6) it is evident that radioactive contribution by water sources to the population equivalent dose is small. Radon 222 and his daughter products contribution by air is far more important than other radioactive pollutants and ways. Engineered cover will minimize affects by air and by water ways.

Long term radioactive pollution by water pathway may act more important role in future due to washout under Mideuropian climate conditions.



**Figure 4: CHEMICAL COMP. IN MINE EFFLUENTS**  
*Monthly composite samples year 1992 ave*



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## REFERENCES

- (1) A. Pavel Florjancic, Exchavated materials disposals at Uranium mine Zirovski vrh - situation 31. 12. 1991  
Todraz, March 1992 (in Slovene)
- (2) Zmago Logar, Material balance of input and output from start till shutdown (09.01. 1984 - 06.30. 1990), IP RUZV  
Todraz, 1991 (in Slovene)
- (3) Covering materials tests and determination of type and thick ness of cover at mill tailing disposal at Borst, Final  
Report, Institute Josef Stefan  
Ljubljana, July 1992 (in Slovene)
- (4) A. Stergarsek and F. Dolenc, Zero discharge of liquid effluents in acid uranium ore treatment, process development and industrial experience,  
Ljubljana, 1989
- (5) A. Pavel Florjancic, Milko Krizman, Uranium (Raw material, extraction and environmental impact,  
Ljubljana, 1992 (in Slovene)
- (6) Radioactivity monitoring in vicinity of Uranium mine Zirovski vrh and environmental impact assessment, Results for year 1992,  
Institute Josef Stefan,  
Ljubljana, may 1993 (in Slovene)
- (7) Cadez Franc, Begus Tomaz, Hydrological conditions in ore deposit referring to the radioactive elements contamination,  
Todraz, Dec. 9. 1991
- (8) Report on measurements and site visit of radioactive waste storage place at Zavratac, Institut Josef Stefan  
Ljubljana, 12. 7. 1988 (in Slovene)
- (9) Preparation for remediation of the low and medium radioactive wastes at Zavratac, SNSA interoffice information  
Ljubljana, 1993 (in Slovene)
- (10) Almanach Community of Skofja Loka for 1990, INDOK  
Skofja Loka, July 1990 (in Slovene)

- (11) Elaborate on ore reserves in Uranium Mine Zirovski vrh status 31.12.1989,  
RUZV R - I - 56  
Todraz 9.03. 1990 (in Slovene)
- (12) Dolenc T., Origin of uranium ore deposit Zirovski vrh,  
Doctor thesis, Ljubljana 1983 (in Slovene)
- (13) Bernik J., Uranium geochemistry and of some other elements  
in ore deposit Zirovski vrh,  
Ljubljana, 1981 (in Slovene)
- (14) Verbal communication with Dr. Pucelj, Institute Josef Stefan  
Ljubljana, Sept. 1993
- (15) Begus T. Hydrological Characteristics of the Mine Waste  
Disposal Jazbec  
Todraz, Oct. 1992 (in Slovene)

**IDENTIFICATION AND RADIOLOGICAL CHARACTERIZATION  
OF CONTAMINATED SITES IN SPAIN.  
ANDUJAR URANIUM MILL SITE**

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**Abstract**

Empresa Nacional de Residuos Radiactivos, S.A. (ENRESA) is remediating an inactive uranium mill facility in the town of Andújar in the south of Spain. The Andújar plant became operational in 1959 and continued in operation until 1981. All solid waste generated during the operation of the plant are contained in a tailings pile, which covers an area of 9.4 hectares and has a total volume of about one million cubic meters.

The remedial action plan proposed for Andújar mill site involved stabilizing and consolidating the uranium mill tailings and contaminated materials in place. The actual tailings pile was reshaped by flattening the sideslopes to improve stability. Tailings from sideslope flattening were relocated around the existing pile and on the top of the lower pile. Mill equipment, buildings and process facilities were dismantled and demolished and place in the tailings pile. Off-pile contaminated soils were excavated and placed on top of tailings pile in order to reduce the radon flux.

The pile has been covered with a multilayer system to meet the three simultaneous demands of erosion control, infiltration and radon control.

In this paper is described the identification and radiological characterization of Andújar Uranium Mill site wich was carried out in three phases:

During the first phase, the radioactive concentration in tailings was established, and site general parameters and potential spreading of contamination were defined.

During the second phase, once stabilization in place was decided three main tasks were performed: a radon flux field test, a radionuclide migration study and radiometric study.

During the third phase in order to establish the final conditions of the stabilized tailings pile and off-pile soils clean conditions after remediation works a radiometric survey was carried out.

## 1. INTRODUCTION

The Andújar Uranium Milling Plant is in the province of Jaén (Andalucía) on the southern floodplain of the Guadalquivir river at 1.5 kilometers south from the urban center of Andújar. The site is trapezoidal in shape, covers an area of approximately 17.5 hectares and is contained within a peripheral wall, which is about 150 m from the course of the river.

The plant was designed for processing low grade uranium ore (0.15% of  $U_3O_8$ ) and produced 80% concentrate of  $U_3O_8$  in the form of sodium and ammonium uranate at a rate of 60 to 80 tons per year. The plant became operational in 1959 and continued in operation until 1981. During this period 1.2 million tons of uranium ore were processed to produce 1350 tons of  $U_3O_8$  with a fineness of 80-85%. Recovery of the uranium involved sulphuric acid leaching followed by ion exchange or by tertiary amine/Kerosene extraction. Solid wastes were stored in the tailings piles and liquid wastes were treated before disposal to the Guadalquivir river.

The configuration of the Andújar mill site includes the following areas: the tailings pile, the processing plant, the waste water treatment area, the auxiliary and administrative buildings and the housing area.

All solid waste generated during the plant's operation (1.2 million tons) are contained in the tailings pile, which sideslopes vary from 25 to 35 degrees.

## 2. ANDUJAR SITE CHARACTERIZATION WORKS

The general characterization scheme for Andujar uranium mill site was carried out in three phases:

### - Phase I

During this phase the radioactive concentration in tailings was established, and site general parameters and potential spreading of contamination were defined. After that a cost-benefit-analysis was made to decide and justify what form of remediation between the following alternatives:

- . No action
- . Stabilization in place
- . Relocation to an alternate site

### - Phase II

Once stabilization in place was decided three main tasks was performed:

- . A radon flux field test
- . Radionuclide migration study
- . Radiometric survey

The results of this studies was used mainly for radon an infiltration barrier design and general condition for tailings pile reshaping, and stabilization of process equipment and building debris.

## - Phase III

This phase was designed to establish the final conditions of tailings pile and off-pile soils clean conditions after remediation works.

### 2.1. PHASE I

#### 2.1.1. 150 Tailings sample program

During plant operation an inventory of the radionuclide concentrations of the ore and processed concentrate was kept. The total activity of the ore processed was 6,021 Ci and 622 Ci was produced in the form of concentrate ( $U_3O_8$ ). Assuming that all remainder radionuclides are contained in the tailings pond, the total activity they contain is conservatively estimated at 5,399 Ci ( $2 \times 10^{14}$  Bq). The average Ra-226 concentration in tailings, estimated from operational records was 13 Bq/gr.

Radium and Uranium activities have been measured in the tailings and underlying unsaturated zone using analytical and gamma spectrometer methods. Average activities for these and other isotopes are:

Radium-226	12 ± 6	Bq/g
Uranium-238	2 ± 1	Bq/g
Lead-214	10 ± 5	Bq/g
Bismuth-214	10 ± 5	Bq/g
Protactinium	1 ± 0.5	Bq/g

The Uranium activities show almost constant levels through the tailings and unsaturated zone. Uranium concentrations beneath the tailings dam in the unsaturated zone, are between 0.004% and 0.008 by weight.

In contrast, Ra-226 measurements indicate activities of 12 Bq/g in the tailings falling to 0.2 Bq/g in the underlying strata. It is evident from these results that attenuation of Ra-226 is occurring in

the unsaturated zone and that Ra-226 is not at present reaching the saturated zone in significant quantities.

Samples from boreholes constructed through the tailings were subjected to particle size distribution analysis. The results show that the tailings consist of silts with clays and fine sands. The maximum concentration of clay and sand size particles in samples was 50%. Particle size distribution in the tailings indicates the finer material to generally exist in the centre of the ponds, as is consistent with usual tailings pond construction methods.

#### 2.1.2. Monitoring Programs

Levels of alpha and beta activity and Uranium concentrations have been monitored in the region of the FUA since 1983. Monitoring of surface water, ground water, river sediments, river organism and vegetables and crops grown in the area has been carried out. The results show considerable variation.

The mean annual analysis for 1986 for wells in the region of the FUA are shown on Table 2.1. The zone of high Uranium concentrations is limited to three wells: PZ-16-A, PZ-17-A and PZ-18-A. Uranium levels in well F12 and F13 are about 50 times background and in F11 approximately 10 times background.

Ra-226 levels are only slightly elevated above background, as indicated by boreholes upstream of the site. These observations confirm the conclusions drawn from the boreholes constructed through the tailings piles, that the Uranium appears to have a high degree of mobility in the saturated zone whereas Ra-226 is retained in the unsaturated zone.

After plant shutdown two main liquid effluent streams flow to the water treatment system of the plant. One from the drainage pipeline system of tailings pile other from the rainfall drainage system.

TABLA 2.1

RESULTS OF ANALYSIS OF GROUNDWATER SAMPLES  
IN Bq/CUBIC METRE (1986)

<u>WELL</u>	<u>ALPHA</u>	<u>BETA</u>	<u>URANIUM</u>	<u>Ra-226</u>
PZ-1-A	213 ± 157	231 ± 79	75 ± 25	12 ± 5
PZ-4-A	259 ± 213	489 ± 72	50 ± 25	14 ± 7
PZ-3-A	255 ± 130	177 ± 72	75 ± 25	11 ± 3
PZ-8-A	248 ± 209	496 ± 87	75 ± 25	14 ± 11
PZ-5-B	237 ± 141	1018 ± 314	125 ± 25	12 ± 4
PZ-6-B	287 ± 309	505 ± 94	175 ± 25	10 ± 10
PZ-7-B	307 ± 172	3009 ± 860	100 ± 25	9 ± 4
PZ-11-B	314 ± 349	467 ± 56	175*	8*
PZ-12-B	240*	98 ± 50	50*	9 ± 4
PZ-10-B	119 ± 1	440 ± 240	50*	13*
PZ-16-A	674 ± 410	1736 ± 746	676 ± 400	14 ± 7
PZ-17-A	2716 ± 1724	3199 ± 1065	5661 ± 2179	13 ± 7
PZ-18-A	1812 ± 768	1930 ± 744	5511 ± 4208	13 ± 5
PZ-19-A	261 ± 328	318 ± 130	175 ± 50	14 ± 12

\* One result with value greater than lower limit of detection.

± Likely level of uncertainty.

There was records of concentration activity and flows prior to treatment for this two streams that shows Radium concentrations up to 1000 mBq/l and 4 mg/l of uranium.

## 2.2. PHASE II

### 2.2.1. Radon Test Program

With the aim of testing the radon barrier ability to reduce radon flux from Ra-226 in tailings pile, a 5000 m<sup>2</sup> test cover was



constructed on the top of the pile during the last three months of 1987.

Calculations with RAECOM code showed that 50 cm of clay barrier was sufficient to reduce the radon flux from  $400 \text{ pCi/m}^2/\text{sg}$  to  $20 \text{ pCi/m}^2/\text{sg}$ , as it was specified in the general design criteria.

In order to protect the radon barrier from erosion a multilayer cover was designed consist of the following layers:

- (1) Radon barrier, composed of clay materials.
- (2) Filter layer, composed of sandy materials.
- (3) Erosion barrier, composed of local soils.

Taking into account the great influence of the radon barrier moisture content in the radon flux, a sensitivity analysis was performed with the purpose of defining the differents thickness of the layers to be used for the tests.

The final test pad consists of six different bands, five meters wide and 90 m. long, each of them divided into three different water content sectors of 30 m. long.

The A, B, C bands were constructed with different thickness of the clay layer, (60, 50 and 40 cm. respectively), while the sand filter and soil thickness were maintained the same at 40 and 120 cm respectively. The D, E and F bands were prepared with 60 to 40 cm of clay thickness respectively and sand filter and soil thickness were maintained the same at 40 and 100 cm respectively.

The optimum water content for the clay materials was determined to be 13,22%, and the three different test sectors were defined as:

Sector I :  $9,22 < H < 11,22$

Sector II :  $12,22 < H < 14,22$

Sector III :  $15,22 < H < 17,22$

Where  $H$  is the molding water content.

This test facility allows the measurement of the radon flux at the surface in 18 different arrangements. In addition 11 wells, nine up to the different clay layers (thickness and water content sectors) and two up to the tailings surface were constructed in order to measure the radon flux directly without the intermediate effects of filter sand and soil. A reference point was located outside the test facility to measure the radon flux in the tailings bare surface.

Prior to initiating the construction works, a three month program was conducted to characterize the bare tailings radon flux at the test facility surface and throughout the pile surface.

The monitoring program, in operation for two years since February 1988 is shown in table 2.2, and provided data for the purpose of radon barrier characterization and RAECOM code calibration.

TABLE 2.2  
MONITORING PROGRAM

Sample Collection					Sample Measurement	
Type of Sample	Number	Location	Type	Frequency	Frequency	Type of Measurement
1. Radon	18	Top Surface	Continuous	Monthly	Monthly	Radon flux
	9	Clay Surface (wells)	Continuous	Monthly	Monthly	Radon flux
	2	Tailings Surface (wells)	Continuous	Monthly	Monthly	Radon flux
	1	Tailings Surface (reference)	Continuous	Monthly	Monthly	Radon flux
2. Moisture Content	18	Clay layers	Continuous	Monthly	Monthly	Water content
	2	Soil layers	Continuous	Monthly	Monthly	Water content
	1	Filter layer	Continuous	Monthly	Monthly	Water content
	2	Bare tailings (wells)	Continuous	Monthly	Monthly	Water content
	1	Bare tailings (reference)	Continuous	Monthly	Monthly	Water content

The main results of this program can be summarized as follows:

1. Thicknesses from 40 to 60 cm of clay reduce the bare tailings flux to levels of  $2 \text{ pCi/m}^2/\text{seg}$ .
2. The flux reduction observed as the layer increases 10 cm from 40 to 50 cm thickness is double than the values registered when the layer increases their thicknesses from 50 cm to 60 cm.
3. The average radon flux through the entire cover system was  $0,64 \text{ pCi/m}^2/\text{seg}$ .
4. The average radon flux through bare tailings was  $271.8 \text{ pCi/m}^2/\text{sg}$

#### 2.2.2. Radiometric Survey

To evaluate levels an extent of contamination in process equipment, buildings and off-pile soils a radiometric survey was conducted throughout the plant site.

The first task performed was the physical and radiological characterization of equipment and facilities of the Plant. The physical characterization consisted of a detailed inventory of equipment and facilities indicating location, construction material, current and estimated volume after cutting, anticipated contamination levels, weight and structural conditions. The radiological characterization was carried out in two phases. Direct gamma radiation inside and outside large items of equipment, (particularly chemical process tanks) and surface contamination (alpha and beta-gamma) were measured during phase I. 1,172 measurements were conducted on 586 measuring points. In phase II a more detailed inspection of the contaminated facilities was performed in order to define the extent and level of contamination. 200 samples were taken using the dry smear test method for determination of alpha and beta surficial contamination.

A complete characterization of levels (alpha and beta-gamma) and type (fixed or loose) of contamination was obtained for all equipment and facilities and materials were classified as contaminated or not in accordance with the free-release limits established by the Regulatory Authorities (i.e.,  $10^{-6}$  uCi/cm<sup>2</sup> for alpha contamination and  $10^{-5}$  uCi/cm<sup>2</sup> for beta-gamma contamination).

The physical and radiological characterization also provided information to identify the radiological and conventional risks involved in the decommissioning operations.

Radiological risks are similar to those involved in the tailings works and result mainly from the handling of tanks and equipment with loose contamination and residues from the ore leaching process.

Conventional risks are associated with the dismantling of asbestos closure panels, stripping of rubber coatings in tanks prior to their cutting to prevent fire risks, generation of aerosols, collection and removal of electric transformers with PCB's and removal and disposal of tanks with traces of heavy fuel.

The extent and level of contamination in buildings and structures was evaluated through a radiometric survey which was carried out in two phases. In the first phase, 4566 direct gamma radiation and surface contamination measurements were performed over 2283 points. In the second phase, a more detailed inspection of the contaminated areas was done and 471 samples were taken for analysis of surficial contamination. In addition, 48 cores of concrete slabs and structural walls were drilled to evaluate the extent of the contamination in depth.

The surficial contamination generally found over buildings and structures was identified as loose and no significant deep contamination was detected as a result of this survey.

A radiometric survey outside the plant was made to evaluate the spread of contamination in soils, in a circular area of 750 m around the facility.

The grid size used was  $18 \times 18 \text{ m}^2$ , and measurements was taken with an INa detector prepared for radium gamma energy of 186 KeV.

Spot of contamination was search by radiochemical determination in laboratory of Ra-226 concentration in soil samples taken at 0.3 m an 1 m depth.

The offpile soils within the plant area, was surveyed measuring the gamma exposure rate in contact an at 1 m from the surface.

After this survey selected points was defined to take samples at different depths up to 1.5 m, for Radium concentration determination in laboratory by radiochemical analysis of soil samples.

#### 2.2.3. Radionuclide Migration Study

A complex study of radionuclide migration through tailings pile, unsaturated underlayer zone and saturated zone, with special emphasis on Uranium and Radium behaviour.

A detailed radiochemical mineralogical, physical and chemical evolution characterization of tailings pile and geologic sustrate was conducted, pintly with a complete characterization of insertitial water in pile, unsaturated zone and saturared zone.

As a result of this work a complete knowledge of the Uranium radionuclides behaviour and distribution in pile, unsaturated zone and saturated zone was kept with a first conclusion that support previous studies (150 tailings sample program) about the statement that only uranium has a high mobility through unsaturated and saturated zone while Ra-226 seems retained in the underlayer zone (unsaturated), mainly because of the carbonate compounds present in the silty clay substrate.

## 2.3. PHASE III

### 2.3.1. Tailings and radon barrier final conditions

To evaluate the final construction thicknesses of the radon silty clay barrier the following verifications was conducted:

- Ra-226 concentration in the first 3 m of relocated tailings.
- Tailings radon emanation fraction determination.
- Silty clay diffusion coefficients determination and longterm moisture content.

Twenty boreholes distributed from top to slopes of the reshaped tailings pile was made to take three samples at 1, 2 and 3 m in depth.

The average Ra-226 concentration from the 60 samples, 292 pCi/gr, was finally used to estimate the thickness of silty clay barrier, instead of the average Ra-226 concentration used in the initial calculations (345 pCi/gr).

The laboratory determination of emanation fraction, diffusion coefficients and long-term moisture content will be described in future workshops.

### 2.3.2. Soil verification survey

A radiological final soil survey is required as a part of the decommissioning process at Andujar uranium mill site. The ultimate goal of the survey is to provide with reasonable assurance that the concentration of radium-226 in land averaged over an area of 100 squared meters shall not exceed the background levels by more than.

- a) 5 pCi/gr, averaged over the first 15 cm below the surface.

b) 15 pCi/gr, averaged over 15 cm thick layers of soil more than 15 cm below the surface.

However, the cleanup criteria applied is the value 15 pCi/g as after final survey the off-pile soils will be covered with a layer of borrow clean soil at least of 15 cm.

Based on the initial radiometric survey (see section 2.2.2) the off-pile area was divided into five zones (which are also divided into subzones) to carry out the final radiological survey activities. These zones are:

Zone 1 correspond to sector NW

Zone 2 correspond to sector NE

Zone 3 correspond to sector E

Zone 4 correspond to sector S

Zone 5 correspond to sector W

Grid system are established at the site on the one hand to facilitate systematic measuring and sampling process and on the other hand to provide measurement and exact sample points unambiguous location.

The grid system is referenced to a site geographic coordinates. It consists of intersecting lines arranged in a perpendicular pattern of 100 square meter approximately (1331 areas of this size).

#### 2.3.2.1. Radiological instrumentation

Survey are performed collection two types of radiological data: direct field measurements using portable instruments and sample for laboratory analysis.

The portable instrumentation used consist of a central process unit and two sodium-iodine scintillation detectors. The detectors are configured to count total gamma radiation and radium-226 emission in the energy 186 KeV with a monochanel analyzer.

Moreover, the detector have a lead shield of 5 cm thick and 8 cm long to avoid tailings pile radiation contribution.

A hand cart has been designed to transport the set, central process unit and detectors.

Radium concentration of soil samples in laboratory was made by gamma-spectrometry. A high purity Germanium detector (HPGe) with a Marinelli geometry is used. The counting is done in the radium-226 energy peak 186 KeV.

The procedure used has a 1 pCi/g as a lower limit of detection and assure a maximum uncertainty of the mean value of 0.5 pCi/g for concentration equal or more than 15 pCi/g.

#### 2.3.2.2. Measurement Methods

##### a) Direct field measurements

There are two main goals of the direct field measurements: a fast assessment of soil radiological state providing a preliminary verification and a initial survey of zones.

The measure set (unit central process and detector) fixed to a hand cart is transported through each 10 x 10 m square area drawing longitudinal lines with East-West and South-North directions.

The measurement is carried out in contact with the soil and the data collection is automatic. The result measurement is an integrate value of the emission recorded in a prefixed time.

##### b) Soil sampling procedure

The soil sampling procedure is based upon areas of 100 square meters and is characterized by nine-plug composite sample.



The 10 x 10 m<sup>2</sup> area is divided into nine sectors. A soil sample is collected from the top 15 cm of soil in the central point of each sector. The nine samples are approximately of equal mass and homogeneous mixed to be representative.

# **A SHORT REVIEW OF SWEDISH URANIUM MINING, MILLING AND RESTORATION IN RANSTAD**

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## **Abstract**

In Sweden large but low grade uranium ore reserves are found in the district of Västergötland. The total uranium content is estimated to be in the order of 1 million tons. A uranium mining and processing plant was taken into operation in 1965 and the capacity was designed for an Annual production of 1275 tons of uranium. In 1984 the Swedish government made the decision to stop all plans for uranium production in Sweden and in 1985 it was decided that the whole Ranstad area should be restored. Through all the years of industrial activities at Ranstad, the environmental consequences have been studied. Today the environmental sampling programme is still in force to ensure that emissions and seepage from the mining area are below acceptable levels.

## **1. INTRODUCTION**

In Sweden large but low grade uranium ore reserves are found in the district of Västergötland. The ore is found in the extensive formations of alum shales and the area of interest covers about 500 km<sup>2</sup>. The thickness of the uranium ore layer is 2.5 to 4 meters and the ore grade 250 - 325 ppm. The total uranium content is estimated to be in the order of 1 million tons of which 30% is estimated to be recovered from the economical point of view.

The interest for the exploration of the low grade uranium shales started in the early 50's. In the 60's a 120 tons per year uranium mill was built in Ranstad. The plant was taken into operation in 1965. As the demand for domestic uranium at that time was small it was not justified to operate the plant at full capacity. The production rate was about 50% of full capacity during the years 1966 to 68.

## **2. PLANS FOR INDUSTRIAL U-PRODUCTION**

As time went by and the demand for natural uranium increased, it was recognized that the Ranstad uranium plant, as it was built, would be too small for covering the domestic requirements. This was especially clear as the production costs for the uranium produced at Ranstad at that time could not compete with uranium produced in other countries.

New and more efficient processes were studied and in combination with results of a feasibility study a new mill with much higher capacity was considered. It was planned that the Annual production would be 1275 tons of uranium, and that it would be sufficient for a major part of the uranium demanded for the Swedish reactor programme. The amount of ore produced would be in the order of 6 million tons per year.

A combination of open pit and underground mining was planned for the first 15 years. The open pit mining was designed as strip mining with a capacity of 3 million tons of uranium ore per year. The capacity of the underground mining would at first be of the same order of magnitude and, as the open pit mining decreased, finally reach 6 million tons.

The planning for the Ranstad uranium factory continued through research and development until 1984. At that time the Swedish government made the decision to stop all plans for uranium production in Sweden and the Ranstad uranium license was cancelled.

Until that date uranium had been processed for four years (1965 to 1969) and a total amount of 215 tons of uranium oxide had been produced.

### 3. ENVIRONMENTAL IMPACT

Through all the years of industrial activities at Ranstad, the environmental consequences had been studied. These studies include dose calculations as well as impact from non-radioactive elements.

Measurements on Uranium and Radium content in water, water vegetation and sediments have been carried out as well as measurements on Radon exhalation and gamma radiation from different areas of interest.

Water from the open pit area enters a small stream, Pösan. For the estimation of the radiological environmental impact water samples up-stream and down-stream are collected weekly and then analyzed for radium and uranium content. The same procedure is used for an other stream, Hornborgarån. Water from the uranium production plant was collected in a special lake, "Collection lake" and later, after sedimentation, released to another lake, "Storage lake", before releasing to the recipient, Hornborgarån. In Hornborgarån uranium and radium contents in water vegetation, as well as in sediments, were measured.

The results of the above mentioned measurements are presented in table 1.

Table 1.

	<b>Uranium</b> Bq/m <sup>3</sup>	<b>Radium</b> Bq/m <sup>3</sup>
<b>Pösan</b>		
up-stream	22-74	< 15
down-stream	850	15
<b>Collection lake</b>	37000 - 55000	<185
<b>Storage lake</b>	370-1200	<7 - 74
<b>Hornborgarån</b>		
up-stream	100-520	1,5 - 30
down-stream	110 - 370	4 - 20
<b>Vegetation</b>	Bq/kg	Bq/kg
Up-stream	9-85	7-37
Down-stream	12-67	13-25
<b>Sediment</b>		
Up-stream	55-140	50-110
Down-stream	37 - 96	25 - 50

Measurements on radon exhalation and gamma radiation were also made. Results are presented in table 2.

Table 2.

	Radon exhalation mBq/s m <sup>2</sup>	Gamma radiation μSv/h
New tailings - not covered	70	2,7 - 2,9
Old tailings - not covered	140	2.5
Covered with 1 m of soil	0.2	0.3
Covered with 0.2 m of soil	0.7 - 1.5	0.2
Untouched area *	0.3 - 11	0.1 - 0.2

\*average from four different locations

In the early 70's a substantial leakage of heavy metals and sulphate had been observed from the tailings. The leakage water was collected and had to be purified before being released to recipients. Due to this, a more comprehensive monitoring programme was carried out for the control of the environmental consequences of releases from the tailings, as well as from the open pit mine. The content of radium and magnesium in water was the limiting element in this early recipient control programme.

#### 4. RESTORATION

In 1985 it was decided that the whole area should be restored. The area consisted, except from the industrial area, of the former open pit mine and the uranium mill tailings. The restoration of the open pit mine was planned to result in a small lake with an area of 0.27 million m<sup>2</sup> and a total water volume of 1.3 million cubic meters. The mill tailings consisted of about 1 million cubic meters crushed alum shale, covering an area of 0.25 million m<sup>2</sup>.

During the years 1987 to 1989 studies on mill tailing passivation was performed and a restoration programme was worked out. The programme was presented to the county administration and accepted in 1990.

As mentioned above, the open pit mine was to be transformed it into a small lake. Before pumping, the natural water flow was stopped, the bottom of the mine was covered by till to prevent the water to come in contact with the alum shale, thus reducing the leakage of pollutants to the lake.

The plan for the mill tailing area was to cover the tailings with layers of different characteristics to minimize the infiltration of rainwater, as well as with oxygen, to reduce the weathering. The layer of tailings was between 6 to 10 meters, with a coverage of a up to 0.3 meter thick layer of moraine from earlier attempts of restoration. The whole area should then be covered by a moraine-clay mixture of 0.2 meters. On top of that, a 0.2 m layer, consisting of crushed limestone which in it's turn was to be covered by 1.2 meters of moraine. On top of everything, a soil-moraine mixture of 0.2 meters should be placed.

In all, the protective layer should be in the order of 1.6 m, which was estimated to withstand the penetration of rainwater into the tailing masses.

The restoration work started in 1990. The mill tailings were covered as planned and the open pit was transformed into a small lake. Old factory buildings, not in use, were demolished and in 1992, the covering was finished. In order to remove the last tracks of the industrial activities, landscaping was started, in order to make the whole area look natural.

## 5. FOLLOW-UP PROGRAM

The aim of all restoration work is to minimize the environmental impact. It is therefore important to firstly, predict the effects of the restoration before the restoration is finished and secondly, after the restoration, have a follow-up programme, to ensure that the work has been successful.

The follow-up programme now in force, is a result of discussions between authorities, local and governmental, and the contractor, Studsvik, used for the restoration work. The programme is a release- and recipient control programme and it includes water sampling and discharge measurements from a number of different sampling stations.

The water samples for heavy metals analyses are collected every week. Furthermore, a physical-chemical program is performed once a month. In order to have a picture of the transportation of heavy metals from the tailings, as well as from the open pit area, a specific calculation programme (compartment model) has been established.

## REFERENCES

- [1] Åke Andesson and Gunnar Olsson: *Uranium recovery from Swedish low grade bituminous shales*. Studsvik AB, S-611 82 NYKÖPING, Sweden
- [2] Björn Sundblad: *The restoration of the uranium mine area of Ranstad, Sweden*. Studsvik AB, S-611 82 NYKÖPING, Sweden
- [3] Björn Sundblad and Yvonne Stiglund: *Efterbehandling Ranstad - Utsläpps- och recipientkontroll (in Swedish)*. Studsvik AB, S-611 82 NYKÖPING, Sweden

## IDENTIFICATION AND RADIOLOGICAL CHARACTERIZATION OF CONTAMINATED ENVIRONMENT IN UKRAINE

C RUDY

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### Abstract

As a result of the Chernobyl accident, 1779 towns and settlements all over the Ukraine were contaminated with level of surface contamination exceeding 1 Ci/sq.km of Caesium-137. In general, Chernobyl accident affected 18 Ukrainian regions. Nowadays in Ukraine are officially registered 104,802 radioisotopic devices, which are used by 2 641 organisations. Also there are 550 gamma-flaw detectors, 78 installations for gamma-therapy and 6 medical isotopic units. Average annual radioactive inventory of open isotopes used in Ukraine comes to 327,166 Ci. Radioactive inventory for closed isotopes is equal to 991 kg-equivalent of Radium. Activity of radioactive wastes disposed annually in Ukraine falls within 14,000 - 22,000 Ci (NPP wastes are not included). All this creates a considerable potential for release of radioactivity into environment and contamination of soil, water and biota. Critical radioecological situation was created in the areas of uranium mining and milling in the South of Ukraine, mainly in Kirovograd and Dnipropetrovsk regions. Significant volumes of tailings and depleted uranium ore waste piles has accumulated in the region. In fifties and sixties this materials were used for civil building purposes and road building in those regions. In total, uranium milling and mining industry occupies 5530 hectares of land, 1340 ga are damaged. Beside the radiological survey programme for Chernobyl affected regions, up to date there is in action the programme for radiological survey of 20 large industrial population centers. This programme is planned to be extended to comprise all population centers having more than 50,000 inhabitants. To coordinate all activities in the area of environmental restoration, and as a response to IAEA Technical Co-operation project RER/9/022, Ministry for Environmental Protection of Ukraine, recently started the development of National Environmental Restoration Programme (NERP), which as a first step includes systematic detalisation of the current radiological situation in Ukraine.

## 1. INTRODUCTION

Severe nuclear accident at the unit 4 of the Chornobyl Nuclear Power Plant caused heavy contamination of vast regions in it's northern part. In general, Chornobyl accident affected 18 Ukrainian regions. As a result, 1779 towns and settlements all over the Ukraine were contaminated with level of surface contamination exceeding 1 Ci/sq.km of Caesium-137.

Chornobyl accident stimulated initiation of the more or less regular and substantial radioecological investigation of the Ukrainian territory.

While Chornobyl contamination affected in even extent both the rural and municipal areas, reflecting prevailing atmospheric dispersion routes in first days and weeks after the accident, the contamination of the technogenic nature reflects the structure of the relevant industrial activities and mostly affects the respective population centers.

Nowdays in Ukraine are officially registered 104,802 radioisotopic devices, which are used by 2 541 organisations. Also there are 550 gamma-flaw detectors, 79 installations for gamma-therapy and 5 medical isotopic units.

Average annual radioactive inventory of open isotops used in Ukraine comes to 227,165 Ci. Radioactive inventory for closed isotops is equal to 991 kg-equivalent of Radium. Activity of radioactive wastes disposed annually in Ukraine falls within 14,000 - 22,000 Ci (NPP wastes are not included).

All this creates a considerable potential for release of radioactivity into environment and contamination of soil, water and biota.

There were several cases of direct harmful impact of lost radioactive sources on human health and human life in Ukraine. It is sufficient to mention the Kramatorsk case, when lost radioactive source came into the concrete wall of the house and caused the death of two children before it was identified.

Also critical radioecological situation have been created in the areas of uranium mining and milling in the South of Ukraine, mainly in Kirovograd and Dnipropetrovsk regions. In total, there accumulated significant volumes of tailings and

depleted uranium ore waste piles. In fifties and sixties this materials were used for civil building purposes and road building in those regions.

Another issue, contribution of which into contamination of environment could be essential, is coal and iron ore mining industry, when there are initiated secondary mechanisms of natural radionuclides concentration (mining waters with considerable content of natural radionuclides).

## 2. CURRENT LEGAL PROVISIONS AND ORGANIZATIONAL STRUCTURES IN UKRAINE DEALING WITH THE ENVIRONMENTAL RESTORATION

Environmental restoration is an extremely complex and cost consuming activity, which involves many organizations and requires considerable resources for implementation. Even superficial analysis of the environmental restoration operations in different countries with different conditions shows that the main prerequisite for ensuring effective environmental restoration is the establishment of an optimal organizational structure and management framework for relevant activities.

Main governmental agencies involved in activities on radioecological environmental restoration in Ukraine nowadays are following:

- 1) Ministry for Environmental Protection of Ukraine (since 1993).
- 2) Ministry for the Mitigation of the Effect of Chernobyl Accident (MinChornobyl).
- 3) Ministry for Geology.
- 4) Civil Defence of Ukraine.
- 5) Ministry for Health Protection.
- 6) State Committee for Hydrometeorology.

Though, there is still no special organizational framework for co-ordinated and optimally planned rehabilitation activities in Ukraine.



It is clear that sound organizational structure should be based on relevant legislative provisions.

Legal provisions that could be used for deriving certain functions with respect to rehabilitation of the contaminated environment are formulated in the "Act on Environmental Protection", 1991. According to this legislation, general responsibility for co-ordination of all environmental activities performed in Ukraine by different governmental agencies, ministries and institutions is imposed on Ministry for environmental protection of Ukraine.

## 2. ENVIRONMENTAL CONTAMINATION AS A RESULT OF THE CHORNOBYL ACCIDENT

Environmental contamination in Ukraine after the Chernobyl accident is investigated comparatively good. Comprehensive work of the IAEA within the framework of the International Chernobyl Project has become well-known all over the world. In addition, a good deal of information concerning the levels of environmental contamination is regularly being presented at relevant international conferences and symposia.

Nowdays the responsibility for characterization of post-Chernobyl contamination is shared between MinChernobyl and Hydromet. All information gained as a result of different radiological surveys performed by different laboratories and institutions are summarized and reviewed by special Intergovernmental Radiation Monitoring Commission, which was created ad hoc after Chernobyl accident in 1986.

To date since 1986 there have been surveyed 22 regions, 262 counties and 12 800 population centers in Ukraine. There were taken and analysed more than 270 000 samples for Caesium-137, nearly 63 000 samples have been analysed for Strontium-90. In general more than 3,700 sq. kilometers of forests and 6,700 sq.kilometers of agricultural land were surveyed. Aerial gamma survey of all the Ukrainian territory has been performed to the date.

As a result there were derived mean contamination levels for each population center within affected regions. This mean figures are reflected in a special annual register ("Ucge

Passport of Population Centers Contaminated After Chernobyl Accident") which is approved by the Minister for Health Protection.

General figures on contaminated areas with Strontium-90 and Caesium-137/134 are summarised in Table I and Table II, and illustrated at Fig. 1 and Fig. 2, respectively.

Number and distribution of population centers in contaminated areas with different levels of Strontium-90 and Caesium-137/134 are summarised in Table III and Table IV.

On the basis of the review of those measurements it was decided to perform in 1993 confirming series of measurements in settlements with a contamination levels exceeding 2 Ci/sq.km, that is, final formal conclusions on contamination levels of post-Chernobyl nature will be made after analysis of 1993 survey data. Besides the environmental contamination there were also investigated the contamination levels of food - milk, meat, vegetables, mushrooms, berries, which contribute the most into the dose of the population. This information in form of mean values for each village is also included in the "Passport...".

#### Restricted zone

Restricted Zone (20-km) represents a special case with respect to environmental restoration. Having the most severe contamination, this area has a potential for spreading nuclides beyond it's boundaries.

In total, in Restricted Zone there is 110 000 Ci of Cs-137, 127 000 Ci of Sr-90, and 800 Ci of different isotops of Plutonium. These figures do not include radionuclide inventory of post-accident radwaste disposal sites as well as within the Chernobyl NPP site. To the date, the major bulk (82%) of nuclides Caesium-137/134, Strontium-90 and Plutonium still is concentrated in the upper 5 cm layer of soil. The rest 18% is within next 5-10 layer. Table I summarizes distribution of radioactive nuclides inventory in the upper 5 cm layer of soil over Zone.

In 5-km zone mean level of Caesium-137 surface contamination is 500 Ci/km. The maximum registered active sample of Plutonium contaminated soil was taken at the left

bank of river Pripiat (Krasne tributary): Cs-137 - 760 Ci/km, Sr-90 - 1720 Ci/km, Pu-239,240 - 40.1 Ci/km, Pu-238 - 19.2 Ci/km. General radioactivity of the soil reduces gradually (appr. 10% annually) at the expense of natural decay of Ce-144, Cs-134 and Ru-106.

### Migration

Spreading of radionuclides beyond the Zone is being proceeded by four major routes:

- surface water carry-over;
- resuspension;
- biogenic transfer;
- technogenic transfer.

Transfer of radionuclides with river waters is the major migration contributor. Annual carry-over for Sr-90 now comes to 235 - 300 Ci, which is 0.5-0.9% of the total Sr-90 inventory in the Zone. Cs-137 annual migration by river waters comes up to 30 Ci, or 0.03-0.05% of it's total inventory. Maximum carry-over of radionuclides takes place in the early spring (spring floods).

Resuspension and air transfer is of secondary importance now. Outside the Zone daily fallout of nuclides with precipitation and other atmospheric mechanisms comes to 0.1-1mkCi/sq.km. For reference, mean activity of global fallouts comes to 0.00019 Ci/sq.km. Growth of nuclide concentrations in air corresponds to seasons with preveiling dry periods - summer, early winter (insignificant). Also significant growth of airborne nuclides concentration takes place during forest fires in highly contaminated areas of the Zone. Thus, during major fire 27 - 31 July, 1992 near Euriakovka - Chystogalivka, concentration of airborne radionuclides increased up to  $4.8 \cdot 10^{-15}$  Ci/l (two orders up the normal magnitude).

It should be noted, that forrestation considerably reduces both spreading modes.

Contribution of other two modes is negligible. For comparison it is worth mentioning that due to factors of technogenic nature annual transfer is well within 1 Ci.

### Contamination of water resources

As it is clear now, the major part of the radioactive inventory is being spread through the Pripjat river run-off into the Dnipro.

In the cooling pond of the Chernobyl NPP with area of 22.9 sq.km and water inventory of 160 mln. cub.m. Sr-90 concentration in water is 200 pCi/l, in bottom sediments there is some 2,500 Ci Cs-137, up to 800 Ci Sr-90, and 2 Ci of different Plutonium isotopes.

In river Pripjat near Chernobyl average concentrations of Sr-90 and Cs-137 during year is within 15 pCi/l and 5 pCi/l, respectively (1992). Substantial growth of nuclides concentration takes place during floods (early spring, or summer, autumn in rainy years). Thus, in a very rainy summer this year Sr-90 concentration in river Uzh reached 29 pCi/l near village Cherevach, and 50 pCi/l - near Chernobyl (at the outlet).

Character of nuclide distribution in bottom sediments of Kyiv reservoir is shown in Fig. 3. Total inventory of radionuclides in series of reservoirs of the river Dnipro is shown in Fig. 4.

### Ground waters

In the close proximity to Chernobyl NPP site, contamination of ground waters with Sr-90 falls within range of 300 - 2700 pCi/l.

Concentration of Sr-90 in eocenic aquifer within 5-km zone is within 1 - 15 pCi/l.

Radioactivity of drinking water used in various points of restricted Zone, extracted from senoman aquifer at the depth of 150 - 230 m, falls within 1 - 16 pCi/l, with prevailing

mean figure of 2 pCi/l (prescribed limit for drinking water is 100 pCi/l). There is evidence that this contamination could take place during boring of the wells early after Chernobyl accident.

#### 4. ENVIRONMENTAL CONTAMINATION OF TECHNOGENIC NATURE

Regular investigation of the environmental contamination of technogenic nature, coming as a result of industrial, scientific and other kinds of human activity is in a very initial stage in Ukraine. As it was already mentioned, this process was stimulated by Chernobyl accident contamination.

Until 1989 nearly all surveys were directed to the detalisation of the Chernobyl accident consequences. In 1989 there were adopted the separate programme for population centers survey, which was aimed to identification and remediation of lost radioactive sources and sites contaminated as a result of antropogenic activity. This survey programme is implemented by prospecting geological organizations within the Ministry for Geology. The first stage comprises 20 population centers of Ukraine, involving those with uranium milling and mining industry, major industrial and scientific centers having more than 100,000 inhabitants, and largest recreation and resort centers.

The second stage of the programme includes also industrial centers with population within 50 - 100,000 inhabitants. The status of the Population Centers survey is shown in Fig. 8. The comprehensive list of population centers to be surveyed is given in Table VI. There reflected also minimal essential planning information and availability of the current results, which are given in Section 5.3 in more detail.

##### 4.1. Overview of the survey techniques

The technique used for radiological survey of the population centers consists of the following elements:

- 1) Vehicle-born gamma monitor screening (VEGMS).
- 2) Hand-held gamma/beta detector survey (HHGES).

## 2) Optional sampling for detailed laboratory analysis

Vehicle-borne monitor system consists of the "NEVA" type spectrometer with two NaI detectors. Measurements are performed in 4 channels, one of which is integral for sensing gamma within the energy band of 0.3 - 2.0 Mev. Other two are nuclide-specific for characteristic Cs-137, Ra-226 and Co-60 energy spectrums. Thus, the monitor is designed for detecting most probable contaminants of technogenic origine.

Information is registered at the graphic recorder, where individual curves from each channel are reproduced. Quality control of the measurements were performed in two ways: first - by surveying reference rout every day, and regular measuring of the reference sources with definite content of Co-60, Cs-137 and Ra-226, second - by independent follow-up survey of 5% already done routs using different vehicle unit. According to data of independent measurements, mean square root error of such measurements is within 1.1 mkrem/h. Measurement records are then quantified and mapped with a gradient move of 5 mkrem/h. Anomalies detected are to be obligatory surveyed using field walk-downs with hand held gamma/beta detectors.

Detailed field gamma/beta detector survey was performed in a regular grid of 20\*10 m using hand-held detectors CPH-68-01 (deviations detection), ДРТ-01Т (gamma dose-rate measurement), and radiometer "Прогноз" (express-diagnostics).

Detector CPH-68-01 is equipped with head phones allowing operator immediately detect the deviation and it's boundaries, which then is measured by ДРТ-01Т, the certified gamma dose-rate meter. Dose rate measurements are performed also in the noddles of the regular 10\*20 survey grid. Radiometer "Прогноз" ("Prognos") is measuring beta flux up to 5000 beta/sq.cm\*min and gamma dose-rate within 0 - 5000 mkrem/hour, and is used for immediate approximate characterisation of typical contaminations, each having more less constant characteristic gamma-beta ratio value. When nature of anomaly is unclear, samples are taken for standard laboratory spectrometry analysis. The typical example of the survey map is given in Fig. 7 (in surface contamination units).

#### 4.2. Current results of the survey programme

Dnipropetrovsk region. Population: 2 936 400 inh.  
Area: 21 900 sq.km

1. Dnipropetrovsk: Population - 1 185 500 inh.  
survey period: 1992-1996  
survey type: hand held walk-downs

Current results: Within boundaries of the city during initial stage of the survey there were identified several thousands of contaminated spots of various dimensions and gamma dose-rate levels. Most of them are uranium ores pieces and powder along the railway tracks having dispersed during uranium ore transportation operations. Mean levels of gamma-dose, measured at the height of 1 meter over the spot, are within the range of 100 - 5000 mkrem/hour.

In addition, there was identified so-called **Taromska anomaly** with 2.8 km long and mean gamma dose-rate levels within 3000-4000 mkrem/hour, and some spots having gamma dose-rate levels from 0.001 to 1 rem/hour. This contamination

was caused by two unsealed radioactive Caesium-137 sources, which were finally retrieved during relevant decon operations performed by local civil defence. The dose-rate levels in the immediate vicinity of the residuals of the sources were of 4 and 30 rems per hour, respectively. It was not identified, which amount of Caesium-137 was dispersed all over the place (Fig. 8).

2. Dniprodzerzhinsk: Population - 287 300 inh.  
survey period: 1990-1992  
survey type: vehicle born gamma-monitor screening,  
hand held gamma dose-rate walk-downs

Results: Within boundaries of the city, including its industrial area, there were identified 381 contaminated spots with areas from 1 to 7000 sq.meters and gamma dose-rate levels within 40 - 4700 mkrem/hour.

176 of these spots are situated at the territory of local metallurgical works, and their contamination is caused by scattered uranium ore pieces, metallurgical slag, gravel, brick debris, concrete, asphalt, contaminated soil and trash.

Other contaminated spots are situated in the populated regions of the town, and have been caused by utilisation of wastes of the local metallurgical industry for building purposes (communal building foundations, roads, private houses walls, etc.).

2. Kryvyj Rig: Population - 737 800 inh.

survey period: 1992-1994

survey type: vehicle born gamma-monitor screening,  
hand held gamma dose-rate walk-downs

Current results: To the date there have been surveyed 120 sq.km of the municipal territory. As a result there were identified 321 contaminated spots (radioactive anomalies) with a gamma dose-rate levels of 50 - 5000 mkrem/hour. 270 out of these have dose-rate levels more then 100 mkrem/hour. All these anomalies are the result of utilisation of the gravel and gangue from the exhausted uranium deposit in the local civil building industry (communal building foundations, roads, private houses walls, etc.).

Donetsk region. Population: 5 265 800 inh.

Area: 26 500 sq.km

1. Donetsk: Population - 1 121 200 inh.

survey period: 1991-1992

survey type: vehicle born gamma-monitor screening,  
hand held gamma dose-rate walk-downs

Results: There were identified 2 spots contaminated with Caesium-137, one of which had the length of 50 m and gamma dose-rate level of 500 mkrem/hour, and the other - of irregular shape and 410 mkrem/hour.

2. Makiyivka: Population - 424 000 inh.

survey period: 1992-1995

survey type: vehicle born gamma-monitor screening,  
hand held gamma dose-rate walk-downs

Current results: Using vehicle born gamma-monitor screening, up to date there were identified 8 insignificant anomalies of technogenic origin with gamma dose-rate levels of 50 - 150 mkrem/hour.



Kyiv region. Population: 1 937 900 inh.  
Area: 26 500 sq. km

2. Kyiv: Population - 2 646 100 inh.  
survey period: 1988-1990  
survey type: vehicle born gamma-monitor screening,  
hand held gamma dose-rate walk-downs

Results: There were identified 13988 contaminated spots of the Chernobyl nature and 7 contaminated spots of the technogenic origin.

Distribution of gamma dose-rate levels of the Chernobyl spots represents the following: gamma dose-rate levels of 75% spots falls below 65 mkrem/hour, of 23% - 65-120 mkrem/hour, and of 1.7% - more then 120 mkrem/hour.

Contaminated spots of technogenic origin had gamma dose-rate levels within 0.26 - 65 mrem/hour, and represented lost radioactive sources filled with Ra-226, K-40, or Caesium-137. In two cases the isotopic content was not identified.

Kirovograd region. Population: 1 251 800 inh.  
Area: 24 600 sq. km

1. Kirovograd: Population - 291 400 inh.  
survey period: 1990-1992  
survey type: vehicle born gamma-monitor screening,  
hand held gamma dose-rate walk-downs

Results: There were identified 800 contaminated spots, 23 of which having GDR exceeding 1000 mkrem/hour, 106 have GDR within 100 - 999 mkrem/hour, the rest - within 45 - 100 mkrem/hour.

All these anomalies are the result of utilisation of the uranium mining wastes from the local uranium reserves in the local civil building industry (communal building foundations, roads, private houses walls, etc.).

Lviv region. Population: 2 776 900 inh.  
Area: 21 800 sq.km

1. Lviv: Population - 810 000 inh.  
survey period: 1992  
survey type: vehicle born gamma-monitor screening,  
hand held gamma dose-rate walk-downs,  
grid of 50\*50m

Results: There were identified 3 radioactive sources of Co-60, with a GDR of 3-5 rems per hour. Because this survey had been performed using non-standard grid of 50\*50m, additional survey is needed, with a standard grid of 10\*20m.

Odessa region. Population: 2 639 200 inh.  
Area: 33 300 sq.km

1. Odessa: Population - 1 086 700 inh.  
survey period: 1989 - 1991  
survey type: vehicle born gamma-monitor screening,  
hand held gamma dose-rate walk-downs,

Results: There were identified 1611 deviations in gamma-dose rate with levels of 30 - 120 mkrem/hour, and 24 deviations with levels exceeding 120 mkrem/hour. 9 of these 24 cases are of the natural origin (utilisation of building materials with an increased natural radioactivity), 15 - contaminated spots of Chornobyl origin (after cleaning of contaminated cars and utilisation of contaminated peat from affected regions of Ukraine). Six cases are of technogenic nature, containing Ra-226.

Kharkiv region. Population: 2 182 100 inh.  
Area: 31 400 sq.km

1. Kharkiv: Population - 1 615 000 inh.  
survey period: 1992 - 1996  
survey type: vehicle born gamma-monitor screening,  
hand held gamma dose-rate walk-downs,

Current results: Survey works are in initial stage. Still up to date there were identified more then 600 deviations, including 2 radioactive sources with GDR about 1 rem/hour.

## E. URANIUM MILLING AND MINING INDUSTRY

### E.1 General description.

Uranium milling and industry is concentrated in Kirovograd, Dnipropetrovsk and Mykolaiv regions of Ukraine. Geologically it is the center of Ukrainian crystalline sheet. Territory is relatively smooth, with developed network of small rivers, creeks and natural pits. It is a region of highly productive agriculture and most valuable soils - chernozem type with thickness of 0.2 - 1.2 m. Region is densely populated and has developed mining and metallurgical industry. Being reach with various mineral resources.

In total, in Ukraine there are 21 explored uranium deposits. Significant volumes of tailings and depleted uranium ore waste piles have been accumulated at three sites during industrial scale mining for twenty-twenty five years. Zhovti Vody, Kirovograd and Smolino mines perform mining in rocks at depth of 300-500 to 1500-2000 m (Zhovti Vody). The in-situ leaching technology of uranium output was applied at two other sites, Devladdovo and Bratske deposits, causing significant contamination of ground waters and transformation of uranium into movable forms. All milling and mining activities are performed by state-owned Scientific & Industrial Consortium "Eastern Mining & Enrichment Combinat" (NVO "SkhidGZK").

Zhovti Vody mining region has one uranium mine which is already exhausted, hydrometallurgical mill plant for converting uranium ore to  $UO_2$ , and number of supporting services. Contamination sources are mining ventilation, uranium mill tailings, mining drainage waters with elevated content of natural radionuclides. There are 2 mill talings at the Zhovti Vody, one of which is out of operation and partly restored, other is in operation. Mines, mill plant and mill tailings are situated in close proximity to living area.

Kirovograd mining region consists of three mines. All mines are situated in town of Kirovograd and its outskurts.

Smolino site is 3 km from town Smolino, Kirovograd region. Sources of radioecological contamination at both sites are very much the same as at Zhovti Vody mining region, except of mill tailings.

Devladove in-situ leaching site is situated 30 km South-East from Zhovta Vody. Uranium reserves in buchak paleogen depositions were produced by underground leaching of the ore depositions. Technological solutions of 10-50 g/l of sulphuric acid and 2 g/l of nitrogen acid were injected through the system of injecting wells into uranium-bearing formations, and recovered through special recovery wells. The same scheme was used for Bratske in-situ leaching site. This practice was stopped in early eighties. There is no definite information about extent and effectiveness of the rehabilitation measures performed after that. Both sites are situated 2.5 - 4 km from nearby settlements, and impact of in-situ practice to hydrogeological medium require further investigation.

In total uranium milling and mining industry occupies 5530 gectars of land, 1340 ga are damaged. The structure of affected lands is summarised in **Table VI**.

#### 5.1. Radiological Characterisation Summary

Main contamination sources at Kirovograd site here are waste rock piles. General information is given in **Table VI**.

Prevailing contamination path is natural leaching of waste rock piles with rains and snow melting waters. **Fig. 9-10** shows processed curves of nuclide content and alpha-activity in 1-meter layer of soil at the site of relocated uranium ore stock. **Table VIII** contains reference figures for samples taken at 10-20 m distance from this former stock. Comparison of given data shows that at the vicinity of ore stock there are elevated quantities of U-238 and Pb-210 at the depth up to 1 m, while Po-210 - only at the surface. Thus, contamination of soil at the site of former stock exceeds 1 m depth, and in the adjacent area of 10-20 m width reaches 1 m depth.

**Table IX** gives results of radiological survey and soil sampling to identify area affected by waste rock piles at Kirovograd mining site. It shows that natural nuclide content in the vicinity of waste rock piles exceeds background concentrations for Ra-226 - up to 200 times, U-238 - 100, and for total alpha-activity - up to 30 times. Zone affected by piles was estimated extending to 100-150 m.

At Govt. Vody site the most powerful source of environmental contamination is represented by two mill tailings. Exhalation of Rn-222 and resuspension of dry tailing beaches provides two major mechanisms of environmental contamination. Rn-222 exhalation rate is 0.35-3.0 Eq/sq.mxs.

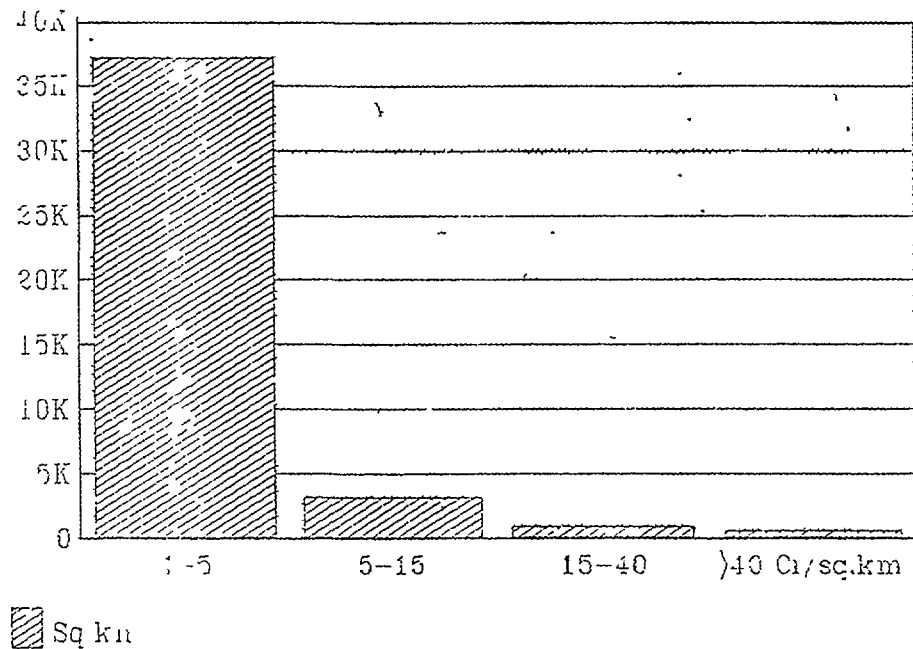


Fig. 1 Areas with different levels of contamination with Cs-137

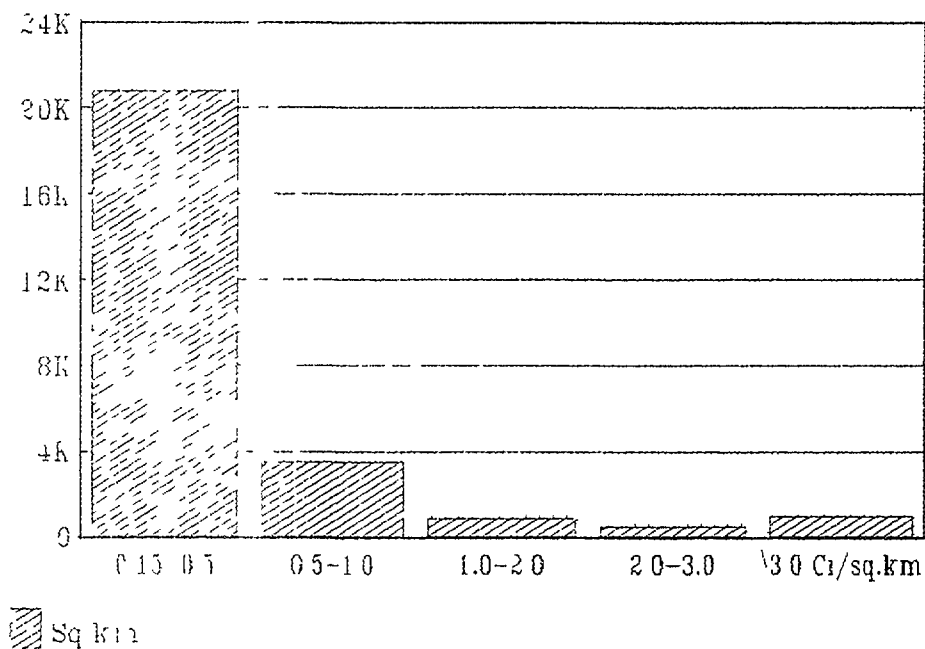


Fig. 2. Areas with different levels of contamination with Sr-90

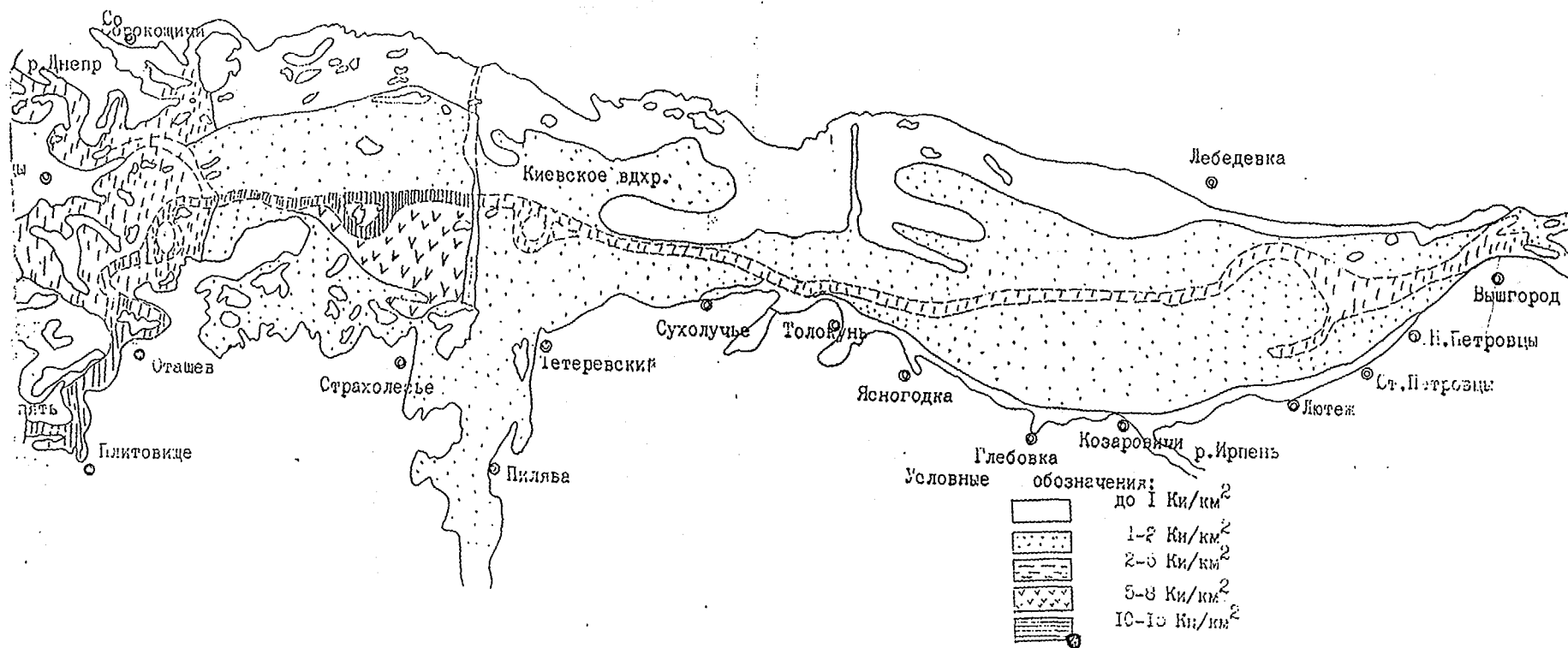


Fig.3. Distribution of Cs-137 in the bottom sediments of Kyiv reservoir

Measured zone of tailings influence for Ra-226 reaches 1200 m, and Po-210 - up to 1800 m (above background levels).

Average venting discharge of Rn-222 and it's daughters from mines of Zovti Vody comes to 2000 - 3000 Ci annually.

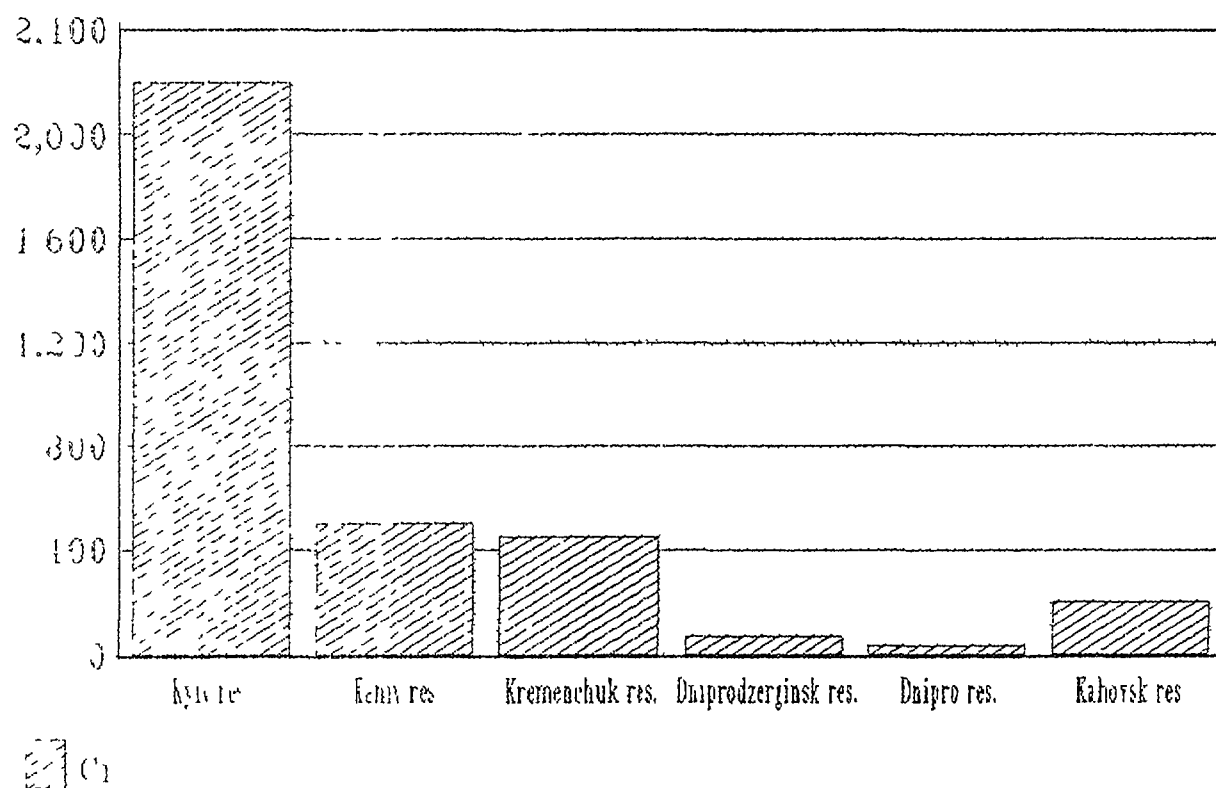


Fig. 4 Caesium-137 inventory in the bottom sediments of Dnipro river reservoirs

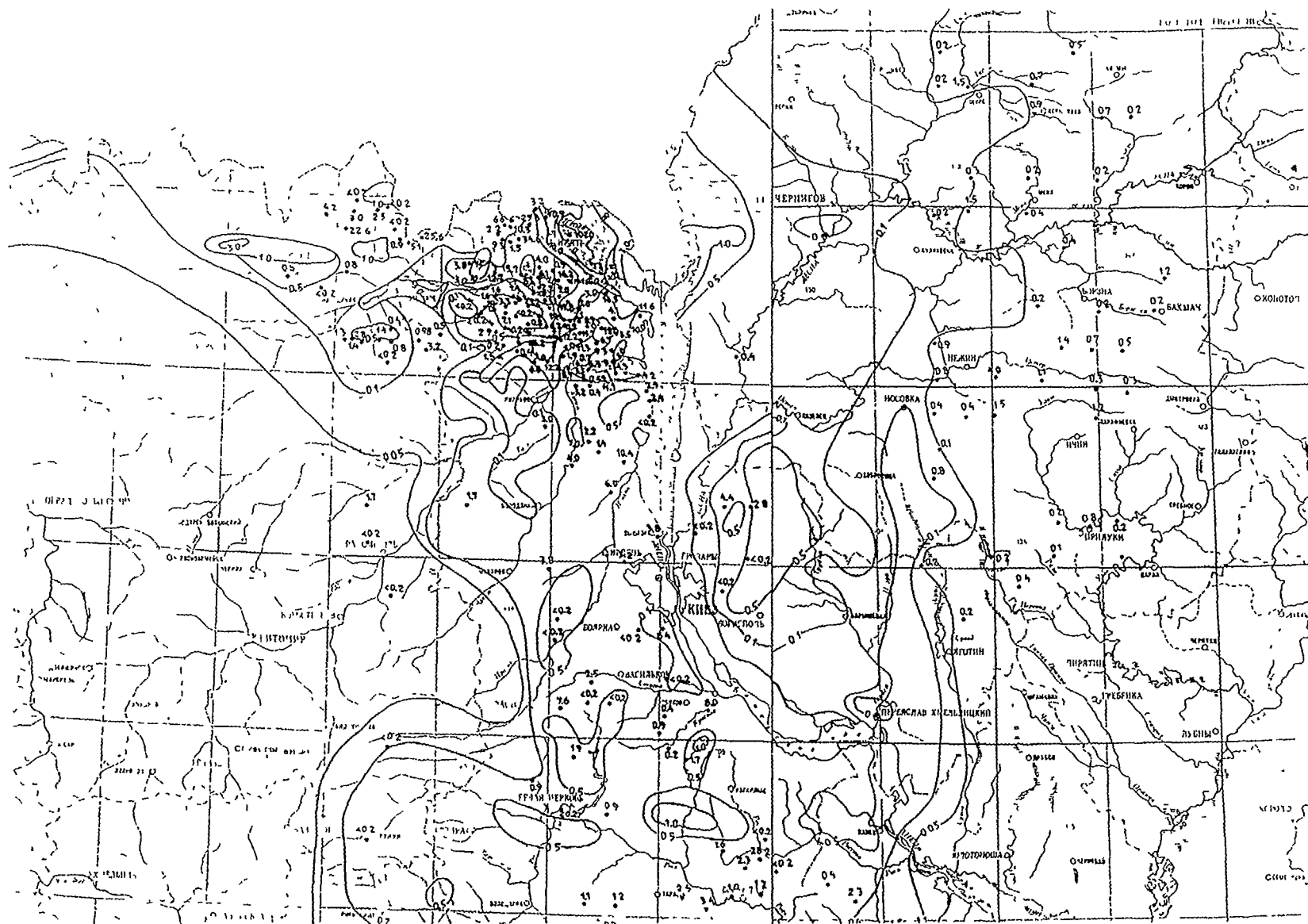


Fig. 5. Distribution of Sr-90 and Pu-239 in the northern part of Ukraine



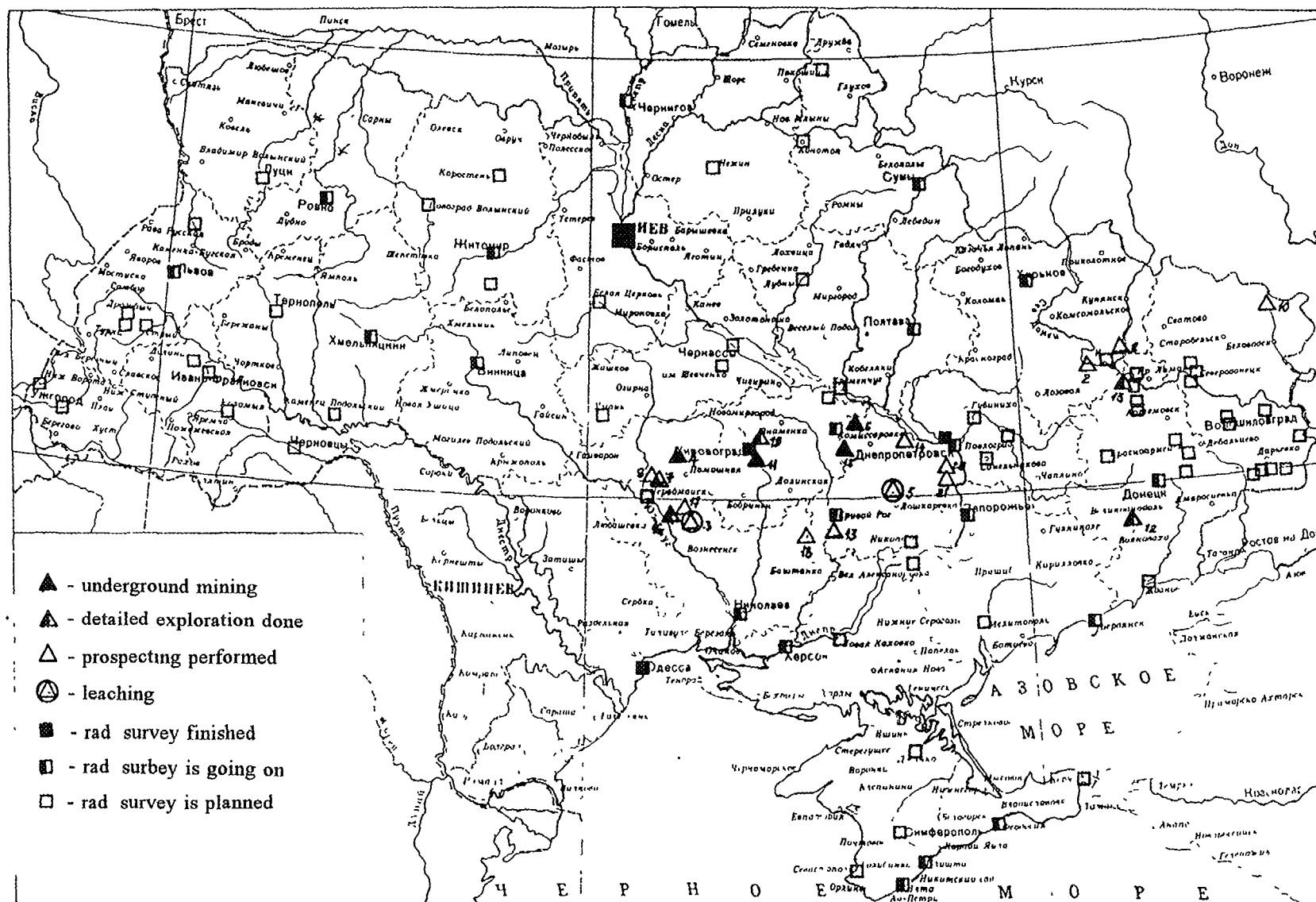
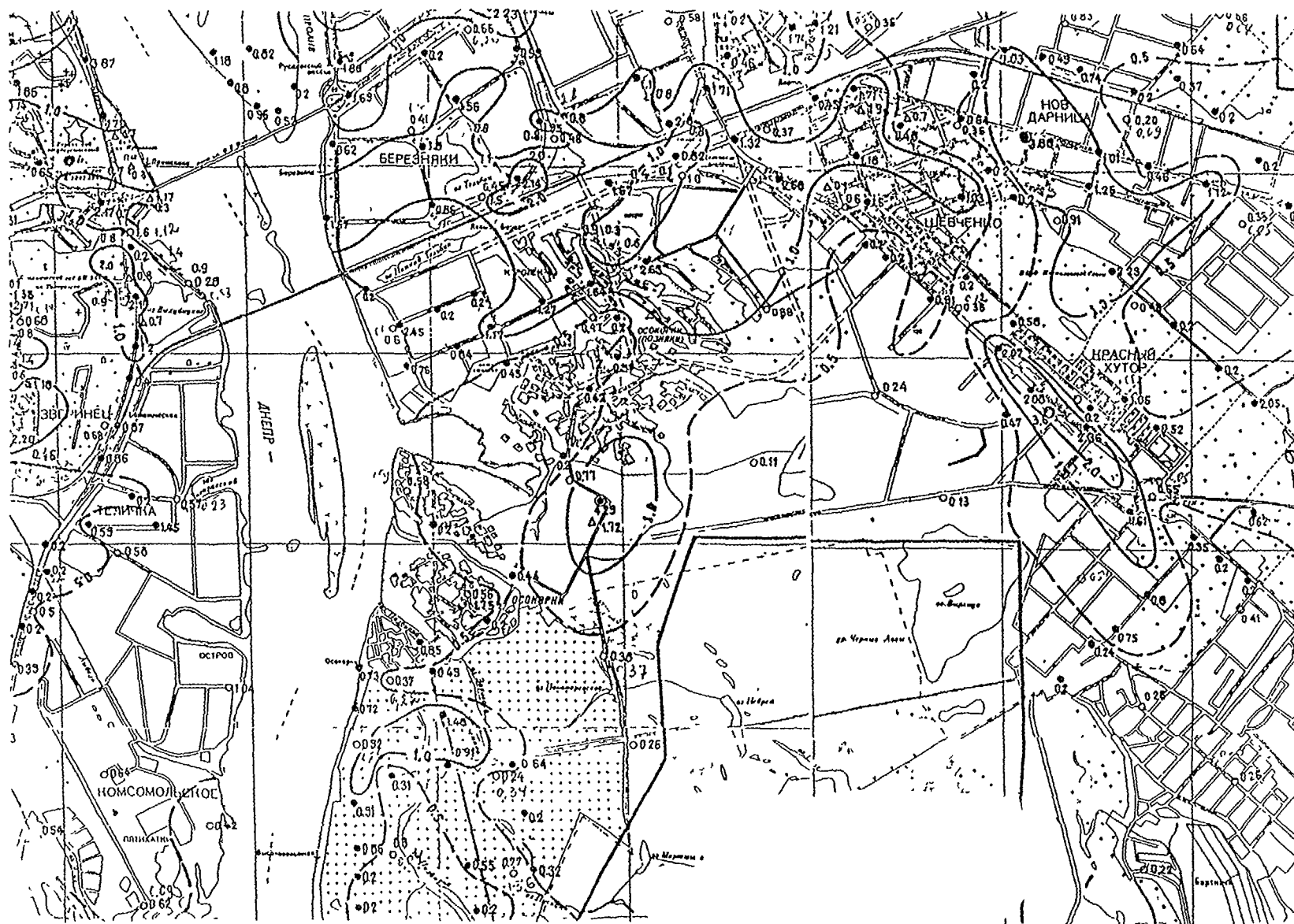


Fig.6. Status of uranium mining and of population centres survey programme



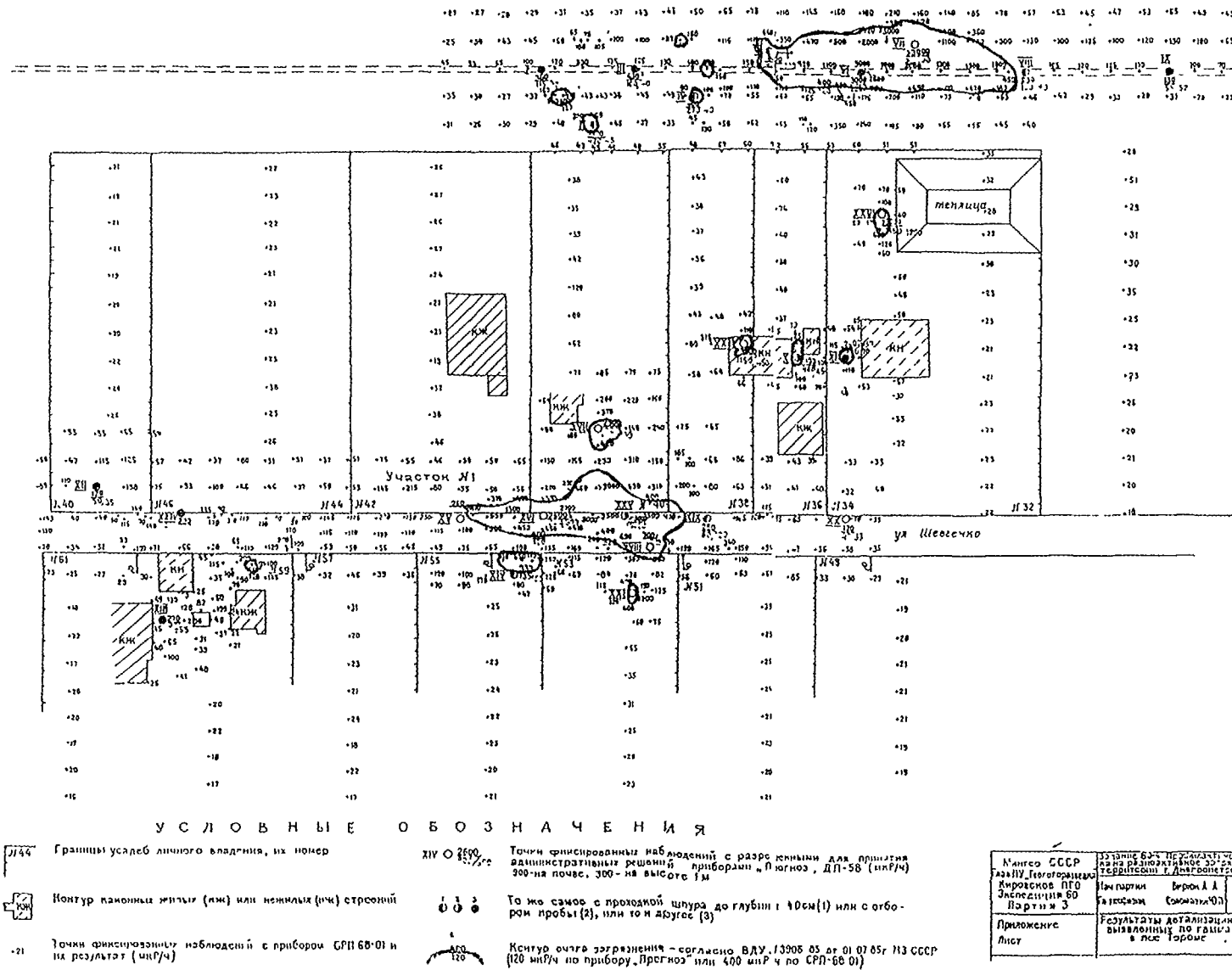


Fig.8. Taromska anomaly (contamination of private housing area by two unsealed Cs-137 sources)

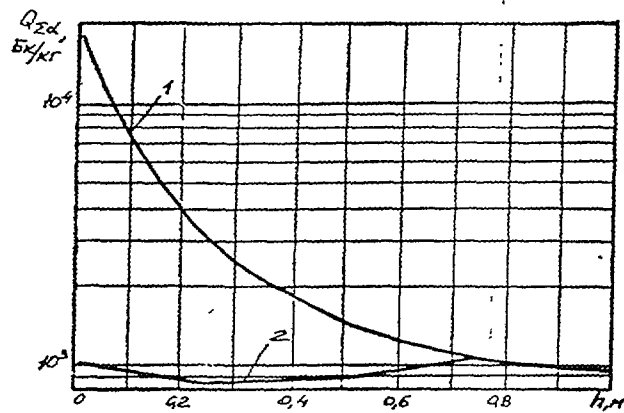


Fig.9. Alpha-activity of soil at the site of relocated uranium ore stock (1), and background curve (2)

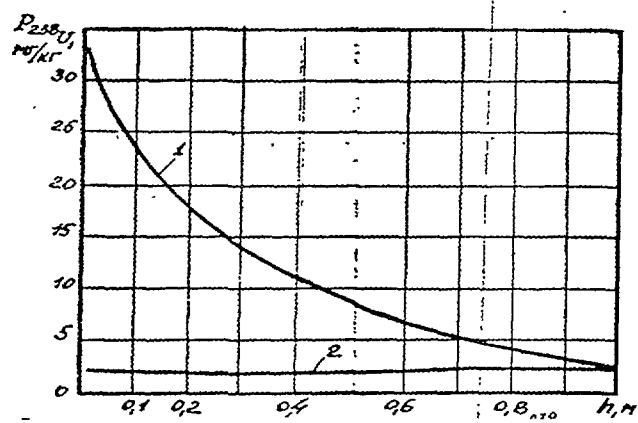


Fig.10. Uranium-238 content in a soil at the site of relocated uranium ore stock (1) and background concentration (2)

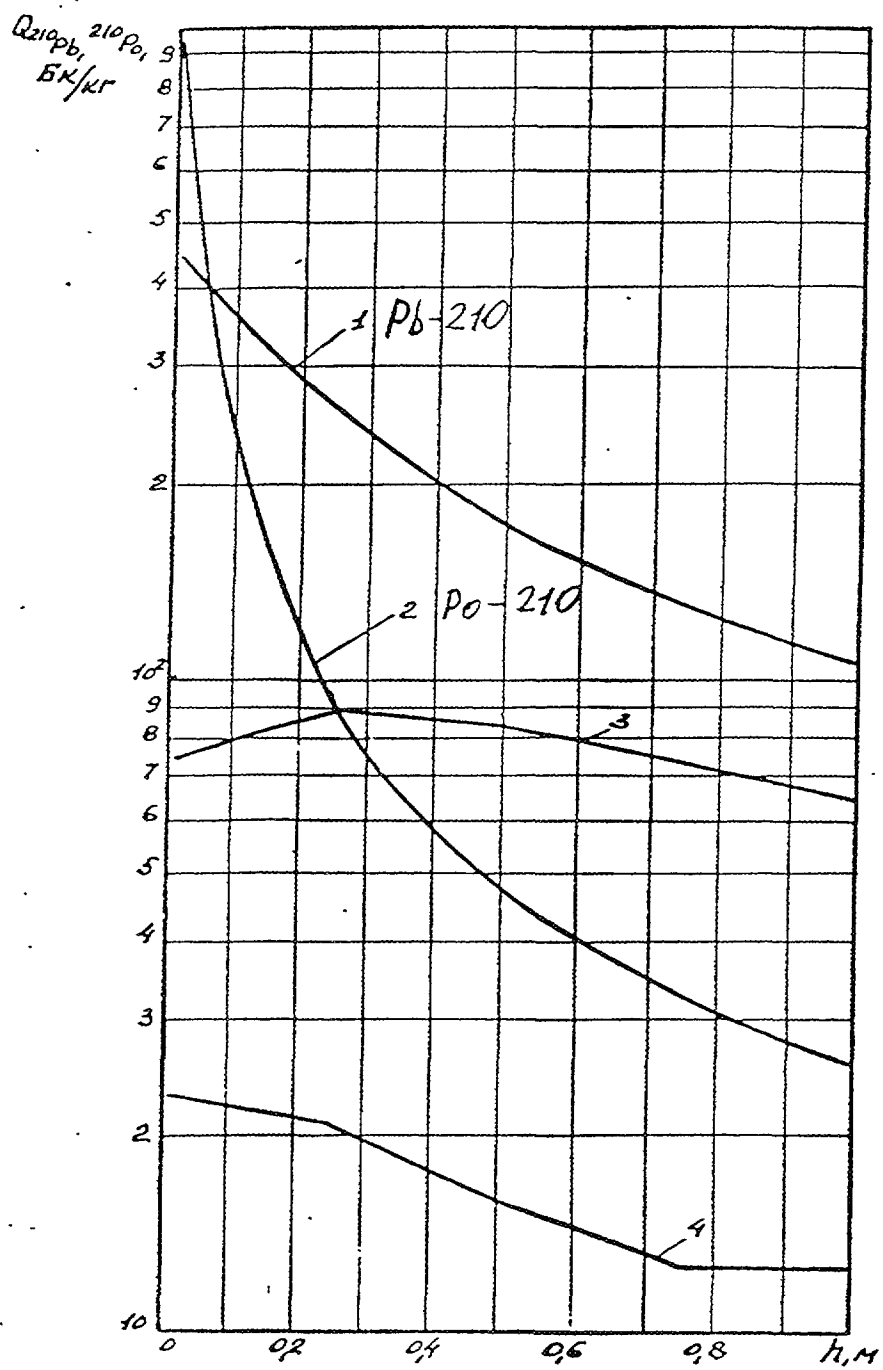


Fig.11.  $Pb-210$  and  $Po-210$  concentrations in a soil at the site of relocated ore stock, as a function of depth. (3), (4) - reference background concentrations

TABLE I. AREAS WITH DIFFERENT LEVELS OF CONTAMINATION WITH Sr-90 ACROSS THE AFFECTED REGIONS OF UKRAINE

Region of Ukraine	Contaminated area, sq. km					Sum
	Level of contamination, Ci/sq. km					
	0.15-0.5	0.5-1.0	1.0-2.0	2.0-3.0	3.0	
Vinnitsa	588	-	-	-	-	588
Zhitomyr	3643	275	187	31	3	4139
Ivano-Frankivsk	101	-	-	-	-	101
Kyiv	11597	2884	684	444	976	16585
Kirovograd	129	-	-	-	-	129
Rivne	19	-	-	-	-	19
Sumy	47	-	-	-	-	47
Khmelnysky	2	-	-	-	-	2
Cherkasy	2292	168	-	-	-	2460
Chernivtsi	321	-	-	-	-	321
Chernigiv	2027	181	-	-	-	2208
Total:	20766	3508	871	475	979	26599

TABLE II. AREAS WITH DIFFERENT LEVEL OF CONTAMINATION WITH Cs-137 ACROSS THE AFFECTED REGIONS OF UKRAINE

Region of Ukraine	Contaminated area, sq. km				Sum
	Level of contamination, Ci/sq. km				
	1.0-5.0	5.0-15.0	15.0-40.0	40.0	
Vinnitsa	1944	38	-	-	1982
Volynska	582	-	-	-	582
Dnipropetrovsk	38	-	-	-	38
Donetsk	410	-	-	-	410
Zhitomyr	9192	1780	336	154	11462
Ivano-Frankivsk	606	-	-	-	606
Kyiv	7695	957	546	417	9615
Kirovograd	219	-	-	-	219
Mykolayiv	24	-	-	-	24
Odessa	27	-	-	-	27
Rivne	9332	181	-	-	9513
Sumy	491	-	-	-	491
Ternopil	357	-	-	-	357
Kharkiv	16	-	-	-	16
Khmelnysky	318	-	-	-	318
Cherkasy	3233	72	-	-	3305
Chernivtsi	500	14	-	-	514
Chernigiv	2221	135	-	-	2356
Total:	37205	3177	882	571	41835

TABLE III. NUMBER OF POPULATION CENTERS IN AREAS WITH DIFFERENT LEVEL OF CONTAMINATION WITH Sr-90

Region of Ukraine	Number of contaminated population centers					Sum
	Level of contamination, Ci/sq. km					
	0.15-0.5	0.5-1.0	1.0-2.0	2.0-3.0	3.0	
Vinnitsa	39	3	-	-	-	42
Volynska	7	-	-	-	-	7
Dnipropetrovsk	2	-	-	-	-	2
Donetsk	4	-	-	-	-	4
Zhitomir	123	29	17	1	3	170
Ivano-Frankivsk	19	1	-	-	-	20
Kyiv	412	73	18	4	4	511
Kirovograd	9	1	-	-	-	10
Rivne	22	1	-	-	-	23
Sumy	3	1	-	-	-	4
Ternopil	6	-	-	-	-	6
Khmelnitsky	9	-	-	-	-	9
Cherkassy	91	16	2	-	-	109
Chernigiv	110	10	1	-	-	121
Chernivtsi	25	1	-	-	-	26
Total:	881	136	38	5	4	1064

TABLE IV. NUMBER OF POPULATION CENTERS IN AREAS WITH DIFFERENT LEVEL OF CONTAMINATION WITH Cs-137

Region of Ukraine	Number of contaminated population centers				Sum
	Level of contamination, Ci/sq.km				
	1.0-5.0	5.0-15.0	15.0-40.0	40.0	
Vinnitsa	74	-	-	-	74
Volynska	19	-	-	-	19
Dnipropetrovsk	2	-	-	-	2
Donetsk	46	-	-	-	46
Zhitomir	559	82	9	-	660
Ivano-Frankivsk	35	-	-	-	35
Kyiv	301	32	7	1	341
Kirovograd	15	-	-	-	15
Lugansk	2	-	-	-	2
Mykolayiv	3	-	-	-	3
Odessa	3	-	-	-	3
Rivne	245	8	-	-	253
Sumy	26	-	-	-	26
Ternopil	33	-	-	-	33
Khmelnitsky	26	-	-	-	26
Cherkasy	116	3	-	-	119
Chernigiv	87	4	-	-	91
Chernivtsi	30	1	-	-	31
Total:	1632	130	16	1	1779

TABLE V. LEVEL OF CONTAMINATION IN DIFFERENT PARTS OF ZONE (Ci/sq.km)

Monitoring area	Caesium-137	Stroncium-90	Plutonium-239/240
t. Pripjat	1.5 - 52	0.85 - 2.3	0.42 - 0.95
t. Chornobyl	0.15 - 24	0.25 - 16	0.01 - 0.36
5-km zone	4.5 - 1800	3.2 - 600	0.05 - 20.4
5-30-km zone	0.06 - 150	0.24 - 48	0.005 - 3.15
* RWDP "Buriakivka"	0.05 - 19	0.24 - 45	0.002 - 0.14
* RWDP "Pidlisnyj"	1.9 - 580	2.7 - 490	0.04 - 2.44

\* radioactive waste disposal point



TABLE VI STRUCTURE OF AFFECTED LANDS AT URANIUM MINING AND MILLING  
REGIONS IN UKRAINE

Mining site	Licensed industrial usage area, ga	Area of affected lands, gectares							usage coefficient
		Total	including definit kinds of damage						
			Waste rock pile	pits	open casts	industr. sites	mill tailings	others	
Zhovti Vody site	4050	968,3	19,1	17	50,6	202	645,6	34	0,23
Kirovograd site	500	52,4	7,4	-	-	45	-	-	0,105
Smolino Site	644,5	40,1	2,4	-	-	27,7	-	-	0,062
Devladovo site (in-situ leaching)	150,5	123,2	-	-	-	-	-	123,2	0,818
Bratske site (in-situ leaching)	182,5	152,5	-	-	-	-	-	152,5	0,836
Total	5527,5	1336,5	28,9	17	50,6	284,7	645,6	309,7	0,242

TABLE VII. GENERAL CHARACTERISTICS ON THE WASTE PILES AT THE KIROVOGRAD SITE

Number of waste pile	Waste rock volume, cub. m	Waste pile area, sq. m	Mean dose rate mkZv/h	Mean Ra-226 concentr. Bq/kg
1	215	23688	1,05	1191
2	448	26324	1,15	1325
3	160	15525	0,8	907
4	132	16715	0,83	939
5	410	19253	0,98	1130
6 (operated)	250	15341	1,20	1389
7 (operated)	400	37512	0,91	1095
Total	2115	154389	0,99	1139

TABLE VIII. CHARACTERISTICS OF SOIL SAMPLES TAKEN AT URANIUM ORE STOCK

Layers of soil samples m	Total activity of soil samples Bq/kg	Content of natural nuclides in soil samples									
		U-238 mg/kg		Th-230 Bq/kg		Ra-226 Bq/kg		PB-226 Bq/kg		Po-210 Bq/kg	
Surface	1260 630	7,8	2,4	37	13	17	7	214	111	98	59
0,20 - 0,30	960 300	6,6	2,2	40	28	26	13	178	95	24	11
0,45 - 0,55	780 110	5,2	2,1	33	21	15	14	148	52	21	13
0,70 - 0,80	670 260	5,2	1,9	33	20	20	9	130	55	15	5
0,95 - 1,05	700 150	4,3	2,0	30	17	14	8	96	30	16	6

TABLE IX. RADIATION SURVEY RESULTS FOR KIROVOGRAD SITE

Survey point	Dose rate mKSv/h	Soil activity Bq/kg	Range of natural nuclide content in soil	
			U-238 mg/kg	Ra-226 Bq/kg
Area adjacent to waste pile No. 5	0,11 - 0,31	570 - 7660	1,8 - 60	30 - 1266
Area adjacent to waste pile No. 5	0,18 - 0,33	850 - 1200	2,65 - 110	46 - 2217
Area adjacent to waste piles No. 2,3	0,13 - 0,45	850 - 3660	1,5 - 7,2	38 - 210
Area adjacent to waste pile No. 7	0,14 - 0,40	740 - 3890	1,5 - 41	50 - 1370
Area adjacent to waste pile No. 1,4	0,14 - 2,30	2900 - 77700	2,9 - 220	50 - 10830
Area adjacent to uranium ore stock	0,13 - 1,01	850 - 38900	5,25 - 240	82 - 4310

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## REFERENCES

1. "Radiological Characterisation of the territory of Ukraine contaminated as a result of technogenic activity" special report of the State Geological Enterprise "Kirovgeology", Kyiv, September 1993.
2. "Radiological Characterisation of the territory of Ukraine contaminated as a result of the Chernobyl Accident", special report of the State Committee for Hydrometeorology, Kyiv, September 1993.
3. "Restricted Zone Radiation Monitoring Data for 1993", report of the Scientific Productive Unit "PRIPYAT" of the restricted zone, Pripjat, 1993.
4. Soroka Y.N., Molchanov A.I., Isayeva N.E., Restoration in Uranium Milling and Mining Industry in Ukraine.

# THE IDENTIFICATION AND RADIOLOGICAL CHARACTERISATION OF CONTAMINATED SITES IN THE UNITED KINGDOM

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## Abstract

The United Kingdom has a large number of sites on which radioactive materials are used and hence where there is potential for radioactive contamination to occur. It has several power generating reactors with supporting facilities for fuel enrichment and fabrication, spent fuel reprocessing, waste conditioning and low-level waste disposal. It has a nuclear defence capability with a submarine nuclear propulsion programme. It is also a major producer and supplier of radioisotopes and associated products. This paper briefly addresses the types, nature and extent of contamination at these sites and the approaches to characterising the contamination.

The problems of radioactive contamination of sites in the UK have generally been very much more limited in number and scale than those experienced in the USA, the former USSR and many of the other countries of Central and Eastern Europe. This has been due to a strict regulatory and planning framework, the location and limited size of the major nuclear sites with the consequent need for more effective operational and waste management practices and the absence of uranium mining and nuclear weapons test facilities within the UK.

Under UK law (Radioactive Substances Act) all sites using or storing radioactive materials with certain limited exceptions must be registered. Currently there are over 10,000 premises and sites so registered. Many of these sites are potential sources of releases of activity. Discharges of radioactive wastes to the environment may only be made, subject to certain limited exemptions, in accordance with authorisations issued under that act. These sites are regulated by Her Majesty's Inspectorates of Pollution (HMIP) in England and Wales and Industrial Pollution (HMIP) in Scotland and Northern Ireland.

The main types of sites have been former luminising workshops and facilities for dismantling equipment containing luminous sources, chemical plants handling naturally radioactive ores, eg., for rare earth, detergent and fertiliser production, and leaks from tanks, process piping and active waste storage facilities, mainly at spent fuel reprocessing plants.

## 1. INTRODUCTION

The United Kingdom currently has a nuclear power generation programme with 7 advanced and 6 operating Magnox, gas-cooled, multi-reactor, power plants with a further one 1100 MW(e) PWR being completed. 3 commercial Magnox stations have been closed and are

currently undergoing the first stage of decommissioning. It has facilities to convert uranium concentrates into pure uranium tetra- and hexafluoride, for enrichment, fuel fabrication and spent fuel reprocessing. It has no uranium mining activities, although very low-grade, uranium-bearing ores do occur in certain parts of the country. It does have facilities for waste treatment and conditioning and has disposal sites for low-level wastes at Drigg and Dounreay. It has a nuclear weapons defence capability with establishments for constructing and assembling components of such weapons at Aldermaston, Burghfield and Cardiff. It has a nuclear propulsion programme with dockyards for refitting nuclear submarines. It also has manufacturing facilities for a very major supplier of radioisotopes and associated products, Amersham International.

## 2. REGISTRATION OF SITES USING RADIOACTIVE SUBSTANCES

It is a requirement of the law (Radioactive Substances Act, 1960) in the United Kingdom that all sites using radioactive materials must be registered. This act provided for controls to be exercised over the use and keeping of radioactive materials and the accumulation and disposal of radioactive wastes. This act and amendments by subsequent legislation, including Part V of the Environment Protection Act of 1990, have since been consolidated as the Radioactive Substances Act 1993. Currently, there are over 10,000 registered premises using and storing radioactive materials in the UK, some 8,000 of which are in England and Wales. These premises include:

- i) nuclear power plants, operated in England and Wales by the electrical utility, Nuclear Electric, and in Scotland by Scottish Nuclear;
- ii) fuel cycle facilities, including those for fuel enrichment, manufacture and reprocessing, operated by British Nuclear Fuels plc (BNFL);
- iii) research establishments, including major facilities for fuel cycle and nuclear power reactor development, operated by the United Kingdom Atomic Energy Authority;
- iv) defence establishments, including those which develop and manufacture nuclear weapons and others which refuel and refit nuclear submarines;
- v) radioactive waste disposal sites;
- vi) radioactive source manufacturers and distributors;
- vii) university and college laboratories;
- viii) hospitals;
- ix) various industrial users of radioactive sources and manufacturers of other products, which incorporate man-made or naturally occurring radionuclides. The latter include luminising and ionising sources, smoke detectors, lightning conductors, safety lighting, and processors and users of ores containing significant levels of radioactivity, such as the phosphate ores used to produce phosphoric acid and various detergents.

### 3. REGULATION OF FACILITIES USING RADIOACTIVE MATERIALS AND SOURCES OF IONISING RADIATION

Many of these facilities are potential sources of releases of activity. Discharges of radioactive wastes to the environment may only be made, subject to certain exemptions, in accordance with authorisations issued under the RSA. Certain premises are exempt from this requirement, notably those occupied on behalf of the Ministry of Defence. In these cases inter-departmental administrative arrangements exist whereby discharges are made in accordance with approvals which apply the same standards as authorisations. During 1992 there were about 1000 premises in England and Wales authorised to discharge radioactive wastes. The major sites were those licensed under the Nuclear Installations Act of 1965 and known as "nuclear sites". They include the nuclear power stations, nuclear fuel fabrication and reprocessing plants, research centres and isotope production centres. The other sites mainly consisted of hospitals, universities and industrial, research or manufacturing centres. Lists of these premises have been published [1].

These sites are regulated in England and Wales by Her Majesty's Inspectorate of Pollution, (HMIP), which is a regulatory branch of the Department (Ministry) of the Environment and in Scotland and Northern Ireland by Her Majesty's Industrial Pollution Inspectorate (HMIPI). HMIP and HMIPI are responsible for licensing all premises which keep or use radioactive substances, or accumulate or dispose of radioactive waste. They issue authorisations, which lay down limits for the amounts of radioactive material to be held, permissible release levels and operating standards. They also oversee compliance, carry out inspection visits and take enforcement action where necessary. The latter includes requiring site owners/operators to clean up land and facilities contaminated with radioactivity from whatever source. These regulatory bodies have enforcement powers, which include prohibition notices forcing the shut-down of processes and premises, fines and even imprisonment for staff, managers or directors found guilty of offences.

Information on radioactive substance authorisations are kept on national Public registers, which are held at the three main HMIP regional offices in Bedford, Bristol and Leeds. Additional copies of the information on sites and processes within their local areas are held by individual district councils.

### 4. EXTENT OF RADIOACTIVE CONTAMINATION ON SITES

The problems of radioactive contamination of land in the UK have generally been very limited and much smaller in both scale and number than those experienced in the USA, the former Soviet Union and many of the other countries of Eastern Europe. This is a result of:

- i) the regulatory framework described above and the associated inspectorate which enforces it;
- ii) the high population density and degree of land use within the UK, which has severely restricted the size of the major nuclear sites. As a consequence their operational and waste management practices have had to be much more rigorous than those of other major users of nuclear power and nuclear weapons states;

iii) a formal system of town and country planning, which has been implemented for the major part of this century and requires that there be careful public scrutiny of any proposed change of land use, new developments or major changes to existing facilities or their uses.

iv) location of almost all of the major nuclear facilities, including all of the nuclear power plants bar the now closed Trawsfynydd and the Sellafield and Dounreay nuclear fuel reprocessing plants, on the coast with the discharge of their treated effluents under authorisation into the sea. This approach has led to very marked reductions in the quantities of liquid wastes which have to be stored on site and hence the potential for leakage of mobile radioactive materials;

v) tight restrictions on sites which can accept radioactive materials for disposal;

vi) the absence of uranium mining facilities; and

vii) the absence of nuclear weapons test sites in the UK with sites formerly used being in Australia (Maralinga), USA (Nevada) and on the Monte Bello and Christmas Islands.

## 5. TYPES OF SITES WITH POTENTIAL RADIOACTIVE CONTAMINATION PROBLEMS

The main types of sites and problems which have led to contamination are:

a) former luminising workshops, primarily using  $^{226}\text{Ra}$ , which was treated as a natural rather than as a radioactive material and hence for many years was not tightly controlled. A number of such facilities, which were often situated in small commercial premises in cities and made luminous dials for aircraft and military vehicle instruments, as well as clocks etc, have been identified and decontaminated. Similarly, some minor contamination has resulted, particularly at military establishments and scrap yards, from the decommissioning of redundant military equipment fitted with luminous dials, markers, etc. Some contamination has also resulted at facilities using  $^3\text{H}$  as the luminising source.

b) storage areas for old Betalites (tritium-filled, phosphor-lined, glass capsules), which were very widely used in domestic and other telephones for dial illumination. These were collected when the telephones were phased out and many were stored in ISO transport containers until a disposal route could be identified. A number of these sources broke and some minor tritium contamination of the storage areas resulted.

c) the radioactive plume resulting from the Chernobyl accident crossed the United Kingdom whilst rainfall was occurring over Cumbria and North Wales. This resulted in precipitation of primarily  $^{137}\text{Cs}$  over these areas at areal concentrations of  $\sim 1\text{Ci/km}^2$ . Clean-up over the large ( $\sim 100\text{'s km}^2$ ) areas involved was not considered to be a practical or cost effective option and has been replaced by management controls, primarily restrictions on livestock movement and sale.

d) chemical plants producing phosphoric acid and various phosphates from naturally occurring, phosphate-bearing ores. Many of these ores contain Ra and Th and their daughters. In some cases the residues from the processing of such ores have been used as the hardcore

foundations for roadways and buildings on the sites and nearby or have just been dumped into spoil heaps on the sites. As a result, low-level contamination has occurred. This has required characterisation and then clean-up.

e) similar problems have occurred on other sites where naturally radioactive ores have been used. These include the smelting of metals, such as Pb, where during the process natural  $^{210}\text{Po}$  and  $^{210}\text{Pb}$  were liberated and discharged via a stack. In this case the radioactivity was characterised, but given the low levels of activity further action has not usually been considered necessary.

f) leaks from tanks, process piping and active waste storage facilities at spent fuel reprocessing plants. Some leaks, which have resulted in the contamination of immediately adjacent land, have occurred at Sellafield and Dounreay.

At Sellafield cracks occurred in 1976 in some of the concrete vaults (B38) used to store the active Magnox cladding removed from spent fuel prior to reprocessing. In the past water was added to these vaults to reduce the rate of  $\text{H}_2$  formation. This water became contaminated with the more soluble fission products, primarily  $^{137}\text{Cs}$  and  $^{90}\text{Sr}$ , associated with small fragments of spent fuel retained with the cladding. The water leaked through the cracks and contaminated the soil beside the silo. This necessitated characterisation and then decontamination by removal of the contaminated soil. Another leak was associated with a sampling annex on a line to some highly active liquor storage tanks. Over a period of years small quantities of liquor leaked out and contaminated the annex and the nearby soil.

Similar leaks from piping have occurred at Dounreay, leading to limited contamination of the neighbouring soil. At Dounreay problems have also arisen with a unlined storage vault which was cut into the rock underlying the site. This vault was used primarily for  $\beta\gamma$  wastes, although low-levels of  $\alpha$  activity were accepted. The lower portion of the vault became flooded and the resulting contaminated liquor has leaked through the fractured rock geology to contaminate the immediate foreshore behind the site, which is situated on the coast.

Beach contamination has also occurred in the past at Sellafield, when low-level, liquid waste was discharged under authorisation to sea through a pipeline. The liquor contained a small quantity of tributyl phosphate in odourless kerosene, reprocessing solvent. This solvent contained  $^{106}\text{Ru}$ . The discharge was made at a period when the sea was very calm and the solvent floated to the surface and was washed back onto the beach contaminating a stretch of over 1 km. This necessitated closure of the area, followed by characterisation and then removal for controlled disposal of the areas of contamination.

g) The use of inappropriate temporary waste storage practices. Some problems have arisen when activated and sometimes contaminated equipment has been stored either in open hard standing areas or inadequate stores. The result has been the spread of contamination to surrounding areas. Such problems have arisen at the naval dockyards at Faslane, Rosyth and Chatham, where the wastes from the refitting of nuclear submarines have been stored.

They have also arisen at Harwell where parts of the Southern Storage Area became contaminated as activity spread from redundant experimental rigs, which had been removed from research reactors. This site had also been used for low-level waste processing operations during the early years of development of the UK's nuclear power programme. As such there



was quite widespread, low-level contamination with fission products and actinides. This site required extensive characterisation and then radiological clean-up [3].

h) A number of laboratories at universities and research institutes, particularly those working with radioactive materials during the early years of nuclear energy have been contaminated to a limited extent and have required both internal decontamination and clean-up of adjacent soil, etc.

## 6. CHARACTERISATION OF RADIOACTIVELY CONTAMINATED SITES

The characterisation of radioactively contaminated sites has not generally been carried out to a prescribed standard in the UK to date. It is only recently that a draft British Standard [4] has been issued on the identification and investigation of any potentially contaminated site. Investigations have initially involved reviews of the past history of each site and almost without exception these reviews have identified the radionuclides of concern. Characterisation has then usually involved a combination of areal monitoring where appropriate and sampling of soil, plants, groundwater and surface water, etc., for analysis. Depending on the radionuclides present, the latter has involved  $\gamma$ -spectrometry,  $\alpha$ -spectrometry, radiochemical analyses for specific radionuclides and liquid scintillation counting. An example of this approach has been described in the clean-up of the Southern Storage Area at Harwell [3].

## REFERENCES

- [1] DEPARTMENT OF THE ENVIRONMENT. List of Premises in England and Wales currently authorised under the Radioactive Substances Act 1960 to dispose of Radioactive Waste. April 1988.
- [2] HER MAJESTY'S INSPECTORATE OF POLLUTION. HMIP Monitoring Programme. Radioactive Substances Report for 1992. June 1994.
- [3] FELLINGHAM, L. R., MAY, N. A. and SNOOKS, W. A. The Characterisation and Remediation of Radiological Contamination at the Southern Storage Area, Harwell, England. Second Int. Conf. on Environmental Restoration in Eastern Europe, Budapest'94. Budapest, Hungary, 1994.
- [4] BRITISH STANDARDS INSTITUTE. Code of Practice for the Identification of Potentially Contaminated Land and its Investigation. DD 175. British Standard. Draft for Development.

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