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

Volume 3: Technologies for, and the implementation of, environmental restoration of contaminated sites

Proceedings of a workshop held within the Technical Co-operation Project on Environmental Restoration in Central and Eastern Europe in Rež, Czech Republic, 12–16 December 1994

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PLANNING FOR ENVIRONMENTAL RESTORATION OF RADIOACTIVELY CONTAMINATED SITES IN CENTRAL AND EASTERN EUROPE: VOLUME 3
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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, Kazakhastan, 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

CONTENTS

Background Paper

TECHNOLOGIES FOR AND IMPLEMENTATION OF ENVIRONMENTAL RESTORATION PROJECTS

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Abstract

A great deal of experience is available related to the cleanup of small and medium sized land areas. A variety of techniques and equipment are available for the cleanup of contaminated areas and for the transportation and disposal of wastes arising from such cleanups. The selection of methods and technical procedures for environmental restoration will be governed by criteria such as:

- external dose rates and the mixture of radionuclides present;
- the nature of the location and of items requiring cleanup;
- mechanical properties of the materials requiring treatment;
- the availability of different methods of cleanup and the technical facilities for applying them;
- the availability of trained staff.

Although a great deal is known about such cleanups, further work is required, especially on the decontamination of urban areas. Most of the information provided in this paper is based on IAEA's Technical Report Series No. 300, Cleanup of Large Areas Contaminated as a Result of a Nuclear Accident, 1989.

1. INTRODUCTION

This paper represents the logical continuation of those presented at the Budapest and Piestany Workshops. It covers practical methods and techniques to decontaminate or rectify radioactively contaminated sites. As for previous project papers, the focus of this paper is on accident-contaminated sites. For uranium mining and milling waste, such as mill tailings and mining debris, different procedures will normally apply for environmental restoration. However, relevant information on how to decontaminate/dispose of contaminated materials from uranium mines/mills can also be found in this paper.

2. STABILIZATION OF CONTAMINATION

Following a serious nuclear accident which results in widespread contamination, the detriment to man from the radioactive contaminants can be reduced by the decontamination methods described below, by interdiction of the contaminated area or by using coatings to stabilize the contamination using the techniques described in this section.

The objective of using coatings to stabilize or immobilize radioactive contamination on soils, buildings, roads and equipment are to:

(a) Reduce the spread of contamination to clean areas.
(b) Reduce the airborne inhalation hazards.
(c) Decontaminate surfaces by incorporating the contamination in a removable coating.
(d) Reduce the volume of radioactive waste generated. If the contamination is in an area which does not contribute to radiation doses and it arises predominantly from relatively short lived radioisotopes, it may be desirable to stabilize and leave it to decay.

In most but not all cases, the application of surface stabilizers is a short term corrective action which would be followed by further decontamination. For uranium mining and milling waste, stabilization is a basic part of the environmental restoration project. It may include dewatering the tailings, building/repairing dams, covering the tailings and neutralizing generated acids.

A large number of stabilizers/fixatives are commercially available and these are generally classified as chemical, mechanical, physical or chemical with mechanical characteristics. The stabilizers are rated according to their:

- preferred applicability to various land types and land use classes
- hazard level
- durability
- application methods, and
- effect on vegetation recovery.

Chemical stabilizers are liquid or solid additives that alter the physical properties of the treated surface.

Mechanical stabilizers are used to physically cover the contamination without modifying the physical properties of the soil or surface. They include concrete and asphalt covers, manufactured materials like polyvinyl films or erosion control nets, sandbags and rock rip-rap.

Physical stabilization of soils can be carried out by using heat, electricity or cold to change the physical properties.

Another approach to stabilization is to combine it with shielding. For example, 5 cm of concrete would reduce the gamma radiation levels from ^{137}Cs by a factor of about 3 and would fix the contaminants. This could be a more cost effective solution for car parks or some roadways than removal and disposal of the contaminated surface, particularly if waste disposal sites are limited.

In urban areas, stabilization of contamination on areas which do not require decontamination and which will not be subject to weathering could be considered. For example, vertical building walls may have lower contamination levels than roof surfaces and may not need to be decontaminated. In this case stabilization of the contaminants by a polymer spray, painting, etc., would reduce resuspension from the surface and should also reduce additional contamination when the roof surfaces are washed down.

3. DECONTAMINATION TECHNIQUES AND EQUIPMENT

Decontamination of materials, equipment, buildings and sites to permit operation, inspection, maintenance, modification or plant decommissioning to be done safely has been an integral part of the nuclear industry since its inception. A large number of decontamination techniques and a large variety of chemical mixtures have been developed over the years to assist in removing contamination from all kinds of surfaces and these are continually being improved. These techniques also include means of decontaminating reasonably large areas of land which have been contaminated by mining/milling wastes, nuclear test fallout, etc.

To achieve a good decontamination factor (DF), a decontamination process must be selected on the basis of site specific considerations taking into account a wide variety of parameters such as:

- type of material: metal, asphalt, concrete, soil, wood, etc.
- type of surface: rough, porous, coated (paint, plastic, etc.)
- composition of contaminant: activation or fission products, actinides, etc.

- chemical and physical form of contaminant: solubility, aerosol, flocculent particles, complex compound with other materials, etc.; for many decontamination processes, the smaller the particle, the more difficult it is to remove from a surface
- the decontamination factor required
- the proven efficiency of the process
- the method of deposition; the distribution of the decontamination and its adherence to the surface can depend on whether the deposition was wet or dry.

Other factors which are important in selecting the method and equipment, but which do not affect the DF are:

- availability, cost and complexity of the decontamination equipment
- the need to condition the secondary waste generated
- occupational and public doses resulting from decontamination
- other safety, environmental and social issues
- availability of trained staff
- the amount of work involved and the difficulty in decontaminating the equipment used for the cleanup if it is to be reused.

In summary, the final decontamination process selected will depend on the best overall balance between the above factors to minimize the overall impact and net detriment to people using the most cost effective means.

In the following sections, the methods available for decontaminating buildings, equipment, roadways and large land areas are described.

3.1. Decontamination of Buildings, Equipment and Paved Surfaces

Much of the past decontamination experience at nuclear facilities relates to the cleanup of buildings, equipment and paved surfaces in or adjacent to nuclear reactors and other facilities during normal operations or decommissioning.

However, there has been less attention to the development of methods suitable for large scale application to urban areas and to urban construction materials following a nuclear accident. Many of the techniques suitable for nuclear plants and sites may be too expensive for application on the scale required in an urban environment. Accessibility and recovery of the radioactive wastes generated by decontamination procedures are also likely to present more difficult problems in an urban environment.

Decontamination methods range from simple physical cleaning techniques, including allowance for natural weathering, to fairly sophisticated physical and chemical procedures. Some of the methods described use industrial equipment such as road sweepers which are readily available in many industrialized countries and which can be operated by relatively unskilled personnel. In other cases specialized techniques such as pavement grading and sand blasting require skilled personnel and special consideration of airborne contamination problems.

In an urban environment, there will be a large number of building designs, surface finishes, roof covers, a variety of outdoor equipment and many different paved surfaces. Building surface finishes can range over smooth tile, concrete, brick, wood and many other surfaces. Paved surfaces can be concrete or asphalt, and may be new, cracked, broken or porous. Outdoor equipment can include motor vehicles, power transformers, bicycles, etc.

The large range of buildings, surfaces and occupancy factors met in an urban environment means that several cleanup criteria would be required since it would be much more difficult to clean certain types of surfaces and in many cases it would not be necessary to clean to the same level. It may be possible to leave relatively inaccessible areas contaminated, for example the tops and sides of high buildings, provided that the contamination is fixed and does not affect those in the building. The cleanup levels required for certain industrial sites having low occupancy and no routine public access could be less restricted than those for areas with heavy public usage, such as shopping centres, which may require very rigorous decontamination to reduce the collective dose equivalent and prevent the transfer of radioactivity into buildings via footwear and clothing.

3.1.2. Motorized Sweeping and Vacuum Sweeping

In urban areas of industrialized countries, motorized road sweepers and vacuum sweepers are used for cleaning roads and parking areas; hence such equipment should be readily available. Vacuum sweeping is the more attractive procedure since it not only cleans the surface but also picks up the displaced contamination more effectively. However, the removal efficiency for small contaminated particles, typical of those from a reactor accident, is likely to be low for these types of equipment.

Although cleanup efficiencies might be low, it is good practice to remove dry loose particulate material using this process before applying a liquid cleaner which could fix the contamination or cause it to penetrate porous surfaces. Even if only marginal decontamination is achieved, the amount of waste produced is minimal because there are no added reagents. Therefore, it is recommended that where access is possible, vacuum sweeping should be considered as the initial decontamination process for buildings, equipment and paved surfaces.

Since many sweepers collect the particulate material in a container on the vehicle, the dose to the operator will increase unless the container is shielded and/or water filled (which prevents dust emission as well as providing shielding).

3.1.2. Firehosing

It seems likely that firehosing could be a potentially useful technique provided it can be applied fairly promptly after an accident and depending on the particle size and texture of the surface. It relies on the contaminants still being in an accessible particulate or soluble form on the surface of materials where it can be redissolved or resuspended into the runoff water created. Obviously, as time elapses the likelihood of rainfall washing contamination further down into the matrix becomes greater.

The practicality of firehosing, and also high pressure water jetting, will depend upon the accessibility of drainage routes. Most road surfaces are provided with adequate storm drainage routes and firehosing of roads as soon as possible after an accident would seem to be a desirable step. However, if the firehosing merely shifts contamination to areas where it can become adsorbed more easily, then it may actually have a detrimental effect. For instance, movement of contamination from roofs or vertical surfaces of buildings to ground level could lead to a higher dose commitment.

Firehosing should also be useful for decontaminating buildings and equipment having smooth impermeable surfaces. It will be less effective for permeable, porous, rusty or cracked surfaces. The big advantage of firehosing is that the equipment is readily available in most areas.

During firehosing, large volumes of contaminated water could be produced. Great care should be taken to ensure that as far as practicable this water does not result in the contamination of drinking water supplies or of other areas. If the technique is used for widespread washing of buildings and roads, containing the water will be a major task.

3.1.3. Aqueous Methods Incorporating Chemical Additives

Numerous proprietary solutions are available for decontaminating surfaces at ambient temperature under non-aggressive conditions. Generally, these reagents contain various combinations of detergents and complexing agents.

The effectiveness of washing procedures can be improved by the addition of various inorganic ions (Na^+, K^+, Cs^+, NH_4^+) to exchange with adsorbed Cs^+. It was found that a dilute solution of ammonium nitrate was effective in removing caesium, adsorbed on a number of common urban construction materials. This reagent, as agricultural fertilizer, is readily available in large quantities, which is an important factor. Spraying with dilute ammonium nitrate solution always resulted in the displacement of some caesium; in some cases as much as 90% of the caesium was displaced in less than three hours. In general, aged weathered materials were most amenable to decontamination with ammonium nitrate. This technique has so far only been applied on a laboratory scale. Further development is needed for full scale application for extended periods, for the collection and disposal of the radioactive waste arisings, and for very large scale use, consideration of the possible contamination of groundwater supplies.

3.1.4. Abrasive Jet Cleaning

Abrasive jet cleaning including both wet and dry procedures with various types of grit has been employed on a large number of occasions in the nuclear industry. These applications range from heavily contaminated pipework with the contamination fixed in oxide on the surface, to lightly contaminated surfaces. Typical abrasive which have been used include sand, glass beads, metallic beads and soft materials such as nut shells and rice hulls. Abrasive jetting has been shown to be a very efficient method, with DFs of 10-100 being obtained. Wet sandblasting of houses has been used as a restoration procedure. However, it is a relatively costly, labour intensive procedure which would be difficult to apply on a large scale. One of the major problems would be containing the wastes produced though equipment is available incorporating vacuum brush techniques. However, careful health physics control of the operation would be required to ensure that people were not exposed to radioactive aerosols and that contamination was not spread. From the aerosol generation viewpoint, wet abrasive blasting may be a better procedure. However, this has the disadvantage that both the water and the abrasive must be retained and monitored for disposal.

One of the advantages of abrasive blasting is that the equipment is commercially available and there is considerable cleaning experience on various surfaces (Fig. 1). For freshly contaminated surfaces with the radioactivity on the outside, good decontamination factors can be obtained. In principle, equipment could be operated remotely, although for complex surfaces involved setting up might be required.

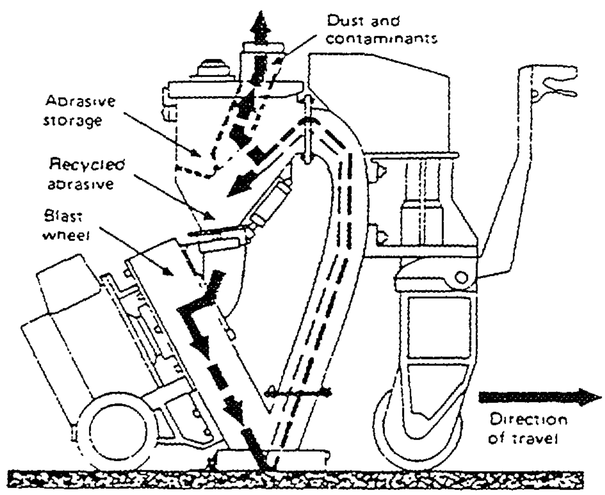

FIG 1: Portable abrasive blasting equipment. (Credit: Blastrac.)

3.1.5. Road Planing/Grinding

The removal of a fairly precisely defined layer, typically 1-3 cm from the surface of asphalt or concrete roads, using commercial equipment is a common procedure during road resurfacing (Fig. 2). Both cold planing for asphalt and concrete and hot planing for asphalt are used. The planers can cut the surface with hard bits at speeds up to 4.5 $km.h^{-1}$ and milling widths up to 2.1 m and load the milled surface rubble directly into a truck. Although the use of such equipment to remove a layer of contaminated material from a road surface has not been reported, it is likely that very effective decontamination could be achieved. Costs for cleaning contaminated surfaces would be higher than for normal road work since methods to keep contaminated dust from spreading would be required, for example wetting surfaces and spraying the rubble. Extra costs would arise if there were special requirements for disposal of the wastes.

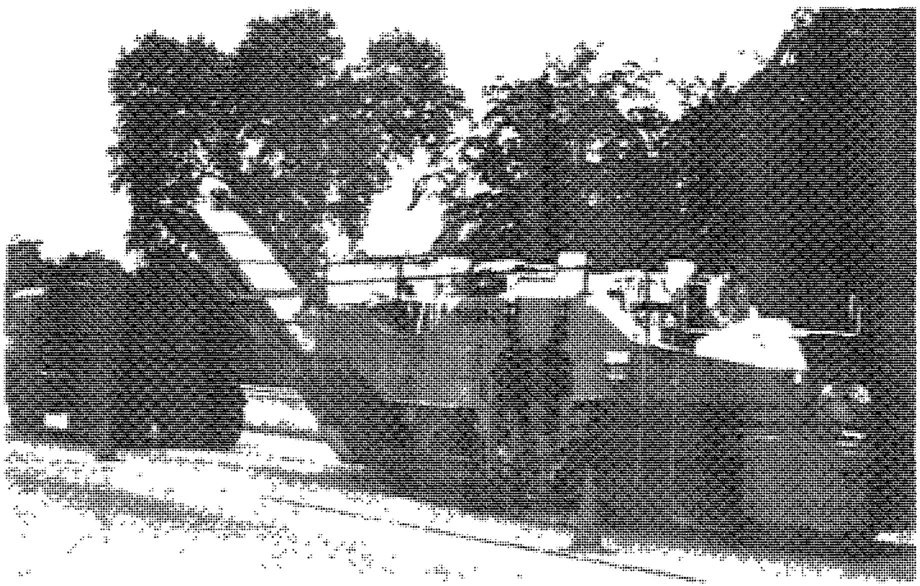

FIG 2: Cold planer for removing a layer from concrete and asphalt surfaces (Credit: Wirtgen.)

Such road planers, using different types of cutters for the removal of layers of earth and direct loading into trucks, might also have application in areas with fairly flat surfaces.

Smaller scale remotely operated scarifers have been used during the cleanup of various nuclear plants (Fig. 3).

A large number of hand held and large commercial grinders (Fig. 4) are available for removing thin layers of contaminated material from the surface of concrete. Some of the technology employed is an extension of highway grinding processes developed in the 1970s.

Road planers and grinders have limited applicability and would be expensive compared to certain other techniques. However, in some cases the use of such equipment may be the only answer.

3.1.6. Strippable Coatings

Strippable coatings are liquids or gels which are applied to surfaces, allowed to dry, and then stripped from the surface, carrying with them the loose contamination. The stripped film must be strong enough to be removed from the surface in as few pieces as possible. In a similar fashion to the gels and foams described above, various decontamination chemicals can be added to the film

Strippable coatings are ideal for large scale recovery operations, especially for structures and large pieces of equipment, since they can be applied easily and quickly to large areas and require minimum equipment and personnel. Although these coatings can be applied by brushes or rollers, a pressurized spray system is best for large areas since it coats without disturbing the contamination. Loose contamination is trapped during the curing process and removed with the layer which is easy to handle and dispose of.

One disadvantage of strippable coatings is that they require careful removal, generally by hand, and thus a considerable radiation dose may be incurred. This may limit their application on a large scale.

3.1.7. Cleanup of Indoor Contamination

Contamination of indoor surfaces in urban buildings is likely to occur by infiltration of radioactive aerosols during dry deposition, by infiltration of contaminated dust particles or by transport of activity indoors by foot traffic.

Cleanup of dust borne and foot borne contamination on smooth surfaces can probably be achieved by vacuuming and/or washing and scrubbing. Cleanup of radioactive aerosols on smooth surfaces could probably be accomplished only by washing/scrubbing. It is unlikely that more severe methods of cleanup, for example firehosing or steam cleaning, would be warranted or acceptable.

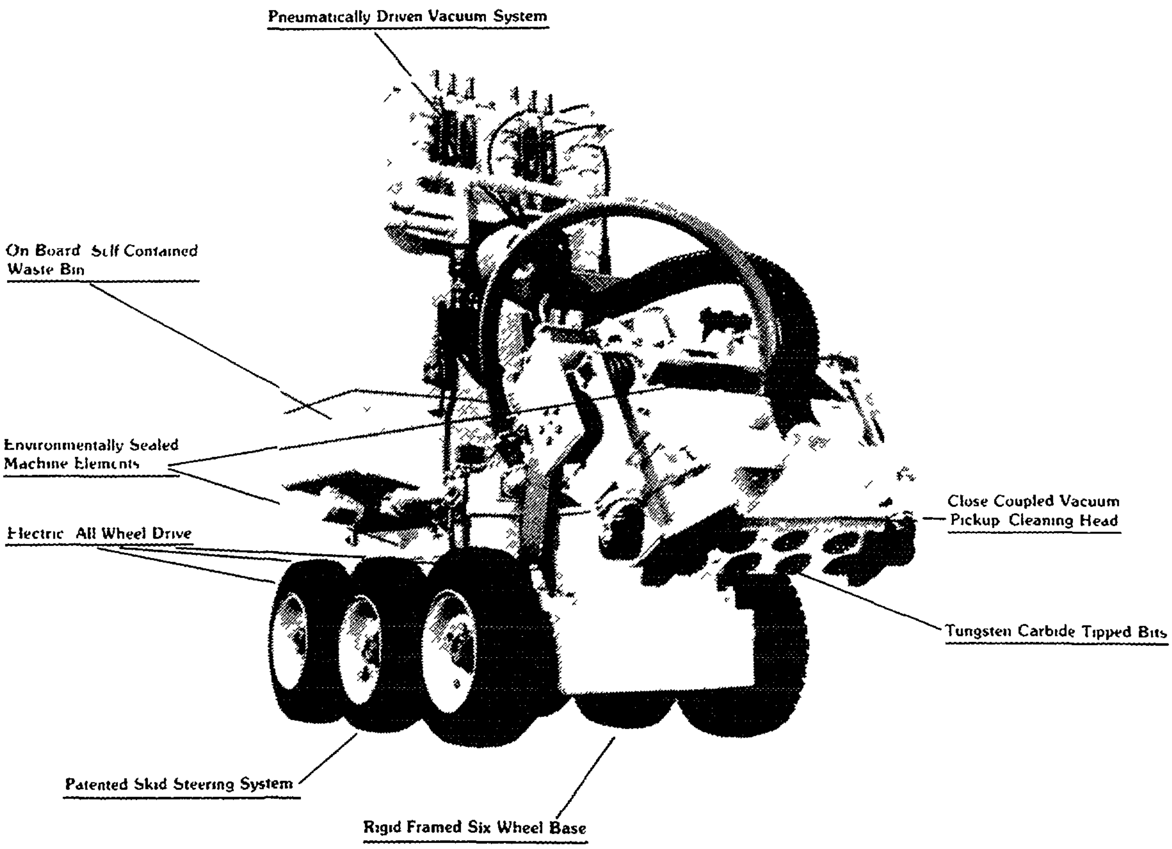

FIG 3: Remotely operated scarifier that vacuums, filters and collects all rubble. During active operation it is wrapped in plastic to minimize contamination of the vehicle (Credit: Pentek Inc./Electric Power Research Institute.)

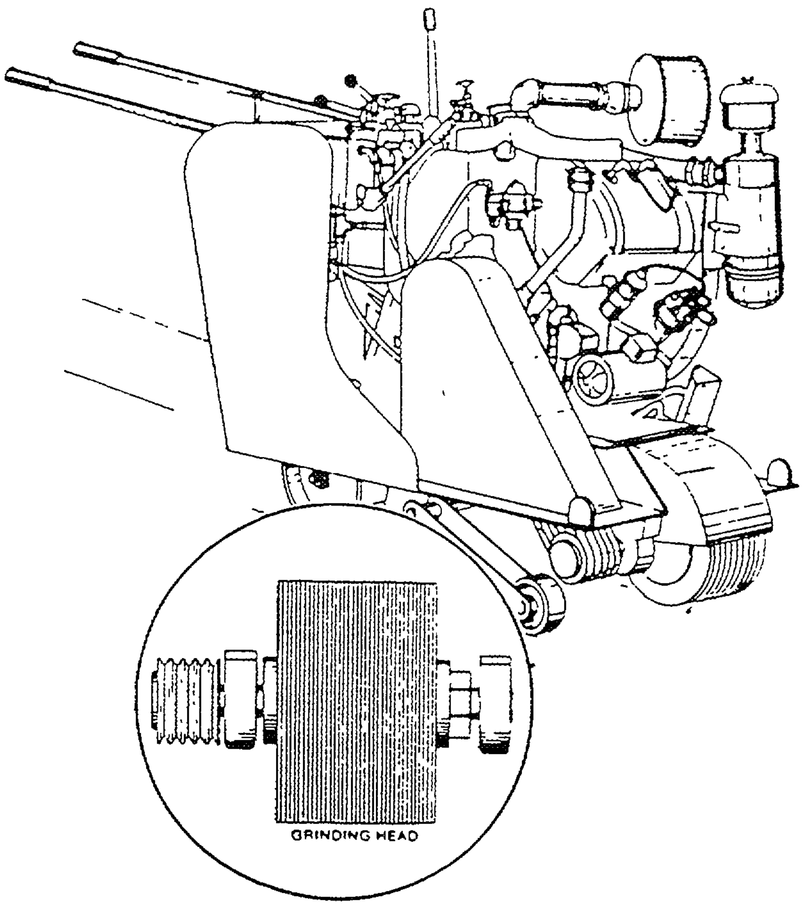

FIG 4: Heavy duty floor grinder

Cleanup of rough surfaces (curtains, rugs, rough wooden floors, etc.) could be more of a problem. Vacuuming may be partially successful for dust borne particles. Removal of curtains and rugs for washing/dry cleaning might be required if excessive contamination remains. The cleanup of activity on indoor household surfaces needs further work.

3.1.8. Decontamination of Equipment

During a cleanup operation, a large amount of equipment will be used, including various vehicles, hosing, pumps, specialized units, instrumentation and clothing. These all run the risk of becoming contaminated, thereby giving an additional dose to operators and requiring further decontamination operations. Where possible, simple protective measures should be used on equipment to facilitate its subsequent decontamination. Painting, strippable coatings and protective plastic covers applied in advance as a temporary protection are possible measures which could be taken.

Well organized decontamination centres for equipment are required, especially at the transition between dirty and clean zones. These may consist of simple monitoring and washdown facilities for trucks at disposal areas and transition zones, in addition to centres having special decontamination equipment.

The organizing team should, where possible, make use of available expertise, equipment and facilities for decontaminating equipment. For example, it may be possible to convert garage facilities or standard car wash facilities to clean vehicles and other equipment since they have high pressure hoses, detergent cleaning, steam cleaning and hoist facilities. However, before wet washing, equipment and vehicles should be vacuumed to remove as much loose contamination as possible. Both wet and dry vacuum cleaners having filtered outlets should be available at cleanup stations.

Planning should include provision for the containment and treatment of waste water generated during cleanup.

Certain cleanup centres may contain specialized equipment for reclaiming valuable pieces of equipment and instruments. Examples are: freon systems for cleaning electrical equipment, instruments, greasy items, clothing, etc., ultrasonic baths for cleaning tools, pumps, components, general equipment, etc.; and various chemical baths. Whether or not these specialized techniques and others such as electropolishing are used, and the timing of such use, will depend on the accident scenario, the availability of equipment and trained staff, the need for such techniques, etc.

During the cleanup of very large areas, the decontamination of clothing, overshoes, respirators and the other types of personal equipment used by the cleanup crews will be a major problem requiring access to laundry and cleanup facilities. Various designs of laundry and cleanup facilities for active clothing and gear are readily available for routine and emergency use. These facilities may also be required to clean up materials which have been contaminated by indoor deposition.

3.1.9. Guidance on the Selection and Application of Decontamination Methods

The previous subsections reviewed a number of decontamination procedures which could be used for various surfaces during large scale cleanup operations. A summary of the techniques (including simple vacuuming and washing) most appropriate to various surfaces is given in Table I. The techniques are shown in order of approximate cleanup cost per unit area. Some techniques such as vacuuming and fire hosing can be applied relatively quickly by unskilled personnel. In other cases, e.g. abrasive blasting, much more planning, especially with attention to health physics precautions and waste disposal, would be required.

Table II lists equipment which would be required or useful for cleanup of an urban environment along with the skill requirements to operate such equipment. Equipment for monitoring or decontaminating personnel is not included in this list.

In general, it is recommended that vacuum sweeping and/or vacuuming be considered as the initial decontamination process, especially if the contamination is in the form of dry loose particulate material. Even if only marginal decontamination is achieved, the amount of waste produced is minimal and the process does not fix the contamination to the surface or cause it to penetrate porous surfaces. Use of this equipment in areas of medium to high activity would not be possible unless shielded or remotely operated equipment is available. The use of vacuum cleaning for the inside or urban buildings and smooth building surfaces should be beneficial.

Firehosing is also recommended under controlled conditions, especially on smooth surfaces such as roads and parking lots which need to be cleaned up quickly. However, it should only be used if suitable drainage routes are available and contamination of drinking water does not occur. Firehosing should also be useful for decontaminating certain types of roofs, buildings and equipment having smooth impermeable surfaces. Care must be taken to ensure that the process does not just shift the contamination from high surfaces to ground level, resulting in higher dose commitments.

If vacuuming followed by firehosing is not successful in cleaning up heavily contaminated ares, more aggressive methods such as abrasive cleaning, road planing or paint removal would be required.

For decontamination of buildings a detailed survey of individual surfaces will be required. It is likely that contamination levels on different roofing materials will vary substantially. During the cleanup of urban areas, every effort should be made to select decontamination processes that minimize the spread of contamination from exterior to interior surfaces. Interior contamination would generally cause higher dose commitments than contamination outside the building. When using road planing,

TABLE I SUMMARY OF DECONTAMINATION PROCEDURES MOST APPROPRIATE FOR VARIOUS SURFACES

	Increasing cost												
	Vacuum cleaning[a]	Washing with detergent	Sweeping or vacuum sweeping	Fire hosing	Water jetting	Steam cleaning	Aqueous with chemical additions	Gels foams	Strippable coatings[b]	Abrasive cleaning[b]	Spalling	Road planing	Remarks
Plastics		A		B	B			A	A				
Asphalt/concrete paving		B	C+	C+	C+					B		A	Use of modified street cleaners should be considered
Concrete walls		B		B	B	B	B	B	B	B	A		
Metal surfaces		B	B	B	B	B	B	B	B				Applicability depend on accessibility of surfaces
Metal machines		B		C	C	B	B	B	B				Reduced efficiency for complex machines
Glass		A		B			A	A	A				
Painted surfaces		B	C	C		B	B			A			Commercial stripping solutions should be effective
Roofs metal		B	(As for metal surfaces but accessibility could be a problem for some techniques)										
Roofs other		C		C	Spray with dilute amonium nitrate								Development of some form of roof irrigation device to keep surfaces wet for a number of hours is required
Unpainted wood		C		C									Scraping/sanding may be effective
Brick walls		C		C+						A+			

[a] Good DFs if surface is smooth or if contamination is in the form of small particles or is attached to dust Much less effective if contamination has penetrated below the surface or is in the form of aerosols

[b] For use on limited areas

A Good DFs

B Good DFs depending on surface finish and type and depth of contamination

C Variable DFs depending on surface condition and type of contamination ,

\+ Further investigations of applicability for surfaces contaminated with reactor accident fallout required

TABLE II: EXAMPLES OF EQUIPMENT WHICH COULD BE REQUIRED/USEFUL FOR CLEANUP OF URBAN ENVIRONMENT

	Likely availability	Skilled personnel required to operate or install
Industrial vacuum cleaners	A	Z
Vacuum brushing street cleaners	B	Z
Fire tenders and hoses	A	X
Water jet cleaners	B	Y
Steam cleaning apparatus	B	Y
Pumps, sprays, perforated pipes	A	X
Chemicals - detergents, ammonium nitrate, strippable coatings, gels, foams, paints	A	X
Scaffolding, ladders	A	Y
Road planers	B	X
Abrasive blasters	B	X
Water tanks - water supply, etc.	A	X
Large trucks, loaders, graders	A	Y
Airborne activity monitors	C	X
Health physics equipment	C	X
Large ultrasonic and freon baths	C	Y
Deep ploughs	B	X

A - Should be easy to acquire/requisition at short notice
B - Limited availability
C - Likely to be resource limited
X - Essential
Y - Desirable
Z - Not essential.

Equipment for monitoring and decontaminating personnel also required.

sweeping, abrasive cleaning and other processes which could raise dust, methods to minimize dust spread should be used.

During preparations for the decontamination or urban areas, detailed attention must be given to the current and future use of the facilities involved. It is possible that where particular surfaces or geometries have led to gross accumulation of contamination, isolation of the area may be a more cost effective solution than decontamination.

3.2. Decontamination of Large Land Areas

Many of the decontamination techniques described in the previous section are not appropriate to the cleanup of large land areas. This section briefly reviews the special methods used to decontaminate such areas.

During the planning stage, it is important to select land cleanup methods that will least affect the viability of the land to produce beneficial crops and minimize the ecological damage to the soil, vegetation and animals. The selection of the proper technique will also make reclamation of the land following cleanup easier.

A generic assessment of the ecological impact of land restoration and cleanup techniques for various land types and land use classes was performed in the USA. The areas examined for cleanup ranged from 0.01 to 10 km^2. Conclusions about the effects of cleanup on the soil, vegetation and animals in an area are summarized in Table III using a ranking of 0 to 5 for each cleanup method. The interpretation of these rankings is:

0 - causes no measurable change in the ecosystem
1 - preferred technique because adverse environmental effects on recovery and side effects of treatment are minimal
2 - conditionally acceptable because of significant impact by the treatment and/or the equipment upon the area
3 - acceptable as a 'last resort' cleanup to remove exceptionally hazardous material while incurring maximum acceptable impact
4 - causes unacceptable damage but can be used as an interim cleanup if the injury is erased during the final treatment
5 - not applicable to the land type for which it is proposed.

The rankings considered the environmental insult generated during the cleanup, the physical possibility of restoring the area to its original productive state, side effects caused by the equipment needed to perform the cleanup, the impact upon the environment adjacent to the cleaned up area, and the social acceptance of the cleanup work. Not all treatments were expected to be evaluated with all land types; the exceptions are indicated in the table.

It should be emphasized that the conclusions from Table III are very specific to the land types discussed in the report and the conclusions only provide general guidance.

The selection of the most suitable methods of cleaning up large areas of contaminated land and restoring to productive use is complicated by:

- the topography of the area to be cleaned up
- the large number of possible natural ecosystems and land uses
- the large number of vegetation types
- the large variation in the characteristics of soil classes
- the complex behaviour of radionuclides with different soils
- the varied response of the contamination to different weather conditions
- the ecological impact that different cleanup techniques have on different natural ecosystems and land restoration.

The final selection of the methods to be used to clean up an area must consider accident specific and site specific factors such as the type of contamination, how it was deposited, soil types, value of the land, alternative land use, population distribution, size of the affected area and the equipment available. Many techniques and types of equipment will be required for cleanup after any serious accident. The methods selected should reduce the beta/gamma radiation to acceptable levels, prevent radioisotopes such as ^{90}Sr, ^{137}Cs and actinides from entering the food chain and have minimal ecological impact. In addition, the methods must be safe, practical and cost effective because of the logistic problems and huge costs associated with the cleanup of such large areas and the need to dispose of the wastes.

In general, the cleanup methods can be classified as physical, chemical and agricultural or some combination of these. The more important methods are described in the following sections.

3.2.1. Physical and Chemical Methods

The cleanup of land can be carried out by selectively separating the radionuclides from the soil matrix, by deep ploughing to remove the contamination from the surface and the root zone or by removing the vegetation and/or top layer of soil containing the contaminants.

The volume of wastes arising from the cleanup would be smallest for deep ploughing and largest for layer removal. The volume of wastes from the separation technique would depend on how well the separation could be done. The cost of storing, transporting, additional treatment and/or disposal of contaminated soils and vegetation is an important factor in selecting the proper method. For example, if the disposal area is a long distance from the wastes, transportation costs could exceed all other costs if the layer removal technique was used.

3.2.1.1. Physical and Chemical Separation of Radionuclides from the Soil

Separation of radionuclides from soil is desirable since it can significantly reduce the volumes of wastes which have to be transported and disposed of. In principle, this technique is applicable only to coarser grained soil or gravel in which the radionuclides are associated with fine grained particles which can easily be separated. The technique is most practical if the area to be decontaminated is relatively small. However, since physical separation of radionuclides is almost always associated with the removal of the clay fraction of the soil matrix, the process will result in a decrease in soil fertility. If the land is to be used for crop production, addition of fertilizers after the cleanup will be necessary to restore land fertility.

Two physical techniques have been investigated, inertial separation and gravitational separation. The decontamination of soils using these methods can be carried out using water, chemical wash solutions or chemical separation processes.

3.2.1.2. Deep Ploughing

Deep ploughing has been investigated to a limited extent in several countries as an alternative to the removal of the contaminated soil layer. Typically, a tractor drawn trenching plough is used to completely invert a thick layer of soil, placing the active top 10 cm at the bottom and moving the deep clean layers to the top. In theory, with this method the major part of the activity would be placed well below the lower boundary of the roots of the crop. However, ploughing does not result in the perfect turnover of soil layers and some mixing of layers occurs. The extent of this mixing has been investigated to some extent but further work needs to be done. Before a decision is made for deep ploughing, an evaluation of the impact on soil fertility and productivity should be conducted. The impact of deep ploughing appears to be influenced by the type of soil and the crops grown.

It is evident that further study is required to determine when and if ploughing should be used as a cleanup procedure. The primary benefit would be the reduction in external radiation levels at the surface. The benefit regarding soil-plant transfer will depend on the depth of ploughing, soil type, how the ploughing affects the vertical distribution, the root depth of plants, etc. Even for acceptable circumstances, the cost-benefit advantage of ploughing versus other methods and the depth of ploughing must be carefully considered. In some areas, the presence of land drainage systems and subsurface items such as cabling may limit the depth to which land can be ploughed.

If deep ploughing is used, the replacement of deep-rooted plants by shallow- rooted plants may be desirable.

3.2.1.3. Removal of Vegetation

Since under certain conditions vegetation can intercept almost all of the fallout, its removal could be an effective method of decontaminating certain areas.

The removal of contaminated vegetation appears to be an effective method of decontaminating land under certain conditions. The effectiveness of the technique depends on the density and type of vegetation, on the nature of the contaminant and the method of application (wet/dry). In any event it may be necessary to remove surface vegetation to permit subsequent treatment of the soil surface.

TABLE III: SUMMARY OF CONCLUSIONS ABOUT THE EFFECTS OF VARIOUS CLEANUP MEASURES ON THE SOIL, VEGETATION AND ANIMALS IN VARIOUS LAND USE CLASSES AND TYPES

	Land use classes		Land types						
	Suburban	Agriculture	Coastal/ intertidal marshes	Tundra	Mountain, subalpine	Coniferous forest	Deciduous forest	Prairie	Desert
Natural rehabilitation	4	4	4	3	4	4	4	3	4
Chemical stabilization	4	3	3	5	2	2	2	5	2
Clear cutting vegetation	4	3	3	5	2	2	2	5	3
Stumping and grubbing	4	3	3	5	3	3	3	5	4
Scraping and grading (<5 cm)	3	1	3	1	2	2	2	1	4
Shallow ploughing (< 10 cm)	4	1	5	5	4	4	3	1	4
Deep ploughing (10-20 cm)	4	1	5	4	4	4	3	1	4
Soil cover (<25 cm)	2	1	2	2	3	3	3	2	4
Soil cover (25-100 cm)	4	1	3	4	4	4	4	3	4
Remove plough layer (10cm)[a]	2	1	3	1	2	1	1	1	4
Remove shallow root zone (<40 cm)	4	1	3	2	3	2	2	1	4
Remove scraping and grading, mechanically stabilize	1	1	2	1	1	1	1	1	4
Remove plough layer (10 cm), mechanically stabilize	1	2	2	2	3	2	2	1	4
Remove shallow root zone (<40 cm), mechanically stabilize	4	2	3	2	3	3	3	2	4

TABLE III: SUMMARY OF CONCLUSIONS ABOUT THE EFFECTS OF VARIOUS CLEANUP MEASURES ON THE SOIL, VEGETATION AND ANIMALS IN VARIOUS LAND USE CLASSES AND TYPES (cont.)

	Land use classes		Land types						
	Suburban	Agriculture	Coastal/ intertidal marshes	Tundra	Mountain, subalpine	Coniferous forest	Deciduous forest	Prairie	Desert
Remove scraping and grading, chemically stabilize	2	2	4	5	3	3	2	1	4
Remove plough layer (10 cm), chemically stabilize	2	2	4	5	3	3	2	2	4
Remove shallow root zone (<40 cm), chemically stabilize	4	3	4	5	4	4	4	3	4
Barriers to exclude people	3	2	1	1	1	1	1	3	1
Barriers to exclude large and small animals	3	3	3	3	3	3	3	3	1
Mechanical stabilization by hard surface	5	4	b	b	b	4	4	3	4
Application of sewage sludge	a	1	b	b	b	0	0	b	b
High pressure washing (<3 cm)	a	a	b	b	3	b	b	b	b
Flooding (3 to 30 cm)	a	a	b	b	5	b	b	b	b
Soil amendments added	a	4	b	b	b	b	b	b	b

a: Increases the severity of scraping and grading.

b: Outside the scope of this work.

For large areas, brush and small trees can be removed using cabling or anchor chaining. In cabling, a 45-60 m long steel cable is dragged between two tractors travelling on parallel courses. The cable breaks off or uproots brush and can be used where the brush breaks easily and is not willowy. In anchor chaining, a heavy chain is dragged by two tractors to break or uproot vegetation including small trees. The ground is more disturbed with anchoring than with cabling. Grassy vegetation can be cut using a mower. Figures 5 and 6 show examples of techniques being developed in France.

When vegetation is defoliated and allowed to desiccate, it may be desirable to apply a bitumen emulsion or synthetic polymer spray to reduce suspension of contamination during collection, compaction, transportation and disposal Dead vegetation and very dry soils can cause severe resuspension problems unless they are stabilized or dampened.

3.2.1.4. Removal of Surface Soil

Studies and decontamination projects in the former USSR, the USA and other countries show that many common types of earth moving equipment such as graders, bulldozers and scrapers can be effective in removing a layer of contaminated soil. The earth moving machines can be used to efficiently remove layers of material (sod, soil, etc.) as thin as 5-15 cm or thicker than 35 cm and transport the soil distances of 150 m without reloading or stopping. The contaminated earth is either moved into piles and hauled away or buried directly in a depression or specially excavated trenches.

The effectiveness of any procedure depends greatly on the type of terrain and soil and the land use class. If the cleanup is done while the contamination lies on the surface of the soil, then careful removal of a layer slightly greater than the irregularities in the surface should remove all of the contamination. The removal of contamination will not be complete if the irregularities and fissures in the surface are deeper than the surface layer removed or if spillage occurs. Removal of a layer of soil will be less effective as a decontamination method if the radioactivity has moved down the soil profile. The rate at which the move down occurs depends on the ground cover, the soil type and the amount of precipitation following deposition

FIG 5: Forage harvester used in experiments to remove all kinds of crops in the French RESSAC programme (Credit: Renault)

FIG 6: Machine used to reduce underbrush and small trees to chips in the RESSAC programme (Credit: Cimat.)

This type of decontamination method is most effective in flat, relatively large areas having fine grain compacted earth. The efficiency of removal of the surface layer is affected by surface unevenness, soil texture, moisture content and vegetation cover In some cases it may be advantageous to remove part of the vegetation cover before removing the layer of soil. If the surface is coarse grained or gravel the contamination may have seeped to considerable depth, making this type of decontamination less effective. Figures 7 to 10 show examples of machines currently used to remove surface soil.

A key element to prevent the spread of contamination during earth removal is dust suppression; this can be achieved by water sprays. Another method to fix the contamination is to spray the earth with an asphalt emulsion (Section 2) which dries and glues the soil components together for removal of the layers.

Past experience with the cleanup of contaminated soil indicates certain features which would be desirable in graders and other earth moving equipment, such as:

- smooth cutting surface (teeth tend to smear contamination)
- ability to skim layers of soil as thin as 10 cm and transport large volumes short distances (up to 200 m) with minimal spillage
- ease of control and good vision by the operator

Just as important as selecting proper equipment for cleanup is the selection, training and supervision of operators and the planning of the campaign to ensure efficiency and thoroughness. The use of mobile monitoring systems is a very time and cost effective means to ensure the effectiveness of the cleanup processes.

Table III summarizes some generic conclusions about the ecological effects of these cleanup methods on the soil, vegetation and animals in various land types and land use classes

FIG 7: High capacity scraper used in the coal industry (26 m^3) (Credit: TEREX.)

FIG 8: High capacity loader (Credit: Dresser.)

In summary, it appears that the removal of surface soil can be an effective method of decontaminating certain types of soil such as clay loam without doing serious ecological damage. However, the application of this technique to fragile ecosystems should only be made as a last resort and only if subsequent rehabilitation actions are conducted.

Equipment must be selected to suit a particular land area and accident situation. There is no method which is best for all circumstances. The use of special large scale industrial equipment in the cleanup of areas contaminated with radioactive and toxic pollutants is worth investigating further.

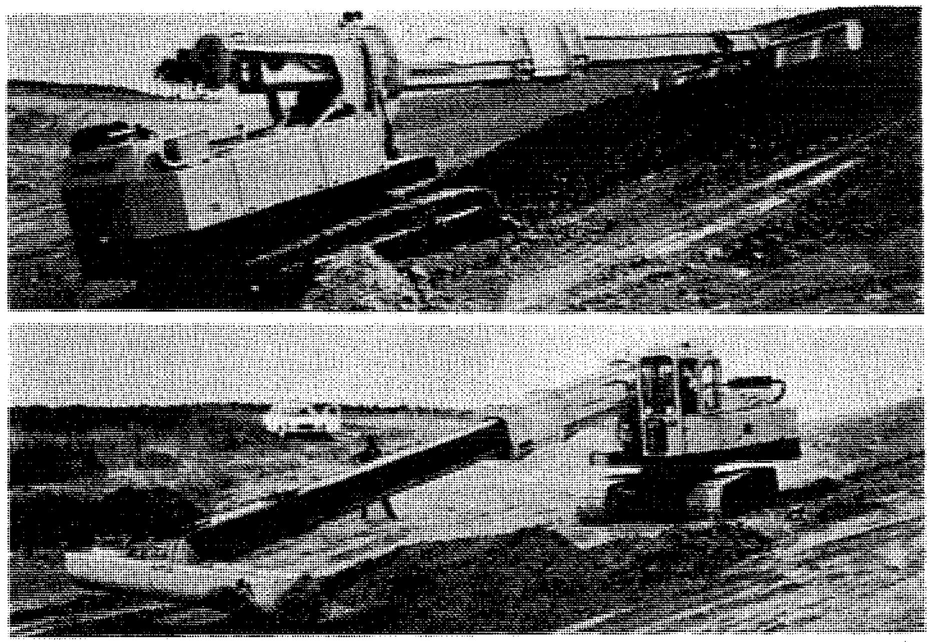

FIG. 9: Machine for removing a layer of soil from steep slopes (Credit: Wieger Maschinenbau GmbH)

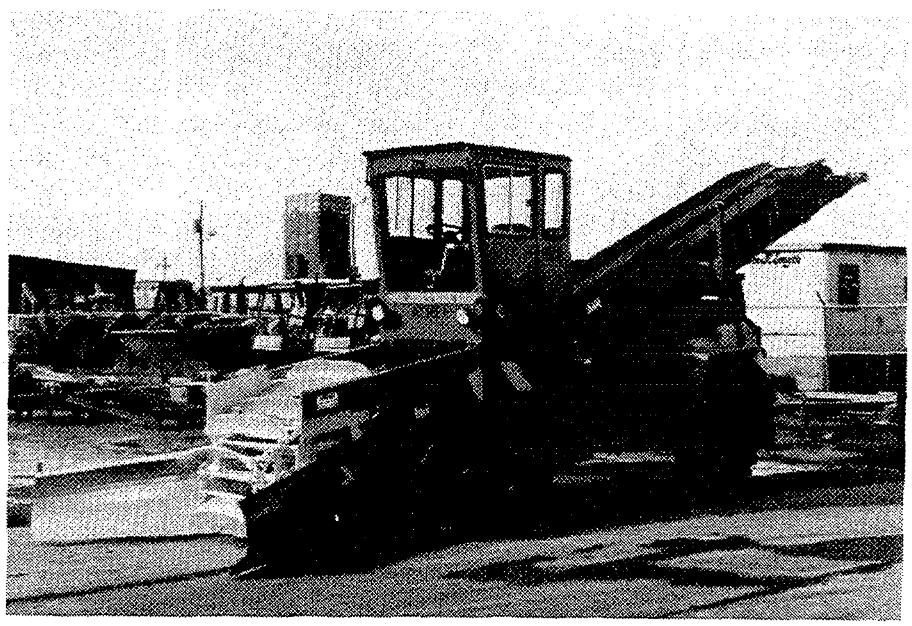

FIG 10: Force feed loader with 25 foot (~ 8 m) main conveyer. The moldboard is adjustable and tapered (Credit: Athey Products Corp.)

3.2.2. Biological Decontamination of Soil Using Plants

On the basis of current literature data, this technique does not appear to be practical for widespread usage even though it is feasible. However, it might have some application in special cases, for example to decontaminate interdicted land in an undisturbed state over a long period of time. Further studies are needed to determine the full potential of this technique. Factors that need to be considered include: the most appropriate plant species, conditions that will maximize radionuclide uptake, the number of crops required to reduce soil concentrations to an acceptable level, harvesting practices and costs, and plant processing and disposal methods and costs. This approach to soil decontamination should be considered in the context of other options that may be available, such as the use of chemicals and fertilizers to reduce the uptake of radionuclide contaminants in soil during the productive use of land following a contaminating event.

3.2.3. Restoring Land to Productive Use

In many cases, contaminated land could eventually be reclaimed and returned to productive use. The return to productive use can be assisted by:

(a) The eventual reduction in residual activity levels in the soil by natural means;

(b) Decontamination of the land followed by reclamation measures such as fertilization;

(c) Deep or shallow ploughing in combination with the addition of chemicals or adsorbents to reduce the uptake of residual radionuclides in plants;

(d) Using the land to grow non-food/feed crops.

To make restoration of land following decontamination easier, it is important in the planning stage to select decontamination methods that will least affect the viability of the land to subsequently produce beneficial crops and minimize ecological effects on soil, vegetation and animals. In addition, the planners should decide on which remedial actions are required to restore productivity to the land after cleanup.

Numerous research workers have addressed the problem of revegetating land following remedial actions and mining activities. Revegetation is particularly difficult in arid areas. Irrigation including drip irrigation with the application of nutrients has been successfully applied. Most of these studies address revegetation from the point of view of stabilization of soils rather than increasing direct beneficial use. However, the land must first be stabilized if it is eventually going to be put to productive use. Various techniques investigated for encouraging growth of vegetation include the addition of topsoil and treatment with fertilizer, straw, clay, minerals, pH modifying chemicals and other substances.

An action required in response to one need may provide a remedy for another problem, for example the addition of fertilizers and minerals to farmland after removal of the top layer will not only reconstitute the soil but may result in decreased uptake of ^{90}Sr, ^{137}Cs and transuranic elements. Overlaying the soil with clean topsoil from nearby lands with an overabundance of topsoil should also be considered in certain cases. This process will not only increase nutrients but will dilute radionuclide concentrations in the root zone of crop plants. However, for certain land types and land use classes this action may not be desirable (Table III).

Various workers have addressed practices that may be effective in returning land to productive use by reducing the uptake and retention in plants of radionuclides following a contaminating incident. Increased availability of isotopic or chemically related elements can reduce the soil-plant transfer of radioactive isotopes. The use of liming and increased pH will decrease the uptake of strontium and the application of potassium/phosphorous fertilizers will reduce the uptake of caesium. The uptake of potassium rich fertilizers reduced the uptake of ^{137}Cs by an order of magnitude in a variety of tropical crops. To get a reliable estimate of the usefulness of such techniques to assist in the reclamation of contaminated land the following considerations should be kept in mind. Adding fertilizers or chemical analogues creates a competition with the radionuclide at the plant root absorbing zone, and therefore a lower contamination level in the plant should be expected. However, a similar competition may also occur at the soil absorbing sites, resulting in an increase in bioavailability and higher levels of contamination in the plant. Therefore, depending on the chemical nature of the radionuclide, the soil type and the plant species, a reduction or increase of the plant contamination level may then occur. Much insight into the basic principles of soil and root absorption has to be obtained before these methods can reliably be applied to reclaim land.

Selective removal of ^{137}Cs from soil poses a more difficult problem owing to the lack of suitable complexing agents. Although compounds such as crown ethers will complex caesium, they are quite toxic and very expensive. Hence they would not be suitable for application on a large scale. These techniques with complexing agents may have serious drawbacks since most of them more effectively bind the micronutrients indispensable for healthy plant growth, and these will not be fully restored by fertilization techniques. The cost-benefit analysis of such practices moreover, will need careful consideration.

Land may be reclaimed and used for productive purposes, even if there is some residual contamination, by the judicious selection of crops. For example, the cultivation of non-food/feed crops such as cotton, flax and timber could be considered if food crops would contain unacceptable concentrations of radionuclides. Again, the content of radionuclides such as ^{90}Sr should be very low in corn since it has one of the lowest mineral contents of all grains and would be safe to grow on contaminated land.

Land could also be restored to productive use by growing sugar and oil producing crops since most of the radioactivity in the refined products would be removed during processing. However, if the by-products, such as sugarbeet pulp, are fed to animals for meat production, the indirect contribution of radionuclides to the human diet would have to be considered.

Changed practices such as the planting of deep-rooted rather than shallow- rooted crops would be expected to reduce the uptake of radionuclides unless the activity has penetrated well below the surface as a result of deep ploughing or for natural reasons.

The available information on the reclamation of land and land use does not constitute a body of facts that can be translated into specific and precise guidance to be followed in agricultural practices after a contamination incident. However, current information and experience that is now being accumulated in the aftermath of the Chernobyl accident should be helpful in selecting practices that will enhance the beneficial use of land.

4. LOADING AND TRANSPORTING LARGE VOLUMES OF WASTES

Large volumes of contaminated soil, concrete, asphalt, equipment, vegetation, etc. could arise from the cleanup of a large area contaminated as a result of a serious accident at a nuclear power plant. The removal of a thin (average thickness of about 5 cm) layer of contaminated material from a 7 km radius around a damaged facility could result in 8 x 10^6 m^3 of waste which has to be transported to a disposal site and buried. The loading and moving of such large volumes of soil is time consuming and expensive but the experience is not unique.

For example, during the construction of large earth dams, millions of cubic metres of inactive soil and concrete have to be loaded and moved. It is also common to load and move large volumes of product and waste rock in mining.

During the cleanup of very large contaminated areas, the loading and transportation of much of the wastes to the disposal site could probably be accomplished using conventional earth moving equipment from the construction industry. Some modifications may be beneficial, such as the addition of shielding between the driver's cab and the box of the dump truck. If the disposal site is located within the cleanup area, much larger equipment such as that used on the site in major civil engineering and mineral extraction projects could be used.

The loading of the contaminated soil could be done:

(a) Using equipment such as wheeled or tracked loaders and excavator loaders with capacities of 30 m^3 or more. The material would first be moved into piles using conventional grader/planers or bulldozers with wide blades.

(b) Using a force feed loader with a conveyor which can pick up a layer of soil or soil from large windrows and dump it directly onto a truck. On flat surfaces it may be possible to use a modified road planer.

(c) Using vacuum pickup systems for certain types of soil under dry conditions.

Water spraying equipment, to dampen soils during handling under very dry conditions, may be useful to minimize dust production.

Highly contaminated soil from locations close to the damaged facility may have to be sealed in appropriate containers for transport. Remotely operated equipment or units with shielded/air filtered cabs would be required.

The contaminated wastes could be transported using one or more of the following techniques:

(1) Moving the layer of contaminated soil directly into depressions or specially excavated trenches using scrapers, bulldozers or graders. The soil can be moved 100-150 m without reloading or stopping.

(2) Loading the soil into dump trucks for transport to the disposal site. Rear dumping trucks are available with capacities of up to 250 t.

(3) Loading the soil into railway cars for transport to the disposal site. The choice of rail transport depends on the availability of railway lines in the vicinity of the cleanup and disposal sites. If double or triple handling of material is required, as in a truck-rail-truck transportation system, Canadian analyses suggest that rail transport is not cost effective for distances less than a few hundred kilometres. However, the economic factor in the decision may be offset by the fact that rail transport results in smaller radiation exposure to transportation workers and involves less interaction with the public than does truck transport.

Effective management and control systems will be required to move and dispose of large quantities of earth safely. The protection of the operational staff and the environment must be important factors during the planning and cleanup. One of the biggest problems on a job of such magnitude may be to ensure continual maintenance of safety and health physics procedures once the job becomes routine.

In planning for the loading and transport of these wastes there are certain basic requirements:

- a modified waybill control technique in conjunction with a data handling system to control the loading, transport and disposal of wastes;

- well defined transportation routes and truck control points to ensure compliance with the routing plan;

- truck cleanup areas and monitoring points either at the dump site or between the contaminated and clean zones;

- an emergency response plan for implementation in the event of a transportation accident.

5. DISPOSAL OF LARGE VOLUMES OF WASTES

The objective of disposing of radioactive wastes is to confine the radionuclides within the repository site until they no longer represent an unacceptable risk to the environment and the public. A repository should fulfil two important and related functions in this regard: firstly to limit dispersion of the radionuclides contained in the wastes by water-borne and airborne pathways and to protect the waste from surface and near surface deteriorating processes such as erosion or intrusion by humans, burrowing animals or deep-rooted vegetation.

The radionuclides of longer term concern in the soil after an accident at a nuclear power plant are ^{90}Sr and ^{137}Cs, both with a half-life of approximately 30 years. After about 300 years, the concentrations of these radionuclides in soil would be about 0.1% of the concentrations after the accident. Therefore, a storage facility capable of containing these wastes for several hundred years should be suitable for most of the soils collected.

The type of facility selected for disposal of the soil will be dictated by many factors, including the availability of equipment to move the wastes, the volumes to be moved, the distances involved, the availability of natural or man-made disposal sites such as quarries, mines or depressions and the hydrogeology and geology of the area. The basic factors which must be considered in order to achieve a suitable disposal repository system are: the quantity and nature of the wastes, the engineering features incorporated into the repository design, the site characteristics and the time period allowed for institutional control. Conditions are combined in the safety assessment (Fig. 11) to achieve a disposal system that will meet the regulatory or desired environmental protection requirements. For example, a special cover to prevent intrusion by humans would not be required if the institutional control period is expected to be longer than the hazardous life of the wastes.

While the specifics of any accident will affect the disposal plan, some general guidance can be offered regarding disposal of large volumes of contaminated soils.

5.1. Methods for Storing/Disposing of Large Volumes of Wastes

A variety of generic designs are available for the storage/disposal of the very large volumes of contaminated soil and other bulk materials arising from the cleanup after a major nuclear accident. These designs include:

(a) *Natural basins or valleys.* For a valley, an embankment may be required at the downstream end to form a basin. Ideally, these impoundments should be situated at the head end of a natural drainage area. Flow diversion channels could be constructed around the area to control erosion and long term seepage.

(b) *Specially dug trenches.* If suitable transportation is not available, it may be necessary to dig many smaller trenches and bulldoze the wastes into these. The clean fill could be used as a cover and/or to raise the trench walls above the normal ground level. With this approach it may be more difficult to delineate the outer perimeter of the trench and keep track of the many facilities. In addition, small trenches do not use land efficiently. The use of large trenches or specially dug pits is being considered in some countries for the long term storage

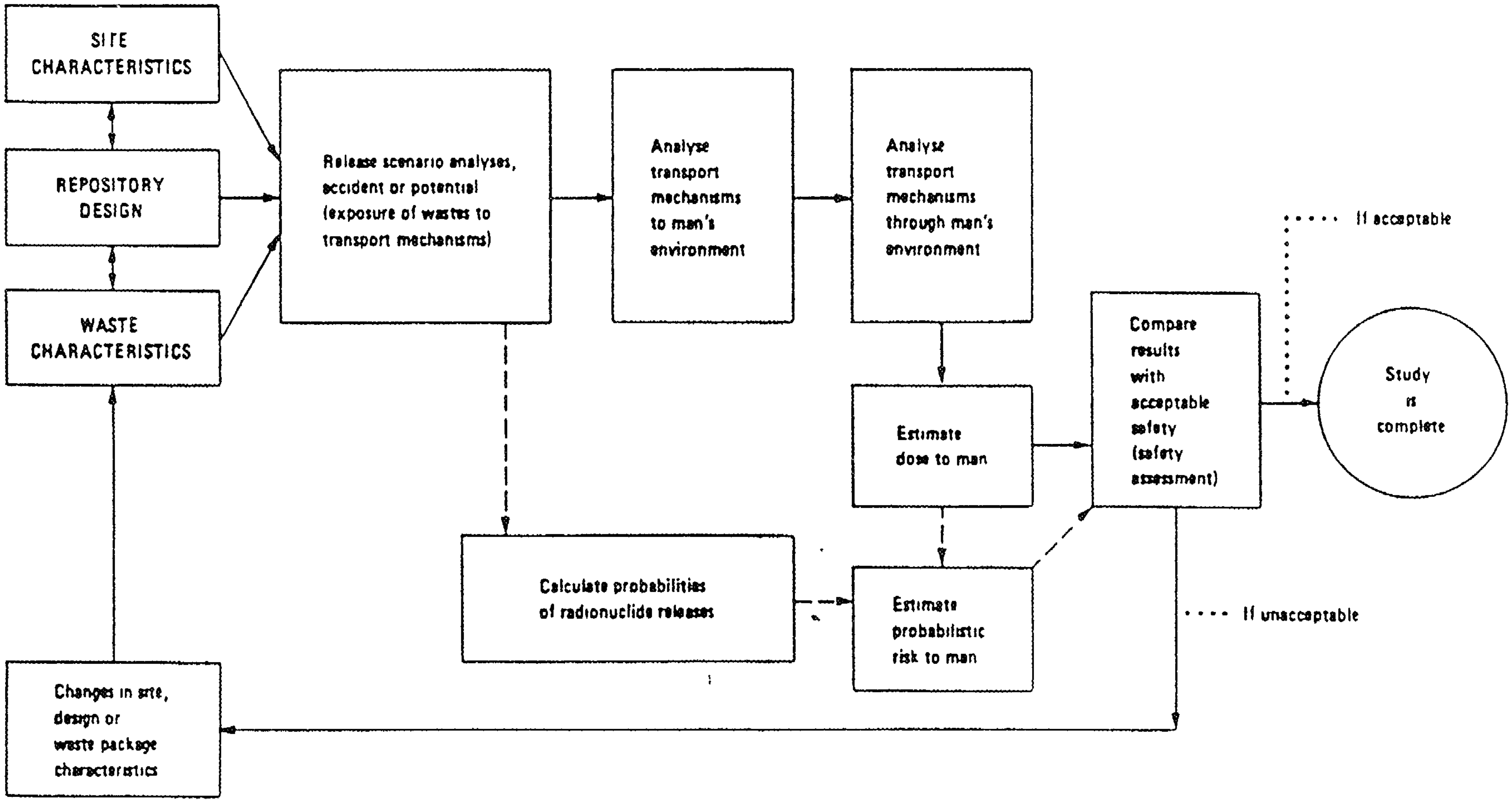

FIG 11: Relationship between safety analysis activities. Dashed lines indicate additional activities when analysising probabilistic safety.

of uranium mill tailings to eliminate the risks associated with possible embankment failures in other facility designs. Large trenches or pits using this engineering technology or that used for well engineered municipal disposal areas could also be constructed for the disposal of large volumes of contaminated soil.

(c) *Mined out quarries or open pit mines.* The possibility of using these depends on climate, groundwater depth and variability, permeability of rock walls, susceptibility of the pit to flooding, etc. If a particular quarry is considered especially desirable, some of the above problems can be reduced by using engineered features such as a rock filled hydraulic bypass, clay lining and a clay-rip-earth cover.

(d) *Underground mines.* Some wastes could be disposed of in underground mines which no longer have any valuable mineral resources. The usefulness of this approach would depend on many factors, including groundwater depth and movement through the mine and susceptibility to flooding. These aspects could be difficult to characterize at short notice.

(e) *Large mounds.* The mounds would be covered with clay, other soil and/or a rip-rap cover of rock.

If necessary, the impoundment facility could be lined with clay (if available) or other impermeable barriers to minimize leakage. Siting of the disposal facility on an area of impermeable clay geology would eliminate reliance on the integrity of an engineered clay liner. Infiltration of precipitation into the waste can be controlled using an impermeable cover such as clay and suitable drainage. Intrusion by man, animals or plants into the wastes can be minimized using a rock rip-rap and/or thicker cover.

Impoundment facilities are currently in use to hold very large volumes of uranium mill tailings during the operational phase of the mill. In these operations, the uranium tailings are pumped as a slurry to fill up impoundments based on some of the generic designs described (Fig. 12). The latest facilities are being designed and closed out so that the release of pollutants such as ^{226}Ra, radon, acids and heavy metals will stay within authorized limits for at least 1000 years. Although the soil arising from a reactor accident will not be in a slurry form, much of the generic information on the construction and closeout of certain designs of mill tailing impoundment facilities would be of great use in designing and building disposal sites for contaminated soils.

The wastes from areas very close to an accident may require special handling and disposal. For example, selected wastes may be collected in containers and buried under the low level wastes. If long lived actinides are present in significant concentrations, the wastes may have to be disposed of in special disposal areas.

In many countries, disposal facilities require institutional control and monitoring programmes until they are finally closed out, using features which prevent intrusion and control seepage within regulatory limits.

The cost to clean up, transport and dispose of large volumes of contaminated material will be high and may have some impact on the selected cleanup criteria through cost-benefit analysis.

5.2. Site Selection

The choice of the location and the method of disposal can be dictated by many factors including economics, availability of equipment, the radionuclides involved, the climate and the availability of disposal sites and their characteristics. The cost of loading, packaging and transporting the very large volumes of wastes from contaminated land can significantly influence the choice of disposal site. Societal implications can also be important but this factor will probably not have a large effect in an emergency situation.

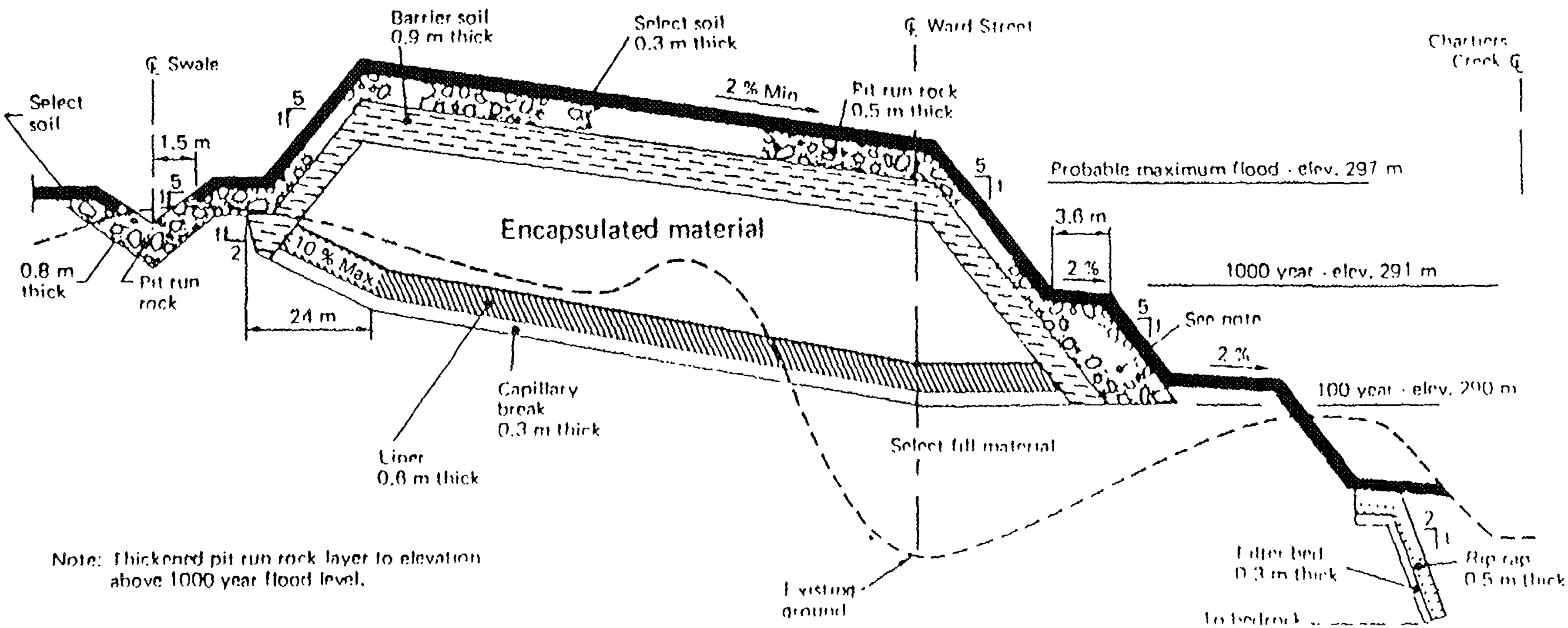

FIG 12: Typical cross-section (not to scale) through encapsulation area of the uranium mill tailings remedial action project site at Canonsburg, PA.

The idealized sequence of investigation for the selection of any waste disposal site has four general phases:

- planning and general studies
- area survey
- preliminary site selection
- site confirmation.

During the preliminary planning, potential sites for the disposal of very large volumes of contaminated wastes could be examined using available data or new core samples if funding is available. However, since even the selection of potential sites could be a very sensitive issue, it may only be possible to do this study in a generic manner and to match repository designs with generic sites in the area. Hydrological considerations in site selection are described in Table IV.

6. COMPLIANCE WITH RELEASE CRITERIA

The decision to implement the cleanup of a contaminated area is made on the basis of the Derived Intervention Levels (DILs) for this protective measure. Once the decision has been made, then cleanup criteria should be available to define the specific radionuclide concentration limit or gamma exposure level which should be achieved by remedial action in a particular area. In addition, re-entry criteria should be established by which it can be decided whether to allow the return of the population and/or reuse of the land for agriculture, etc.

The development of such criteria which relate the dose to humans to contamination levels using pathway analysis is difficult for small sites and extremely difficult for large diverse regions. In practice, different acceptance criteria may be set for different zones or situations in large contaminated areas. Fortunately, by the time large scale cleanup is initiated, only a few longer lived radionuclides would need to be considered in setting criteria.

It is beyond the scope of this report to give detailed guidance on the development of such criteria since it is a specialized task. However, the criteria should be based on risk levels translated into acceptable dose limits. For rural areas, concentration limits for radionuclides in soil, water, air and food or acceptable radiation levels can be derived using suitable pathway analysis and, where possible, realistic site specific parameters. For urban areas, an integrated evaluation of the radiation from various surfaces should be undertaken.

Just as important as the cleanup and release criteria are the validation and quality control protocols required to ensure compliance with these criteria.

6.1. Basic Steps

The basic steps in developing and implementing a plan to ensure that areas, buildings, materials and equipment being released for reuse comply with release criteria include:

(a) Selection of release criteria to be used for each application;

(b) A preliminary survey to assist in defining the scope of the cleanup and what instruments are required;

(c) An assessment of the monitoring and sample analysis requirements and preparation of the required protocols for an efficient and comprehensive compliance survey;

(d) Selection and calibration of instruments;

TABLE IV: HYDROGEOLOGICAL CONSIDERATIONS IN SITE SELECTION

Map/Literature review

Geology
Topography
Precipitation
Evapotranspiration
Nearest surface water
Nearest water use or discharge point

Field reconnaissance

Preliminary

Type of disposal media
Prevailing wind direction
Relief
Subsidence
Slope stability
Flooding potential
Erosion potential
Depth to water table
Depth to fractured bedrock

Intermediate

Existing geological faults
Disposal media
- Sorption capacity
- Thickness
- Engineering properties
- Permeability
- Effective porosity

Structure
Hydraulic gradients
Hydrological budget
Hydrological complexity
Adequate water supply
Monitorability
Remediability

Detailed site analysis

Three dimensional head distribution
Disposal media and underlying site geology (including nearest confined aquifer)
- Water chemistry
- Stratigraphy
- Ion exchange capacity
- Moisture content of unsaturated zone
- Soil moisture tension
- Transmissibility

Natural fluctuation of water table
Flow data for nearest streams including underflow
Water table contour map
Possible measures for groundwater manipulation

(e) Determination of background radiation levels (done during the preliminary planning);

(f) The final survey to ensure compliance with release criteria;

(g) Documentation;

(h) A quality assurance programme which should ensure that the sampling, analysis, monitoring, documentation, interpretation, use of data, etc., would not result in the release of an area having activity levels greater than the designated criteria.

6.2. Costs and Number of Measurements

The cost of ensuring that areas, buildings, materials or equipment being released for reuse comply with release criteria can be highly variable and depends on many factors such as the type and size of the component, the release criteria and labour and analytical costs.

Depending on factors such as future land use, population density, soil type, uniformity of contamination, type of topography and accessibility, and equipment availability, the number of measurements required for verification may vary to a great extent.

Since the number of samples taken during the cleanup operations may be very large, statistical sampling plans should be developed for various zones to minimize the number of samples required and increase the probability that unacceptable levels of contamination are not missed. This sampling plan should be backed up by an appropriate quality control programme. For measurement sets having a large number of samples, the quality control programme, measurement validation function, type of measurement, etc., would be strongly influenced by costs.

Depending on the intended use of a specific type of measurement, differing quality control requirements may be appropriate. For example, less stringent quality control need be applied to initial aerial survey data, since (for various reasons) interpretation of such measurements is difficult, and measurements are in general used either for preliminary direction of cleanup or for final checks of cleanup effectiveness on a broad scale only. On the other hand, quality control for final surveys and for sample and laboratory analyses, including sample preparation, should be stringent, since the instruments are capable of good accuracy and the results could be critical for release of sites.

Selection of quality control criteria should be performed in advance, based on an evaluation of such factors as:

(a) Variation of the parameters being monitored;

(b) The purpose of the particular data set, for example for final dose estimates or preliminary gamma exposure estimates;

(c) The costs of sampling and the funding available.

To control costs and maintain adequate accuracy when quality control for a large number of individual measurements is required, a logical sequence of measurement quality control should be considered in advance. In general, only a small fraction of quality control is performed using expensive laboratory chemical separation analysis. This level of quality control is used only as a final check on the absolute accuracy of well designed field analytical systems, which in turn are used to regularly corroborate inexpensive scan type systems that produce the vast majority of the cleanup measurements.

TECHNOLOGIES FOR RESTORATION OF ENVIRONMENT CONTAMINATED WITH RADIONUCLIDES IN BELARUS

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Abstract

The state of work on creation of technologies for clean-up of the territories of Belarus contaminated as a result of the Chernobyl accident is considered in the report. It is pointed out, that the technologies for decontamination of pre-school medical and prophylactical institutions, schools, zones for recreation, industrial and agricultural objects are used in Belarus.

On the whole, the strategy of changing the residence of population and supervision over the radiological situation is carried out in the Republic.

Clean-up of contaminated soils of large territories is not realized in the Republic on industrial scale.

Presently, the methods have been developed for radiation forecast and determination of advisable extent of decontamination.

The description of worked out technologies for decontaminaton, waste management and disposal is given. The need in development of industrial methods for the soil clean-up is shown.

There are about 1 million Curie of activity at the territory of Belarus. Caesium - 137, strontium - 90, plutonium - 239, 240 are the main contaminants. For the purpose of evaluating the level of contamination in Belarus, the concept of density of contamination has been introduced. This value defines the number of disintegrations on a unit area.

The territory of the Republic of Belarus is divided into some regions depending on density of contamination.The living conditions and measures for decontamination have been developed for each region. The data on development of technologies for decontamination of various surfaces during clean-up operations in Belarus are given in Table I. Three stages of work are considered in it:

- scientific research;
- experimental;
- industrial.

The main fraction of radionuclides is in soil, forest and water ecosystems. The territories with high density of contamination cause the greatest anxiety. Total volume of contaminated arable land, gardens, pastures accounts for 1.645 milliard cubic metres. The major proportion of them are radioactive wastes. Table II gives the data on the volumes of the contaminated soil.

Up to now the large scale clean-up of the territories haven't been carried out in Belarus.

Chemical and physical methods for decontamination procedures, which will be used for soil clean-up operations, are being developed under laboratory conditions. In 1995, it is intended to develop the project on environmental restoration of contaminated territories jointly with French companies.

The special comprehensive method has been worked out at the Institute of Radioecological Problems of the Academy of Sciences of Belarus for determination of advisability of decontamination.

The method determines the advantageous technologies of decontamination and the scope of works. The decontamination experience relates to the clean-up of the soils, water systems, outdoor equipment, buildings, paved surfaces, etc.

The developed technologies cover the waste management cycle, including the problems of decontamination, treatment and disposal.

Radioactive wastes are sorted according to their types and classes.

A new concept has been introduced in Belarus, the concept of conventionally radioactive wastes. They don't fall within the category of radioactive wastes in line with the code of practice. However, they can be dangerous for environment and man.These wastes are defined by the data given in Table III.

The decision about carrying out works on decontamination is made when the permissible levels of contamination are exceeded. The basic data on the normalized levels are given in Table IV.

Let us consider the technology for pulling down contaminated buildings. The demolition of buildings and constructions is carried out according to the design. All due measures on dust suppression are taken before and while carrying out works. The State Sanitary Inspection and the Ministry for Emergencies are carrying out the control over adherence to the norms of radiation safety.

All materials are divided into three categories:

Clean materials are the materials with the level of contamination by beta-particles up to 20 beta-particles/cm^2 min. Clean materials are stored in special sites before moving away to the clean zone. Their removing is carried out through the control posts between dirty and clean zones with monitoring at the outlet. The materials with activity 20 - 50 beta-particles /cm^2 min can be used in building trade only after special treatment. The treatment is carried out for reduction of activity below 20 beta-particles/ cm^2 min.

The materials with the level of activity from 20 to 50 beta-particles/cm^2 min are called, as it has been mentioned above, the conventionally radioactive wastes. Such wastes are stored in organized disposal areas.

Materials with the level of activity exceeding 50 beta-particles/cm^2 min are disposed in special facilities.

Decontamination of pre-school medical and prophylactic institutions and schools is carried out in the following order:

- decontamination of sites with anomalously high level of contamination;
- decontamination of buildings and rooms;
- decontamination of roads.

In decontamination of grounds for games and sport activities, they are removed and replaced with clean ones. The replacement of the soil is ensured for the sites used for growing plants.

TABLE I: THE LIST OF TECHNOLOGIES FOR DECONTAMINATION DEVELOPED IN BELARUS

		Scientific study	Experimental specimen	Industrial method
1	Removal of radioactive soil	X	X	X
2	Technology for soil clean-up	X		
3	Decontamination of pre-school medical and prophylactic institutions and schools	X	X	X
4	Decontamination of the zones for recreation and residence sites	X	X	X
5	Clean-up of private plots of cultivated land	X	X	X
6	Decontamination of industrial and agricultural objects, populated areas	X	X	X
7	Integral treatment of liquid radioactive wastes	X	X	
8	Integral technology for decontamination of organic wastes	X	X	
9	Integral technology for decontamination of ash	X	X	
10	Decontamination of food products	X	X	X

TABLE II. VOLUME OF CONTAMINATED SOIL

Land	Density of contamination, Bq/km^2	Area, thousands of hectares	Volume of soil, mln m^3
Arable lands	$3.7\ 10^{10}$ – $18.5\ 10^{10}$	1045	1045
Gardens, private plots of cultivated land	$18.5\ 10^{10}$ – $55.5\ 10^{10}$	395	395
Pastures, meadows	$55.5\ 10^{10}$ – $148\ 10^{10}$	141	141
Homesteads	up to 40		64.6
TOTAL			1645.6

TABLE III. CHARACTERISTICS OF CONVENTIONALLY RADIOACTIVE WASTES

Type of wastes	Level of contamination (LC)		
Beta-active materials	7.4 kBq/kg ($2\ 10^{-7}$ Ci/kg)	< LC <	74 kBq/kg ($2\ 10^{-6}$ Ci/kg)
Gamma-active materials for caesium-137	$9.62\ 10^2$ Bq/kg	< LC <	$9.62\ 10^3$ Bq/kg
Alpha-active materials	0.74 kBq/kg ($2\ 10^{-8}$ Ci/kg	< LC <	7.4 kBq/kg ($2\ 10^{-7}$ Ci/kg)
(For transuranic elements)	37 Bq/kg ($1\ 10^{-9}$ Ci/kg	< LC < < LC <	$3.7\ 10^2$ Bq/kg ($2\ 10^{-8}$ Ci/kg)
Surface (determinetion on the area $100 cm^2$)	20 beta-particles/cm^2 min < LC < < 20 beta-particles/cm^2 min		
Surface (determination on the area 100 cm^2)	3 alpha-particles/cm^2 min < LC < < 3 alpha-particles/cm^2 min		

TABLE IV. NORMALIZED VALUES

No	The investigation objects	Derived limits: Exposure dose rates (mkR/h)	Derived limits: Beta contamination (part./cm^2 min)
1.	Territories of pre-school medical and prophylactic institutions, schools	35	isn't normalized
2.	Territories of private plots of cultivated land	40	isn't normalized
3.	Indoors of pre-school medical and prophylactic institutions and schools, living quarters	25	isn't normalized
4.	At working places and in office premises:		
	- permanent residence	50	isn't normalized
	- temporary residence	100	isn't normalized
5.	Territories of the objects of industry and other open territories of populated areas	60	isn't normalized
6.	Inner surfaces of dwelling houses and private property inside them	isn't normalized	15
7.	Inner surfaces of pre-school medical and prophylactic institutions and schools and the surfaces of equipment inside them	isn't normalized	15
8.	Inner surfaces of office premises, social, industrial buildings and the surfaces of equipment istalled there	isn't normalized	20
9.	External surfaces of buildings	isn't normalized	20
10.	Outdoor equipment (internal and external surfaces)	isn't normalized	20
11.	Roofing of buildings	isn't normalized	40

Unpainted wooden constructions, such as townships for children games, household buildings are not intended for decontamination. They are subject to dismantling and replacement.

Exceeding the control levels isn't permitted even in the isolated sites.

The sites with anomalous high level of contamination are usually decontaminated by removal of surface soil. The contaminated layer is removed using common outdoor equipment and specially designed techniques.

Decontamination of the zones for recreation and the people residence sites is carried out in two stages:

- decontamination of anomalous spots with high activity;
- decontamination of the entire site.

With low levels of activity, the overlaying the soil with clean topsoil can be used, followed by sowing the perennial plants.

Decontamination of household sites is carried out in three stages:

- decontamination of anomalous spots with high activity;
- decontamination of household buildings;
- decontamination of private plots of cultivated land.

The household buildings are completely demolished, if wooden, porous blocks and asbestos cement have been used as building materials.

In the places of domestic animals maintenance, the complete removal of manure and the under-layer is made.

When realizing the works, separate collection of liquid and solid wastes generated in decontamination is arranged. Liquid wastes are collected into leak-tight reservoirs and sent for treatment. Solid wastes are graded into three types:

- domestic wastes;
- conventinally radioactive wastes;
- radioactive wastes.

The following measures are foreseen for prevention of the secondary contamination:

- the works are carried out in dry windless weather;
- the wastes are collected at the end of each day;
- wastes are transported skirting the populated areas.

In the process of work the DRGZ - 0.2, DRG - 01T1, DRG - 05, DVG - 06T devices are used for measurement of the dose rate.

The MKS 01P1 radiometer - dosimeter and the universal RUP - 1 radiometer are used for measurement of beta - particle densities.

For decontamination of places with hard covering, sweeping, high pressure jetting from the firehosing are applied.

The removal of the soil is primarily practised for decontamination of surfaces without hard coverings.

Decontamination of industrial and agricultural enterprises begins with anomalously contaminated sites.

Decontamination of the spots at the sites with hard coverings is implemented by simple mechanical cleaning and surfactants. When there is no results, mechanical stabilizers are used to physically cover the contamination. They include concrete and asphalt protective covers. Decontamination of spots at the sites without hard coverings is executed by removal of the contaminated spot. The isolation of the spot is admitted as an exception.

Decontamination of equipment is performed as follows:

- mechanical removal of radioactive dust;
- decontamination of the outer surface;
- decontamination of separate components in special bathes.

If the permissible levels of contamination are not achieved after decontamination is carried out three times, the decision is made about the replacement of equipment.

Technological process of decontamination of machinery is conducted in two stages:

- removal of oils and soil with the help of brushes and scrapers;
- removal of contamination using decontaminating solutions.

In order to decontaminate rubber, decontamination is carried out two times:

- 5 % soda solution; 0.1 % potassium permanganate and 0.4 % potassium hexametaphosphate;
- 2 % nitric acid solution, 0.2 % oxalic acid, 0.2 % sodium fluoride and 0.5 % detergents.

Decontamination of the ventilation systems of industrial enterprises is carried out with the help of decontaminating solutions. Decontamination of heaters in ventilation chambers is accomplished by steam with addition of surfactants. Table V summarizes the recommended detergents for decontamination of equipment surfaces, machinery and ventilation systems.

Presently, paste-like compositions are prepared for decontamination of contaminated metal unpainted surfaces. They make it possible to remove 93 - 97 % of caesium-137 and 83 - 95 % of strontium-90.

The data for painted surfaces are 91 - 95 % and 79 - 95 %, correspondingly.

The efficiency of decontamination substantially depends on the deadline of putting the equipment in dead storage. When this date is 5 - 10 days, the total reduction of activity is 95 %. With the deadline equal to 5 - 6 monthes, the reduction is only 35 %. In addition, it has been established, that the waste water is not contaminated with radionuclides.

In the process of decontamination, liquid and solid radioactive wastes are generated.

In the Institute of Radioecological Problems of the Academy of Sciences of Belarus two installations have been designed for treatment of liquid radioactive wastes.

The first installation is operated on the principle of chemical co-precipitation followed by separation of solid and liquid phases.

The second installation takes as the basis the evaporation of rotatable liquid flow.

The process of volumetric evaporation is carried out in this apparatus. Liquid is preheated and then the evaporation of whirled liquid occurs, when pressure is reduced. It should be noted, that such eddy evaporator is the separator at the same time. It decontaminates liquid radioactive wastes of any composition.

Fig. 1 gives the basic elements of the installation for processing liquid radioactive wastes. The installation embodies the following principle:

Liquid is heated and conveyed by the tangential channel into the evaporation chamber. On the certain radius of rotation liquid comes to the boil. Vapour bubbles move to the axis of the evaporator with the velocity 10 - 20 m/sec and liquid moves to the periphery. The main technical characteristics of the installation are as follows:

1. Initial temperature of liquid radioactive wastes is 278 - 298 K.
2. Capacity is 0.05 - 0.01 kg/sec.
3. Operating conditions for pH are not specified.
4. Decontamination factor is 10^5.

TABLE V. RECOMMENDED DETERGENTS FOR DECONTAMINATION OF SURFACES OF EQUIPMENT, MACHINERY, VENTILATION SYSTEM

Composition	Component of solution	Quantity	Notes
N 1.	soap-powder alkali water	3 g 10 g 1 g	Removal of unfixed and weakly fixed contaminations
N 2.	DS - SAC water	10 g up to 1 l	Removal of unfixed and weakly fixed contaminations
N 3.	DS - SAC oxalic acid sodium chloride water	10 ml 5 g 50 g up to 1 l	Removal of unfixed and weakly fixed contaminations
N 4.	DS - SAC oxalic acid potassium hexametaphosphate water	5 g 5 g 7 g up to 1 l	Removal of unfixed and weakly fixed contaminations
N 5.	potassium permanganate alkali water	5 g 50 g up to 1 l	Removal of strongly fixed contaminations when decontamination is carried out in two bathes
N 6.	potassium permanganate sulphuric acid water	40 g 5 g up to 1 l	Decontamination of surfaces, which are not subject to cleaning with solutions N 1 - 4
N 7.	alkali water	10 g up to 1 l	For processing the surfaces of material non-resistant to action of acids
N 8.	citric and oxalic acids water	10 - 20 g up to 1 l	Recommended for decomposition of expensive equipment
N 9.	trisodium phosphate or sodium hexa-metaphosphate water	10 - 20 g up to 1 l	Damaged surfaces are to be restored
N 10.	DS - 10 water	5 - 10 g up to 1 l	Degreasing the surfaces

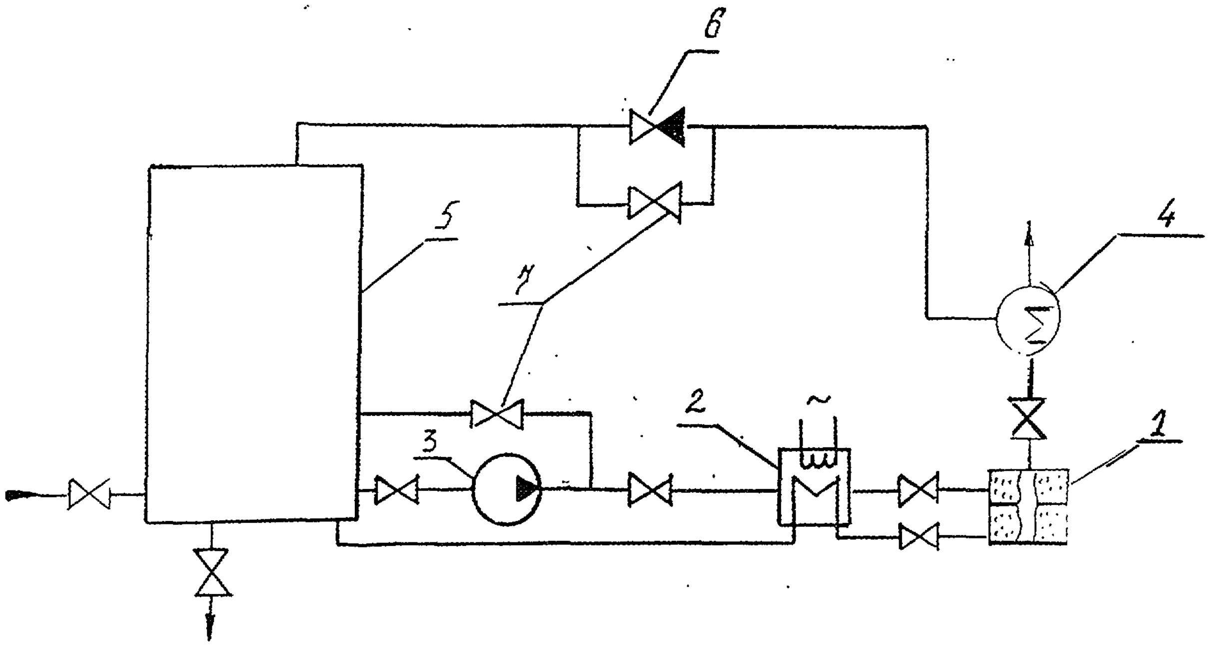

Fig. 1. Basic elements of the installation for processing liquid radioactive wastes:

1 - eddy evaporator;
2 - heater;
3 - pump;
4 - condenser;
5 - reservoir;
6 - safety valve;
7 - locking fitting.

Special reagents have been prepared for dust suppression. They can be used in carrying out the decontamination and for prevention of radioactive dust moving from the soil. The reagents have been prepared on the basis of the by-products and wastes of food and forest industries and other enterprises of Belarus.

Investigation have been carried out under laboratory conditions, and experimental units on reclamation of organic radioactive wastes have been developed. The unit with the boiler in the boiling bed is of interest.

The works on development of sorption systems for decontamination of milk and juice from radionuclides are carried out. Such systems are selective in their effect.It has been found out, that clean-up by sorption reduces the milk activity from 40.000 Bq/l to 185 Bq/l. In addition, the main edible properties of the product are preserved. The conditions for application of this method in industry and in the private sector have been perfected. The batch of sorption filters has been manufactured at the enterprises of Belarus and the plant for the clean-up of milk has been built in Gomel'.

The conducted investigations have shown, that when burning up firewood, the active ash is generated. Thus, in towns with density of contamination by caesium-137 equal to 3.7 10^{10} - 18.5 10^{10} Bq/km^2, 7.1 thousand tons of radioactive ash are generated, and in towns with density of contamination 18.5 10^{10} Bq/km2 it is 4.6 thousand tons. Every year 44.1 thousand tons of contaminated ash is generated in the towns, 14.8 thousand tons are radioactive wastes. Table VI gives the results of studying the ash generation in 14 populated areas of Belarus, and Table VII gives the quantity of ash being radioactive wastes.

TABLE VI. QUANTITY OF ASH WITH VARIOUS LEVELS OF ACTIVITY GENERATED ANNUALLY IN 14 TOWNS of BELARUS

Town	Number of private houses	Quantity of ash for a house	Quantity of ash, t			
			Total	3.7 10^4 Bq/kg	3.3 10^3 Bq/kg	3.7 10^2 Bq/kg
Zone 3.7 10^{10} - 18.5 10^{10} Bq/kg			100%	16%	76%	8%
Stolin	676	0.72	487	78	370	39
Luninets	1569	0.72	1130	181	859	90
Kostyukevichi	1815	0.72	1307	209	993	105
Krasnopol'e	1053	0.72	758	121	576	61
Total in zone	5113		3682	589	2798	295
Zone 18.5 10^{10} - 55.5 10^{10} Bq/kg			100%	18%	74%	8%
Slavgorod	1282	0.72	927	167	686	74
Cherikov	1566	0.72	1128	203	835	90
Bykhov	2566	0.72	1847	332	1367	148
El'sk	1208	0.72	870	157	644	69
Khojniki	1800	0.72	1296	233	959	104
Total in zone	8427		6068	1092	4491	485
Zone more than 55.5 10^{10} Bq/kg			100%	33%	65%	2%
Bragin	987	0.72	711	235	462	14
Narovlya	1092	0.72	768	259	511	16
Vetka	1621	0.72	1167	385	759	23
Chechersk	1141	0.72	822	271	535	16
Korma	661	o.72	478	158	310	10
Total in zone	5505		3964	1308	2577	79
Total:	19045		13714	2989	9866	859

TABLE VII. QUANTITY OF ASH, WHICH IS SUBJECT TO DISPOSAL, IN 14 TOWNS OF BELARUS

Zone of contamination	Number of ash samples: Total in zone	Including more than 10^3 Bq/kg: number	Including more than 10^3 Bq/kg: %	Quantity of ash, t: Total in zone	Quantity of ash, t: Including radioactive wastes
3.7 10^{10}-18.5 10^{10} Bq/km^2	25	11	44	3682	1620
18.5 10^{10} -5.5 10^{10} Bq/km2	62	36	58	6068	3520
55.5 10^{10} Bq/km^2	66	51	82	3964	3250
TOTAL:	153	98	61	13714	8390

Presently, the comprehensive technology for ash decontamination and disposal is being developed.

The collection of ash into special containers is foreseen, including its compaction and disposal.

Special compositions for covering the constructions of repositories for ash have been created. It is shown, that coatings based on bituminic emulsions are the most reliable. The two-layer coatings based on bituminic emulsions increase their efficiency. Ground - bituminic emulsion compositions possess high protective properties. They are used for walls and bottoms of repositories for ash disposal.

Sediments of sewages are of great radiation danger. The part of them are radioactive wastes. Total volume of sewages amounts to 32840 m^3. It should be noted, that sediments of sewages are evaluated in terms of presence of more than 50 % of soluble forms of caesium-137. Owing to this, the comprehensive technology ensuring radiation safety of sediments of sewages are being developed.

Fires in contaminated forest are of great danger. In this connection, the aids for putting out fires and fire-protective means have been developed. They have been tested in the special fire testing grounds. The ability to fire retardation for 3 - 6 days with the amount 1.5 l/m^2 has been shown.

The works on selection of the location of perspective disposal sites have been carried out. The principles and the methods of shallow - ground and surface disposal have been worked out. Six types of sites have been assigned, ranging from favourable to useless. Ecologically safe sites have been found out. The following areas have been chosen for radioactive waste disposal sites:

The Gomel' Region:
- Narovlya area - "Karpovichi" site;
- Khojniki and Bragin area - "Babchin - 4" site;

The Brest Region:
- Stolin area - Ol'myanskaya Koshara" site;
- Luninets area - "Dobraya Volya" site

Fig. 2 shows the location of the disposal sites on the map. The codes of practicies have been worked out for carrying out the works on decontamination. They determine the sequence and technologies for works on decontamination, demolition of buildings and waste management.

On the basis of above, the following conclusions can be made:

1. The strategy of changing the residence and the control is carried out in Belarus.
2. Decontamination is of local character.The populated areas, industrial and agricultural objects are the main subjects for decontamination. The most contaminated territories have been turned into strict nature reserves.
3. For ensuring radiation safety, it is necessary to develop the advantageous methods of decontamination of large territories.

Fig. 2.

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URANIUM INDUSTRY IN BULGARIA AND ENVIRONMENT: TECHNOLOGIES AND IMPLEMENTATION OF ENVIRONMENTAL RESTORATION PROJECTS

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Nuclear Power Plant at Kozloduy, Kozloduy

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Abstract

The paper presents a summary of the environmental restoration process in Bulgaria before the termination of the uranium industry (August 1992) when some sites due to depletion had been closed and also the restoration procedure which are currently applied. The methods for rehabilitation depend on the type of site and therefore the uranium mines, the milling plants and the auxiliary units are discussed separately. At present all sites have been ecologically assessed and for most of them the restoration technologies are selected. Part of the land is restored and returned to its owners. For the in-situ leaching sites long term monitoring of underground water is necessary. The restoration activities must be synchronized since residual ore has to be processed and waste has to be dumped in tailing ponds and underground mines. It has been pointed out that there is a need for cheap and proven technologies since the closed uranium industry has to provide the funding for the restoration process and there are difficulties due to the general reduction of industrial production in the country.

1.Introduction - review of the uranium industry in Bulgaria [1,2]

The uranium industry in Bulgaria started immediately after the II World War with classical underground mining and hydrometalurgical processing of the ore to uranium concentrate. The total amount of uranium concentrate produced in the period 1947-1992 is approximately 35000 t or 800 t/annually in the last years before the closure of the uranium industry.
Approximately 34 uranium mining sites exist which include 4 open pit mines, underground mines and in situ-leaching mines (after 1968-69).

The total amount of tailing pond wastes of the two milling plants in Buhovo and Eleshnitsa (3 tailings) is 16 000 000 t and approximately 3.1 PBq of stored activity. The total number of the waste heaps is 298 with approximately 13 720 000 t of dumped mass which covers approximately 845 000 square meters.

Up to 1958 no tailings pond existed at the Buhovo plant and the result is 1 200 000 square meters contaminated with radium land along the rivers Yanishtitsa and Lesnovska.

Along the valleys of the rivers Maritsa, Tundzha and Struma approximately 15 low grade ore deposits were processed by in-situ leaching with sulfuric acid. The surface communications (tubes) cover approximately 16 000 000 m^2 agricultural land and forests. Some of the heaps of the underground mines have been processed by combined in-situ leaching technologies.
The total contaminated area affected by the uranium industry is approximately 20 000 000 m^2, including 4 000 000 m^2 forest.

At present the uranium industry is terminated by a government decision and the necessary funding for restoration of the former uranium sites is expected to be raised by selling of equipment.

2.Technologies and procedures for environmental restoration applied before the termination of the uranium industry.

A number of uranium sites were closed before the decision for total termination of the uranium industry (see Table 1 in [1]). Many mines have been closed in the Buhovo region, mines near the towns of Sliven and Melnik, near the Haskovo mineral baths, above the Rila monastery. In-situ leaching sites have been also closed - near the village of Prepechene and Zlatolist in South-West Bulgaria (along the Struma river), some sites of the mine "Pioneer" near the village of Orlov dol and also sites in the Upper Tracian Plane.

A common feature for all sites is that no environment remediation actions followed after the technical closure. In some cases the monitoring had also been terminated. The environmental monitoring was resumed within the procedures for radioecological assessment and decision taking for site remediation.

The only procedure which has been in implementation is the closure of the entrances of shafts and adits but long afterwards poorly closed exits used to emerge as caving or downfalls occurred.
There are still abandoned trolleys, broken equipment, repair shops, administrative buildings. Only some reloading sites being close to railroad stations have been cleaned-up.

The operation of the first tailing pond of the Buhovo plant has been terminated long before the closure of the uranium industry but no actions followed - no fence was built and it often served as pasture ground for cattle. The migration of water due to tailing seepage is controlled by regular sampling of water in control shafts down the tailing pond. Control of the water is necessary since no tailing impoundment (liners, cover layers) had been implemented when the pond was built.

No water treatment facility has been built after the closure of any site, no actions were taken against wind and sheet and rill erosion of the waste heaps. Only some sorption facilities for excess water have been used during the operation of several mines in order to extract the uranium from water which otherwise will be lost ("Deveti septemvri", "Druzhba", "Dospat").

In the period 1985-1990 remediation procedures were applied for closed sites of the mine "Pioneer" according to a method developed in the Institute of Soil "N.Pushkarov" (Sofia). The method is intended for a period of approximately 5 y and includes radioecological survey, cleaning-up, neutralization, intensive natural fertilization.

Before the termination of the uranium industry a project was started for restriction of the propagation of the contaminated underground water of sites "Okop" and "Tenevo" (mine "Pioneer") and site "Cheshmata" (Mine "Parvi mai"). The project has been developed in the former USSR and included pumping of chemical agents - e.g. sodium silicate solution (soluble glass) around a leaching site in order to "seal" the underground contaminated water. The project was not realized , only some preliminary experimental observations can be used for further estimation for the migration velocity of contaminated water.

3.Technologies for environmental restoration applied after the total termination of the uranium industry.

At present all uranium industry sites are radioecologically assessed and most of summary information was reported in the previous meetings [1,2].

Most of the restoration work has been carried out for the sites of in-situ leaching. Data on rented areas, remediated areas up to 1 of January 1992 and projected areas for remediation after January 1992 are summarized in Table 1.
During the operation of sites of in-situ leaching the humus layer had not been removed along the piping and the boreholes and therefore detailed radiometric mapping and sampling for chemical analysis is necessary. Only the forest lands of the in-situ leaching sites (approximately 25%) cannot be restored and it is useless to restore them.

According to us the most serious hazard comes from the chemical contamination of subsurface layers and contamination of water. The most dangerous sites are "Cheshmata" (Haskovo) and "Tenevo-Okop" (Topolovgrad) near which there are catchment facilities for drinking water. In all hydroecological assessments passive systems for monitoring of the underground migration of contaminated water are suggested hoping that the hydrobarriers

Table 1
Rented land, remediated land and returned to the owners land up to 1 of January 1992 and projected land for remediation after January 1992, square meters.

SITE	*AREA *10^3M^2*	*REMEDIATED mechanically + biologically*	*REMEDIATED mechanically*	*PROJECT mechanically + biologically*	*PROJECT biologically*
mine CARIMIR	*918.7*	*238.7*	*155***	*182*	*155*
-Carimir	*680*	-	*155***	*182*	*155*
-Ceretelevo	*238.7*	*238.7*	-	-	-
mine HASKOVO	*2113.5*	*246*	*272*, 32***	*851*	*321*
-Cheshmata	*621*	*175*	*72*, 256***	*194*	*256*
-Maritza	*71.5*	*71.5*	-	-	-
-Navusen	*622*	*200*, 65***		*557*	*65*
-Debar	*799*	-	-	*100*	-
mine TOPOLOVGRAD	*4393.2*	*1538*	*786*, 855***	*786*	*855*
-Orlov dol+Iztok	*2422.5*	*1054.5*	*696*,665***	*596*	*665*
-Vladimirovo	*201.2*	-	*190***	-	*190*
-Mudren	*483.5*	*483.5*	-	-	-
-Chukarovo	*190*	-	*190**	-	*190*
-Tenevo	*230*	-	-	-	-
-Okop	*656*	-	-	-	-
-Dobroselec	*210*	-	-	-	-
mine BELOZEM	*7342*	*2091*	*155*, 258***	*718*	-
-Belozem	*1978*	*848*	-	-	*258*
-Trilistnik	*498*	-	*258***	-	*258*
-Momino	*2155*	*660*	-	*284*	-
-Rakovski	*2711*	*583*	*155**	*795*	-
mine SELISHTE	*3853*	-	-	*450*	-
Total TRAKIA RM ltd	*18620.4*	*4114.1*	*1213*, 1589***	*4062*	*1589*

**technical restoration without hole sealing;*
*** technical restoration without neutralization*

will seal the water layer and consequently the contaminated water will be desalted. We are also afraid that the calculated rates of migration are optimistic although we understand that construction of adequate purification facilities for enormous water volumes is very expensive and there are no sound arguments that they are absolutely necessary.

Data on quantity of uranium and specific activity of radium in water from different mines (Table 2) show that uranium is below the accepted limit for surface water (0.6 mg/l) but the activity of Ra is generally above the limit (0.15 Bq/l). Continuous observations show that several years after the termination of mining there is a trend for water purification and Ra reduction since Ra is transported by the mechanical particles.

At the present moment (end of 1994) two projects are being financially supported within the PHARE programme - restoration of the tailing pond region of Eleshnitsa and restoration of the Bay of Vromos radioactively contaminated by dumping of slurry of copper mines [1]. The projects are in very initial stages and neither the methods nor the organization which will perform the restoration have been selected.

Table 2
Content of uranium, mg/l, and specific activity of radium, Bq/l, in uranium mine waste water

		uranium, mg/l				*radium, mBq/l*		
mine	*N**	*min*	*max*	*average*	*N**	*min*	*max*	*average*
Deveti spetemvri	*25*	*0.04*	*0.37*	*0.19*	*12*	*52*	*5143*	*1162*
Seslavtsi	*24*	*0.01*	*1.05*	*0.20*	*12*	*592*	*1628*	*696*
Smolyan	*22*	*0.03*	*0.97*	*0.34*	*16*	*118*	*13209*	*3537*
Eleshnitsa	*19*	*0.02*	*1.12*	*0.31*	*9*	*599*	*20460*	*7130*
Dospat	*20*	*0.02*	*0.43*	*0.12*	*8*	*9.3*	*6327*	*1898*
Selishte	*19*	*0.04*	*0.80*	*0.14*	*8*	*140.6*	*18315*	*8095*
Sliven	*18*	*0.01*	*0.37*	*0.10*	*13*	*344*	*26085*	*5990*
Melnik	*11*	*0.01*	*0.97*	*0.26*	*7*	*203.5*	*8584*	*270*

N - number of samples*

A small experimental facility for bacterial sulfate reduction for the excess water from the tailing pond in Buhovo is now in the process of construction [3]. The bacterial removal of sulfates from the washwater results also in simultaneous precipitation of heavy metals.

There are also other propositions but a major obstacle is the lack of financing.
Another major obstacle for accurate assessment of the impact of radioactive contamination is the lack of national levels for natural radionuclides in soil, plants and animal products. Only lately such a project is under development - development of levels and criteria for protection of the population which live in regions with increased natural radioactivity.

4.Technologies for environmental restoration of uranium mining and milling sites

The methods for rehabilitation depend on the type of site and therefore the uranium mines, the milling plants and the auxiliary objects will be discussed separately. All methods which are implemented or are intended to be implemented are "classical" and described in earlier publications [4,5]

The restoration activities must be synchronized since:

all existing ore and ion-exchange resins have to be processed in the milling plants;
radioactive waste has to be dumped in tailing ponds;
part of the mined rock and equipment can be dumped in underground mines before sealing.

4.1. Uranium mines - underground mines, open pit mines, in-situ leaching sites.
4.1.1. Underground mines.

The methods for restoration include:
- closure of adits, shafts, ventilation shafts;
- transportation of mined ore (if any) to the chemical plants;
- water precipitation or purification,;
- heap stabilization , leveling and afforestation., or leveling , isolation and afforestation. In the case of heap leaching , a neutralization is necessary;
- ban on house building on the heaps, prevention on the use of rock material for use in buildings, especially for houses for living. possible use as rock for roads.

There are different alternatives for heap stabilization. For the isolation technologies because of the large total area (845 000 m^2) the experts are looking for cheaper covering material which has been experimentally verified.

4.1.2. Open-pit mines.

The methods for restoration include:
- site leveling, filling when possible;
- treating of the water collected in the region - precipitation or purification;
- ban on house building,
- use of the pit, if possible, for lake for recreation activities.

4.1.3. In-situ leaching sites

The main method for leaching in Bulgaria is the acid method, the soda methods had been applied only in experiments. For the in-situ leaching additional reagents have been used for intensification of the process - iron sulfide, potassium permanganate, sodium nitrite. Some metals - Mo, Cu, Zn, Ni, W, Cr, Fe, Al, V, and also rare earths - La, Ce, Yb, are also present in the solutions.

The methods for restoration include mechanical and biological technologies:
- removal of radioactive wastes of contaminated earth;
- deep sealing of boreholes in order to prevent direct water contact;
- neutralization of the contaminated with acid or soda areas (liming);
- leveling and intensive fertilization with natural fertilizer;
- "green" fertilization - ploughing of lucerne etc. before ripening;
- crop rotation, e.g. corn-rye-sunflower-rye etc.;
- drying or irrigation;
- grass, or planting fruit trees;

The methods for water restoration include:
- recycling of the solution for increasing the pH;
- neutralization of the water in the layer or on the surface;
- salt purification;
- generation of hydrochemical barriers;
- control of the eventual spread of contamination;
- ban on hole drilling for irrigation or drinking water.

4.2. Uranium milling plants.

The milling plants process ore and the ion exchange resins from the in-situ leaching sites. The wastes - slurry and water are deposited in the tailing ponds. The rehabilitation procedures concern the plant site and the tailing pond. For the specific case of the Buhovo plant, the Yana-Bogrov region needs remediation and counter-measures because of the contamination of large areas with Ra [1].

The methods for restoration of the plant site include:
- cleaning-up of the radioactive wastes and dumping the wastes in the tailing pond;
- deactivation of the plant premises;
- disassembling of filters, mills, transport belts etc.;

The methods for restoration of the tailing pond include:
- strengthening of the tailing dam wall;
- routing of the clean water away from the tailing pond;
- covering the pond with a layer for prevention of radon exhalation - clay, asphalt, polymers;
- covering the isolation layer with soil;
- planting of grass or trees;
- management of the tailing seepage of surface or underground water - neutralization (if necessary), purification for sulfates, heavy metals, radium;
- creation of a system for long term monitoring.

The methods for restoration of the radium contaminated areas of the Yana, Gorni Bogrov, Dolni Bogrov region [1] include:
- removal of the most radioactive spots;
- afforestation of the contaminated areas;
- covering with soil of tailing pond wastes;
- control of utilization of the low contaminated areas.

4.3. *Auxiliary sites - warehouses for ore and chemicals, repair shops, transport units, research laboratories, drill core samples store.*

The remediation measures are site-specific but the most common procedures are:
- clean-up of the sites of radioactive wastes;
- transportation of highly radioactive ores, solutions, resins, drill cores etc. to tailings ponds;
- disassembly of contaminated facilities;
- deactivation of buildings;
- excavation and transportation of sludge from purification facilities to tailing ponds or underground mines;
- planting of grass or trees;

5. *The case of the Bay of Vromos - coastal radioactive contamination from copper floatation plant*

The case of the Bay of Vromos has been described in [1]. During the period 1954-1977 the waste from a flotation plant which concentrates the ore from a copper mine has been dumped in the sea near the coast. The total waste is estimated to be approximately 8 000 000 t. On the coast the waste is 1300 m long, 120 m wide and the thickness of the layer is 2 - 3 m. The increased exposure rate is associated with increased content of U-238 (by a factor of 10 - 300 the natural concentrations) and Ra-226 (5 - 200). The measured exposure rates are shown in Table 3.

In 1991 a project started (partially supported by the PHARE programme) for recycling the dumped mass since preliminary experiments showed it will be economically profitable. The sand mass is recycled in the copper plant and the results is a copper concentrate (13-15%), iron concentrate (55%), gold (4-5 g/t). The waste varies within 70-85% from the initial mass

Table 3
Exposure rate and associated area in the Bay of Vromos

exposure rate, μSv/h	*area, m^2*
max 2	*200*
>1	*7000*
>0.3	*208000*

[6]. The preliminary experiments confirmed the assumption that natural processes have "enriched" the dumped mass.
The mass which is expected to be reprocessed is 216 000 t, from which ~700 t copper and ~32 000 t iron will be obtained.

The sand is transported to the copper concentration plant by trucks, the sand from the sea (10-20 m) will be dragged by chain-drag.
The measured values of the exposure rate shows that already from the places where the dump has been reprocessed the background is reduced twice.

6. Conclusion

The procedures for environment restoration of the uranium industry sites have been started. The first steps are radioecological and hydroecological assessment. Some of the sites are in the process of restoration but for the further actions there is not enough funding - the uranium industry after its termination with a government prescription is expected to supply the necessary financing. Due to the general reduction of industrial production, classical mining is also in a difficult position and therefore there is no buyer for the equipment from uranium mining industry.
International projects can help the restoration process especially by exchange of knowledge on experimentally verified cheap restoration procedures.

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TECHNOLOGIES FOR AND IMPLEMENTATION OF ENVIRONMENTAL RESTORATION IN CANADA

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Abstract

This paper discusses the technologies for, and implementation of, environmental restoration at three types of sites present in Canada:

- at historic low-level radioactive waste (LLRW) sites, resulting from the early years of the radium and uranium industry in Canada, which are the responsibility of the Low-Level Radioactive Waste Management Office (LLRWMO).
- the Chalk River Laboratories (CRL) site of Atomic Energy of Canada Limited (AECL);
- decommissioning and waste management at uranium mining and milling sites.

The latter section deals primarily with sites for which the uranium production companies are responsible for decommissioning. It has been included to describe Canadian experience in this area since, from a technical perspective, there are many factors in common with restoration programs at inactive or abandoned sites.

1. INTRODUCTION

This is the third of three papers presenting Canadian experience in environmental restoration at radioactively contaminated sites. The first two papers dealt with identification and radiological characterization of contaminated sites in Canada [1] and planning for environmental restoration [2].

The sites which are the subjects of these papers have been primarily, but not exclusively, associated with mining, processing, transport and disposal of radioactive ores and their by-products. In particular, events associated with the radium industry in Canada resulted in contaminated sites, which, although dating back to the 1930s, continue to be addressed today.

An historical overview of these activities is included in the first of this sequence of papers [1], and is not repeated here.

This paper is divided into three sections:

- environmental restoration at historic low-level radioactive waste (LLRW) sites;

- environmental restoration at the Chalk River Laboratories (CRL) site of Atomic Energy of Canada Limited (AECL);

- decommissioning and waste management at uranium mining and milling sites.

The latter section deals primarily with sites for which the uranium production companies are responsible for decommissioning. It has been included to describe Canadian experience in this area since, from a technical perspective, there are many factors in common with restoration programs at inactive or abandoned sites.

2. ENVIRONMENTAL RESTORATION AT HISTORIC LOW-LEVEL RADIOACTIVE WASTE SITES

2.1 Background

The Low-Level Radioactive Waste Management Office (LLRWMO) was established by the federal government in 1982 to resolve historic waste problems (those for which the original producer can no longer reasonably be held responsible) that are a federal responsibility, to ensure that a user-pay service is established for the disposal of low-level radioactive waste (LLRW) produced on an on-going basis, and to address public information needs concerning LLRW [3]. In Canada, low-level radioactive wastes are defined as all radioactive wastes other than nuclear fuel wastes and uranium mill tailings. These latter types of wastes are not within the mandate of the LLRWMO.

Historic low-level radioactive wastes date back to 1933 in Canada, when a radium refinery began operation in Port Hope, Ontario. The problem of residual wastes and contaminated buildings and soils in Port Hope, resulting from the practices in the early years of radium and uranium production, was discovered in the mid-1970s, and a large scale cleanup program carried out. This work was concentrated on developed properties. As a result, substantial quantities of contaminated materials remained in a number of large undeveloped areas. A number of additional historic waste sites have subsequently been discovered at other locations in Canada, where buildings and/or soils were contaminated with uranium ores or concentrates spilled during transport, or with processing residues, or as a result of the use of radium containing materials. This section of the paper describes the technical approach being used at these sites by the Low-Level Radioactive Waste Management Office. Conceptual design studies and cost estimates for facilities designed specifically for disposal of large volumes of bulk LLRW and contaminated soils are also discussed. These include both work done previously by the LLRWMO, and current work being by done by independent Siting Task Forces established to locate new sites for the long-term management of specific historic waste inventories.

A major distinguishing factor between different sites is how the wastes resulting from cleanups and related activities are managed. This section of the paper is organized on this basis, with the sites described in the first paper [1], and several others, described as examples.

2.2 Historic Waste Volumes Relative To Total LLRW Volumes In Canada

Table 1 (from next page) portrays the current and estimated future inventory of LLRW in Canada over the next 35 years or so [4].

It can be seen that low-level radioactive wastes to be managed in Canada fall into two broad categories. The first is the large inventory of soils contaminated with natural long-lived radionuclides and, in most instances, heavy metal constituents including arsenic. These are primarily located in the Port Hope area.

In addition to historic waste sites in Port Hope a number of other sites have been discovered. These include sites in Scarborough, Ontario and Surrey, British Columbia which, although the volumes of contaminated soils are much lower (less than 10,000 m^3 at each area) have had a lengthy history of difficult social and political issues [5].

The most recent discoveries have been in northern Alberta and the Northwest Territories, along the transportation route from the old Port Radium mine. Water transportation by barge along a 1,400 mile route of lakes and rivers was used to move supplies in, and uranium ores and concentrates out to the railroad at Waterways, (now Fort McMurray) Alberta. A number of transfer points were needed, due to portages at rapids, and the need to use different sizes of equipment for different parts of the route.

A comprehensive survey program to identify residual contamination at these transfer locations has been completed. Cleanup work started in 1992, with the priorities being any currently inhabited sites and a series of old industrial properties in Fort McMurray where extensive redevelopment is planned. Although the total volume of soil involved at the Fort McMurray sites is significant (in excess of 100,000 m^3), new procedures for soil characterization and soil segregation have resulted in both volume reduction and early resolution of the problem.

The second category of wastes are those produced mainly by the nuclear industry, which contain primarily man-made radionuclides having a large range of half-lives. These are identified as "ongoing" wastes and are currently stored by the major producers.

It can be noted that, in terms of the estimated volume to the year 2025, about 75% is existing inventory.

2.3 Small Scale Sites

These are defined as historic waste sites where the waste volumes are small.

This is the typical situation at buildings where there is residual contamination from the past use of radium paint. These cleanups have many similarities with decommissioning and decontamination projects at nuclear fuel cycle and radioisotope facilities. Waste volumes are minimized by surface decontamination techniques and by careful segregation of contaminated

TABLE 1

ESTIMATED VOLUME OF LLRW TO THE YEAR 2025

Existing Inventory	Thousands of Cubic Meters	Millions of Cubic Feet
- Nuclear Fuel Cycle & Radioisotope Use		
- AECL Research (CRL)	300 42	10.5 (Soils) 1.5 (LLRW)
- AECL Research (WL)	17	0.6
- Ontario Hydro (BPND)	25	0.9
- Other	<3	<0.1
- Port Hope Area		
- LLRWMO Historic Sites	270	9.5
- Port Granby & Welcome Sites	610	21.5
- Other LLRWMO Historic Sites	~100	~3.5
SUBTOTAL	1370	48
Projected Arisings		
- Ongoing Wastes from Nuclear Fuel Cycle & Radioisotope Use	170-340	6-12
- Decommissioning of Nuclear Fuel Cycle & Radioisotope Facilities	100	3.5
- New Discoveries of Historic Waste Sites	Uncertain(1)	Uncertain(1)
SUBTOTAL	270-440	9.5-15.5
TOTAL	**1640-1810**	**57.5-63.5**

(1) Not expected to significantly affect totals shown.

materials. Although gamma radiation fields are low, the surface decontamination techniques and/or removal of complete pieces of contaminated materials frequently cause substantial concentrations of airborne contaminated dust. Worker protection thus involves both protective clothing and breathing protection. Temporary containments, with filtered exhaust ventilation systems, have to be established.

Wastes are transferred from the project sites to a warehouse storage facility operated for the LLRWMO through a contract with Chalk River Laboratories (CRL) of AECL. The LLRWMO has completed 15 projects of this type, generating about 1,000 barrels of waste. In addition to building decontamination wastes, small volumes of contaminated soils (nominally 20 m^3 per site or less) are also transported to the warehouse. These have filled the initial warehouse at CRL, and a second one is in operation.

2.4 Cleanups With Local Area Temporary Storage

Cleanup with temporary storage at, or near, the contaminated site is an interim remedial strategy. It is a standard approach now used by the LLRWMO for sites where remedial action should be undertaken, but where the volumes of contaminated soil are too large for transfer to the warehouse type of storage facility at CRL, [6][7].

Although final disposal is still required for these contaminated materials, these interim remedial actions have eliminated health risks and remedied environmental problems [8]. There are also general benefits to the community, in terms of eliminating negative perceptions and removing constraints on land use.

Work practices and protocols have evolved over many years. Some aspects now included in all LLRWMO projects are presented below.

Waste Delineation and Characterization. Thorough initial site investigations, including radiological surveys of the surface and subsurface conditions, are undertaken and factored into engineering designs. In 1992 the LLRWMO, together with the Borehole Geophysics Section of the Geological Survey of Canada, experimented with the installation of boreholes and use of spectral logging protocols at a municipal landfill with historic waste contamination [9]. This project has produced an improved field protocol for subsurface investigations in radon gas influenced environments.

Over the past two years, the LLRWMO has been developing large area surface gamma survey protocols. A battery powered detection system with twin sodium iodide scintillating detectors and an on-board lap-top computer mounted on a simple cart comprised the hardware. The software and field scanning protocol are undergoing statistical sensitivity analysis and software quality assurance. This approach is integral to survey work at Scarborough, Ontario and Fort McMurray, Alberta remedial sites, where the equipment has to be manually portable, but able to cover large areas in a practical time.

However, supplementary investigations during the progress of the excavation work, as well as direct supervision at the work face, is now common practice in LLRWMO projects. Rigorous attention to define and limit the materials excavated reduces volumes for future management, while still ensuring site cleanup to criteria levels.

Cleanup Operations. Standard construction equipment is used, with contamination control and health physics procedures which have been developed and proven over fifteen years. Clear delineation of the contaminated work zones, thorough briefing of workers, and auditing of compliance are featured. Where practical, site-dedicated equipment and vehicles are preferred. A predetermined staged approach to excavations and material movement on the remedial site is used. Continuous environmental monitoring is performed during the work.

In Situ Storage Facilities. LLRWMO experience with in situ consolidation and interim storage projects has shown a number of advantages [6]. Containment of the waste prevents further spread of contamination thereby limiting the environmental impact and reducing remedial costs. Barriers applied over the waste protect potential intruders from any hazards and also lessen the probability that unsuspecting parties will relocate and then further spread the problem. Covering the waste affords physical shielding to reduce gamma fields and provide a barrier against radon emanation. In fact, the act of excavating and stockpiling waste provides self-shielding layers. An accurate understanding of the characteristics and volume of the waste requiring further long-term management is obtained when the materials are delineated and excavated for interim consolidation. Progress is made toward final completion of remedial activities since most of the affected areas are cleaned and restored to the ultimate desired level. Material is also prepared for easy future removal.

In situ consolidation and storage sites have much simpler designs than permanent disposal facilities since, typically, a containment period of only a few years is expected. The LLRWMO has used a number of interim storage approaches as part of its remedial work program at sites in Ontario, British Columbia, and the Northwest Territories. These include: engineered mounds, concrete block walled bunkers, and drummed storage within a fenced area. Such storage sites hold as little as 50 and as many as 30,000 m^3 of contaminated soil, but typically apply to inventories of a few thousand cubic meters.

Monitoring. Monitoring programs have also been developed over a number of years and encompass four broad areas:

- normal background studies

The major contaminants at historic waste sites are naturally occurring radionuclides and heavy metals. Determining normal background values, and more importantly, their variability is thus important to both assessing and remediating contaminated sites.

Previous LLRWMO work in Ontario [10] is now being extended, through a complementary project to a major program by the Ontario Ministry of the Environment to establish typical ranges for a wide range of inorganic elements and organic substances. Local area studies are also now carried out for all major historic waste sites. The LLRWMO also participates in joint projects relevant to determining natural background radioactivity in Canada [11].

- worker protection

Worker exposures during remedial projects at historic waste sites have been sufficiently low, due to the relatively low average radioactivity concentrations, that there has been no need to use Atomic Radiation Worker classifications. Standard radiation protection techniques are used to implement the ALARA principle. Personnel monitoring, using standard practices, is nonetheless carried out to demonstrate that radiation doses are well below dose limits for members of the public [12].

- environmental monitoring

Comprehensive environmental monitoring programs are carried out before, during, and after major remedial projects [8]. Components are described in Table 2.

- compliance (with cleanup criteria) sampling

Standard protocols, acceptable to regulatory agencies and property owners, have been developed. They feature both field measurements of radiation, and collection and analyses of soil samples.

TABLE 2

ENVIRONMENTAL MONITORING PROGRAM COMPONENTS

COMPONENT	MONITORING APPROACH
Surface Gamma	- Scintillometer Surveys & Scans - Worker & Visitor Dosimetry (TLD)
Airborne Dust (Including Long-Lived Alpha)	- Suspended Particulate Measurement - Sample Analysis
Radon & Radon Daughters	- Passive Radon Monitors - Grab Air Sampling and Analysis - Monitoring with Continuous Reading Equipment
Soil	- Scintillometer and Field Gamma Spectroscopy Surveys - Laboratory Gamma Spectroscopy Analyses of Samples - Laboratory Chemical & Radiochemical Analyses of Samples
Surface Water	- Surface Stream Sampling and Analysis - Work Site Runoff Collection, Sampling & Analysis
Groundwater	- Borehole Water Sampling & Laboratories Analysis

2.5 Technology For Waste Segregation Into Different Inventories

One of the technical factors at some sites is the random, heterogeneous distribution of contaminated materials. For example, in Malvern, small discrete contaminated objects became distributed through an area now representing hundreds of individual private yards. A similar situation existed at Fort McMurray, where pieces of high grade uranium ore, or small amounts of concentrates, were randomly located over substantial areas of industrial land. At other sites in Fort McMurray, and elsewhere, the wastes have relatively low average radioactivity concentrations with only pockets of heavily contaminated soil, or discrete artifacts, which require an AECB license for their possession or disposal. Local residents are thus not willing to consider disposal at facilities in their locality. However, moving these wastes long distances, at considerable cost, is also not a desirable solution given that the majority of the material represents comparable, or less, hazard than common industrial or municipal wastes.

The approach now being used at Fort McMurray is to segregate the AECB licensable materials from the waste, resulting in the major volume fractions being either clean soils, or mildly contaminated soils meeting all criteria for classification as industrial waste. This approach was initially tried in 1990 at a new historic waste site in Scarborough, Ontario and appeared to be helpful in gaining public acceptance for the work. It is also now being proposed for the wastes at Surrey, British Columbia, where a mineral processing slag contaminated with thorium is the original waste, and for completion of all work in Scarborough, Ontario. Details are shown in Table 3 for the Surrey wastes.

TABLE 3

PROPOSED CLASSIFICATION FOR
SURREY, B.C. NIOBIUM SLAG AND SOILS

COMPONENT	PROPOSED DISPOSAL METHOD
ANVIL WAY INVENTORY - 6,000 tonnes, bulk slag and soils	INDUSTRIAL OR MUNICIPAL WASTE SITE - Not radioactive or chemically hazardous waste, with respect to regulatory limits
116th AVE. INVENTORY - 90 tonnes, drummed slag	SPECIAL WASTE SITE - Radioactive and Chemically hazardous, with respect to regulatory limits

Equipment developed, or being developed, is used in several ways.

Recovery of Small Discrete Artifacts Randomly Distributed In Surficial Soils. Surface gamma radiation surveys have been performed at over 500 individual private properties, and at public properties such as schools and parks, in the Malvern area of Scarborough in the past two years. Compared to the traditional manual surveys, the computer assisted gamma radiation data collection and analysis system provides increased objectivity in data collection and analyses, reduces manual data handling, and is more amenable to quality assurance procedures.

Three types of analysis are performed on the collected data. One concentrates on identifying anomalous readings that indicate the possible presence of a discrete radioactive source, while another shows larger areas of possible contamination. The third produces a summary characterization of the surveyed area and the survey itself.

Several methods of identifying larger areas of possible radioactive contamination have been developed, each relying on the interpretation of a type of site map.

This is a gamma choropleth map of a public school property. The areas occupied by buildings are shown in white indicating that they were not surveyed. Although there are no indications of contamination on this property, there are other points of interest shown on this map. The areas of lowest gamma radiation readings are all covered with asphalt. The grassed areas are easily distinguished by their generally higher readings. Elevated readings immediately surrounding the main building are from brick used in the school's construction.

Another type of map used in the analysis of some sites is a discrete source density map. This type of map highlights areas in which there were many recovered discrete radioactive sources.

Sites are generally characterized by a number of summary statistics such as the maximum, mean and variation of gamma radiation measurements.

In Situ Segregation During Excavation of Contaminated Soils. This approach has been used extensively in Fort McMurray, with the soil classification shown in Table 4.

Segregation is done using a combination of the computer assisted gamma survey system and manual inspections/surveys. Contaminated soils are excavated in relatively thin (approximately 15 cm) layers, so that each layer may be surveyed, until the full depth of the contaminated soil has been excavated.

Characterization of Segregation of Excavated Contaminated Soils. In 1990, an experimental soil sorting operation was used at a newly discovered site in Scarborough, Ontario. Mechanical and manual techniques, followed by a detector-controlled compliance conveyor, were used to handle soil contaminated with discrete particle contamination. Details have been previously described [1]. An automated system for bulk soil characterization and segregation, with a throughput rate of 20 tonnes per hour, is now in the final stages of development. It will be operated in either of two modes - separation of soil with licensable concentrations of contaminated material from mildly contaminated soils, or separation of mildly contaminated soil from clean soil (ie. soils with radium concentrations within the normal background range). The first major application of this equipment will be in the Malvern Remedial Project, where approximately 25,000 tonnes ($\approx$12,500 m^3) of contaminated soils are to be excavated and sorted in 1995.

2.6 Long Term Management of the Port Hope Area Historic Wastes

2.6.1 Background

The historic wastes in the Port Hope area represent Canada's largest challenge with respect to cleaning up and disposing of low level radioactive wastes produced over the period 1932 to 1988. The wastes comprise at least 880,000 m^3 of process residues and soils. They are currently stored at the Port Granby and Welcome waste management facilities, which are operated by Cameco (formerly Eldorado Nuclear Limited) as well as numerous locations within the Town of Port Hope, which are the responsibility of the LLRWMO.

In the early 1980s, Eldorado Nuclear Limited, a federal Crown Corporation (ie. a company owned by the government) at the time, attempted to site a disposal facility for all of the wastes and soils in the local area. At that time, two disposal options were proposed: underground mined caverns in the shale bedrock underlying the area and an engineered burial mound which took advantage of a zone of low permeability till to contain and isolate the wastes. The nuclear regulator, the Atomic Energy Control Board, was on record as favouring the underground mined caverns.

Vigorous local opposition to the proposed sites halted the project and caused the federal government to assume responsibility for finding a publicly acceptable site. In 1987, an independent task force (Siting Process Task Force) recommended a unique voluntary, cooperative site selection process to obtain community agreement to consider hosting a disposal facility and then to identify a potential site [13]. In 1988, a successor task force (Siting Task Force) was established by the Minister of Natural Resources Canada to implement the recommended siting process [14]. To date, two communities in Ontario, Port Hope and Deep River, have agreed to consider hosting a facility. A referendum on whether or not to host a facility is likely to be held in each community in 1995.

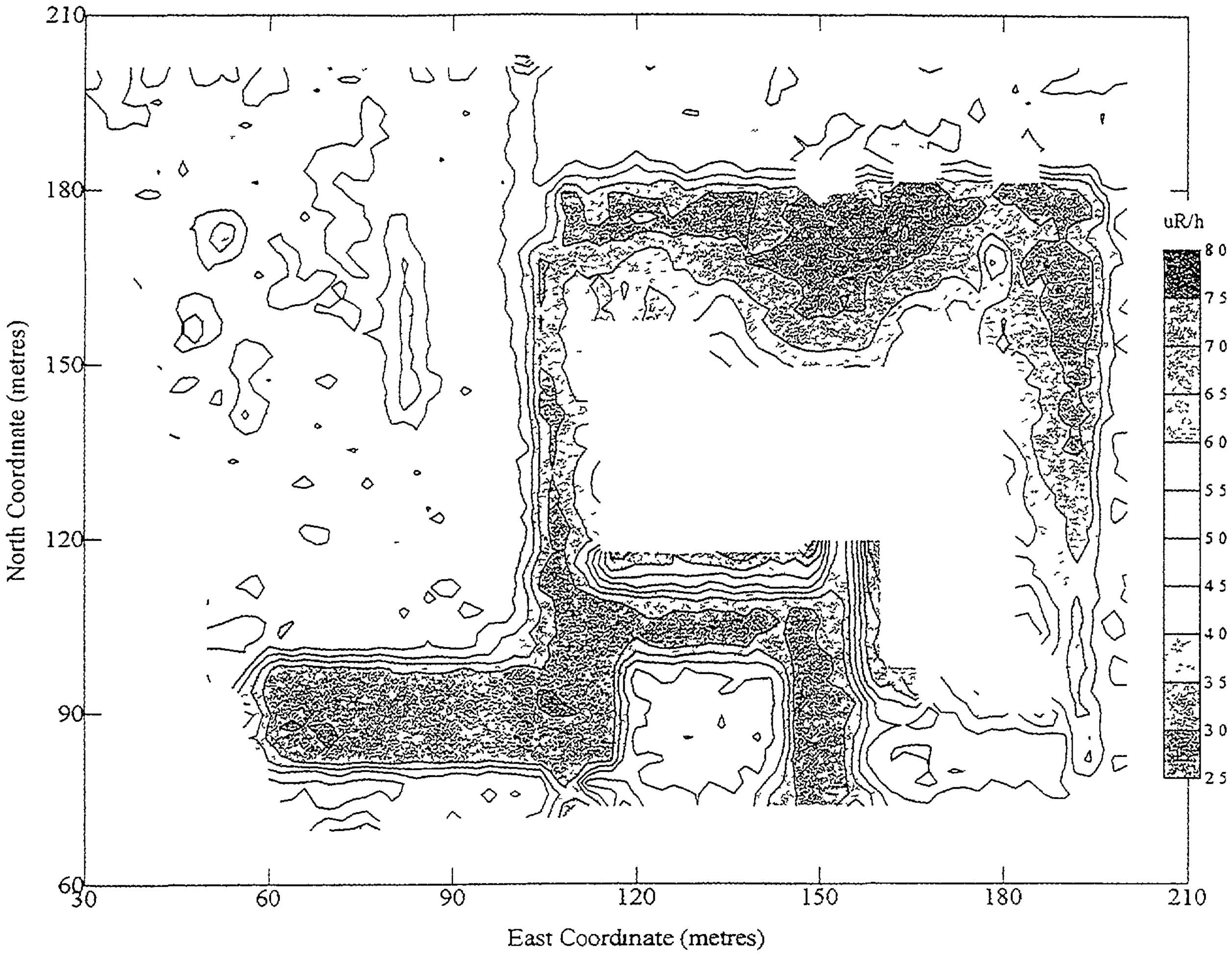

FIG. 1. An Example Gamma Choropleth Map

2.6.2 Facility Design and Cost Estimates

As part of the voluntary, cooperative siting process, preliminary conceptual design and costs were developed for a number of possible waste disposal facility designs. These conceptual studies have examined a very broad range of options including containerizing the wastes. The initial study [15] was performed for the LLRWMO. Its objective was to develop consistent comparative cost estimates for all concepts identified by the Siting Process Task Force. Conceptual designs ranged from those providing permanent disposal without institutional controls to those requiring ongoing long-term monitoring and maintenance. An original in situ waste volume of about 800,000 m^3, relatively favourable generic site conditions, a four-year disposal schedule, and a consistent costing basis were assumed for all concepts. Results are shown in Table 5, and range from \$77.m^3 (1988 Cdn\$) of waste for shallow land burial on unlined trenches, to \$350/m^3 of waste for disposal in concrete canisters.

Most recently, these conceptual designs have been refined and tailored by the Siting Task Force to meet the general site conditions in the volunteer communities. Specifically, four concepts appear promising.

- Engineered burial mounds involving placing the wastes in mounds constructed on or slightly above ground surface.

TABLE 4

CLASSIFICATION FOR FILL MATERIALS AND CONTAMINATED SOILS AT FORT McMURRAY SITES

Category A - Licensable LLRW

Material exceeding an uranium concentration of 500 ppm and therefore requiring a licence from the Atomic Energy Control Board (AECB). This is mainly uranium ore. The volume of Category A material that results from the cleanup work at Fort McMurray will be several hundreds of cubic meters or less. This material is transferred to a LLRWMO storage facility at the Chalk River Laboratories of AECL.

Category B - Mildly Contaminated Soil

Material that contains elevated amounts of uranium that are below licensable concentrations. This is mainly contaminated soil which exceeds one or more of the cleanup criteria for radium (0.1 Bq/g), arsenic (30 ppm) or uranium (30 ppm). The present volume estimate for Category B material is about 40,000 m^3 . Category B material is disposed locally as industrial waste, in a separate cell at the municipal landfill.

Category C - Restricted Use Fill

Soil which meets all of the cleanup criteria, but which may contain occasional rocks or pieces of ore that are elevated in uranium concentration. This material is used as fill for specific purposes.

- Shallow burial trenches, which involves excavation of the trenches, are located below existing ground surface in natural low permeability soil deposits with good engineering characteristics in terms of strength and compressibility.
- Open pit in bedrock with pervious surround concept which could be applied to containment in existing or new open pit excavations in bedrock and/or in small rock-bound lakes. The containment would be below the permanent groundwater table or lake level.
- Underground mined caverns where the waste containment units would typically be located at a depth of 100 m or so below the ground surface. The containment units are excavated entirely in bedrock.

Cost estimates were developed for each of the conceptual designs listed above for a reference waste volume of 880,000 m^3. In addition, a cost sensitivity analysis was carried out for the engineered burial mound, open pit with pervious surround and underground mined cavern concepts. The shallow burial trench option was not evaluated because the site conditions do not exist in the potential volunteer communities. Total costs, and costs per unit volume, are shown in Table 6.

TABLE 5

COMPARISON OF TOTAL PROJECT COSTS

Disposal Option	Cost (millions of 1988 $)	Unit Cost ($/m^3)[a]
Unlined Trenches	61.31	77
Lined Trenches	67.84	85
Engineered Storage Mounds	85.54	107
Open-Stoped Caverns	129.72	162
Shrinkage-Mined Caverns	140.38	175
Above Ground Concrete Vaults	170.52	213
Below Ground Concrete Vaults	230.99	288
Concrete Canisters in Trenches	279.95	350

(a) Based on approximately 800,000 m^3 original in situ volume.

TABLE 6

EFFECT OF PORT HOPE AREA WASTE VOLUME ON UNIT DISPOSAL COST

Technology	Unit Disposal Cost ($/m^3)		
	Decreased Volume (688,000 m^3)	Reference Volume (880,000 m^3)	Increased Volume (1,082,000 m^3)
Engineered Mound	96	82	72
Pervious Surround	87	75	67
Mined Cavern	187	168	155

2.6.3 Alternative Methods For Marginally Contaminated Soils

A large portion of the material at the Port Granby and Welcome waste management facilities consist of native in situ subsoils that have become contaminated by groundwater transport of uranium, arsenic or radium. Recent estimates by the Siting Task Force show that this category termed "marginally contaminated soil" may comprise up to 400,000 m^3, depending upon the cleanup criteria selected. This has led to discussions on whether there are more economical alternatives for this marginally contaminated soil than excavation and disposal in a new LLRW facility.

Table 7 shows the average concentration of arsenic, uranium and radium in the marginally contaminated soils. The recommended cleanup criteria and natural background levels in the immediate area are provided for comparison.

A preliminary costing study has been done which compares the marginal cost of disposal in a LLRW facility with alternative management approaches. Nobody has questioned that the wastes, themselves, and the most contaminated soils must be placed in a new facility.

Specifically, the costing study examined three alternative strategies for managing the estimated 400,000 m^3 of marginally contaminated soils at the current Port Granby and Welcome waste management facilities, namely:

- *manage in place* by using engineered barriers or in situ treatment to contain and isolate the contaminated soils to the maximum extent possible. Costs have been developed for two possible approaches:

TABLE 7

CONTAMINANT CONCENTRATIONS IN MARGINALLY CONTAMINATED SOILS AT PORT GRANBY AND WELCOME

Contaminant	Average Concentration in Marginally Contaminated Soils		Recommended Cleanup Criterion	Background Soil Concentration in Area
	Port Granby	Welcome		
Arsenic (ppm)	30	40	20	3.45
Uranium (ppm)	46	12	100	1.99
Radium-226 (Bq/g)	0.22	0.07	0.133	0.033

  - an engineered cover using a combination of synthetic and natural soil materials
  - in place chemical stabilization using large-scale, soil mixing techniques.

- *treat and manage on site* by excavating the contaminated soils by either immobilizing or removing the contaminants through solidification or removing them by washing the soils. The solidified or washed soils would be returned to their original location.

- *excavate and dispose commercially* by relocating the material to a commercial industrial waste disposal facility licensed and approved for similar, though non-radioactive, industrially contaminated soils.

The results of the study are shown in Table 8 where the marginal costs of disposal in one of the three facilities under consideration are compared with the costs of the alternative management strategies.

The costing study shows that the marginal cost of disposal ($42.9 to $71.1 million or $66 to $109/t) is considerably less expensive than the technological solutions such as fixation/stabilization ($210/t) or soil washing ($128/t). Indeed, the only management technologies that appear economically competitive are those that treat the marginally contaminated soil in place, ie. covering the soils with an engineered cap and cover or a soil mixing technique ($28 to $42/t).

TABLE 8

INCREMENTAL COSTS OF ALTERNATIVE MANAGEMENT STRATEGIES FOR APPROXIMATELY 400,000 M^3 OF MARGINALLY CONTAMINATED SOIL

Soil Management Strategy	Total Cost (Cdn $ M)	Cost Per Tonne (Cdn $)
1. Disposal in New LLRW Facility		
- mined caverns	71.1	109
- open pit with pervious surround	42.9	66
- engineered burial mound	42.9	66
2. Manage in Place		
- in situ soil mixing	27.4	42
- cap and cover	18.2	28
3. Treat and Manage on Site		
- fixation/stabilization	136.3	210
- soil washing	83.5	128
4. Commercial Disposal	53.8	83

3. ENVIRONMENTAL RESTORATION AT CHALK RIVER LABORATORIES

3.1 Background

Atomic Energy of Canada Limited (AECL) is the federal Crown Corporation responsible for research and development for the uses of nuclear energy in Canada. AECL develops and markets CANDU reactors, supplies CANDU and light water reactors (LWR) support services, develops and applies radioactive waste management and site remediation technology, and provides associated products such as research reactors and industrial accelerators.

AECL has two major research sites - Chalk River Laboratories in Ontario where operations started in the mid-1940s, and Whiteshell Laboratories in Manitoba where operations started in the mid-1960s. The current focus of AECL's LLRW program is to make the transition from interim storage to permanent disposal at the CRL site [16]. A complementary program is directed as assessing the need for remedial action at old CRL storage sites dating back to the 1940s and 1950s, and implementing the required remedial activities.

Waste management practices evolved over the years of operation of nuclear facilities at CRL. Initially there were no established practices and various approaches were taken with both liquid and solid wastes. In some cases these practices, which were acceptable at the time, have led to radionuclide discharges to soil and the formation of groundwater contaminant plumes. Some specific trials were also conducted to determine the rate and extent of subsurface and ground water contamination that might result from inadvertent spills and to gain specific knowledge from wastes placed purposely in the ground. There is no danger to the general public as the specific waste sites are well within the boundaries of the laboratory, nor is there any risk to employees since the sites are well delineated with signed fences.

A strong and active hydrogeological program is in place at CRL to observe and to model radionuclide releases and to develop a thorough understanding of contaminant transport in saturated and unsaturated media. Recently, a remediation program was initiated to set priorities on the cleanup and restoration of the contaminated sites on the CRL property, to meet the environmental standards of stewardship outlined by the Federal Government. Part of the remediation program is to develop a suite of technologies that can be applied to the removal of contaminants from soils and ground water in an efficient and cost-effective manner.

The radioisotope and fission product wastes, and associated contaminated soils, at these old sites are of relatively short half-life compared to the natural radionuclides in the Port Hope area, and other, historic wastes. Consequently, thee will be some differences in the approaches to remedial action.

3.2 Ground water Remediation by Selective Contaminant Removal

Remediation technology for the removal of low concentrations of radionuclides, heavy metals and organics from ground water began several years ago by applying novel technology developed for radionuclide removal from waste waters. The process involved adding water soluble polymers to create macromolecules by attaching dissolved ions to the polymers. The macromolecules were then removed from solution by using cross-flow ultrafiltration. The process provided high removal efficiencies, it was highly selective in the removal of hazardous substances, and generated a minimal volume of secondary waste. Unfortunately, the technology is generally applicable to low ionic strength waters and the presence of large concentrations of iron caused the membranes to foul rapidly.

Building on the experience gained in laboratory and field investigations, the technology to selectively remove contaminants was improved by switching to a more porous membrane and by altering the chemical treatment. By inducing precipitation and adding fine sorption materials, similar removal efficiencies were achieved without the membranes being fouled by the presence of iron while attaining low volumes of secondary waste requiring immobilization and eventual disposal. The effluent quality achievable with the process can be made to comply with drinking water standards.

The technology has been successfully demonstrated on a contaminated site at CRL and is now in routine operation. Over the past two years, more than 2 million litres of water have been treated to reduce Sr-90 concentrations from about 2,500 Bq/L to < 1 Bq/L, with typical effluent values at 3 Bq/L, well below the Canadian drinking water standard set at 10 Bq/L.

The process in place involves the sequential addition of chemicals and adsorption/ion exchange materials to remove contaminants. The combination of chemical conditioning, cross-flow microfiltration and dewatering by filter pressing is effective for treating various ground waters containing mixed wastes having diverse physical and chemical properties. The filtrate water is discharged once it meets the specified water quality. To achieve high quality water, up to three steps of chemical treatment and microfiltration may be employed to remove contaminants. The secondary waste volume is typically 1/500 the volume of the feed.

The chemical conditioning and microfiltration process has significant technical advantages and economic benefits for site remediation. The combined action of precipitation, co-precipitation, adsorption and ion-exchange coupled with cross-flow microfiltration effectively removes dissolved contaminants into a concentrated suspension. The direct contact of contaminants with iron and other metal precipitates provides high contaminant removal efficiencies and fast kinetics. Low cost ion exchange/adsorbent materials are utilized in a continuous operation. Less space is required than conventional systems and the use of modular construction permits flexibility and adaptability to different flow requirements as well as providing portability and ease of movement to a contaminated site. One of the features of the process is that it is sufficiently generic to permit treatment of waste solutions containing a variety of radioactive and hazardous substances.

3.3 Recent Advances

Further progress in site remediation has taken place with acidic soil leachates, generally created by oxidation and dissolution of sulfide-bearing wastes which in turn dissolve heavy metal contaminants. The application of ultrasonics after the addition of pH adjustment chemicals, oxidants and precipitants leads to the removal of contaminants more rapidly. With subsequent separation of solids by cross-flow microfiltration and filter pressing, the overall time for processing is generally reduced by an order of magnitude. Ultrasonic mixing in place of mechanical agitation in large tanks increases the conversion of contaminants to precipitates and affects the rate by which oxidation and ion exchange takes place. Without large tanks required for sufficient time to allow processes to take place, the use of ultrasonics permits the system to be more compact, more portable, more energy efficient and requires less capital for construction. The technology generates minimal fugitive emissions and also produces a treated effluent that meets applicable discharge limits. The technology has also been able to treat waste containing small quantities of dissolved or suspended organics.

Soil washing has been evaluated to speed up the removal of contaminants from ground water, by applying chemicals in solution directly to the soil. By stripping the contaminants from the soil, the leachate can then be treated to extract the contaminants. Application of in situ soil stripping could reduce the time required for treating contaminated ground water by eliminating the source.

Studies are underway at CRL to develop injection and recovery methods that will allow soil treatment without having to disturb the soil. A key factor is having a good understanding of the hydrogeological properties of the contaminated site to properly situate the injection and recovery wells. One of the successful materials for injection and removal of radioactivity from CRL soils is dilute ferric chloride. Other dilute solvents can also be employed and are dependent on the contaminant to be extracted. Soil leaching tests established that the passage of five to six pore volumes of leachant solution removed close to 100% of all leachable Sr-90.

This leachate was then passed through the chemical treatment/microfiltration system to extract the leached Sr-90. Further tests are planned at other CRL contaminated sites to improve the removal of contaminants including uranium, cobalt, cesium, lead and other radionuclides and heavy metals.

There appears to be potential for application of these technologies to LLRWMO projects. An initial laboratory scale test for removal of arsenic and uranium from groundwater from historic sites in Port Hope has produced good results.

4. DECOMMISSIONING AND WASTE MANAGEMENT AT URANIUM MINE AND MILLING SITES

4.1 Background

There are currently over 225 million tonnes of uranium mine tailings and mine waste rock on the surface in Canada. These wastes have been the subject of extensive research under two programs:

- National Uranium Tailings Program (NUTP) 1983 - 1988; and
- Mine Environmental Neutral Drainage (MEND) 1989 - 1997.

The NUTP was a federal government funded program of contract research, costing $8.5 M, that focussed on developing predictive techniques for disposal technology. MEND is a cooperative industry-governments program that focusses on one very important issue for uranium mine wastes in Canada, acid mine drainage.

Also, the Canadian industry has been developing site-specific technology that minimizes the short and long-term impact of uranium mine wastes. The most significant development has been the progress from development to implementation of the pervious (or porous) surround technique that permits the disposal of wastes below surface and below the water table.

4.2 Research and Technology Development

4.2.1 The National Uranium Tailings Program (NUTP)

The NUTP focussed on developing predictive geochemical models that would predict long-term water quality from various decommissioned tailings management areas. The major model developed was the UTAP (Uranium Tailings Application Program) model which has been continuously revised by its users since first developed in 1986. A major subset of the UTAP model development was a predictive model named RATAP (Reactive Acid Tailings Application Program) which predicts the progress and impact of acid-generating sulphides in tailings.

The NUTP research had confirmed that a major environmental threat from uranium tailings in Canada was acid generation from residual sulphides, particularly in the Elliot Lake district. This acid-generation is particularly severe because the very strong acid leaching solutions needed to attack the brannerite-uraninite minerals had destroyed all residual natural alkalinity.

Among the decommissioning technologies investigated under the NUTP was the use of simple soil and enhanced vegetative covers for tailings. For stacked tailings sites, the simple covers were shown to have minimal impact on the geochemical activity of the tailings, particularly acid generation.

4.2.2 The Mine Environment Neutral Drainage Program

Acidic drainage is not only a problem for the uranium industry but it is the largest single environmental problem facing the world's metal mining industry today. Technologies to prevent, or substantially reduce, acidic drainage from occurring in waste rock piles and tailings sites and mine walls need to be improved and demonstrated. These new technologies will substantially reduce the operating and closure costs at existing mine sites and in the rehabilitation of abandoned mine sites.

A decade ago, the Canadian mining industry and government laboratories had conducted research into rehabilitation of mine sites, with a special focus on uranium tailings sites by establishing sustainable vegetative growth on tailings and waste rock. It was believed that this technology would alleviate acid drainage problems from these sites, thus allowing mining companies to abandon these sites without future liability. However, after several years, the quality of water drainage from vegetated waste sites had not improved, and property owners were faced with the prospect of continuing to operate and maintain lime and barium chloride treatment plants indefinitely. It was clear that more knowledge and expertise needed to be developed and new remedial technology needed to be developed and demonstrated.

As a consequence, in 1988 the Canadian mining industry, 5 provincial governments and the government of Canada cooperated to form a tripartite consortium organized under the **Mine Environment Neutral Drainage (MEND) Program**. Since then, the two levels of government and the Canadian mining industry have together committed $18 million on MEND projects to find ways to reduce the liabilities caused by the natural acidification of mineral wastes.

MEND is focussing on the development of new technologies that will reduce the estimated $5 billion liability that would be incurred if the known, reliable, but costly remedial technologies were used. For example, it is currently estimated that multi-zone earth covers would cost between $15 and $30/m^2, and even if installed, could reduce acid production by only 90%. This reduction, although significant, would not permit the treatment plants to be shut down for a long time.

Indefinite collection and treatment of acidic waters may be an option at many sites, but the significant annual cost plus the storage problem of accumulated sludges is also costly and problematic. Also, Canadian regulators do not favour long-term treatment as a decommissioning option.

Some examples of the technology being developed in over 100 MEND projects that are applicable to uranium mine wastes are:

- Refined chemical prediction procedures have been developed to determine if waste rock or tailings will acidify.

- Predictive geochemical models for tailings and waste rock are being modified and new ones developed. Models that will predict the performance of dry soil covers on stacked tailings and rock piles are being field-evaluated.

- Underwater disposal is being confirmed as the best prevention technology for unoxidized sulphide-containing wastes. Field and laboratory studies have confirmed that reactive sulphide tailings and waste rock do not react underwater in the natural environment. Natural or engineered water cover systems with an organic surface or sedimentation layer prevent the minor amounts of oxygen dissolved in water from oxidizing sulphides. Underwater disposal in engineered impoundments to prevent acid production is now a common feature in proposals for new mines in Canada.

- Water covers to control acid generation in already oxidized tailings and rock is also being investigated and implemented. Results to date on Elliot Lake uranium tailings show that oxidation is effectively stopped and techniques are being developed to minimize water contamination so that treatment plants can be shut down in a few years. Two methods have been used to engineer the water cover:

  - "move the wedge by dredge" - Denison Mines
  - "rice paddy" concept - Rio Algom

  The Denison Long Lake 280 hectare tailings area has been levelled by dredging the beach tailings area into lower areas of the impoundment, and the whole area has been flooded. In the few months that the tailings have been flooded, surface water quality has substantially improved.

  The Quirke 190 hectare tailings area is being flooded by establishing a series of internal dykes. The first cell, Cell No. 14, has been extensively monitored, and the water quality is approaching natural background quality.

- Unfortunately, many of the uranium and base metal mine waste sites are not physically suitable for water covers. MEND has extensively investigated multi-zone earth covers for tailings and waste rock. These covers are effective, but are very costly to install in many areas of Canada. Innovative "dry" cover research is indicating that several materials, including waste materials from other industries provide excellent potential at lower cost for generating moisture retaining, oxygen-consuming surface barriers.

- Several other disposal technologies that will reduce acid generation are being investigated:

  - permafrost covers about 40% of Canada, and cold conditions inhibit oxidation;

  - an elevated water table, if it can be maintained in the long-term, can be a significant part of a tailings decommissioning scenario. Thickened tailings discharge is being investigated by an active uranium producer in Saskatchewan.

  - Depyritized and fine tailings are also showing excellent potential as covers. Depyritization has been extensively investigated by the Elliot Lake uranium companies and has been rejected for economic and environmental reasons;

- Pervious surround is a very promising method for "walk-away" for tailings deposits located below the water table. This technology is being used at a Saskatchewan uranium mine and new mines are proposing this method for below grade disposal of uranium mill tailings.

- Passive treatment systems such as engineered wetlands have been shown to have some application to metal mine acidic drainage in Canada. As a result of a tailings spill many years ago, and because of the action of beavers, a natural wetland was established over acid-generating uranium mine tailings. Extensive field investigations have shown that natural sulphate reduction in this wetland has reversed the acid production process and a truly passive treatment system has been established.

- MEND has sponsored the adaptation of geophysical methods including eletromagnetic (EM) methods for the tracking and monitoring of subsurface acidic seepage, and the method is being successfully applied at mine sites across Canada. A "sediment probe" that will detect acidic seeps that emerge in a stream or river bed has been successfully demonstrated. These methods have been shown to be well suited to uranium mines wastes sites and are now routinely used at Canadian locations.

4.3 Site Specific Decommissioning Programs - Completed Sites

The decommissioning of four sites has been completed; the Bancroft region mines (3), the Agnew Lake site in the province of Ontario and the Beaverlodge mine in Saskatchewan, and the Port Radium mine in the Northwest Territories. All locations had site specific characteristics. In general all tailings were "decommissioned" in situ, and contaminated building rubble was incorporated in the wastes. The following Table 9 summarizes decommissioning activities:

TABLE 9

SUMMARY OF DECOMMISSIONING ACTIVITIES

Site	Operated, Years	Tailings Tonnes 10^6	Decommissioning Action			
			Cost ($\$10^6$)	Tailings	Waste Rock	Buildings
Bancroft	1957-1963 1977-1982	2.4 2.0	0.4	In Situ** Soil Cover	In Tailings	Tailings
Agnew Lake	1977-1983	0.4*	3	In Situ Soil Cover	Mix into waste precipitates	Mine Shaft, Tailings
Port Radium	1933-1960	0.9	0.5	Soil, Rock Cover	Tailings	Demolish, Rock Cover
Beaverlodge	1947-1982	6.0	6.0	Underwater	In Situ, Contour	In Situ, Waste Rock Cover

* Chemical precipiates from in situ leaching operation
** Some tailings moved to reduce gamma exposure

All sites except Bancroft sites are remote from extensive habitation and this resulted in intrusion not being a major factor in the decommissioning programs.

4.4 Development of Plans for Decommissioning of Elliot Lake Mine Tailings

4.4.1 Background

In 1953, uranium deposits were discovered along the northern shore of Lake Huron in Ontario. Over the next four years (1955 to 1958) twelve mines were brought into production and, by 1959, a town of more than 25,000 people had been located on the eastern shore of Elliot Lake in northern Ontario.

Tailings were slurried and discharged, usually to low-lying areas of small lakes. Dams were used at some locations to assist in the containment of discharged materials. Tailings effluents were neutralized to a pH of 6 to 7, the range of values found in the lakes and rivers of the areas. At the time, these waste management techniques satisfied the government agencies responsible for the licensing and monitoring of such practices.

In addition to uranium (and daughters), the Elliot Lake ores contain pyrite (about 6% FeS_2).

Underground, bacterial oxidation of pyrite turns mine water acidic, subsequently permitting the dissolution of various heavy metals and radionuclides also present in the ore. One of the earliest environmental concerns identified in the Elliot Lake area stemmed from the initial practice of discharging untreated acidic mine water to nearby lakes and streams. The most notable example of this situation, was Quirke Lake which received untreated mine water from several mines in the late 1950s. Within a few years the pH of the lake, the largest in the region's drainage basin, had decreased dramatically to pH 2-3. Approximately two years after the start of mining, the practice of pumping mine water back to the mills for treatment was started. Since that time, mine water has been recycled into the milling circuits. This reduced both the amount of fresh water used in the mill and the amount of effluents that eventually discharged to the environment.

The hectic years of the 1950s were followed by a downturn in the 1960s. The early 1970s saw an increased demand for uranium which resulted in expansion to Elliot Lake uranium mining activities. More recently, the diminishing ore grades and consequent higher cost of Elliot Lake uranium has lead to progressive shutdown of mines. Today only Rio Algom's Stanleigh mine remains in operation. Two companies, Rio Algom and Denison Mines are in the process of decommissioning properties in Elliot Lake. The plans described in the following sections have been proposed to the public review currently in process through the federal Environmental Assessment and Review Process (EARP). The projects were referred to this environmental review by the Atomic Energy Control Board (AECB), the nuclear regulatory agency in Canada.

4.4.2 Development of Decommissioning Plans

Numerous decommissioning studies have been carried out for the Elliot Lake properties. Decommissioning plans for mines typically involved removing stationary and mobile equipment, piping, electric and all hazardous materials. This is followed by staged sealing of underground workings and the shutting off of ventilation systems. Underground raises and shafts are capped as they are no longer needed. Salvageable equipment is recovered from and decontaminated prior to sale. Surface facilities are demolished and rubble placed in the tailings basins or in vertical mine openings as appropriate. The mines were then allowed to flood.

Various tailings management facilities decommissioning options have been studied. These include:

Wet Cover Options. Wet cover involves the placement of water over the entire exposed tailings. The objectives are to:

- prevent acid generation and the associated leaching of metals and radionuclides;
- provide a barrier to intrusion onto the tailings;
- essentially eliminate radon and dust emissions.

Dry Cover Options. Dry cover involves the placement of fill over the exposed tailings. The objectives are to:

- raise the water table above the acid generating tailings to prevent the acidification and associated leaching of metals and radionuclides;
- provide a physical barrier to intrusion onto the tailings; and
- reduce radon and dust emissions.

For the soil cover option, soil cover could be an inert material including sand, gravel, till, or crushed rock. The cover would be vegetated to minimize erosion and stabilize the surface.

Dry cover can also be achieved by the placement of pyrite-reduced tailings in lieu of soil.

For the pyrite-reduced cover, depyritized tailings would be placed to a depth of about three meters over the exposed tailings with the objective of elevating the water table above the surface of the reactive tailings and thereby halting acidification.

Another dry cover concept is the application of a radionuclide-reduced cover. Several options for the removal of radionuclides from tailings were considered. However, none of these processes are fully effective.

4.4.3 Lake Disposal

Lake disposal requires the relocation of tailings at depth in a natural lake. The objectives are the same as those for the wet cover scenario.

The concept includes dredging the tailings at existing tailings areas and thickening the tailings slurry, followed by pumping of the thickened tailings to Quirke Lake for disposal. Quirke Lake covers an area of 20 km^2, with a mean depth of 30 m.

4.4.4 Underground Disposal

The placement of tailings back into the mine is called backfilling. The objectives are the same as those for wet cover. There are three methods for placing tailings into a mine: conventional engineered mine backfill techniques; past backfill; and slurry backfill.

For placing as an engineered fill during mining operations, the tailings would have to be segregated and thickened. The fine tailings slimes would be pumped back to the tailings pond and the coarse stream would be mixed with cement and "placed" underground. Capacity for underground backfill, is limited, typically, the maximum quantity of backfill that could be placed would be about 28% of the total tailings volume.

The long-term objectives for decommissioning include:

- minimizing acid generation and radionuclide migration and hence input on watershed and users;
- minimizing radiation exposure from casual access, and
- ensuring stability of structures.

Taking these objectives into account, the relative merit and disadvantages of the various alternatives decommissioning concept can be compared in terms of environmental, social and economic impacts. Such a comparison is illustrated for two Elliot Lake tailings management facilities in Table 10.

4.4.5 Comparison of Technical Issues of Canadian Uranium Mine Wastes With Other Countries

Most of the uranium mine wastes in Canada are relatively remote from large population centres. In general, this is similar to other current and former uranium producing areas in the world, except central and eastern Europe. A significant difference between Canadian sites and international locations is the cold Canadian winter climate and the abundance of water - net positive precipitation over evaporation in all regions of the country. Therefore, the prevention of contamination of surface and groundwaters by mine wastes is a primary waste management focus. In the case of pyritic tailings, water is being used as an effective barrier for oxidation and intrusion. Water covers are rarely an option outside Canada.

In addition to the normal concerns about radioactivity from the uranium series, the presence of significant concentrations of Th^{232} in the pyritic tailings in the Elliot Lake district emphasises the need to prevent acidification.

Newer uranium mines in the Athabasca basin region of Saskatchewan are mining higher grade deposits than previously mined elsewhere in the world. Also, significant concentrations of heavy metals and arsenic are typically present. The waste volumes are small, but the metallurgical operations produce considerable amounts of slightly soluble sulphates and hydroxides that must be isolated from the environment. Engineered subsurface impoundments, so called porous or pervious surround is the preferred disposal technology.

TABLE 10
COMPARISON OF VARIOUS DECOMMISSIONING ALTERNATIVES

Option Type	Wet Cover	Dry Cover	Lake Disposal	Underground Disposal[5]
Water Treatment				
- short term	Yes	Yes	Yes	Yes
- long term	No	No	Yes	No
Seepage Losses	Base Case	Same as Base Case	Eliminated	Decreased
Air Emissions				
- short term	Eliminated	Increased	Increased	Increased
- long term	Eliminated	Eliminated	Eliminated	Eliminated
Stability of Tailings	Modest Maintenance	Major Concern	No Issue	Modest Maintenance
Intrusion Considerations	Minor Concern	Major Concern	No Issue	Minor Concern
Disturbance of Areas Outside WMAs	Minimal	Major	Major	Minimal
Resource Recovery				
- mine	Available	Available	Available	Major Constraint
- tailings	Available	Available	Major Constraint	Minor Constraint
Employment Opportunities	Minimal	Short Term	Short Term	Short Term
Radiation Exposure				
- public	Base Case	Decreased	Increased	Same
- worker	Base Case	Increased	Increased	Increased
Cost ($M)[1]				
- A	$4(+$26)[2]	$38-$58	}	}
- B	($15)[2]	$20-$28	} $238[3]	} $67-$223[4]

Notes:

(1) Costs do not include engineering, long-term monitoring or expenditures for dam construction to date.
(2) Numbers in parenthesis represent total expenditures to date to implement flooded tailings concept.
(3) For removal of both A and B tailings to Quirke Lake.
(4) The low estimate ($67 M) applies to use of slurry backfill while the high estimate is for use of engineered mine backfill.
(5) Not a "stand alone" option. Only disposes of 25% to 35% of tailings.

TABLE 11

COMPARISON OF RECEPTOR DOSES
TO BACKGROUND LEVELS

Source of Exposure	Radiation Dose (μSv/y)
Typical Background Levels	
- Typical natural radiation exposures from all sources	2,000 to 3,000
- Background radiation from gamma level of 8 μR/h (low range Elliot Lake)	424
- Background radiation exposure from gamma level of 45 μR/h (high range Elliot Lake)	2,385
Limits	
- Proposed regulatory limit for exposure from nuclear facilities	1,000
Exposures from Decommissioned Facilities	
- Exposure to Elliot Lake resident from Quirke & Panel mines	1.9
- Casual assess to mine sites 200 h/y	13.2
- Living on Serpent river at Quirke Lake(1) and casually accessing the site (peak dose)	34

Notes:

(1) Assumes the receptor lives at the inlet to Quirke Lake, drinks water from the Serpent River, eats fish from Quirke Lake, spends 200 hours at the mine site, and resides 365 days per year in the area.

4.4.6 Conclusions and Summary

Several uranium mine sites have been "closed out" in Canada, however, long-term monitoring of site conditions and water quality continues at some sites. The sites have been closed out by the property owner without financial assistance from the government. All decommissioning technology is subject to rigorous regulatory review, and after all decommissioning and monitoring has been completed, the property will eventually be turned back to the government.

In general, waste management areas are decommissioned in situ, and tailings areas are used for the disposal of contaminated rubble from metallurgical and mine buildings.

Options are currently being evaluated for large volume tailings areas in the Elliot Lake region of Ontario. Because of favourable geography (hydraulically tight basins of rock), and because of long-term concerns about acid generation, water covers are being favoured by the property owners. Risk assessments indicate that risks are low if long-term monitoring and maintenance is considered. For these sites, no complete "walk away" technology has been determined to be economically achievable.

New, high grade mines in Saskatchewan are designing and installing waste management systems that will, upon mine closure, require minimum remediation and long-term monitoring. The porous surround is the technology that fits best the local geography and environmental objectives.

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RESTORATION OF RADIOACTIVELY CONTAMINATED SITES IN THE REPUBLIC OF CROATIA

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Abstract

This report brings results of performed investigations and analyses, but also shows information of relevant past researches referring to three highly prioritised sites: INA-VINIL Plant, PLOMIN Power Plant and INA-PETROKEMIJA Plant. This information serves as a suitable start-point for further investigations, which are - according to the programme schedule - foreseen to be completed by the end 1997. The report also gives some recommendations for the personnel of INA-PETROKEMIJA Plant, being in this way useful for everyday practice in the only fertiliser factory in Croatia.

1. SITES SUSPECTED TO BE RADIOACTIVELY CONTAMINATED

As it is previously mentioned, there are four main groups of sites in Croatia which are suspected to be radioactively contaminated: (1) sites containing coal slag/ash piles; (2) sites containing phosphates and waste gypsum (i.e. phospho-gypsum) from fertilizers industry; (3) geothermal springs and oil/gas drilling sites; (4) sites containing natural radioactive materials used in human activities.

Consequently, following sites were initially examined as possible radioactively contaminated spots in our country:

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

It should be also mentioned that additional quantities of coal-slag and ash have been used at numerous fire-rooms 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 phosphate ore

C. Geothermal springs and gas/oil drilling sites
   1. Istarske toplice spa
   2. Topusko spa
   3. Velika Ciglena gas drilling site

These *geothermal springs* (Velika Ciglena, in particular) show that considerable attention should be payed also to *gas- and oil drilling sites* where contamination of pipelines and separators by occasionally high-radioactive scale is possible (the problem is evaluated in more details below). Therefore, it seems reasonable to accomplish preliminary measurements not only at the mentioned geothermal springs, but also at gas- and oil exploitation fields. As it has not been possible to carry out any kind of measurements in the frames of this programme so far, below are given results of past investigations at referring sites.

It is known that separation of uranium and thorium occurs during crystallisation 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 water 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 Table 3, whilst composition of radionuclides contained in geothermal waters (often high-mineralised) at some of investigation drilling sites, is given in the Table 4. 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 drilling sites. [Editor's note: Tables 1 and 2 are not referred to in the text]

Table 3. Radionuclide content in natural geothermal water (Bq/m^3)

Site (borehole)	^{40}K	^{228}Ra	^{226}Ra	^{238}U	^{235}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
Source: Archives of performed measurements, Institute for Medical Research and Occupational Health, Zagreb

Table 4. Radionuclide content in geothermal water from boreholes (Bq/m^3)

Site (borehole)	^{40}K	^{228}Ra	^{226}Ra	^{238}U	^{235}U
Zagreb ("Mladost")	1,060	305	400	1,340	64
Podsused (INA)	131	9	54	40	2
Velika Ciglena 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-1 B (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)*
Source: Archives of performed measurements, Institute "Ruđer Bošković", Zagreb

However, another type of radioactive contamination, possible to occur at gas- and oil drilling sites should be also discussed. During gas- and oil exploration, the parameters (pH, pressure, temperature, etc.) of the "in situ" stable stratum-water could be disturbed, generating "scale", reported to be occasionally high-radioactive. The scale, including in many cases 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 [11] on the surface of pipelines in cases of the high-radioactive scale (i.e. 10^7 Gy/h higher than natural gamma-dose rates). There are no data on this phenomenon, which would be measured in Croatia. Nevertheless, in order to find out the situation at gas- and oil drilling sites, and - if needed - to improve protection measures of operating workers, it is recommended to organise and start the scale sampling at the gas- and oil drilling sites in Croatia.

Recommendations for further actions:

(1) Sampling and measuring of drilling scale at gas- and oil exploration fields should be started, and a long-term co-operation with people from the INA-NAFTAPLIN oil company should be set up aiming the performance of occasional or periodical sampling of material at oil- and gas drilling sites, suspected to be considerably radioactive.

(2) As the most serious problem in radioactive scale figures the content of ^{226}Ra (in less extent also ^{228}Ra) and its decaying products. Possible clean-up actions will depend on results of further radium measurements.

(3) As the highest concentrations of uranium are expected in heavy oil distillation fractions (i.e. asphalt and bitumen), they should be sampled in order to determine the uranium activity. Further actions will depend on results of analyses.

D. Sites containing natural radioactive materials

There have been measured increased natural radiation at some substances used in industry of building materials (cement, bricks), as well as at substances composing ceramics (e.g. zircon-silicate, silicon sand etc.). Special attention in cement industry should be payed to cement admixtures (e.g. coal slag/ash etc.), which were reported to be radioactively contaminated.

Hence, the performance of periodically repeating measurements in ceramics industries (e.g. INKER-Zaprešić), brickworks (e.g. "Zagorka"-Krapina, Virovitica, Sladojevci etc.), but - above all - at cement plants (e.g. those at Našice, Solin, Kaštel Sućurac, Pula, Koromačno, Umag and Omiš), is suggested (Tabs. 5 and 6).

Table 5. Radioactivity of some natural materials used in industry (Bq/kg)

Site / material	^{40}K	^{228}Ra	^{226}Ra	^{238}U	^{235}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
Source: Archives of performed measurements, Institute "Ruđer Bošković", Zagreb

Table 6. Radioactivity of some waste materials used in industry (in Bq/kg)

Site / material	^{40}K	^{228}Ra	^{226}Ra	^{238}U	^{235}U
Dross (imported from Austria)	437	50	85	145	7
White slag (JUCEMA)	14	20	62	87	4
Dross (imported 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
Source: Archives of performed measurements, Institute "Ruđer Bošković", Zagreb

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

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 (in terms of check whether their radioactivity does not surpass the limits defined by the law).*

(3) *The present legislation in the field seems to be too mild. Some modifications resulting in more stringent law are expected to be more convenient.*

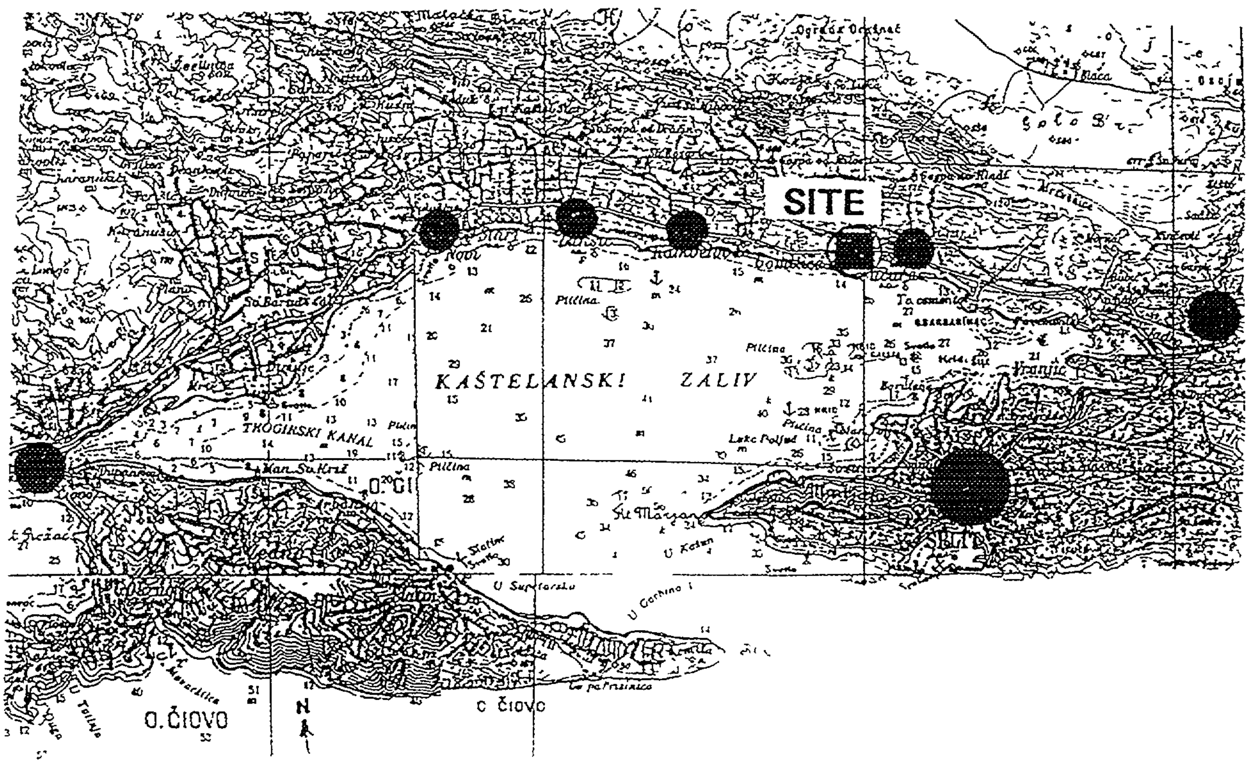
2. HIGHLY PRIORITISED SITES

It should be initially mentioned that facilities generating contaminated waste dumps at all highly prioritised sites are still in operation, presents considerable limitation for planning of clean-up actions at these sites. Namely, it is not probable that shut-down of further accumulation on the piles or simply their removal can be realised until some additional way of waste release will be operating.

2.1. INA-VINIL Plant in Kaštel Sućurac

<u>Site Description</u>
Site: 2 coal slag/ash piles (10,000 m^3 + 2,000 m^3)
Quantity of contaminated material: approx. 12,000 m^3 total in both piles
Geology: flysch (Eocene), limestones and dolomites (Cretaceous, Jurassic)
Facility: PVC factory (in operation)
Population in 10 km radius: approx. 300,000
Possible contaminated area: surrounding ground and littoral sea of Kaštela bay
Transportation route of contaminated material: coal has been transported by the sea from Rijeka and Bay of Kotor (Boka Kotorska) as well as by railway from adjacent north-Dalmatian coal-mine basin in hinterland of Šibenik (Dubravice, Širitovci)

There are two coal slag- and ash piles with increased radioactivity, situated at the INA-VINIL plant in Kaštel Sućurac, some 5 km northward of Split (population 250,000). While the older and larger pile (No. 1) was closed and covered by soil and PVC sheet, the pile No. 2 is still in operation. First pile, having dimensions 100 x 100 m (i.e. 10,000 m^2), is fairly organised and periodically controlled. As the average depth of stored material is about 1 m, total slag and ash quantity in this pile is some 10,000 m^3. The other, operating pile is considerably smaller. Position of both piles presents a remarkable environmental problem: since they are situated close to the seaside, slag and ash are accumulating in littoral zone and, in the case of the operating pile (No. 2) are being filled up directly into the sea. Slag and ash,



being actually deposited in the recent INA-VINIL pile, have remained after the burning of coal used as energy source for this, PVC synthesis and treatment plant.

Pile 1

Slag and ash were accumulated in this pile from 1950s until early 1970s. The material only partly originates from the PVC facility energy-plant (i.e. fire-room) itself, because remarkable quantities were transported by the sea from Rijeka and Boka Kotorska. After available data it is realistic to suppose that coal was mined mostly at small brown-coal mines in the karst area (carbonate lithology) of the Šibenik hinterland (Dubravice, Širitovci). Deposits shipped from Rijeka are in fact residues remained from combustion of black coal, mined at Labin and Raša in Istria. There is no reliable information on origin of deposits delivered from Boka Kotorska.

Table 7. Coal slag and ash stored in INA-VINIL's pile 1 (in Bq/kg)

Sample	^{40}K	^{228}Ra	^{226}Ra	^{238}U	^{235}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

Source: Archives of performed measurements, Institute Ruđer Bošković, Zagreb

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 being temporary applied in Croatia [8], the analyzed material is classified as low-radioactive solid waste).

After some calculations of uranium content in the slag and ash (by the Institute "Ruđer Bošković"), some 7.5 t ^{238}U and about 55 kg ^{235}U are contained in the piles. Some attention should be payed to surveying of adjacent family houses, which foundations were in some cases filled by contaminated material (referring to this, some health problems at locally living population have been reported). However, the opposition of local population and, especially, the "greens" against the further operation of industries situated in Kaštela bay has been derived not only for possible radiation contamination from the pile sites, but also for other possible harmful effects as it is pollution by mercury, vinyl-chloride monomer, "PVC-sludge" etc. Anyway, the idea of removal of the piles to some more convenient place is very popular in local population. Nevertheless, we have not got yet sufficient data to be decisive to recommend the removal of the piles.

In order to get current, accurate and reliable data on real radiation contamination at the site, we have planned to carry out sampling on both pile sites at grid 20 x 20 metres, i.e. to pick up some 30 samples (above all, gamma-spectrometry). TLDs and GM probes are foreseen to be set up on the site, and contamination should be measured during a period of some 6 months.

The sampling requires piercing of surface plastic sheet and digging of holes in overlying protection soil-cover for each sample. During on-site sampling and set up of measuring devices, operating members of the project co-ordination team will be accompanied by the state inspector for radiation protection. All measurements are foreseen to be performed at coal piles itself, as well as at concerned slag- and ash piles. After preliminary elaboration the site INA-VINIL is thought to be most delicate among all high-priority sites.

Pile 2

The site is located in-door the INA-VINIL facility area. The pile is partly attached to the above mentioned site (pile 1). It is worth mentioning that deposited material (i.e. coal-slag and ash) has been partly dumped into the sea. There are some rumours that certain quantities of slag has been exported in Italy. For a difference from the pile 1, the slag accumulating at this pile originates completely from the facility's energy-producing plant (i.e. fire-room). The pile contains slag and ash remained after burning of coal mined at Dubravice was used during the 1980s, as well as slag and ash from coal from Herzegovinian mines (probably the mine of Tušnica near Livno), which has been preferred in last few years. The results of measurements, performed during late 1980s, are given in Table 8. There is no information on radioactivity of slag and ash from coals, which have been used more recently.

It seems some quantities of the slag and ash to be 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 9 (measurements were accomplished in late 1980s). No additional measurements have been carried out during last few years.

Table 8. Coal slag and ash stored at the INA-VINIL plant /former JUGOVINIL/*

Sample	^{40}K	^{228}Ra	^{226}Ra	^{238}U	^{235}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

* *samples were being taken in the period 1988-1990 from both the fire-room and pile 2; given in Bq/kg)*

** *measured material is accumulated beneath the floor, i.e. in the foundation of a family house*

Source: Archives of performed measurements, Institute "Ruđer Bošković", Zagreb

Table 9: Content of radionuclides in cement and concrete produced at "Dalmacijacement" industry (Bq/kg)

Sample	^{40}K	^{228}Ra	^{226}Ra	^{238}U	^{235}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
Source: Archives of performed measurements, Institute "Ruđer Bošković", Zagreb

Note: Concrete samples present a mixture of cement and coal-slag and ash (in various shares), as well as limestone. Cement samples contain various percentage of coal and ash.

Recommendations for further actions:

(1) It is necessary to perform sampling and radiation measurements of coal being currently used, as well as at the slag and ashes landfills. Further measures depend on obtained results.

(2) Further sea-dumping of slag and ashes or their rejection anywhere into the environment must be stopped immediately, if no radiation measurements of the material are not previously performed.

(3) Coal for further fuelling of the plant fire-room must contain as low share of radionuclides as reasonably possible.

(4) It should be introduced an obligatory permanent radiation control of slag and ashes, used as admixture in cement industry.

(5) There are some indications that slag and ashes from the site INA-VINIL 2 (former JUGOVINIL) were used for filling up the foundations of family houses in the area of Kaštela (see the site Kaštel Lukšić in Table 8). This statement must be carefully examined.

2.1.1. Preliminary Risk Assessment for Slag & Ash Pile 1

In accordance with the Work Breakdown Structure, Task 7 (see Part I, ch. 2.1 of this report), we made some progress in risk assessment at INA-VINIL site, slag/ash pile 1 (as it is described previously). This assessment is rather preliminary and partial, so that only rough conclusions can be derived from it. Furthermore, the assessment covers only one, the closed slag/ash pile (No. 1) and does not refer to the other, operating one (No. 2). Nevertheless, we have got some useful indications from this part of foreseen activities on risk assessment at highly prioritised sites. Continuation of this task is expected to proceed in accordance with previously presented schedule.

This preliminary risk assessment was performed by the group of experts in the field, operating at Faculty of Electrical Engineering and Computing in Zagreb.

<u>Brief Description on Used Risk Assessment Tools</u>

The analysis was carried out by applying computer code RESRAD ("**Res**idual **Rad**ioactivity"), version 5.0. The code was developed by the Argonne National Laboratory - Environmental Assessment Division, in 1993, and was tested during the preparation of the US DOE document "Radiation Protection of the Public and the Environment" in 1990 (DOE Order 5400.5, Feb.). It is worth mentioning that part of the document which refers to "Recommendations on materials characterised by elevated and residual radioactivity", is prepared in March 1993 and has been subsequently included into 10 CFR (Code of Federal Regulations, Part 834).

Computer code RESRAD is organised in the way which enables calculation of radionuclide concentrations in ground (soil), water and air, as well as effective dose equivalents for individuals (EDE - Effective Dose Equivalent for outdoor radiation; CEDE - Committed Effective Dose Equivalent for indoor radiation) and consequent cancer hazards.

Calculation of radionuclide and dose concentrations is performed by means of so-called "concentration factor" method, based on summing up of "path factors" products like e.g. Dose Conversion Factor (DCF), Environmental Transport Factor (ETF), Source Factor (SF) and Branching Factor (BF). Product of these four factors represents Dose/Source Ratio, which - multiplied by specific activity of respective radionuclide - gives the dose value.

Risk assessment done by the RESRAD code is based on co-called EPA '92 method (in the following table indicated as EPA), which is derived from the "Slope Factor" concept, i.e. by distributing a specific factor to each radionuclide. By this factor the dose is converted into risk. This method is somewhat different from the ICRP methodology, which defined in a special publication (ICRP, No. 60; published 1991) sum factors for conversion of risks from both occupational and public dose values.

The RESRAD code is able to analyze nine scenarios, respecting three different paths of possible radionuclide migration:

A. *Outdoor gamma-radiation:* A1. soil (three-dimensional source, two-dimensional source); A2. air (dust, radon, radon daughters, other gaseous radionuclides); A3. water.

B. *Inhalation of radionuclides:* B1. dust; B2. radon and radon daughters; B3. other gaseous radionuclides.

C. *Ingestion of radionuclides:* C1. food (vegetarian food, meat, milk, sea-food); C2. water (groundwater, surface water); C3. soil.

<u>Parameters Used in Preliminary Risk Assessment of INA-VINIL, Pile 1</u>

Preliminary Risk Assessment of the INA-VINIL Pile 1 (Tab. 10) includes only firstly mentioned scenario, i.e. outdoor individual whole body radiation from soil (A1). The site (i.e. pile) was treated as an ideal cylindrical body (three-dimensional source) covering an area of 10,000 m^2 and being 1 m thick.

Presented analysis, done in accordance with the mentioned scenario, includes following options:

(a) an individual stays every day for 4 and 8 hours on the site (pile);

(b) calculated soil thickness covering contaminated materials is: (1) 0 cm, (2) 10 cm, and (3) 20 cm;

(c) the dose, i.e. risk is calculated (1) for the first year after performance of the analysis (1995/96) and (2) cumulative, for first five years after the analysis (i.e. the period 1995/96 - 2000/01). Calculation is based on presumption that contaminated material has been permanently stored on the site during past 40 years.

Note: According to recent Croatian legislation, the maximum allowed annual dose for individuals is 1 mSv, which would be in some cases higher if does not surpass the 5-year cumulative dose of 5 mSv (which is, in fact, based on mean annual dose of 1 mSv).

Tab. 10. Results of Preliminary Risk Assessment at INA-VINIL Plant (Slag / Ash Pile 1)

Coverage thickness	Daily exposure											
	4 hours						8 hours					
	Total exposure						Total exposure					
	1 year			5 years			1 year			5 years		
	Dose (mSv)	*Risk*		*Dose (mSv)*	*Risk*		*Dose (mSv)*	*Risk*		*Dose (mSv)*	*Risk*	
		EPA	*ICRP*		*EPA*	*ICRP*		*EPA*	*ICRP*		*EPA*	*ICRP*
0 cm	***1.50E+00***	5.58E-05	7.48E-05	***7.48E+00***	2.79E-04	3.74E-04	***2.98E+00***	1.11E-04	1.49E-04	***1.49E+01***	5.57E-04	7.46E-04
10 cm	8.29E-01	3.12E-05	4.15E-05	4.15E+00	1.56E-04	2.07E-04	***1.65E+00***	6.21E-05	8.27E+00	***8.27E+00***	3.11E-04	4 13E-04
20 cm	3.12E-01	1.19E-05	1.56E-05	1.56E+00	5.97E-05	7.76E-05	6.22 E-01	2.38E-05	3.11E+00	3.11E+00	1.19E-04	1.56E-04

Remarks:

(1) Dose values typed bold/italic are higher than maximum allowed values prescribed by our regulations (1 mSv/year or 5 mSv/5 years). It means that these risks are not acceptable.

(2) This preliminary risk assessment respected only one of possible realistic scenarios, i.e. outdoor radiation scenario. However, some of other scenarios, presented in PRA methodology description, could be also significant.

(3) There are some differences in risk assessment methodology between EPA 92 and ICRP apporaches. Results vary up to 20 %; the ICRP method is more conservative, i.e. it gives higher risk values than the EPA 92.

(4) This preliminary risk assessment shows that the INA-VINIL site deserves a special attention from the viewpoint of possible radiation impact to the environment, and it is not recommendable to leave it out of any control.

2.2. Power Plant PLOMIN

Site Description

Site: coal- and slag/ash piles

Quantity of contaminated material: approximately 900,000 tons

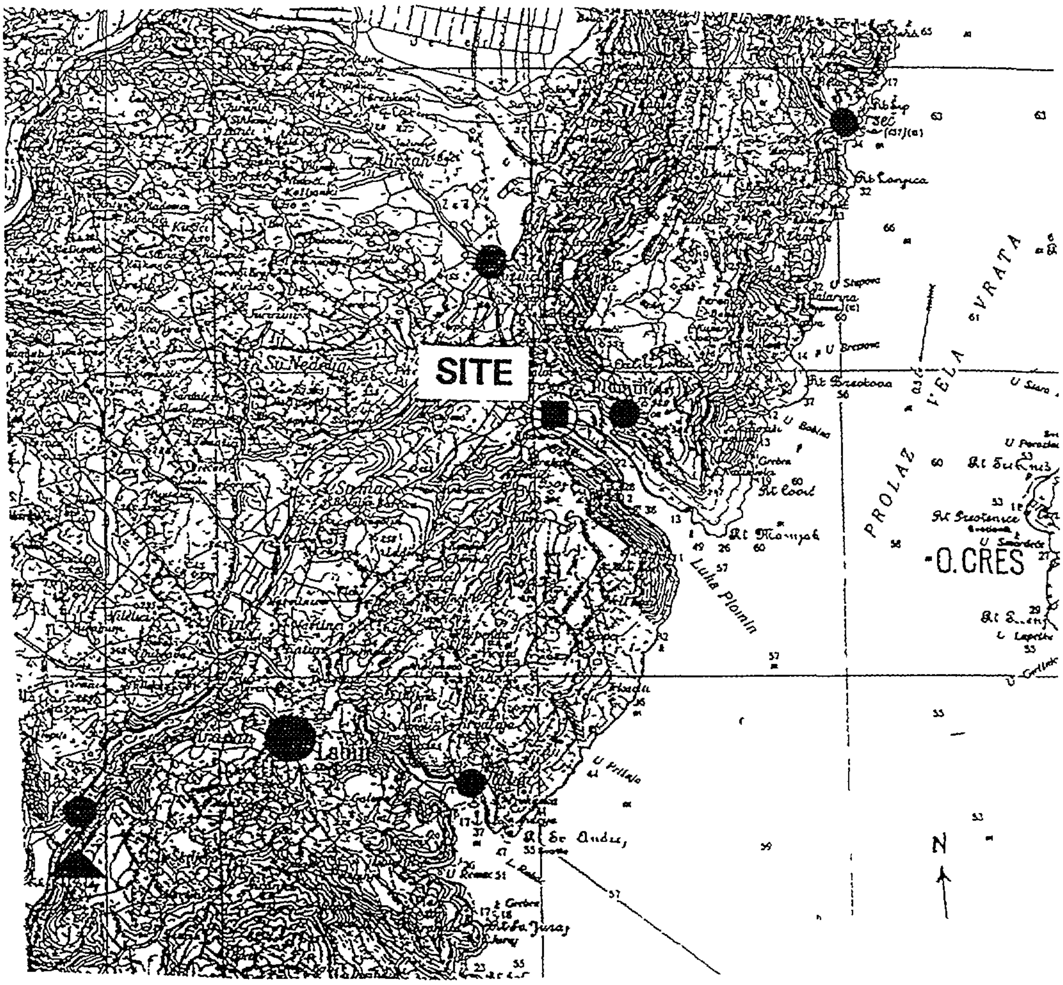
Geology: flysch (Eocene), limestones and dolomites (Cretaceous)

Facility: coal-fired power plant (in operation)

Population in 10 km radius: approx. 25,000

Possible contaminated area: neighbouring settlements, local streams, Plomin bay

Transportation route of contaminated material: coal has been mined mostly at adjacent Raša coal mine area (about 10 km from the plant site)



The coal slag/ash pile is situated close to the power plant site. Slag and ash is accumulating continuously, consequently to regular operation of the power plant. There have been performed some measurements of natural radiation (in marine and fluvial sediments) in vicinity of Raša coal mine and Plomin power plant, as well as in ashes generating at the power plant (Tab. 11). It should be added that besides the stored slag and ash, there is another source of pollution acting at the power plant: release of gas and contaminated smoke into the atmosphere.

Table 11. Radioactivity of stream sediments, ash and slag in Istria (in Bq/kg)

Site	^{40}K	^{228}Ra	^{226}Ra	^{238}U	^{137}Cs
FRACTION 0.063 mm					
(a) stream sediments					
Raša (bridge)	680	28	26	18	27
Potpićan (bridge)	471	38	46	30	8
Gologorički potok	516	27	31	35	hd
Floričići (cascade)	497	21	22	24	14
Raša 1	571	49	39	32	hd
Raša 2	674	47	39	36	hd
(b) marine sediments of Raša bay					
Target (- 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) stream sediments					
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	19	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

Source: Archives of performed measurements, Institute "Ruđer Bošković", Zagreb

Although we have not done any on-site measurement at Plomin power plant so far, the following facts referring to the site can be useful for further programme implementation:

(a) *Slag and ash* accumulated on the site have been generated by burning of coal mined in Raša coal-mine area (mines Ripenda, Tupljak, Koromačno). Unfortunately, this coal is characterised by remarkably high percentage of sulphur (up to 10-14 %) and naturally elevated share of uranium (^{238}U).

(b) Total quantity of *slag and ash* which has been accumulated on the site so far is about 900,000 tons.

(c) Past investigations [12,13] showed that *black coal* from the Raša mines, which was burned in the power plant Plomin I, is rich in uranium: ^{238}U concentration reaches in some samples 400 ppm, i.e. specific activity of ^{238}U in this coal is up to 4,900 Bq/kg. Radium (^{226}Ra) concentration in the coal is 21 ppm (260 Bq/kg).

Specific activity of ^{238}U in *collected flying ash* (at electro-filters) vary between 500-8,600 Bq/kg (concentration 40-700 ppm), whilst its mean activity is 2,260 Bq/kg. Specific activity of ^{226}Ra in flying ash is up to 2,600 Bq/kg (concentration about 210 ppm) [12,13].

Accumulated *slag or "bottom ash"* on the pile, which has remained after being burned in the plant, is characterised by ^{238}U specific activity 400-1,800 Bq/kg (concentration 33-147 ppm), and ^{226}Ra specific activity 800 Bq/kg (concentration 66 ppm) [12,13].

Activity of radioactive potassium (^{40}K) and thorium (^{232}Th), measured in slag and ash samples on the plant pile, is not remarkable and can not have any impact to the environment and human health.

Calculations performed by the Institute "Ruđer Bošković" [12], show that *annual activity of uranium and thorium,* being emitted through the plant chimney into the atmosphere, is 7.61 x 10^8Bq.

The activity concentration of accumulated ash at the plant pile, derived from specific activity of ^{238}U, is about 1.4 x 10^7 Bq/m^3. This activity represents in Croatian legislation [14] just a limit value for solid radioactive waste (for alpha-emitters). However, it should be emphasised that requirements contained in the mentioned regulation are "mild" in relation to current regulations of European Union countries and the United States. For this reason, possible radiation pollution from the Plomin power plant facility and slag/ash pile must be perceived seriously. Nevertheless, it is worth mentioning that past radiometric investigations in Istrian peninsula [15] show at fairly high background activity in this region, so that e.g. the content of thorium and radium in Istrian stream sediments is remarkable.

According to another regulation [16], slag and ash collected on the plant pile are not allowed to be used as house-building material, since its specific activity surpasses the maximum allowed limit values (400 Bq/kg for ^{226}Ra).

(d) The slag/ash pile is situated on low permeable Eocene flysch. However, this sediment could be very easy weathered and affected by proluvial processes (torrents). Geological setting of the broader area is characterised by Mesozoic carbonates (limestones, dolomites) which are due to irregular circulation of groundwater environmentally very sensitive and, thus, the monitoring of radionuclides which would possibly migrate in the groundwater from this pile, is not easy. Besides the immediate surrounding area of the plant, radioactively polluted material affects also the zone between the plant and the seaside, as well as submarine area of the Plomin bay.

(e) Environmental preservation and human health protection measures, performed on the site so far, have been directed basically to *covering of contaminated material* by soil and clayey material (which can remarkably prevent ingression of rainfall into contaminated material, but also disable deflation, i.e. blowing the material away). The existing fence surrounding the pile is an additional protection measure: it prevents the access of uninvited persons and possible carrying the stored material away. Finally, the pile site is provided by drainage system which considerably lowers erosional, derasional and proluvial processes, whilst retention pool - situated between the plant and the coastline - diminishes sedimentation of eroded terrestrial material (including stored slag and ash) into adjacent Plomin bay.

(f) Environmental clean-up of the site, i.e. slag/ash pile, should be based on the above mentioned facts, but necessity of further plant operation must be also considered. As local coal mines has been almost exhausted, it has been already decided that imported coal with low share of sulphur and uranium will be used as fuel for plant operation. Detailed site characterisation and consequent risk assessment will show which clean-up method would be preferable for safely insulation of contaminated slag and ashes. Pile closure, conservation or removal are some of options, but - disregarding the finally chosen method - the monitoring of the site should start as soon as possible.

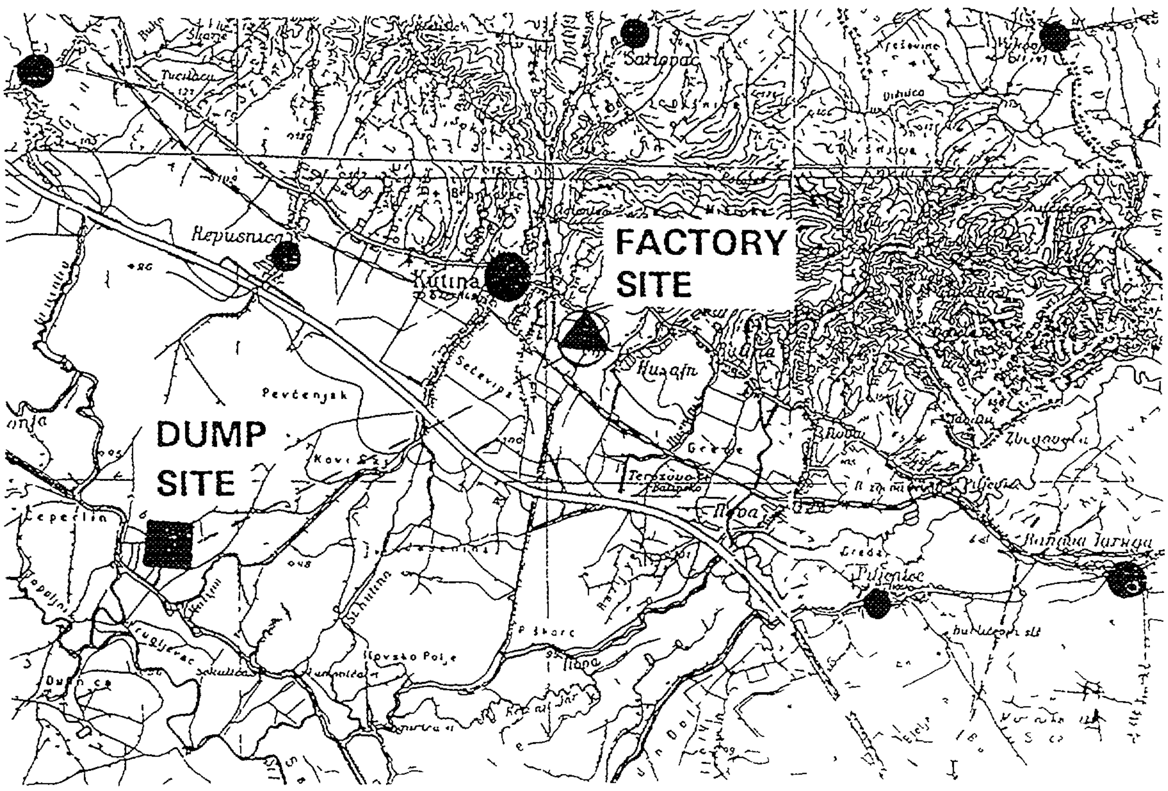
In order to identify real recent pollution, the following sampling and measurements are about to start:

* gamma-spectrometry and radiochemical analysis of coal-, slag- and ash-samples from the piles at the power plant;
* measurements of natural radionuclide concentrations (6 air-samples) taken inside the 20 km radius around the power plant;
* measurements of natural radionuclide concentration in soil (4 samples) taken within the 2 km radius around the power plant;
* measurements of radionuclide concentrations in pedological horizons at few vertical profiles (3-5 samples per profile) in order to determine vertical migration of radionuclides (especially uranium) which could be caused by acid rains.

Finally, there has been taken into consideration measurement of possible radiation contamination in marine sediments in the Plomin bay. This idea is initiated by the fact that long-term pollution of the bay seabed is very probable due to continuous accumulation of polluted terrestrial sediments which has been entering the bay by activity of local streams.

Stream sediments in the plant vicinity are also supposed to be sampled in order to prove radioactive contamination caused by inadequate disposal of slag and ash at the plant.

2.3. INA-PETROKEMIJA Fertilizers Plant in Kutina



Site Description
Site: waste/phospho gypsum landfills (4 pools)
Quantity of contaminated material: 3.5 million cubic metres
Geology: Quaternary fluvial sediments (alluvium) - mud, sand, gravel
Facility: phosphate fertilisers plant (in operation)
Population in 10 km radius: approx. 35,000
Possible contaminated area: phospho-gypsum landfills, arable land where fertilisers are applied, streams running through fertilised croplands, groundwater beneath phospho-gypsum landfills.
Transportation route of contaminated material: (a) phosphate ore /railway Rijeka-Karlovac-Zagreb-Kutina; formerly, railway Šibenik-Knin-Sisak-Kutina/; (b) fertilisers /throughout Croatia, but mainly in the interior of the country - e.g. Slavonia; transported by railway or lorries/

The fertilizer plant INA-PETROKEMIJA in Kutina consists of two sites where increased radiation is expected: (1) the factory indoor area (phosphates as raw material, phosphate acid, fertilizers as final products), and (2) phospho-gypsum landfills, lying some 5 km southward from the factory (see the map). As the sites of fairly increased radiation contamination are identified agricultural lands where fertilizers are used. Basic difference between a nature of contamination of these two types of sites is derived from the fact that ^{238}U is identified as a basic radiation pollutant in fertilizers, whilst ^{226}Ra prevails in phospho-gypsum. Although routes of phosphates (raw materials) have been precisely defined, it is not realistic to expect any considerable contamination along transportation routes from the entering Croatian ports (Šibenik, Rijeka) to the factory in Kutina. Basic input data on radioactive contamination point at increased radioactivity of all components consisting processing cycle, as it is shown on the following tables:

Table 12. Radioactivity of raw material (in Bq/kg; sampled in 1988)

Raw material (origin)	^{40}K	^{228}Ra	^{226}Ra	^{238}U	^{235}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-USSR)	16,400	nm	nm	nm	nm
Potassium salt 6 (ex-USSR)	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 (Source: Institute "Rudjer Boskovic", Zagreb)

Table 13. Radioactivity of phosphoric acid and mono-ammonium phosphate /MAP/ (in Bq/kg; sampled in 1988)

S a m p l e	^{40}K	^{228}Ra	^{226}Ra	^{238}U	^{235}U
Phosphoric acid 1	24	3	6	3,020	140
Phosphoric 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

Source: Archives of performed measurements, Inst. "Ruđer Bošković", Zagreb

Table 14. Radioactivity of waste /phospho/ gypsum (in Bq/kg; sampled 1988)

Sample	^{40}K	^{228}Ra	^{226}Ra	^{238}U	^{235}U
Gypsum 1	122	19	708	576	27
Gypsum 2	90	25	624	545	25
Gypsum 3	148	17	733	566	26

Source: Archives of performed measurements, Inst. "Ruđer Bošković", Zagreb

Table 15. Radioactivity of phosphate fertilisers /final products/ (in Bq/kg; sampled in 1988)

Sample / type	^{40}K	^{228}Ra	^{226}Ra	^{238}U	^{235}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	38

Source: Archives of performed measurements, Inst. "Ruđer Bošković", Zagreb

Due to significant variations in contents of ^{226}Ra and ^{238}U, it seems reasonable to introduce a permanent control of radioactivity in imported phosphates.

Investigations of phosphate fertilizers used in eastern Slavonia [17] pointed at following radioactivity: 75 Bq/kg ^{226}Ra, 9 Bq/kg ^{228}Ra, 52 Bq/kg ^{235}U and even 1,120 Bq/kg ^{238}U. The estimated annual deposition of uranium and radium in soils of agricultural fields in the area of Vinkovci is 4.5 Bq/m^2 for ^{226}Ra, 0.5 Bq/m^2 for ^{228}Ra, 3.1 Bq/m^2 for ^{235}U and 67 Bq/m^2 for ^{238}U. The highest concentrations of both uranium isotopes, measured in drainage channels water, have mean values of 120 Bq/m^3 for ^{238}U and 5.5 Bq/m^3 for ^{235}U.

Anyway, the most environmentally sensitive point in the fertilizer production are landfills of phospho-gypsum. There are four pools (landfills) organised in the floodplain of Sava river, some 5 km southward from plant in Kutina. Their size is 43 hectares (ha), 33 ha, 28 ha and 32 ha respectively, i.e. the landfill covers a total surface of 136 ha. Pools are arranged along an area 1 km long and 700 m wide. Total capacity of pools is 20 million cubic metres, but only 3.5 million cubic metres of phospho-gypsum have been stored so far. Waste gypsum, mixed with water, is transported from the factory to pools by special pipeline. Radionuclide contents of phospho-gypsum itself, groundwater and waste-water is controlled continuously (their highest concentration values are given above). Nevertheless, it is out of any doubt that clean-up of these pools, which are indispensable for regular operation of INA-PETROKEMIJA factory, represents highly recommendable action necessary for improvement of environmental quality and, consequently, human health protection. Some improvement in fertilizer production should be also discussed since generation of 4-5 tons of phospho-gypsum (as waste material) from production of 1 ton of phosphoric acid does not seem to be reasonable and represents considerable ecological burden.

2.3.1. Basic Results of Measurements at INA-PETROKEMIJA Plant, Done in the Frames of the Programme Performance

Most of planned data on radiation characterisation at the INA-PETROKEMIJA plant indoor area in Kutina (i.e. excluding phospho-gypsum landfills) have been collected so far (sampling and measurements were performed in the period March-October 1994). Presented numerical results of the performed measurements are referring to the following topics:

(M 1) radiochemical analysis of ^{226}Ra in groundwater and well-water;
(M 2) radiation doses in airborne samples (measured by TLDs);
(M 3) gamma-spectrometry of phosphates (raw material), fertilizers (final products), phospho-gypsum (waste material) and airborne samples;
(M 4) measurement of Working level (^{222}Rn daughters)
(M 5) sampling and measurement of radon concentrations;
(M 6) estimate of ^{226}Ra activity at the phospho-gypsum landfill;

M.1. Radiochemical Analysis of ^{226}Ra in Groundwater and Well-Water

<u>CaF_2-$Si(OH)_4$ /sediment/ landfill</u>	
* vertical piezometer D1	112.2 +/- 12.2 mBq/l
<u>Phospho-gypsum landfill</u>	
* horizontal piezometer D2	112.7 +/- 12.2 mBq/l
* vertical piezometer D3	165.9 +/- 31.5 mBq/l
* vertical piezometer	87.5 +/- 12.9 mBq/l
* horizontal piezometer	101.4 +/- 13.4 mBq/l
<u>Well-water samples</u>	
* family house in Radićeva street 388	36.5 +/- 11.4 mBq/l

Comment:

Alpha-spectrometrical analysis, carried out after radiochemical separation, found ^{226}Ra in all water samples. Activity of ^{226}Ra determined in piezometers analyzed in 1994, varies between 88 and 166 Bq/m^3. These values lie within the span of results of former measurements, i.e. between 20-170 Bq/m^3. Such a wide span is caused by different starting activities of raw materials, which were used in the plant processing technology. The ^{226}Ra activity in well-water 1994 was 37 Bq/m^3 in 1994. For comparison, a mean value of ^{226}Ra-activity in past few years were 32 Bq/m^3 (i.e. even for a value order higher than mean value for water-pipe water, which is about 2 Bq/m^3). Higher activity in well-water is a consequence of specific position of the sampled well, which is situated in immediate vicinity of phospho-gypsum landfill.

The "Law on Taking Over the Federal Laws in the Field of Health Protection, Applying in the Republic of Croatia as Republic Laws" [1] defines the upper level of allowed radioactivity concentration in drinking water, following the concentrations in drinking water for individuals. For ^{226}Ra this derived concentration is 1,000 Bq/m^3.

The measured activity of ^{226}Ra in well-water, given in the above presented table, is only 4 % of maximum allowed derived concentration for drinking water.

M.2. Radiation Doses in Airborne Samples

Radiation doses in airborne samples, measured by thermo-luminescent dosimeters /TLDs/ (which are presented below), are obtained during the measurement (exposing) period 8 March - 5 October 1994, i.e. in 211 days. Doses are given in micro-grays (1 µGy = 1 µSv):

TLD No.	LOCATION	MEASURED DOSE (in 211 days)	ANNUAL DOSE (365/29 x D_{211})
44/1	Town Sport Hall (2,5 km from INA-P)*	101	1,271
42/1	Public Restaurant (Kutina)	490	848
37/1	INA-PETROKEMIJA Lab. (r.19/1)	440	761
49/1	The nearest house to the phospho-gypsum landfill (Radićeva street 388)	570	986
14/1	Phosphoric acid warehouse	**1,270**	**2,197**
17/1	MAP/NPK - new facility (warehouse of white KCl)	**820**	**1,418**

* TLD No. 40/1, which was supposed to be used after 211 days, disappeared and - therefore - the preliminary value obtained after 29 days (in April 1994) is given here.

TLD No.	L O C A T I O N (in 211 days)	MEASURED DOSE (365/29 x D_{211})	ANNUAL DOSE
10/1	MAP/NPK - new facility (warehouse of red KCl)	**880**	**1,522**
27/1	MAP/NPK - new facility (warehouse of BOUCRA phosphate & quartz sand)	**940**	**1,626**
36/1	MAP/NPK - new facility (granulator)	660	1,142
22/1	MAP/NPK - new facility (command room)	480	830
23/1	MAP/NPK - new facility (northern warehouse of final products)	420	727
48/1	Phosphoric acid facility (vice-director room)	560	969
01/1	Phosphoric acid facility - phosphate milling plant (command room)	500	865
39/1	Phosphoric acid facility - filtration (filters)	**1,040**	**1,799**
05/1	Phosphoric acid facility (command room)	440	761
28/1	Packing area III - NPK lines 8 & 9	390	675
15/1	Packing area I (old) - lines 5 & 6	420	727
47/1	NPK-1 (old) - command room	480	830
04/1	NPK-1 (old): at spherodizer	480	830
25/1	NPK-1 (old): warehouse of final products**	**120**	**1,510**
07/1	Phospho-gypsum landfill: pumping station	440	761

** TLD No. 26/1, which was supposed to be used after 211 days, disappeared and - therefore - the preliminary value obtained after 29 days (in April 1994) is given here.

Note: For comparison pay attention to the following annual doses at meteorological observing points: Bjelovar - 1,023 micro-Sv, Daruvar - 1,062 micro-Sv, Sisak - 981 micro-Sv (measured values which are considerably higher than background radiation are printed bold).

Comment: Doses were measured by TLD based on CaF_2Mn. Dosimeters were read out after exposing period by reader TOLEDO 654 (Vinten). Since annual doses of neighbouring towns (Sisak, Daruvar, Bjelovar) are about 1,000 micro-grays, it is reasonable to expect similar background value in Kutina as well.

The above presented doses are even lower than all preliminary doses measured in April 1994, after 29 day-exposure period (excluding the sample 39/1). It can be explained in the following way:

(a) results of preliminary measurements were obtained after only 29 days, when the one-day error contributes to aberrance of even 10 %;

(b) the highest doses were registered in warehouses, which had been mostly full of phosphates at the beginning of measurement (i.e. in the moment of dosimeter set up). However, the quantity of phosphates was varying during summer period, so that no phosphates or potassium chlorides were stored in vicinity of dosimeters. Hence, the latest values were slightly lower that preliminary ones (those, read out in April 1994);

(c) dosimeters were exposed during extremely hot summer, what resulted due to "fading" in lower values.

M.3. Gamma-Spectrometry of Phosphates, Phosphate Fertilizers (Final Products) and Phospho-Gypsum (Waste Material)

The following results, obtained by gamma-spectrometry of phosphate samples, final nitrogen-phosphor-potassium (NPK) products and waste (phospho) gypsum, were performed in April 1994 (results of gamma-spectrometric measurement of airborne samples are presented in the section M.3.1., after the below given comments on possible improvement of the plant processing technology). Specific activity of samples shown in the following table is given in Bq/kg:

S A M P L E	^{40}K	^{228}Ac	^{226}Ra	^{235}U	^{238}U
KCl (white)	15,939	nd	nd	nd	nd
KCl (red)	16,132	nd	nd	nd	nd
K-sulphate	13,824	nd	nd	nd	nd
BOUCRA phosphates					
Sample 1	11	7	848	26	565
Sample 2	27	12	1,229	43	938
Sample 3	15	8	830	27	577

SAMPLE	^{40}K	^{228}Ac	^{226}Ra	^{235}U	^{238}U
MOROCCO phosphates					
Sample 1	16	11	1,140	44	951
Sample 2	28	8	1,120	42	917
Sample 3	9	9	1,423	51	1,103
Phosphoric acid					
High concentrated - sample 1	32	4	12	77	1,680
High concentrated - sample 2	28	3	10	77	1,681
Low concentrated - sample 1	3	2	4	39	853
Low concentrated - sample 2	6	1	7	41	897
Phospho-gypsum					
Landfill (1/94)	17	5	377	1	4
Filter (3/94)	25	6	702	2	47
Mono-ammonium phosphate (MAP)	32	2	31	83	1,797
NPK-fertilizers					
20-10-10	2,438	2	95	20	440
13-10-12	3,153	3	192	10	210
13-13-21	5,296	2	46	9	202
15-15-15 (1)	3,795	3	94	18	392
15-15-15 (2)	3,885	3	114	18	392
08-16-24	6,358	2	130	18	384
18-18-18	4,924	3	10	28	611
08-26-26	7,016	3	7	36	780
Soot	4	nd	nd	1	22

nd = not detectable

Comment : In accordance with the given gamma-spectrometry results, the members of Project team have given *the following statements and recommendations related to improvement of the plant processing technology* (in both economic and ecologic senses) to the INA-PETROKEMIJA Plant Management:

A. RAW MATERIALS

A.1. Potassium chloride (KCl)

Both KCl samples show high purity (white KCl about 98.3 %, red KCl about 99.5 %; possible deviation is not higher than 1 %). Namely, the measured specific activities of ^{40}K are roughly identical to specific activity of chemically pure KCl (i.e. 16,220 Bq/kg). Due to insignificant content of admixtures, all other observed radionuclides (^{238}Ac, ^{226}Ra, ^{235}U, ^{238}U) have not been detected in any of samples.

A.2. Potassium sulphate (K_2SO_4)

The measured specific activity of ^{40}K in the potassium sulphate sample shows at very high purity of raw material (99.6 %, i.e. 13,824 Bq/kg in the sample, related to 13,880 Bq/kg in chemically pure potassium sulphate). Specific activity of other observed radionuclides is equal to zero, as it was expected due to almost quite chemically pure sample.

Conclusion and recommendations

On the basis of the performed sampling and measurements (but referring also to the measurements done in the late 1980s), it is obvious that potassium concentrations in raw materials are in harmony with their chemical composition. Further determination of potassium specific activity in potassium salts is not considered to be necessary, with exception of control measurements if source of raw materials is changed. Potassium load degree of agricultural lands can be simply identified and calculated in accordance with the annual plant consumption of potassium salts alone.

A.3. Phosphates

The term "phosphates" or "phosphorites" is used for rocks abundant in phosphorus containing minerals (e.g. monazite, triphyline, copite, colinsite, lithophylite, vivianite, guanite, monetite, phylovite, pyro-phosphorite, natriphylite etc.). Main natural phosphor bearing minerals are apatite, collophane and dalite.

Apatite, $Ca_5(F,Cl)(PO_4)_3$, has either igneous or sedimentary origin. Apatite consisting igneous rocks originates through crystallization of magma as an accessory mineral, and often presents an admixture in biotite and quartz. Apatite found in sedimentary rocks is known as basic mineral of phosphorite and - in opposite to igneous apatite - does not include higher radionuclide concentrations of thorium series.

Collophane, $3Ca_3(PO_4)_2 \times n\ Ca(CO_3, F_2O) \times H_2O$, is calcium-carbonate-phosphate, mostly known by amorphous structure, although there are also samples of crypto-crystalline collophane. It is mainly solid, and in some cases oolitic. Mineral fragments of glauconite, carbonate and biogenous opal, as well as remnants of organisms are often found incorporated into the mineral. It is sedimentary (marine) mineral, found in phosphorites as white, yellowish-white to brown matrix.

Dalite, $3Ca_3(PO_4)_2 \times CaCO_3$, generates by recrystallisation of collophane and is known as a crust on phosphate rocks.

From the above mentioned it gets obvious that phosphates can be described by the general formula $(Ca_3(PO_4)_2)_3CaF_2$, but content of admixtures (mainly carbonate component - $CaCO_3$ can be fairly high and is usually indirectly proportional to the content of pure phosphor.

A.4. "Boucra" phosphates

Measured specific activities of ^{228}Ac show at marine sedimentary origin or raw material wherein the phosphoric component is probably completely related to collophane matrix and - in less extent - maybe to dalite. Radiochemical balance of ^{226}Ra and ^{238}U is disturbed "in favour of" ^{226}Ra (which prevails in shallow or even surface layers) at all samples. This situation can be explained by migration of ^{238}U in deeper formations. Concentrations of ^{226}Ra and ^{238}U are very similar in the samples 1 and 3 (both samples originate probably from the same, surface or very shallow layer). Concentrations of ^{226}Ra at the sample 2 are elevated (46 % higher than at other samples), while concentrations of ^{238}U are higher for some 64 % than a normal value (the layer is slightly deeper but also fairly shallow). The same conclusion can be derived from relation ^{226}Ra/^{238}U in the sample 2 (1.31) referring to the samples 1 and 3 (where the same relation is 1.47).

A.5. "Morocco" phosphates

Similar to the "Boucra" phosphates, "Morocco" phosphates are undoubtedly of marine sedimentary origin. Their radiochemical balance is disturbed "in favour of" ^{226}Ra, but not so apparently (about 1.25) as in the case of the "Boucra" phosphates. Concentration rates of ^{226}Ra and ^{238}U are somewhat higher than at "Boucra" phosphates, but they are considerably lower in relation to concentrations measured in late 1980s at "Morocco" phosphates. In distinction from the sample 3, the samples 1 and 2 originate probably from the same layer.

B. PHOSPHORIC ACID

Specific activity of all analyzed radionuclides, excluding uranium isotopes, at all samples is expectedly low. High specific activity of uranium in all samples of phosphoric acid are caused by the fact that uranium is "bounded" with phosphor. Thus, uranium concentrations in phosphoric acid are proportional with uranium concentrations in raw phosphates and phosphor content in phosphoric acid, i.e. phosphates. According to the content of phosphor in pure phosphates (containing no carbonatic admixtures) and phosphoric acid, it is possible to conclude that 1.72 tons of phosphates are required for production of 1 ton of phosphoric acid.

<u>*Conclusion and recommendations*</u>

Control of specific activity of uranium in phosphoric acid is not necessary if ^{238}U concentration in raw phosphate is not higher than some 1,000 Bq/kg. Namely, the uranium content in phosphoric acid in pure phosphates (content *of P^2O^5 is about 42.2 %) is simply detectable by multiplication of uranium content in phosphate by factor 1.72 if no uranium is transferred into waste gypsum. In addition, if phosphates are not pure and uranium concentrations are lower than 1,000 Bq/kg, it is also possible to calculate content of uranium in unit-amount of phosphor, and resulting value multiply with 31.6 (expected error is negligible). Measurements of specific activity of ^{238}U in phosphoric acid and phospho-gypsum, performed at the site in late 1980s, showed that uranium transfer from phosphates with fairly high content of ^{238}U (2,000-2,500 Bq/kg) into phospho-gypsum, is considerable. Hence, it is obvious that the uranium transfer from phosphates into phosphoric acid is lower than the factor value (1.72).*

C. WASTE (PHOSPHO) GYPSUM

Waste gypsum (or "phospho-gypsum") generates in production of phosphoric acid as it is described by the following reaction:

$$(Ca_3(PO_4)_2)_3CaF_2 + 10H_2SO_4 + 2H_2O \text{-----} 6H_3PO_4 + 10CaSO_4 \times 2H_2O + 2HF$$

In the case of pure phosphates some 3 tons of waste gypsum remain after production of 1 ton of phosphoric acid. As most of admixtures in phosphates are carbonate compounds, the resulting amounts of waste gypsum exceed about 4 tons per 1 ton of produced phosphoric acid (if P_2O_5 content in phosphates is about 33 %). ^{226}Ra, contained in phoshpo-gypsum, is fully incorporated into qypsum, replacing the homologous calcium in chemical structure of gypsum. Some previous measurements of waste gypsum radioactivity showed that ^{238}U comes in less amounts also to gypsum if uranium concentrations in raw phosphate are higher than 1,000 Bq/kg. Anyway, this interesting problem is not yet known in details, and for more accurate conclusions additional investigations are necessary. Hereby, some possible disturbances of processing of phosphoric acid could be additional cause of ^{238}U removal into waste gypsum. In that case, a considerably elevated content of uranium into gypsum could point at phosphor losses in processing of phosphoric acid. Therefore, we suggest the radium and uranium concentrations to be continuously monitored, in particular in case of increased uranium concentrations in raw phosphates (i.e. if the values are remarkably higher than 1,000 Bq/kg).

Conclusion and recommendations

Annual rate of ^{226}Ra generation at waste gypsum landfill can be assessed on the basis of radium concentration in raw phosphate. Since no systematic measurement of uranium and radium concentrations in imported phosphates has been performed so far, the estimate of presently accumulated amounts of radium and uranium can be done only through detailed sampling at the phospho-gypsum landfill. Due to accumulated quantities of waste materials and high variability of ^{226}Ra and ^{238}U in raw phosphates, the emplaced radionuclides could be estimated more accurately by radioactivity measurements of at least some fifty gypsum samples, taken from the entire landfill area in accordance with convenient sampling network.

D. NITROGEN-PHOSPHOR-POTASSIUM (N-P-K) FERTILIZERS AND MONO-AMMONIUM-PHOSPHATE (MAP)

Uranium and radium concentrations in fertilizers and mono-ammonium-phosphate (MAP) are in keeping with their concentrations in treated phosphate ore. It is worth mentioning that uranium and radium concentrations in series of measured fertilizer samples are considerably lower than in fertilizers measured in late 1980s. According to measurements of uranium and radium concentrations in phosphates and nitrogen-phosphor-potassium (N-P-K) fertilizers, it gets clear that fertilizers have been produced from phosphates containing uranium and radium concentrations even lower than those, identified in phosphates. The only exceptions are probably two samples of N-P-K 15-15-15, a sample N-P-K 8-26-26 and a sample N-P-K 8-16-24 (see the Table in ch. M.3), where declared content of phosphor convenes entirely to ^{226}Ra and ^{238}U shares in Boucra-phosphates /samples 1 and 3/ (Table in ch. M.3), assuming the phosphate purity is 80-85 % . All measured samples were produced directly adding different portions of raw phosphates; exceptions are two samples: N-P-K 18-18-18 and N-P-K 8-26-26 (Table in ch. M.3) - which were produced exclusively from phosphoric acid.

Conclusion and recommendations

Measurements of radionuclide concentrations in fertilizers are not necessary if content of radionuclides in treated phosphate ore and processing method is known. In accordance to the mentioned findings, our strong recommendation to the Plant management staff is to use phosphate ore containing the higher possible share of phosphorus and the lower possible portion of uranium and radium in further operation. Thus, the maximum possible economic benefit with minimum environmental burden would be achieved.

E. SOOT

Concentrations of all measured radionuclides in soot samples are expectedly low or even equal to zero value (the only exception is ^{238}U). Since the carbon content in soot is high due to incomplete combustion, the measured content of ^{238}U can be accepted as normal and additional control is not necessary. Namely, increased concentrations of ^{238}U can be expected only in solid residuum after complete combustion.

M.3.1. Gamma-Spectrometry of Airborne Samples (HVS-High Volume Sample)

The analysis is based on on-site sampling (in the operational area of INA-PETROKEMIJA plant), which was performed in the period 13-16 June 1994. Values are given in Bq/m^3:

^{7}Be	^{40}K	^{226}Ra	^{232}Th	^{235}U	^{238}U
1,48 E-2	< 1,48 E-3	< 5,82 E-4	< 2,22 E-4	< 3,32 E-4	< 1,02 E-2

Comment: Airborne sample is obtained by pumping at 1 m above ground. Sampling is carried out by "Glass-fibre" filters, and measurements were performed using Ge(Li) detector during 80,000 seconds period. All measured values were not detectable with exception of ^{7}Be. However, this radionuclide is cosmogenic and cannot be created in production of phosphate fertilizers. Hence, the presence of ^{7}Be is not influenced by the INA-PETROKEMIJA plant operation.

M.4. Measurement of Working Level (^{222}Rn Daughters)

The following results were obtained from the measurements performed in the period 13-16 June, 1994:

LOCATION	mWL*	WLM**
Phosphoric acid facility		
Command room	4,86	1,67
Phosphate milling	3,31	1,13
Phosphate warehouse	10,10	1,72
NPK - new facility		
Command room	8,04	1,37
Front-side of granulator	6,70	1,14
KCl warehouse	5,36	0,91
NPK - old facility		
Command room	2,30	0,39
Spherodizers	3,88	0,66
Phosphate warehouse	3,19	0,54
Landfills		
Phospho-gypsum landfill	6,14	1,09
SiF landfill	9,68	1,65

* mWL = WL E-3 = WL x 10^{-3} = 0,001 WL
** WLM = 170 WL (calculation based on 170 work-hours a month)

Comment: Calculated values are similar to those, obtained by measurements performed by the "Institute for Medical Research and Occupational Health" in the 14-years period.

M.5. Sampling and Measurement of Radon Concentrations

Following values are obtained by sampling carried out in the period March-October 1994:

	LOCATION	CONCENTRATION (Bq m^{-3})
01	Phosphate warehouse	165 +/- 7
03	MAP/NPK: New facility - phosphate warehouse	20
04	MAP/NPK: Granulator	53
05	MAP/NPK: Command room	25 +/- 3*
06	MAP/NPK: North warehouse	122
07	Phosphoric acid facility: Office	19 +/- 2
08	Phosphoric acid facility: Phosphate milling - command room	13
09	Phosphoric acid facility: Filtration	12
10	Phosphoric acid facility: Command room	12 +/- 2
14	NPK-1 (old facility) - at spherodizer	121 +/- 6
32/1	Package room 1 (old): line 5/6	detector damaged
38/1	Package room 3: NPK lines 8,9	detector damaged
	Laboratory (room 19/1)	26 +/- 1
	Restaurant	53 +/- 2

* *Measured by detector placed in diffusion chamber.*

C o m m e n t :

Radon detectors (open and placed in diffusion chambers) were set up in chosen indoor places of the "INA-PETROKEMIJA" Plant in March 1994. Radon measurements were performed by solid detectors containing films KODAK LR-115. These detectors enable measuring of radon concentrations, as well as calculation of risks from radon inhalation by occupational population.

On the basis of surficial trace density on open detectors, which were exposed for a month, specific activity of radon varied between 12-123 Bq/m^3. For a more accurate estimate of radon concentration, open (cassette) and sealed detectors (diffusion chambers) have been exposed for a longer period - approximately for 7 months. Namely, the estimate of received dose for occupational population was made possible by reading out of two detectors. Obtained data (few films have been unfortunately lost) show at somewhat elevated radon concentrations at three measuring points, but they are all remarkably below the allowed limits for working areas.

Recent ICRP recommendations for limits of specific radon indoor activity (ICRP 65, [18]) define the "action level", i.e. the radon activity which requires additional measures to be decreased. The action level for resident areas (houses, flats etc.) varies between 200-600 Bq/m^3, whilst the same value for working areas is 500-1,500 Bq/m^3.

Although past measurements did not precisely estimated radon specific activity (another measuring method should be applied), it is obvious that *measured radon activity is below the upper limit of "action level".* However, performed measurements do not indicate occupational doses in INA-PETROKEMIJA indoor area, which would be received by inhalation or radon and its daughters, because detectors in diffusion chambers have been lost. It is reasonable to conclude that these low specific radon activities - in spite of stored material having increased radium concentration (e.g. phosphates) - are a consequence of permanent openness of warehouses, i.e. continual ventilation of these areas.

M.6. Estimate of ^{226}Ra Activity at the Phospho-Gypsum Landfill

This estimate is based on 3.7 million tons of phospho-gypsum being currently stored at INA-PETROKEMIJA landfills. Specific activity of stored phospho-gypsum is 537 Bq/kg, but total ^{226}Ra activity contained in phospho-gypsum is about 1,987 billion Bq (i.e. 1.987 E12 Bq). In fact, this activity is equal to some 53.7 grams of ^{226}Ra.

YEAR	MEAN ACTIVITY (Bq/kg)	NUMBER OF SAMPLES	ANALYZED BY
1984	337	8	IMI*
1985	402	4	IMI
1988	726	1	IMI
1988	688	3	IRB**
1989	1,130	1	IMI
1990	1,160	1	IMI
1993	674	2	IMI
1994	540	2	IRB

* Institute for Medical Research and Occupational Health
** Institute "Ruđer Bošković"

Comment: The estimate is performed under supposition that all available measurements were related to samples which are characterised by the same weight and same annual quantity of generated waste gypsum. Since the ^{226}Ra activity in waste gypsum is a consequence of radium activity in phosphates - and for some periods (1986-87, 1991-93) data are missing or number of samples is very restricted (1-2 per year) - the estimate is extremely rough. A more precise estimate is not possible before suggested sampling at phospho-gypsum landfill will be performed (according to /3/ in below given "Recommendations for Further Radiation Protection Measures at the Plant").

Recommendations for Further Radiation Protection Measures at the Plant

In order to improve processing methods, i.e. facility operation, and to decrease environmental risks at the INA-PETROKEMIJA Plant, we have recommended to the plant management staff to introduce following actions:

(1) to measure all imported phosphate shipments continuously (3-5 samples per ship);
(2) to measure periodically radiation contamination of waste (phospho) gypsum (1 sample from filters monthly);
(3) to carry out detailed sampling of phospho (waste)-gypsum landfill (some 50 samples per each pool);
(4) to control regularly possible groundwater contamination in piezometers adjacent to the phospho-gypsum landfill;
(5) to measure Working level periodically;
(6) to measure exposure doses of gamma-radiation by TLDs every six months or, at least, once a year at all sites (locations) where elevated doses were detected, as well as at the phospho-gypsum landfill.

N o t e : It is reasonable to expect that a type of clean-up action (e.g. conservation, insulation, removal etc.) will be necessary at the phospho-gypsum landfill, but final decision can not be made before the activity (3) is performed. In the plant itself nothing more than some minor improvements in processing technology is needed.

Final Statement on INA-PETROKEMIJA Plant

All presented results, as well as findings of investigations which have not yet been done, are expected to give a reliable input for performance of detailed risk assessment study and cost-benefit analysis for possible remediation options. They are also the background for final decision on most convenient clean-up action(s). Most of these planned activities depend on circumstances at the sites of concern.

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TECHNOLOGIES FOR AND IMPLEMENTATION OF ENVIRONMENTAL RESTORATION IN THE URANIUM INDUSTRY IN CZECH REPUBLIC

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Abstract

This contribution is a logical continuation of the first and the second parts of the Regional Technical Cooperation Project on Environmental Restoration in Central and Eastern Europe. The first part was concentrated on identification and radiological characterization of contaminated sites (Budapest 1993); the second part, then, on planning for environmental restoration (Piešťany 1994);this third part has been directed to particular technologies for environmental restoration. Problems of uranium industry only has been dealt in this contribution.

As documented in the previous parts, protection of water is the fundamental problem in the field of environment protection in the uranium industry. This is the reason why we have concentrated our attention to main technologies which are used for decontamination of water in the uranium industry in the Czech Republic.

1. Types of water

By origin, water may be divided into two main groups:

1) Water connected with classic mining technology and uranium processing (mining and milling). This kind of water is divided to the mine water and to the free tailing water. The mine water has been cleaned by means of decontamination plants at entire mines of the uranium industry. We present here in this contribution the Central Decontamination Plant which belongs to the deposit called Hamr as a representative of decontamination plants because that is the biggest and the most complete plant. Combined technology of electrodialysis and evaporation at the processing plant in Dolní Rožínka has

been chosen as an example for decontamination of the tailing water. Technology at the processing plant MAPE in Mydlovary is an example for the classic decontamination of drainage water from a tailing.

2) Water connected with the underground leaching technology means a very complicated hydrogeological and hydrochemical system. As for composition, the water forms a continuous series beginning at concentrated technological solutions, continuing at dispersion solutions up to background water. We present a description of technology for decontamination of dispersion solutions at a neutralization station (NDS-6) and a technology being prepared for restoration of technological solutions in the following section of this contribution.

2. Mine Water And Tailing Water

At classic procedure to get uranium, that means at uranium ore mining and at uranium ore processing at the processing plant, two kinds of waste water are developed:
- the mine water, with a low TDS concentration
- the tailing water, with a high TDS concentration

2.1. Mine Water-Central Decontamination Station

The biggest and the most modern waste mine water treatment plant in the uranium industry is the Central Decontamination Station (CDS), state-owned company called DIAMO in Stráž pod Ralskem. The plant was put into operation in 1988. This plant processes mine water of volume approx. 400 $l.s^{-1}$.

In the course of technological operations realized by the CDS, first of all, insolubled substances (TSS), radium, uranium and heavy metals are removed.

A block technological diagram of the CDS is shown in Fig. 1. There are two parallel technological lines at the CDS, which differ in a practical way only in the fact that there is, in

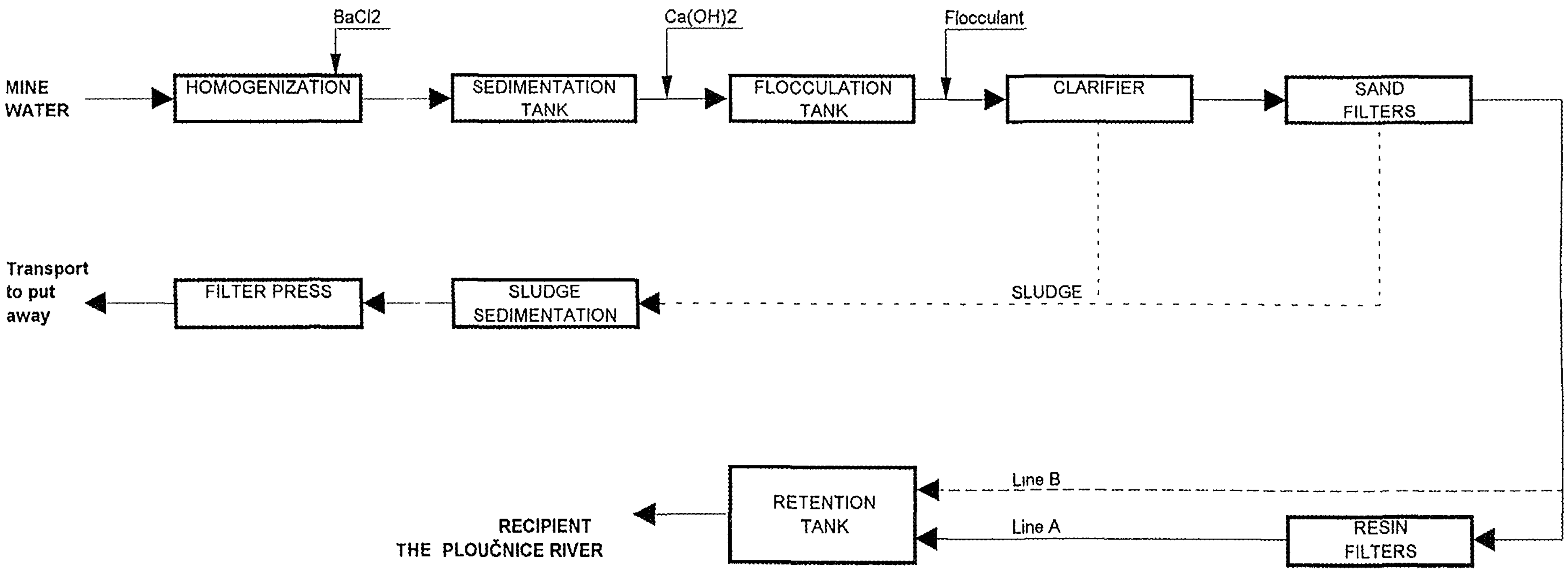

FIG. 1 BLOCK TECHNOLOGICAL DIAGRAM : CDS

TAB. 1.: COMPOSITION OF INPUT AND OUTPUT WATER AT THE 'CDS'

	INPUT	OUTPUT	PLOUČNICE RIVER
pH	6.5-7.5	7.6	7.3
TSS $mg.l^{-1}$	14-24	0.4	-
TDS $mg.l^{-1}$	500-1200	500-1200	234
NH_4 $mg.l^{-1}$	3-5	3-5	0.2
SO_4 $mg.l^{-1}$	300-500	300-50	57
Ni $mg.l^{-1}$	0.7-1.0	0.04	0.02
Zn $mg.l^{-1}$	0.5-1.0	0.02	0.06
U $mg.l^{-1}$	1.4-1.1	0.09	0.02
Ra $Bq.l^{-1}$	22.1	0.1	0.1
Q $l.s^{-1}$	400	400	810

addition to the equal equipment, a technological section to catch uranium by means of ion exchanging resin columns on the strong basic anion exchanging resin in case of one of these two technological lines. Both lines have the same capacity.

Waste water from different pits is mixed in a homogenization tank and the barium chloride solution is added there to it. From there, the water flows into sedimentation tanks. Rough TSSs are sedimented in these tanks.

At outputs from the sedimentation tanks, the calcium hydroxide suspension (or a lime suspension) and a ferric sulphate are dosed.

A precipitate which arises here is separated by means of a clarifier in the 1st degree and by means of sand filters in the 2nd degree. The water goes on then to the ion exchanging resin filters where uranium is caught. After leaving the ion exchanging resin filters, the water is joined to the water flowing out of the sand filters belonging to the second line.

2.2. Tailing Water

In the course of working of the processing plants in the Czechoslovakia Uranium Industry, the overbalance tailing water was treated at three processing plants in the following ways:

- **Processing plant called MAPE (near the town České Budějovice):** from 1962 to 1982 - after uranium, radium and manganese decontamination, the water was drained-off to the Vltava river; from 1982 till 1991 (finish of working) - a closed cycle with an accumulation of free tailing water; from the beginning of 1994 - decontaminated drainage water is drained off to the Vltava river (the decontamination technology is described below)

- **Processing plant called DIAMO in Dolní Rožínka:** from the beginning of working in 1968 - an accumulation of overbalance water;

  from 1974 up to now - evaporation of overbalance tailing water and production of a saleable product - sodium sulphate

- **Processing plant in Stráž pod Ralskem:** without growing volumes of tailing water. As proved later, free tailing water seeps through the coniac horizon and ground water is contaminated.

2.2.1. Liquidation of the Overbalance Water at the Processing Plant in Dolní Rožínka

An evaporation station is used for the liquidation of the overbalance at the water system of this processing plant. An eight-effect parallel-flow evaporator with forced circulation has been built, of capacity 30 - 50 $m^3.hour^{-1}$. The last two levels are connected in parallel to the same level and they work as continual classification crystallizers. The evaporator has been working since 1972. The overbalance tailing water presents the basic volume of the evaporator input.

During 1985-1986, within the frame of intensification, an electrodialysis station as a pre-concentration unit was built. The three-effect electrodialysis unit made by the firm Asahi Glass and the four-effect electrodialysis unit MEGA have been installed. A permeate from the electrodialysis unit is connected with a processed condensate from the evaporator and this product is drained off to the recipient. A concentrate from the electrodialysis unit is joined to the evaporator input.

The solid crystalline sodium sulphate is the evaporator output. This product is sold and it is used for washing powder production. A mother solution is used for preparation of an elution agent at the uranium processing plant.

Composition of the evaporator input is shown in Tab. 2. Block technological diagram is shown in Fig. 2.

2.2.2. Drainage Water Decontamination Station

When activity of the processing plant MAPE had been finished at the end of 1991, volume of free tailing water was growing considerably because of drainage tailing water pumping.

A technological project has been worked up for the drainage water decontamination and the construction of the decontamination station was finished in 1993. From June 1994 a trial operation has been running which is analysed at the present time.

TAB. 2.: COMPOSITION OF OVERBALANCE TAILING WATER

SO_4	$mg.l^{-1}$	15 000-20 000
Mo	$mg.l^{-1}$	0.1-0.5
Ca	$mg.l^{-1}$	40-50
Cu	$mg.l^{-1}$	0.1-0.5
Fe	$mg.l^{-1}$	0.1-0.5
NH_4^+	$mg.l^{-1}$	70-220
Cl	$mg.l^{-1}$	100-500
NO_3	$mg.l^{-1}$	600-900
CHSK	$mg.l^{-1}$	30-50
U	$mg.l^{-1}$	6-8
TSS	$mg.l^{-1}$	6-8
Ra	$Bq.l^{-1}$	0.4-1.0

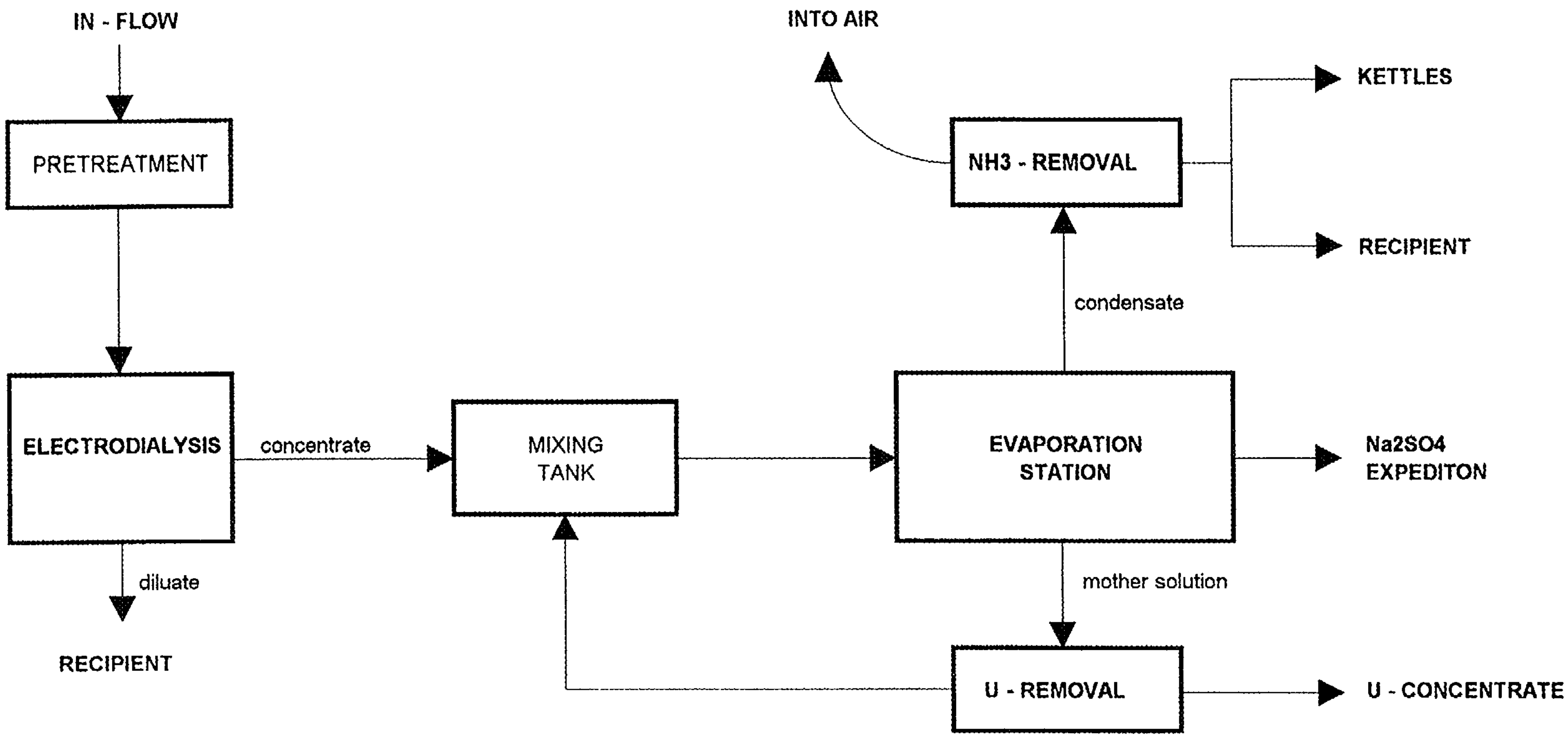

FIG. 2 BLOCK TECHNOLOGICAL DIAGRAM : EVAPORATION STATION FOR OVERBALANCE WATER OF THE TAILING IN THE PROCESSING PLANT Dolní Rožínka

A technological diagram of the decontamination station is shown in Fig. 3. Capacity of the station is 5 $l.s^{-1}$, values of input and output concentrations and composition of water in the Vltava river are shown in Tab. 3.

Water from the drainage system is pumped into an accumulation tank to ensure a regular pumping into the decontamination station. Volume of water which flows into the accumulation tank may be different since it depends on weather conditions - from 0 to 100 $m^3.hour^{-1}$. The accumulation tank is used for the homogenization of the pumped water as well. Nitrites are removed, the pH-factor is treated to the value 2 - 3 and the sodium sulphite solution is added. Barium chloride solution is added to the output from the reaction tank for the removal of nitrites. At dosing into the pipes, mixing inside the pipes is ensured by means of a stator blender. Suspension of lime is added into the water in the precipitation tank and, after the necessary delay in the second precipitation tank, solution of soda is added. The precipitate which arises is separated in a circle sedimentation tank. The precipitate is separated in full by means of sand filters then. The pH-factor is treated by means of the sulphuric acid. In case of higher contents of uranium in the input water, the water goes through ion exchanging resin filters with the strong basic anion exchanging resin in order to catch the uranium. After the decontamination, the water is gathered in a retention tank. The water is checked in order to comply with parameters for drained-off water and then the water is pumped through the nine-kilometer-long polypropylene pipe to the recipient.

3. Underground Leaching

3.1. Neutralization Decontamination Station NDS-6

In continuity to a resolution of problems connected with lowering of negative influence caused by technological solutions being used at the leaching, and with lowering of volumes of these solutions, the dispersion solutions have been decontaminated since 1987 at the neutralization decontamination station (NDS).

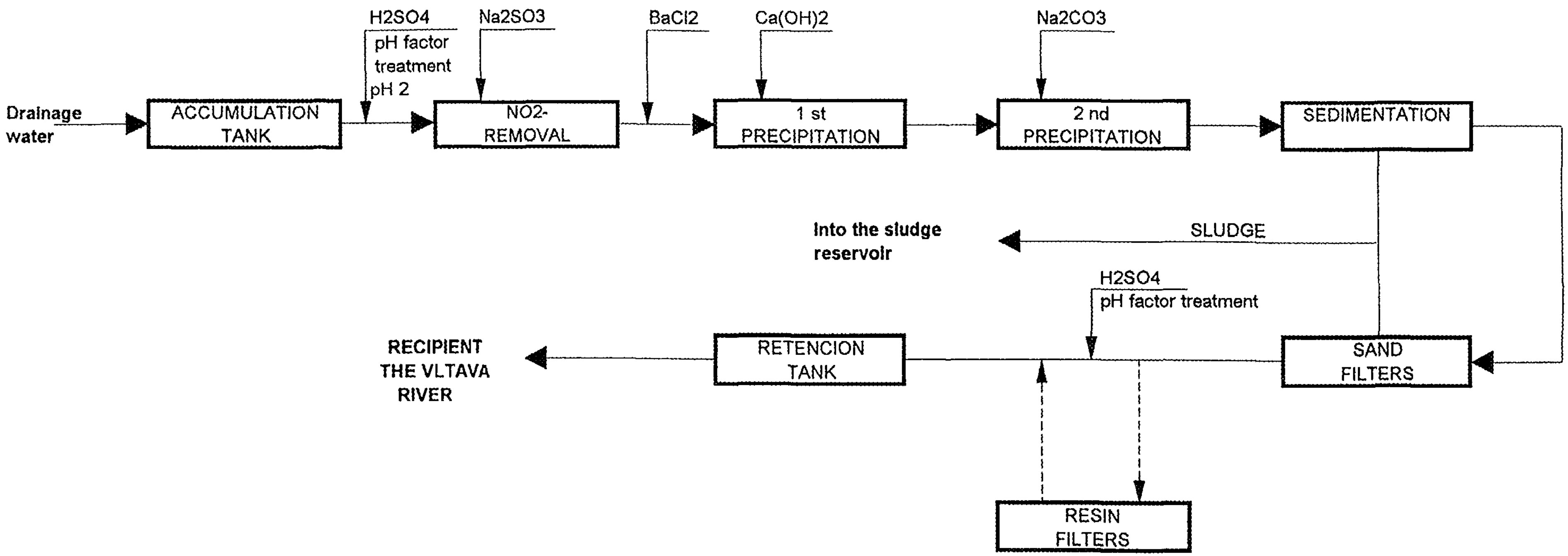

FIG.3 BLOCK TECHNOLOGICAL DIAGRAM : DRAINAGE TAILING WATER DECONTAMINATION - THE TAILING "MAPE"

TAB. 3:

	INPUT	OUTPUT	VLTAVA RIVER
pH	7.0-7.5	6.5-8	-
TDS $mg.l^{-1}$	6500	7600	282
NH_4 $mg.l^{-1}$	100	100	2.89
Fe $mg.l^{-1}$	40	0.4	-
Mn $mg.l^{-1}$	26	0.05	0.18
SO_4^{2-} $mg.l^{-1}$	3900	4000	67
NO_3^- $mg.l^{-1}$	320	320	6.6
NO_2^- $mg.l^{-1}$	50	< 0.1	-
Cl $mg.l^{-1}$	65	65	25
U $mg.l^{-1}$	0.6	0.02	0.0004
Ra $Bq.l^{-1}$	0.5	0.08	0.13
Q $l.s^{-1}$	5	5	10000

This station has been working since 1987 and 100 $l.s^{-1}$ of the dispersion solutions is processed on average.

Technological diagram of the NDS is shown in Fig. 4. Water is processed by means of two technological lines at the station. Both technological lines have the same capacity and the same technology. Values of input and output concentrations are shown in Tab. 4.

The dispersion solutions from underground, after catching of uranium on the ion exchanging resin station, go on to the NDS-6. At the NDS-6 station, suspension of lime is added in the precipitation reactor to get the pH-factor 7,5. Water with lime goes through blended reactors at delay approx. 30 minutes and it is led to the second precipitation - suspension of lime is added there to get the pH-factor of the solution at value of 12. The precipitate which arises is separated in the sedimentation tank. Strained-off substance is led, after the pH-factor is treated, to the chlorination. Ammonium ions are oxidized by the gaseous

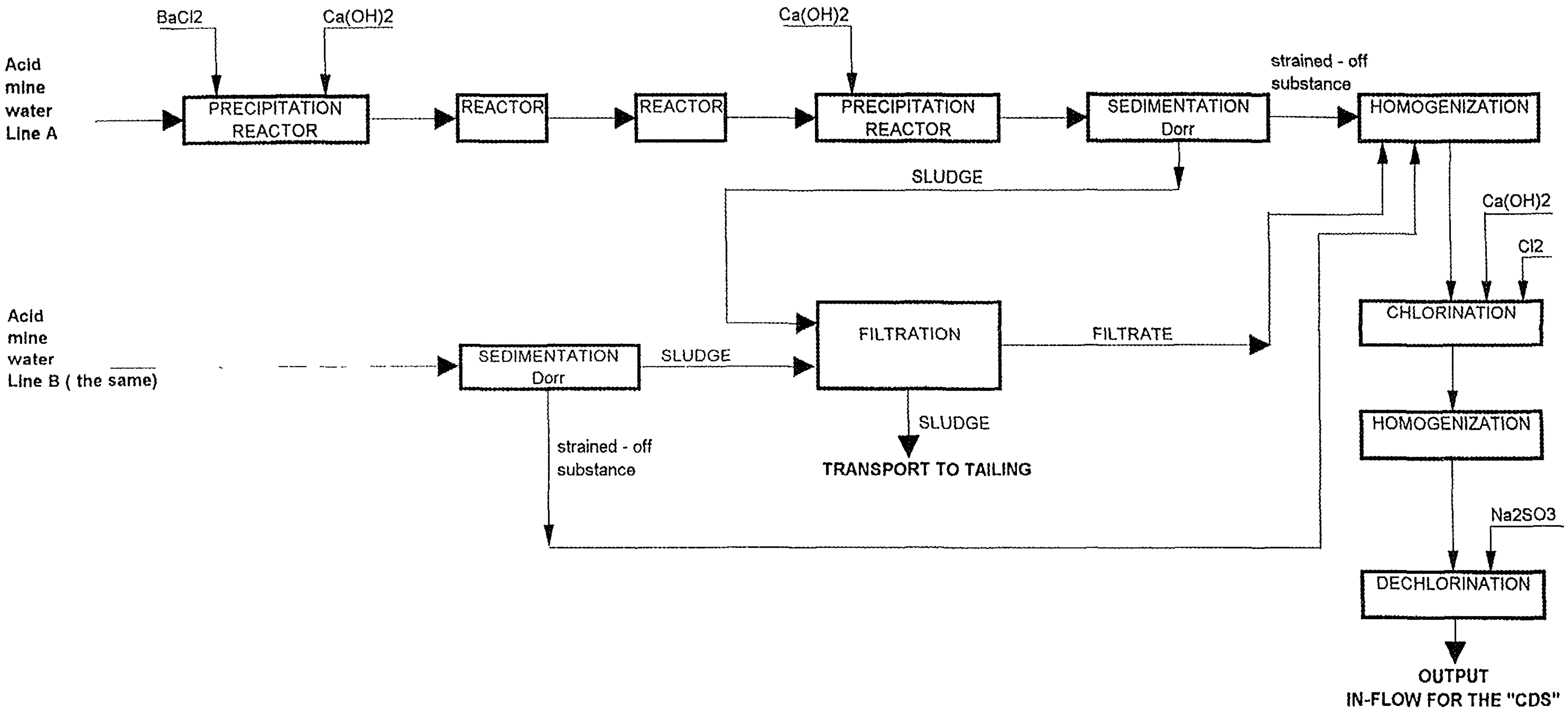

FIG.4 BLOCK TECHNOLOGICAL DIAGRAM : "NDS - 6"

TAB. 4.: COMPOSITION OF INPUT AND OUTPUT OF THE 'NDS'

	INPUT	OUTPUT	
pH	2.6	6.7	
TDS $mg.l^{-1}$	5100	3000	
Ca $mg.l^{-1}$	105	580	
NH_4 $mg.l^{-1}$	100	1.6	
Al $mg.l^{-1}$	420	0.6	
Fe $mg.l^{-1}$	130	0.4	
SO_4 $mg.l^{-1}$	3400	950	
Cl $mg.l^{-1}$	8	650	
Ni $mg.l^{-1}$	3.2	0.02	
Zn $mg.l^{-1}$	10.5	0.05	
U $mg.l^{-1}$	0.3	< 0.02	
Ra $Bq.l^{-1}$	38	1.5	

chlorine. Free chlorine after chlorination is removed by adding of the sodium sulphite solution. The solution decontaminated this way is led to the input of the CDS, then is is mixed with the input mine water and, after a finishing decontamination at the CDS it flows out to the recipient.

3.2. Evaporation Station in Stráž pod Ralskem

To liquidate the concentrated leaching solutions of the underground leaching process, an evaporation station has been built of capacity 92 $l.s^{-1}$($5,5$ $m^3.min^{-1}$). Construction was started on September 1994, a trial operation will be open during the 1st quarter 1996.

Composition of solutions which go through the evaporator is shown in Tab. 5. Guaranteed output values are shown in Tab. 6. Block technological diagram is shown in Fig. 5.

TAB. 5. COMPOSITION OF CONCENTRATED CENOMAN'S SOLUTIONS

Component	Unit	Average comp. of the sorp. solution [3]	Input solution on the evaporator [4]
RL (105 °C)	$g.m^{-3}$	64 000	68 000
RL (180 °C)	$g.m^{-3}$	-	55 000
Insoluble substances	$g.m^{-3}$	-	40
pH-factor	-	-	1.2
Conductivity	$mS.cm^{-1}$	63.7	64.7
Na	$g.m^{-3}$	14	15
Ca	$g.m^{-3}$	253	240
Mg	$g.m^{-3}$	41	56
K	$g.m^{-3}$	35	67
SiO_2	$g.m^{-3}$	150	160
Al	$g.m^{-3}$	5 030	6 000
Fe	$g.m^{-3}$	1 320	1 040
NH_4^+	$g.m^{-3}$	1 175	1 060
SO_4*	$g.m^{-3}$	49 000	48 000
SO_4*	$g.m^{-3}$		49 500
NO_3^-	$g.m^{-3}$	470	1 150
F	$g.m^{-3}$	240	300
Cl	$g.m^{-3}$	-	40
P (total)	$g.m^{-3}$	95	105
S (total)	$g.m^{-3}$	-	16 500
H_2SO_4	$g.m^{-3}$	20 000	14 800
As	$g.m^{-3}$	10.8	7.6
Ba	$g.m^{-3}$	< 0.1	0.028
B	$g.m^{-3}$	-	0.14
Cd	$g.m^{-3}$	0.36	0.33
Cr	$g.m^{-3}$	7.7	11
Cu	$g.m^{-3}$	4.7	2.5
Pb	$g.m^{-3}$	0.34	< 0.2
Mn	$g.m^{-3}$	12	12
Ni	$g.m^{-3}$	23	24
Se	$g.m^{-3}$	-	< 0.5
Ag	$g.m^{-3}$	-	< 0.02
Sr	$g.m^{-3}$	20	23
Zn	$g.m^{-3}$	50	50
Co	$g.m^{-3}$	-	7.1
V	$g.m^{-3}$	12	15
Be	$g.m^{-3}$	0.8	0.6
Ti	$g.m^{-3}$	2.5	2.1
Zr	$g.m^{-3}$	0.26	
Mo	$g.m^{-3}$	0.6	

TAB. 6: GUARANTEED VALUES

Parameter	Guarantee
Quality of the condensate	< 10 $mg.l^{-1}$
Quality of the sulphate	> 99.3 %
Quality of the waste product going into the air	< 500 $mg/Nm^3 NO_x$
Capacity of the plant in relation to the volume of evaporated water	> 330 m^3/year > 2 779 920 m^3/year
Capacity of the plant in relation to the production of ammonium--aluminium sulphate	33 000 kg/hour max. 277 990 t/year max.
Electric energy consumption	34.1 kWh/m^3 of the distillate (drained off) 30.4 kWh/m^3 of the distillate (from evaporation)
Steam consumption	8.5 t/hour
Chemical compound consumption	
NaOH 40 %	0.6 t/day
H_2SO_4 92 %	0.3 t/day
Quantity of concentrate from evaporation process	< 43 m^3/hour with production of salt < 74 m^3/hour without production of salt

In compliance with the given total conception for the liquidation of solutions at the underground leaching of uranium and on a basis of results arising at checking of individual technological steps, the project 'Liquidation of solutions of underground leaching - 1st stage', has been realized. The aim is - by means of the evaporation - to ensure:

a) at the first period of operation from 1996 - a volume underbalance of solutions at mining area of the 'Důl chemické těžby' (Mine of chemical processing), state-owned company, DIAMO;

b) after realization of the 2nd stage of the project - a complex procedure for liquidation of solutions of the underground leaching.

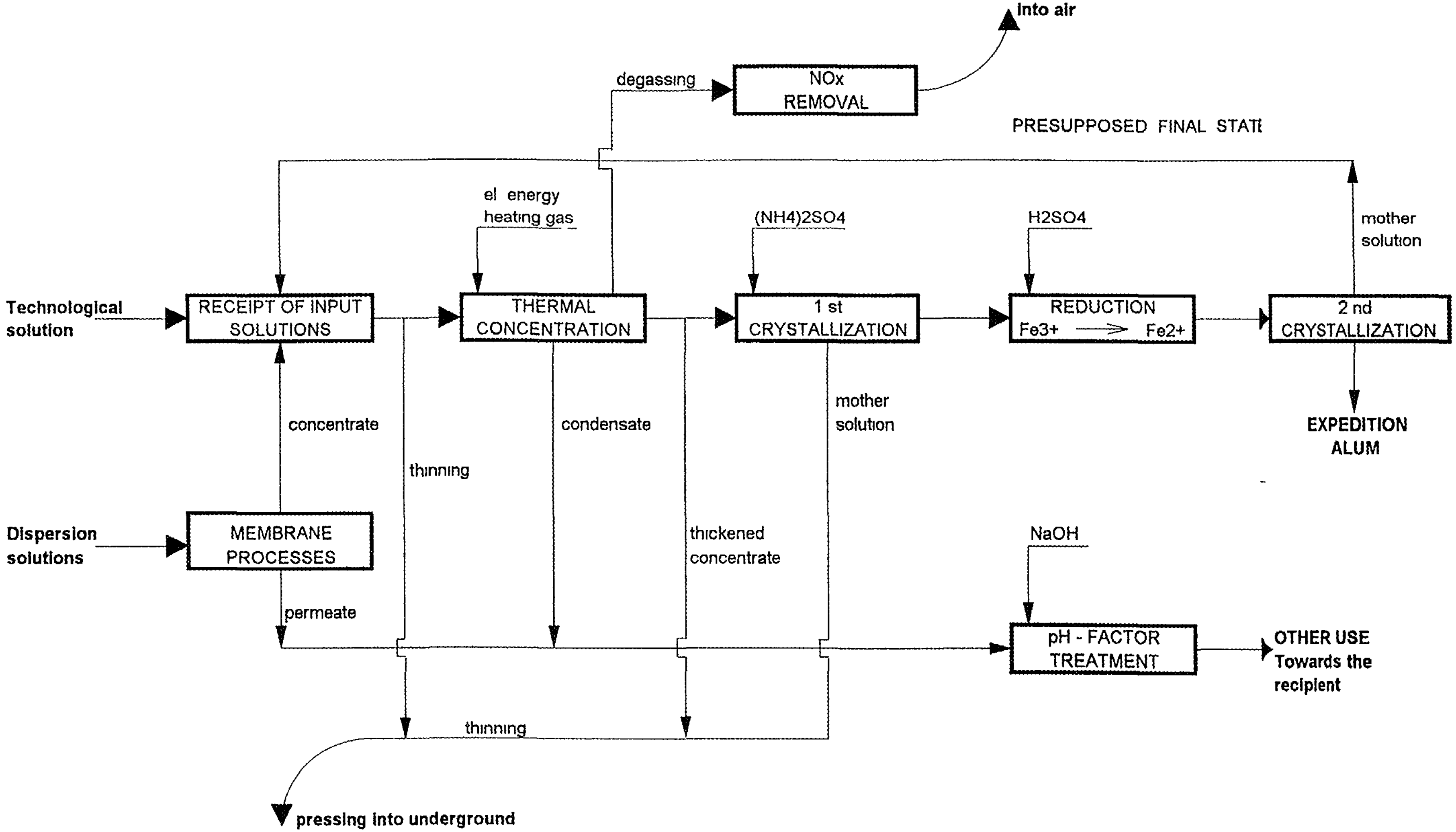

FIG. 5 **BLOCK TECHNOLOGICAL DIAGRAM : EVAPORATION STATION FOR LEACHING SOLUTIONS -" Stráž pod Ralskem "**

The first stage includes all technological plant to ensure the following technological steps:

* Concentrated solutions of the underground leaching of uranium are pumped from the place of rise. They are concentrated in a membrane process unit and the concentrate is gathered in reception tanks. Joined concentrates are pumped to a thermal concentration unit.

* The solution is concentrated in the thermal concentration unit so that conditions for crystallization of a solid portion from the thickened concentrate after cooling are created.
  The distillate is treated (neutralized) to the values which enable to drain it off to the recipient. The concentrate goes on to the crystallization of salts.

* After cooling, a crystalline portion is separated from the concentrated solution, which consists mainly of ammonium - aluminium sulphate. In case of need, an ammonium anion in the form of ammonium sulphate is added in the place of the crystallization. Mother solution from the crystallization is pumped into expedition tanks. The separated crystalline portion is led to a re-crystallization.

* The crystalline portion is dissolved and, after cooling, it is separated again and goes through a washing process. Mother solutions are pumped into expedition tanks, or, if need be, they are recycled to the input of the thermal concentration unit. Recrystalled ammonium-aluminium sulphate is dewatered and taken away of the system for another use.

* The solution which consists of waste solutions after the crystallization, recrystallization an, if need be, after the washing of crystals, is gathered in expedition tanks and, after thinning by the origin solution is pumped to the place of consumption.

The technological plant enables - in compliance with the two-level conception for the realization of technology for the liquidation of solutions - an operation in two basic regimes:

Regime I : includes the technological procedure explained above in full extent

Regime II : The crystallization unit and the recrystallization unit are not in operation in this regime. The concentrate from the thermal concentration unit goes on to the expedition unit directly. This way, the plant will be used till the time of realization of the technical plants belonging to the 2nd stage of the project.

Both regimes may be operated either with use of a membrane process unit or without this unit. Interconnection among all these three operational sets enables an operation in four variants:

1. Regime I including the membrane processes (the complex operation)
2. Regime I without the membrane processes
3. Regime II including the membrane processes
4. Regime II without the membrane processes (minimum)

Authors of this contribution have been the main designers of the decontamination technology for the central decontamination station, for the neutralization decontamination station and for the drainage water decontamination station MAPE.

REMEDIATION OF ECARPIERE URANIUM TAILING POND BY COGEMA (FRANCE)

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Abstract

Division of Vendée has been operated by COGEMA from 1954 to 1991. The main site is named ECARPIERE where underground and open pit mining fed a mill and heap leaching facilities which produced all together 15000 t of Uranium.
The frame of the methodology was presented during the second workshop in Piestany [1]. Specific informations and details concerning inventory of ECARPIERE, the materials used on this site, results of the studies are given.
Main points for implementation are mill dismantling, resloping of the dykes, covering of the impoundment and water management. Every step needs a careful radiological and topographical follow up.
Post remediation monitoring is adapted from the initial network to the new situation of the site.

I - HISTORICAL BACKGROUND

ECARPIERE, located at Gétigné (Loire Atlantique - France) on the border of river Moine has been one of the three main uranium extraction site of COGEMA's mining division of Vendée.

Prospection started in 1950 allowing underground mining to begin in 1953 : maximum extension was 3 kilometers long and 500m deep producing 3600 tU.
Three open pit (maximum depth 50m - total production 475 tU) mark out the upper part of the mineralised structure.

Ore treatment developed in two stages :

- from 1957 to 1991 a mill (acid pulp leaching - maximum capacity 450000 t/year) treated 9000 kt containing 14000 tU,
- in 1967 heap leaching pads have been built for poor ore (grade <600ppm - 4000 kt) producing 6 millions cubic meter of uranium bearing solutions containing more than 1200 tU,
- total output has been 14761 tU.

Remediation of the mill facilities is the chalenge for this site. The frame for the operations described hereafter is set in the updated operating license issued on May 16th 1983 :

- drying of the pond,
- covering and reshaping of the site,
- seeding,
- monitoring,

The general principles and the main objectives have been listed in the previous presentation [1].
Remediation started in 1992 and should be completed next year.

II - CHARACTERISTICS OF THE SITE AND THE PRODUCTS

II.1 - The site

Total area is 240 ha divided in units (see figure 1) which characteristics are different and need specific management.

II.1.1 - Open pits

Remediation of these areas - partial backfilling, mining debries damps recountering, revegetation - does not leave any material left on site.

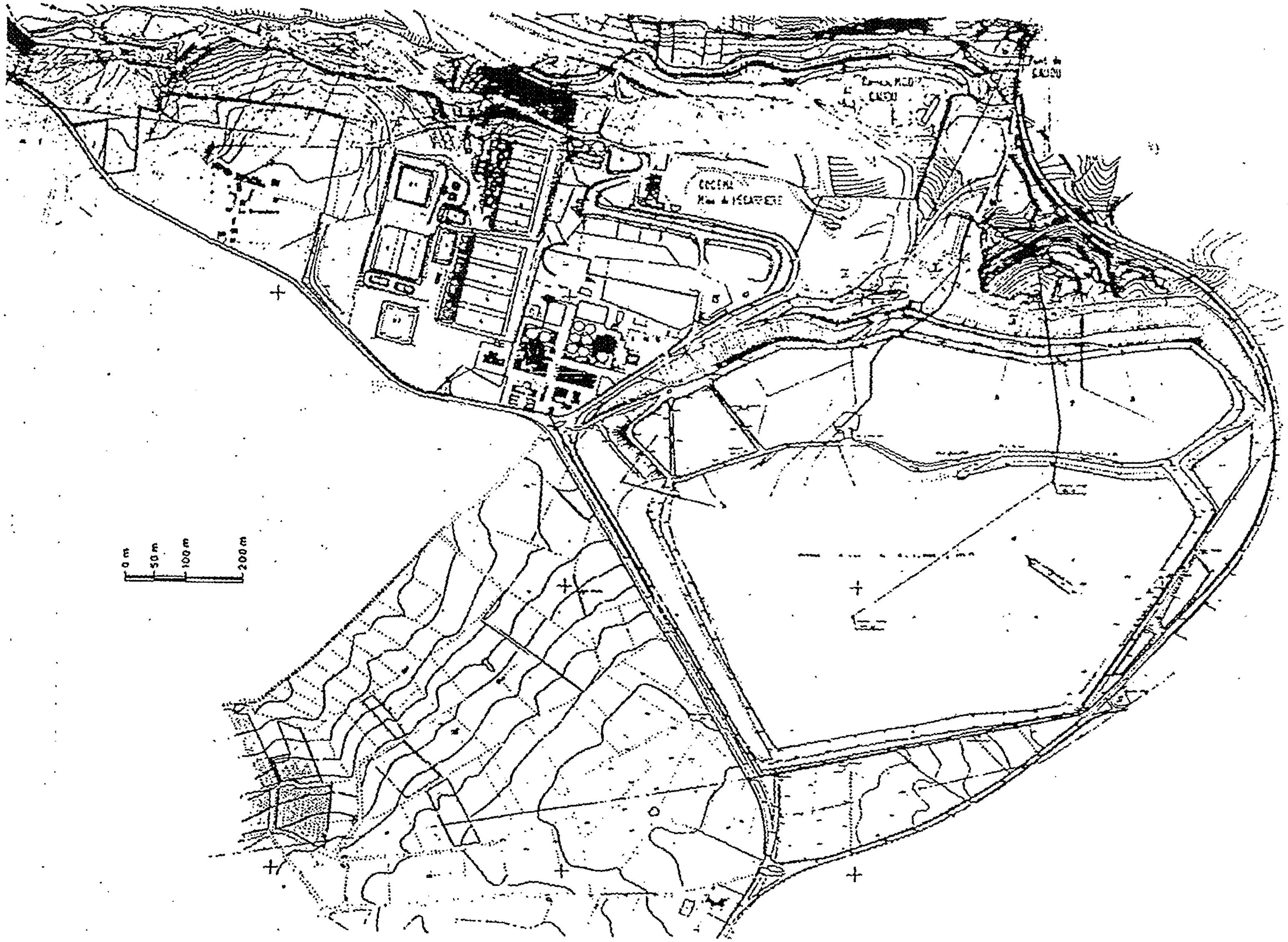

FIGURE 1

Table I : Main subdivision of the site of ECARPIERE

LOCATION	AREA (ha)	TONNAGE(Activity) kt (TBq Ra 226)
Open pit mining	115	
Mill	6	
Heap leaching facilities	16	
Undergr. Mining installations	12	
Heap leaching waste damp	9	4000 (16)
Mill tailings impoundent	73	7575 (170)
Waste water collecting zone	9	

II.1.2 - Underground mining surface facilities (mine heads)

In this area the ore storage damp and heap leaching wastes used for various purposes give residual surface radioactivity between 1000 and 5000 c/s SPP2 - far above the average natural background (600 c/s)

II.1.3 - Mill

For treatment, ore was going through crushing and grinding to 500 µm, in pulp hot and acid attack, solid liquid separation in raked classifier, ion exchange resins to purify pregnant solutions, final precipitation of yellow cake ("diuranate d'ammonium") and waste water treatment.
The equipment and building materials are partly contaminated. Highest contaminations are located in the resins and the scale (tartres) lining some tanks.

II.1.4 - Heap leaching facilities

The initial facility had been built in 1967. The final total designed capacity was 105000 tons of ore including 13 leaching areas, 24 collecting reservoirs, 3 pumping stations and an acid solution preparation facility.
Most of it had been built with leached ore and the whole has to be scraped.

II.1.5 - Impoundments

Two main impoundments have to be considered :

- after flushing of U pregnant solutions, wastes from heap leaching have been piled on the side of the mine-head with bottom collection of acid water soaking out of the damp.

- the mill tailings impoundment is a large settling pond resulting from linkage of four initial ponds (1957-1982) and a big southern extension built in 1983. The wastes used to be pumped (pulp density $1.23 t.m^{-3}$ - 25% solid) and cycloned, the coarse fraction (42 % >150 micron) beeing used to build the dykes (a part of the sands was used for mine hydraulic backfilling).

Table II : Main characteristics of the tailings pond

Material used		sand > 150µm
construction		vertical + upward
basement		granitic arena
Height		15 - 50 m
crest	length	3000 m
	width	10 m
slope		30-45 % (16-25°)
area		73 ha

Monitoring equipment for the dykes include :

- piezometric boxes (in the lower parts of the dyke)
- and three shafts (lower parts),
- piezometer drills in the upper part.

II.2 - The materials

II.2.1 - Wastes from leaching of uranium ore

Chemical and mineralogical characterisitics are typical of granites and do not show any risk for acid generation :

Table III : Chemical and mineralogical characteristics of the wastes

SiO2	60 - 90 %
Fe 2O3	2 to 10 %
A12O3	2 to 20 %
F, P2O5, CaO, MgO, K2O, Na2O, TiO2	
U (mill tailings/heap leach.wastes)	70/70 ppm (<2Bq/g)
Ra (mill tailings/heap leach.wastes)	22/4 Bq/g

Two types of leached wastes have been produced :

- heap leaching residues which are medium size (60-150 mm) as they were crushed (in addition similar mixed products come from dismantling of miscellaneous facilities),
- mill tailings which have been partly cycloned (to build dykes behind which the overflow settles and to produce sand for mine hydraulic backfilling)

The main geotechnical characteristics have been listed in the last presentation [1].

II.2.2 - Altered gabbros

As all barren materials had been used for remediation of the open pits, we considered discarded altered gabbros coming from the overburden of a nearby gabbros quarry.
This material gave good results to compaction as regard to permeability ([1] and § table IV).

II.2.3 - Material from dismantling of the facilities

A tonnage and Ra226 evaluation of the contaminated and non contaminated equipment and materials coming from dismantling of the mill give the following figures (table V):

Table IV : Physical characteristics of mill tailings and altered gabbros used for ECARPIERE

	MILL TAILINGS	
	ECARPIERE	Possible range
Grain size % < 500μ	100	100
% < 80μ	80	40 - 100
Water content w%	35 - 130	25 - 130
Dry specific weight $T.m^{-3}$	0.6 - 1.2	0.6 - 1.3
Consolidation Cv $cm^2.s^{-1}$.	$6.5 \cdot 10^{-3}$	10^{-5} - 10^{-2}
Cohesion Cu $T.m^{-2}$	0.5 - 8	
Permeability K $m.s^{-1}$	10^{-8}	$< 10^{-7}$

	Altered Gabbro	Heap leached ore
Grain size max. mm	pit run overburden	60 - 150
< 80μ %		6 - 12
Water content w %	5 - 16	5 - 8
Dry specific weight $kN.m^{-3}$		
non compacted	13.9	14.3
after compaction	21.9	21.7
Permeability K $m.s^{-1}$		
non compacted	1.2 10^{-4}	8.4 10^{-5}
after compaction	3.0 10^{-7}	2.5 10^{-8}

Table V: Evaluation of materials from dismantling

TYPE OF MATERIAL	TONNAGE (t)	ACTIVITY (GBq Ra 226)
Contaminated scraps	1900	667
Non contaminated scraps	1100	0
Resins and charcoal	270	116
Concrete	1750	22
TOTAL	5020	# 0.8 TBq Ra226

Separate evaluation of the activity coming from the mill give less than 1TBq Ra226. That is less than 1% of the total activity stored in the impoundment (170 TBq) and was already included in the activity of the impoundment. Such a small quantity of fixed radioactivity is allowed to be disposed of with the tailings.

III - STUDIES

Studies include better knowledge of the residues we have to deal with, of the materials which are going to be used for the required cover and of the environment of the impoundment.

III.1 - Petrography of the wastes and leaching tests.

A complete study of the tailings recovered by drilling through the complete pile down to the underlying granite gave valuable informations :
- deepest samples show an increasing proportion of argilous minerals (smectite) which is proof of a real diagenesis comparable to any natural rock evolution [2],
- of course gypsum linked to neutralisation is observed,
- radioactivity is linked to smectite and gypsum,
- 60% of the radium is located in the fine fraction (<30µm) and is not moved by lab leaching tests,
- no radium migration is observed in the overlaid granite.

One can conclude that radium is fixed in the pile and that natural evolution leads to an even better chemical containment.

III.2 - Hydrogeology

Water balance of the impoundment, of the underground mine (feeding by deep granitic circulation) and its environment show that the impoundment is watertight.
Moreover,
- altered surface granite give very low permeability measures : 10^{-10} m.s^{-1},
- this layer has been kept at the bottom of the impoundment and drilling show it is now compacted by weight of the overburden,
- bottom residues are not yet consolidated and piézometric level is ten meters above the original topography.

III.3 - Settlement

- Lab measurements (§ II.2.1 - Table IV) show variation with depth of the density and cohesion. The first three meters are consolidated with cohesion over 2 t.m^{-2}.
- The natural settlement of the tailings under their own weight is not finished. Calculations conclude that in 25 to 30 years time, total reduction of the height will reach 5 to 12% meaning up to 5 meters for the thickest part of the impoundment.
- The final settlement is taken into account for determination of the thickness of the cover.

III.4 - Test plots : compaction and the final cover

Aim of the plots was to test the efficiency of the cover as regard to :

- decrease of the radiological impact,
- increase of permeability in order to limit the seepage of rain water and radon diffusion,
- reduction of gullying and prevention of intrusion.

The plots were 50 meters long and 10 meters large built on a naturally dried part of the impoundment :

- a first metric layer of compacted heap leaching wastes,
- a second cover of different thickness of compacted and non compacted gabbros.

Radioactivity and radon flux measurements give the results in Table VI.

Compaction measurements (Table IV) show a considerable decrease in permeability and more than 50% increase in density. Radioactivity and radon flux are reduced to values comparable to the background.
For the typical cover (figure 2), derived evaluation of the annual rate of exposure (ATAER) give 0.12 (on site) which is much less the prescribed limit for the population in the environment.

Table VI : Measurements on the test plot

RADIOLOGICAL IMPACT	UNCOVERED TAILINGS	COVER*	BACKGROUND
Gamma SPP2 c/s **	910	135	100 - 200
Radon flux (10^5 at.$m^{-2}.s^{-1}$)	40	0.6	0.6
Running water	(heap leaching waste)		
Ra Bq.l^{-1}	0.30	0.02	
U mg.l^{-1}	< 0.10	<0.10	

* 1 meter heap leaching waste + 0.3 m compacted gabbro
** focussed measures

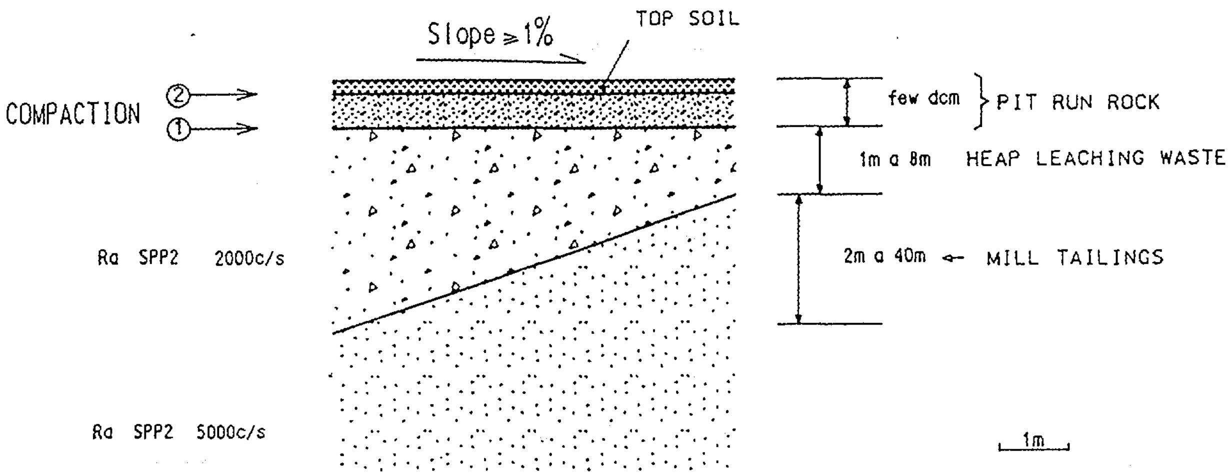

FIG. 2. Typical cover (Ecarpiere).

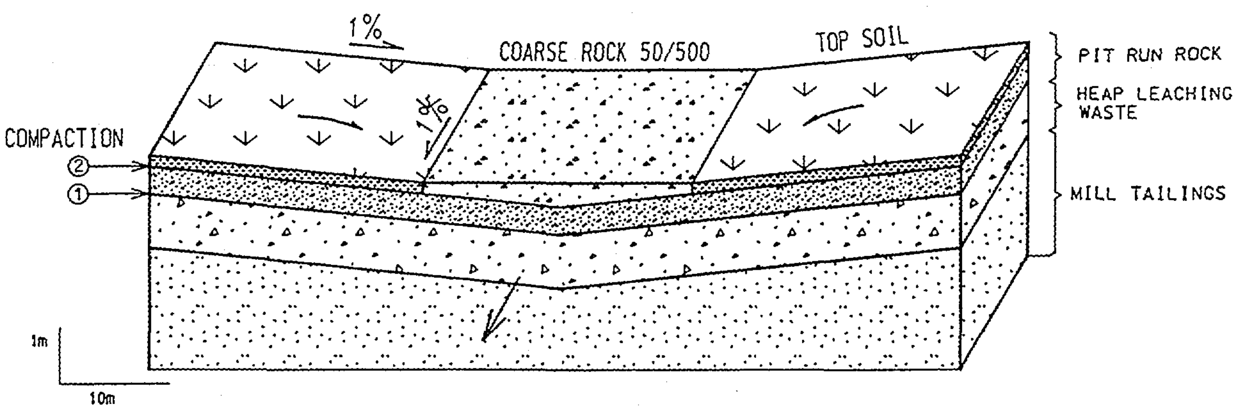

FIG. 3. Drainage channel (Ecarpiere).

III.5 - Stability

During operation behavior of the dykes was always good with stability coefficient of 2.
After resloping stabilty calculations show that under normal conditions the security coefficient is 2.6.

- in case water would rise to the maximum possible level, it is still 2.3,
- the historical seism, with a horizontal acceleration of 0.15xg reduce the coefficient to 2.4 : all values beeing much higher than the usual 1.5 value for such dykes.

IV - OPERATIONAL OBJECTIVES FOR THE REMEDIATION

The basic radiological objective is a total added exposure of 5 mSv [1].

For Ecarpière, standard gamma measurements give 350-450 c/s SPP2 for the defined cover.
According to surface contamination equipments can be reused : average alpha and beta surface contamination limits used are 1.85 $Bq.m^{-2}$. Maximum value is 18.5 $Bq.m^{-2}$. Contact gamma measurement with a scintillometer should be limited to 500 c/s SPP2.

Effluent quality is fixed by the license (Ra226 0.37 $Bq.l^{-1}$, U 1.8 $mg.l^{-1}$) but could move up to 0.74 Bq $Ra226.l^{-1}$ according to the general regulation. Final pH must be between 6.5 and 9.5.

Required compaction is obtained after six applications of a V3 compactor.

The stability was calculated for slopes less than 35%

V - IMPLEMENTATION OF THE REMEDIATION

V.1 - Dismantling of the mill

There are three main categories of dismantling products :

- part of the equipment found reuse on other sites of the company,
- some equipments could be sold for public reuse according surface contamination limits (§ IV),
- as mentionned before, over limits dismantlement products (scraps and concrete) were allowed to be disposed of with the tailings. Location was choosen where the planned cover was the thickest.

Two areas of one hectare were delineated and prepared with a first layer of heap leaching waste. The products were trucked up to the storage area.
Big pieces have been cut, some sheets have been pressed, the whole has been laid in order and voids filled with sand in order to minimize settlement,
Final cover is part of the engineered cap of the tailings impoundment.

Description and radioactivity of each equipment beeing sold or each load of dismantlement product stored with the tailings has been carefully registered.

V.2 - Management of water on the site

The tailing pond was the central point for all the waste waters on site and the first operation is "soaking" of the residues :

- the northern pond was used as storage for the water pomped out of the mine : it started drying in late 1990.

- the southern part, settling pond for the mill tailings till 1991, was used to store water before feeding the waste water treatment plant till 1993 : since, water is beeing pumped. The final situation is a natural outlet for the whole remediated impoundment built on the bedrock for long term stability.

- a double set of ditches around the whole pond collects separetely the seepage water and surface running water either running on the surface of the site and collected by the draining trails (figure 3) toward the outlet or running from outside the site. Seepage water are directed to the treatment plant whereas the surface running water might only need some decantation for suspended matter.

V.3 - Resloping of the dykes

The main dyke was covered by ten years old pine trees and acacias. It was decided to cut the whole in order to reslope the dykes and achieve long term stability.
Reasons for resloping of the dykes are :

- achieve long term stability,
- reduce surface gullying (erosion),
- make easier implementation of the cover (specially compaction),
- improve integration in the environment.

Final slopes have been determined according to stability studies. In order to reduce transportation costs two types of slopes have been determined :

- slopes dipping less than 35% are beeing dug down to 20% (figure 3 - removed sand is put back inside the impoundment),
- above 35-40% the higher part is dug down to 20% and, in the lower part, benches are anchored (figure 4)
The benches are made of compacted heap leaching wastes covered by barren rock as the typical cover (figure 2).

V.4 - Cover and final topography

The final cover determined earlier (figure 2) is a combination of heap leaching wastes compacted before spreading a layer of compacted gabbro which is finally covered by top soil or altered gabbro.

Different materials are first gathered on top of the impoundment :

- heap leaching wastes,
- sand coming from resloping of the dykes,
- all contaminated materials gathered from dismantling of the heap leaching facilities, remediation of miscellaneous areas and contaminated equipment, scraps and concrete resulting from dismantling of the mill.

According to the surface cohesion a first metric layer of heap leaching wastes is carefully spread on the tailings. On the top part of the impoundment thickness of heap leaching wastes depends on the compaction and the final topography : the resulting slope is minimum 1% to prevent water retention and maximum 20%.

Important structures are the draining trails dipping inward. They divide the area in subbasins and collect the surface running water toward the draining channels (figure 3) and the final outlet.

The final situation is mapped on figure 6.

V.5 - Water treatment plant

The main type of water can be distinguished on the site :

Table VII : Characteristics of waters on the site.

ORIGIN	FLOW RATE $m^3.h^{-1}$	pH	Ra226 $Bq.l^{-1}$	Uranium $mg.l^{-1}$
Mine water	50	5.4	2	0.8
Seepage water	18	6.2	1.5	0.5
Surface running water	0 - 60	6.2	0.02 - 0.4	< 0.1
River MOINE	9000	7.0	0.03	<0.1

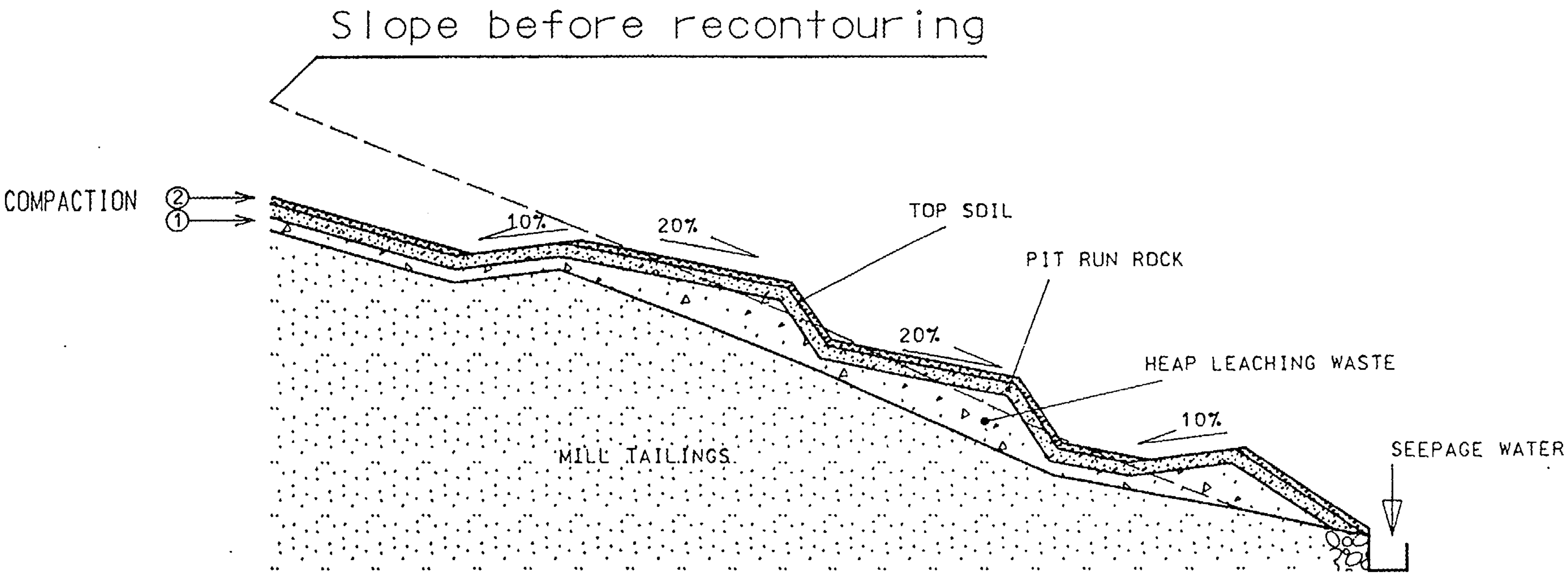

FIG. 4. Cover for slopes dipping > 40 % (Ecarpiere).

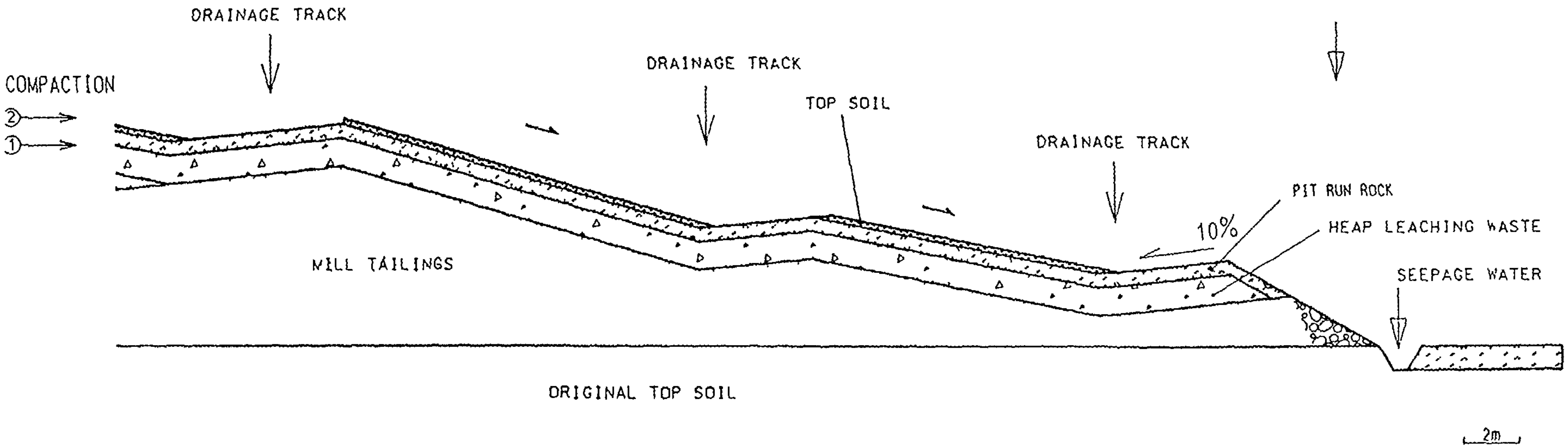

FIG. 5. Cover for slopes dipping 20 % (Ecarpiere)

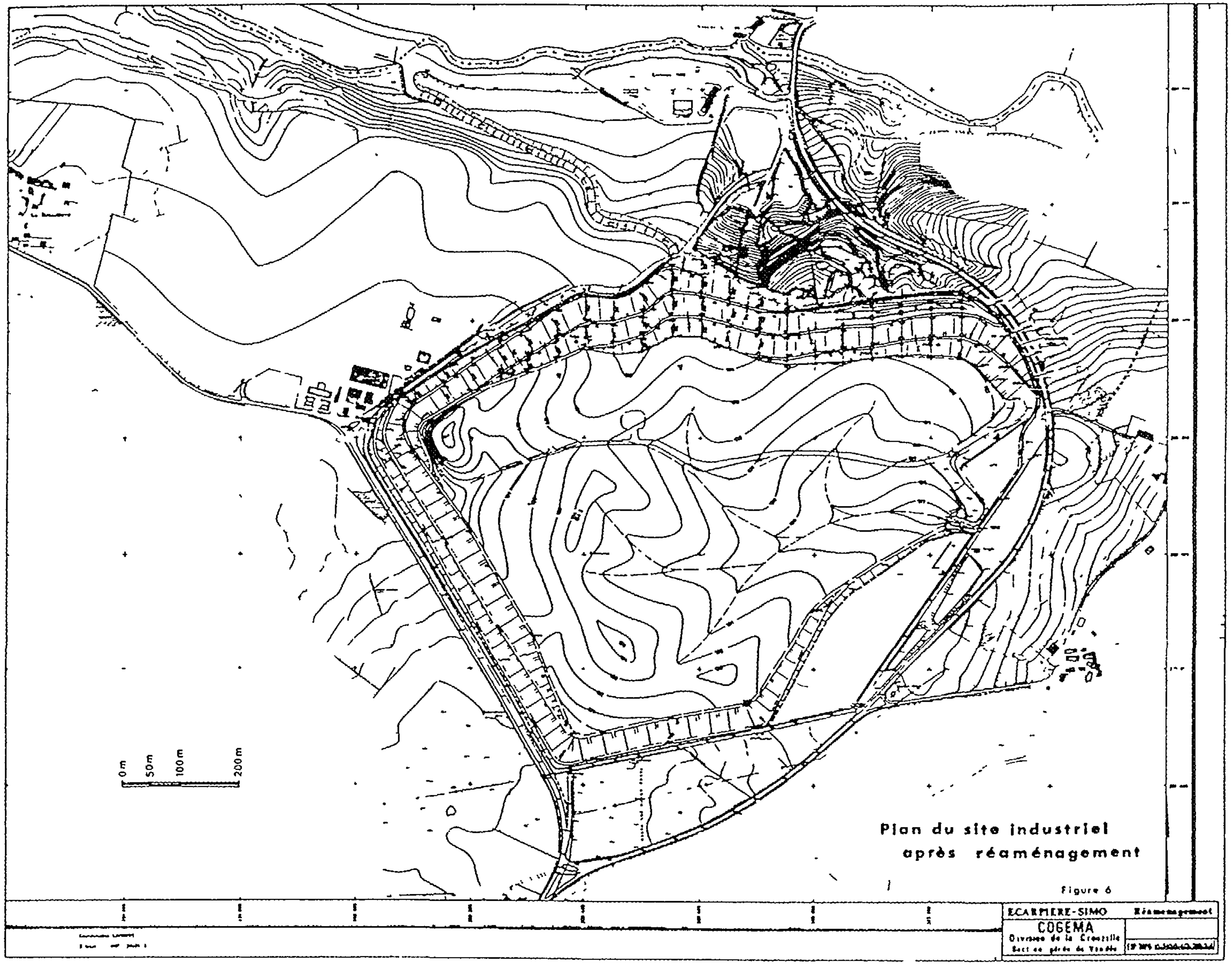
0m
50m
100m
200m
Plan du site industriel
après réaménagement
Figure 6
COGEMA

FIGURE 6

Direct discharge in the river Moine is not possible according to the limits included in the license.
Waste waters are collected to adapt pH (objective 6.5 to 9.5) and reduce Ra226 (objective <0.37 to 0.74 $Bq.l^{-1}$) and U238 (objective <1.8 $mg.l^{-1}$) content.
The water treatment plant is still located near the former mill: a new one may be built on the lower part of the site to allow gravity collection of the water.

V.6 - Quality control

Radiological control are mainly external radiation gamma control and may include a few radon flux measurements. The most important are gamma measurements of the materials used for the cover : this control must be done before beginning transportation. All along implementation of the final covering radiometric control can be done. At the end of the job a final systematic measurement is done on a 20 meters grid.
Radioactivity of each equipment leaving the site or transported to the tailings pile is carefully registred.

Topography : thickness of the different layers and position of the main land marks are pegged out on site and regular survey allow to check conformity with the project.

Compaction measurements confirm that permeability are comparable to those on the test plots.

Table VIII : Compaction measurements after remediation.

	Heap leached ore (test plot)	Gabbro (test plot)	Gabbro (remediated dyke)
Dry specific weight $kN.m^{-3}$	20.0	20.6	19.5
Permeability $m.s^{-1}$	$3.5\ 10^{-8}$	$0.5\ 10^{-7}$	$1.8\ 10^{-8}$

VI - POST REMEDIATION

VI.1 - Geotechnical monitoring

During remediation all the equipments necessary to assess the water level inside the residues and specially in the dykes is preserved. Measurements go on after remediation in order to make sure of the normal evolution of the settlement and global stability.

VI.2 - Radiological monitoring

On site measurements are made to assess the evolution of the quality of running water, seepage water from the tailings pile, underground and open pit waters as well as air quality.

Around the site a network of stations give the measurements necessary to :
- give an evaluation of the exposure due to natural environment (background - this is necassary in the case of Ecarpière as no on site evaluation has been made before beginning of operations),
- give an evaluation of the added exposure due to the past industrial activity and the remediated storage,
- evaluate the impact of the site on the critical population for which a scenario is applied.

The network used during operation and remediation is usually kept for a while after the end of remediation. According to the results, the license may allow later a reduction in the number of sampling site.

The network in the environment of ECARPIERE comprises now :
- 13 alpha integrated dosimeters (ALGADE devices filtering air during a month to measure radon and long live alpha emitters in dust associated to a DTL for gamma radiation),
- water sampling station upstream and downstream as well as on the final effluent,
- well waters are also analysed.

Every six months to two years sampling of the food chain is implemented : sediments in the river, soils in the fields or gardens and the associated plants (weeds, grass and vegetables) as well as milk, wine and fishes.

The main contribution to the added exposure is linked to air and water pathways which measures are given in table IX. In this case the exposure and ATAER (Added Total Exposure Rate) of the critical group is evaluated with the following parameters :

- annual residence time : 7000 hours
- standard breathing rate : 0.8 m3.h-1
- daily amount of ingested water : 2.2 liters of the downstream water. This figure includes water ingested through food consumption.

Table IX : Impact on the environment - ECARPIERE 1993

STATION	AIR PATHWAY				WATER PATHWAY		TAET	TAETA
	EXT.EXP	INTERNAL EXPOSURE						
	Gamma ray nGy.h-1	Rn220 nJ.m-3	Rn222 nJ.m-3	Dust Bq.m-3	Ra226 Bq.l-1	Uranium mg.l-1		
HAUTEGENTE	200	18	51	1	0,11	0,1	0,53	0,12
AVERAGE 13 st.	211	15	45	1	0,13	0,1	0,53	0,12
BEL AIR (Background)	150	13	35	1	0,12	0,1	0,41	

Hautegente is the nearest station to the site. According to evaluation of the exposure due to the background by measures at Bel Air, the ATAER is 0.12, equivalent to the average of the 13 stations.
The impact on the environment is definitely very limited.

Analyses of the food are used to calculate the possible daily consumption necessary to reach the annual limit of ingestion for U, Ra and Pb210 (for Ra226 or Pb210 daily consumption of 100 liters of milk is necessary to reach the annual limit of ingestion).

VII - CONCLUSION

Petrographic and leaching tests show that radium is confined in the tailings pile. Moreover the site is isolated from the environment.
This allows on site remediation which is mainly recontouring followed by covering with local materials to protect from erosion and infiltration. Seepage water are collected for water treatment.

Total radiological impact has always been within the prescribed limits.

Uranium mining and milling site remediation is expensive and time consuming and has to be carefully planned.
The experience gained by COGEMA's Uranium Division joined to soil decontamination and engineering can be shared worldwide through specialised subsiduaries SGN and DSR.

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REHABILITATION TECHNOLOGIES TO BE USED IN THE DECOMMISSIONING OF URANIUM MINING SITES IN THE FEDERAL REPUBLIC OF GERMANY

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Abstract

Rehabilitation technologies to be used in the decommissioning of uranium mining sites in the Federal Republic of Germany

The world's largest uranium mining operations in Saxony and Thuringia produced a total of 220 000 t of uranium. In 1990 production came to an end and decomissioning began. The clean-up effort focusses on the sources of contamination: waste piles covering a total area of 1517 ha, tailings ponds of 720 ha, 9 underground mines, one large open pit mine, as well as plant areas, structures and facilities.

In 1990/91 emphasis was on activities aimed at reducing population exposure and on development and planning of rehabilitation work.

The concept chosen by Wismut GmbH for final rehabilitation comprises the following elements:

- removal of hazardous materials from and partial backfilling of mine workings,
- mine flooding,
- in situ rehabilitation of tailings ponds,
- treatment of mine waters and tailing waters,
- in situ rehabilitation or relocation of waste rock piles
- dismantling and demolition of contaminated structures and facilities, rehabilitation of plant areas,
- development of an environmental monitoring system

Total costs are estimated at 13 thousand million German marks to be funded by the federal budget.

Introduction

The world's largest uranium mining operations were conducted in Saxony and Thuringia from 1946 to 1990. In the course of the 44 year mining period, a total of 220 000 t of uranium was produced, which represents some 13 % of wordwide production after WW II.
When uranium mining started in these regions in 1946, it was a purely Soviet venture. In 1954, a joint Soviet German stock corporation, SDAG Wismut, took over. The GDR government then held 50 % of the shares. The uranium concentrate produced was exclusively shipped to the then USSR.

Wismut-owned mine fields produced some 90 % of that uranium. Uranium deposits mined by Wismut can be subdivided into five geological types:

- lenticular and stockwork deposits in Palaezoic shales, limestones and diabases ("Ronneburg" type);

- hydrothermal vein deposits ("Schlema" type);
- deposits in Upper Cretaceous sandstones ("Königstein" type);
- seam-like deposits in fluvio-lagoonal, carbonate sediments of Upper Permian age ("Culmitzsch" type);
- uraniferous bituminous coals in molasses of Lower Permian beds ("Freital" type).

The company also operated two processing plants: one at Seelingstädt near Gera, and the other at Crossen near Zwickau.

An agreement concluded between the German and the USSR governments came into force on 20 December 1991. It provided for the transfer to the Federal Republic of Germany of the 50 % Soviet shareholdings and for the Soviet Union's exemption from sharing costs for the decommissioning of the mines and the rehabilitation of all contaminated plant areas.

Uranium production operations by Wismut ceased on 31 December 1990.

Today, the restructured Wismut GmbH is striving to reach optimum conditions, ecological, economic and social, in decommissioning and clean up activities.

The sudden and unprepared switch from production to decommissioning and clean up following German unification has put Wismut in a somewhat unique situation. As no provisions had been made in the past, nor any planning been done with regard to decommissioning and rehabilitation activities, we had to move on parallel lines and advance step by step.

Planning, conceptualization, and the development of technologies for decommissioning and clean up coincide with the implementation of appropriate measures which would not hamper future activities.

Sources of exposure

<u>Plant areas</u>
These comprise, among others, an area of 1517 ha covered by waste piles, 720 ha of tailings impoundments, 9 underground mines, and one large open pit mine.

Current exposure is essentially due to the following sources:

<u>Tailings</u>
Release of contaminated seepage to receiving streams and groundwater, radon and air-borne radioactive dusts;

<u>Waste piles</u>
Essentially the same type of source, partly lower concentrations, but located closer to residential areas;

<u>Mine ventilation</u>
Radon, dust;

<u>Mine waters</u>
Radioactive and non-radioactive contamination;

Contaminated facilities
Structures and equipments used in former mining and milling activities.

Contaminated soils

Preparation for rehabilitation
In 1991 and 1992, the main emphasis was put on actions to reduce current population exposure, for example by covering exposed beaches in tailings management areas and collecting seepage waters.

Development and planning for rehabilitation activities was another important feature.

During the period 1991-1992, an environmental register was established that covered Wismut plant areas and vicinity properties. Ambient dose rates were measured using a grid of 20 x 20 m. An analysis of the results showed that some 20 % of the total property and 40 % of the plant areas needed clean up. Measurements went on in greater detail and were enhanced by drive core and soil sampling.

Rehabilitation technologies

The conceptual design developed and updated by Wismut GmbH for the final rehabilitation of the sites consists of the following key elements:

- **clean up of mine workings:** removal of all hazardous materials; backfilling of mine workings which in the long run might constitute a hazard to aquifers or cause surface subsidence;

- **mine flooding**, including, if need be, treatment of discharge for regulatory compliance;

- **in situ rehabilitation of tailings ponds:** removal of water cover, dewatering of sludges and cover building, treatment of water before discharge;

- **in situ rehabilitation or relocation of waste piles;**

- **dismantling and demolition of contaminated facilities; clean up of plant areas:** deposal of contaminated scrap metals and debris into tailings ponds; removal of waste piles into open pit mine;

- **development of an environmental monitoring system.**

Mines clean up and backfilling

Wismut's underground mine workings constitute a network of galleries, drifts, shafts, and rooms branching out in all directions. At the end of 1990, when production ceased, the number of shafts amounted to 56, with open mine workings stretching over 1 400 km and covering an area of 110 km^2.

Prior to the flooding of the mine workings, before the water is "allowed to come in", hazardous materials must be removed. This concerns in particular materials like oils, greases, fuels, acids, paints, solvents, lead, mercury, and other chemicals. To further reduce the risk of subsequent mine water pollution, former shops, fuel depots, and explosive stores are not only cleaned up, but all contaminated areas will in addition be covered by layers of concrete.

Partial backfilling of mine workings will be required to protect the surface, groundwater, and the atmosphere by reducing the radon flux.
The backfill material used is a mixture of sand, binding agent and water. Apart from cement, ashes from lignite-firing power stations are used as binder. The use of ashes is subject to stringent quality control. Ashes are a cost-effective alternative to cement; and their physical properties for backfilling purposes are equal or superior.
The backfill material is mixed at the surface and placed in the mine workings via boreholes and pipes. Once in place, it takes some time to set.
From 1991 to 1994, Wismut placed some 4.5 million m^3 of backfill in underground mine workings.

To avoid surface subsidence, backfilling, down to a depth of 100 m, was required of all outlets such as shafts, as well as of large-diameter boreholes, rise headings und near-surface mine workings. All mine-related cavities beneath the township of Ronneburg were completely backfilled down to the depth of 240 m.
Due to the solid structure of the rock mass, the major part of mine workings at greater depths could be left open.

Mine flooding

Both economic as well as ecological reasons clearly speak against indefinite dewatering of underground mine workings. Therefore, controlled flooding of the mines is the only feasible long-term option.
Once mine drainage is shut off, all mines will be flooded by the natural influx of infiltration and ground waters.
What we want to achieve is the maximum possible flooding level, which, we assume, will best restore original hydrogeological conditions and help minimize the thickness of the aeration zone which as the result of oxidation and infiltration processes is a source of increased contaminant discharge.
To prepare for the flooding, modelling is under way to determine relevant hydrodynamic processes and contaminant transport.
In addition to these general measures which apply to all mines, site-specific action is also required.

In the Ronneburg mining fields, underground barriers will be put in place to hamper contaminant transport between workings having diffently contaminated waters.
At the Niederschlema-Alberode site near Aue, there is a direct relationship between flooding and mine ventilation. Up to the present time, return air from the mines is exhausted via a single upcast shaft situated at a distance from surrounding communities. Once the flooding waters reach the -540 m level this system will no longer work, and there is a risk of fugitive emissions of radonrich mine air from non-flooded near-surface mine workings which would concentrate on valley floors of the Schlema community. Radon flux would be in the order of 5,000 to 8,000 kBq/sec. To eliminate this risk, a new ventilation system has to be installed

and made operational before flooding reaches the -540 m level in order to ensure permanent evacuation of mine air from the Marcus-Semmler-Stolln level via other shafts outside the Schlema perimeter.

Flooding of the Königstein mine is a problem apart, as four aquifers are running right across the deposit with sandy limestones and clays separating them. The three upper horizons serve for drinking water purposes at some distance.
Until the early 80es, there was conventional room and pillar mining in the lowermost aquifer. Running on parallel lines with mining activities, studies on underground acid leaching of low-grade ores started in 1968. Production switched to 100 % in situ leaching in 1984. Less permeable rock was blasted and prepared before sulphuric acid was injected from drifts whereas boreholes were used to inject acid into more easily permeable rock.
The pregnant liquor was collected at the bottom of the leaching blocks and pumped to the surface via pipes. After sorption of uranium, the barren liquor was then recycled to the leaching circuit.
When uranium production ceased in 1990 some 750,000 m^3 of solution were in the circuit. Following neutralization and precipitation of radium by barium chloride the effluent is discharged to the receiving streams.
Quite more complex is the removal of approximately 2 million m^3 of leach solution left in the pores of the rock. During the flooding of the mine this solution will be displaced by and mix with incoming waters. For that reason, flooding waters will be collected at the northern end of the deposit, treated and then pumped back into the mine workings; this process will go on and on until contaminant levels fall below regulatory standards.
The rationale of this process is that separations between the mined lowermost aquifer and the higher drinking water horizon will not be completly watertight. Sure, all accessible mine workings will be sealed, but access cannot be excluded via natural geological structures.

Rehabilitation of tailings management areas

At the former milling sites, emphasis is put on the decommissioning of tailings management areas. They contain a total of some 160 million t of fine-grained mill tailings; their thickness is in the order of up to 70 m; and they contain 10 Bq/g radium.
As the major portion of seepage from the ponds is being collected and pumped back, there is next to no risk to nearby aquifers. As a preventive measure and within the framework of preparations for clean up, a comprehensive hydrogeological investigation program was started in 1992 to study the environment of the TMAs and improve the data base for site modelling.
In the current state of knowledge, the preferred option would favour rehabilitation in situ: (i) removal of the water cover, (ii) gradual covering of exposed beaches as a precaution against erosion and sand-blown dust, (iii) dewatering of the slimes by means of gravitation wells, vacuum, and additional load in combination with geotextiles and textile wicks, and (iv) covering of dried-up tailings using cohesive soil materials in order to reduce infiltration from precipitation and radon flux.

Treatment of contaminated waters

Waters from flooded mines and TMA rehabilitation are contaminated and need being treated to meet regulatory standards for discharge. Relevant contaminants are uranium, radium and arsenic. In addition to these specific contaminants we have to deal with relatively high levels of water-hardening substances and salts.
In contrast to treatment facilities available for the municipal and industrial sectors, no operational technology is at hand to deal with these specific contaminant configurations.
Therefore, Wismut GmbH is currently busy to identify technologies for the treatment of waters collected at different sites and for the safe disposal of treatment wastes rich in radionuclides. At this moment, two water treatment plants are under construction.
At the Pöhla site, for example, the following procedures is employed:
In a first stage, following the addition of hydrochloric acid, radium is precipitated by addition of barium chloride. In a second stage, ion exchange resin is used for uranium separation. Ferric chloride separates arsenic in a third stage.
The treated effluent is discharged into the receiving streams. Precipitation sludges are dewatered and drummed for intermediate storage in mine workings.
Simular concepts are being developed for the other sites.

Waste pile rehabilitation

Mining wastes were dumped in four ways: as conical piles, as hillside dumps, as table piles and as valley fill.
The piled up materials contain radionuclides and toxic chemical materials in a wide range of concentrations. Uranium mining wastes in the western part of the Ore Mountains, for example, have average ^{226}-Ra concentrations between 0.6 and 0.9 Bg/g, while concentrations at mining sites in Eastern Thuringia vary between 0.3 and 3.0 Bq/g. Precipitation infiltrating these waste rock piles mobilize pollutants and cause pollution of surface and ground waters.
In 1993, for example, seepage from waste piles in the mining district of Aue/Ore Mountains contained an annual average of up to 3.8 mg/L uranium and up to 1.4 Bq/L radium.

Waste rock piles will either be rehabilitated in situ or removed.

In case of in situ rehabilitation, slope angles will be graded from a ratio of 1 : 1.3 to 1 : 3 or 1 : 5.
Piles will then be covered with soil materials in order to inhibit radon exhalation and pollutant release due to infiltration of precipitation.
The cover may be built as a single or multiple layer system. Depending on the location and exposure situation of the waste rock pile, the single layer cover may be a thin layer of topsoil or a thick soil cover of up to 1.5 m.
A number of experiments have been conducted by Wismut, and the measurements made of resulting exhalation and infiltration rates have helped identify the following best cover design option from top down:

0.2 - 0.5 m	topsoil
0.5 - 1.0 m	water storage layer
0.2 - 0.5 m	drain layer
0.3 - 0.6 m	liner.

Wismut operates a number of test plots for studies on revegetation and on the performance of cover systems. Different species have been tested with regard to their evapotranspiration capacity, compatibility with cover materials, root penetration, and stability of slope seeding. Wismut's current studies also include the identification of optimum plant communities.
Experience from waste rock piles teaches that besides seeding of selected species one has to reckon with natural seeding of species whose root penetration makes them unfit for cover vegetation. That is why we have to consider perpetual maintenance of rehabilitated waste pile surfaces.

One example to be mentioned for waste pile rehabilitation is the Hammerberg waste pile at the Niederschlema/Aue site.
Situated at the northern edge of the Schlema community, the Hammerberg waste pile was, by its sheer dimension of some 0,35 km^2, a major source of radon and dust. In addition, steep sideslopes made it instable. To improve slope stability, 485 000 m^3 of waste material were relocated at the site and 450 000 m^3 of waste rock were imported from other waste piles. Then the entire pile was graded, terraced and covered using inert material. Finally, a top cover was constructed using 160 000 m^3 of clay material and the site was revegetated.

The relocation of piles is considered in cases when the original site is close to residential areas or when the waste rock is required at other sites. It is our aim to meet these two objectives.
Up to now, relocation of wastes piles took place at Niederschlema and at Ronneburg.

At the Schlema site, materials taken from two waste piles was used to fill mining damage at the surface and create a parkland area.

At the Ronneburg site, decommissioning and relocation of waste piles is part of the rehabilitation of the open pit mine.

That open pit mine was in operation from 1958 to 1977. When production ceased, the open pit mine had a final volume of 84 million m^3 and a depth of 160 m. Average slope angles are 50 ° which renders them potentially unstable. The major part of the open pit volume will be flooded once flooding of the mines will be under way.
Some of the waste rock piles in the Ronneburg area will be removed into the open pit mine for decommissioning. The sequence of the placement of this material will depend on the mineralogical and chemical characteristics of the rock.
Materials having high levels of sulphur will be placed below the anticipated ground water level in order to inhibit oxidation and acid generating processes. Materials having high carbonate contents will be placed above the anticipated ground water level.
After placement of the materials, a multiple layer cover system will be built over the filled up open pit.

First to be removed for placement into the open pit are waste materials from a former heap leaching site where acid mine waters and/or sulphuric acid were used to lixiviate uranium from low grade ores. As a result of the lixiviation process, Ra concentration is between 2.3 and 2.9 Bq/g, Rn exhaltion amounts to 11 - 12 Bq/m^2. Seepage waters from those wastes contain up to 8 mg/L of uranium and 22 g/L of sulphate. In order to avoid contaminant release, this material is being placed at the bottom of the open pit in shifts of 0,6 m with granular calcined limestome being added to neutralize the acid generating potential.

The relocation of this material from the heap leaching pile is being monitored by a measuring program serving the radiological survey of the operation. It comprises measurements of air-borne pollution in nearby communities, emission measurements at the waste pile site and in the open pit mine as well as dosimetry of the workforce. Throughout the relocation operation, concentrations of long-lived alpha emitters in nearby communities must not exceed 2.5 mBq/m^3.

Dismantling and demolition of contaminated facilities, decommissioning of plant areas

As many structures and plants are out of repair and radioactively contaminated their after-use would require extensive upgrading and clean-up. Unless the expenditure for the required cleaning is reasonable, structures and facilities are pulled down.

Temporary storage sites for contaminated building rubble and scrap metals are being provided on plant areas. Their final management will be a matter of disposal facilities and regulatory approval.

In the clean up of plant areas, natural soils and fills are excavated to the extent contaminated. Ambient dose rates are measured for verification. After excavations have been filled with topsoil, ambient dose rates are again to be measured. Soil-geological and radiological reports then will certify the clean up.

Environmental monitoring system

All Wismut sites, whatever their status of decommissioning, are sources of emission. Clean up activities such as the relocation of waste rock piles generate additional emissions. An extensive network of some 1,300 measuring stations has been put up to monitor emissions of radon and its long-lived daughter products, air-borne dusts and qualities of receiving streams and ground waters. Soil and biomass sampling are an integral part of this system.
Collected data are transmitted to a central environmental data base for storage and processing.

Prospects

The Wismut reclamation project is - compared with similar challenges worldwide - unique in the fields of mine decommissioning and environmental protection. Total decommissioning costs are estimated at 13 thousand million German marks to be funded by the federal budget.

Allocations from the federal budget over recent years permitted Wismut GmbH to advance to the leading edge of rehabilitation performance. The clean up of former uranium mines operated by SDAG Wismut is opening up new economic, ecological and social vistas in the affected regions.

THE RESTORATION WORK ON THE HUNGARIAN URANIUM MINING AREA

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Abstract

In Hungary the uranium mining and milling activities are close to the shut-down, so the planning for restoration works and implementation of different remedial action has been undertaken in the last years. The restoration planning and works were begun in 1992-93, and at the first step the mining piles have been restored. The main goal is that the restored residues on mining and milling area are fitted into the surrounding topographic features, and the other important aspects, like radiological situation, water management and revegetation are also taken into account. The plan of the pile 3 includes main experiences of the earlier restored piles, so it takes into stronger consideration the optimization of pile relocation and sloping, mining cavities and activities and hydrography.

1. INTRODUCTION

In the last years because of the exhausting of good quality uranium ore and the decreasing of the price of it on the world market, the uranium producer has to change its strategy. Besides the reduced production and the plan for closing, the restoration works have also been placed into the limelight. The restoration project for many years has a step by step trend, it is getting more and more difficult task. The first task was to carry out a precise survey of the amount of mining and milling residues and a lot of investigation for remedial action, for example like a radiohygienic analysis. The next task has been to plan for the actual works of restoration project. Among the actual work for restoration series the remedial action of the waste rock piles is the first and then heaps for leaching and then the retention ponds and at the end the mining and milling facilities. The tasks are very huge as seen from the list of quantities [1, 2, 3]:

- waste rock piles: 1.8 10^7 t
- heaps for leaching : 7 10^6 t
- ponds: 2.5 10^7 t

2. RESTORATION OF WASTE ROCK PILES

The task for remedial action of waste rock piles is also characterized by a progression toward more and more difficult work. So at the first step the least piles have been restored, where the restoration work is still lesser, however a lot of experiences have been obtained for later works (like a pilot plant). The restored site has continuously been checked in order to get information about the needs for correction and goodness of plans. The two little piles have 3 10^6 t of waste rock.

The restoration plan has a big problem, namely the waste rock piles have been emplaced on a hilly countryside, so the plan has to take account of the topography and erosion problem [4, 5].

2.1 Main experiences of the restored site

The most important experience is the angle of slope of the restored piles that is not to be greater than 7°, because above the angle of 7° the erosion is very early beginning.

The second learned lesson is that perfect surface water collection is needed. A proper pile arrangement has to be performed in order to avoid water erosion and a ring collector ditch is to be built so that big rainfall water is diverted in an orderly way.

The covering layer must be compacted very well in every place by using heavy machines. So the erosion problem, the rainwater infiltration and the local increase of the radon concentration can be easy eliminated.

The control radiological measurement above restored pile verify that the measured values (dose rate, radon flux, radon concentration) are below the limit, which is prescribed by the planning. So the covering layer in depth of 70-100 cm is enough to reach the goal of radiation protection. However, above clay a thicker humus is to be provided so that grass grows promptly [6, 7].

2.2 Special aspects of restoration of the pile 3

The waste rock pile 3 is one of the greatest piles, it has 4.6 million m^3 of waste rock. A lot of special aspects have been taken into consideration so the remedial action will provide acceptable conditions for a long period of time.

2.2.1 Hydrography

Near the pile 3, streams gave a little more complication to the pile restoration. The catchment area of these streams is 4.7 km^2 . The annual precipitation of this area is about 3.2 million m^3 that is 8700 m^3 / day. The summarized yield of the streams is only about 600-800 m^3 / day, the great portion of the rainwater is filtered into soil, as well as moving in the soil. In the case of unrestored pile the rainwater can get to the nearest stream (Zsid stream) through the infiltration process, meantime a lot of radioactive isotopes are dissolved into water. Because the infiltration water arising from the pile 3 adds to the yield of Zsid-stream, so water movement is diverted and it needs a select handling of this water.

2.2.2 Geology

On the basis of the results of discovery wells there is a geology formation under the pile 3, which has a great extension and a good 'waterproof' capability. This formation hasn't connected with the subjacent karstic water stratum, so it's sure to stop the surface water before it reaches the karstic water.

The mining and milling area is geodynamically stable. Regarding the geology surveying, it's stated that this area has very low seismic process and so the 7° earthquake in a Mercalli scale can occur at the maximum. In a 100 year period the 6° earthquake can happen below a probability of 10 %, when a slope of pile will be destroyed. Afterwards, it's concluded that the effect of earthquake isn't to be taken into account.

2.2.3 Mining activity

Between 1959 and 1963 under the pile 3 the exploitation of the mine 1 was carried out and the mine galleries (cavities) can be found in depths from 370 m to close to the surface. From the beginning of 1965 the rate of subsidence has been registered. The maximum of subsidence was about 1 m and the average one was 0.3-0.5 m. In the last decade the study of this subsidence verifies that there is no further subsiding in this area.

It's stated the basement of the pile 3 hasn't had to be strengthened before restoration work.

3. RESTORATION TASK OF PILE 3

3.1 Short list of the main task

During the remedial action of the pile 3 the next important viewpoints were included:

- providing adequate area for restoration
- considering the topographic features
- ensuring the panorama of surrounding hills
- performing the proper ridge of pile
- creating a similar slope degree to surrounding hills
- avoiding a lot of dust production
- ensuring the radiation level
- creating a proper cover layer
- building the divert water system
- performing the revegetation
- public relations
- considering the routine mining activities

3.2 More details of the restoration

3.2.1 Adequate area

As mentioned in chapter 2.2.1, this area was suitable for the in-site restoration, because the former mining activity, hydrology and geology problems didn't perturb the final solution. The removement of the pile 3 to the other place which should have been perhaps more suitable, but it should have been a very expensive solution according to transportation of about 5 million m^3 of waste rock.

3.2.2 Topographic features

In spite of that the pile 3 was emplaced on a hilly side, after the restoration work the contour of restored pile couldn't give a large obstacle in a view of hill behind the pile. The different view (panorama) denoting the letters A, B, C are shown on Figure 1.
The optimization of relocation (smoothing) procedure had to be carried out so that the slope degree and the height of the restored pile ensure the least shadowing of behind hills.

3.2.3 Direction of pile

At the smoothing arrangement the direction of ridge of the pile had to be justified to the surrounding hills, namely this direction line is from north to south (Figure 2).

3.2.4 Slope of pile

The value of slope degree also had to be justified to hills. However, the range of degree had to be analyzed for the acceptable erosion solution.

3.2.5 Dust production

Both during the restoration work and after the restoration the dust production should be minimized to avoid unnecessary contamination of environment and radiation burden of the popula-

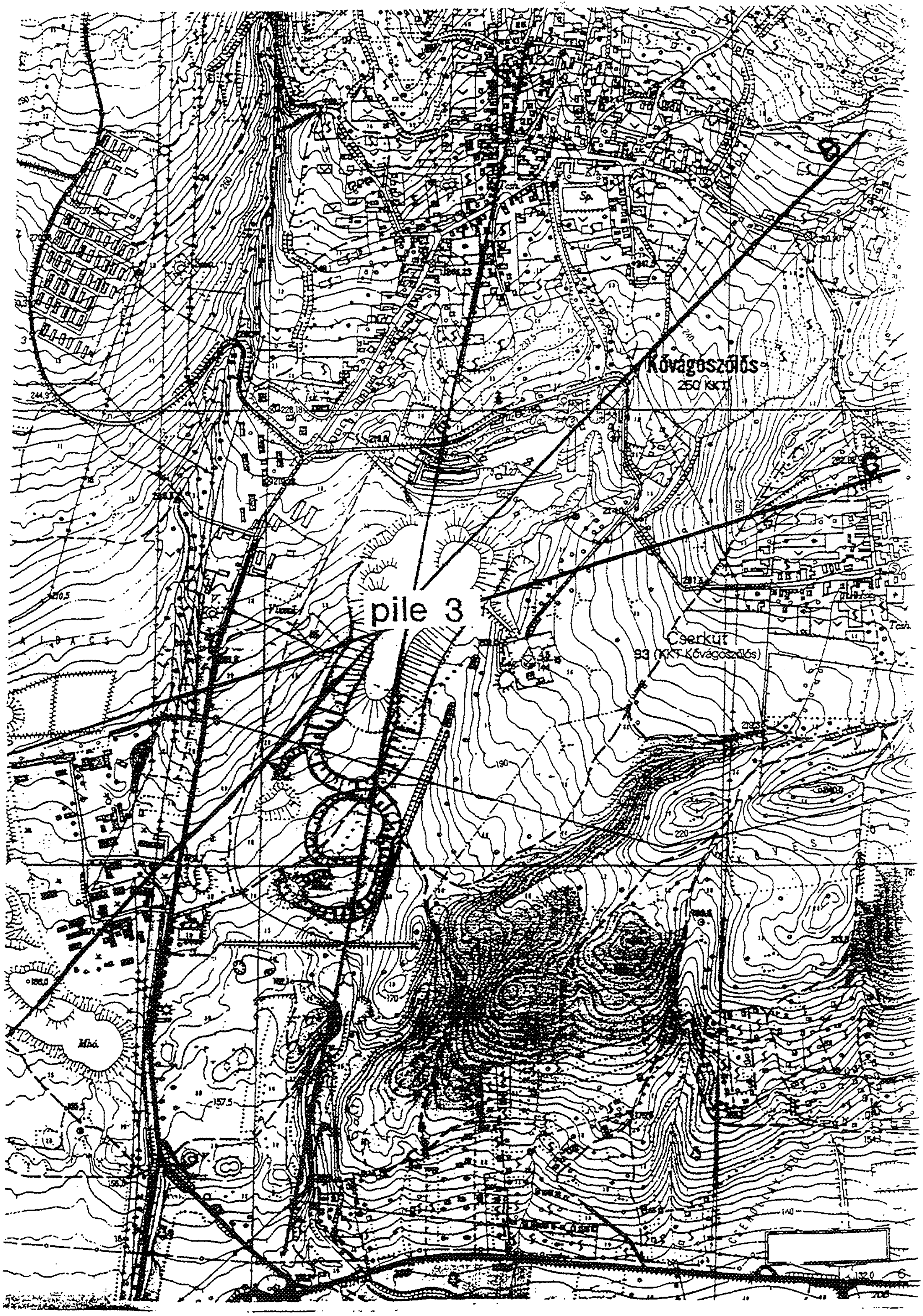

FIGURE 1

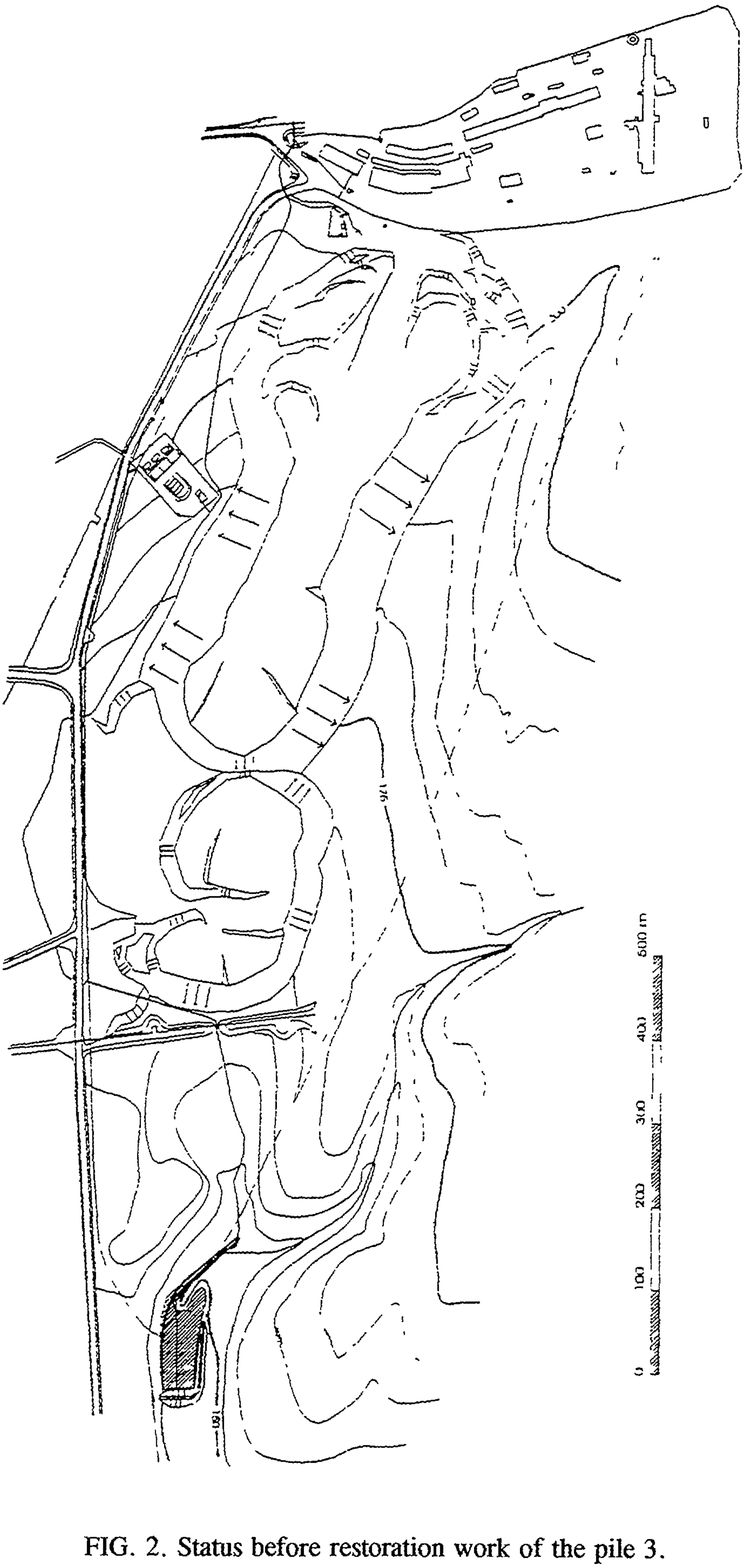

FIG. 2. Status before restoration work of the pile 3.

tion. Because the sandstone, where the uranium ore occurs, is very easy turn into dust form, the relocation of pile should be carried out during or after wet weather. And after the smoothing arrangement as soon as possible the pile is to be covered with clay layer.

3.2.6 Radiation protection

For requirements of radiation protection, the main assumption is that someone stays 1700 hours (0.2 occupational factor) in a year, then the radiation burden doesn't exceed the 1 mSv. The derived value for typical radiation pathway is:

- external dose rate : 400 nGy/h
- radon concentration (at 1 m) : 20 Bq/m^3
- radon flux : 0.7 Bq/cm^2/s

3.2.7 Cover layer

The cover layer has to be created in a depth of 70-100 cm properly in order to ensure the next aims:

- to stop the dust production
- to decrease the external dose rate to acceptable level
- to decrease the radon flux and radon concentration in air above the pile
- to carry away the rainwater with minimal infiltration
- to be capable to grow plants.

3.2.8 Water system

One main function of water system is to collect the surface and a portion of infiltration water and to avoid this water increasing the flow of streams. Building of the ring ditch and the pond are good solution to collect the whole surface water of this area. From the pond the collected water is led into the lower mining cavities through a boring hole. In the mining cavity the discharged water is filtered by a natural process. At the end the main portion of rainwater that gets through the pile also reaches the lower mining cavity considering the hydrogeology survey and calculation. In that way the surface water is diverted, so the dissolution of radioactive isotopes from the pile is minimized and the near streams won't be contaminated by radioactive elements.

Otherwise, the most important limits for the releasing of water to streams are:

- Uranium content : 2 mg/ dm^3
- Ra concentration : 1 Bq/ dm^3

and in addition the other chemical component content has to comply with the authorized limits.

3.2.9 Revegetation

The vegetation and return to natural condition is important to be brought about as soon as possible considering the erosion process and the climate condition. The cover layers are formed out in a such way that the selected vegetation can grow in this soil. So the clay layer which has depth of 70 - 100 cm is to be covered with a humus layer, which is suitable for cultivation of the grass and shrubs and later trees, too.

3.2.10 Public relations

It's very important that the restoration works are known to the public through the local authority. Information are given through the public media to interested people or organization, so that uninformed individuals do not interfere with the restoration activities.

3.2.11 Mining activity

The current mining activities are also taken into account during the remedial action, so the restoration work doesn't interfere with the mining process (i.e. transportation).

4. PRACTICAL RESTORATION WORKS

4.1 Pile arrangement (on site relocation and smoothing)

On site arrangement were carried out on the waste rock of 4.6 million m^3 and in addition this amount is supplemented with 300.000 m^3 heap leaching residues.

The pile 3 was divided into 22 sections and the arrangement was performed between these sections (Figure 3).

The most relocation works on the shapeless pile were:

- reshaping the north to south direction line of ridge
- creating the proper slope for the west to east and the north to south direction

During the relocation and smoothing works the high dose rate area of pile was taken into account, namely where the dose rate above the pile exceeded the value of 600 nGy/h, then this area was covered with waste rock arising from a low radiation level (100-600 nGy/h) area. Because there was a lot of low dose rate area on the pile, it didn't cause any special problem for the arrangement of rock.

The total amount of relocation and smoothing works was about 700.000 m^3, and 300.000 m^3 of the rocks had to be transported by heavy truck from another place (Figure 4).

4.2 Building the water system

The ring ditch was made from concrete and led to a pond that can be found at the lowest point of pile area. From the pond the water flows into the mining cavity by gravity.

Because the Zsid-stream is flowing very close to edge the pile 3, a new bed was built for this stream in farther distance of 20 - 30 m of the pile in a length of about 1.5 km.

4.3 Cover layer

During the covering (smoothing) of the pile each clay layer of 15-20 cm was compacted by heavy machine. The final depth of covering clay is about 70 - 100 cm. This compacted clay layer is enough to ensure low radiation levels above pile.

On the north to south ridge of the restored pile a service road was built for the later restoration and control works.

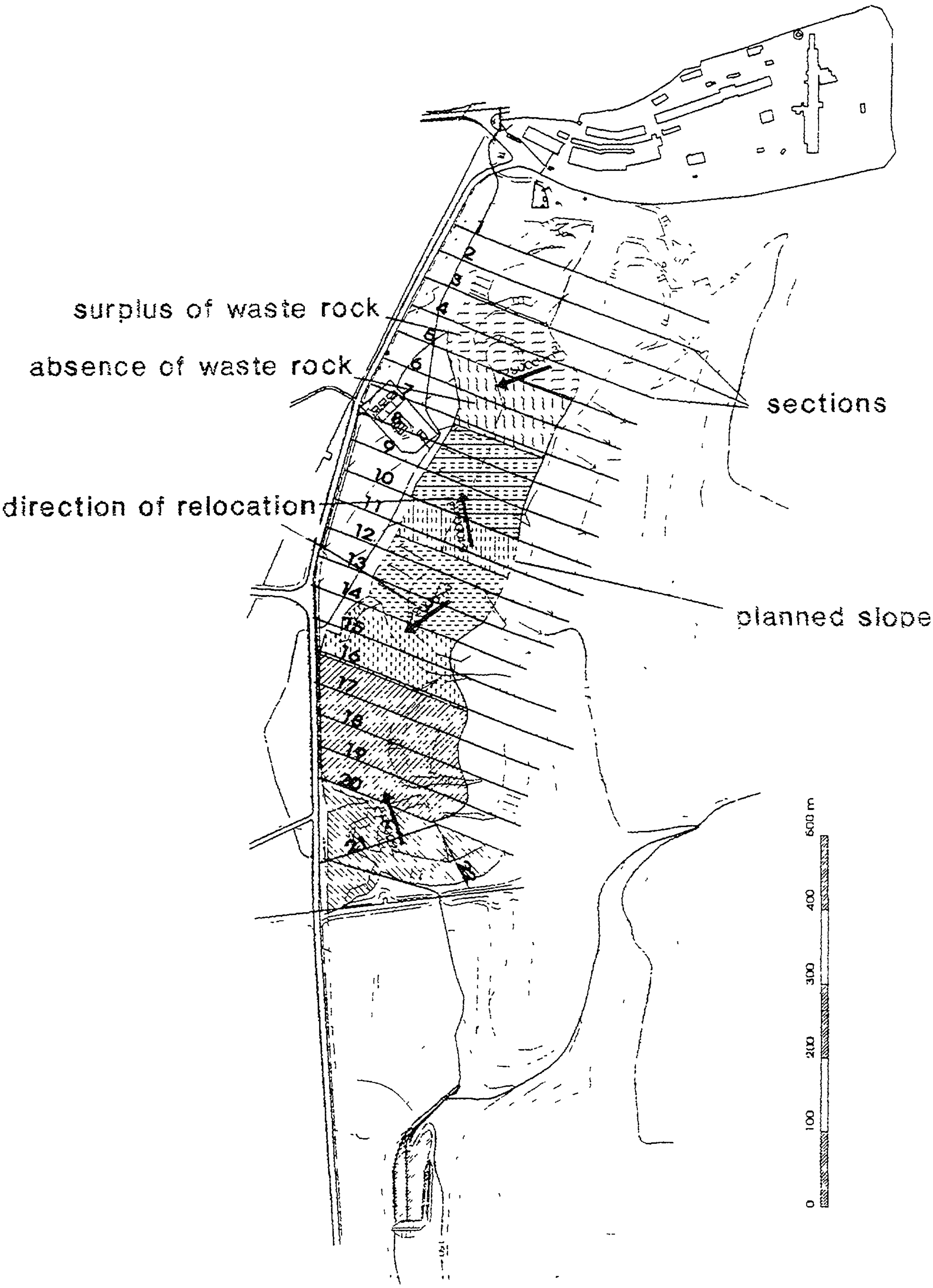

FIG. 3. Relocation plan of the pile 3.

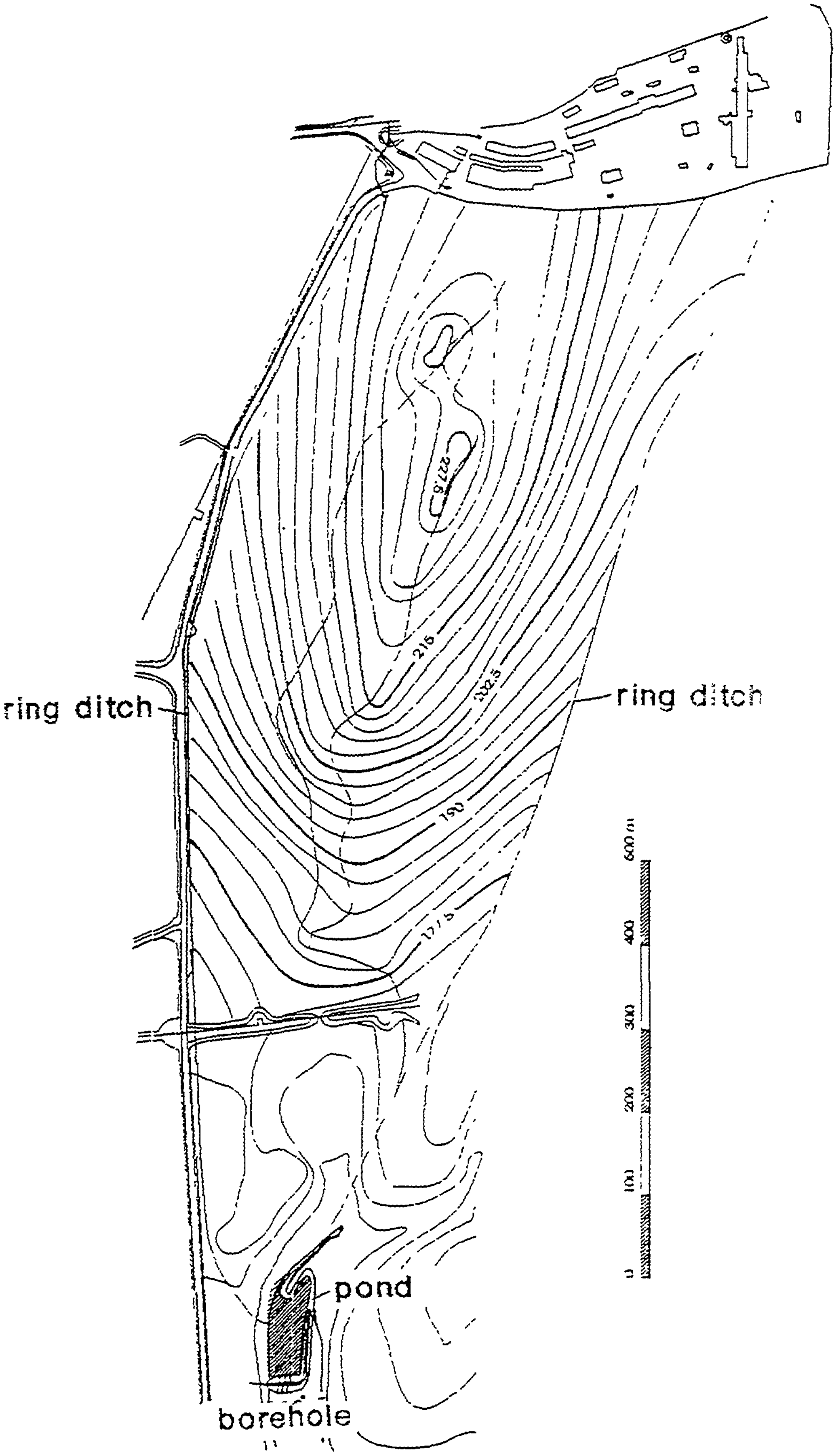

FIG. 4. Topography of the restored pile 3.

4.4 Radiation protection

During the remedial action of the pile 3 the next radiation levels characterize it:

	Requirement	Before restoration	After restoration
Dose rate at 1m above the pile in nGy/h	400	100-1800	100-200
Radon concentration in Bq/m^3	20	20-300	10-20
Radon flux in $Bq/cm^2/s$	0.7	0.1-0.4	< 0.2

The calculated dose from these data is the following:

	Before restoration	After restoration
	mSv/year	
External exposure	0.95	0.21
Radon inhalation	1.3	0.27

It is stated the radiation situation on the restored pile is acceptable and it is assumed that this state will remain for few hundred years.

4.5 Water quality

According to the assessment, rainfall infiltrates an uncovered pile having the uranium content of 70 - 100 g/t, then the uranium content of the released water is 10 - 15 mg/ dm^3. This value is much more greater than the authorized limit of 2 mg/ dm^3. Therefore from the point of view of safety it's practical that the collected water is led into the mining cavity after extraction of uranium by an ion exchange process. Similarly, the removal of the stream bed has caused the concentration of the radioactive isotopes released to water near the restored pile to be below the authorized limit.

4.6 Revegetation

Surrounding the pile 3 is a woodland, therefore the main aim is long term remediation.

The slowness of revegetation is also taken into consideration, so at the first step the grass has to be planted and then shrubs, and at the end different trees.

However, soil (clay) has to be improved for the revegetation task. The clay cover is acceptable for radiation protection, but it is not suitable for the plant cultivation. A humus layer could be produced over clay, if it's overlaid with sewage mud. Nevertheless, the grass-covered restored pile is needed to halt the erosion process, too. And the next time shrubs and trees will be planted on the pile.

Besides the climate and spreading of species the shrub and tree plantation will be taken into account:

- humidity of soil
- root extension
- no collection for it or for its harvest.

The planting distance is also important because

- surface water distribution
- reduction of soil erosion
- fire protection.

Therefore the suggested distance for stock is 40 cm and for spacing is 2.2 m.

4.7 Control after restoration work

After the actual remedial action a continuous control measurement program is to be implemented. According to this program on the restored pile 3 the next measurements are carried out:

- gamma dose rate measurements at 50 points in a mesh at 1 m above the pile
- radon flux measurements at the same point
- radioactive isotopes study in the cover layer considering the migration process
- water sampling from near streams, ring ditch and pond, the analysis is extended for uranium and radium content.

At present the background level measurements are performed above the restored pile 3.

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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 final phase of the works carried out to secure the industrial waste disposal situated near Brescia (Italy) contaminated by Cs137 of unknown origin, and represents the logical continuation of the papers presented at the Budapest and Piestany Workshops.

After the campaign survey undertaken to evaluate the amount of the radioactivity on the surface of the facility, the deposition of a first coating, in order to temporary stabilize and immobilize radioactive contamination, and the drilling campaign undertaken to investigate the quantity and the distribution of the contamination inside the mass of waste, the whole surface of the waste disposal was eventually coated with a physical cover and protected with an erosion control net.

In particular, the lecture focuses on the technologies involved, the description of the works undertaken and the results obtained.

INTRODUCTION

The matter of this report is the description of the final intervention carried out in order to secure an industrial waste disposal contaminated by Cs137 of unknown origin.

It is useful, anyway, to briefly remind here the main steps of the intervention already reported in the Budapest and Piestany Workshops.

The contamination of the site was evidenced by geochemical and administrative controls started during the second part of 1989, after an increase of Cs137 in the water of Po river, just near the nuclear Power Plant of Caorso (Piacenza, Italy), had been pointed out during some routine controls. The level recorded was about five times the background value: so, even if that value was far below the safety limit, the presence of that radioisotope in the power plant site of Caorso was immediately monitored, but luckily no contamination was there evidenced. By Cs137/Cs134 ratio determination, it became then clear that the radiocontamination could not be due to any fission products correlated with used nuclear fuel, and then with any nuclear power plant accidents (Chernobyl included).

Using a well known geochemical survey method applied to the local river network it was possible, going upstream from the signalized place on Po river, to locate the source of the contamination, found to be two aluminium scraps refineries near Saronno, about fifty kilometers north-west of Milan.

Through further administrative investigations on some correlated foreign suppliers, it was then pointed out that two more scraps aluminium refineries near Brescia, about 90 Km east of Milan, were highly contaminated .

The last step of the control was then toward the industrial dump facility (fig.1÷2) where they used to discharge the melting salts from the drum ladles of the latter two refineries, in the aluminium extraction process: and, as a matter of fact, that facility was found to be contaminated, not uniformly, but with some piece of solid salt blocks along the slope highly contaminated (28 Bq/g).

On the other hand, following the hypothesis of an uncontrolled Cs 137 source mixed to the aluminium scraps and submitted to the same refinery process, melting in a drum ladle included, for chemical reasons Caesium could not be linked to aluminium and stay in the final aluminium ingots, but necessarily it had to remain in the melting salts. In fact, aluminium ingots always resulted with very low activity (<1 Bq/gr) and could so be regularly distributed, while salts used in the melting process resulted, more or less, contaminated.

At the same time it was evident that, as a consequence of the refinery process, the Cs137 fragments could not but be scattered everywhere in the mass of waste, in such a way that certainly it could not be possible to separate and pick them all up.

Anyway, by the radiological survey on the surface of the waste plant, it was possible to see that radioactivity was present mostly in some blocks along the slope of basin n° 3; and the blocks were the melting salts directly coming from the drum ladles of one of the two refineries, situated in a buried layer some meters below the surface.

By the drilling campaign (fig.3) in every basin it was then possible to state that significative radioactivity was present only in the basin n° 3, some meters under the surface; the total radioactivity of C_s137 was valued in about 1.1×10^{12} Bq (~29 Ci) in that basin, while in the basins 5 and 6 the average concentration of Cs137 was only, respectively, about 125 Bq/Kg and 6 Bq/Kg.

Moreover it was confirmed that the most active samples were the ones from the sault blocks located in the intermediate layers of waste, according to the information on the discharging schedule and according to the previsional model.

Furthermore, 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.

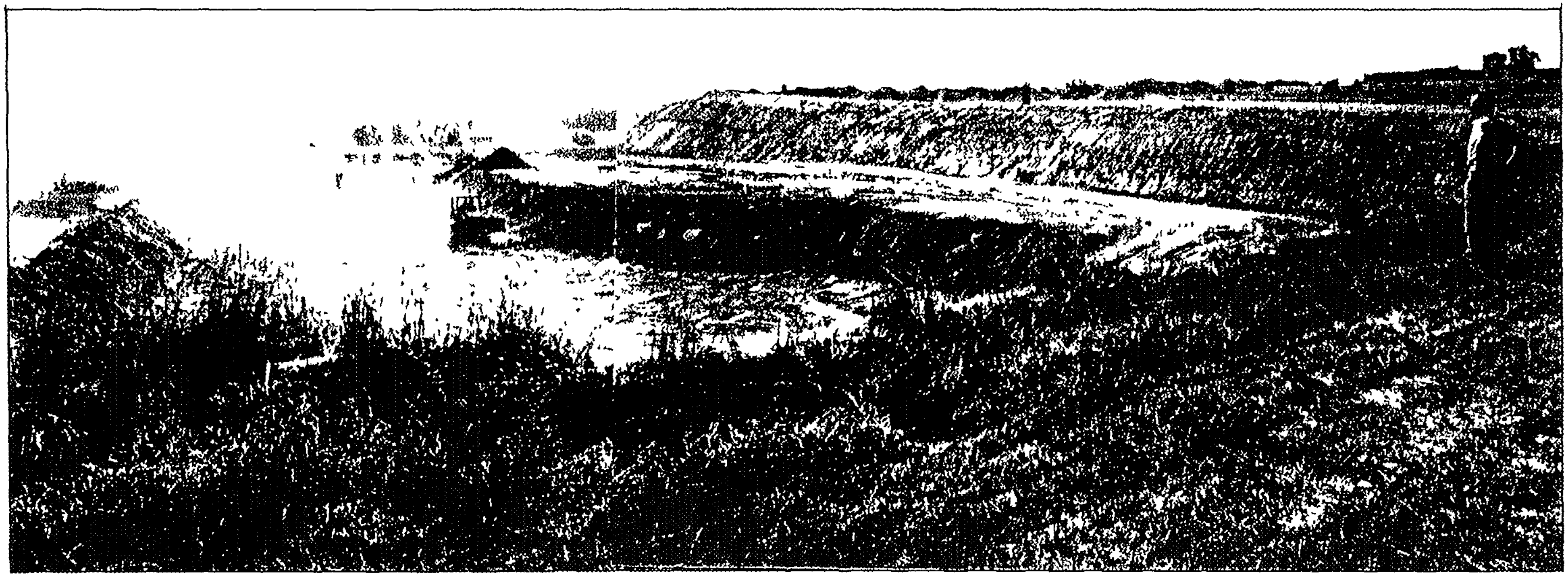

Fig. 1 - The contaminated waste disposal near Brescia

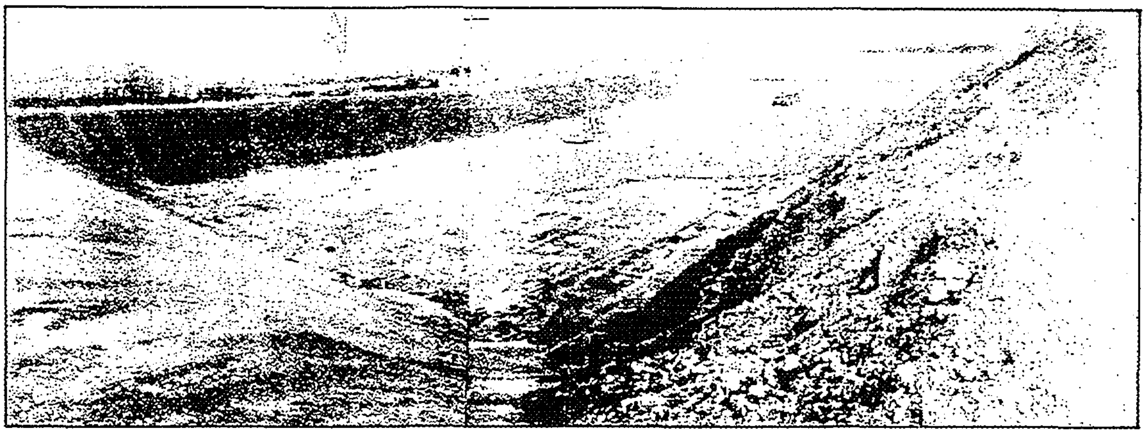

Fig. 2 - Another view of the contaminated waste disposal

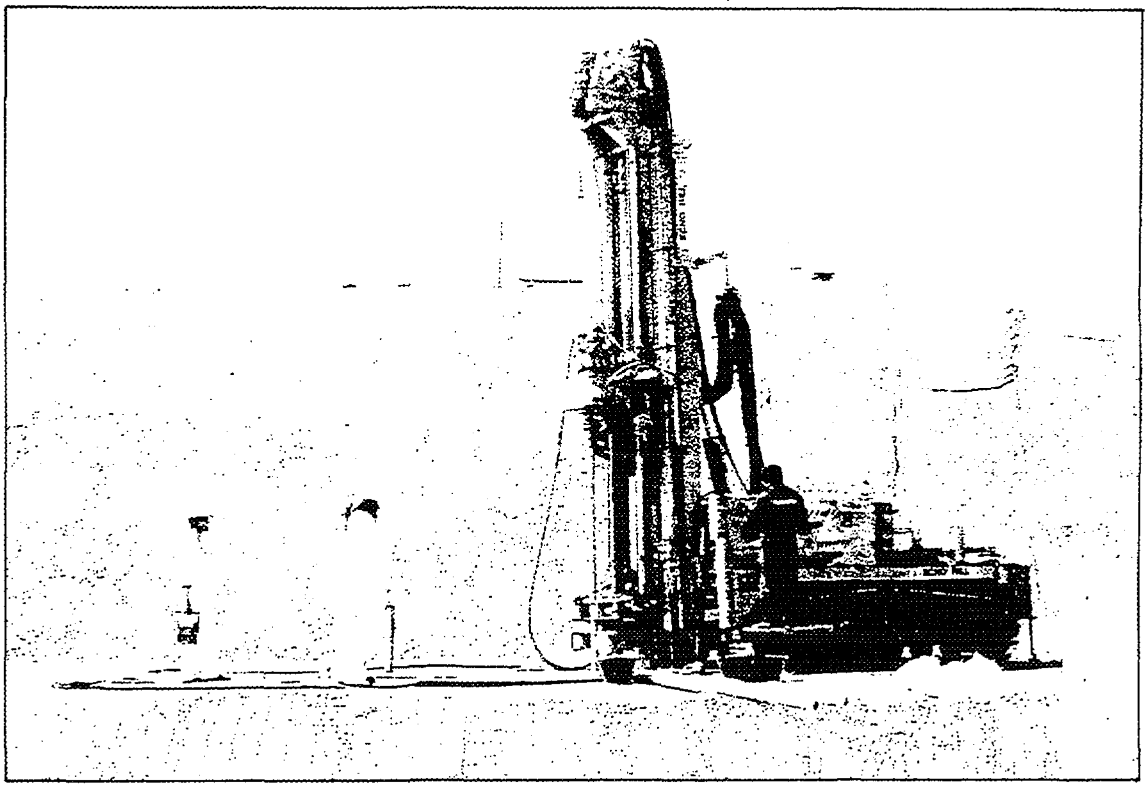

Fig. 3-The drilling campaign

In that situation, as it was impossible to separate the contaminated material, too scattered in something like thousands of cubic meters of waste, and remove it, it was then decided to apply a physical cover, made of different natural coatings, in order to immobilize radioactive contamination.

THE FINAL COVERING

The final securing intervention was based upon a good cover of the whole surface of the waste plant, with particular attention to the part in correspondence of the waste of basin n 3, then adding to it an erosion control net.

Fig. 4 is just the schematic standard cross section, direction N-S, of basin n°3.

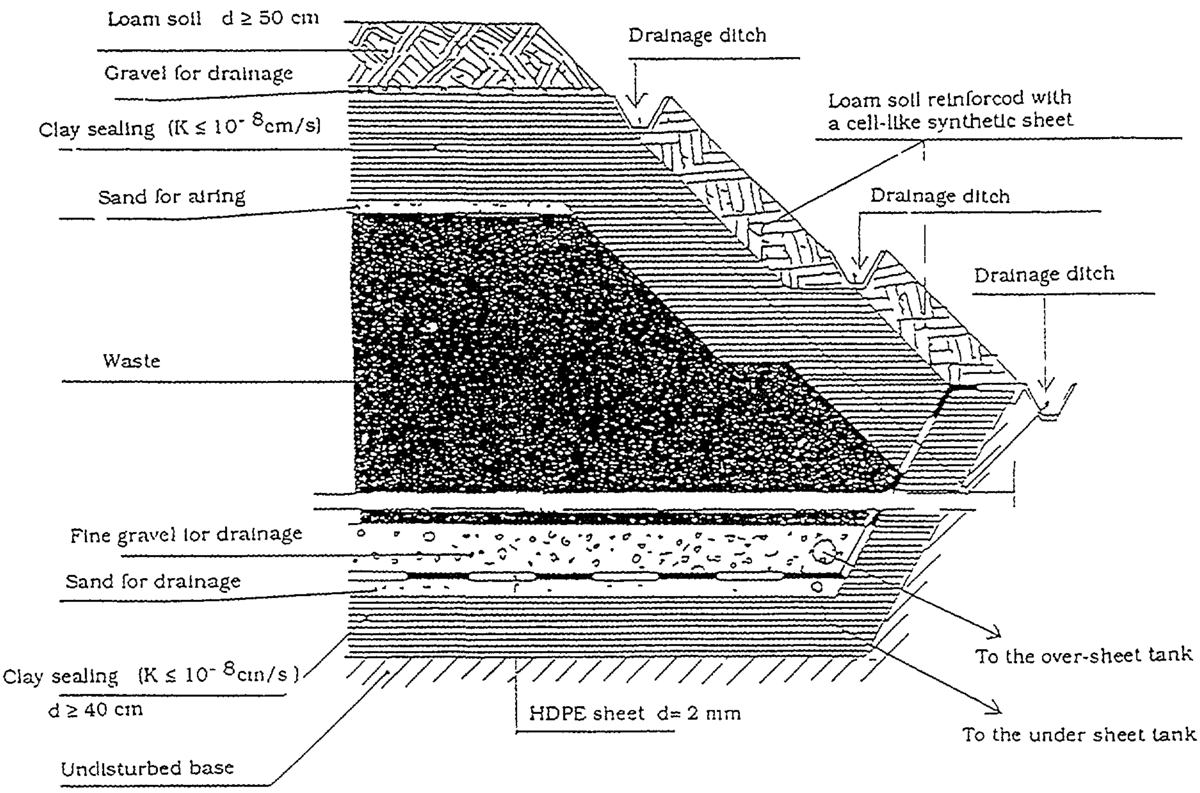

Fig.4 - Cross section of the cover

The intervention was based on a multilayers cap of natural materials, laid on the waste plant surface. The heart of the intervention was of course the clay layer, that was made with minerals of very good quality. In that way it was drastically decreased any percolating water inside the waste, and sealed the contaminated material with a very effective geochemical barrier, due to the absorption characteristics of clay toward caesium.

Very important were some erosion control nets, that is to say a surficial rain-water collecting network, able of conveying all the surficial rain water outside the waste facility, in order to prevent any consistent erosion of the protective layers.

Fig. 5 represents the starting point for the final intervention and the first clay layer (20 cm) deposited during the interim action is visible: it had been carried out in order to make the

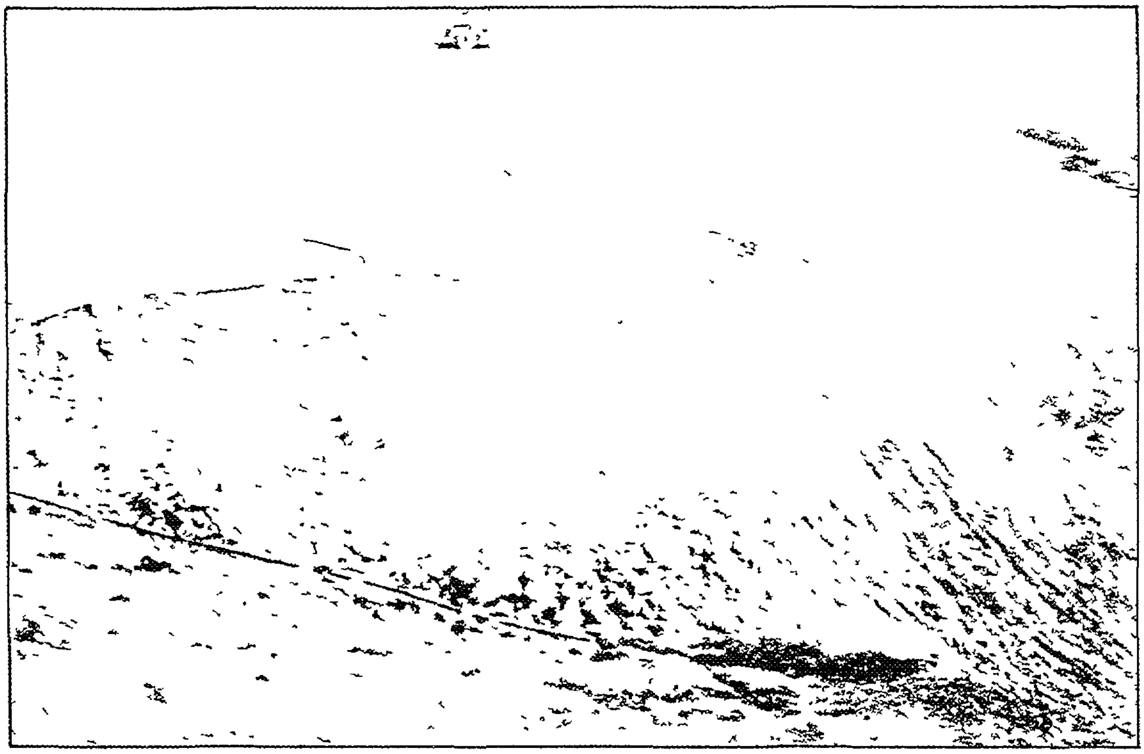

Fig. 5 - The situation after the interim intervention

surface of the site safe from the radiological point of view, and in order to let people and vehicles go freely all around to take measurements and samples.

The new layers were deposited after a scarification of the whole previous surface, in order to link them to the mentioned first clay layer. Then, three more clay layers, 20 cm each, were deposited in correspondence of the basin n° 3, and one clay layer, same depth, in correspondence of the other basins (fig. 6).

The works were carried out according to a proper planning: the whole area involved was divided in twenty-four portions, and for each of them the duration, the beginning and the type of works planned.

Great care was placed in the sequence of actions, in order to expose every clay layer to air only for a short time.

Moreover, some precautions against risks of swelling that could rise for gas production inside the mass of waste were taken by interposing a thin sand-gravel layer between the clay and loam layers on the top, and between the waste and clay layer in the middle, both connected with some proper ventilation openings.

The upper layer consisted in a continuous deposition of 50÷80 cm loam soil layer, on which some grass was planted.

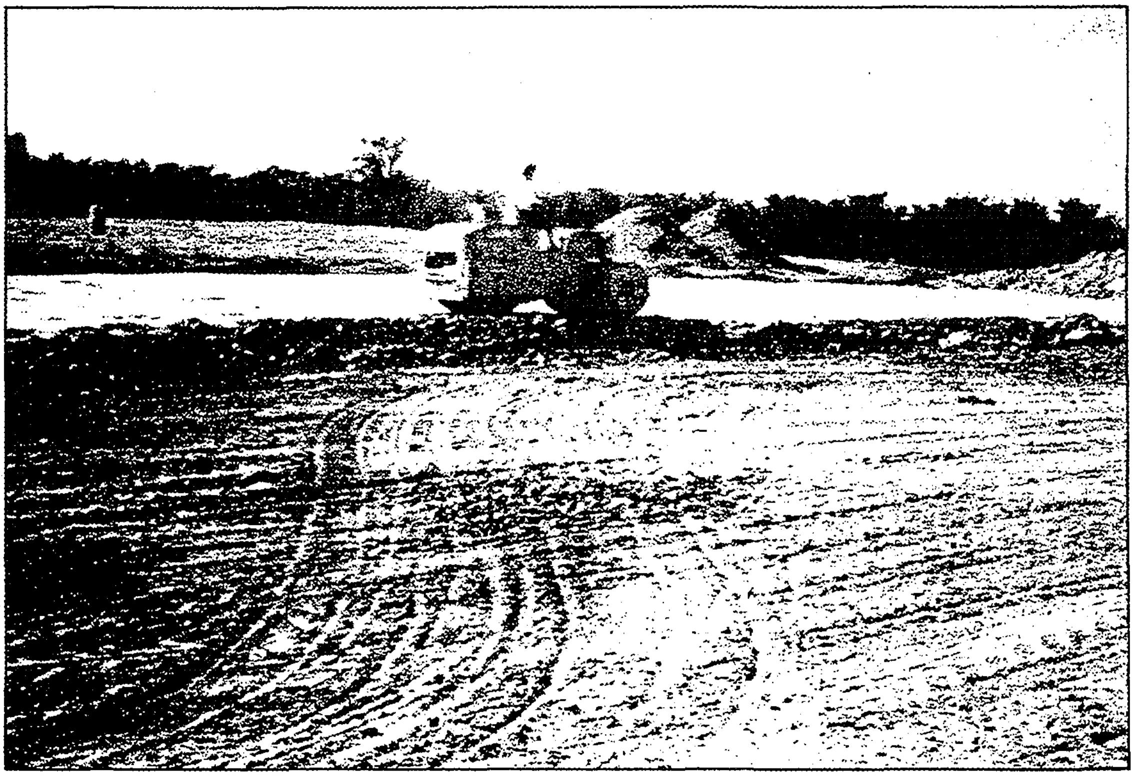

Fig. 6 - Scarification of the first protective clay layer on the top surface

CONCLUSIONS

So far, the contaminated material is well confined inside the mentioned waste disposal, well protected by suitable physical and geochemical barriers, capable of avoiding any contamination in the environment for a very long time.

The methodology used reached the aim of both localizing which part of the waste was contaminated and measuring Cs137: everything inside a great quantity of material.

Moreover, all the actions were carried out in safe conditions and without any (or almost any) contact of the contaminated material with the environment.

The project of the final covers was realized utilizing natural material and conventional and cheap technologies.

The physical multilayers cover has been very well and successfully tested during the last two years by very bad weather conditions.

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TECHNOLOGY FOR RESTORATION OF CONTAMINATED SITES; REVIEW OF AVAILABLE EXPERIENCE IN THE FIELD OF ENVIRONMENTAL RESTORATION IN ROMANIA

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Abstract

In Romania as a result of human activities there are several contaminated sites with low specific activity widely dispersed. Among these uranium mining and milling, fertilizer industry and coal-fired power plants are the most significant. Particularly, for heaps resulting from uranium mining and milling activities the environmental restoration is justified but no comprehensive optimization process (multi-attribute cost benefit analysis) was implemented so far. The lack of specific technology and necessary financial resources are major problems in the implementing of environmental restoration work. Regarding contaminated sites from the fertilizer industry and coal-fired plants the justification for restoration is not complete. The same difficulty is foreseen with respect to available technology and necessary funds. However, a number of research projects are under development and could be considered in the light of international co-operation programmes. Limited remedial actions were performed in order to do something with respect to smaller areas of contamination with higher specific activity.

1. PRELIMINARY

In Romania there are a number of important issues related to environmental restoration:

(i) regulatory and scientific aspects; the most important problems are:

a. a clear definition of contaminated sites in terms of specific activity and/or effective dose;

b. the definition of action levels in terms of specific activity, effective dose or risk values. A definition of action levels in terms of risks appears to be more appropriate because other associated non-radioactive risks are involved. Moreover the heaps from uranium mining and milling activities present potential exposure, which is defined in terms of risk limits and risk constraints, rather than occupational exposure which is associated with dose limits and dose constraints [1]. In the same way actual regulations [2] offer only general principles on closing of the extraction activities for raw materials;

c. guidelines for remedial action plan.

(ii) organizational problems associated with a diversity of structures involving human activities which could lead to environmental restoration. Due to the lack of national and international regulations it is hard to have a consensus on this subject.

(iii) available technology and funding of environmental restoration work. Mainly, as a result of uranium mining and milling, the fertilizer industry and coal-fired power plants we have widely dispersed contamination with low specific activity. This situation led to large areas and volumes of low activity contaminated soil which requires specific technology and large financial resources.

Presently, there is a lack of funding for the remediation of contaminated sites.

Concerning higher specific activity in smaller areas of contamination, limited remedial actions were performed in order to clean-up an old facility used for temporary storage of radioactive waste. Its use was due to the lack of a repository.

2. ENVIRONMENTAL RESTORATION RELATED TO URANIUM MINING AND MILLING ACTIVITIES IN ROMANIA

2.1. Review of actual situation

Presently, there are about 173 contaminated sites as a result of uranium mining and milling, containing about 5,350,000 waste rocks (annually predicted quantity is 300,000 tonnes), and 30,400 tonnes low grade uranium ore (ore heaps) [3].

The distribution of the waste rocks and the ore heaps is summarized in table I, [3,4,5,6]. The radiological characterization and the restoration criteria laid down for each side is offered in table II, [4,5,6].

TABLE I

The distribution of waste rocks and ore heaps in Romania

Area	Type	No. of heaps	Surface (m2)	Volume (m3)
Bihor	ore waste rock	5 8	16,600 529,300	18,960 1,251,100
Banat	ore waste rock	1 17	1,000 209,500	7,000 1,817,100
Crucea	ore waste rock	- 29	- 364,000	- 1,390,500
Tulges	ore waste rock	3 75	1,400 156,847	2,898 414,987
Alba-Tulia	ore waste rock	1 32	500 122,300	1,500 472,142
Feldioara (*)	mill tailing	2	350,000	1,750,000

(*) Milling Plant

TABLE II

Radiological characterization of contaminated sites following mining and milling activities in Romania

Area	Uranium (%)	Dose rate min-max x E-o6 Sv/h	Present waste condition treatment	Forecasted measures	Funds needed x E+O3 $ 1990
Bihor	0.03 - 0.04	0.10-10	no	vegetal soil strata	500
Banat	0.02	0.12-10	no	vegetal soil strata	150
Crucea	0.02 - 0.04	N (*)	no	vegetal soil strata	250
Tulges	0.01-0.02	N (*)	no	vegetal soil strata	100
Alba-Tulia	0.02	N (*)	no	vegetal soil strata	n (**)
Feldioara	0.02-0.04	n (**)	no	vegetal soil strata	500

(*) No recent measurements

(**) No available information

2.2. Related technology for environmental restoration

The contaminated sites following uranium mining and milling activities are characterized by widely dispersed low specific activity. Thus very large areas and volumes should be considered in the environmental restoration plans. So far technology used for environmental restoration of the heaps is an extension of classical equipment adopted to radiological conditions. In the past the low specific activity of these heaps was considered an argument for simple remedial plants and classical technology usage. The lack of national regulations with respect to action levels at which remedial actions should be implemented was a reason for doing something. In the same connection it is worth mentioning the lack of international guidelines in this respect. For instance at Barzava for a low activity uranium ore heap, classical technology was adopted for covering it with a 5 cm thick concrete layer to prevent wind and rain corrosion. After a while the concrete cracked in some places and following these the gamma dose rate of 9 mSv/h was measured (as compared to 1-2 mSv/h over the concrete shield and 0.17 mSv/h in the background in the nearest village).

These facts raise the question: for how long a time are these measures appropriate? As time passes the greater the uncertainty becomes. So, variation over thousands of years in the meteorological conditions, population distribution, natural disasters and human intrusion make our attempts quite difficult. Besides the money needed for clean-up these sites are not available now since other sectors required large amounts of funds.

2.3. Current trends in research and development

Generally, technology is not available and the lack of financial resources precludes large environmental restoration projects.

Currently, there are no national action levels established for remedial actions related to environmental restorations. Moreover, there is no agreed upon definition of a contaminated site. However this situation could be reconsidered through international co-operation programmes: IAEA's Coordinated Research Programmes (CRPs) and Technical Assistance Projects.

On the other hand a number of national research programmes are under development. Some of these are related with the covering of the heaps resulting from the mining and milling industry with a vegetation strata. This should include the following steps:

(i) justification: at this stage the environmental restoration is justified for most of the heaps following mining and milling activities,

(ii) optimization: multi-attribute cost-benefit analysis should be carried out

(iii) safety assessment of the site with specification of:

a. possible internal events within the heaps
- structural failures
- consolidation
- radon emanation
- direct radiation
- chemical hazards

b. possible external events affecting the heap
- human intrusion
- water erosion
- wind erosion
- earthquake

c. remedial action plan including
- stabilization and consolidation of the heaps
- excavation of nearby contaminated soils and placing them on the tops of heaps for radon flux reduction
- choosing layers for control of erosion, infiltration and radon. The top layer will be vegetation strata which can reduce the visual impact of the remediated heap.

In addition to this research effort there are some regulatory and design aspects:

(i) The Regulatory Authority should lay down clear concepts on what comprises a contaminated site and what is the action level at which remedial actions should be implemented.

(ii) International agreement on action levels could be quite helpful in the future.

In the new International Basic Safety Standards only action levels for radon are offered.

3. ENVIRONMENTAL RESTORATION RELATED TO CHEMICAL PHOSPHATE FERTILIZER INDUSTRY

3.1. Review of actual status; related technology and trends in research and development.

It is recognized that by-products of the fertilizer industry enhance the public exposure. The main radioactive by-product is phosphogypsum. This is produced in large quantities (from 1 tonne of raw material 600 kg phosphogypsum are produced).

In Romania 500,000 tonnes are annually produced. Four main sites are candidates for environmental restoration, table III. This problem consists also in widely dispersed low-level activity. However the heaps are located near population centres and thus remedial action could be justified.

TABLE III

Contaminated sites from chemical phosphatic fertilizer industry in Romania

Area	Amount (mil.t)	Surface (ha)	Specific activity Ra (Bq/g)
Valea Calugareasca	5	29	0.319
Navodari	4-5	40	0.340
Bacau	5	17	0.530
Turnu Magurele	5	20	0.481

No site-specific remedial action plan has been prepared so far. Anyhow, action levels must take into account:

- the individual and collective exposures,
- the radiological and non-radiological risks, and
- the financial and social cost.

If remedial actions are justified then further studies on ecological impact and cost-benefit analysis are expected. In any case financial resources are limited and only some research studies are under development, so far. Of course, there is no special technology at hand.

4. ENVIRONMENTAL RESTORATION RELATED TO COAL-FIRE POWER PLANTS

4.1. Review of actual status; related technology and trends in research and development.

The remedial actions concerning the deposition of the fly-ash due to elimination of organic compounds in the thermoelectric power plants is not yet justified. This case is also one of widely dispersed low specific activity (U-238, Ra-226, Th-232, K-40).

A comprehensive research programme [8] has been implemented since 1983 around a number of both old and modern power plants. The individual and collective dose received by the population in these areas were assessed. For instance the collective effective committed dose was found between 76 man x Sv/GW annually and 0.24 man x Sv/GW annually.

If remedial actions are justified further studies on ecological impact and cost-benefit analysis are expected. In any case financial resources are limited and only some research studies are under development, so far. Of course, there is no special technology at hand.

5. CONCLUSIONS

1. The main concern with respect to environmental restoration is widely dispersed low specific activity due to uranium mining and milling activities; there are 173 contaminated sites placed in 6 main regions. The remedial actions are justified in this case but the lack of special technology and financial resources are major problems in the implementing of environmental restoration work. However, a number of technologies are under development and could be improved and applied taking into account good experience from other countries. Thus, IAEA's CRPs and Technical Assistance Projects are welcome in order to make the remedial actions as cost efficient and successful as possible.

2. International Organizations involved in environmental restoration as well as Regulatory Authority should provide guidelines on how to define contaminated sites and related action levels.

3. For contaminated sites, the overall problem should be considered including non-radioactive risks.

4. Fertilizer industry and coal-fired power plants release large amounts of by-products and fly-ash with increased concentration of radioactive nuclides. Low specific activity is widely dispersed around the facilities. Environmental restorations still needs justification, and a number of projects are under development.

5. A number of regulatory and scientific aspects should be defined in the future such as the contaminated site concept and action level values.

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TECHNOLOGIES OF ENVIRONMENTAL RESTORATION IN RUSSIA

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Abstract

In present study the attempt is undertaken to compile, systematize and analyze the data on technologies of radioactively contaminated sites restoration. The principal attention is paid to the methods and tools already employed in the practical activities. Some generalized considerations and suppositions on the subject are also presented.

1. INTRODUCTORY REMARKS

The environmental legacy of industrialization and the "cold war" strongly influences both the everyday life of inhabitants and economy of the countries as a whole - that's a truism now. As to the impact of "atomic rubbish," this fact became an absolutely obvious and even threatening after 1986: to some extent because of Chernobyl catastrophe, directly or indirectly affected predominant majority of the countries; but mostly because of radical changes in the world political and morale-psychological climate (see in detail [1-3]). In such or another way the curtain of secrecy, which concealed the data on the facts associated with the great and often uncontrolled radioactive contamination of many sites, was raised, and the problem of nuclear legacy came up in a practical plane. The first and the natural reaction of the public, specialists and officials was to restore radioactively contaminated sites a.s.a.p. As a result, in many countries, including Russia. ambitious environmental restoration programs have been developed and approved on the governmental levels as a high priority activity.

However, very soon it became understandable that these programs require such a great expenditure of material and financial resources which are not available even in the highly developed countries. For example, representative of US DOE Mr. C. Frank recently reported that remediation of nuclear weapons production sites in USA will require $600 billion for 30 years. According to [4] the costs to vitrify nuclear wastes in the former USSR alone will run about $ 500 million annually in the next ten years, using local personnel, etc.

Enormous cost of site remediation and the financial risk of environmental liability presented more than serious barriers to cleaning up contaminated properties

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and encouraged responsible institutions to build upon undeveloped open space, rather than clean up and reuse these sites.

Thus, initial euphoria was rapidly replaced by very restrained expectations, if not by full skepticism in relation to the original plans of environmental restoration.

However, it looks as if situation can be changed in the nearest future and, at the first glance -- quite unexpectedly. Economics again is a key point of the proofs adduced, but from absolutely opposite point of view. To put it briefly, there exists opinion that environmental restoration technologies should be viewed as a stimulus, rather than an obstacle, to economical revitalization [5,6]. The proponents of this concept give an impressionable figures and facts [5-10]:

- the global market for environmental products and services has been assessed as $ 240 billion in 1993, and it is expected to reach around $600 billion by the year 2000;

- DOE alone has 1.5 million tons of radioactive scrub metal in storage around the USA, a contaminated heap that is expected to grow at least tenfold as nuclear weapons facilities are decommissioned. For example, the old Oak Ridge GD Plant contains $ 400-million worth of contaminated nickel to salvage, millions of dollars worth of copper, steel and other metals, and enough concrete to lay a roadbed from New York City to Los Angeles;

- each 1100 MW light water reactor operated for its full 40-year license life, will generated 18,000 m^3 of contaminated metals and concrete, about 98% of which will be the least radioactive form of low-level waste, making this a market ripe for companies with market - tested cleanup and recycling technologies to deploy;

- the OECD reported Germany to be the leader in environmental exports with a 1992 surplus of $ 10 billion, followed by the USA with a $ 4 billion and Japan with $ 3 billion. The conference on Asia-Pacific Economic Cooperation recently made stimulating green industries a linchpin policy, stating that "...sound environment and sound economic policies are mutually supportive" and that "preventing environmental degradation is fundamental to sustainable development".

Summarizing all the above described, it is logically to make up a question: what is environmental restoration? The heavy burden of objectively forced, time and money - consuming technological activities, or profitable business? Apparently, the truth is somewhere in between of these two extreme points of view. The ultimate answer depends, among others, on the global political climate, and on the concrete combination of technological, economic, environmental and political variables in each country. It is not difficult to predict that the rates and effectiveness of environmental restoration activities will differ from country to country, and that these differences will be intensified in due course. Commercialization of technologies gives to the "rich" countries an advantage of significant opportunities abroad - opportunities to test environmental technologies; opportunities to buy or license foreign technologies; and opportunities to market their environmental technologies and expertise in other countries [6]. On the other hand, countries with so called transfer economy but with good scientific potential obtain, in principal, an opportunity to sell technologies and know-how, or to lease an experimental ranges for technological trials, and by this way - to improve both economic and environmental conditions.

If so, one could formulate the following reasonable suppositions:

(1) there are some grounds to expect that in the not very distant future activities on rehabilitation of radioactively contaminated sites will obtain a new impulse for development;

(2) apparently, technologies of environmental restoration will become (or already became) a subject of commercial secret, and it's unlikely that delicate technical details will be widely available. This is the more so that a number of technologies employed in environmental restoration have been enlisted from the military programs, and such data are classified up to now.

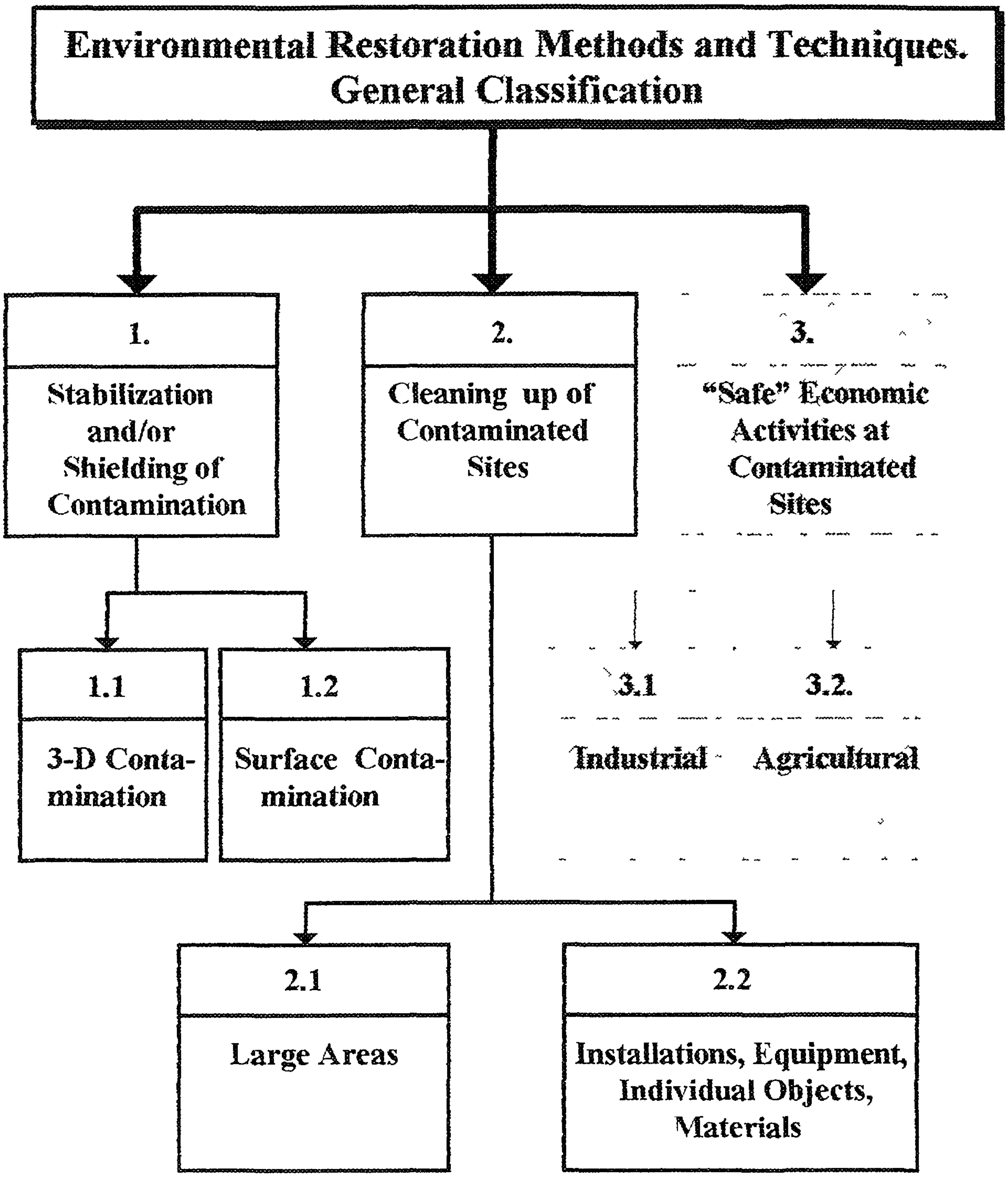
These trends have to be taken into consideration under attempts to analyze present situation and to evaluate technological prospects of environmental restoration

2. GENERAL CLASSIFICATION OF ENVIRONMENTAL RESTORATION'S TECHNOLOGIES AND RUSSIAN EXPERIENCE

With methodical purposes it is reasonably to systematize all known technological approaches to the practical realization of environmental restoration projects. Multitude of technologies being employed can be divided into three principal groups (see Scheme 1).

Inclusion of the 1-st and the 2-nd groups in proposed scheme should not give rise to objection - it's logical and well substantiated step. Group 3, at the first glance, has nothing to do with the case, and apparently some explanations are needed.

The key point is that environmental restoration is aimed at ***reducing or eliminating the risk to human health - in the first place,*** and to environment - in the second turn. Complete cleanup of contaminated site should never be considered as the end in itself. The problem of human health, including moral and psychological aspects, has to be a corner-stone of any responsible decision. In specific, this question is extremely important for the territories affected by Kyshtym and



Scheme 1

Chernobyl disasters. It was shown, for example, that evacuation of local inhabitants from the settlements with surface activity of ^{137}Cs around 40 $Ci \cdot km^{-2}$ will allows to prolong their life for ~35 day at the expense of the dose reduction, while up to 8 years (!) will be lost as a result of forced resettlement [11] In the same context one could view serious criticism in respect to the active environmental intervention undertaken by developers in Kyshtym region [12] So, do we like it or not, but today hundreds thousands people in Russia live at radioactively contaminated territories.

And these people must live as safely as possible. Such possibilities are (or should be) ensured by the complex of special measures including some technological methods. This is not a technology of rehabilitation in the true sense of this word . Nevertheless, so far as it concerns the main problem of environmental restoration - the problem of human health, ***both safe economic activity at contaminated sites and environmental restoration of these sites can and have to be analyzed in the same framework.***

Before proceed to further detailing of the Scheme 1, let us note the following features:

- 3 - D contamination (sub-group 1.1), in practice, have to do with uranium mines and natural and engineering reservoirs filled by the liquid radioactive waste;
- surface contamination (sub-group 1.2) means the case when radionuclides are distributed in the spatial layer of soil or other material of the finite and enough small ($n{\cdot}10$ cm) thickness.

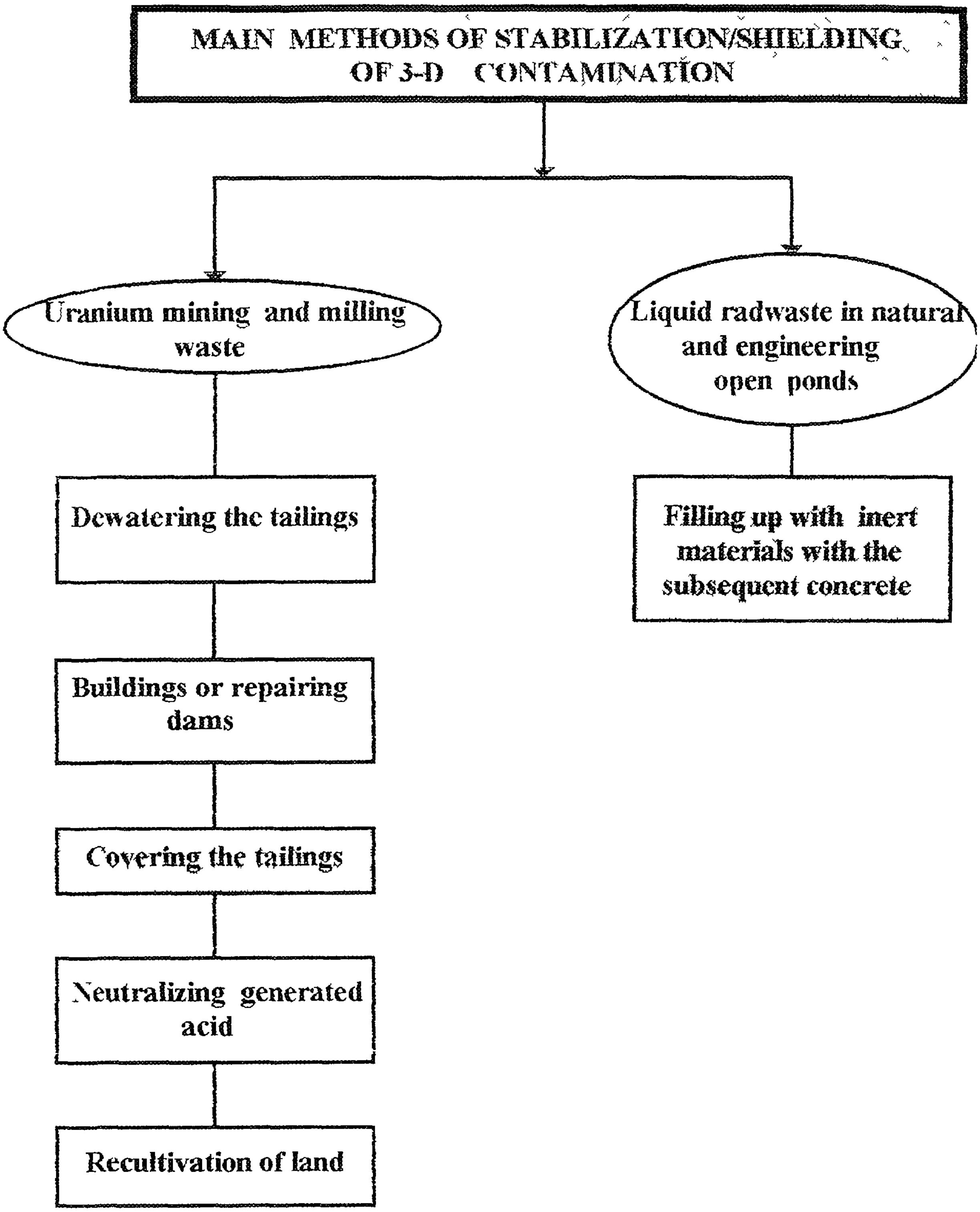
2.1.1. STABILIZATION AND/OR SHIELDING OF 3-D CONTAMINATION

Decontamination of uranium mining debris and mill tailings is nonsense. Low-level uranium waste at production sites should be stabilized and reliably isolated from the biosphere. The following methods are applicable for these purposes (see, also Scheme 2):

- deswatering the tailings;
- building/repairing dams;
- covering the tailings;
- neutralizing generated acid;
- recultivation of territory.

These technologies with some variations are used in all the countries involved in uranium business.

As regards the Russian Federation, situation is the following. At present only two uranium production centers are in operation: Production Association "Almaz" near Lermontov (Northern Coucas) and Priargun Mining - Chemical Combinat (Eastern Siberia) [13]. At the same time a few scores of abandoned uranium enterprises, mines and open pits are distributed all over the country. The tailings in a part of them are covered with a several layers of isolating materials - mainly with shingly - sandy mixtures and clay. As usual, the thickness of the covering materials is ranged from 0.5 to 1.0 m on the top and from 2 to 3 m on either side of the



Scheme 2

tailing pond. However, in a number of obsolete production centers a proper precautionary measures are not undertaken, and information on another mining and milling 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) is not available

Taking into account an importance of the problem discussed, recently special Federal program was elaborated by the All-Russian Institute of Chemical Technology. This program envisages careful inspection of all abandoned production sites with a subsequent development and implementation of a proper environmental restoration technologies. Now this work is in initial stage [13], so that description of the methods and techniques expected to be employed is premature.

Natural and engineering open reservoirs contained a thousands and millions cubic meters of high-level liquid radioactive waste - is another type of 3-D contaminated sites in Russia [14,15]. Sophisticated technologies of waste processing are not applicable in this case because of enormous volume, unstable and unregulated composition of radioactive water and some other reasons. The only possibility to improve radiological situation is mechanical stabilization and shielding of reservoirs contents.

This labor - consuming work has been started 1.5 years ago at the radiochemical combinat "MAYAK". Natural lake Karatchai contained 400,000 m^3 of radioactive waste with a total activity around 120 MCi is gradually filled up with a broken brick and then is covered by concrete. It is difficult to predict now when the all ponds in Tchelyabinsk and some other production sites will be shielded (if it is possible at all), and whether the ultimate goal will be reached.

2.1.2. STABILIZATION AND/OR SHIELDING OF SURFACE CONTAMINATION

Stabilization and shielding of contaminated surfaces can be applied as an effective technology practically for any kind of materials/installations in various conditions. However, in the most cases, the application of surface stabilizers is a short term action which requires further decontamination. In emergency conditions of radiological accident stabilization of radionuclides on soils, buildings, roads, etc. is the most essential initial measure.

All the methods of stabilization can be divided into three classes (Scheme 3). The methods used by Russian specialists are reflected in shaded boxes of the scheme.

Chemical technologies elaborated by NIKIMT, VNIIChT, GIPCh and IPhCh, have been employed at Chernobyl site and Bryansk region mostly for dust fixation. A large variety of chemicals has been tested for these purposes. The best results were received with compositions on the base of latex and with water-resistance sulfit-cellulose barda.

It should be noted that chemical fixatives (in the form of removal coatings) were effectively used for surface decontamination as well. In specific, polyvinilbutiral (VL-85-03-77) with additives of TCPAA, plastificators and anti-adhesive substances (EAP-30, KEP-2A, OP-7, OP-10) provided decontamination factor ~2.0. Mixture of PVA+HF allows to remove fixed radioactive contamination with factor 20, etc.

As to mechanical stabilization of surface contamination all stabilizers enumerated in Scheme 3 have been employed with a greater or lesser success. The proper choice depends on many factors, including availability of a certain material, the level and the type of contamination, economics, etc.

From the three physical methods mentioned in the Scheme 3, only one - electrokinetic - has been employed in the national practice. In electrokinetic stabilization/remediation, electrodes are implanted in soil, and a direct current is imposed between the electrodes. The application of direct current leads to a number of effects: ionic species and charged particles in the soil - water solutions will migrate to the oppositely charged electrode (electromigration and electrophoresis), and contaminant with this migration, a bulk flow of water is induced toward the cathode (electroosmosis). The combination of these phenomena leads to a movement of contaminants toward the electrodes (Fig.1). As a result, radionuclides in various form concentrate in a local space near electrode (**stabilization**), and their concentration between electrodes is diminished (**remediation**)*.

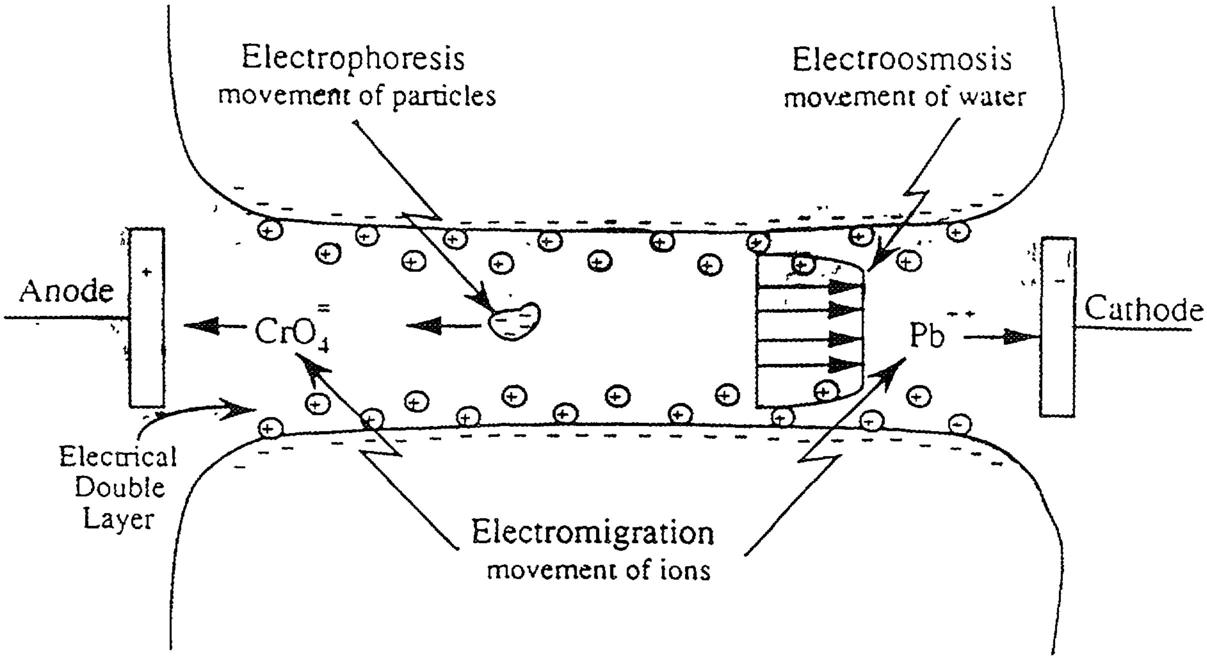
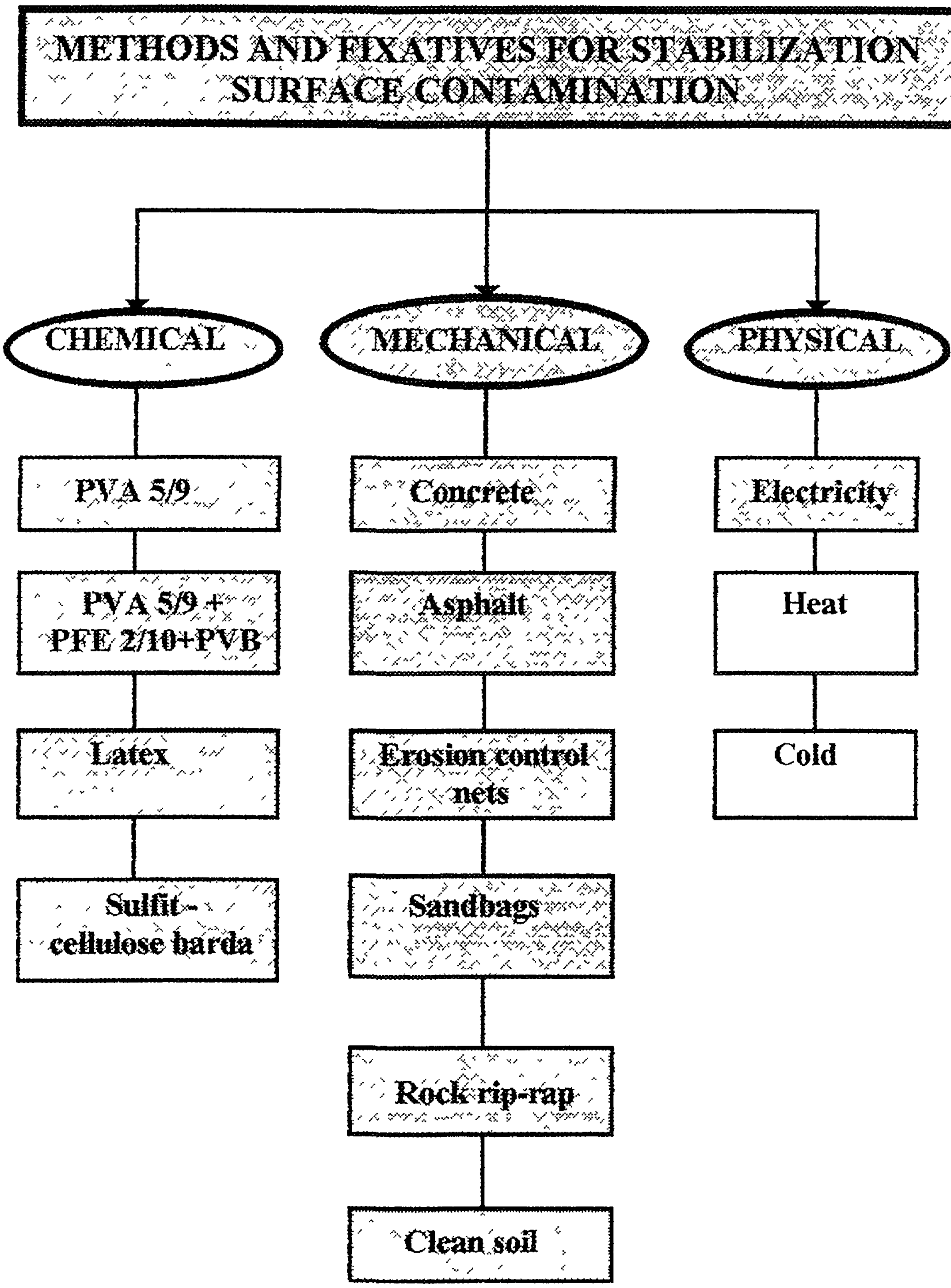


Fig.1. Electrokinetic phenomena pertinent to *in situ* stabilization/remediation. Credit to Lingren and Kozak (1993)

* Potentially contaminants arriving at the electrodes may be removed from the soil by one or several methods, such as electroplating at the electrode, precipitation or co-precipitation at the electrode, pumping of water near the electrode, or complexing with ion-exchange resins (restoration) [16,17]



Scheme 3

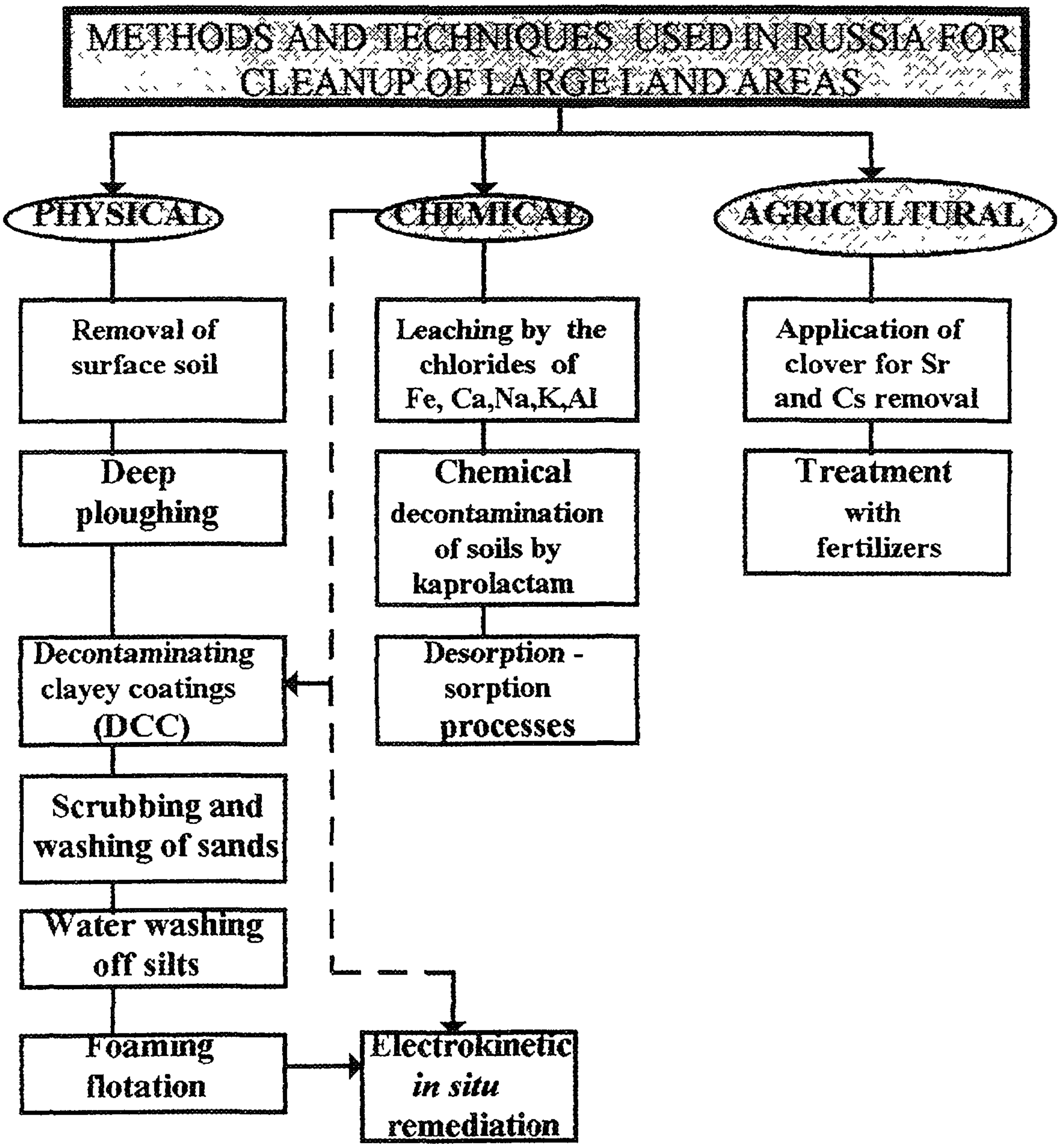
This technology has been successfully tested by the specialists of "Promstroytechnology" at uranium M&M sites, and now an experimental program is underway at SPA "RADON" to determine the feasibility of using electrokinetic processes to stabilize ^{137}Cs in soil.

2.2.1. CLEANUP OF LARGE LAND AREAS

Decontamination of large land areas is one of the most complicated problems in Russia, both from the technical-economic and strategical (risk-benefit) points of view [12]. Although a large variety of technologies are potentially available (Scheme 4), the scope of environmental restoration activity does not correspond to the real requirements.

In practical sense the most attention historically has been paid to ***removal of surface soil and deep ploughing***. These methods have been developed, tested and used more than 30 years ago - after Kyshtym desaster.

Recently a removal of contaminated upper layer of soil was practiced in Chernobyl area and in Bryansk region with employment such "standard" equipment



Scheme 4

as graders, scrapers and bulldozers. As usual contaminated earth was moved into piles and then hauled away in a specially excavated and isolated trenches.

Two problems were met by developers when ***removal and storage of soil*** from large areas was applied for decontamination purposes:

- first, **the problem of safe storage of contaminated soil.** For example, examination of underground waste storage in Nikolaevka (Bryansk region) placed at a pine forest (contamination level 60-70 $Ci \cdot km^{-2}$) with dimensions 45×40 m and formed by a sand showed that underground water radiation level did not exceed PL. At the same time it was demonstrated that in Novozybkov area, where the levels of ground water range from 0.5 to 10 m, the problem of radioactive waste isolation both from ground water and atmospheric falls (to prevent infiltration) is of vital importance and very difficult.
- second, **impossibility of the total territory decontamination by this technology because of enormous volumes of soil removed.** Therefore, in Bryansk region coverage of contaminated soil with clean sand was practiced as a more effective means of decontamination/stabilization, since it does not require its transportation and storage.

Technology of ***deep ploughing*** was extensively applied in Kyshtym and Bryansk region for the restoration of lands used for agricultural purposes.

In Bryansk region nearly 101 200 ha (14.8%) of all agricultural lands are situated in a zone with a level 15-40 $Ci \cdot km^{-2}$, and 17,000 ha (2.5%) have a contamination exceeding 40 $Ci \cdot km^{-2}$. These include 7,300 ha of a ploughed land and 9,800 ha of meadows and pastures.

In 1986-1987 in order to decontaminate agricultural lands they were reploughed. Ploughing lands to a depth of 20 cm reduced radiation dose to 1.5-2 times. The ploughing with a full overturn of a furrow was more effective when radionuclides excluded from a 30-cm ploughed layer. It made it possible to reduce ^{137}Cs storage in harvested plants. By the end of 1990 lands with contamination exceeding 40 $Ci \cdot km^{-2}$ had been banned for agricultural purposes [18].

Nevertheless, deep ploughing, as a method of decontamination, can not be recommended **a priory** for any soil type, any root depth of plants, etc. In each concrete case additional study is required to determine when and if ploughing should be used as a cleanup technology.

Other physical methods included in Scheme 4 can be considered rather as prospective elaboration than market - tested technologies of environmental restoration.

Decontaminating clayey coatings (DCC) on the base of natural clay and clayey minerals are an effective and cheap sorbents, and they are able to form thin films on the surfaces. These properties have been tested in a large-scale in-field experiments with employment of the "standard" military equipment: ARS-14 and ARS-14A. The following compositions were investigated at Chernobyl site:

- DCC-1 incorporated in a "liquid glass" and peat-extractants;
- DCC-2 contained sulfit-cellulose barda;
- DCC's contained humus extractants.

This method has been assessed as an universal, economic, satisfactory in respect to decontamination factor and enough simple in technological sense. However, this approach was not used in a "routine" practice of environmental restoration.

Good results have been obtained in extended experiments on mechanical separation (scrubbing and washing) of the fine sand fraction from the bulk of contaminated quartz sand. Standard equipment for sand classifying have been used. It was shown that around 90% of plutonium is concentrated in the separated fine fraction, and only 5% - in a coarse sand. The mass ratio "sand-humus" in the soil investigated was 2:1.

The specialists of All-Russian Institute of Chemical Technology elaborated and tested in on-site trials prospective methods of radioactive contaminants removal from the soil on the basis of

- ***foaming flotation*** and
- ***washing off silts*** after separation of roots.

In the flotation experiments the fraction with γ -activity 2-5 times less than in initial samples has been obtained with the yield about 70%. Application of hydro-cyclon technology (triple washing of the pulps) made it possible to separate 50-70% of the sand fraction with 25% of initial radioactivity.

As to electrokinetic technology, this subject was already discussed in a previous chapter.

Chemical, or better to say, physico-chemical methods of soil decontamination have been tested on a pilot scale both in Chernobyl zone and Bryansk region. The data on decontamination in a settlement of Swyatsk (Novozybkov district) indicated the purification of soil amounting to 45% when a decontaminating solution effluent was applied. This method of chemical purification was assessed, however, as very expensive in comparison to a physical one, as 1 m^2 costs more than $1 [18]. There also exists another problem associated with potential necessity to process a large volume of the secondary liquid waste, contained both radionuclides and chemical toxins. And finally, it is known that from 40% to 90% of the total amount of ^{137}Cs

(the most important contaminant at the territories affected by Chernobyl fallout)* is practically insoluble in water, strongly fixed by soil and couldn't be mobilized even in hard acid conditions [18,19], what is, undoubtedly, important for evaluation of chemical methods effectiveness.

Therefore, in spite of good results obtained in experiments with metal chlorides, mixture of mono- and bicarbon acids as well as composition on the base of bentonit+peat+HNO_3 +H_2O (desorption - sorption process), it is unlikely that chemical methods will be widely used in a visible future.

R&D on agricultural methods of soil decontamination have been started in the end of 1950s - beginning of 1960s. However, this technique does not appear to bee practical for widespread usage.

As an example it could be mentioned here some experiments with clover which extracts from soil up to 5% of ^{90}Sr and up to 0.5% of ^{137}Cs. But in general, the so called biological or phyto-technology is characterized by relatively low effectiveness and by large volumes of the secondary plant waste, which require proper processing/disposal.

In the same context one could mention some other well known measures applicable for decreasing of the soil-plant transfer of radioactive isotopes (e.g., liming for reducing of ^{90}Sr uptake ; treatment with fertilizers for decreasing the uptake of ^{137}Cs; application of ferrocyanides and bioactive substanses to reduce ^{137}Cs transfer in agricultural products [20, 21], etc.). This subject as well as the problem of the plants selection will be discussed in detail in Chapter 2.3.

2.2.2. DECONTAMINATION OF EQUIPMENT, BUILDINGS AND PAVED SURFACES

Russia has an extensive experience in decontamination of various objects in or adjacent to nuclear facilities during normal operations. These knowledge and techniques have been partly applied to cleanup materials, equipment, buildings and paved surface after radiological accidents.

However, very soon it became clear that "normal" decontamination technologies and compositions, in the most cases, are too sophisticated and too expensive for application on the scale required in an urban environment.

At present this question is solved, at least at practical level. Although, research in this field still continue, there are a number of able-bodied and effective technologies for cleanup of large contaminated areas. So far as technical principles

* In Bryansk region range of ^{137}Cs contamination is 5-9 $mCi{\cdot}m^{-2}$ while ^{90}Sr contamination does not exceed 0.7 $mCi{\cdot}m^{-2}$, and the level of Pu-isotopes activities is no more than 0.01 $2mCi{\cdot}m^{-2}$.

of technologies discussed are almost the same all over the world, only brief factual description is presented below (Scheme 5).

Aqueous methods (based on chemical interaction of decontaminated materials with working solutions) showed low effectiveness for paved surfaces. The *following compositions have been tested in real conditions:*

- preparation SF-2U + $H_2C_2O_4$ + H_2O;
- preparation SF-2U + $H_2C_2O_4$ + EDTA + H_2O;
- 1% HCl
- H_2O + 0.5% OEDFA + (2-5)% H_2O_2

Decontamination factor was a little bit higher than 1.0.

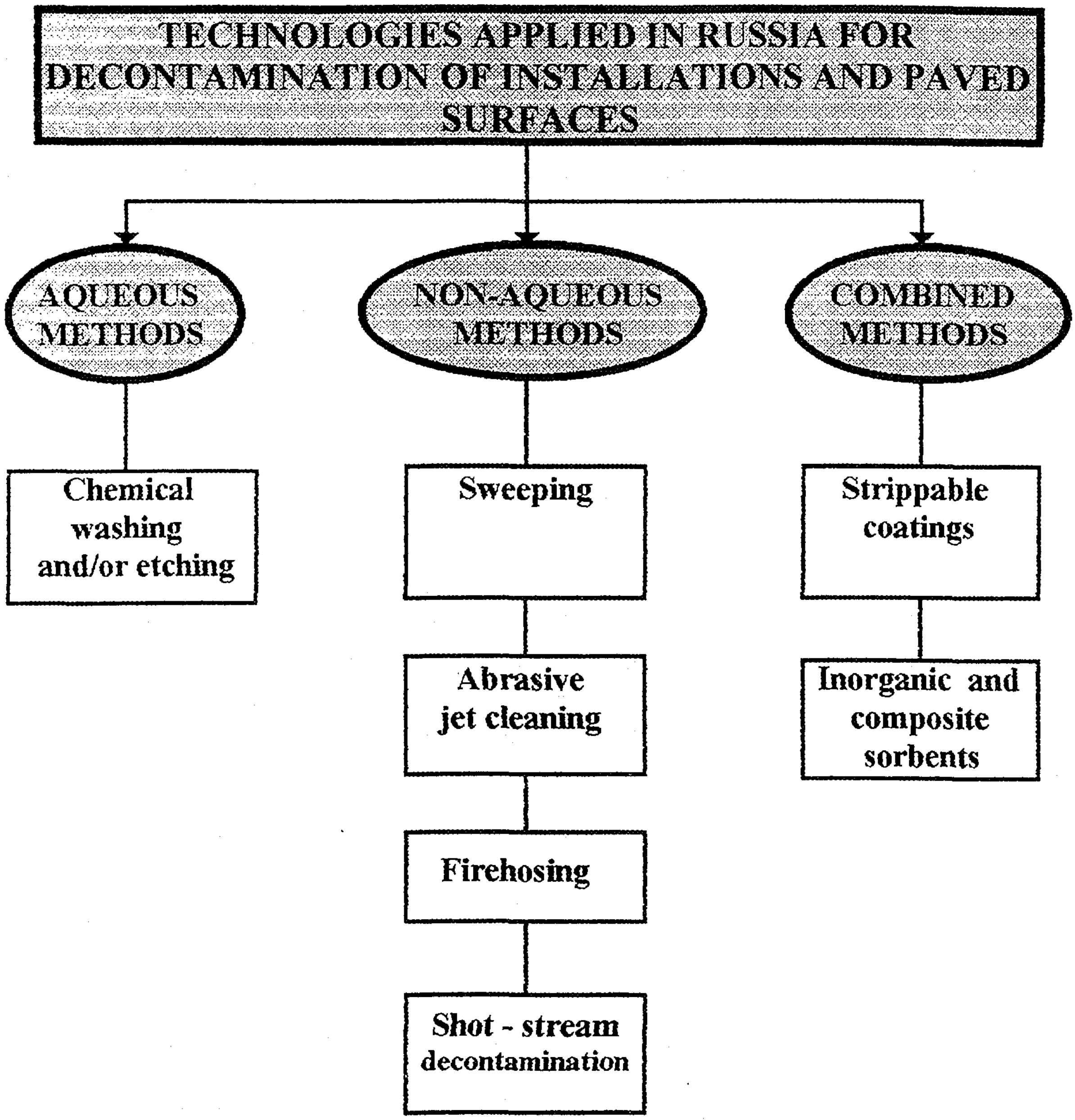
At the same time, aqueous technology has been successfully applied to decontamination of steel and alloys for recycling purposes. The principal parameters of the process are the following:

- decontaminant - (5-25%) HBF_4
- temperature - from 40 to 90° C
- duration of processing - from 4 to 6 hours
- decontamination factor - from 10 to 100.

Non-aqueous (mechanical) methods have been used for cleanup of large areas the most intensively. In sweeping and vacuum sweeping technologies the following equipment were employed:

- facility "Typhoon"
- water-vacuum cleaning machine KU-005
- whirlwind decontamination facility VDU
- industrial vacuum-cleaner PO-11M
- aerosol vacuum-sweeper on the base of the truck KRAZ-256
- equipment of "Nuclear Services".

Abrasive jet cleaning technology has been realized with the usage of sand as an abrasive, and with employment of the standard machine OM-22612. Decontamination factor did not exceed 6.



Scheme 5

Firehosing technology did not give an expected results: decontamination factor ranged from 1.5 to 3.0, when the standard equipment OM-22612 has been used.

Another possibility to decontaminate concrete and brick is application of dust-protective shot-stream equipment BDU-EZM or "Kaskad". Throughput of such machines is around 1-2 $m^2 \cdot h^{-1}$; the depth of the surface layer removed is about 1 mm; decontamination factor ~5-8.

The above described mechanical technologies are simple; relatively cheap; allow to use standard commercial equipment. At the same time, in the most cases heightened attention should be paid to the collection and disposal of radioactive waste arising and to radiation protections of operators.

Strippable coatings and decontaminating clayey coatings, representing the class of so called combined methods, are described in Chapter 2.1. These coatings can be applied easily and quickly to large areas and require minimum equipment and personnel. Loose contamination is trapped during the curing process and removed with the layer which is easy to handle and dispose of. Big advantage of DCC' is that their removal is performed by standard equipment and do not require a manual labor.

In addition to DCC, special compositions contained inorganic desorbents, have been applied for decontamination purposes. They are the following:

- klinoptilolit + potassium silicate + H_2O
- klinoptilolit + $H_2C_2O_4$ + SF-2U + H_2O
- montmorillonit + peat + H_2O.

The mechanism of decontamination is based on adhesive - sorptive - ion exchange properties of compositions. This technology allows to remove contaminants from the cracks, clearances, junctions and even from macropores. Decontamination factor depends on the properties of surface and contaminants, and ranges from 2 to 5.

In conclusion it is reasonable to note once again that decontamination of large objects, equipment and paved surfaces is specific task, and positive experience accumulated in a 'routine" nuclear activities is not always applicable in this case.

2.3.1. INDUSTRIAL ACTIVITIES AT CONTAMINATED SITES

This subject is highlighted here just in provisional term. Any industrial activity is strongly regulated by existing norms and rules of radiation protection. There are no exclusions from these regulations even at accidentally contaminated territories. However, conversion of the defense industry can leads to necessity (or desirability) to organize new "non-nuclear" works at the former military or nuclear fuel cycle facilities. For Russia such situation is more than realistic. If so, the question will raise about the limits of occupational dose for the personnel of new plants, and consequently - about special procedures of work, about special privileges for staff, and about additional efforts on environmental restoration of these sites. We must be ready to meet such problems and to solve them properly.

2.3.2. "SAFE" AGRICULTURAL ACTIVITIES AT CONTAMINATED SITES

This unusual but very actual issue has already been mentioned in Chapter 2.1. The first forced and extensive experience in this area was gained after Kyshtym nuclear accident. Unfortunately, this experience come in handy to us again, when a large territory of the country was contaminated in consequence of Chernobyl fallout.

Some common actions on restoration of lands used for agricultural purposes have been described in Chapter 2.2. In present chapter the main attention will be paid to the agro-ecological issues aiming at providing safety of the commonplace life of villagers. All the data presented are compiled from the reports and publications connected with the program of many years study at the territory of Eastern-Ural Radioactive Trail. Recently these activities as well as conclusions and recommendation formulated have been called in question and even subjected to serious criticism (e.g., see [12]). Nevertheless, we hope that these data will be interesting and useful for the unprejudiced reader.

Some notion about the real radiological situation can be obtained from the Tables 1-3 [22].

The most significant isotopes is ^{90}Sr. Therefore all semi-empirical correlation were obtained namely for this long-lived radionuclide.

Behavior of ^{90}Sr in the system "soil-plant" is seriously influenced by the concentration of Ca in soil. When concentration of Ca amounts 2-8 $g{\cdot}kg^{-1}$, factor K_p is connected with Ca-concentration by the following equation:

$$K_p = 0.162 \exp\{-0.259[Ca]\}, \qquad (1)$$

where K_p = concentration of ^{90}Sr in grain/concentration of ^{90}Sr in soil; [Ca] - concentration of Ca in soil.

TABLE I. Soil and Vegetation Contamination Dependence on the Distance from Release Place [22]

Inspected Object	Distance from Place of Release, km					
	12.5	18	23	35	55	75
Soil, $MBk{\cdot}m^{-2}$	1,100	930	740	170	40	5.6
Grass, $MBk{\cdot}g^{-1}$	360	340	100	26	6	1
Pine-Needles, $MBk{\cdot}g^{-1}$	-	-	90	-	-	-

TABLE II. Types of Contaminated Area [22]

Type of Land	Level of Contamination, $MBk{\cdot}m^{-2}$			
	>3.7	>0.37	>0.037	>0.0037
Forests, km^2	120	160	560	9,200
Virgin Soil, km^2	40	80	280	4,600
Arable Land, km^2	120	160	560	9,200

TABLE III. Dose Rate Dependence on the Distance from Release Place One Day after Accident [22]

Distance from Release Place, km	Dose Rate, $R \cdot h^{-1}$
0.1	360
6.0	2.5
12.5	1.4
18	1.1
23	0.6
35	0.09
55	0.002
75	0.002
105	0.001

Type of the soil influences K_p in the following way:

$$\lg K_p = -0.264[Ca] + 0.0086h - 0.027pH + 1.66Z, \qquad (2)$$

where h - concentration of humus, %; Z - concentration of silt-fraction, %.

Analysis of the data presented in Tables 4 and 5 allows to conclude that

(i) concentration of Ca is the main factor influencing uptake of ^{90}Sr by the cereals;

(ii) average value of K_p for cereals is equal ~0.7.

For potatoes and vegetables these correlation differ from those for cereals (see Table 6).

For the system "soil-grass" corresponding concentrations of ^{90}Sr are correlates as follow

$$[Sr]_{grass} = 0.5[Sr]_{soil}.$$

It means that for the fattening of the beef cattle and dairy cattle the natural pastures and meadows with the levels of contamination around 7-10 $Ci \cdot km^{-2}$ and up to 2 $Ci \cdot km^{-2}$, correspondingly, can be used safely.

On the basis of experimental data, and taking into account the factors of ^{90}Sr transfer in a different food chains (see Table 7), it was calculated that at the territory with the reference contamination level 1 $Ci \cdot km^{-2}$ concentration of ^{90}Sr in a "normal" daily ration will amounts 225pCi.

According to the factual data, at the 3-d year after accident each inhabitant obtained with food around 300 pCi of ^{90}Sr daily. National regulations limit daily uptake of ^{90}Sr by 800 pCi. On the basis of these requirements the territory of EURT has been divided in the following zones:

(i) sanitary-protective zone - > 4 $Ci \cdot km^{-2}$

(ii) zone A - 4-2 $Ci \cdot km^{-2}$

(iii) zone B - 2-1 $Ci \cdot km^{-2}$

(iv) zone C - < 1 $Ci \cdot km^{-2}$

In respect to the "safe" agricultural activities the following official recommendations were formulated:

Plant-growing

Zone A: cereals (fodder grain and seeds), grass for seeds

TABLE IV. Average Values of Parameters in Equation (2) [22]

Parameter	lg K_p	[Ca], $g \cdot kg^{-1}$	h, %	pH	Z,%
Average Value	-(1.38±0.38)	4.2 ± 1.4	4.3±1.5	6.1±0.7	4.9±1.2

TABLE V. Average Values of K_p for Some Cereals [22]

Crop	Type of Soil	
	Gray Forest	Black Soil
Oats	0 09	0.06
Barley	0.08	0.05
Rye	0.08	0 05
Buckwheat (Corn)	0.12	0.10

TABLE VI Concentration of ^{90}Sr in Potatoes and Vegetables on the Soils with Contamination Level 0 037 $MBk \cdot m^{-2}$ [22]

Crop	Concentration of ^{90}Sr, $Bk \cdot kg^{-1}$
Potatoes	0 74 ± 0.10
Cabbage	9.2 ± 0.6
Beetroot	11.1 ± 0.8
Cucumbers	7.4 ± 0.4
Tomatoes	5.6 ± 0 4
Onion	14.8 ± 1.1
Carrot	16.6 ± 0.8

TABLE VII. Factors of ^{90}Sr Transfer in a Food Chains [22]

Chain	Factor
Soil - Grain	0.1
Soil - Hay	1.3
Soil - Milk	0.14
Soil - Meat	0.25
Water - Fish	25.0
Soil - Ration	0.25

Zone B: cereals (fodder grain and seeds), grass for seeds + cereals for food and fodder crops

Zone C: unrestricted agricultural activities.

Cattle - breeding

Zone A: dairy stock-raising with processing of milk in butter; pasture of younger animals; pig-breeding and poultry-raising (chickens only)

Zone B: liked in zone A + pasture of dairy cattle and laying-in of hay

Zone C: laying-in of the coarse fodder and pasture of the dairy cattle.

These recommendations have not been realized in practice because of organizational reasons. However, this comprehensive study has shown that **it is practically possible to use large areas contaminated with ^{90}Sr up to 100Ci·km^{-2} (?!) for agricultural purposes without any special restoration measures** [22].

3. CONCLUSION

In Russia, environmental restoration of radioactively contaminated sites is an urgent and high-priority task for Government and other responsible authorities. The Federal program's main challenge is to balance technical and financial realities with the public expectations and develop a strategy that enables the Government to meet its commitments to the Russian people. High scientific level of R&D and large practical experience in this field provide good basis for such projects. However, at present the scope of environmental restoration activities do not correspond to the real requirements. The main reason is a chronic deficit of financial resources. For the time being it is very difficult to predict situation even for 1-2 years. Taking into account the fact that the same problem (maybe not so severe) exist in other countries of Central and Eastern Europe, one can suppose that mutually beneficial international co-operation is the best way out of present difficulty.

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TECHNOLOGIES FOR AND IMPLEMENTATION OF ENVIRONMENTAL RESTORATION PROJECT IN THE SLOVAK REPUBLIC

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Abstract

This paper represents the logical continuation of those presented at the Budapest and Piestany Worshops. After finishing the monitoring activities in the contaminated site near Bohunice Nuclear Plant the need for reconsidering the old restoration project arose. To solve this task new principles for the evaluation of remedial measures was developed in close cooperation with the national hygiene authorities. This principles as well as the resulting evaluation and proposal of a justified extent of the contaminated banks restoration are described and discussed in the paper.
The re-evaluated extent of the banks restoration project include removing and safe burial of about 1100 m^3 of contaminated soil from, and overlaying by clean soil cover on about 10 000 square meters of contaminated flat area of thes banks. The total cost can be estimated by about US \$ 100 000.

1 INTRODUCTION

In Slovakia, no direct experience with the implementation of techniques for a restoration of the environment exists. These problems, however, have been continuously addressed at the design level since 1991 in relation to identification of a ^{137}Cs contamination during preparation of a flood control project implementation for an unengineered bank section of the Dudváh River (from Bučany to Trakovice).

This section with the length of 2.5 kilometers would extend the engineered downstream section (regularly widened and deeped bank profiles) of the Dudváh River (Fig.1), and the protection against flooding in Trakovice village would thus be resolved.

However, ^{137}Cs contamination was identified not only in this section, but also on the banks of Manivier canal having discharged released water from the Bohunice Plant A1 into Dudváh. A design for removal of the contaminated soils from the banks which was established subsequently thus included also Manivier canal with the expected volume of disposed soils of approximately 5000 m^3.

Planned decontamination of the banks corresponded to the character of the contaminated places (steep slopes of irregular banks, 90% of ^{137}Cs activity concentration in 15-20 cm thick surface layer of soil) and to the working limit specified ad hoc by the value of 1 Bq^{137}Cs/g of soil. Prepared design included a surface disposal facility on the Bohunice site the parameters of which were described in a previous work [2].

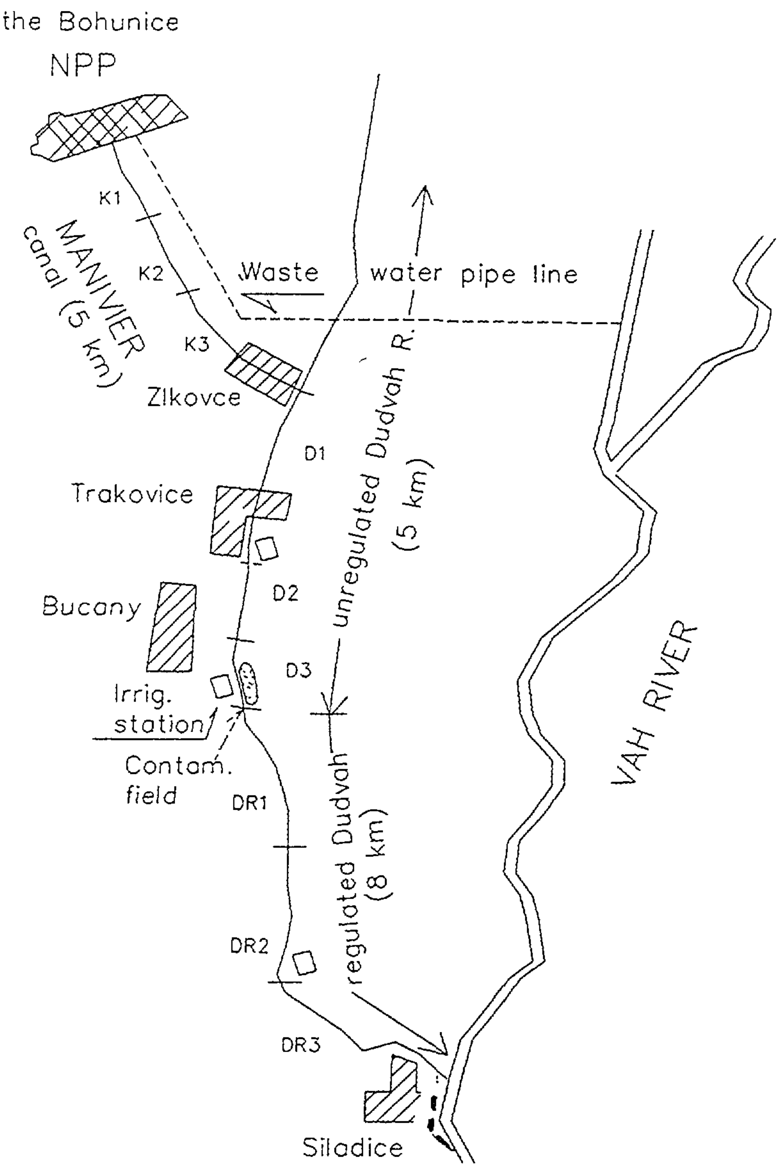

Fig. 1.a. Scheme of the water system taking out the waste water from Bohunice NPP to the Vah River.

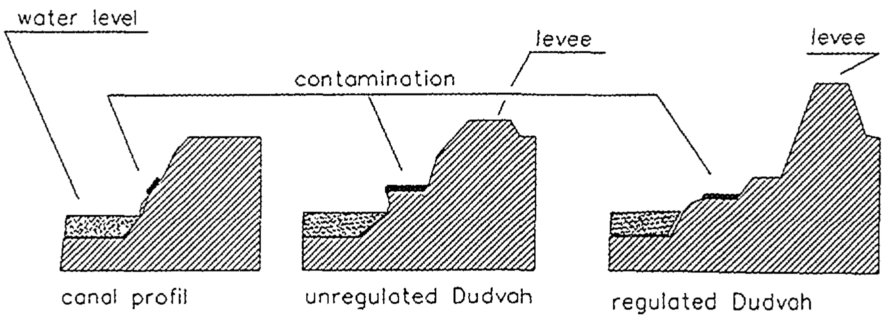

Fig. 1.b. Width profiles and the location of contamination in typical sections of flow.

It was found in the course of approving the construction that the most significant problems of the construction relate to the disposal facility which has not been approved up to now due to a disagreement of the owner of the appropriate land register (Pečeňady village). Meanwhile in 1992 to 1994, independently from the design, monitoring activities went on in other non-mapped places of the impacted banks of the Dudváh - Váh river system.

1.1 Finalisation of monitoring of the contaminated site

A ^{137}Cs contamination from the accidentally damaged A-1 Bohunice Plant was gradually identified also along the entire downstream engineered section of Dudváh River, as well as along an one kilometer long section in the Váh River flood plain area which formed a mouth delta of Dudváh River prior to the reconstruction of the flow body.

The contamination in the engineered section of Dudváh River was described in [1]. On the present banks of Váh River close to the Siladice village, the contamination used to occur in the places with sediment depositions in the former Dudváh River's delta bed. Processed data about ^{137}Cs activity concentration obtained by detailed in-situ mapping of gamma radiation and thickness of sediment/soil layer in 1994 are shown in Tab.1 of this work.

On these new places, another approximately 11 000 m^2 of contaminated surfaces were found with the average activity concentrations from 1.3 up to 3.7 Bq ^{137}Cs/g of soil. On other places of the Váh River banks, no other contamination was identified by monitoring. Only after completing these monitoring activities, the entire extent of the contamination could be considered as a final one. According to the data in [1] and Tab.1, the overall identified contaminated area on the affected banks around Bohunice NPP with the ^{137}Cs activity concentration above 1 Bq/g is approximately 67 000 m^2 and the corresponding volume of contaminated soils is about 12 000 m^3.

It was gradually recognized that it is impossible to store the entire volume of the contaminated soils on the Bohunice site due to capacity reasons. In this context, it became necessary to reevaluate comprehensively the design of remedial measures in the Bohunice vicinity including all contaminated places on the banks and to account for all circumstances including large costs for the restoration techniques and benefits obtained.

1.2 A comprehensive reconsidering of remedial measures

The objectives of the comprehensive considering of remedial measures for the contaminated banks were specified as follows:

- to propose the implementation of appropriate restoration techniques and of its optimum scope related to the development of proper cleanup criteria based on proposed and authorized principles
- to propose rules for manipulations with below-limit contaminated soils on the banks in case of their transposing during bank maintenance or reconstruction,
- to propose criteria for the application of less expensive prohibitory and administrative remedial measures.

The solution is mostly complicated by the fact that a clear legislation is absent in the field given. Although, an environmental ministry act (1992) exists, where a limit of ^{137}Cs concentration in soil with a value of 0.5 Bq/g is given for a case of land-field transfer from the state to private ownership, it is not usable in the subjected context.

According to the experience with the present solution, as well as with the knowledge obtained in the course of the solution of this IAEA Project (RER/2/022 Environmental Restoration in Eastern and Central Europe), it was obvious that the choice of restoration techniques and derivation of proper acceptance levels depend on a number of parameters (dose limits, parameters of scenarios...) that necessarily should be harmonized and clearly declared.

It was thus necessary first of all to develop certain principles and rules for evaluation of the extent of necessary remedial measures, including development of contamination acceptance levels, and to achieve their authorization by hygiene authorities. Such principles, as well as the evaluation itself were elaborated recently in the VÚJE Institute and submitted for comments into the National Institute of Hygiene and Epidemiology (NÚHE). After the first round of discussions with this authority, in October 1994, it was agreed in this context that the ICRP dose limitation system for recovery of contaminated places will be used accounting for a net benefit from the implementation of remedial measures or for a reduction of radiation risk below a declared limiting value.

When proposing the scope of remedial measures, it was then possible to start from the following agreed principles:

- justification of restoration techniques (cleaning/covering) is conditioned by exceeding the limit of individual effective dose for public, 1 mSv/y (according to the ICRP/91 recomendation) for authorized scenarios with non-zero probability,
- costs associated with the application of a particular technique decide its choice,
- decision making about the extent of restoration starts from derived intervention levels of average ^{137}Cs concentration in soils and of contaminated soil volumes and surfaces comparable with those resulting from corresponding scenarios of radiation risk,
- averted detriment from a stay on contaminated banks does not justify the implementation of cost consuming techniques, its optimization is used for the application of less expensive remedial measures, only
- the residual contamination of banks will be considered as a contamination under control for the time period of 50 years as a minimum. It will be assured by administrative steps that a planned translocation of contaminated soils (maintenance, re-engineering) within this whole time is under the control of hygiene authorities.

More detailed results obtained in the application of these principles for the evaluation of remedial measures and for developing proper cleanup criteria for contaminated banks are described further in the paper.

2 ANALYSIS AND SELECTION OF RADIATION RISKS SCENARIOS

Two stay scenarios were selected for the evaluation of an actual risk from banks and from contaminated field, and another two residential scenarios for the evaluation of a potential risk from the use of contaminated soils supposed to be fully (about 200 m^3) and partially (about 50 m^3) spread on the site surface around a living house with a garden. Critical individuals were chosen based on an analysis in the following way:

1) a fisherman with the duration of his stay on the bank of 300 hours in sitting position (dose in 30 centimeters above the ground, corrected by the factor of 1.4 against the

dose at the height of 1 m) and consuming 200 liters of milk and 10 kilograms of meat (goat),

2) a farmer spending 500 hours in growing vegetables on a field and consuming 110 kg of potato and 110 kg of root and leaf vegetables from his own field,
3) a resident on a fully contaminated land with the area of 800 to 1000 m^2 spending 500 hours in the garden and 1500 hours around the house, consuming the entire annual consumption of potatoes (110 kg), a semiannual consumption of root and leaf vegetables (110 kilograms), 100 L of milk and 10kg of meat (goat) from his own garden,
4) a resident on a partially contaminated land (50 m^3 of soils), spending 250 hours in the contaminated part of the garden (100 m^2) and 1700 hours around the contaminated house (300 m^2 x 0.1 m) consuming the same contaminated food as in case No 3.

When determining the effective annual dose from external exposure $E_{ext} = H_E(0.7)$ for critical individuals, either measured data (E_{ext} = dose x 0.7) for banks and fields , or factors for a limited source according to Oztunali [3], and for a limited layer of an indefinite source, according to Cocher [4], in the case of scenarios with a resident, were used.

When modelling the ingestion, transfer factors were used either according to measurements (grass) or to "expected" values for goats milk and meat, and loamy soils, in case of vegetable according to [5]. The residential scenario with fully contaminated soil was described in more details in the previous work [2]. For soil dilution, in the scenarios, it was agreed upon to use the dilution factor c_w = 1.0 in case of a surface contamination on the banks.

Making certain parameters in the scenarios more harmonized has not been completed yet, however, resulting dose factors, shown in summary in Tab.3 according to the first draft,

Tab.1 Contaminated area and Cs 137 activty concentrations on the affected banks near Bohunice NPP

Contam. section	S,>1 [m^2]	As [Bq/g]	S,>8 [m^2]	As [Bq/g]	A-resid. [Bq/g]	Note
K1	10000	6.7	2000	9.5	4.9	strip,1-1.5m
K2	5730	16.2	5730.0	16.2	3.3	strip,0-2.5m
K3	9725	2	0	-	2	near village
D1	1200	1.8	0.0	0.0	1.8	
D2	1500	3.5	0.0	0.0	3.5	
D3	3725	4.7	1400.0	8.4	2.5	
Dpo	1500	2	0.0	0.0	2.0	
DR1	5940	1.9	0.0	0.0	1.9	
DR2	6050	3.2	0.0	0.0	3.2	
DR3	10500	9.6	9450.0	10.2	0.9	soil cover
VPK1	4760	2.1	10.0	8.7	2.1	<min area
VK35	2530	2.8	25.0	9.2	2.8	<min area
VK6	4260	1.8	0	-	1.8	
SUM	67420		18615.0	0.0	2.5	

S,>1,>8 - area with activity conc. >1,>8 Bq/g should not change significantly. From Tab.3, one can see that the most critical scenario with the full use of soils (200 m^3) poses a potential radiation risk at the level of effective dose approximately 210 $\mu Sv.y^{-1}/1\ Bq^{137}Cs.g^{-1}$ of soil to which the dose related activity concentration factor of 4.8 $Bq^{137}Cs.g^{-1}/1\ mSv.y^{-1}$ corresponds.

The probability of occurrence of such types of scenarios, however, is very small (0.01) and is conditioned by:

1) uncontrolled removal of soils from banks, whereas large volumes (200 m^3) can be removed only in the event of extended planned reconstruction activities, and smaller volumes (up to 50 m^3) can be removed in case of a common maintenance of banks, or for example in case of a bridge construction across a river etc.
2) uncontrolled translocation of those soils to the vicinity of a resident's house.

The small likelihood P of this scenario is taken into account when determining the risk and related intervention levels in the estimate of time for which it is possible to consider the probability of the scenario given as zero. These times were estimated, on the basis of consideration and agreement, in the following way: the uncontrolled removal of contaminated soils from banks is improbable (P=0):

- for volumes approximately 50 m^3 within the time of 5-10 years as minimum (for the calculation 5 years)
- for volumes approximately 200 m^3 within the time of 10-15 years as minimum (10 years).

When optimizing less costly remedial measures, agreed scenario is used with the pre-estimated factor for collective dose (milk + external exposure from banks) $2x10^{-7}$ man $Sv.a^{-1}/(m^2.Bq^{137}Cs.g^{-1})$. This simpler approach seems to be justified in this case due to the absence of correct quantitative data about the use of contaminated banks. Besides the

Tab.2 Cs 137 spots distribution in a strips on the Manivier canal banks depending on their position (0 - water level)

section	strip position	Aav. Bq/g	area m^2	Asp.>10 fraction	Asp.>25 fraction
K1	0 - 0.5 m	6.7	2000	0.24	0.12
	0.5 - 1	4.2	2000	0.24	0.12
	1 - 1.5	9.5	2000	0.35	0.23
	1.5 - 2.5	6.5	4000	0.11	0.11
K2	0 - 0.5 m	16	1150	0.42	0.24
	0.5 - 1	13.5	1150	0.42	0.24
	1 - 1.5	25	1150	0.68	0.5
	1.5 - 2.5	14	2300	0.26	0.26
K3	0 - 0.5 m	5.7	1950	0.18	0
	.5 - 1	3.2	1950	0.18	0
	1 - 1.5	1.3	1950	0.05	0
	1.5 - 2.5	1.3	3890	0.05	0.05
SUM	0 -2.5m		25490	0.21	0.13
estim. area of spots [m^2]				5440	3300

Asp.>10 - activity concentration >10 Bq/g

Tab.3 DOSE FACTORS (DF) RELATED TO 1 Bq/g of Cs 137 IN SOIL FOR SELECTED SCENARIOS FOR CONTAMINATED BANKS

SCENARIO	To [y]	geom. f texp [h/y]	INGESTION [REL. UNIT]	DF(1Bq/g) [mSv/y]	DIL(1),To [Bq/g]
STAY ON BANKS	0	300x1.4 g=0.54	0.4 milk+ meat	0.035	28.6
STAY ON CONT. FIELD	0	500 g=0.67	1 veg.+ potato	0.078	12.8
USE OF SOIL 50 m^3	5 0	1950 g=0.39	1.2 ve+po+mi+me	0.14	8 7.1
USE OF SOIL 200 m^3	10 0	2000 g=0.67	1.2 ve+po+mi+me	0.21	6 4.8

unit of ingestion = 0.04 mSv/y (potato 110kg+ root veg. 55 kg + leaves veg. 55 kg)
g = used dose rate / 0.118 microSv.h^-1/1 BqCs 137.g^-1 of soil

DIL(1),To = 1/DF*exp(lambda x To), where
To - minimum time from which the scenario likelyhood is considered as non-zero

optimization, also a limitation of individual effective doses according to the stay scenario is used with the value of 0.25 mSv/y when evaluating less costly remedial measures (warning signs, ...).

3 ACCEPTANCE LIMITS (DILs) FOR SOIL ^{137}Cs CONTAMINATION AND FOR CLEANUP OF BANKS

The maximum acceptable level of soil contamination (MCAL) can be defined, in line with previous chapters, as an average ^{137}Cs activity concentration in the proper volume of soil which results in the first year of the exposure scenarios in the limiting effective dose 1 mSv/y with a non-zero probability. The MCAL values were derived from the dose relation factors in Tab.3 and their values are according to Tab.3 (DILs) 6.0 Bq/g for 200 m^3 and 8.0 Bq/g for 50 m^3 of soil used.

The cleanup criteria AL can be defined also as due average activity concentrations of ^{137}Cs in a surface soil layer in a continuous strip of bank with the area which is proportional to the defined soil volumes. Assuming a thickness of 20-25 cm of the contaminated top soil layer, the criterial minimum area or length of banks, where the average activity concentration should not exceed the limiting values, are as follows:

- 800-1000 m^2, or a section with the length of 300 m for
  $MCAL_{200}$ = 6.0 Bq^{137}Cs/g = AL_{200}
- 200-250 m^2, or a section with the length of 80 m for
  $MCAL_{50}$ = 8.0 Bq^{137}Cs/g = AL_{50}

When making decision on the application of technical remedial measures, the belonging limits are compared with the average activities in these areas of contaminated banks defined in such a way.

The decision on the extent of bank restoration can be made according to the monitoring results obtained up to now which are summarized in Tab.1 and Tab.2. The cleanup criteria and the criteria for residual activity concentration on the banks expressed in a directly measurable term being depending on the detector used, will provide a part of the bank restoration design.

It can be seen from the monitoring results evaluated, as it is introduced in Tab.1, that the surface distribution of ^{137}Cs on the banks is strongly non-uniform. On Dudváh River banks, for example it is sufficient to apply the cleanup criterion AL_{50} = 8.0 Bq/g, only, because the average residual activity concentration on larger areas of the bank sections already will meet the limit of AL_{200} = 6.0 Bq/g.

On Manivier canal where the contamination consists of small but intensive spots, the situation is more complex as it also results from the estimate of the average ^{137}Cs distribution on particular 0.5 m wide bank strips, shown in Tab.2. The extent and procedure for this bank cleanup have been proposed as follows:

- according to a shielded detector response, spots of contamination above 25 Bq/g have to be identified and removed, at first, from steep slopes of bank (0.5-2.5 m) (controlled spreading of contamination to water)
- in a similar way, the spots will be removed from a strip of eroded soil covering paving stones including spots above 10 Bq/g (inverse depth activity distribution is expected),
- finally, due to process reasons, also remaining soil, covering the paving stones, will be removed and it will be used for filling up the holes remaining from the spots removed from the bank.

The value of 25 Bq/g is close to the non-rounded value of the proposed exemption criterion, according to [6].

4 COST OF RESTORATION TECHNIQUES AND THEIR DESCRIPTION

4.1 Decontamination by bank cleaning

With regard to an older origin of the contamination on banks, and to the in-depth distribution and its location found in lower parts of the steep slope in the unengineered section of Dudváh River and Manivier canal, the only reasonable way of removing the declared over-limiting radiation risks is the decontamination of banks by reducing the contaminated top soil layer.

The removed soil is to be safely disposed in an isolated disposal facility which makes this restoration technique cost consuming. The costs for removal and storage of 1 m^3 of soil from the above mentioned banks were recalculated on the cost level of 1994 according to the design in [2] and they are, on the average, approximately 1500 Sk (US $ 45) per m^3.

The structure of the considered costs is also shown in Tab.4. When removing soil from the banks, equipements commonly used for maintenance and reconstruction of flows are assumed to be used. There is mainly a walking excavator Shaef HR40 made in Germany, and dumping trucks Tatra 815 with the capacity of 9 m^3 which should transport the soil loaded from the banks directly into the storage facility.

The minimum thickness of the soil layer removed from banks is limited technologically as thin as 15-20 cm. For an average width of contamination of 2.2 m, this means to remove 330-440 m^3 of soil from one kilometer of decontaminated banks. On Manivier canal, in the length of about 5 km, we propose only to remove individual spots of contamination which would save a part of costs for the soil storage against the removal of continuous strips (Tab.2). Also, due to the possibility to detect the individual spots, the cleanup criterion for spots on the slopes was proposed on the level of 25 Bq ^{137}Cs/g of soil.

Tab. 4 THE TOTAL COSTS AND THEIR STRUCTURE FOR SOME REMEDIAL MEASURES

Remedial measures	Sk/m (*)	Sk/m^3
Fencing of banks	700	-
Soil Cover (DR)	92	-
Removing + disposal, total	(1473)	1473
removing from banks (K+D)	(400)	400
consolidation of banks	(320)	320
transport	(60)	60
filling during disposal	(22)	22
building of disposal fac. underground, concrete basin, 5800 m^3 / 2880 m^2	(678)	678
above ground landfill, asphalted stone liner, 4700 m^3 / 3100 m^2	(632)	632
above ground landfill, clay liner, 4700 m^3	(1221)	1221

(*) for 1 m of banks to be remediated
for data in brackets 2.5 m wide strips of contamination
and 0.2 m of soil layer is assumed

4.2 The storage of contaminated soils

The only acceptable place for the storage facility with regard to the public opinion expressed by the mayors of villages is the site of the Bohunice Plant. A confined storage area is available there, on which an underground storage facility would be implemented with isolation layers according to the design, already described in [2]. The isolated layers of the storage facility are shown in Fig.4. The costs and capacity relations for investigated options of the storage facility are also shown in Tab.4.

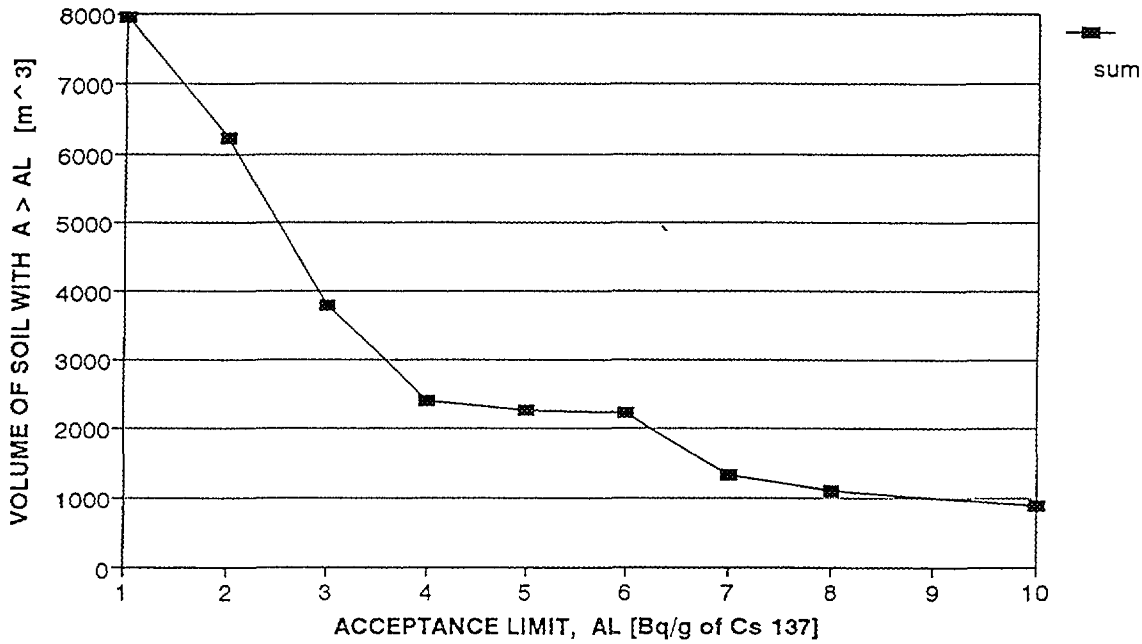

Fig.2 Soil to be removed from overall contaminated banks depending on AL

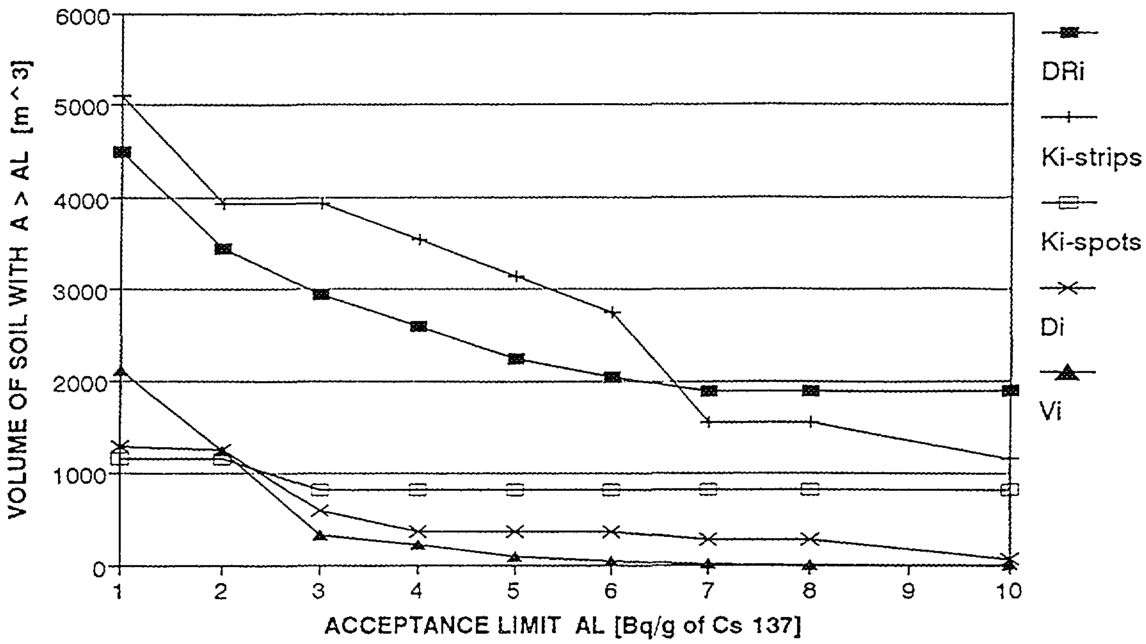

Fig.3 Soil to be removed from partial sections of banks depending on AL

4.3 Dilution and fixation of contamination by clean soil cover

This remedial technique was proposed for the engineered section of Dudváh River where the upper 20 cm layer of soil on flat 2.5 m wide terraces built in the bottom parts of banks is contaminated. Clean gravel cover may be considered also for the flood plain of Váh River where smaller flat areas in natural depressions are contaminated.

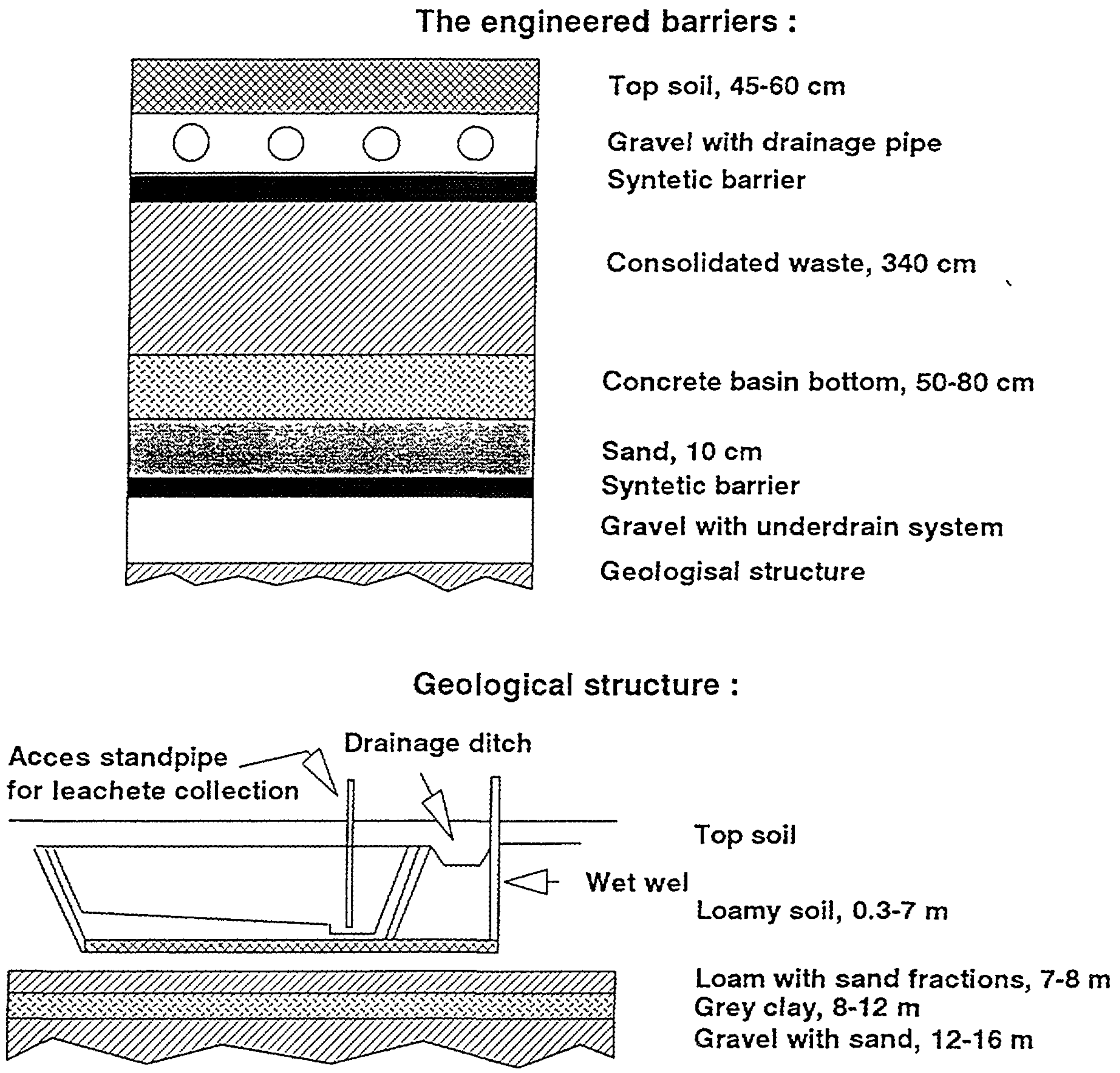

Fig 4. The underground disposal facility with engineered isolation barriers.

The large time scale protection effect of clean cover technique results according to the soil use scenario, mainly, from the dilution of contamination achieved in the top 20-30 cm soil layer, which is given by the removing technique commonly used. The associated protection effect of clean soil cover through bank use scenario is also evident, but it is not a main reason for its implementation on the contaminated banks.

Regarding to the engineered character of subjected sections (DRi), it is justified for an acceptance level derivation, only to consider the reduced (50 m^3) residential scenario and due acceptance level AL_{50} for cleanup. This AL_{50} was modified by a factor ($1/c_w$) of two for set up an acceptance level for the use of the covering technique, in this section of banks.

In this way the clean soil cover technique could be considered as an alternative against the cost consuming bank decontamination, however, up to maximally about twofold higher levels ($2*AL_{50}$ = 16 Bq/g) than the proper cleanup criterion AL_{50}.

On these engineered sections, only, the DR3 one does not comply the cleanup criteria for average concentrations (9.6 Bq of ^{137}Cs/g versus AL_{200}=6.0 Bq/g - Tab.3) . Also, according to the obtained monitoring results [1], even on the smaller parts of these sections the ^{137}Cs concentration does not exceed the mentioned 16 Bq/g . Consequently, the soil removing from this section is not necessary.

The cost for the implementation of this technique is approximately 10 times lower in comparison with the alternative decontamination by soil removing technique (Tab.4).

5 THE SCOPE OF THE CONTAMINATED BANKS RESTORATION

According to the criteria developed, it is necessary to subject to the restoration approximately 11 000 m^2 of contaminated area on the Dudváh River banks (Tab.1.) The most of this area are in the engineered section DR3, where clean soil cover is sufficient to be applied on an area of approximately 9 500 m^2 as it was described earlier.

In the contaminated sections of Váh River's flood plain, the activity concentrations found do not exceed the defined cleanup criteria, even though all specifications of this place were not taken into account up to now (contamination of gravels and underground water). However, 30-50 cm clean gravel cover on smaller areas is considered, only, in these places as a maximum.

The extent of the entire contamination of banks is also shown in Fig.3 in the graph of assumed volume of contaminated soil to be removed from the partial bank sections depending on a variable cleanup limit AL. It can be seen from this figure that this volume of soil strongly depends on the value of cleanup limit AL.

From Fig.2 with a like graph as in Fig.3, but for the summary of the sections, it is seen that in case of the proposed cleanup criteria (6 and 8 Bq/g) about 1 100 m^3 of soil is to be removed from the Manivier canal and unengineered Dudváh River's contaminated banks already taking into account application of the cover technique in the engineered part of the Dudváh River.

6 CONCLUSIONS

In the subjected field, clear legislation absents in the Slovak Republic. To consider planning for restoration of the contaminated banks near Bohunice NPP, new principles for the evaluation was developed in close cooperation with the national hygiene authorities. The principles as well as the resulting evaluation and proposal of the justified extent of banks restoration are being now under final authorisation.

The ICRP dose limitation system for recovery of contaminated site was used as a basis for these purposes. The value of 1mSv/y was set up as a basic maximum acceptable dose limit, exceeding of which only justifies implementation of a cost consuming restoration techniques. Cleanup and contamination acceptance criteria was developed as ^{137}Cs concentration in soil (8.0 or 6.0 Bq/g depending on the size of contaminated area/volume) on the basis of authorised radiation risk scenarios for potential use of the contaminated soil around a residential house.

The reevaluated extent of the banks restoration project would include removing and safe burial on the Bohunice NPP site of approximately 1100 m^3 of contaminated soil from, and the overlaying by clean 10-15 cm soil cover on about 10 000 m^2 of contaminated flat area of the banks.

During the restoration of banks commonly available equipment from water-service industry is planned to be used. The total cost for the proposed bank restoration project which would be implemented in years 1995/96 can be estimated by the sum of about US $ 100 000.

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TECHNOLOGIES FOR AND IMPLEMENTATION OF ENVIRONMENTAL RESTORATION PROJECTS IN SLOVENIA

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Abstract

Not all environmental restoration technologies have been tested and evaluated in Slovenia and implementation of environmental restoration projects will not start before 1995. Information is presented in this paper on Slovenia's state-of-the-art in this field. The aim is to present shortly the state, design and implementation of rehabilitation works. Particular emphasis is given to the following aspects: regulations, planning, research, investigations and implementation.

1. INTRODUCTION

1.1. Identification and radiological characterization of contaminated sites in Slovenia. *(Summary from the 1st workshop contribution)*

There are no sites accidentally contaminated with radioactivity in Slovenia in the verbal meaning of the title.
Compared to the large extent of contaminated sites in other countries, this problem arises in Slovenia only on a small scale and at low level.

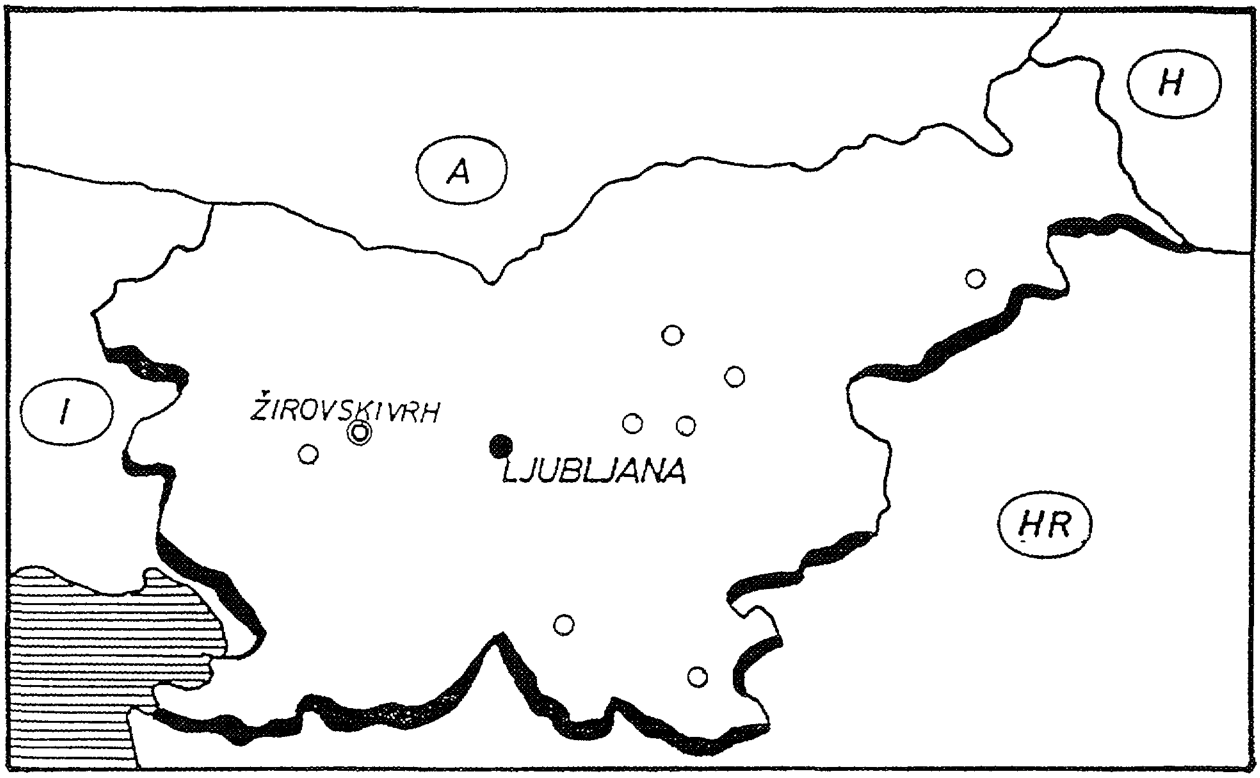
- The former *uranium mine* Žirovski vrh with its mining and milling facilities and waste deposits is at the moment a unique case in urgent need of restoration.

- Beside the U-mine, numerous local disposal sites with technologically enhanced natural radioactivity, with moderately low level U and Th content also need restoration. These are deposits in some *mining districts* (mercury, coal and coal-ash), large *coal-ash deposits* near thermal power plants, and large deposits near various *ore processing* factories (ilmenite, bauxite, phosphates)

- Out of the scope of this paper but worthy of mention: the central state storage for low level radioactive sources and wastes, mainly from medical and industrial use (Podgorica); an isolated storage place with accidentally contaminated material due to medical use of radium needles (Zavratec); the temporary low and medium radioactive waste storage facility at the Krško nuclear power plant;
  The planning of a new low and medium radioactivity waste storage facility is in progress.

The main attention related to contaminated sites in Slovenia is focused on the restoration activities at the former uranium mining and milling plant at Žirovski Vrh. Relatively low grade ore was excavated and treated (less than 0.1% U_3O_8) there in the period 1985-1990. Radioactive wastes, such as chemical tailings of about 600,000 tons were deposited on the slope of a hill on an area of 4 hectares, about 100-150 m above the small and narrow valley. The waste rock deposit (1.5 millions tons of mine rock waste on the area of 4 ha, with a content of 70 ppm U_3O_8) and a temporary deposit of some thousand tons of uranium ore are located near the bottom of the main valley. The radiological characterization of the uranium mining area was discussed already *(1)*, and some basic facts about its radiological impact on the environment in terms of enhanced radioactivity and the related dose calculations were also presented *(2)*. Radon daughters were found to be the main radioactive pollutant at the "Žirovski vrh" uranium mine, from the dosimetric point of view. The current status of annual public exposure: the additional exposure due to the contaminated sites is about 0.30-0.35 mSv, superimposed on a natural background of about 5.5 Msv.

The Žirovski Vrh uranium mine and other industrially contaminated sites in Slovenia



1.2. Planning of Environmental Restoration
(Summary from 2nd workshop contribution)

The Žirovski Vrh uranium mine was at first temporarily closed by an order of the Government of the Republic of Slovenia in the second half of the year 1990, but two years later, in July 1992, the Slovenian Parliament passed a law on the definite closure of the uranium exploitation facilities.

In the same year, a project entitled "Programme on the permanent cessation of uranium ore exploitation and on prevention of mining consequences to the environment at the Žirovski Vrh Uranium Mine" was started by the Consulting Engineering Agency "ELEKTRO-PROJEKT LJUBLJANA" in collaboration with the University of Ljubljana, Faculty of Natural Sciences and

Technology *(3)*. The Programme (see paragraph 2.2. later) covers all aspects of decommissioning of the mining and milling facilities and was accepted by the Government in April 1994. Its aim was to present the scope of the rehabilitation work, the time schedule, the necessary investigations, and costs and funds needed to complete a restoration.

In this period some pilot studies and other investigations (modelling, field studies) related to restoration of the tailings pile and waste rock disposal were also made (see Piešťany contribution *(2)* and further paragraph 2.3.). It also became obvious that there was a need for a relevant regulatory framework, not yet existing for cases for restoration of contaminated sites (see Piešťany contribution *(2)* and paragraph 2.1.).

2. TECHNOLOGIES FOR AND IMPLEMENTATION OF ENVIRONMENTAL RESTORATION PROJECTS IN SLOVENIA

There will be no clear distinction in this paper between the subjects of the last workshop (chapter 1.2. -PER) and the subject of this chapter (2. - TIER) and some overlapping will occur. Namely, not all technologies have yet been elaborated and the implementation of environmental restoration projects will start not before next year. We shall present here information on national state-of-the art activities in the field. The aim of this chapter is to present shortly the status, design and implementation of rehabilitation works.

The further discussion in this paper is concerned with the following items:
regulations, projects, investigations and research, and implementation.

2.1. REGULATIONS

A regulatory framework concerning environmental aspects of restoration has been in preparation since 1993, to set authorized limits for emissions from tailings and wastes and for residual environmental radioactivity, including all categories of contaminated sites, including radioactivity deposited from non-uranium related practices.

Standards for environmental protection against ionizing radiations (a draft version)

When preparing the standards for environmental protection against ionizing radiations *(4)* account was taken of the specific situation: i.e. due to the low level of contamination in Slovenia, more severe limitations (relative to other European countries) and stricter requirements for future waste disposal for non-nuclear radioactive wastes with natural radioactivity, are demanded on account of the relatively high population density and limited area of the country (territory).

Standards are provisional and are nowadays still in a draft version. Standards cover four items, including *nuclear facilities*; facilities and activities, where *unsealed radioactive sources* are used; *permanent storage of low and medium levels of radioactive waste* and finally, *waste disposal* (mining and industrial tailings) from processed raw ores *with enhanced natural radioactivity* - (U and Th ore, coal-ash, other residues from industrial processing plants).

General limitations

Annual dose limits for public exposure for the reference group are the following:

(1) **0.10 mSv** for the effective dose or whole-body dose
(2) 0.15 mSv for the organs gonads, uterus, red bone marrow
(3) 0.60 mSv for skin and bone surface tissue
(4) 0.30 mSv for other organs and tissues, excluding those in (2) and (3).

If there is more than one object (source) that contributes the to exposure of the reference group, then the above limitations are applied to the sum of exposures caused by these objects.

Special requirements

Future owners and users of each new open disposal site should have an operating and a closure plan, that should be submitted to the competent governmental authority to acquire a radiological license for the beginning of operation. This procedure is obligatory for all owners of
- mining and milling tailings related to uranium exploitation,
- to other mining and industrial tailings (with enhanced radioactivity) with a deposit area larger than 1 ha or with a total volume greater than 20,000 m^3,
- to coal-ash deposits with a total area larger than 1 ha and a total volume greater than 20,000 m^3.

The expression "total area" means the sum of areas of all locally separated disposal sites of the same owner and user and all tailings of the same kind of material.

All users of already existing disposal sites with the above characteristics should perform radiological measurements within a period of 12 months. After that all tailings should be classified according to these regulations. Radiological measurements should cover external gamma dose rate, average specific activities of natural and artificial radionuclides, measurements of the radioactivity of emissions and surveillance measurements of inputs (imissions). During the operation of a disposal site, tailings should be determined periodically; in already deposited material control of radioactivity should be measured in the following layers: (0-0.3 m, 0.3-0.6 m, from 0.6 m in steps of 1.0 m to the natural ground level).

Criteria for *the classification of tailings* concerning deposited material with enhanced contents of radionuclides from the uranium and thorium decay chain were established. Intervals of classes were defined according to the available data on known industrial disposal sites in the country.

The last group (IV) is further considered in more detail, because it is concerned in uranium mining and milling wastes.

IV. class of tailings (related to uranium production)

The uranium mine and mill disposal sites are under systematic control in the regulated (controlled) area and in the nearby surroundings. This control comprises external radiation, water and pathways, the food-chain transfer. Removal and use of deposited material is not allowed without the special permission of a competent authority.

Table 1. Tailings classification related to radionuclides from U-238 or Th-232 decay chain with the maximum specific activity

1	2	3
Class	Limit values of average specific activity **Bq kg^{-1}**	Tailings characteristics (description)
I.	less than 100	Tailings below the critical level
II.	100-200	Tailings with moderately enhanced radioactivity
III.	200-700	Tailings with enhanced radioactivity
IV.	more than 700	Low level radioactive waste(*)

(*) ... The term low radioactive waste - also for tailings - is used according the current Slovenian regulations *(ref 4.)*

Restoration of the site should take into account that projected doses will not exceed the general dose limit and in addition to this other limits are set:

- Wind erosion should be controlled to the extent to prevent air particulates at the border of the controlled area from exceeding 0.2 mg/m^3 of air.

- The maximum exhalation rates from the tailings must not exceed 0.7 Bq/m^2 s, (finally depending on the population exposure limit),

- The annual average of outdoor radon concentrations at the fence of the controlled area must not exceed the value 15 Bq/m^3 in any sector.

- The annual effective dose due to ingestion of drinking water must not exceed 0.05 mSv for members of the reference group.

- If the maximum gamma dose rate exceeds the value 0.4 μGy/h, then the controlled area must be physically protected. That means the total area of a disposal site should be situated in a physically protected controlled area, where all interventions (not related to maintenance of the site) are forbidden to conserve the integrity of barriers and cover layers.

- The use of vegetation from the disposal area in allowed conditionally, depending on the acceptability of additional radiation exposure.

Passive and active administration of the disposal site must be introduced, putting into force limitations to preserve the safety measures (integrity of barriers) and concerning the future use of the land.

The general characteristics for further categories of wastes are presented in Table 2.

Technical solutions for covering and remediation arrangement of waste disposals which are based on the results of pilot studies - considering the impact of waste disposal sites to the environment

Table 2. Provisional limiting standard values for all categories of wastes, containing uranium - after restoration (see also Table 1)

Class of waste	External radiation oper./restor. $\mu Gy/h$	Dose-drinking water $\mu Sv/a$	Air particul. mg/m^3	Exhalation rate oper./rest. $Bq/m^2\ s$	Rn-222 conc. enhancement Bq/m^3
IV.	> 0.4; 0.2	50	0.2	> 0.7 0.1-0.7	15
III.	0.2-0.4 0.2	50	0.2	0.2-0.7 0.1	10
II.	0.15-0.2 0.15	50	0.2	0.05-0.2 0.05-0.1	5
I.	0.15 -	-	0.2	< 0.05	-

- were already designed so as to fulfill the proposed standard requirements *(6, 7)*. Recapitulation of these findings is presented in Table 3, for mine waste deposits and for a tailings pile, respectively.

Table 3. Projected limiting values of radon flux, enhanced radon concentrations and dose constraints and corresponding provisional standard values

Disposal site	Rn-flux (actual) $mBq/m^2.s$	Rn-flux (after rest.) $Bq/m^2.s$	Rn-conc. (after rest.) Bq/m^3	Dose constr. (after rest.) mSv/a	Clay barr. (thickn.) m
Mine waste pile	0.7	< 0.1	1-2	0.05	0.25
Tailings pile	5-7	0.7	1-2	0.05	0.35
Standard (proposal)	-	**0.7**	**15**	**0.10**	-

2.2. RESTORATION DESIGN

In general there are three distinct areas at the Žirovski Vrh to be rehabilitated; these are the mine, the processing plant, and all the waste disposal sites. The programme on the permanent cessation of uranium ore exploitation and on prevention of mining consequences to the environment at Žirovski Vrh Uranium Mine *(3)* consists of the following sub-projects:

- project for permanent closure of the uranium ore *mine* exploitation facilities,

- project for cessation of the uranium ore *processing plant* with permanent environmental protection against the consequences of uranium concentrate production,

- project for restoration of *the waste disposal sites*: mine waste piles and mill tailings pile,
- permanent environmental protection against the consequences of disposal and storage with *long-term environmental monitoring* after restoration of the site

The time schedule, manpower and costs of closing down are also important parts of the programme. Roughly 50-70 million ECU are provided for the restoration of the site within a period of 5-7 years (provisionally by 2002).

Main attention at present is focused on restoration of the tailings pile; a major problem is slippage of the slope (7 millions tonnes) with dry tailings on the top (0.6 million tonnes) downwards to the valley at a velocity of 0.3 m per year. Three alternatives were elaborated to solve the problem of mill tailings slippage, each of them is dependent on the technical solution chosen for the mine closure. No decision has been made yet.

The restoration of sites should actually start at the beginning of 1995.

2.3. INVESTIGATIONS

The following investigations have been performed or are still going on in connection with rehabilitation of the affected area:

Radiological research and measurements (ref. 6,7,8):

- Gamma dose rates (on mill site and inside buildings, in mine facilities, at all waste disposal sites and in their surroundings, in the vicinity of the exploitation area, on industrial roads used during operation of the mine etc.),
- Radon-222 exhalation rate (on mill tailings, on mine waste piles, transmission of radon through provisional clay and soil layers (radon barriers),
- Alpha and beta surface contamination (mill site, mill buildings, equipment etc.),
- Indoor radon-222 concentrations (in process buildings, in mine facilities),
- Exposure of public (dose to the critical groups),
- Radioactive pollutants in surface waterflows,

Hydrogeological research work (ref.9) :

- hydrology and hydro-geology of the mine area,
- reduction of the degree of pollution of mine water,
- investigation of underground waters (area of the mill site, mill tailing site, and mine waste disposal site), and of the Brebovščica stream underground aquifer,
- study of the biological and chemical quality of surface waterflows,

Geo-mechanical and soil-mechanical stability studies (ref.10):

- geo-mechanical stability of mine galleries against collapse (to avoid surface deformations)
- geo-mechanical stability (mill tailings, mine waste disposal),
- status of the underground structures at disposal sites (PEHD drainage pipes, concrete water ducts,
- mechanical properties of mixtures of concrete with mill tailings or mine waste aggregate, their leachability and permeability,
- sand blasting tests on concrete slabs,

Technological investigations (ref.11):

- computer modelling of long-term water pollution from the mine, and from the mill tailings pile and the mine waste disposal,

- Tests of ammonia removal from processing water (tailings run-off water),
- Organic solvent treatment tests,
- uranium and radium removal from impounded water by biotic processes

2.4. IMPLEMENTATION

Irrespective of the fact that permission for rehabilitation of the uranium site has not been given to the Žirovski Vrh Mining Company, some planning and the following implementation work was done for reasons of plant and environmental safety:

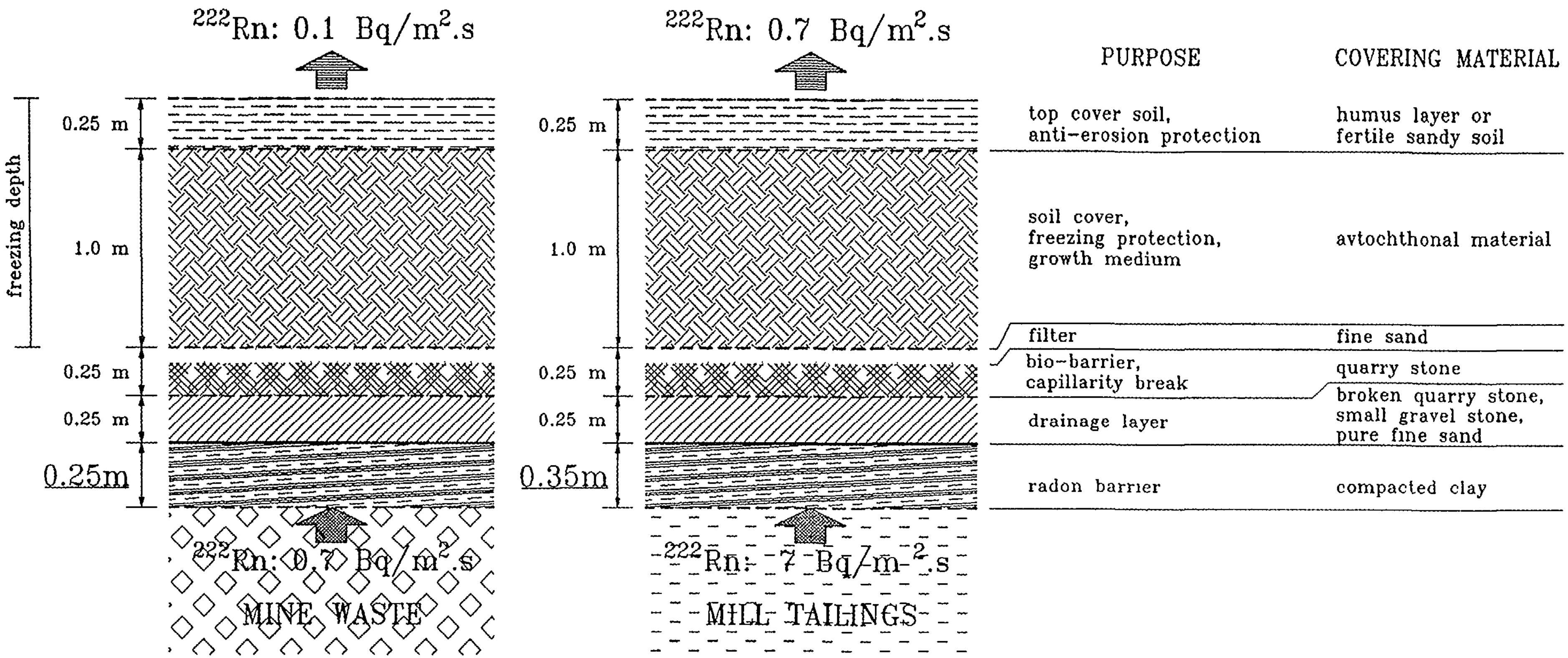
Rehabilitation work on mine waste disposal sites

- a part of the U-ore stockpile was transferred back into the mine,
- large quantities of mine waste from two smaller disposal sites were moved to the main permanent waste disposal site,

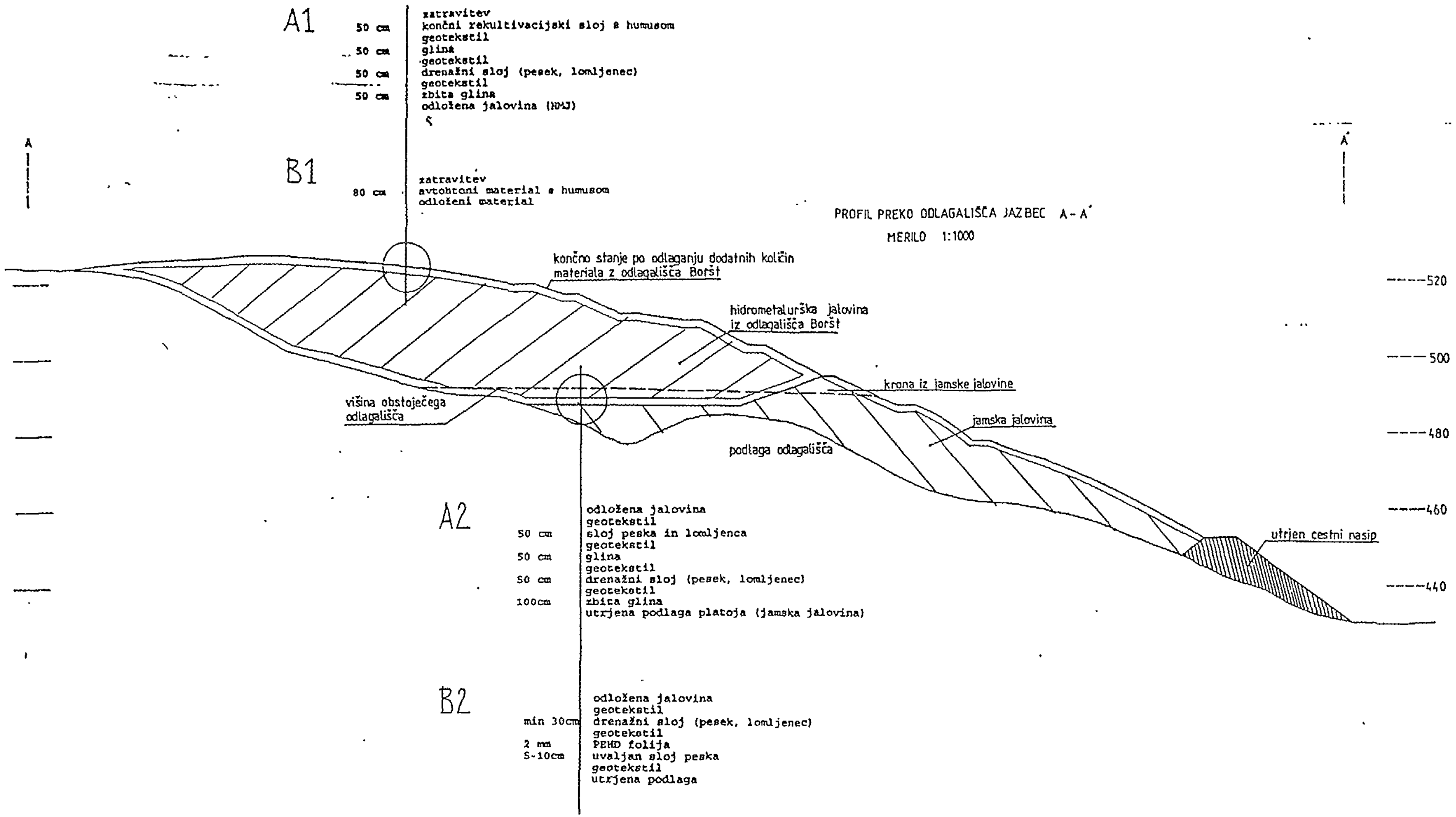
- The necessary permits were obtained and work was started on the construction of a water drainage tunnel under the mill tailings.

Decommissioning work in the processing plant

- corrosive liquids and chemicals from the processing plant were treated or removed,
- radioactive materials from the processing plant were removed,
- organic solvent was treated and removed,
- ammonia was removed and purified from concentrated liquid wastes,
- ammonia was removed (stripping) from processing water,
- equipment in the solvent-extraction plant was dismantled,
- vacuum-room equipment was dismantled,
- some other processing equipment was dismantled and decontaminated



Proposed constructions of complex covering layers



The Žirovski Vhr uranium mine - design of combined waste disposal
(cross section: mine ***waste disposal*** (lower) with ***tailings*** pile (above))

The decontamination procedures applied at the processing plant site have been shown to meet the following decommissioning criteria, set by the competent governmental authorities:

- Gamma dose rate - everywhere in decontaminated area	< 0,2 μG/h
- Alpha surface contamination, equipment and plant surfaces	< 4 Bq/100 cm^2
- Beta surface contamination, equipment and plant surfaces	< 40 Bq/100 cm^2
- Indoor radon gas concentrations, at work-places	< 250 Bq/m^3.

3. CONCLUSIONS

The beginning of restoration work at the Žirovski Vrh uranium mine has been delayed and shifted to 1995. The rehabilitation dynamics will depend on available funds (state budget). Some decommissioning work has begun already, and the whole restoration of the site should be finished provisionally in 2002. Technical solutions are not completed for all aspects of the work.

In the following years some other restoration plans and activities, not related to the uranium mine, are expected to start in Slovenia.

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TECHNOLOGIES FOR AND IMPLEMENTATION OF ENVIRONMENTAL RESTORATION OF THE URANIUM MINE IN RANSTAD SWEDEN

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Abstract

In 1985 the planning of the restoration of the closed-down uranium mine of Ranstad started. The plan was accepted in 1990 by the authorities and the main part of the plan was implemented in 1990 to 1992. The procedures and techniques for the remedial actions are described for the open pit mine and the mill tailings. A multilayer barrier cover system was implemented for reducing the weathering of the pyrite in the tailings and minimize the leachate of uranium, radium and heavy metals. Performance control of the cover system and especially the leaktight barrier was carried out by groundwater levelling and use of lysimeters. The open pit mine was transformed into a lake for recreation and wildlife.

1. INTRODUCTION

As described in the two earlier parts of the TC Regional Project on Environmental Restoration in Central and Eastern Europe the former uranium mine in Ranstad was remediated in 1990 to 1992. This paper will present the technical performance of the remediation of the mill tailings, as well as the open pit mine.

2. THE COVER SYSTEM FOR THE MILL TAILINGS

To eliminate the need for long-term purification of leachate, the tailings were covered with a sealing system to prevent further weathering caused by infiltration of precipitation water and air penetration into the tailings. It is of vital importance that the pyrite weathering reaction is stopped since the acid solutions produced, leach out heavy metals and other pollutants from the alum shale. Besides, the cover system will reduce the radon emanation to a very low rate.

The sealing system consists of a barrier with a low hydraulic conductivity and a protective layer (Fig. 1). The selection and evaluation of the material for the barrier was an issue of great importance. To find the most economical solution, one had to take into account; available material, required hydraulic conductivity and long-term stability.

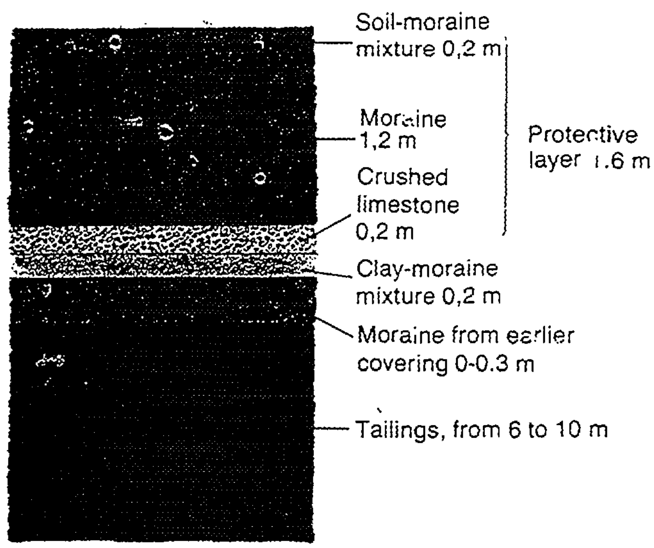

Figure 1 The cover system

The hydraulic conductivity was estimated to be equal to or less than $5x10^{-9}$ m/s, resulting in an infiltration of about 25 mm per year. The infiltration of water into the tailings at this low rate of conductivity is very low. Some water will be standing in or drained by the crushed limestone layer on top of the leaktight barrier. It is very important that this barrier always is saturated to prevent air penetration. The diffusion rate of oxygen in water is very low, so that the oxygen present in the deposit will be reduced. Most of the infiltrated water is expected to be transported down through the underlying moraine into the limestone horizon.

The situation concerning available material for the barrier was very favorable because of the presence of a clayey moraine close to the tailings. Laboratory measurements and field tests showed that the moraine contained a satisfactory fraction of finegrained soil. Thus no addition of bentonite had to be done as was foreseen.

The longterm stability is very important for the performance of the total cover system. A natural material such as moraine, formed during the latest Ice Age has a proven geological history that verifies its stability. Besides the technique of using natural soil for dams is well established in Sweden.

The application of the sealing layer was carefully made. Material with grain size more than 50 mm was sorted out in a screening plant and the rest was homogenized in a mixing plant. With dump trucks the material was transported to the prepared area to be covered. The material was spread with a bulldozer and the right thickness of the layer was finally obtained by using a grader controlled by a laser beam. With a 6 ton vibrating roller the specified degree of compaction could be reached.

The quality control consisted of checking the fine content of the mixed soil, the moisture content and the degree of compaction with both isotope instruments and watervolumetric determination. All measured values were documented and filed. The final condition of the surface of the leaktight barrier was documented on video film.

As soon as an area was checked and accepted, the moraine was covered with a 0.2 m layer of crushed limestone as draining layer and for neutralizing the acid precipitation. Together with 1.2 m of moraine and 0.2 m of soil-mixture, a protective layer of 1.6 m was obtained, preventing the sealing layer from damage due to draught, frost or root penetration.

To evaluate the performance of the leaktight barrier, various control systems were installed. To observe the establishment of groundwater table on top of the barrier, about 80 monitoring pipes were installed. Five infiltration lysimeters give the degree of water infiltration through the barrier. Also ten special oxygen diffusion lysimeters were placed underneath the barrier.

3. OPEN PIT MINE

The aim of the restoration of the open pit mine was to reshape it into a lake called Tranebörssjön with an area of 270 000 m^2. The about 15 m thick alum shale constitutes an almost leaktight bottom, thus allowing the lake to be filled by emerging groundwater (Fig. 2).

The alum shale debris surrounding the open pit area has been moved to the bottom of the lake and covered by moraine.

A considerable effort to transform the former mine into an attractive landscape harmonizing with the natural environment has been done. The embankment formed during mining has been turned into soft hills to match the surroundings. Marshlands, small islands and open beaches will promote bird nesting.

4. ENVIRONMENTAL EFFECTS OF THE RESTORATION

The environmental control programme is presented in part II concerning the planning of restoration. It is a comprehensive programme including macro-constituent, heavy metal and radionuclide

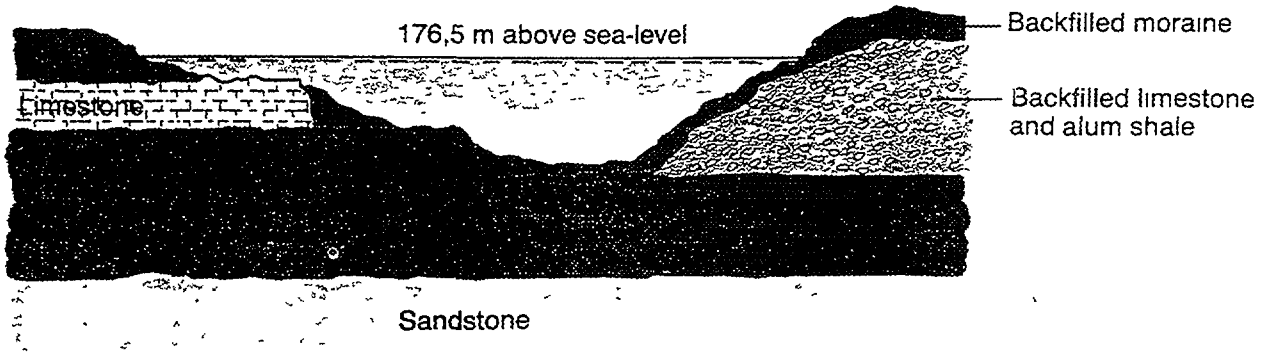

Figure 2 The open pit mine system

analyses. Besides the heavy metal concentration in water-moss is analyzed. The discharge is also measured at all sampling stations to obtain the transport of different elements.

Some elements, such as nickel and sulphate, have been chosen from the control programma to illustrate the initial effects after the restoration.

The concentration of some of the elements in the surface water in the tailings area as well as in the former open pit mine, the present Tranebärssjön are discussed below.

The concentration of sulphate within the tailings, called leakage, as well as in the surrounding ditch is presented in Figure 3. The sealing of the mill tailings took place during spring and summer seasons, because it was necessary with relatively dry climatic conditions to obtain a cover system with good quality. The sulphate content in the leakage water has decreased steadily from 1991 and onwards with a yearly rate of 450 mg/l. The total reduction of the sulphate in the surrounding ditch is about the same size i.e half of the content in the beginning of 1991. However, the decrease is much slower after 1992.

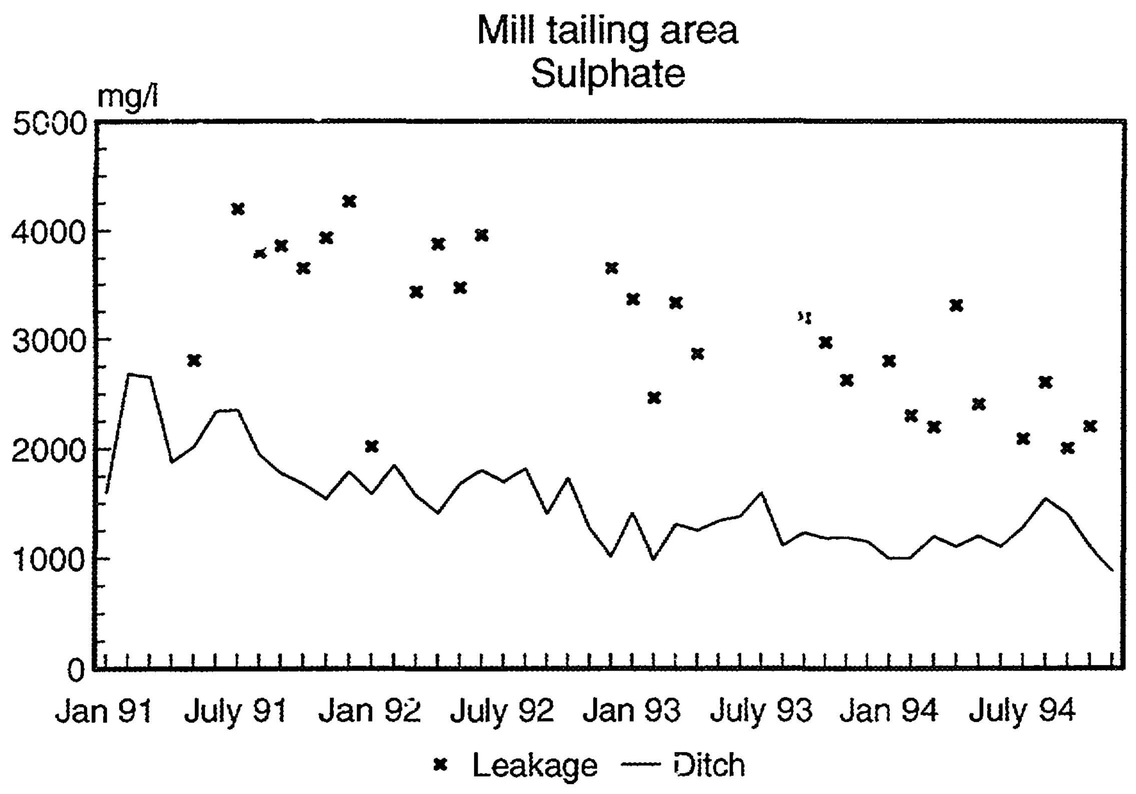

Figure 3 The concentration of sulphate, mill tailing area

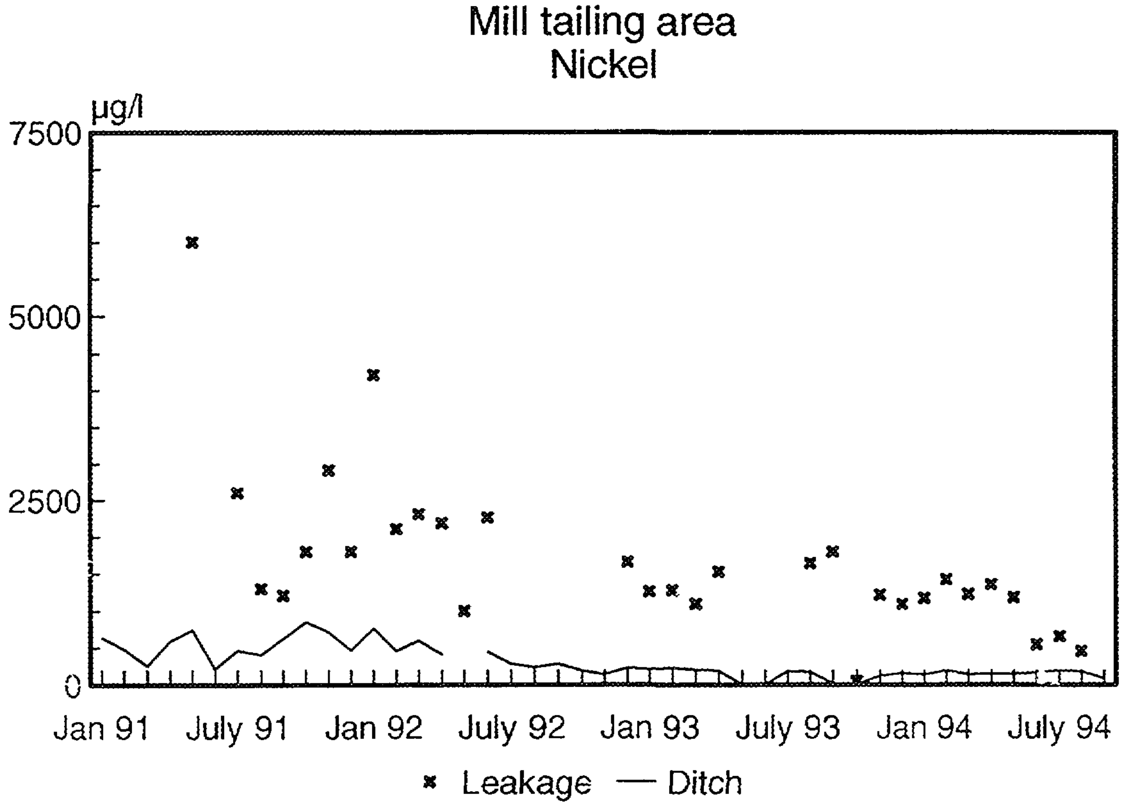

Figure 4 The concentration of sulphate, open pit mine area, Tranebärssjön

The decrease of the nickel content shows the same pattern as the sulphate. Furthermore, the present concentration is just about 20 percent of the highest observed in the leakage water in 1991. The decrease in the ditch water is in the same order as for the leakage water.

The open pit mine was filled with inflowing groundwater during a one-and-a-half year period that was completed in the spring 1993. The surrounding ground, consisting of a mixture of peat, till, limestone and top alum-shale, has been exposed to weathering during the 30 years it was kept dry by pumping the pit. The weathering products are at the moment entering the lake by the groundwater. As the groundwater does not contain oxygen, reducing conditions are prevailing in the bottom water of the lake.

The concentration of sulphate in Tranebärssjön is shown in Figure 5. The concentration in the bottom water is more than 50 percent higher compared to what is observed in the surface water. This is true for most of the time except for the circulation periods when thermal stratification does not exist and the exchange is facilitated. No direct trend of the sulphate concentration is observed, either in the bottom water nor in the surface water.

The concentration of nickel in the bottom water has increased during the transition period from open pit mine to a lake. Eventually the maximum concentration has already been reached. However, the redox conditions are complex i.e. the microbiological activity influences the mobility of different elements. Fast changes in the surface concentrations of for example iron have been observed causing precipitation of iron complexes.

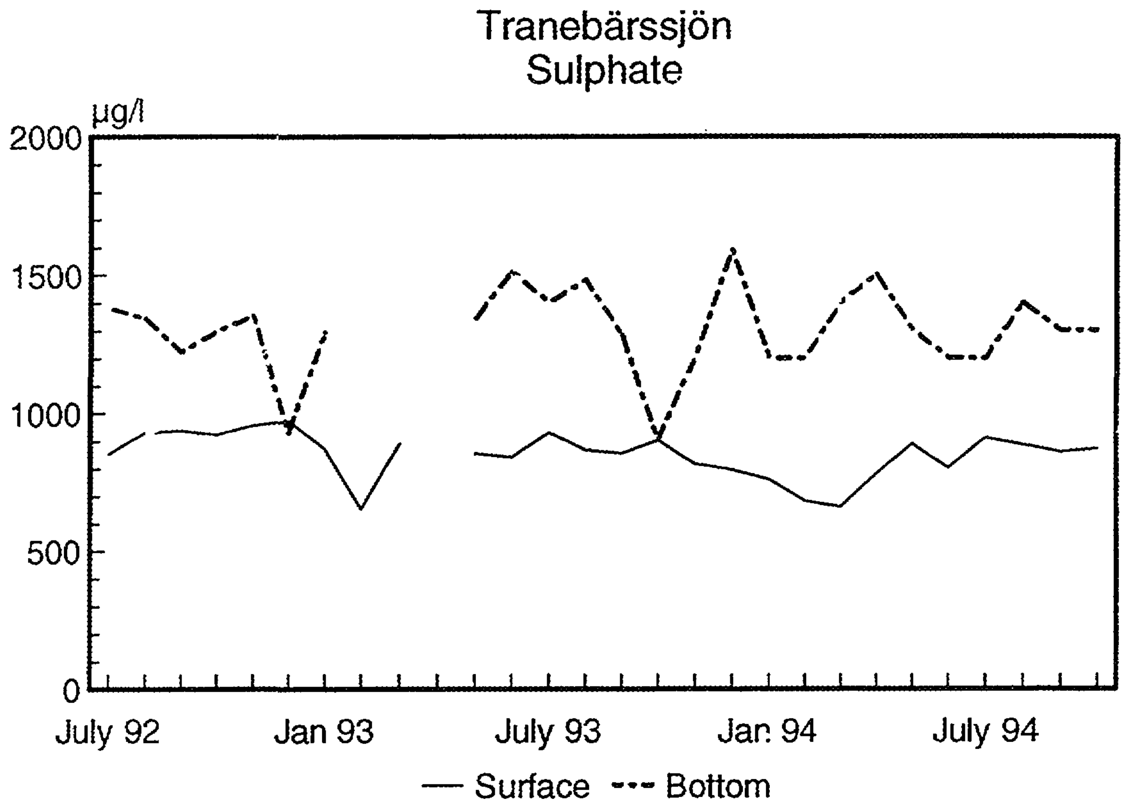

Figure 5 The concentration of nickel, mill tailing area

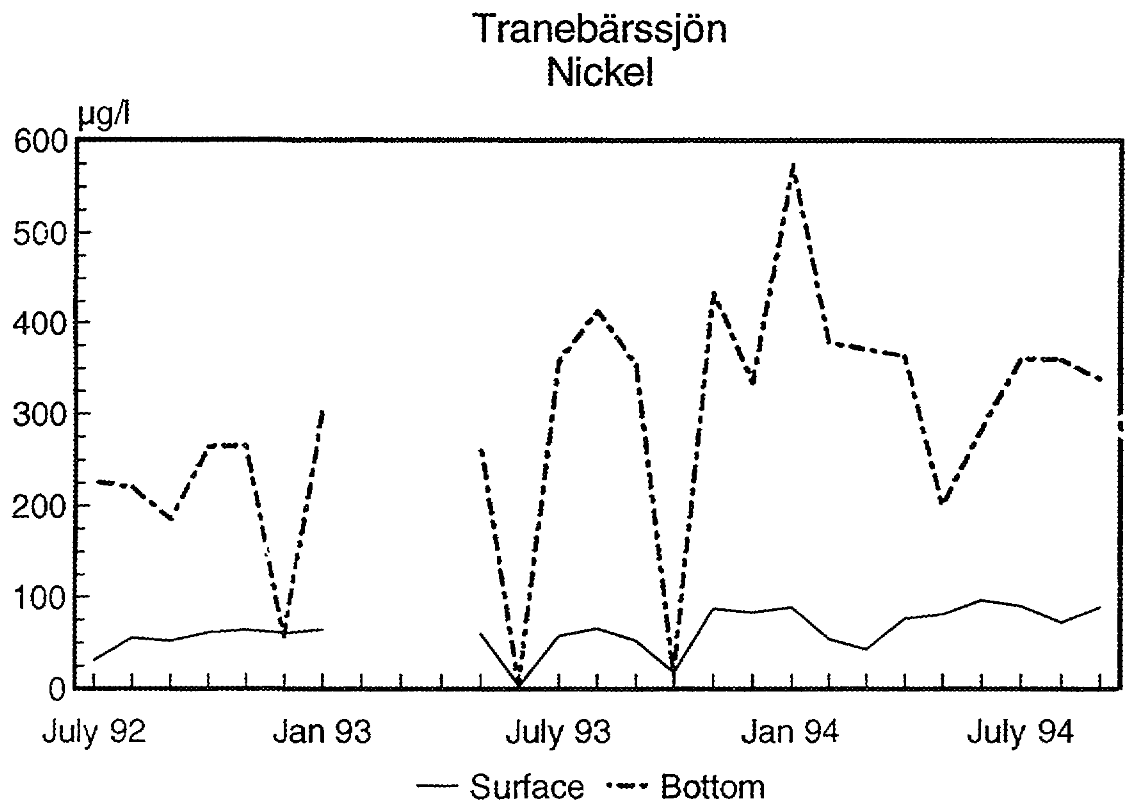

Figure 6 The concentration of nickel, open pit mine area, Tranebärssjön

5. CONCLUSIONS

We can conclude, two years after the sealing of the mill tailings, that the cover system works properly. The observed infiltration of precipitation is very low, less than five percent of the precipitation. No oxygen penetration through the leaktight layer has so far been observed. The concentration of pollutants as uranium and heavy metals is decreasing. However the reduction is slow and it will take some years before the purification of the leakage water can be terminated.

TECHNOLOGIES FOR ENVIRONMENTAL RESTORATION IN UKRAINE

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Abstract

This paper provides examples of restoration approaches in Ukraine for contaminated sites of various nature and origin. Advantages and disadvantages of such approaches are also described.

Forestation proved to be the most effective counter measure to bind radionuclides 'in situ' and prevent resuspension mechanisms in the Chernobyl restricted zone. Another protective measure in this zone is the erection of dams to prevent contamination spreading floods. The restoration programme in living ores mainly include upper soil removal, demolition and removal of roofs, fences and wall plasters, asphalting, and transportation of secondary wastes to the waste disposal site. Additional measures to reduce internal exposures include agrochemical measures and milk filtration. Such aspects are described in detail in this paper. A separate section deals with restoration of sites contaminated by uranium mining and milling activities.

I. INTRODUCTION

During previouse stages of the RER/9/022 it was confirmed and illustrated that restoration of environment, contaminated with ridionuclides is not a mere decontamination process, but rather a complex planned activity involving policy level decisions, sophisticated system of criteria, comprehansive financial arrangements.

Restoration includes application of tecnologies for decontamination, in-situ stabilisation and isolation of radionuclides, as well as special agrochemical measures to reduce nuclide transformation into chemically movable forms, their release and migration into food chains. Restoration technologies were applied in Ukraine in uranium industry starting from the early period of the uranium output, but only comparetively recently the development of systematic criteria was began. Chornobyl accident recovery operations provided the unique information for environmental impact of radiation as well as experience of application of wide range of environmental restoration activities.

Below are given examples of restoration approaches for contamination cases of different nature and origine as they were realized or planned in Ukraine with all positive and negative features proven by practical experience.

2. POST-CHORNOBYL RESTORATION

2.1. Restricted Zone.

Experience of the Chornobyl zone recovery activities demonstrates extremely low efficiency of the direct decontamination measures. Thus, it was shown that with average immediate decontamination factor of 1.2 - 1.5 the estimated collective dose for personnel participating in decon work exceeds 15,000 Sv. There is no data on the ultimate efficiency of those decon operations, because it was shown that in an early post-accident stage, after the certain period, decontaminated anclaves normally were recontaminated practically to the previous levels. For settlements some effect became visible after the nomber of the similar decon cycles.

Up to date decontamination works were performed on 2250 hectars of land in the Restricted Zone, complex restoration measures were done on 2400 hectars. It was shown that forrestation is the most effective countermeasure to bind radionuclides "in-situ" and prevent resuspention mechanizms. Though pine-tree forrest fires represent the other mechanism for radionuclide release. During fire up to 90% of radionuclide inventory could be released with smoke gases and carried to the long distances.

Example of another environmental restoration approach is a complex protective measures of the river Pripiat flood-planes in the vicinity of Chornobyl site, which was highly contaminated after the accident in 1986.

More detaled information on the contamination levels is given in the relevant report presented at the First RER/9/022 workshop in Budapest.

There is no inhabitants and economic (agricultural) activity directly connected with these areas, as they are within so-called Restricted Zone, with all population evacuated in 1986 year. Still, considerable risk is connected with Sr-90 and Cs-137 migration to the basin of river Dnipro, which is the main source of drinking and irrigating water supply in Ukraine, especially for it's Southern regions.

The main problem with planning of restoration activities for restricted zone connected with the vast dimensions of the highly contaminated flood-planes areas, and the need for immediate actions, because every next year reduces the effectiveness of such measures. The first factor requires an application of water protective measures directed to isolation of nuclides in-situ and prevention of flooding the most contaminated parts. Water protection approaches include erection of flood protecting dams along the main channel of river Pripiat and surrounding dams around most contaminated flood-plane parts. Still, the ultimate effectiveness of these measures, including collective dose reduction is not proven yet. The main income of Sr-90 into Pripiat and Dnipro basin takes place from less contaminated but much larger areas of Ukraine, Belorus and Russian outside the Restricted Zone. General picture of contamination flood-planes near the Chornobyl site is shown in Fig. 1.

2.2. Living Areas Restoration Programme.

Restoration Approach

Chornobyl accident caused contamination of 1779 settlements in Ukraine with level of contamination exceeding 1 Ci/sq.km. In accordance to Chornobyl legislastion special protective countermeasures should be implemented to the populated areas where dose limit due to Chornobyl accident exceeds 1 mSv/year. This criterion was put into basis of a living area restoration countermeasures.

Below is given a typical example of restoration technologies and countermeasures for one of the villages in the Rivne region of Ukraine. The contamination level for this zone falls within 37 - 185 kBq/sq.m (1 - 5 Ci/sq.km). Major bulk of radionuclides are concentrated in the upper 10 cm level of soil. Typically, the main constituent of the total dose is due to internal exposure (85 - 95%). Thus, for the example illustrated on Fig. ...-..., average contamination level is 3.95 Ci/sq.km (146 kBq/sq.m), annual individual equivalent dose comes to 3,78 mSv/year, 0.51 mSv/year of it is due the external exposure, and 3.27 mSv/year - internal exposure, 80% of the latter is due the local milk and diary products consumption.

Measures for external dose reduction include:

- upper soil removal in spots were dose rate exeedes 24 mkR/h;
- roofs, fences and wall plaster removal;
- asphalting of the yards and rain drainage areas;
- transporting of the secondary wastes to the waste disposal site.

Internal exposure reduction is perfomed by following measures:

- agrochemical measures (ploughing, fertilising, liming);
- filtration of milk;
- administrative and organisational measures (clean products supply, local food products monitoring and control, change of local habits and traditions, etc.).

Restoration area includes the territory of the village, and ajasent 500-m "protective zone". Fig. 3 illustrates typical contamination level characterisation plan which is the base for restoration works.

At each individual farmer estate restoration measures include removal of the strip of soil adjacent to the buildings with width of 1 m to the depth of 0.1 m. Resulting wastes with the levels of exposure dose rate more than 24 mkR/hr are transported to the waste disposal sites (to be described later). If exposure dose rate from wastes does not exceed 24 mkR/hr, this soil is used for road sides and land filling.

It should be noted, that in view of dominating internal exposure dose fraction, the most effective dose-reducing measures are the agrochemical ones, which include fertilizing, liming and sorbent adding (zeolite, etc.), which in more detail are highlighted in the section 2.5.

Waste Management

The plan view of the typical waste disposal point (WDP) is given at Fig. 4.

The main requirement for the WDP siting is an isolation of wastes from the environment for the period of 10 Cs-137 decay half-lives. Water table depth should not be less then 4 m from the bottom of the trench. WDP is usually sited at the elevated points of the local relief (hills, etc.).

WDP typical design features are:

- dimensions in plan: 100x60 m;

- slope angle: 1 : 5;

- depth of the trench: 2.5 m.

The cross-section of the WDP is shown on Fig. 5. The bottom of the trench is covered with lower isolation clay layer with thickness of 0.5 m. Resulting filtration coeffitient **k** is 10E-4 m/day, and provides full decay of radionuclides while they finally penetrate through the isolation layer. The depth of the nuclide migration through the clay sheat before the full decay for Cs-137 and Sr-90 is approximately 0.02 m and 0.36 m, respectively.

Upper isolation layer has multilayer structure. Radioactive wastes are covered by gas drainage sand layer, with variable thickness from 0.5 m in the center to 0 m at the edges. In this layer ceramic drainage duct is placed. Sand layer is covered with 0.5 m hydroisolation clay layer which is similar to the bottom one. Next is a 0.3 m sand layer for drainage of the precipitations into outer circular drainage trench. Upper layer is a fertile soil layer of 0.3 m. Vegetation is planted over the upper layer to prevent erosion.

Waste loading into WDP is perfomed sequentially by tracks. Each layer is temped by roller. To prevent dusting the surface of the layer is sprinkled with water. Radioactive wastes of organic nature are put inside the bulk of the non-organic wastes with a layer of 0.5 - 0.4 m. In more detail filling technology of the waste disposal point is illustrated in Fig. 6.

After filling of the trench and covering it with fertile soil, vegetation is planted on the top surface of the depository to prevent erosion.

2.4. Selective Soil Removal System

Experience of the early stage of the Chornobyl accident recovery operations shown the need in an effective soil removal units which would allow selective removal of contaminated soil depending on the contamination level of the soil suface. This would allow to reduce the quantity of radioactive wastes to be disposed or processed.

Fig. 7 shows the schematic diagram of the selective soil removal unit which is designed to perform removal of the upper soil layer in correlation with the dose-rate level in four individual strips.

It has four independent measuring and control channels allowing selective removal in four parallel strips of soil. System has a train configuration with tree principal elements: hauler (1), soil removing unit, and changable trailing truck (13) to accumulate and transport removed soil.

Hauler (tracktor, tank, etc.) has a shielded cabin and air conditioning system to prevent unacceptable exposure of two operators, one of which is a driver. In front of the hauler there is a detector group (2) consisting of four sensors in collimating cylinders (3). Viewing angle of each sensor is 20 . To let control of the soil removal mechanism a detector transducing unit (4) and hydrosystem (5) to drive the soil removal mechanism are also installed at the vehicle.

Soil removal part is in more detail shown at Fig. 8. It consists of four through bowls (12), each having individual hydrodriving mechanism (14) controlled in correlation with dose rate measurements in each individual channel. Each bowl has limiting apron (13), which prevents bowl from overdeeping into the soil and controls the thickness of the removed soil. To suppress dust that could be raised with dry soils, water sprinkling unit is mantled consisting of two water tanks (19), piping with valves (20) and two sprinklers (21).

As it was shown that even eight years after the accident the main bulk of radionuclides (up to 95%) is concentrated in the upper 5-cm layer of the soil (see Table I), the device for soil removal does not need to have capability to change the thickness of the removed layer during the same run. It can be ajusted prior the definit set of runs on the basis of more detailed investigation of the soil properties of the particular area to be developed. The last part of the system is an open changable trailed truck to accumulate removed soil and deliver it to the waste disposal point.

2.5. Agrochemical Measures.

Fig. 9 demonstrates that 5-7 years after Chornobyl accident major fraction (70-95%) of the total dose in contaminated areas is due to internal exposure. Nearly 80% of this part is caused by milk consumption, thus making of special importance the problem of production of clean forage crops.

The main agrochemical countermeasures include:

Mechanical treatment:

- Ploughing, cultivation

- Drainage

Chemical:

- Liming

- Addition of fertilizers

- Addition of absorbing compounds (clay, zeolit).

The effectiveness of these counermeasures depends on the specific hydrogeological and soil characteristics and land use class of the each particular site of application. Regions contaminated with Chornobyl radionuclides generally have poor soils of soddy-podzol, sandy and peaty type with poor top-soil layer, and in original condition are characterized with relatively high soil-plant transfer factors.

Ploughing is a comparetively cheap and practical countermeasure allowing to achieve the reduction of surface contamination from 2 up to 20 times, depending on the thickness of the inverted layer of soil. Combination of ploughing with application of some compounds, such as a mixture of sodium carbonate and isopropylphenyl carbonate, which prevent roof intrusion, can reduce uptake of Sr-90 by agricultural crops up to 1000 times.

Drainage is an effective countermeasure on the moisturous peaty meadow soils, and could reduce the nuclide uptake by a factor of 3.

Addition of fertilizers could have different effect depending on soil characteristics, type of the fertilizer and composition of the fertilizer-additives mixture. Addition of potassium fertiliser though generally positive, could be not effective for soils reach in potassium or clay soils. Addition of either nitrogen or phosphate fertilizers increases Cs-137 uptake. Fig. 10 illustrates the effect of different fertilizers to the yield and uptake of Cs-137 by oats and potato. It should be noted that this effect is generally vary depending on the fertilizer doses.

Natural meadow melioration investigation with respect to soil-plant transfer ratio for Cs-137 gives the following results. Drainage reduces uptake up to 3 times, soil treatment - up to 4 times, addition of potassium fertilizer - up to 3 times, the same with lime - 4 times, respectively. Complex countermeasures including all listed measures could reduce Cs-137 uptake to 16 times. Table II gives illustration of the comparetive effectiveness of different agrochemical countermeasures in application to a peaty meadow soil.

Table III gives comparative figures of soil-plant transfer factor for different forage crops for the same conditions of drained peaty soil. Table IV gives illustration of the impact of different meliorants to soil-plant transfer factor for different forage crops. It can be seen that the maximum effect has the elevated doses od potassium fertilizers in a mixture with complex mineral fertilizers, lime and manure.

Addition of lime is a universal effective measure, but it could be of low effect in an already calcareous soils.

3. EXPERIENCE OF ENVIRONMENTAL RESTORATION IN REGIONS OF URANIUM MILLING AND MINING INDUSTRY

Uranium milling and industry is concentrated in Kirovograd, Dnipropetrovsk and Mykolaiv regions of Ukraine. In total uranium milling and mining industry occupies 5530 hectars of land, 1340 ha are damaged.

Prevailing contamination path for waste rock piles is natural leaching of radionuclides with rains and snow melting waters. Fig. 11 - 14 shows processed curves of nuclide content and alpha-activity in 1-meter layer of soil at the site of relocated uranium ore stock. Table V 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 the soil at the site of former stock exceeds 1 m depth, and in the ajiacent area of 10 - 20 m width is within 1 m depth. Fig. 15 shows results of soil, plant and air contamination survey in terms of exceeding the backround reference level. It demonstrates that the area affected by waste rock-piles reaches 700 - 800 meters.

At Zovti Vody site the most powerful source of environmental contamination is represented by two mill talings. Exhalation of Rn-222 and resuspention of dry tailing sandy beaches provides two major mechanisms of environmental contamination. Rn-222 exhalation rate is 0.05-3.0 Bq/sq.m*s. Avarage zone of mill tailing influence for Ra-226 reaches 1300 m, and Po-210 - up to 1800 m (above background levels).

Restoration of lands affected by uranium milling and mining practice in former USSR started in the mid-seventies. The main restoration critera were introduced by special Sanitary Regulation (СП-1324-75). According to this guidance, the final contamination after finishing restoration activities should not exceed background level by 2 times. At limited portion of restored area (limited by 20% of total area) residual contamination could exceed background level by 3 times.

Project for restoration of waste rock piles was developed in Zovti Vody including two options of restoration measures. First option provides for smothening of pile slopes, covering with restoring layer, and finally, plantification of the restored area. Second option propose relocation of the waste rock into nearby natural pit, covering with restoring layer, and plantification of the restored area in the same mode. Table VI shows cost characteristics of both options in more detail. Restoration layer reduces Radon exhalation from 0.95 to 0.084 Bq/sq.m*s, or 10 times.

Waste rock pile that existed in Zovti Vody in late seventies was removed into the giant cave-in pit that was created at the site of former iron underground mining cavity. General view of the waste rock pile before the relocation is shown in Fig. 16 and 17. The general volume of waste rock relocated in the cave-in pit is 400,000 cubic meters. Fig. 18 shows the general view of the cave-in pit were this waste rock was relocated, and Fig. 19 shows the process of filling the pit. Partially materials from the pile were used as a depleted uranium ore to extract residual uranium, thus reducing the volume of waste rock and increasing the mill tailing part.

Cross-section of the mill tailing restored in Zovti Vody is given in Fig. 20. As a result of covering with the first clay layer of 0.4 m thickness the dose-rate on the surface of the tailing was reduced from 450-600 mkR/h to 24-56 mkR/h (background level is 24-56 mkR/h); radon exhalation was reduced from 0.05 - 20.0 Bq/sq.m*s to 0.028 - 10 Bq/sq.m*s. Planned technical and cost characteristics for different options of mill tailing cover layer are given in Table VII.

The main problem for the operated tailings is dusting of their dry beaches. Because the town of Zovti Vody is located in the close proximity to mill tailings, this problem has the special importance. Fig. 21 illustrates the jet monitor unit to sprinkle polymeric compound over the dry surface of the tailing to suppress dust.

Another approach that was used in Zovti Vody was an addition of the polymeric compound into tailing pulp during discharg into tailing pond. Illustration of this process in action is shown in Fig. 22.

Fig. 23 presents a schematic diagram of the underground mining cavity filling system using waste rock and mill tailing materials as a main components of the filling compound. This is a very effective approach which considerably reduces the environmental impact of uranium mining and milling industry, simultaneously allowing more effective use of the available uranium resources. The only negative feature of this technology is the elevated radon exhalation that would require additional anti-radon protective measures in undeground workings and faces.

Another example of restoration measures in uranium industry is the rehabilitation of the surface contamination at Devladove in-situ leaching site in 1973-1976. Contamination of surface was caused mainly by leackages of uranium-loaded lixiviant pumped out of recovery wells. Decontamination technology implied the replacing of surface soil layer for clean one. One patch of land was left intact, and during years was monitored to investigate processes of natural decontamination. Results of the study shows that expected self-decontamination had not took place. U-238, Th-230 and Ra-226 concentrations had not changed; slight decrease of Pb-210 concentration was due natural decay.

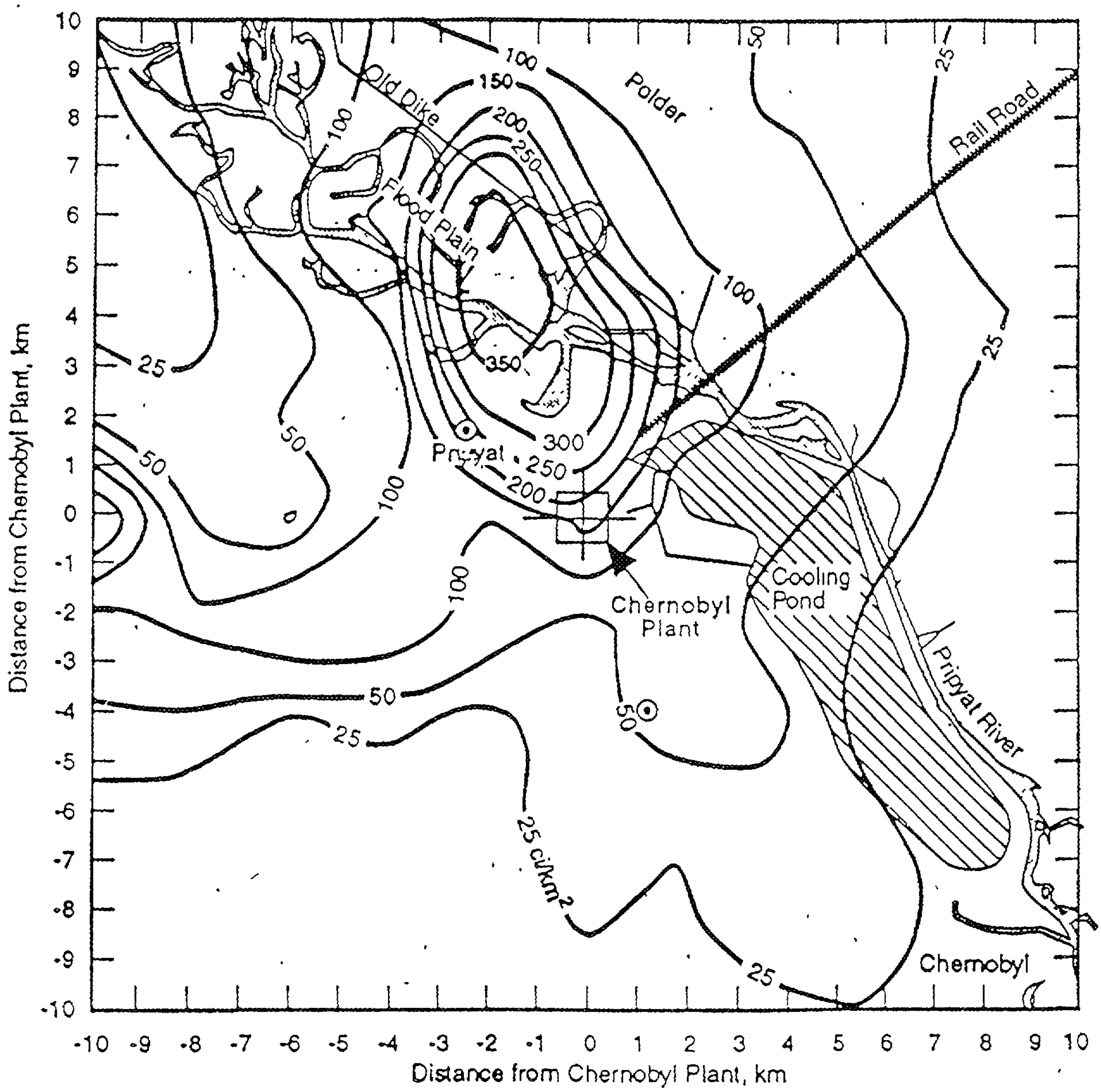
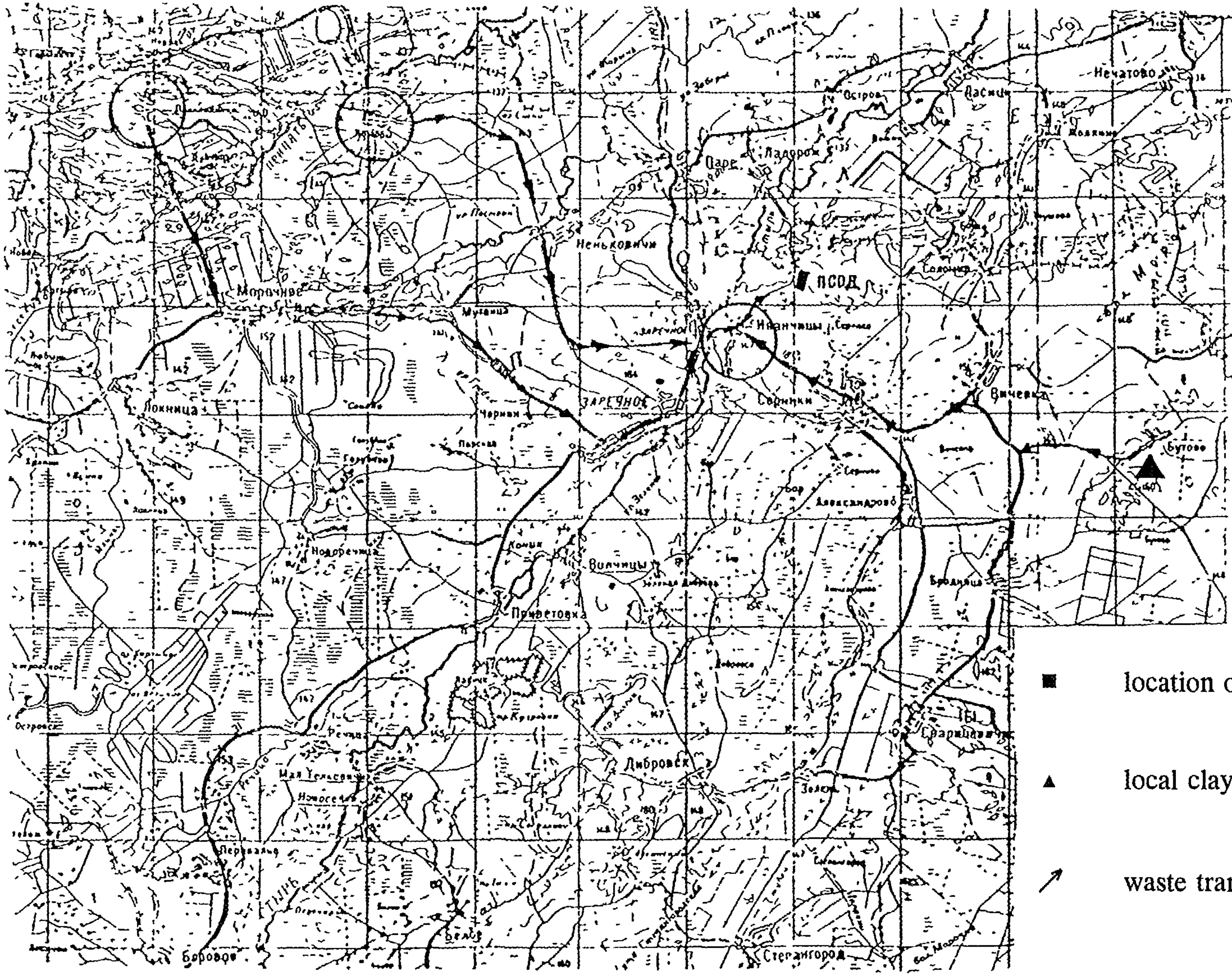


Fig. 1. Contamination of flood-plane areas in the vicinity of the Chernobyl NPP with Sr-90, Ci/km²



■ location of the waste disposal point (WDP)

▲ local clay output point

↗ waste transportation routes

Fig. 2. Plan view of the restored region

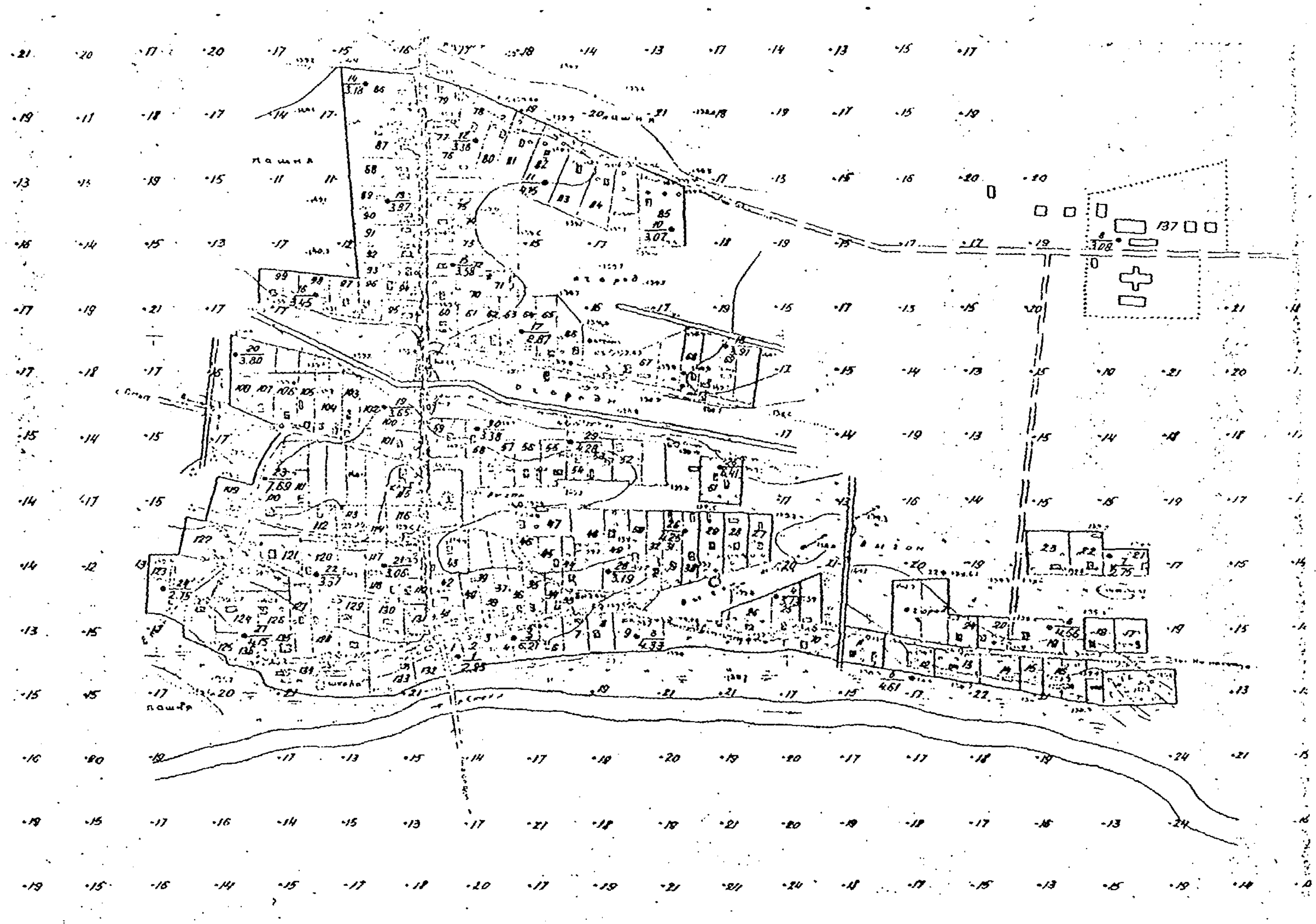


Fig. 3. Contamination characterization plan of the settlement to be restored

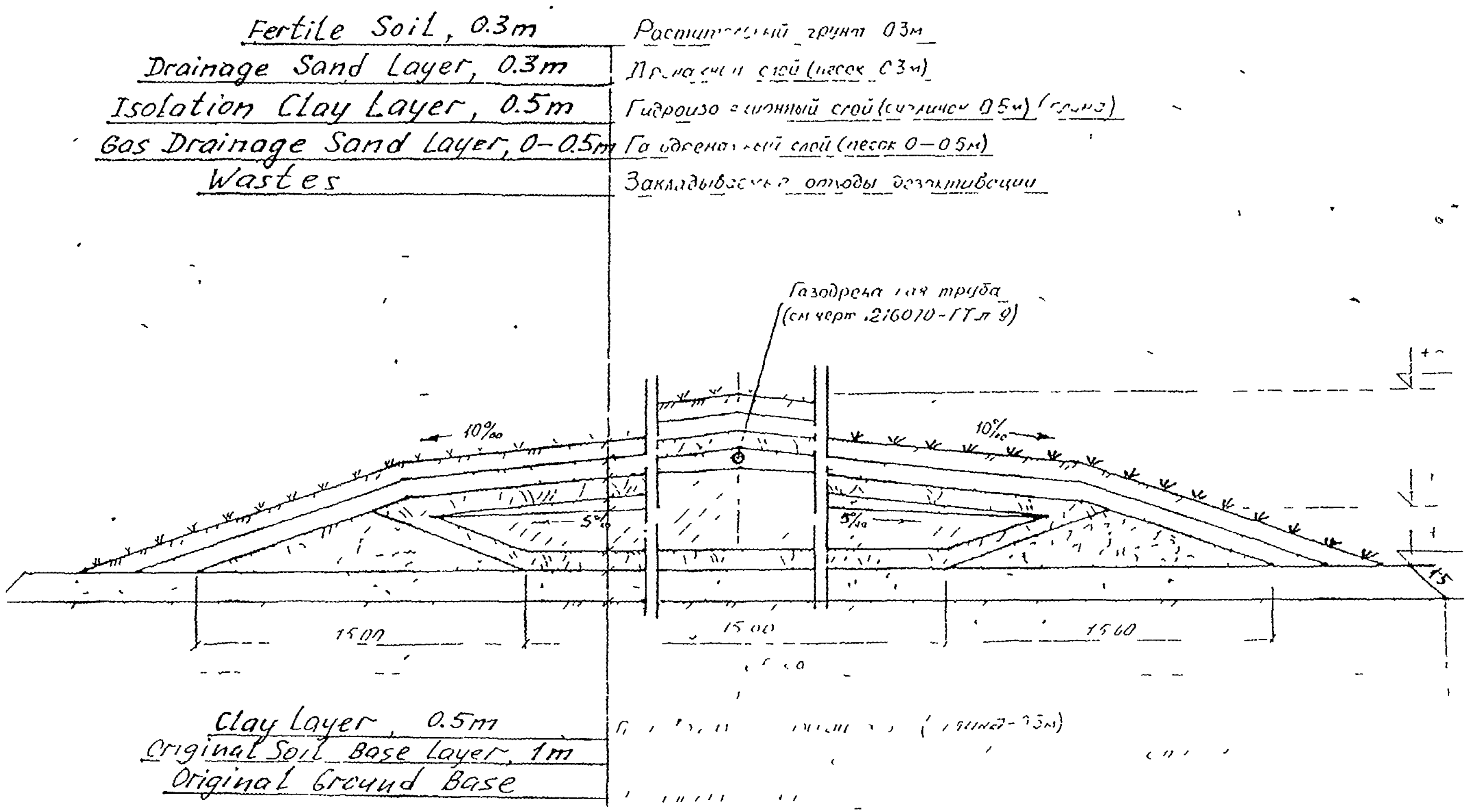


Fig. 4. Cross-section of the Waste Disposal Point (WDP) for wastes arising during restoration activities

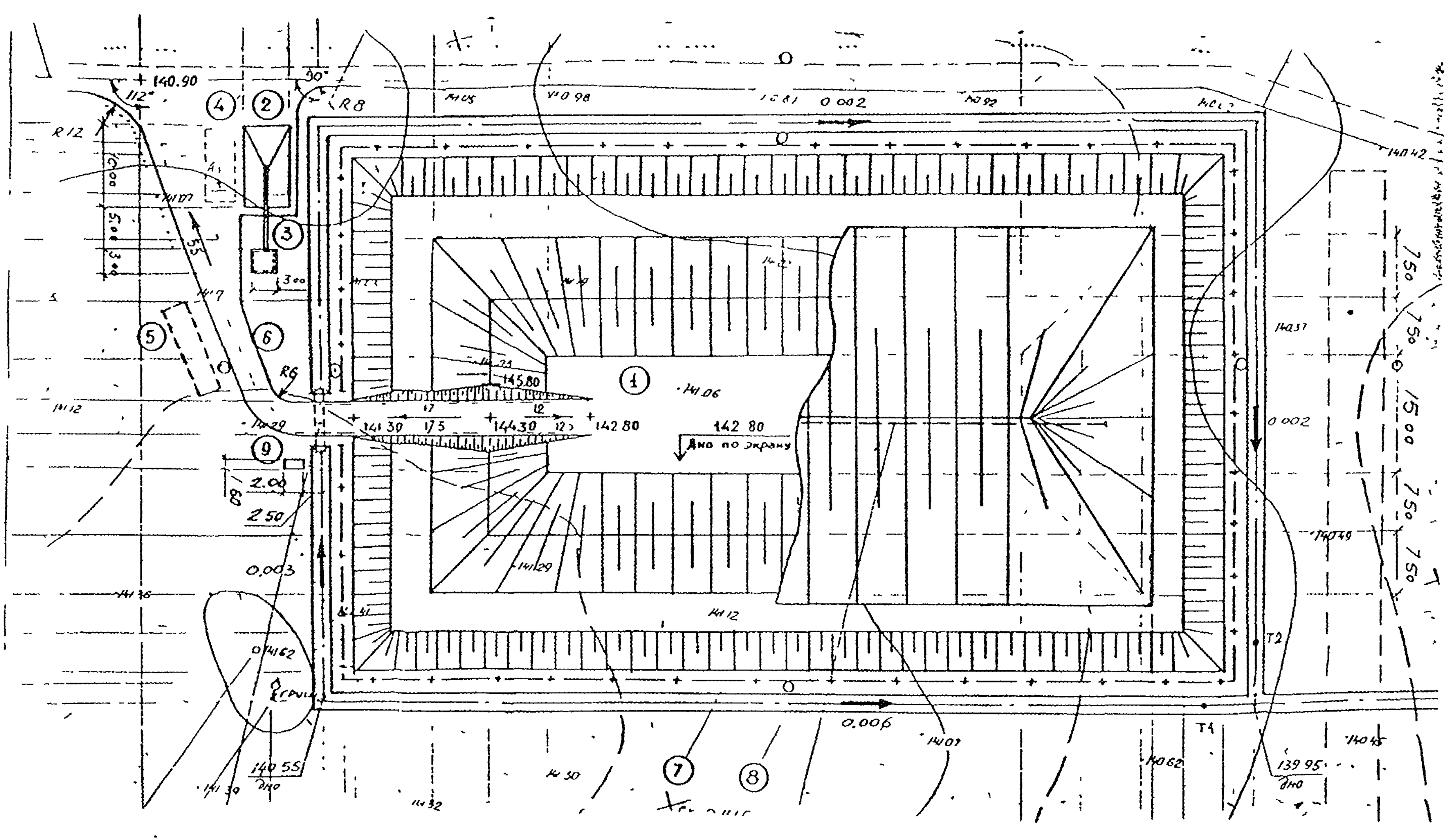
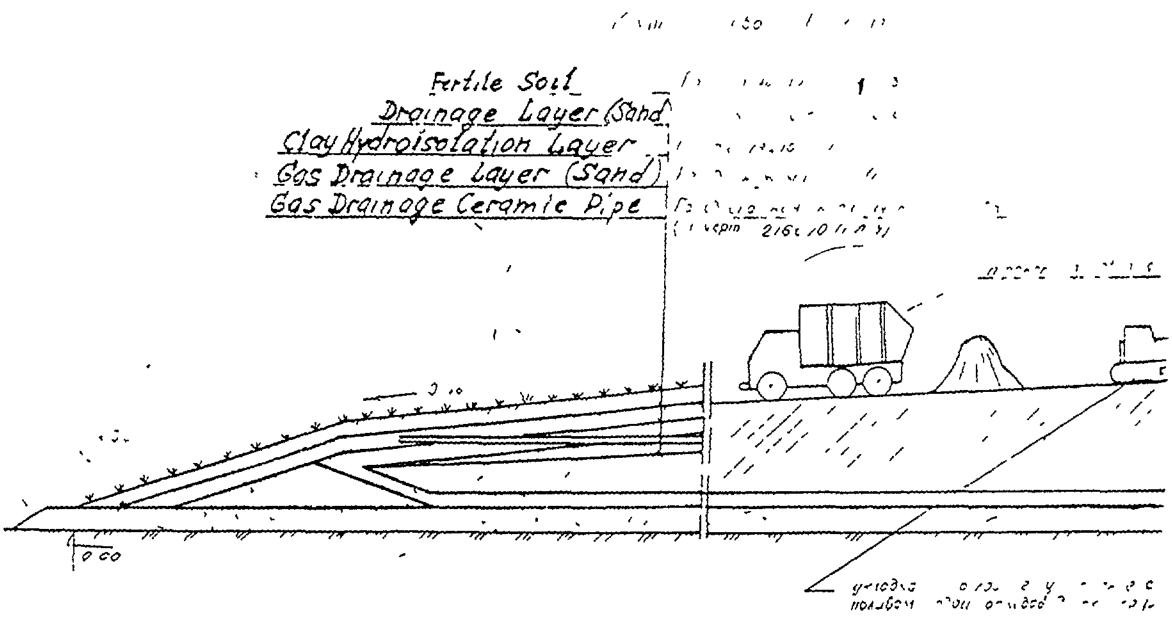


Fig. 5. Plan view of the waste disposal point trench



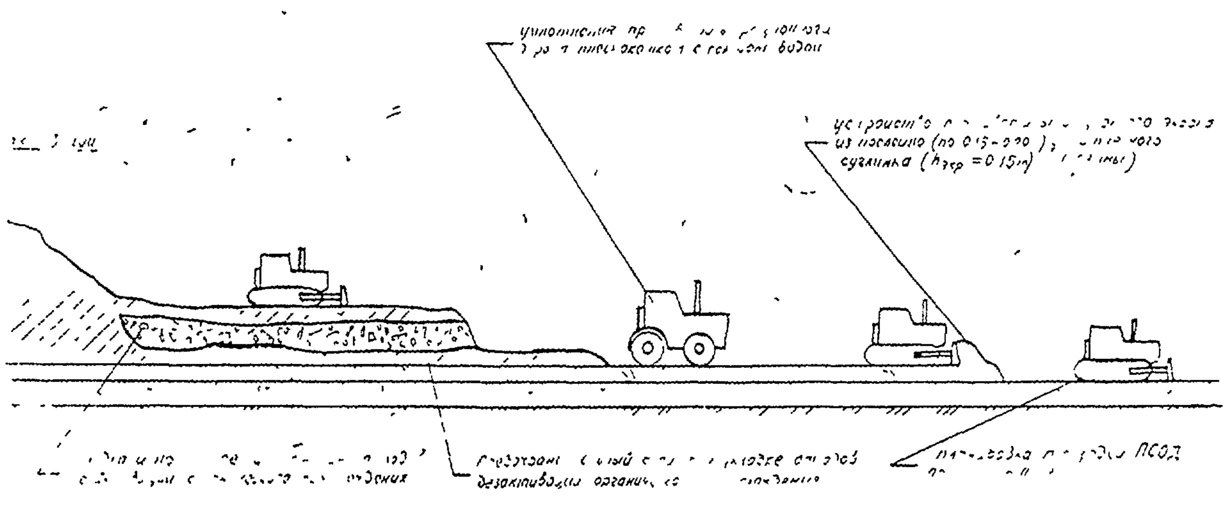


Fig. 6. Waste Disposal Point (WDP) preparation and filling scheme

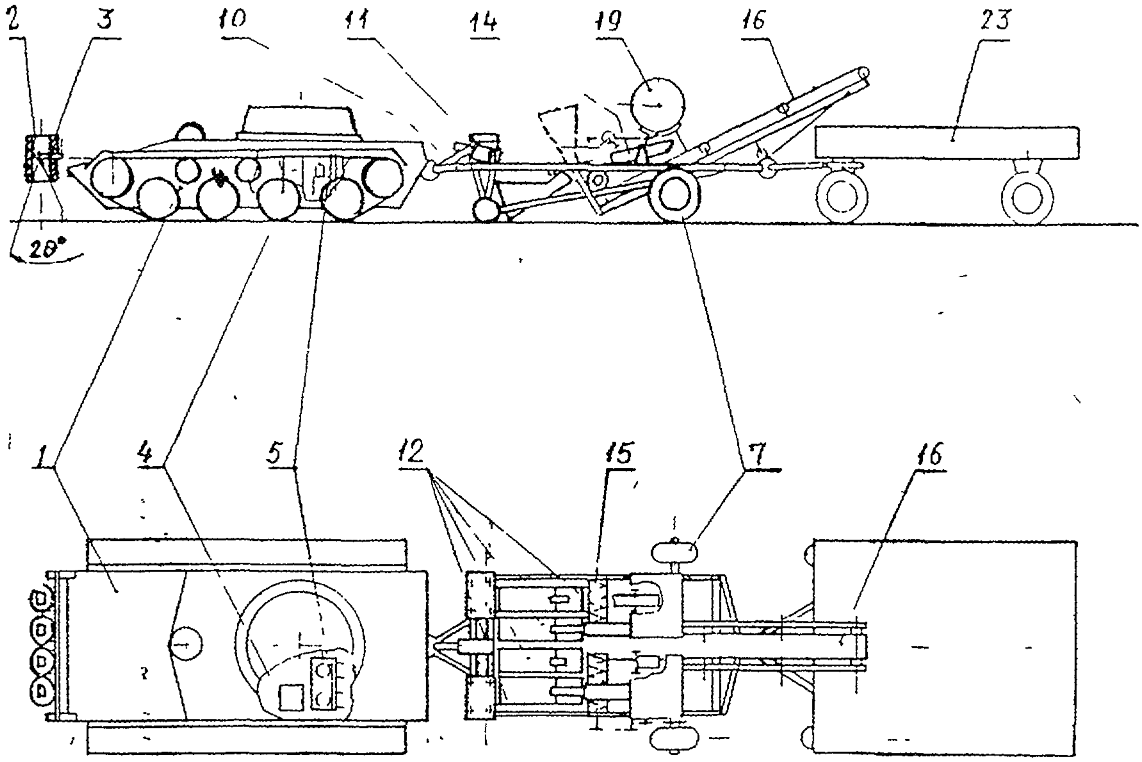


Fig. 7. Scheme and plan view of the soil removal installation (four - channel selective removal)

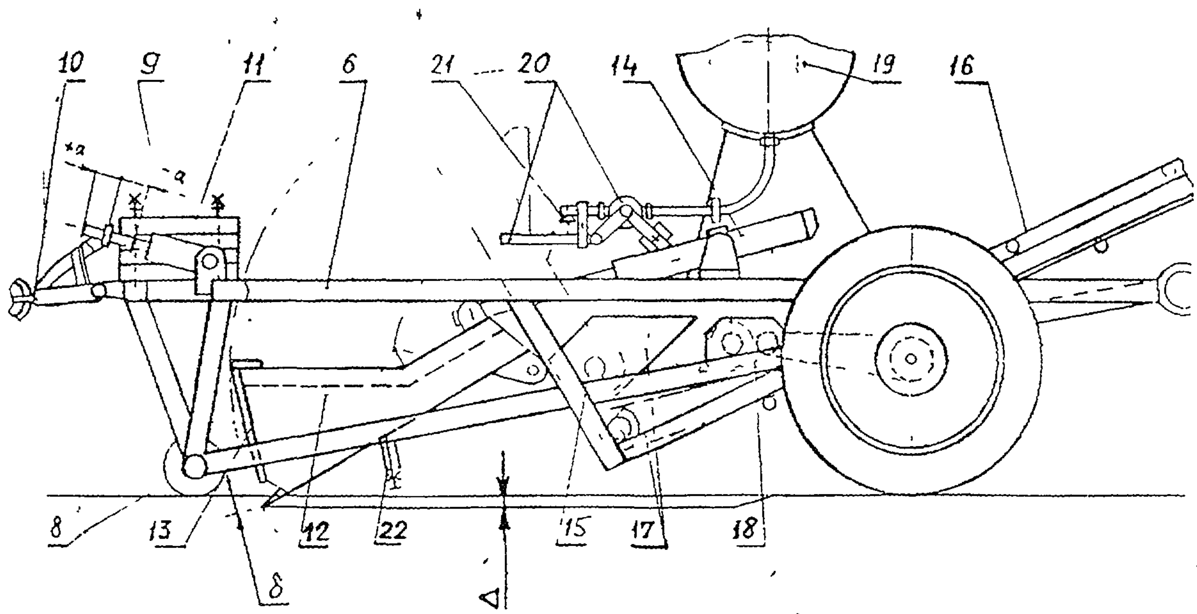


Fig. 8. Schematic view of the soil removal unit

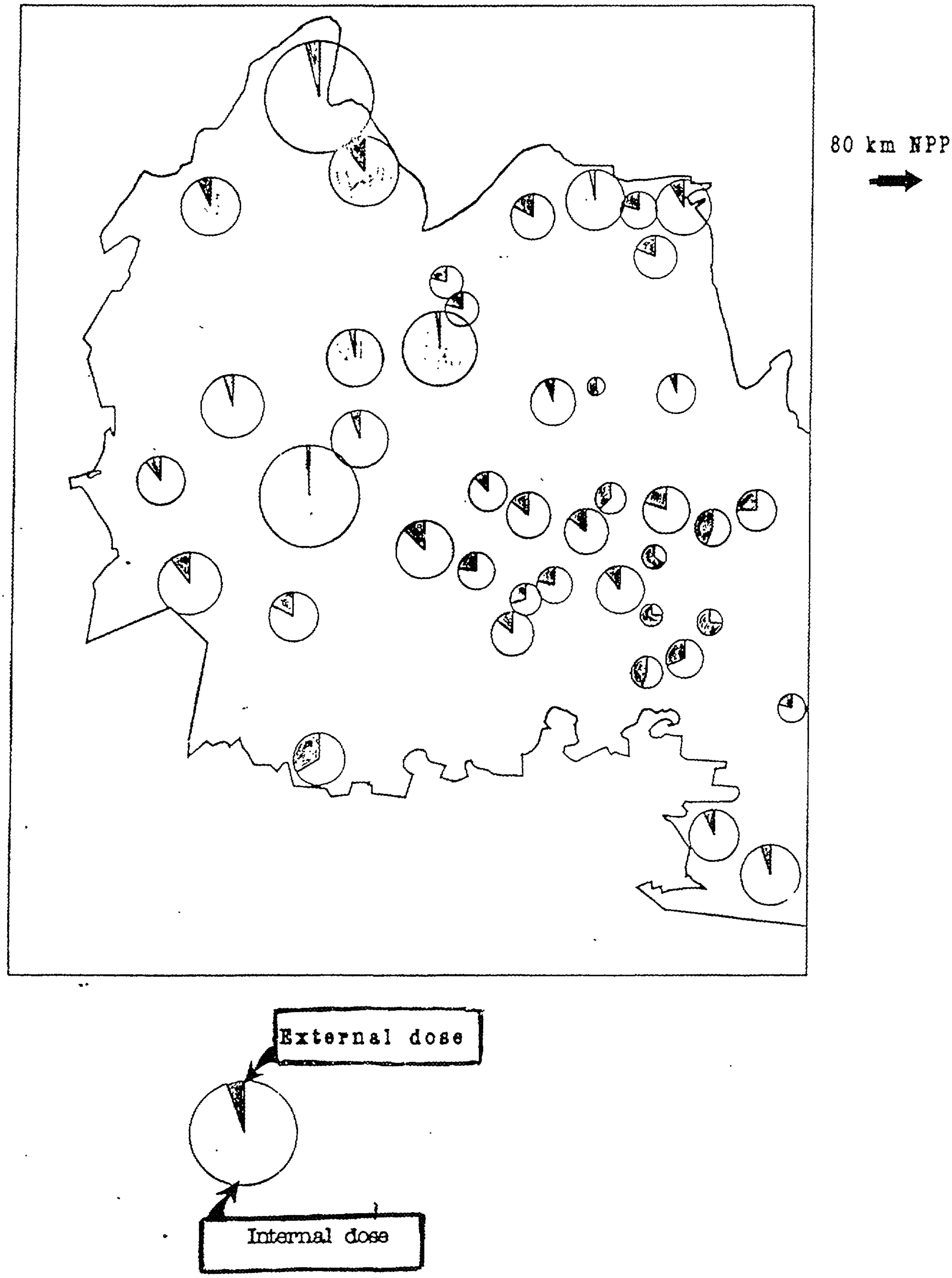


Fig. 9. Annual doses due to Chernobyl accident in Narodychy district (1991)

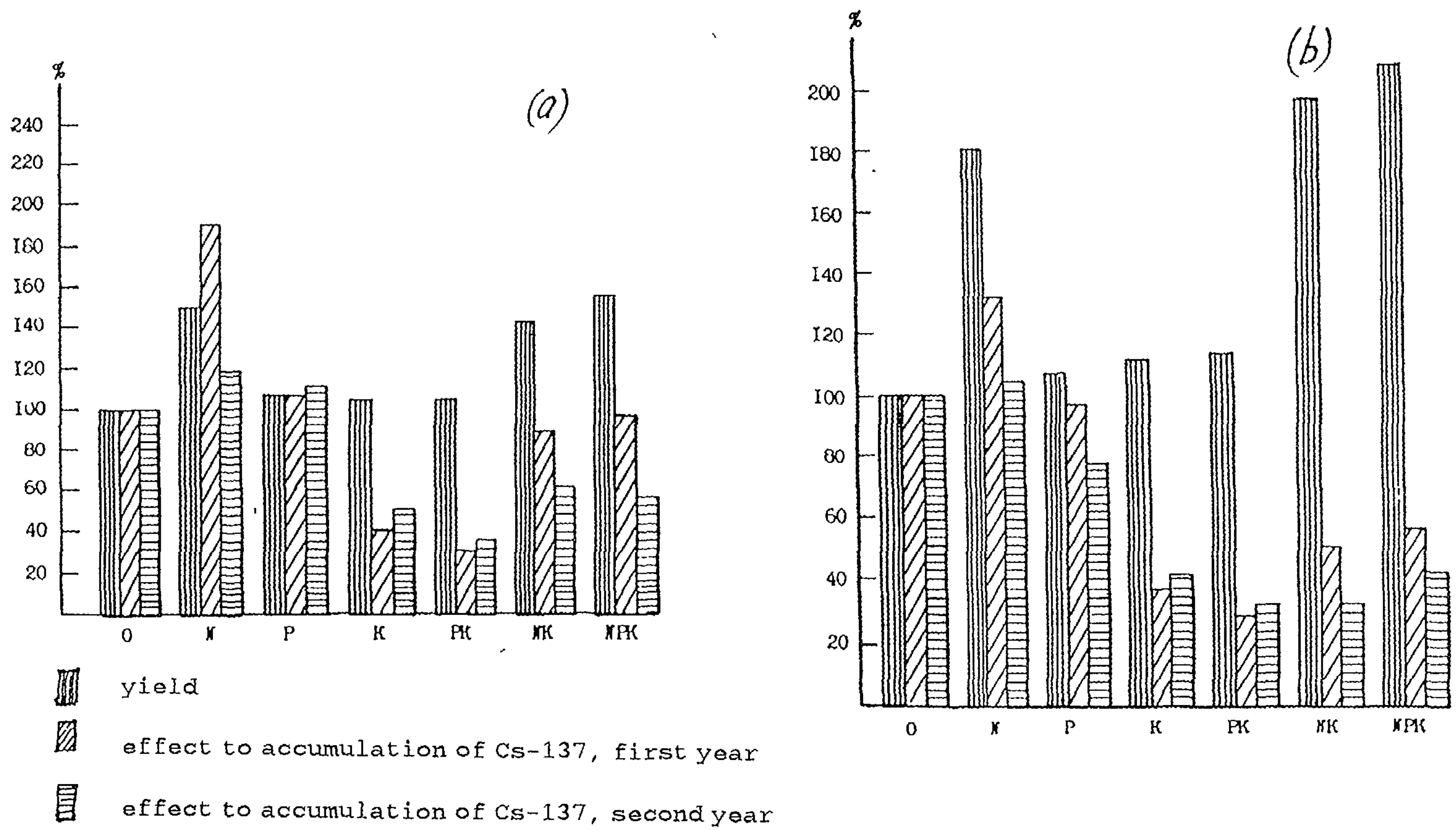


Fig. 10. Effect of different fertilizers to the yield of oats (a) and potato (b), and to the accumulation of Cs-137 in the first and following year after addition of the fertilizer

TABLE I DISTRIBUTION OF Cs-137 IN UPPER 25-CM SOIL LAYER OF NATURAL MEADOWS OF UKRAINIAN POLISSIA

Soil type	Content in soil layers, %				
	0-5см	5-10см	10-15см	15-20см	20-25см
Soddy-podzole subsandy	98.9	0.6	0.3	0.1	0.1
Soddy-podzol sandy	97.1	2.0	0.6	0.2	0.1
Drained peaty	97.0	1.8	0.6	0.4	0.2
Peaty meadow	94.9	2.9	1.1	0.7	0.4
Chornozem meadow	93.0	4.8	1.3	0.6	0.3
Light-grey podzole	86.7	12.0	0.7	0.3	0.3
Dark-grey podzole	84.0	14.9	0.4	0.4	0.3
Peat-boggy	77.5	17.3	3.3	1.5	0.4

TABLE II MELIORATION MEASURES IMPACT TO SOIL-PLANT Cs-137 TRANSFER FACTORS (TF) FOR PEATY-MEADOW SOIL

Measure	TF = Bq/kg в.-dry mass of pl. / kBq /sq.м		Caesium-137 concentr. decrease in plants, times
	before	after	
Drainage	189	71,0	2,7
Ploughing	71,0	25,9	3,0
Cultivation	71,0	17,5	4,0
N60	17,5	39,1	-2,2
P90	17,5	30,4	-1,7
K120	17,5	5,9	3,0
Manure, 50 t/ha	17,5	13,1	1,3
Liming, 1,5 t/ha	17,5	11,7	1,5
N60P90K120	17,5	13,8	1,3
N60P90K120+liming	17,5	4,3	4,1
Substantial melior.	71,0	4,3	16,5

TABLE III Cs-137 TRANSFER FACTORS FOR DIFFERENT FORAGE CROPS

Crops	TC*
Forage cabbage (green mass)	22,0 + 5,0
Klover (green mass)	9,2 + 0,7
Bean (green mass)	8,9 + 0,3
Raps (green mass)	8,4 + 0,9
Turneps (roots)	8,0 + 1,0
Cereals (green mass)	4,1 + 0,2
Oats (green mass)	3,9 + 0,3
Forage beat (roots)	2,7 + 0,3
Mais for silage	1,6 + 0,2
Oats (straw)	1,5 + 0,1
Barley (green mass)	0,8 + 0,1
Potato (dubers)	0,7 + 0,2
Oats (grain)	0,7 + 0,1

$$* \quad TF = \frac{\text{Bq/kg в.-dry mass of the plant}}{\text{kBq /sq. м}}$$

TABLE IV Cs-137 TRANSFER FACTOR FROM PEATY SOIL TO FORAGE CROPS

Trial option	$TF = \frac{\text{Bq/kg, humid mass}}{\text{kBq /sq. m}}$				
	cabbage forage	mais silage	beat forage	turneps	oats
Reference *	90,0	10,0	3,0	6,3	16,0
N60P90K120 (backgr)	20,6	2,4	1,0	2,0	3,7
N60	45,0	9,5	2,0	6,9	15,4
P90	61,0	8,0	1,2	6,3	21,0
K120	45,6	1,3	0,5	3,5	10,6
N60P90	31,0	5,6	0,5	2,6	6,3
P90K120	16,5	2,4	0,6	3,0	4,0
N60K120	28,0	2,2	1,0	2,8	4,8
Liming, 1,5 Hr	44,0	9,3	2,8	7,8	12,4
Manure, 50 t/ha	36,0	3,0	1,3	2,2	5,0
Backgr + manure, 50t/ha	9,0	0,6	0,5	0,9	1,6
Backgr + liming 1,5 Hr	10,0	1,0	0,6	1,2	1,5
Backgr + liming 1,5 Hr+ + manure 50 t/ha	12,4	1,8	0,5	1,0	2,4

* - without fertilizers

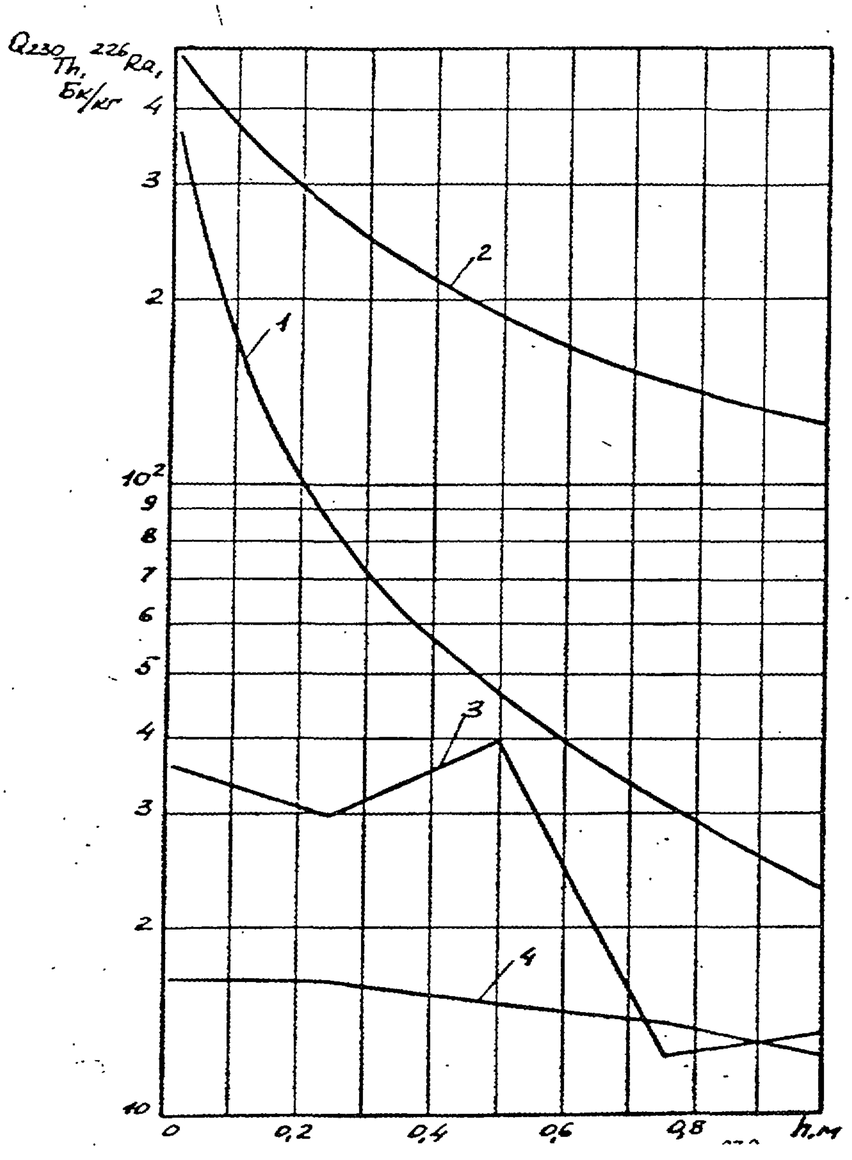


Fig. 11. Th-230 and Ra-226 concentration in the soil at the site of relocated depleted uranium ore stock (1) and (2), respectively; (3), (4) - background curves

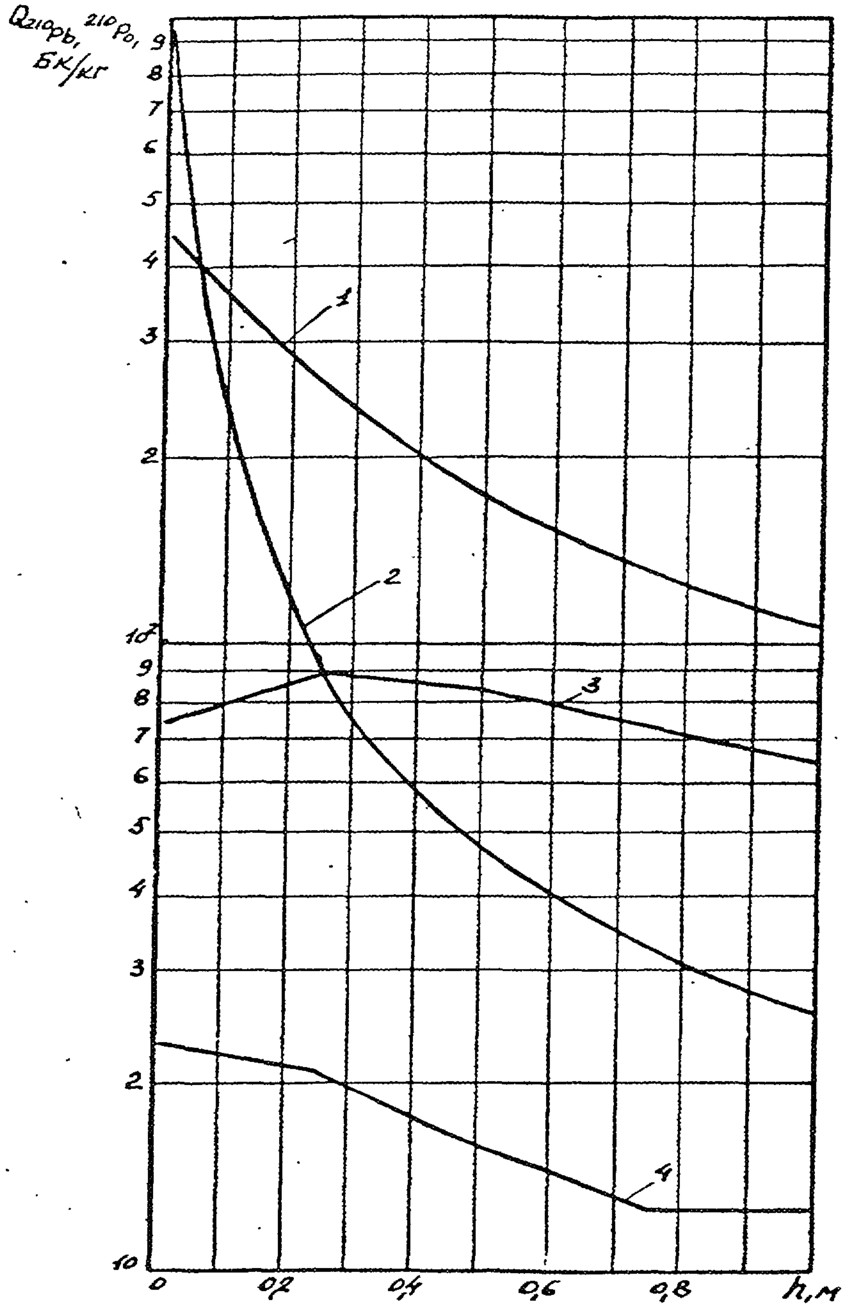


Fig. 12. Pb-210 and Po-210 concentrations in a soil at the site of relocated ore stock, as a function of depth. (3), (4) - reference background concentrations, respectively.

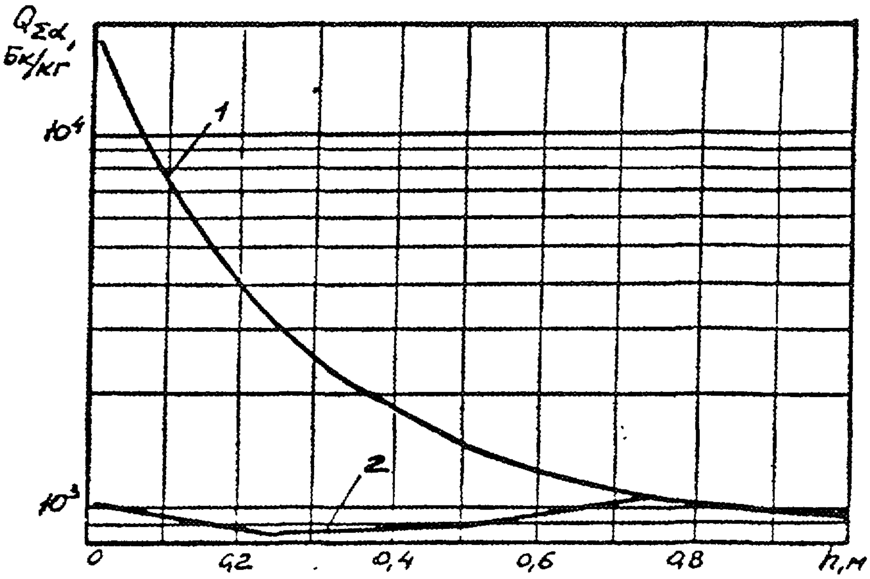


Fig. 13. α-activity of soil at the site of relocated depleted uranium ore pile (1), and background curve (2)

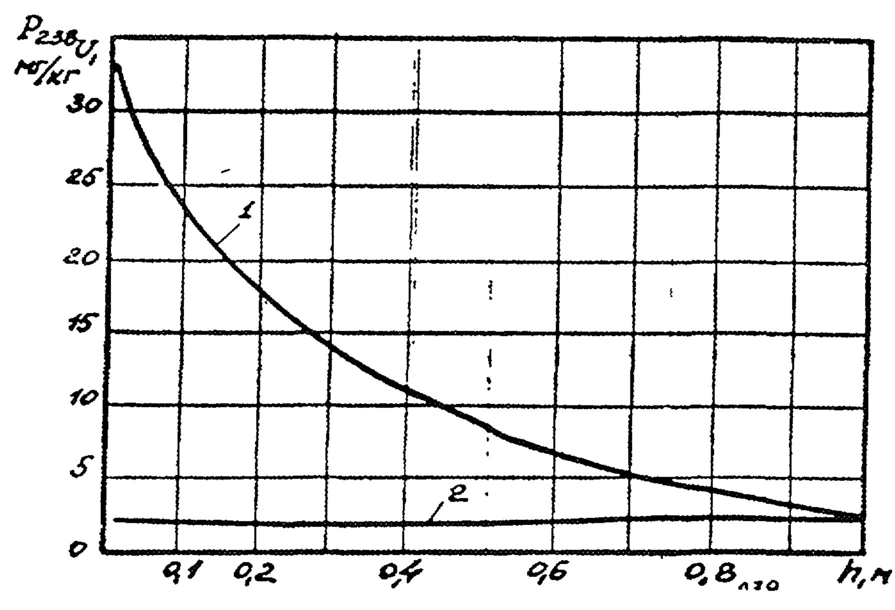


Fig. 14. Uranium-238 content in a soil at the site of relocated depleted uranium ore pile (1), and background concentration (2).

TABLE V 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-210 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 ± 2,8	40 ± 28	26 ± 13	178 ± 85	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

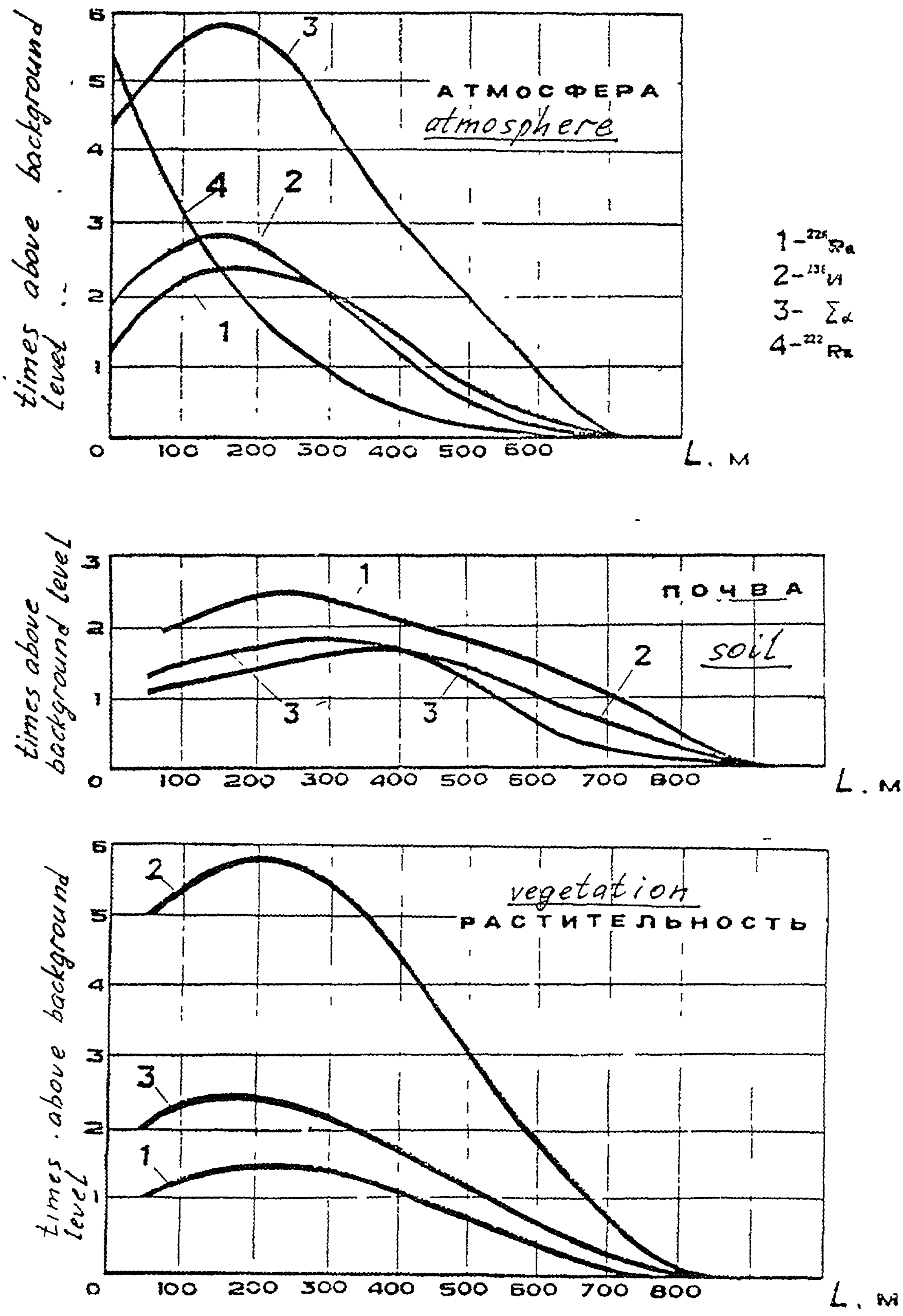


Fig. 15. Contamination of the Uranium waste rock pile vicinities (against the background level)

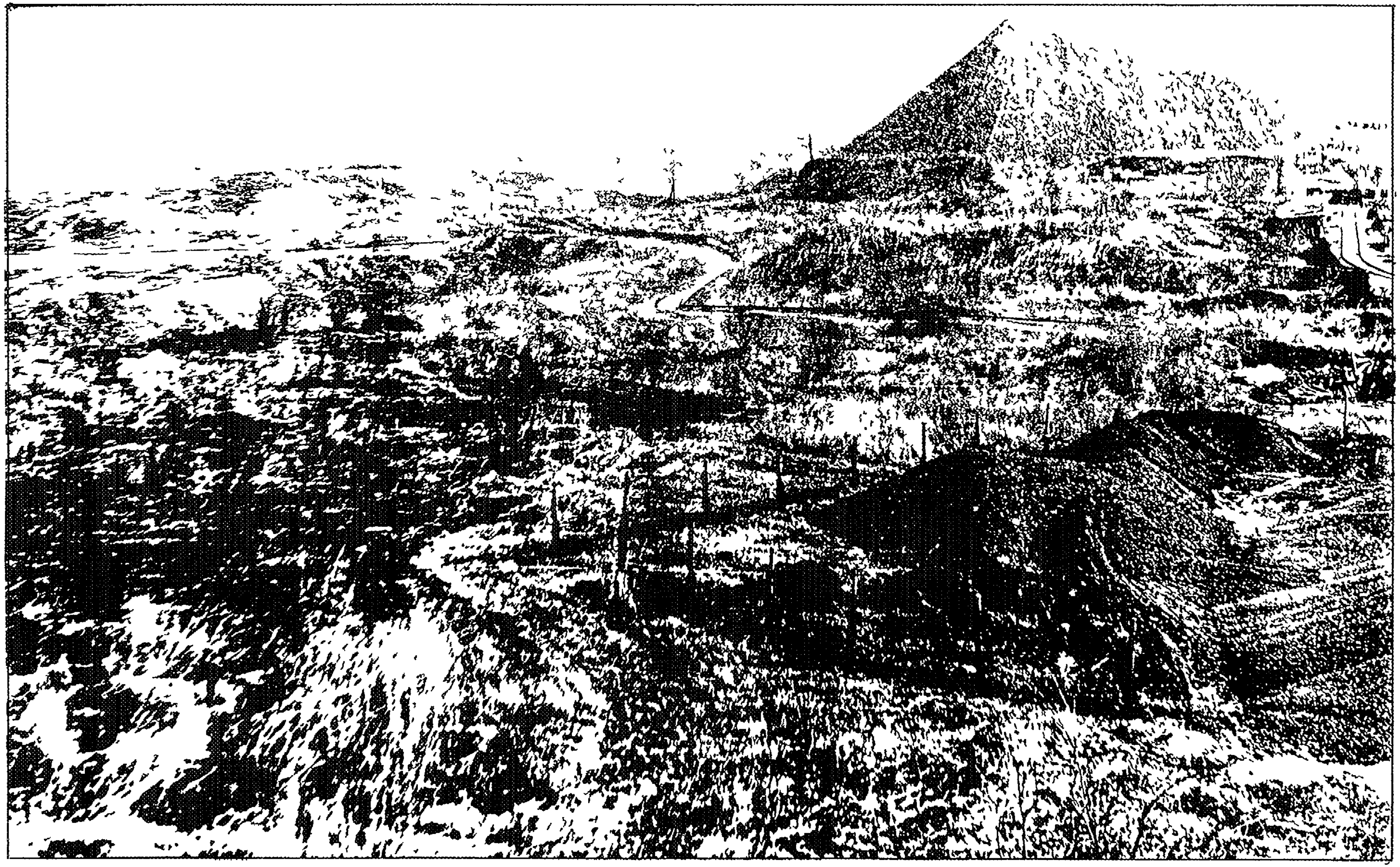


Fig. 16.

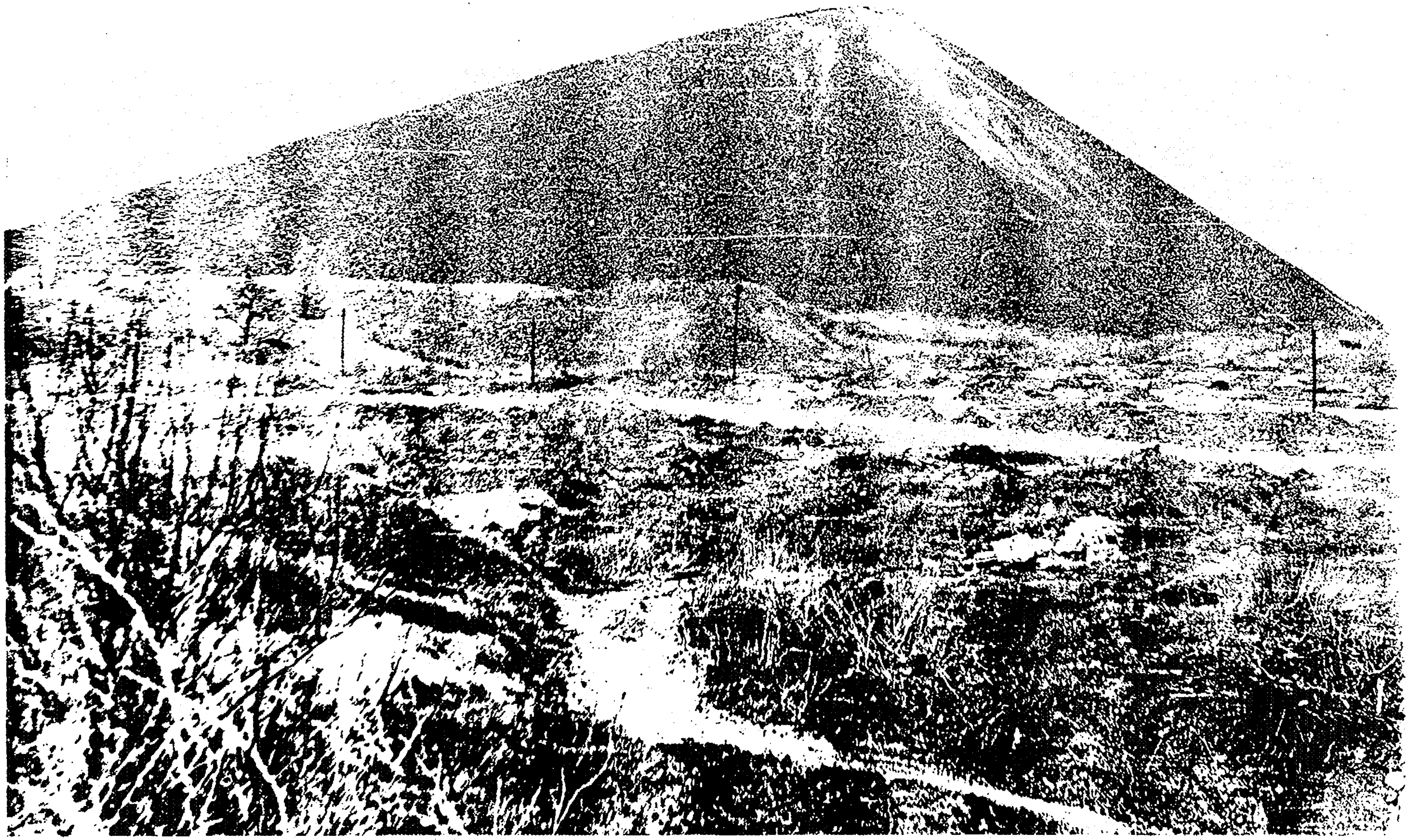


Fig. 17.

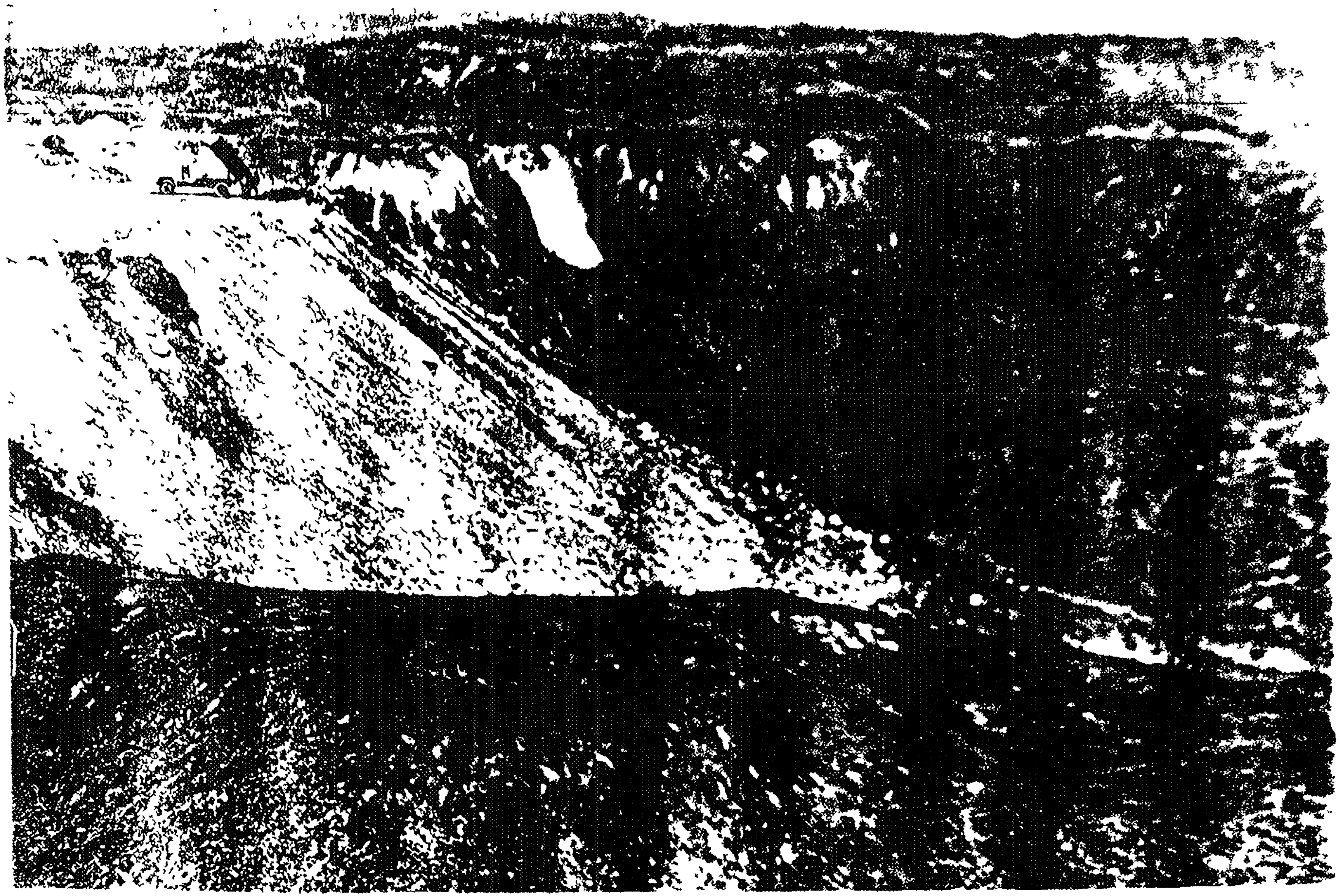


Fig. 18.

Fig. 19.

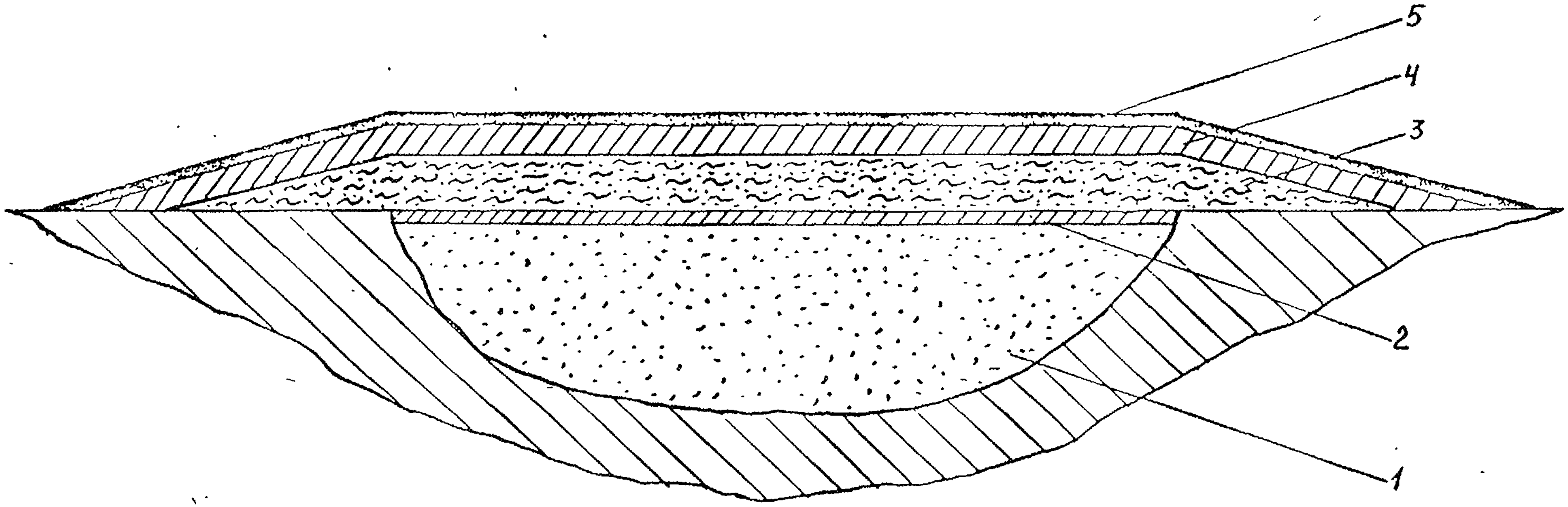
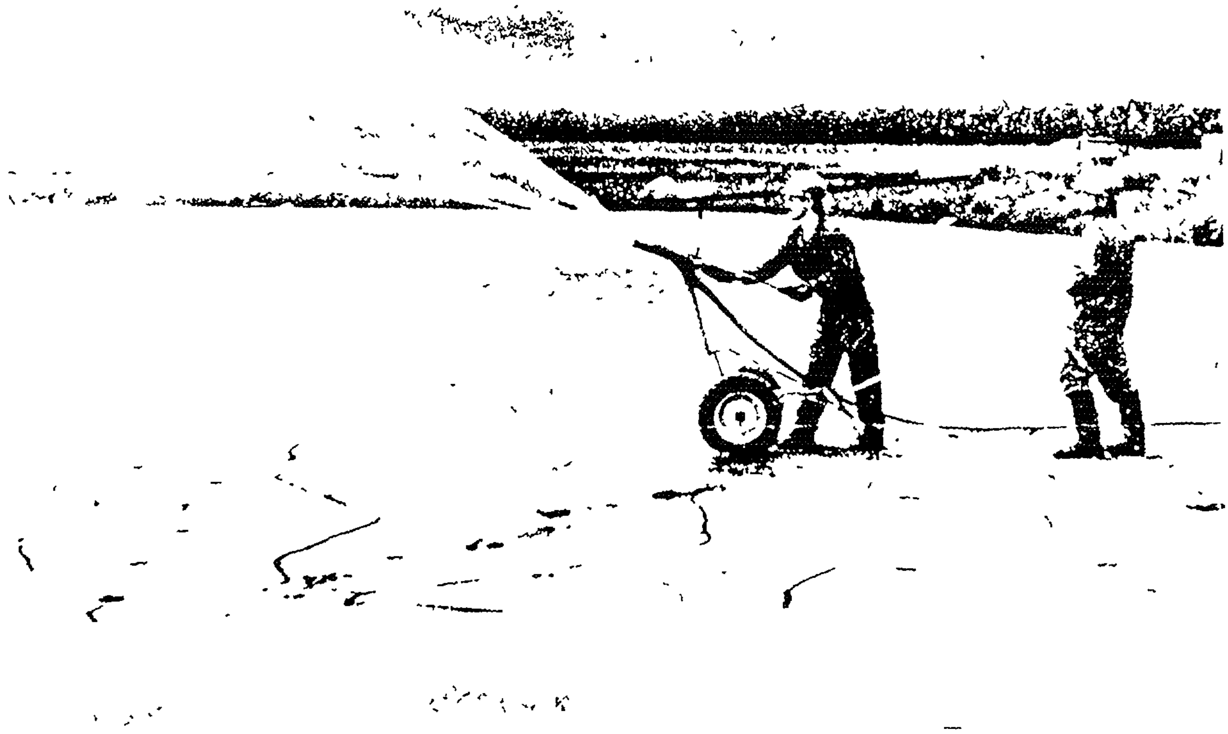


Fig. 20. Cross-section of the mill tailing in Zovti Vody, after restoration works:
1 - tailings
2 - clay layer (0.4 m)
3 - conventional waste (2.0m)
4 - clay layer (1.1m)
5 - fertile soil layer (0.4 m)



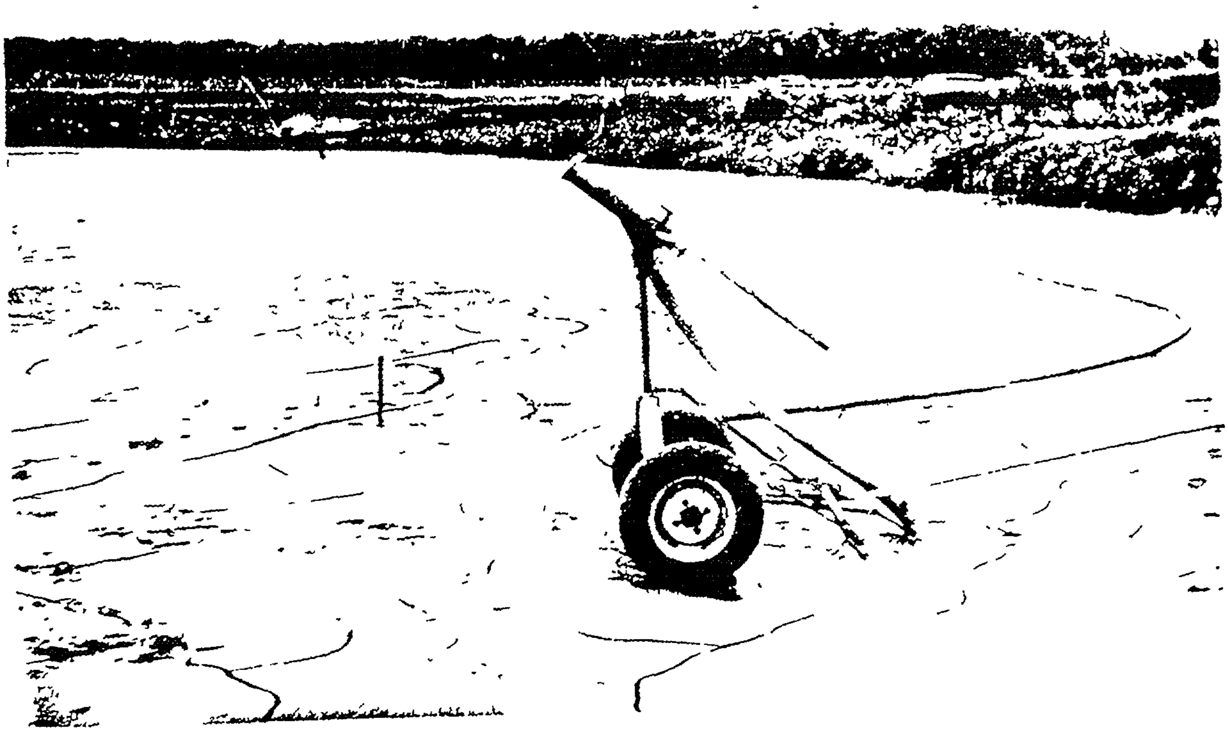


Fig. 21. Jet monitor unit for covering of the dry mill tailing beaches with polymeric compound

Fig. 22. Mixing of mill tailing discharge with a water solution of "K-9" polymeric compound

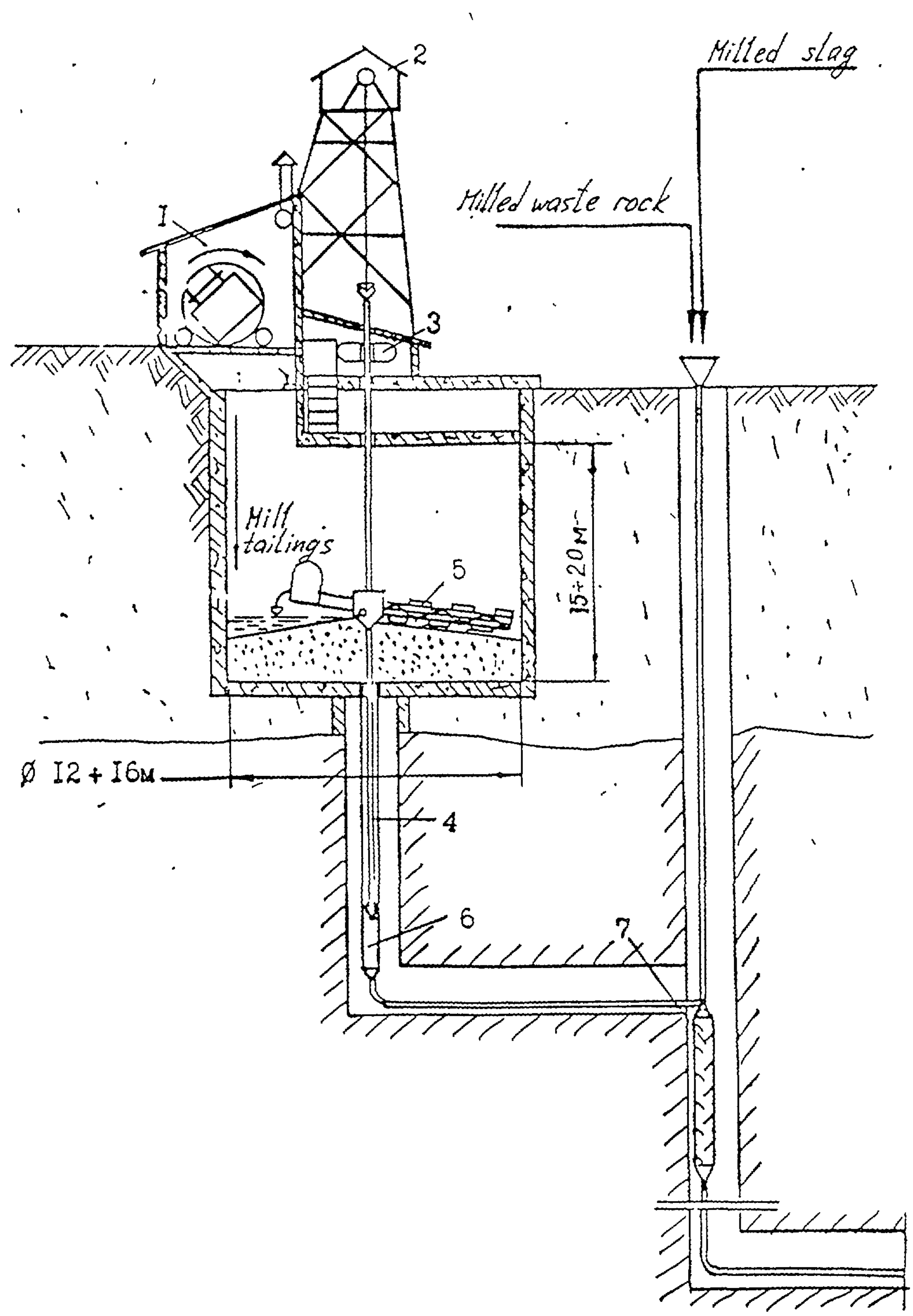


Fig. 23. Schematic diagram of filling complex using uranium mill tailings:
1 - railway car turnover mechanism
2 - lifting mechanism
3 - turning unit
4 - movable pipe with a
5 - mill tailing slurry supply system
6 - slurry supply control unit
7 - mixing header

TABLE VI COSTS FOR TWO OPTIONS OF WASTE PILE ROCK RESTORATION, KIROVOGRAD SITE

Parameter	Unit	Value	
		Option I	Option II
1. Land area damaged by mining	ha	40	40
2. Land area under waste rock pile after rehabilitation	ha	31	28
3. Additional areas nesessary for disposal of waste rock from piles	ha	-	11,4
4. Duration of restoration works	month	36	48
5. Costs for restoration (1984)	ruble	3815	11637

TABLE VII PLANNED CHARACTERISTICS OF DIFFERENT COVER LAYER COMPONENTS FOR ZHOVTI VODY URANIUM MILLING TAILING (PRICES OF 1984)

Cover blanket structure	Blanket thikness	Specific cover blanket volume, thousand cub. m per 1 hectar	Price of work per 1 cub. m, rubles	Price of work per 1 hectar, thousand rub.	Specific price of 1 ha rest. 1000 rubl.	Total restoration expenditures of the mill tailing (45.1 ha), thousand rubles
Fertile Soil	0.4	4.0	0 72	2.88		
Clay	1.1	11.0	0 66	7.26		
Waste rock	0.4	4.0	0.70	2.80	15.58	702.7
Thermal PP ash	0.4	4.0	0.66	2.64		
Fertile Soil	0.4	4.0	0.72	2.88		
Clay	1.1	11.0	0.66	7.26	19.94	899.3
Waste rock	0.4	4.0	0.70	2.80		
Clav	0.4	4.0	0.66	2.64		
Polyethilene film (0.2mm)		10.0*	0.436	4.36		
Fertile Soil	0.4	4.0	0.72	2.88		
Clay	3.0	30.0	0.66	19.80	28.12	1268.2
Waste rock	0.4	4.0	0.70	2.80		
Fertile Soil	0.4	4.0	0.72	2.88		
Clay	1.1	11.0	0.66	7.26	43.23	1949.7
Asphalt concrete	0.1	1.0	33.09	33.09		
Fertile Soil	0.4	4.0	0.72	2.88		
Clay	1.1	11.0	0.66	7.26	75.54	3406.9
Cement concrete	0.2	2.0	32.70	64.40		

* Thousand sq. m

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METHODS AND TECHNIQUES USED TO REHABILITATE RADIOACTIVELY CONTAMINATED SITES IN THE UNITED KINGDOM

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Abstract

In the early years of use of radioactive materials the quality of control and waste management practices were of a considerably lower standard than those acceptable today. As a consequence a large number of sites in the UK became contaminated to varying degrees with radioactive materials. The vast majority of these sites were very small in size and were linked to either radium or thorium operations or research applications. The net consequence of the above has been that the vast majority of the rehabilitation work in the UK has been on a small scale and has not merited the use of innovative techniques. However, a number of innovative remediation techniques are under development in the UK for future cleanup programmes and some have attained full-scale application. These will be described in this paper. Three development areas are described in detail, namely (1) the application of separation processes from minerals processing (2) the BNFL 'Cacitox' process and (3) electroremediation.

1. INTRODUCTION

Radioactive materials have been extensively used throughout the United Kingdom since the last century. Initially these were naturally occurring materials, such as thorium and radium, which were used on account of their luminising or incandescent properties on clock and instrument dials, signs, gas mantles, etc. With the advent of nuclear power and weapons programmes and the use of radioisotopes in research and medicine, the range of materials and applications and the scale of their use has increased dramatically. The United Kingdom has a substantial civil nuclear power programme and is a major fabricator and reprocessor of nuclear fuel. It has also had an independent nuclear weapons programme since the late 1940's. As a consequence it has a large number of major sites where nuclear materials are handled, used and stored.

In the early years of use of radioactive materials the quality of control and waste management practices were of a considerably lower standard than those acceptable today. As a consequence a large number of sites became contaminated to varying degrees with radioactive materials. The vast majority of these sites were very small in size and were linked to either radium and thorium operations or research applications. On the major nuclear sites the control of radioactive materials has generally been very much better and any contamination has been from inadequate long term waste storage practices or leakages from process plant. Contamination on these sites has generally been localised and has been cleaned up upon detection. Alternatively, where it poses no risk to the general environment or the workers on site, it has been left under control and will be cleaned up during final decommissioning of the associated plant or site. All testing work associated with the UK's development of nuclear weapons has been carried out on test ranges located in the USA or in the southern hemisphere. As a result, it has not led to contamination requiring remediation in the UK.

The net consequence of the above has been that the vast majority of the rehabilitation work in the UK has been on a small scale and has not merited the use of innovative techniques. Indeed, the majority of sites have been cleaned up by simply removing the contaminated soil and other materials and segregating them in terms of their activity levels for disposal.

A number of innovative remediation techniques are under development in the UK for future clean-up programmes and some have attained full-scale application. These will be described in following sections.

2. CATEGORIES FOR ACTIVE WASTE DISPOSAL IN THE UNITED KINGDOM

A key factor in the rehabilitation of radioactively contaminated sites in the UK is the identification of acceptable and cost effective disposal routes for any resulting wastes. The categories for wastes in the UK are:

- VLLW - Very low level waste ("Dustbin" level)
  < 400 kBq $\beta\gamma$ and < 0.1 m^3 or < 40 kBq $\beta\gamma$ per item. Such wastes do not require specific authorisation under the Radioactive Substances Act (RSA), 1960.

- LLW - Low level waste
  < 4 GBq α/te and < 12 GBq $\beta\gamma$/te

- ILW - Intermediate level waste
  $>$ LLW and $<$ HLW

- HLW - Wastes which generate a significant amount of radiogenic heat.

The VLLW wastes can be disposed of to selected landfill sites, which receive domestic rubbish and are supervised by local district councils. Some wastes, which are slightly higher in activity than the VLLW limits and hence are in the LLW category, can also be disposed of in authorised local landfills. These wastes are covered by Exemption Orders under the amended RSA 60 of 1993 and are listed under individual Statutory Instruments, which are regulatory orders.

There are 22 Statutory Instruments in this category and they cover materials, such as phosphatic substances and rare earths, etc. (No. 2648 of 1962); prepared uranium and thorium compounds (No. 2711 of 1962); gaseous tritium light devices (No. 1047 of 1985); luminous articles (No. 1048 of 1985) and smoke detectors (No. 953 of 1980). As an example of their use, Statutory Instrument No. 2648 treats Ra at levels between 0.37 Bq/g and 4.59 Bq/g and Th at levels between 2.59 Bq/g and 7.4 Bq/g as radioactive, but exempts them from the requirements of RSA 60. As such these materials can be disposed of by controlled burial in landfills.

LLW can be disposed of in the near surface repositories at Drigg in Cumbria and Dounreay in Caithness, Scotland. These two repositories are operated by BNFL and UKAEA, respectively. Given its location and limited size, the Dounreay repository has been used almost exclusively for UKAEA wastes arising in Scotland. Hence, it has not been used generally as a disposal site for wastes from the clean-up of other sites. In the next decade it is planned that low level wastes will be disposed of in the new deep geological repository being developed by Nirex.

ILW are currently being kept in engineered stores throughout the UK until the new Nirex repository is available early in the next century. HLW, which primarily arises from the reprocessing of spent power reactor fuel, is being vitrified at Sellafield and held in air-cooled stores for 50 years before disposal in an as yet to be defined repository.

The costs associated with the conditioning, storage, transport and disposal of these wastes increase by at least an order of magnitude as the waste category rises. With these costs being of the order of £1-3k/m^3 for low-level waste, it is quite apparent that waste costs can be very significant in any rehabilitation programme. As a consequence a very important objective in any rehabilitation programme is to minimise the quantity and category of any waste. This has led to great emphasis being placed on very careful segregation of wastes at source to avoid increasing active waste volumes.

3. CONVENTIONAL SITE REHABILITATION PRACTICES

The conventional approach to the rehabilitation of radioactively contaminated land in the UK is to use detailed prior planning coupled with rigorous project management in execution to ensure that the objectives of the remediation programme are fully met. Extensive consultations are undertaken with the various regulatory bodies, such as Her Majesty's Inspectorate of Pollution (HMIP), Department of Transport (DTp), Nuclear Installations Inspectorate (NII) for nuclear licensed sites, Ministry of Agriculture and Fisheries (MAFF), etc., who have statutory responsibilities for ensuring the protection of workers, the public and the environment.

The approach involves:

- detailed characterisation of the contaminated site to determine the nature and extent of the contamination.

- assessment of the risks to workers, the public and the environment from the contamination and each proposed remediation option. These assessments of risk can be semi-quantitative, involving comparisons acceptable levels of contaminants in soil, the air and ground and surface waters. They can also be quantitative, involving assessment of potential pathways and risks to most exposed individuals and groups, as well as the general population.

- selection of the preferred remediation strategy and approval of that strategy by regulatory bodies. The selection is usually based on a cost-benefit analysis and is affected by the location of the site, the proximity of receptors and the proposed end use of the land. In selecting clean-up technologies "best available technologies not entailing excessive cost" (BATNEEC) and "best practicable environmental options" (BPEO) are used. The clean-up standards and the methodology to be used are agreed with regulatory bodies.

- detailed design of selected processes.

- implementation of selected strategy to rehabilitate the site.

- verification of achievement of agreed clean-up targets by monitoring, sampling, etc.

In practice for most radioactively contaminated sites, this has led to the wastes being excavated and then disposed of where possible as VLLW or LLW. Excavation has routinely been carried out using conventional earth moving equipment, such as bulldozers, backhoes, excavators, front end loaders and scrapers, or by using hand tools if the areas involved are very limited. All excavations are performed under a very strict health physics control regime. Where necessary dust suppression techniques, such as wetting down using sprays or water carts or the use of temporary containment structures, have been applied. As the wastes are removed they are carefully monitored either by hand-held instruments [1] or if the volumes justify by the use of dedicated monitoring systems using arrays of detectors and conveying systems [2]. Staff are supplied with protective clothing and respiratory protection commensurate with the risks posed and personal and areal air sampling regimes are often employed. Examples of this approach have been given by Drury[1] and Fellingham et al[2].

Even for the clean-up of very large areas, such as occurred at the former British nuclear weapons test site at Maralinga in South Australia [3], a similar approach to rehabilitation was adopted except that the wastes were buried on site. For that clean-up operation, soil was only removed around the firing pads and elsewhere surface activity levels were reduced by ploughing to mix and hence dilute surface contaminated layers with clean underlying soil. In addition, in the most contaminated areas clean topsoil was deposited over the ploughed soil. Such measures were aimed at reducing inhalation risks to nomadic people passing through the semi arid areas.

4. ADVANCED REHABILITATION TECHNOLOGIES

A number of new soil treatment technologies have been developed or are under development in the UK. Several of these are derived from conventional minerals processing technologies and result in substantial reductions in the quantities of active waste requiring storage and disposal.

4.1 Application of Separation Processes from Minerals Processing

Physical processing techniques are being used to develop multi-stage, integrated processes for the ex-situ treatment of contaminated land with the aim of separating, isolating and concentrating the contaminants to leave bulk streams which are much less contaminated [5]. These streams can then be either returned to the site as "clean" material or sent for disposal in landfills for non-active waste.

The basis behind the success of physical separation techniques is that contamination often occurs selectively on the finer fraction particles. These can be separated from the larger, bulk of the particles by exploiting differences in grain size, settling velocity, particle density, surface chemical properties, magnetic susceptibility, etc. The validity of this approach has been demonstrated in the US clean-up of the Johnston Atoll nuclear test site [6] and in results of Pu enhancement in finer soil fractions reported in studies of the Maralinga test site [3].

A wide range of particle separation equipment is being evaluated. These include by exploitable feature:

- Size - sieves and screens;

- hydraulic size (settling velocity) - classifiers, hydrosizers and hydrocyclones;
- specific gravity - jigs, sluices, dense media separators, spirals, shaking tables, tilting frames, vanners, duplex and multi-gravity separators;
- surface chemistry - froth flotation systems; and
- magnetic susceptibility - low intensity magnetic drums, induced and high intensity magnetic separators.

Pilot scale work is currently in progress at the UK National Environmental Technology Centre at Harwell [5] to develop the most effective combinations of these separation techniques for different types of soil and contaminants.

4.2 The BNFL 'Cacitox' Process

A very comprehensive soil treatment system is BNFL's EXCEL*CR™ [4], which is shown schematically in Figure 1. This is a skid-mounted, modular system, which can be rapidly deployed as a transportable or permanent facility. It includes modules that enable it to treat all types of soils, including clays, and it can process soils with radioactive, heavy metal and organics contamination. The system involves five main stages:

- size classification. This stage concentrates contamination when it is primarily associated with the fines fraction. It uses commercially available and proven plant, such as screens, wet classifiers and flotation units, to remove the oversize material. Wet attrition is used to free gravel and sand particles from clay. Hydrocyclones can then separate the clean coarse fraction from the contaminated fines fraction.
- organics treatment. This is particularly relevant for some mixed waste sites. The properties of the organic contaminants determine what treatment stages might be used, but typical examples would be air or steam stripping for volatile organics, surfactant scrubbing for non-volatiles with UV catalysed oxidation for destruction.
- leaching. In this stage the contaminants are dissolved using conventional solid/liquid contacting processes. Batch or continuous countercurrent systems can be used to enhance process performance. Equipment which can be used for this stage includes percolators, stirred tanks and attritors. The leaching stage uses BNFL's patented CACITOX™ leaching process, which involves carbonation at near neutral pH combined with the use of various complexants and oxidants. This system dissolves a wide range of metals without using aggressive chemicals or extreme conditions. This makes it ideally suited to applications where attack on the matrix is to be avoided. It also limits secondary waste arisings. Thus this approach is much more benign to soil than many other leaching agents.
- soil/leachate separation. This stage can be carried out using gravity settlers, hydrocyclones, centrifuges, pressure filters, vacuum filters or ultrafilters.

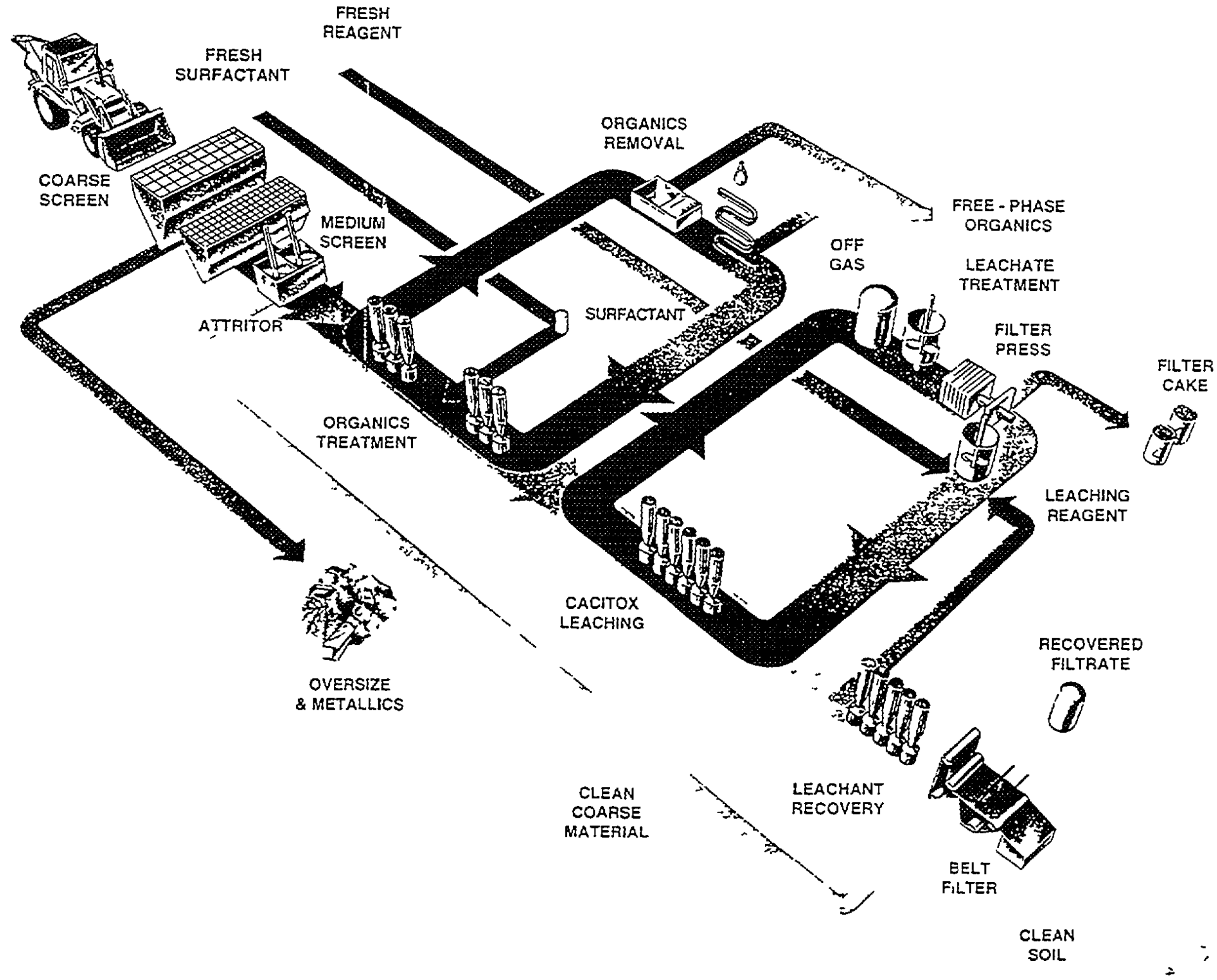

FIGURE 1 THE BRITISH NUCLEAR FUELS "CACITOX" SOIL TREATMENT PROCESS

- leachate treatment. Cacitox reagents are mild. The low concentrations allow the use of conventional effluent treatment processes, such as ion exchange, evaporation, floc precipitation and reverse osmosis.

4.3 Electroremediation

Electrokinetic remediation is an in-situ separation and removal technique, which can be used to extract heavy metals, including radionuclides, and even some organic contaminants from soils. It involves the application of a DC electric field across an array of electrodes inserted into the ground. Under these conditions three distinct electrokinetic effects; electrophoresis, electro-osmosis and electromigration, occur. Electrophoresis is the movement of particles within the soil moisture or groundwater under the influence of the electric field and may be applied to all electrically charged particles, including inorganic and organic colloids and organic droplets. Electro-osmosis is the movement of liquid relative to a stationary charged surface under the influence of an applied electric field. The surfaces of soil particles are usually negatively charged and hence the water layer in contact has an excess of positively charged ions and tends to migrate towards the cathode. Electromigration is the movement of dissolved ions and complexes under the influence of the electric field. Due to electrolysis accumulations of cations and anions occur respectively at the cathodes and anodes. In addition, water splitting electrolytic reactions also occur at these electrodes. Thus to ensure that the processes continue the pH in the system has to be controlled and the contaminants removed.

The concept of electroremediation is currently being extended in the UK, utilising a continuous electrochemical ion exchange plant with both cathodic and anodic exchangers to treat the secondary wastes generated and condition the cathode and anode streams. This results in a much simpler conditioning and extraction plant. It offers the potential for treating radioactively contaminated soil in-situ with very substantial reductions in waste volumes as compared to soil removal and disposal options. It also offers the potential for treating contaminated soil under leaking storage areas and buildings without the need for costly excavations and even demolition. The technique also has potential for preventing the spread of plumes of radioactive contamination from old disposal sites, etc. by an electro-fencing approach.

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CONCLUSION

PROBLEMS (ENCOUNTERED AND FORESEEN) IN RELATION TO THE PROJECT

Site specifications

Siting and specification of radioactively contaminated sites in CEE is probably the most difficult problem in relation to the project. The IAEA has a good knowledge of the location of mines, mill tailing sites, areas connected to nuclear plants, larger waste repositories and so forth. However, dumping of radioactive wastes in connection with other industries, as the military and hospitals have done in some areas, has been done extensively. This has been done without any registration information being given regarding either the sites or the quantity and character of the waste. The IAEA hopes that the co-operation between it and the member countries in CEE will take on another phase so that the environmental restoration programme can start as soon as possible.

The data available is not only incomplete but also somewhat questionable. Estimates from Non Governmental Organization (NGOs) are often overestimates of the problem. Conversely, figures provided by the governments can be underestimates. Not only is there little information but the data available is in many cases inaccurate or questionable. The data presented in some cases may therefore be either obsolete, incomplete or erroneous, even though the best available sources have been surveyed. A premise for the environmental restoration project must be the availability of sources referring to and characterizing radioactively contaminated sites, otherwise the efforts and resources put into the process are useless.

Human resources

From a policy implementation standpoint the political change that has swept the region has produced a number of side effects. The on-going change has led to a stall in most of the actions that the governmental institutions perform, or a prioritization to a narrower aspect of the governmental duty spectrum. The somewhat rapid change of government officials does not make co-operation easier. Another problem in this reconstruction of governments has been that the body in charge of important aspects of waste management is dissolved and that its responsibilities have not been replaced. Previously it was clearer who was in charge of issues relating to the realm of waste management.

Increasing differences among the CEE countries

Due to the individualization of the former communist countries it is imperative that the IAEA take action as soon as possible. These countries were brought into the contamination problems together and should come out of it together as well. A large co-ordinated project is likely to be more cost efficient and beneficial for the region that separate programmes. Nevertheless, there are tendencies that these countries will go different ways due to among other factors dissimilarities in the nature of their present economic and political objectives. This is a tendency that is not beneficial in the realm of environmental restoration projects. Close geographical proximity, political structure, and the similar character of the waste, call for co-operation and the use of the same technology and experience.

Attitude

Another problem with this project is the governmental, scientific and public view of the problem of radioactive waste. Since radioactively contaminated substances have been around for nearly fifty years and bad practices in the handling of the material have unfortunately been shown to be the rule, the public has often accepted that it has radioactive waste around them. It should also be mentioned that the public in many, if not most, cases did not know that these substances were in such close proximity to their neighbourhoods. This apathetic attitude is fortunately about to change as people in the CEE countries get to know more about such hazardous waste. This may be helpful in the IAEA's work in identifying such sites.

The different governments often have not been successful in planning and managing clean-up or restoration of contaminated areas. If this attitude persists, the environmental restoration of these areas will be hard to accomplish.

Another group in society that has an attitude problem regarding radwaste is the scientific community. As the 1992 International Symposium on Environmental Contamination in Central and Eastern Europe demonstrated, the subject of radioactive contamination was surprisingly of very little interest to the scientists from the contaminated countries. Radiological problems are not perceived by some as a real issue in the region. This might make the location and surveying of the prospective sites more difficult. There is no doubt that the expertise to deal with the problem is present, but the willingness to deal with the issue may not be there.

Funding and infrastructure

The most severe problem with this project is the lack of financial resources that the CEE countries have. Given the fact that many CEE countries are in difficult financial conditions, the priorities that are made are carefully worked out in order to make the best use of available resources. Only that which is considered most important is funded. As our work has shown so far, the severity in both the quantity and quality of the radwaste is so acute that remediation and restoration of these areas must be among the priorities these countries set. Some countries have, unfortunately, not fully recognized this and therefore are not willing to pay for the restoration. An option might therefore be to find countries willing to fund such a project. Some restoration projects have already started, independent of this project, and others are set to start. However, these projects are of a smaller scale and with a very limited targeted area. A co-operative project, if well planned, is more likely to succeed than a smaller operation.

A premise for a successful clean-up is to have facilities that can dispose of the generated waste. As of today, such facilities are few. The management of very low level waste is even a problem in the Russian Federation. It is therefore important to look at the aspect of storage/processing sites before the remediation process starts.

Duplication of assistance

The IAEA is not the only organization that seeks to assist the CEE countries in the area of environmental restoration. Rather, many institutions and companies have looked into the issue from an aid perspective (foreign governments) or from a profit perspective (private enterprises). In general terms, these efforts can be said to be independent of each other and are not part of a general and systematic plan. This lack of co-operation leaves room for duplication and inefficiency of the efforts. The ideal situation would be a close joint participation between the different institutions taking part in he restoration so that the resources made available would be used more effectively. Also, the countries that otherwise would not have been able to gain from the acquired knowledge would be able to do so. One can get the impression that most efforts directed toward CEE are isolated projects. These are not to be disputed if part of a larger plan, but it makes little sense for two organizations to run similar projects in the same area independently of each other.

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