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Site characterization techniques used in environmental restoration activities

Final report of a co-ordinated research project
1995–1999

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FOREWORD

The International Atomic Energy Agency (IAEA), in response to the needs of its Member States, has initiated a co-ordinated research project (CRP) in the area of site characterization techniques used in environmental restoration activities. This CRP extended over a four year period, spanning the years from 1995 to 1999.

The CRP is one element in the comprehensive, ongoing IAEA programme on environmental restoration of radioactively contaminated sites. The programme covers both technical and management issues:

- factors important for formulating a strategy for environmental restoration;
- site characterization techniques and strategies;
- assessment of remediation technologies;
- assessment of technical options for cleanup of contaminated media;
- post-restoration compliance monitoring;
- non-technical factors influencing the decision making process in restoration;
- assessment of the costs of restoration measures;
- remediation of low level disperse radioactive contaminations in the environment.

These tasks are supported by the continuing development of a worldwide directory of contaminated areas, covering *inter alia* environmental contamination arising from past practices, such as production and processing of nuclear materials, weapons testing, inadequate waste management, and from accidents involving nuclear materials. The tasks are also complemented by activities addressing specific origins of contamination, such as uranium mining and mill tailings (UMMT) and technically enhanced naturally occurring radioactive materials (TENORMs).

The co-ordinated research programme emphasises the identification and transfer of specific technical information and technologies between Member States seeking solutions for their nuclear related environmental problems. Its focus is on characterization techniques including relevant management procedures used during remediation/restoration activities.

The first research co-ordination meeting (RCM) was held in Vienna, 22–26 April 1996, and was followed by meetings in Madrid, Spain, 3–7 November 1997, and in Rio de Janeiro, Brazil, 12–16 April 1999.

The IAEA officers responsible for this publication were D. Clark and M. Laraia of the Division of Nuclear Fuel Cycle and Waste Management. The final report was compiled and edited by W.E. Falck of the same Division.

The IAEA wishes to express their 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 research co-ordination meetings.

CONTENTS

ANNEX. SUMMARIES OF WORK REPORTED AT RESEARCH CO-ORDINATION MEETINGS

1. INTRODUCTION

Although the use and handling of radioactive materials has been regulated in most countries for many decades, inadequate practices and accidents have led to radioactive contamination of the environment. Thus in some countries, very challenging problems exist for government authorities and the technical community due to the contamination of soil and water. Instances of radioactive contamination of the environment may be the result of, *inter alia*, accidents, spills and inadvertent airborne releases at nuclear installations, inadequate waste handling/ treatment/disposal practices as part of the nuclear fuel cycle, past practices and inadequate radioactive waste management at (now sometimes abandoned) facilities where radioactive substances are/were used (e.g., radium factories, hospitals, research facilities), nuclear weapons testing, man-enhanced radioactivity in residues from mining and processing of ores and other natural materials, and so on.

Proper site characterization is an essential prerequisite in the process leading to decisions on possible remediation actions. Although the problems have been recognised in most countries, for various reasons (e.g., the magnitude and extent of the contamination, the location, the affected populations), some situations may pose particularly difficult technical, economic and political challenges to the authorities.

Low level wastes have been in some cases disposed of as conventional waste or at sites where the population can be exposed to it through air or groundwater pathways. Radioactive waste like this is often not registered and it is hard to identify the sites at which it has been disposed of. Under the circumstances, the suspected sites cannot be released in their present condition and should therefore be subject to one or another form of site investigation and eventually remedial action. If nothing else, contamination should be identified and secured. Any spreading of contamination should be stopped along with the making of plans for the lasting confinement or removal of the radiation sources.

Radioactive fallout or dispersion of radionuclides as a result of a nuclear accident or atomic weapon test is another source of radioactive contamination. Radioactive fallout contamination is generally not intense but spreads over a vast area. This makes the problem truly international because many countries can be affected by the same accident or event. Due to the somewhat unpredictable natures of these events and their evolution, identification and characterization of contaminated sites is often a difficult job.

Another category of conditions resulting in environmental contamination is related to disused factories where radioactive products (e.g., luminescent paints) were manufactured or handled at times when radiation protection criteria were not as stringent as they are today. This has in many cases resulted in extensive contamination of urban areas. Also in this case, contaminated sites are often difficult to identify and to delineate.

Environmental contamination may also occur due to man-enhanced concentration of natural occurring radioactive materials (NORMs). Conventional ore mining (e.g. iron or copper ore) and chemical processing (e.g. phospho-gypsum production in the fertilizer industry, water treatment) may result in increased activity levels in uncontrolled areas.

In view of the importance of adequate and effective site characterization for the design and efficiency of remedial actions, it was considered appropriate to encourage R&D work on this subject. Efforts to improve characterization methods and techniques are considered of high priority as full characterization is an essential prerequisite to further remedial actions.

2. DEALING WITH RADIOACTIVELY CONTAMINATED SITES

The problem of contaminated sites in general and of radioactively contaminated sites in particular requires a structured approach in order to deploy in a most economic way the available resources and to apply the most appropriate restoration option. A typical sequence of events leading towards remediation can be outlined as follows:

(1) Evaluation of historical records on site use, past practices or accidents;

(2) Field screening survey with respect to likely contaminants;

(3) Planning of detailed site characterization using statistical and/or deterministic models;

(4) Detailed site characterization [1];

(5) Determination of remediation/restoration goals considering all relevant factors [2];

(6) Evaluation of applicability of techniques, remediation options assessment, and planning of remedial action [1];

(7) Remediation/restoration [3] [4];

(8) Post-restoration verification and/or monitoring [5].

Some of the above steps, in real cases, may be taken iteratively or concurrently. The present reports addresses mainly steps (2), (3) and (4) from above scheme.

As discussed in this context, the characterization of radioactively contaminated sites is an integral activity for all aspects of environmental restoration. The IAEA’s environmental restoration project includes activities associated with the characterization of a contaminated site, as well as others such as the technologies for cleanup and remediation, factors to be considered in the formulation of a strategy, and the monitoring of a decommissioned site.

3. CHARACTERIZATION AND ASSESSMENT OF RADIOACTIVELY CONTAMINATED SITES

3.1. Variables to be determined

The characterization of a contaminated site is considered to be the methodical scientific process used to determine the extent and nature (i.e., physico-chemical-radiological properties) of the contamination as well as the important site parameters, geotechnical characteristics, geological features, and other properties likely to determine the behaviour of the contaminants [1]. To provide a detailed picture of the nature of the contaminated site, this could include:

— determining the distribution of the contaminants in the various media, e.g. whether the contamination is limited to the surface soil, is distributed to significant depths, or whether the groundwaters are affected;

— determine the chemical and physical characteristics of the contaminants and their immediate environment;

— determining whether the contaminants will be mobile under any of the relevant environmental conditions, and if so, what the likely source term would be.

3.2. Objectives

A good radiological survey plan is an essential element in planning and implementing the cleanup of contaminated areas and could constitute the essential element in step (2) from the above scheme. This plan has the objective:

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

3.3. Screening surveys

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

3.4. Detailed site characterization

A detailed characterization of the contaminated site and assessment of the consequences of various actions is essential before embarking on a programme for its remediation and ultimate restoration. Thus, reliable and suitable data must be obtained regarding the distribution and physical, chemical and nuclear properties of all radioactive contaminants. In this way, characterization data support the required engineering design and project planning for environmental restoration. For large or complex facilities and sites, the sampling and measuring of radioactivities may become quite complicated, timeconsuming and expensive. Therefore, it may be advisable for remediation activities to be started even before complete knowledge on the radioactive contamination is available and in such cases the characterization can be carried out in iteration with remediation steps, i.e. steps (3) to (7) from above would be repeated in several loops. Indeed, continuing characterization can provide information regarding efficiency and efficacy of the cleanup methods, and if necessary help to redirect of work efforts.

Similarly, at end of the remediation phase, characterization and ongoing monitoring can be used to demonstrate completion and success of the cleanup process [5].

While the basic technology necessary to characterize contaminated sites is reasonably well developed, there are still several areas worthy of further studies. To provide one example, full characterization may be very costly if a large site is involved. Advanced techniques like statistical methods, deterministic models simulating the migration of radionuclides in environmental media, or special equipment to optimize resources and perform quick measurements may become indispensable. Due to the international need for environmental restoration, a rapid exchange of information and co-ordination of work are required to foster technology, reduce duplication of efforts and provide useful results for those Member States planning environmental efforts.

The characterization end products are utilized in site assessment activities, that is, for the thorough evaluation of contamination at a site and the alternatives for cleaning it up or otherwise addressing the problems associated with it.

4. IAEA ACTIVITIES IN ENVIRONMENTAL RESTORATION

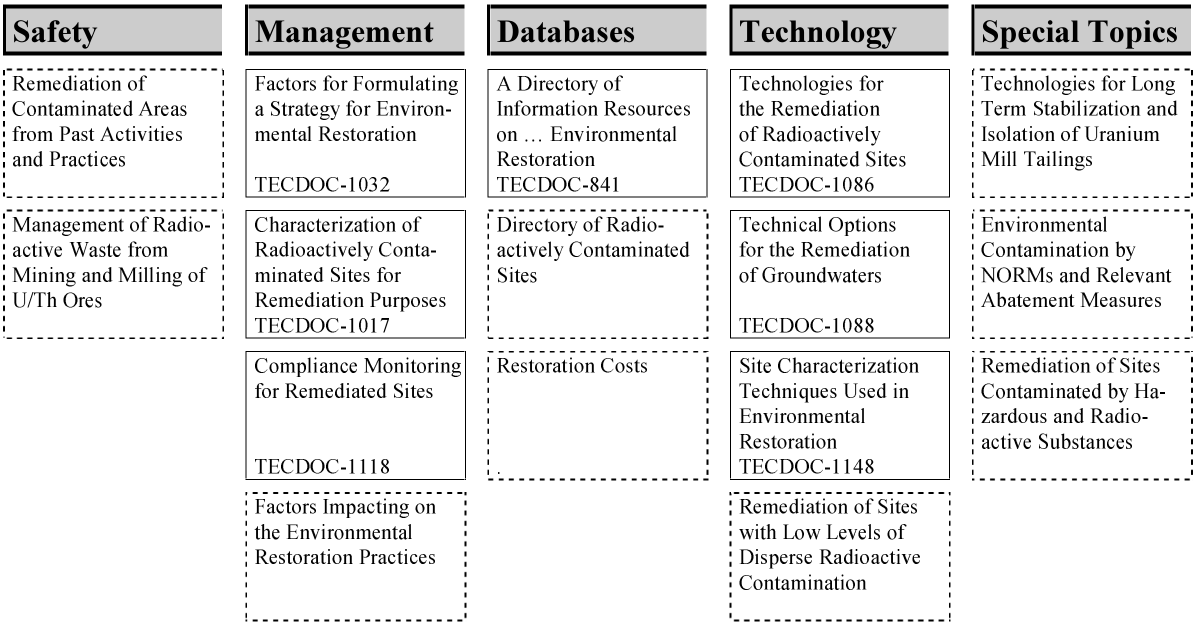
The IAEA has an active programme to assist its Member States in the development of technologies for environmental restoration of radioactively contaminated sites. Within the IAEA, the environmental restoration projects are closely tied to the broader field of radioactive waste management and to the decontamination and decommissioning (D/D) of nuclear facilities.

The IAEA efforts prior to 1995 have largely focused on responses to accidents (e.g. Chernobyl, Khyshtym) and weapons testing (e.g. Mururoa, Bikini, Semipalatinsk). In addition to site or event specific reports, several documents in the IAEA Technical Report and Safety Series have been written, published and revised on subjects relating to environmental restoration [6] [7][8]. These were intended to contain, at the time of writing, the most advanced knowledge and experience in the field of this subject. Relevant Safety Guidance and Requirements are currently under development [8].

A Technical Co-operation Project on Environmental Restoration in Central and Eastern Europe was initiated in 1992 and completed in 1994 with the intention to gain an overview over the extent of the problem. The Central and Eastern European countries involved were Belarus; Bulgaria; Croatia; the Czech Republic; Estonia; Hungary; Poland; Romania; the Russian Federation; the Slovak Republic; Slovenia; and Ukraine. At three different workshop meetings, representatives from these countries presented papers providing considerable amounts of information on their radioactive contamination problems. Also, papers from Canada, France, Germany, Italy, Spain, Sweden, the United Kingdom and the United States of America were included to share their experiences with environmental restoration. The results of this Regional Technical Co-operation Project have been published in three volumes as an IAEA Technical Document [9].

A worldwide survey of radioactively contaminated sites is being undertaken by the IAEA. The initial survey was conducted in 1996 through distribution to all Member States of a questionnaire on contaminated sites. The results indicate a wide range of situations among the Member States, from no such problem to cases where many such sites have been identified and cleanup/remediation work is underway. Work on this survey continues in 1999. It will be used by the IAEA to 1) provide an international directory of radioactively contaminated sites; 2) provide a forum for environmental restoration technology exchange for Member States; 3) assist the IAEA's and funding IAEA's strategic planning in environmental restoration; and 4) assist in focusing the IAEA's R&D programmes in environmental restoration technology.

The IAEA's activities in environmental restoration technology have since developed into an integrated programme, focusing on the characterization of contaminated sites, the formulation of strategies for environmental restoration [2], and the techniques for cleanup and remediation of radioactively of such sites (Figure 1). In addition to more general subjects, projects on specific issues have been developed, including appropriate restoration techniques for soils [4],



Safety	Management	Databases	Technology	Special Topics
Remediation of Contaminated Areas from Past Activities and Practices	Factors for Formulating a Strategy for Environmental Restoration TECDOC-1032	A Directory of Information Resources on … Environmental Restoration TECDOC-841	Technologies for the Remediation of Radioactively Contaminated Sites TECDOC-1086	Technologies for Long Term Stabilization and Isolation of Uranium Mill Tailings
Management of Radioactive Waste from Mining and Milling of U/Th Ores	Characterization of Radioactively Contaminated Sites for Remediation Purposes TECDOC-1017	Directory of Radioactively Contaminated Sites	Technical Options for the Remediation of Groundwaters TECDOC-1088	Environmental Contamination by NORMs and Relevant Abatement Measures
	Compliance Monitoring for Remediated Sites TECDOC-1118	Restoration Costs	Site Characterization Techniques Used in Environmental Restoration TECDOC-1148	Remediation of Sites Contaminated by Hazardous and Radioactive Substances
	Factors Impacting on the Environmental Restoration Practices		Remediation of Sites with Low Levels of Disperse Radioactive Contamination	

FIG. 1. The suite of IAEA work areas on environmental restoration.

groundwaters [3], and the assessment of compliance with restoration criteria [5]. Activities which are specifically related to the characterization and assessment of radioactively contaminated sites are discussed below.

Environmental restoration is also a part of the IAEA's ongoing Technical Co-operation Programme. These activities include within their scope the characterization and assessment of contaminated sites.

5. SCIENTIFIC SCOPE AND GOALS OF THE CRP

The objective of the CRP was to promote the exchange of information on the practical experience gained by Member States in characterization of radioactively contaminated sites. Within this CRP, sites have been included which are contaminated due to:

- waste dumping practices, spills or transport of airborne pollutants;
- accident or nuclear weapon test fallout;
- disused factories where manufacturing of radioactive products has taken place; and
- enhanced concentrations of NORMs from conventional ore mining and processing.

Contamination associated with or originating from decommissioning of nuclear installations were not part of the CRP scope, nor were directly the problems associated with the mining and milling of uranium ores.

Special emphasis was placed in the CRP on the development of methods and techniques for the optimization of radiological characterization. In particular, the scope included:

(a) Definition of a strategy for site characterization, i.e.:
  - identification of relevant contamination boundaries;
  - protocol development;
  - screening surveys;

- determination of background radiation levels;
- costs and number of measurements;
- quality assurance programmes.

(b) Sampling and measurement techniques, in particular in the following areas:
  - vehicle-borne monitors;
  - semi-portable and hand held instruments;
  - air, water and (surface and subsurface) soil sampling;
  - laboratory capabilities;
  - methods to automate radiological surveys;
  - models for evaluating detector responses to complex contaminations geometries.

(c) Data management, including statistical analysis and deterministic radionuclide migration modelling; and

(d) Post-cleanup radiological surveys and assurance of compliance with release criteria.

6. RESULTS FROM INDIVIDUAL PROJECTS

The above methodological developments within the individual projects of this CRP were undertaken while addressing a wide range of actual problems. These problems can be divided into four groups, namely:

(1) Sites, where radioactive materials have been handled intentionally or unintentionally at a time when radiation safety procedures were less well develop as today:
  - radiological characterization of the surroundings of former radium production and processing facilities (Belgium, Canada);
  - characterization and restoration strategies for an urban site affected by a monazite processing plant (Brazil);

(2) Nuclear research sites:
  - characterization of environmental radioactivity around nuclear power plants and nuclear research centres, including their associated waste handling and storage facilities (United Kingdom, Romania, USA);

(3) Sites, where accidents or weapons testing have resulted in disperse contamination of larger surface areas:
  - measuring techniques for characterisation of ^{137}Cs contaminated river banks (Slovakia);
  - characterization of particulate plutonium contamination at a former weapons test site (Australia);
  - assessment of the environmental behaviour of accident-derived plutonium (Spain);
  - site characterization techniques used in restoration of agricultural areas contaminated by the Chernobyl accident (Russian Federation);

(4) Mining sites, where radionuclides have been either mined on purpose or occur as by-products:
  - identification and characterization of radioactively contaminated sites in a mining town as driven by the planning for environmental restoration activities (Ukraine);

6.1. Sites where radioactive materials have been handled

The planned release for unrestricted use of the site of a former plant for extracting rare earth elements from monazite in **Brazil** (Annex BR) required an efficient strategy to assess and verify residual contamination from radionuclides associated with the monazite. The strategy to verify the requirement for unrestricted site release comprised a preliminary soil survey by direct total gamma measurement with a sodium iodine scintillator detector, direct local gross alpha and beta counting of soil samples for monitoring during cleanup, and germanium detector gamma spectrometry of soil grab samples for the validation of monitoring methods. The proceedures established for on-site radiation measurements resulted in a cost reduction and the speed up of the analyses.

The **Canadian** participants approached the problem of estimating subsurface contaminations from above surface measurement by developing models and algorithms for solving the inverse problem first. This means estimating surface gamma radiation spectra for a fully known subsurface radium distribution. The techniques employed fell into three broad categories: empirical (using laboratory and field data), analytical (using mathematical derivations of relationships), and computer simulation (using Monte-Carlo photon transport simulation methods). Methods of analyzing surface spectra to estimate certain source parameters have been studied. The most fully developed methods are those involving the ratio of the areas of two peaks of differing energy from the same radionuclide to determine the source depth.

For a point source of radium and its progeny, peak ratio techniques are able to reliably estimate the source depth from a single gamma radiation spectrum taken at the surface directly above the source. The only significant uncertainties in this case are the soil density and uncertainties introduced as a result of counting statistics. Estimates of source depth may be useful themselves in some specific circumstances. In general, a more important use of the estimated or effective source depth is its use in improving the estimate of source activity.

The participants, however, state that further work is required to develop a comprehensive suite of tools for the improved interpretation of surface gamma radiation spectra from subsurface distributions of radium contaminated soil.

A similar approach was taken in **Slovakia**, where Monte-Carlo simulations of detector responses were compared with actual readings for different measurement geometries in the field and for different shapes of the contaminated areas. The technique is based on a dose rate measurement, taking the ratio of two detector readings at two different distances above ground and Monte-Carlo simulations of the plastic scintillation detector responses.

6.2. Nuclear research sites

Former nuclear research facilities can provide a serious challenge to site characterisation, owing to their usually varied history of nuclear materials handling and to the experimental nature of many practices.

While there appears to be limited contamination of only short lived elements around the IFIN-HH nuclear research centre in **Romania**, former waste handling and disposal practices and subsequent cleanup attempts led to significant problems. Detailed site surveys and their repetition after cleanup actions were considered essential.

A similar challenge was faced in planning for the decommissioning of facilities at the various sites of the **United Kingdom** Atomic Energy Authority (UKAEA). The final stages of decommissioning require confidence that the land on which the facilities stand is clean enough to allow unrestricted public access. One of the crucial questions when establishing cleanup criteria is the problem of defining the level of background radioactivity on sites. Progress has been made in determining the presence of natural radioactivity for the purpose of identifying background levels. Tight controls need to be placed on the analytical methods used in characterising the contamination against natural background levels.

The advances made in the use of accurate radiation mapping using satellite global positioning systems are expected to bring about savings in terms of minimising the volumes of wastes segregated for disposal. The development of soil treatment techniques may assist in the eventual clean up options for some type of contaminants, and is also assisting in the understanding of how some characterisation methods may overestimate contamination concentrations.

Work continues in developing a multi disciplinary approach to characterisation. The main area where advances were most urgently required were in the development of techniques which allow detection of below ground radiation (see 6.2).

The theme of a wide variety of radioactive contaminants and their complex distribution patterns re-occurs for the many military and non-military sites in the **USA**. The importance of redirecting characterizing efforts on the basis of incoming results on a “real time” basis was recognised. Research and technology development conducted at the Idaho National Engineering and Environmental Laboratory (INEEL) has resulted in the ability and state of the art equipment required to obtain real time, densely spaced, *in situ* characterization data (i.e. detection, speciation, and location) of various radionuclides and contaminants. The Remedial Action Monitoring System (RAMS), developed, consists of enhanced sensor technology, measurement, modeling and interpretation techniques, and a suite of deployment platforms which can be interchanged to directly support remedial cleanup and site verification operations.

On-site characterization techniques have advanced to the point where they are being actively deployed in support of remedial operations. The deployment of on-site characterization systems during environmental restoration operations has shown that this approach results in several significant benefits versus conventional sampling techniques. A flexible characterization system permits rapid modification to satisfy physical site conditions, available site resources, and cleanup requirements.

The rapid and precise collection of field measurements can reduce operational costs by reducing the volume of material requiring remediation. This approach can also reduce uncertainty by obtaining a “total area field screen” versus a representative subset of hand collected laboratory analyzed samples. It can lower on-site health risks by reducing human exposure to contamination through reduction of hand sampling during the field sampling process.

6.3. Accident and weapons testing sites

Another type of contamination requiring special characterisation and remediation techniques is the geographically widely dispersed contamination in the aftermath after the heavy radiation accidents such as in the former Soviet Union, or following atmospheric nuclear weapons tests.

Both, characterization and remediation, require efficient strategies in order to efficiently deploy resources.

In the **Russian Federation** special site characterisation techniques and strategies for the restoration of large areas of contaminated agricultural lands had to be developed. In the years following the accidents (e.g. Chernobyl, Kyshtym), various remediation measures had been implemented and a vast amount of data on their effectiveness has been generated, together with information on ancillary factors, such as the required resources and costs. These measures vary considerably with respect to their effectiveness, cost and practicability in actual situations. Their effectiveness depends on many factors, including soil and climatic conditions, or specific features of the agricultural production management. Owing to this, the implementation of the restoration measures on the basis of general expert estimations often results in inadequate decisions. An assessment of this situation underlined the importance of the role of site characterisation in the selection of adequate restoration strategies. It also demonstrated the need for implementing a flexible decision support system, which is capable to organise a comprehensive set of site specific information, and which provides a practical tool for advising on agricultural land restoration measures.

(Unsuccessful or inadequate) restoration measures or other activities leading to the displacement of soil, e.g. civil engineering projects, can significantly complicate the site characterization. Many characterization strategies are, explicitly or tacitly, based on assumptions about the mechanisms effecting the dispersion patterns of radioactive contaminants. River corrections and flood defense construction resulted in a more or less random and discontinuous re-distribution of river bed sediments previously contaminated by accidental discharges from the Bohunice NPP in **Slovakia**.

The sometimes difficult terrain, such as the steep river banks, also required the development of sophisticated field instrumentation and means for their deployment in order to obtain reproducible and consistent results. Reproducibility and consistency in characterization are essential for avoiding the excessive removal and disposal of soil during a restoration project.

The difficulties in site characterization added by the effects from earlier remediation attempts was also addressed by the **Spanish** project. Here, in 1966 as a consequence of an accident involving two US military aircraft, two nuclear weapons were dropped and their radionuclide contents partially scattered. The behaviour of plutonium and americium associated with the soil for more than 30 years has been studied. These studies included chemical (sequential extraction) and mineralogical investigations of plutonium associated with different fractions of the soil, lung solubility of plutonium associated with the breathable size fraction of the soils and respective inhalation dose factors, and the statistical estimation of the inventory of plutonium remaining in the top first 45 cm of the soil at the most contaminated area where remediation work was carried out shortly after the accident.

The Maralinga test site in **Australia** had already seen a cleanup operation in 1967. It was found in scenario analyses that Aboriginal children playing on the ground were the criticial group for exposure from residual radioactivity. Hence, the site characterization programme and the techniques employed were designed such that they cover the top 10 mm of the soil which is prone to become re-suspended by the playing children. Vehicle-borne γ-ray survey using a calibration model which assumes exponential decrease with depth of radionuclide concentrations proved the most suitable characterization technique. However, on rough terrain which had not seen previous remediation, surveying with handheld detectors proved the most efficient method to identify residual plutonium particles. Survey data were stored together with data from a GPS on a computer, allowing the revisiting of the sites for verification of cleanup efficiency.

6.4. Mining sites

Complex site histories, such as are often found in the context of uranium mining, require a range of investigation and assessment tools to complement each other. In the Pridniprovsk-Krivoy Rog region in the Ukraine the main methods for characterizing various sites contaminated by the mining and milling of, inter alia, uranium, were:

- dosimetric surveys;
- radiometric fast measurements of total alpha-activity in samples;
- emanation surveys;
- radon emission survey (measuring the density of radon flux from surface);
- measuring radon and its equivalent equilibrium volume concentration in houses and in the ambient atmosphere;
- soil sampling and analysis for concentrations of natural radionuclides.

The experience from applying the various methods shows that no single one can be used separately for characterizing the contaminated land. In any given case it is necessary to apply a certain set of methods, tailored to the expected contamination and radionuclides involved. The site characterization also showed that it is expedient to perform efficacy analyses before and during implementation of remediation measures. The continuing characterization also allowed to establish reasonable trade-offs between resouces availability, efficiency of remediation methods, and dose limits for the population. From this a target residual gamma dose rate was developed.

The CRP made it possible to learn from an international approach to the identification and characterization of radioactively contaminated sites. The experience gained will be used both for design of a new regulatory basis for the remediation of the contaminated sites and in carrying out the remediation measures themselves.

7. CONCLUSIONS

Complex radioactive contamination problems require complex and complementary assessment methods; this is the perhaps obvious, but main overall conclusion.

Approaches integrating field surveys, laboratory determinations and modelling are advocated. Field dosimetric methods together with computerized data capturing and processing allow rapid mapping of surface contaminations. However, these methods, like many geophysical methods, only record integrated signals. The detector response must be calibrated against known source geometries and strenghts. Computer simulations allow to interpolate and extrapolate detector readings to differing geometries.

The combination of non-invasive techniques and field sampling effects a reduction of exposure for field staff and minimises the problem of having to dispose of contaminated samples.

On-site techniques improve management decisions by providing quasi real time, densely spaced, and repeatable characterization data.

Reproducibility and consistency in characterization are essential for avoiding the excessive removal and disposal of soil during a restoration project.

Exact and repeatable positioning of sampling equipment is an important means to reduce uncertainty in characterization, and can be achieved today by using advanced positioning tools, such as the satellite-based global position system (GPS).

Deployment techniques are another important factor controlling the reproducibility of results and the efficiency of resources use. Vehicle-borne-manned or remotely controlled — scanning methods also help to reduce exposure of operating personnel. Detectors can be mounted on digger's arms or similar, allowing to maintain specified measurement geometries even in difficult terrain.

Careful three-dimensional mapping of a contamination (i.e. an effective delineation of clean from contaminated material) helps to minimise the amount of material to be removed and/or treated, thus reducing the operational costs. Repeating the mapping during the course of remediation helps to verify the previous assessment and the efficacy of the remediation action, as has been discussed in more detail elsewhere [5].

The importance of a structured, and preferrably computer supported, decision making approach has been demonstrated. Such approach helps to break down complex problems into more manageable problem units and makes decisions retraceable.

The CRP has resulted in a speed up of applicable methodological developments by fostering the exchange of experience between participants with seemingly different contamination problems, but essentially similar characterization challenges.

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Annex

SUMMARIES OF WORK REPORTED AT RESEARCH CO-ORDINATION MEETINGS

CHARACTERIZATION OF PLUTONIUM CONTAMINATION AT MARALINGA — DOSIMETRY AND CLEANUP CRITERIA

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ABSTRACT

An area of South Australia remained contaminated following British atomic tests at Maralinga during 1955–1963. Of importance is the long lived ^{239}Pu of which some 24 kg was explosively dispersed in several 'minor trials'. The extent, quantities and physical characteristics of the plutonium have been assessed and estimates of dose, dominated by the inhalation pathway in the critical group of Aborigines living a semi-traditional lifestyle, have been made for potential occupants. Dosimetry, together with social and economic factors, underpins the setting of cleanup criteria in terms of activity concentrations averaged over large areas and permissible concentrations of contaminated particles. The possibility of intentional behaviour such as fragment scavenging has also influenced limits on particulate contamination. Rehabilitation of the most contaminated areas is underway, with scraping of surface soil and burial on site completed. Vehicular-mounted radiation detector systems for widearea and particle monitoring have been developed, and procedures established for determining cleanup boundaries and for the verification monitoring to ensure that the cleanup process has met the specified criteria. Data are being obtained for a final dose and health risk assessment of the cleaned up site.

1. INTRODUCTION

1.1. Major trials

The United Kingdom conducted a programme of nuclear weapons development trials at Maralinga in South Australia between 1955 and 1963, including seven atomic explosions. The smallest was about one kiloton yield, and the largest was of 27 kiloton. All were atmospheric tests, generally exploded on 31 m high towers. The sites of these major trials no longer present any significant health risk, because all the radioactivity released in the explosions was either widely dispersed (i.e. worldwide) at the time, or has decayed sufficiently [9].

1.2. Minor trials

The UK also conducted several hundred 'minor trials' at Maralinga over the years 1955 to 1963. These minor trials were essentially developmental experiments designed to investigate the performances of various components of a nuclear device, separately and in combination. Almost all involved radioactive materials with conventional high explosives, and resulted in radioactivity being dispersed to the local environment.

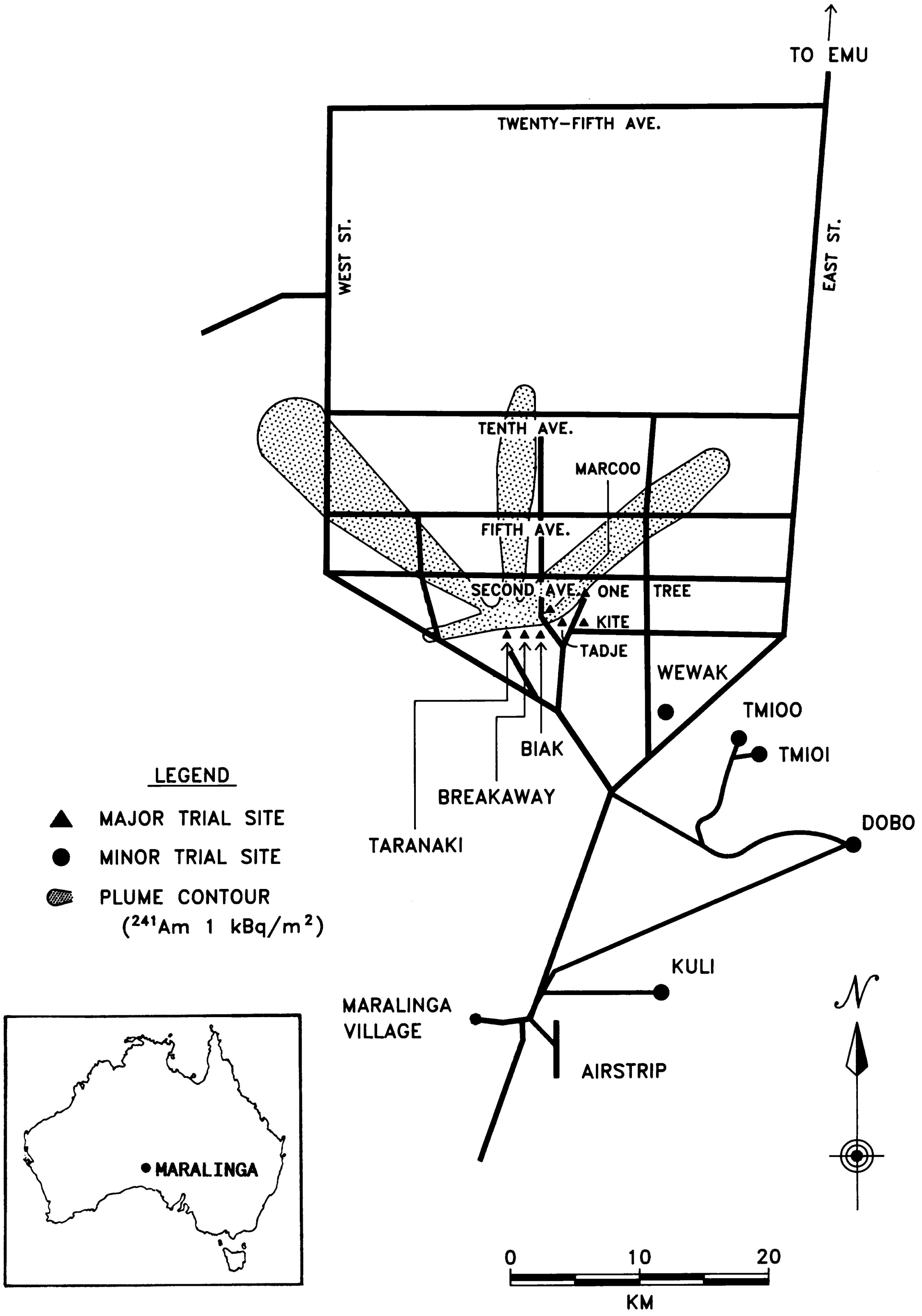

FIG. 1. The Maralinga area showing major and minor test sites and main features.

The Australian Radiation Protection and Nuclear Safety Agency (ARPANSA) has surveyed the minor trial sites at Maralinga, and a report was presented to the Royal Commission into British Nuclear Tests in Australia [9]. In many cases the sites had already been adequately cleaned up, or the radioactive materials used were of sufficiently short half lives that they were no longer detectable. The major and minor trial sites are shown in Fig. 1 and those which were significantly contaminated with plutonium are detailed below.

Wewak — Burnings and explosive dispersals of beryllium, uranium and plutonium occurred at Wewak in trials code-named ‘Vixen A’. Two plutonium burnings (involving a total of 405 g plutonium, of which 395 g was returned to the UK in 1959) and four explosive dispersals of a total of about 570 g of plutonium took place at the site. Surrounding the site are fragments of metal contaminated with plutonium.

TM100 and TM101 — Explosive dispersals of plutonium (about 600 g at each site) took place at both of these locations. Some 500 g of plutonium from TM101 was returned to the UK in 1979. There was a high concentration of plutonium-contaminated fragments and smaller friable particles close to the firing sites.

Taranaki — Taranaki is the site at Maralinga which was most extensively contaminated with plutonium, and which therefore represented the greatest remaining potential hazard to health. It was the site of the final major atomic detonation at Maralinga in October 1957. This was a balloon-borne test of 27 kiloton yield at 300 m, which left very little contamination nearby. Between 1960 and 1963, the area to the north of the ground zero was used for 12 ‘one point safety trials’ (code-named ‘Vixen B’) in which about 22 kg of plutonium was explosively dispersed in a sector extending from the west, through north, to northeast of the site. As well as plutonium, uranium-235 and beryllium were also dispersed in these trials.

In these one point safety trials, jets of molten plutonium were projected up to 1000 m into the air, and the contamination was dispersed by wind in narrow ‘plumes’. The main plumes extend to the west, northwest, north and northeast of Taranaki. The most extensive of these is the northwest plume which can be detected up to 100 km from the firing pads at Taranaki. These trials were very similar in nature to those conducted in 1963 as a joint project between the UK and the USA in ‘Operation Roller Coaster’ at the Nevada Test Site. The purpose of Roller Coaster was to study the effects of transportation and storage accidents involving nuclear weapons, and in particular to investigate the nature and fate of the plutonium aerosol.

The plutonium contamination close to Taranaki occurred mainly in three forms [2] — as a fine dust, as small sub-millimetre particles, and as surface contamination on larger fragments (where a fragment is defined as visually identifiable foreign matter). It is likely, however, that most of the plutonium was dispersed as an aerosol over very wide areas at low levels of contamination.

1.3. Radionuclide composition

The plutonium contamination contains isotopes ^{238}Pu (88 y), ^{239}Pu (24,110 y), ^{240}Pu (6,600 y) and ^{241}Pu (14 y). The minor trials involved negligible fission yield so the isotopic composition of the source material was unaffected by the explosion but radioactive decay has substantially removed the ^{241}Pu and replaced it with ^{241}Am (433 y). The 59.5 keV γ-ray from ^{241}Am serves as the most useful indicator, in the field, of plutonium in soil once the ratio of plutonium to americium has been determined experimentally. This ratio and the isotopic composition of the plutonium vary from site to site, and even from one trial to another at the same site [3]. At Taranaki, ^{239}Pu/^{241}Am activity ratios vary between 6 and 22 (1988 values), with the most common value 6.8. At sites TM100, TM101, and Wewak the ^{239}Pu/^{241}Am ratios are 20, 7.6 and 20 respectively (1988 values).

At present, the americium content is slowly increasing as the remaining ^{241}Pu decays. The ^{241}Am levels referred to above for determining cleanup boundaries based on the agreed inhalation dose limit, and as criteria to be met following clearance, are based on Pu/Am activity ratios pertaining in 1988. Levels measured in the field today are some 10% higher.

1.4. Previous cleanups

During the period that the Maralinga Range was in use, various radiation surveys and cleanup operations were performed [10]. Once the decision was taken to close the Maralinga Range, a cleanup of all sites was undertaken by the UK in Operation Brumby in 1967.

In Operation Brumby, the surface soil in the central area at Taranaki was treated by mixing to reduce average contamination levels, and plume areas were ploughed. Beyond the ploughed area the plutonium contamination tends still, some 30 years later, to be on the surface. Within 500 m of the firing pads there were still many thousands of contaminated fragments large enough to attract attention as potential souvenirs. The range of types of fragments included wire, rusty steel plate, lead, pieces of a grey metal alloy of low density, bitumen and yellow bakelite.

1.5. Maralinga Aborigines

The traditional occupants of the Maralinga lands are the Maralinga Tjarutja (Pitjantjatjara) Aboriginal people. Currently the Maralinga Tjarutja lands cover some 80,000 km^2, with a pool of about 2000 Aborigines who have traditional obligations to parts of these lands. The area that is currently denied to the Aborigines due to the former atomic weapons tests comprises 3,200 km^2. In recent times, between 60 and 200 of the Maralinga people have established a semi-traditional lifestyle at Oak Valley, some 100 km northwest of the Maralinga range.

2. THE HEALTH RISKS

2.1. The inhalation pathway

The inhalation of plutonium dust presents the most significant potential health hazard arising from residual contamination of the Maralinga area, due to the very low solubility of the plutonium oxide at Maralinga and the dusty, dry conditions. ARPANSA has performed a study to determine the input data for dose and health risk assessments for the inhalation pathway [11] [8] [4]. The study included a survey of ambient concentrations of radionuclides and dust in air, artificial resuspension studies, the characterisation of the contamination in the Maralinga soil, particularly particle size, and a dose assessment for the inhalation pathway. Ambient levels of airborne radionuclides are very low except during dust storms, and risk is dominated by possible exposure to isolated events such as a severe dust storm or from dust resuspended by human or mechanical activity.

Anthropological examination of the Aboriginal lifestyle showed that resuspension of dust by everyday actions of adults, children, and their dogs was much more important than natural resuspension. Aboriginal children, being closer to the ground and playing at activities which raise a lot of dust, living a semi-traditional lifestyle were shown to be the critical group for radiation protection purposes [8].

Given the relatively low importance of natural resuspension, the dust was taken to be produced from soil in the top 10 mm. This is appropriate for dust raised by vehicle tyres and human feet, whereas a much thinner layer might be resuspended by wind. Thus the average radionuclide concentration in this layer, scaled by an empirically determined enhancement factor and average dust loadings, was used to calculate the activity concentration of the inhalable fraction of each radionuclide in air.

2.2. Other exposure pathways

Other exposure pathways include ingestion, either directly of individual particles or of contaminated soil, or through eating contaminated foodstuffs, and incorporation of plutonium into open cuts and wounds. In determining cleanup criteria, dosimetry for these other pathways has only been considered in detail for ingestion of individual particles (or soil containing the same activity). However, a general allowance has been made for dose from pathways other than inhalation. For example, at sites contaminated with plutonium, for adults and children, all dose pathways other than inhalation contribute approximately an extra 10% of dose [6] [7].

2.3. Aboriginal health and lifestyle

There are several lifestyle aspects and related health issues, which might alter doses received by residents of an Aboriginal community living in contaminated areas. It is noted that the Aborigines at Oak Valley are commonly mouth breathers, many are heavy smokers, and upper respiratory tract infections are endemic. Mouth breathers tend to have lower deposition in the naso-pharyngeal region of the respiratory tract, which can affect dose intake conversion factors. Smoking further complicates the matter, but generally seems to have a small effect on regional deposition while substantially altering mechanical clearance capability. Endemic respiratory tract infections also complicate the situation, probably causing changes in regional deposition and clearance capability. These infections may also be related to high dust exposures and possibly are a cause for the high incidence of mouth breathing.

Other factors, which may affect dose intake conversion factors, are the general health and dietary habits of the population. The dose conversion factors used in our assessment have been computed from a model designed for essentially healthy workers, a scenario which is not directly applicable to many of the Oak Valley residents. Unfortunately, while these factors are obviously important in assessing doses that might be received by an Aboriginal resident in a contaminated area, little or no data exist to quantify their importance.

2.4. Short term visitors

A casual visitor to Maralinga is unlikely to remain in contaminated areas for very long and almost certainly would be exposed to lower dust concentrations than a member of an Aboriginal community. If it is assumed that the visitor is only exposed to ambient sources of resuspended dust, the greatest hazard would arise if that person were present during a dust storm (and unable to shelter in a vehicle or move out of the area while conditions were unpleasant). Calculations for this scenario, pre cleanup, indicate that there was no significant inhalation hazard for casual visitors to even the most contaminated sites at Maralinga, so long as dust raising activities were avoided [11].

2.5. Intervention philosophy

Calculation showed that doses well in excess of 100 mSv per year were possible (albeit unlikely) if continuous occupancy was to occur in some localised regions before the cleanup. While very few areas presented a significant hazard to the casual visitor even when dust raising activities were considered, doses in excess of 1 mSv per year were possible over a large area for continuous Aboriginal occupancy; this would be the case wherever the levels of ^{239}Pu exceed about 5 kBq/m^2. Because of the narrow plume structure of the contamination, 100% occupancy of contaminated areas by the very mobile Aboriginal people is most unlikely. However, the presence of plutonium in visually identifiable pieces of debris in megabecquerel quantities meant that deliberate collection by a visitor was possible and malicious misuse must be considered.

In circumstances like this, where the contamination is already in place and not subject to the normal controls of radiation protection, international principles of radiological protection state that dose limits are not directly applicable. Rather, a strategy aimed at minimising the overall harm should be formulated based on what is reasonable, having regard to economic, social and environmental factors. The cleanup strategy adopted involves a judgement based on weighing the benefits of a cleanup (e.g. doses averted by the public, ability to reuse land) against the detriment in cleanup (e.g. doses to workers, ecological cost, financial cost). The contamination level used to determine the soil removal areas was based upon knowledge of the present lifestyle of the semi-traditional Aborigines and their life expectancy, and took into account their values. Occupational exposures, however, are treated as for any ongoing radiation activity with full protection and personal monitoring being given to workers.

The Maralinga Aboriginal people have indicated a general acceptance of this approach. They were well aware that, pre cleanup, some of the contaminated areas were considered dangerous for continuous traditional occupation by them. The dilemma faced by the Maralinga people was what form of cleanup could adequately deal with this situation, and they have been very concerned about the gross environmental damage that the removal of too much top soil from large areas would cause — as it would involve the removal of every tree and blade of grass from an area of verdant bush land, with potential for subsequent erosion problems. The decision of the traditional Aboriginal owners has been that they are not prepared to solve one environmental problem by creating another [1]. They have thus reluctantly accepted the 'fencing off' of several hundred square kilometres of their traditional lands as being unsuitable for permanent occupation.

3. THE CLEANUP PROGRAMME

A Technical Assessment Group (TAG), set up by the Government of Australia following the report of the Royal Commission into British Nuclear Tests in Australia, identified a range of options for rehabilitation of the Maralinga lands based on a series of scientific studies. After consultation with the South Australian Government and Maralinga Tjarutja, a preferred programme of remediation was decided. This involves removing surface soil from the worst contaminated areas, and restricting Aborigines living a semi-traditional lifestyle from permanently occupying a further area of about 400 km^2 of land. The Maralinga Tjarutja has been compensated for denial of full access to this area.

The standard for this intervention is that the annual committed dose, for any scenario involving permanent occupancy by semi-traditional Aborigines, will be less than 5 mSv. In fact, following the cleanup, annual doses are not expected to exceed 1 mSv for any realistic scenario. The possibility of intentional behaviour, such as fragment scavenging, has led to limits on particulate contamination.

3.1. The 'non-residential area'

A non-residential area will be established comprising those areas in which the expected annual dose by inhalation for 100% occupancy to the critical group (Aboriginal children living a semi-traditional lifestyle) exceeds 5 mSv per year. At Taranaki, this corresponds to approximately 3 kBq/m^2 of ^{241}Am and, because campsites are moved frequently, averaging over 3 km^2 is appropriate. An annual committed dose of 5 mSv and its associated risk of fatal disease of approximately 10^{-4} per year at age 50 have been accepted by the TAG and the potential Aboriginal inhabitants as a reasonable limit. The boundary of this non-residential area has been marked at close intervals with signs to indicate that the area is suitable for hunting but not for camping. In practice, the boundary follows existing roads and tracks and includes a larger area than is strictly necessary.

Within the non-residential area, transitory activities such as hunting and travel will be perfectly acceptable. However, routine use will be discouraged by the removal of some defined tracks and by re-vegetation of some areas. Alternative routes, passing around the area, will be improved to encourage their use.

3.2. Soil removal boundaries

Taranaki — Within the non-residential area, close to the Taranaki test site, short duration visits prior to the cleanup, particularly if they involved dust raising and large respiratory volumes, could still have given rise to unacceptably high doses. Moreover, the presence of highly active fragments and particles made the contamination of wounds and the deliberate collection of plutonium possible. Such hazards are difficult to quantify. Therefore, it was decided to remove, entirely, the contaminated soil along with contaminated debris from areas in which:

— the average level of ^{241}Am over a hectare exceeds 40 kBq/m^2; and/or,
— particles and fragments exceeding 100 kBq ^{241}Am are present; and/or,
— particles of >20 kBq ^{241}Am exceed a surface density of 1 per 10 m^2.

By limiting the activity of the remaining soil to below 40 kBq/m^2 of ^{241}Am, and by limiting occupancy factors to those typical of hunting activities in a particular location (0.8%), worst-case annual doses of less than 5 mSv can be anticipated. An area of about 1.5 km^2 has been treated by removal of surface soil at Taranaki. The need to satisfy the second and third criteria relating to particles and fragments has meant that the worst-case dose will be, in fact, lower than expected.

TM100, TM101 and Wewak — As these three sites are to remain outside the area of restricted occupancy ('fenceline'), cleanup levels are required to be more stringent than for Taranaki; essentially, the areas that would otherwise have been enclosed within the fenceline have been cleared of surface soil to levels acceptable outside the fenceline. The appropriate average americium levels for TM100 and TM101 are 1.8 and 4.0 kBq/m^2 respectively, and for the plumes at Wewak the appropriate average level is 1.8 kBq/m^2. The criteria for particle/ fragment densities are the same as at Taranaki.

Areas of about 0.46 and 0.31 km^2 have recently been treated by removal of surface soil at the TM sites and at Wewak, respectively.

3.3. Clearance criteria

The criteria to be met following soil clearance for particles and fragments were the same at all sites as the criteria used to set the soil removal boundaries. With regard to dispersed activity, at Taranaki contaminated soil was removed to achieve levels less than 3 kBq/m^2 (averaged over one hectare). This has the virtue that should a future reassessment dictate lower levels for the cleanup boundary, then it is unlikely that further removal of soil would be required from the area already treated. In circumstances where the achievement of the 3 kBq/m^2 clearance criterion was practically very difficult, ARPANSA was able to authorise an upper limit of 10 kBq/m^2. In fact, the highest value approved was 3.8 kBq/m^2.

At the sites TM100, TM101 and Wewak, contaminated soil was removed to achieve levels of americium at or below the levels used to define the soil removal boundary, with averaging over one hectare.

3.4. Soil removal

After trials to assess the suitability of various types of plant for removing the sandy Maralinga soil in thin layers, the standard scraper was identified as the most efficient for soil removal, transport and effective deposition of soil in burial trenches. All plant involved in the dusty soil removal operations had modified cabins to protect the operators, who worked within a sealed and pressurised cabin, with filtered air intakes and extracts, without the need to wear special personal protection equipment.

The soil removal areas were divided into individual Lots of 3–4 ha area. In the soil removal process, cuts of about 100 mm depth were taken except where windrows were present from earlier ‘ploughing’ operations. In this case, the total windrow plus a further 100 mm depth was removed as the windrows contained much of the contamination. After such treatment, there was a need for ‘operational monitoring’ to be conducted (by someone other than the regulator responsible for the signing off of Lots) over the cleared area, to give a first estimate of whether further treatment was needed to meet the clearance criteria. Based on the results of the operational monitoring, the Lot was either thoroughly monitored to check for compliance with the clearance criteria, subject to a further general cut of 100 mm depth, or given spot treatment such as the removal of individual particles or fragments or small scale soil removal.

The contaminated soil and debris has been buried well above groundwater levels in trenches excavated close to each of the sites and covered with a minimum of 5 m of clean fill. Currently, contaminated debris in 21 pits is being rendered practicably inaccessible by exhumation of the pit contents and burial at depth.

3.5. Field monitoring

3.5.1. Detector systems

In order to define the boundaries of the soil removal areas, or to demonstrate that a site conforms to certain clearance criteria following the removal of contaminated soil (‘verification monitoring’), the quantity of ^{241}Am per unit area in the surface soil had to be measured and the number and activity of particles and fragments estimated. These tasks required that two sets of measurements be made.

The first, derived from the risks associated with the inhalation pathway, measures the average level of americium, and hence plutonium, contamination in the surface layer of soil. The photon emissions from ^{239}Pu itself are too low in energy and intensity to be of much practical use in surveying large areas of soil. The 59.5 keV γ-ray from ^{241}Am, almost universally associated with ^{239}Pu, penetrates several centimetres of soil and metres of air and allows large areas to be surveyed, even from altitudes of tens of metres in aircraft.

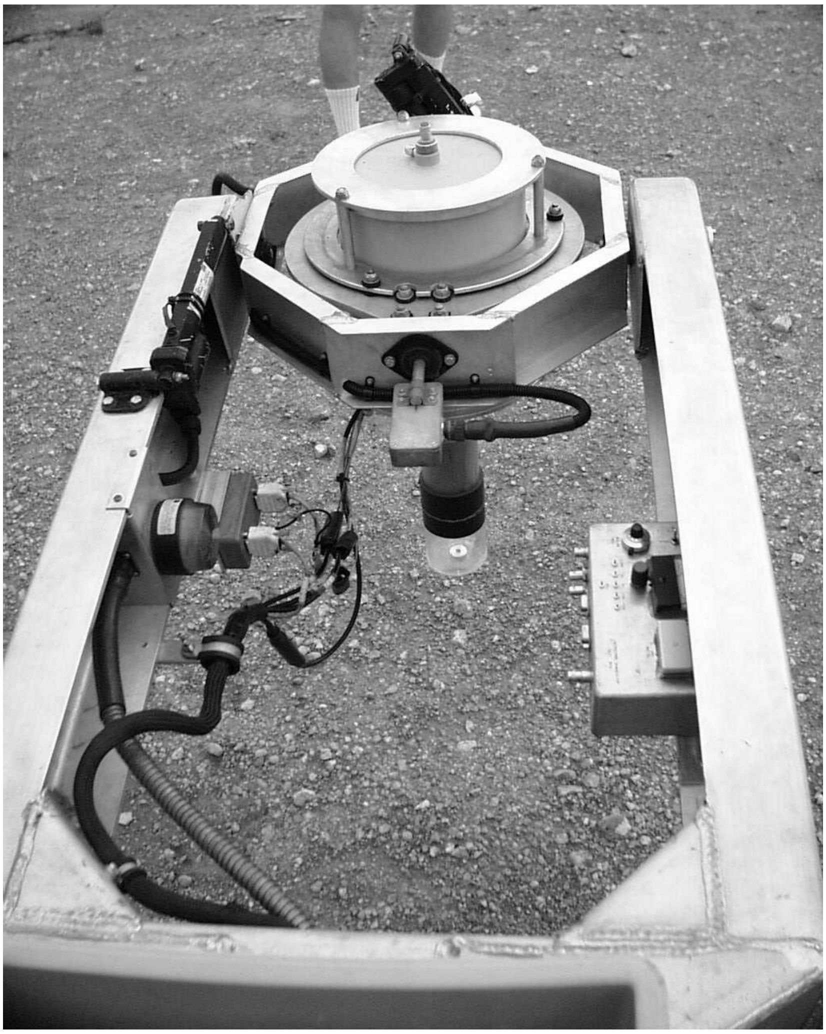

FIG. 2. Germanium detector mounted on gimbal assembly at end of boom, showing calibration source being measured, and test electronics.

The average level of contamination is measured with a closed-end coaxial instrinsic γ-ray detector (Fig. 2), held at 4 m above ground level on a boom mounted on a light truck (Figs. 3 and 4). Both the front face and cylindrical sides of the detector intercept the γ-ray flux emanating from the ground, and a single measurement effectively averages over an area of thousands of square metres. The good resolution (approximately 0.8 keV FWHM at 60 keV) allows easy separation of the ^{241}Am γ-ray from the background contributions of natural radionuclides (^{40}K, U- and Th-series), the Compton continuum from these radiations, and the contributions from fission products and neutron-activation products remaining from the major trials.

A full spectrum to at least 900 keV is acquired and stored by use of a portable MCA system and notebook PC. All the electronics is powered from a stabilised DC supply derived from the 12 volt vehicle supply. The vehicle provides an air-conditioned environment for the operator and electronics but the detector itself remains exposed to the outside temperatures which can approach 50°C in summer. Some reflective protection from direct sunlight has been found useful but generally the 5 or 7 litre portable dewar has proved adequate for a full day of operation in the field. The detectors used are ORTEC POP-TOP detectors of nominally 25% efficiency. The electronics comprises a CANBERRA INDUSTRIES INSPECTOR together with an IBM THINKPAD computer. A custom-built hydraulically operated boom allows the detector to be positioned at 4 m height at about 4 m in front of the vehicle, or to be brought right down to ground level for close measurements or access during calibration and setting up. For transport, the boom pivots back over the vehicle where the detector can be reached from the rear tray of the truck. The boom may be raised or stored in about 30 seconds. The vehicle is a diesel-powered, four-wheel drive 5 tonne OKA truck built in Australia. Measurement times of 600 – 1000 seconds are normal allowing a minium detectable ^{241}Am activity of approximately 0.3 to 0.5 kBq/m^2 depending on the background at the location.

FIG. 3. OKA γ-ray spectroscopy vehicle with boom and detector in retracted position for movement between sites.

FIG. 4. OKA γ-ray spectroscopy vehicle with boom extended in measuring position (detector not fitted).

In assessing the contamination level from the high-resolution γ-ray measurements, it is usually assumed that the americium (and plutonium) is distributed uniformly over the surface and is distributed exponentially with depth in the soil. In particular, a depth distribution with a characteristic depth of 0.5 cm is taken as standard. This agrees with measurements of the soil depth profile from undisturbed areas but is, of course, very different from that found in areas where soil mixing or other disturbance has occurred. However, because the inhalation pathway involves primarily dust raised by mechanical action on only the top 1 cm of soil, this assumed depth distribution provides a realistic and conservative calibration. Where the contamination is distributed to much greater depth,, the measurement will underestimate the total inventory of americium in the soil but will lead to a slight overestimate of the inhalation dose. Where the contamination is much shallower, the measurement will overestimate the amount of contamination but, again, overestimate the inhalation dose.

The calibration is derived from a mathematical calculation and from the measured response of the detector in the laboratory. It takes into account the variation with angle of the detector response, the inverse-square behaviour of the count-rate from a distant source, attenuation in soil and in air. A numerical integration provides a calibration which, for the situation at

Maralinga, is approximately 0.4 counts per second in the 60 keV peak per kBq/m^2 of ^{241}Am on the ground. This semi-empirical value has been confirmed by measurements made in the field at a site where numerous soil samples have been taken and analysed. Unfortunately, due to the particulate nature of much of the contamination, there is very wide variation between one soil sample and the next and this intercomparison only allows verification of the calibration to, perhaps, ±30%. This is, however, adequate for assessment of dose for risk assessment purposes.

The second measurement technique is required to determine the presence of particulate contamination. While the results of the high-resolution spectrometry set an upper limit on the numbers and activities of particles, the clearance criteria require that particles and fragments be surveyed in a separate operation. For most of the survey work to define the soil removal boundary, this was done by use of handheld 5 cm diameter by 3 mm thick sodium-iodide detectors incorporating a single channel analyser tuned to the 60 keV γ-ray. By scanning the ground with the detector held only a few centimetres above the soil, particles down to few Kilo–Bequerels of americium may be detected. Once located, the activity of the particle is estimated by comparison of the count-rate with that from standards. This method is, however, labour intensive and tedious. Automated methods were examined, but replacing the ability of a human operator to detect and then confirm slight increases in an audible count-rate proved difficult. However, the exhaustive particle searching required for the verification monitoring has necessitated the design and construction of an automated system.

FIG. 5. Nissan particulate contamination detection system.

Four detectors, 12.5 cm diameter and approximately 2 mm thick, have been placed at 0.5 m centres at about 28 cm above ground level on the front of a four-wheel drive Nissan utility vehicle (Figs. 5 and 6). In this case thin-crystal sodium iodide detectors (BICRON FIDLER G5) are used. The thin crystal provides significant rejection of the high energy background and is still fully efficient at 60 keV. When driving at a nominal 1.7 m/s the system can reliably detect every 100 kBq particle in its 2 m wide track and cover a hectare in 1–2 hours. The detection of 20 kBq particles is essentially statistical with a significant background rate (several hundred per hectare) and only approximately a 50% detection efficiency. This is somewhat increased by the multiple coverage provided by adjacent detectors, overlapping passes, and successive counting intervals.

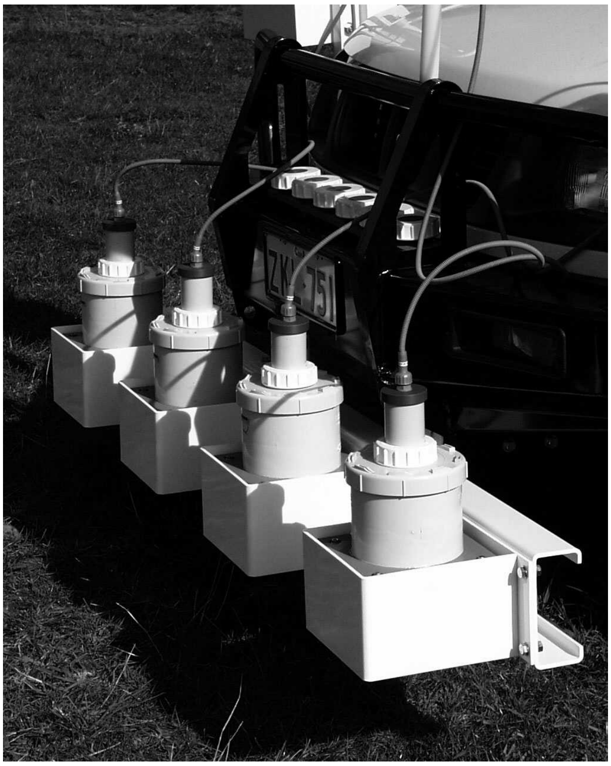

FIG. 6. Four large area NaI detectors mounted in protective canisters on the Nissan vehicle.

A differential GPS system provides accurate position information. The front end electronics used with the FIDLER detectors comprises modified commercial portable scaler/ratemeters made by LUDLUM. This equipment runs from 3 volts DC and provides the high-voltage photomultiplier supply, the pre-amplifier, and pulse–height discrimination. The rate-meter gives useful information for diagnostic purposes. Pulses are derived from a single channel analyser window set around the 60 keV peak, approximately from 40–80 keV. The threshold and window voltages are set either by multiturn trim potentiometers built in to the LUDLUM ratemeters or by external voltages controlled by the computer interface. Under computer control, the threshold and window can be wept from zero to about 100 keV and a 50 channel spectrum obtained. This provides the necessary information to adjust the high-voltage and to set the built-in threshold and window. The threshold, window and high-voltage supply voltages, as well as the battery supply voltage, are all monitored continually by the computer to ensure correct operation. A later version provides full computer control of the threshold, window and HV and eliminates the need for screwdriver adjustments.

In normal operation the pulses from each of the four counting channels are counted continuously together with a 1000 Hz clock as a timing reference. Every time the vehicle advances by a nominal 20 cm all five counters (four detectors plus timing reference) are read and stored. The data in three successive counting intervals are summed and examined to determine whether or not a high-activity particle has been observed. In this way a 60 cm path over the particles and roughly centred on it is used to measure its activity. The alternative of using fixed duration counting intervals can include unnecessary amounts of background when the particle is not under the detector. Background rates of 10–20 counts per second are observed in the field and the sensitivity is approximately one count per second per kBq of ^{241}Am. The actual efficiency depends on the lateral displacement of the particle relative to the detector. At the nominal forward speed of 1.7 m/s, the effective counting time is 0.35 s giving background counts of about 5 and counts for a 100 kBq particle of 35. The 100 kBq particle will be usually detected several times as it approaches and leaves the detector. The efficiency for detection of 100 kBq particles is effectively 100% and the false positive rate only a few per hectare. Variations between one detector and another, changes in detector height due to vehicle movement and other factors mean that particles in the range 50 to 100 kBq may need to be investigated by handheld equipment to show whether or not they actually exceed the 100 kBq limit. The large detectors at 30 cm from the ground will also sum the effect of several smaller activity particles producing false signals in some areas.

3.5.2. Field survey methods

3.5.1.1. Determining cleanup boundaries

There are two main requirements for defining the soil removal area. The quantity of ^{241}Am per unit area in the surface soil must be measured and the number and activity of particles and fragments must be estimated. In-situ γ-ray spectroscopy is the main tool for measuring the overall level of contamination. As explained above, the inhalation hazard may be estimated from the apparent concentration of ^{241}Am as determined by the γ-ray that escapes the surface.

Previous surveys of contamination by soil sampling, by ground based high-resolution γ-ray spectroscopy, and by helicopter based aerial survey making use of a large array of sodium-iodide detectors [5], gave guidance as to the approximate locations of the soil removal boundaries at the appropriate ^{241}Am activity concentrations for each site. These boundaries were defined in detail by systematic measurements using the vehicle-mounted germanium detector at 4 m height. Boundaries thus determined were checked, largely by the handheld thin-crystal NaI detectors, for compliance with the particle criteria, and where these criteria were breached, extensive statistical surveys were require to define the new location of the soil removal boundary.

The vehicle-mounted detector system for particle searching was not available early in the boundary-marking phase of the work, and it is doubtful of its advantage due to difficulties of the generally rough terrain (Unlike the relatively smooth surface available after soil removal treatment). A prototype trolley-mounted system suffered due to both difficulties with the terrain and also its weight which made pushing it around in a hot environment difficult.

The lower levels of americium (approximately 3 kBq/m^2) at the boundary of the non-residential area (outer ‘fenceline’) will require longer counts but do not require high spatial resolution. As this boundary will follow existing roads and tracks, most field monitoring will be for verification purposes. The germanium detector and truck will again be used with satellite GPS navigation for determining locations.

3.5.1.2. Verification monitoring

For surveying treated areas of land, high-resolution gamma ray measurements are made on a roughly square grid of approximately 35 m spacing. Measurements are made as close as 15 m to a boundary where it is recognised that some contribution to count-rate will come from outside the measured area. This can be estimated, when it is important, by making a second measurement at 1 m from the ground. Where the activity is distributed uniformly over the ground the expected count-rate would then be approximately double that obtained at 4 m. If a lower or similar value is found, it may be deduced that significant input from far afield is occurring. Nine adjacent points on the grid cover an area of approximately one hectare and the clearance criteria at Taranaki, for example, specify that the contamination be less than 3.0

kBq/m^2 averaged over one hectare. In assessing an area, all sets of nine such points are taken and the measurements averaged. The worst-case location of the square of nine points is considered but contrived shapes are not. No account of uncertainties is made in calculating the average. Where no 60 keV peak is detected, the net peak area, be it positive or negative, is included to give an unbiased average.

Where the average for an area exceeds the clearance limit, some remedial action is usually required. Normally any active particles will already have been removed so the only course of action is further removal of contaminated soil from the most active regions. Detailed information from the particle searching scan described below can assist in locating the areas from which soil can most beneficially be removed. In cases where further removal of soil is impossible or the effort required is disproportionate, ARPANSA (as the 'regulator') is able to authorise levels of up to about three times the clearance limit.

The second set of measurements, usually performed first, is to search for, and remove if necessary, any highly localised pieces of contamination. Where the contamination is found on a visible piece of debris (a 'fragment') the activity is not usually taken into account although a value of a few kBq is a practical lower limit. For vehicle-borne equipment covering large areas, 20 kBq of ^{241}Am is about the lowest that can be detected with better than 50% reliability. Where the contamination is not visible (a 'particle') two levels of activity are considered under the clearance criteria. No particles exceeding 100 kBq are permitted to remain, and an average of less than one particle exceeding 20 kBq per 10 square metres is permitted. In order to detect such particles in the presence of background, it is necessary to place an appropriate detector close to, at least briefly, every part of the area to be surveyed.

All the count data, together with differential GPS position information, are displayed and stored on the notebook computer. Analysis of the computer records allows re-visiting the position of a potential 100 kBq particle for verification. Using handheld NaI equipment the activity can be measured more reliably and a decision made on what action is required. If the offending activity arises from a fragment, or a particle exceeding 100 kBq, it will usually be removed using hand equipment and the area rescanned to ensure it is now free of contamination. The GPS equipment provides coordinates accurate to about 1 m which ensures that only a small area around each potential particle must be scanned by hand.

Examination of the computer files produced by the particle scan allows checks for total coverage of the area. In practise, small drifts in the GPS coordinates can mean that a few gaps may occur in the coverage but generally better than 99% coverage is expected. A computer display shows the operator where areas require re-scanning.

The computer files also can show the average contamination levels throughout the area being scanned. Some uncertainty in the absolute level of contamination will exist due to the very variable background observed in the field. However, localised hot-spots of perhaps 10 – 20 kBq/m^2 are easily observed. Where the germanium detector results show the need for further remediation, the NaI results can produce useful spatial information.

4. CONCLUSIONS

The cleanup criteria were guided by conservative principles and by estimated doses for realistic scenarios. These included the possibility of an Aboriginal group living for a whole year on the edge of the non-residential area in regions of the highest activity permitted outside it (~20–35 kBq/m^2 of ^{239}Pu depending on the site). This could lead to an annual dose of 5 mSv. If, more realistically, the group spent its time randomly over the Maralinga lands outside the restricted area, or even randomly around its perimeter, average activity levels and hence doses may be confidently expected to fall by at least an order of magnitude.

Similarly, the criterion for soil removal at Taranaki was based on 0.8% occupancy of the most contaminated remaining land within the non-residential area. This was consistent with a tenth of the typical hunting time being spent in this one region. Most of this land will contain levels of plutonium far below the maximum residual level and randomly distributed hunting would incur much lower intakes of radionuclides.

While arguments about occupancy levels and consequent doses are subject to a certain amount of arbitrariness, the rehabilitation, when complete, will have removed three of the present possibilities for very high doses:

— Soil will have been removed from areas where the production of large amounts of dust in locations of high plutonium concentration could lead to doses exceeding acceptable limits.
— Areas containing highly active particles or large numbers of particles will have been cleaned to prevent the deliberate collection of contaminated fragments and particles.
— Burial pits containing debris contaminated with plutonium will be rendered practicably inaccessible by the process of exhumation and burial at depth.

In the case of short term visitors making intermittent forays to the cleanedup site, for example tourists, geological prospectors and surveyors, inhalation doses of very much less than the 1 mSv annual public exposure level can be confidently assured.

During the field monitoring work being conducted in support of the actual cleanup process, data are also being obtained from remediated areas to allow a final dose and health risk assessment to be performed for various scenarios and potential occupants once the cleanup is complete and the area is released from institutional control. These data include remaining levels of contamination, both within and outside of the treated areas, and a study of resuspension parameters (including plutonium concentrations and particle size characteristics in resuspended material) from the remediated areas. However, one can confidently predict that not only will the cleanup have achieved all its objectives, but it will have actually done better in that predicted annual doses for all realistic scenarios are expected to be very considerably lower than the required value of 5 mSv.

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SITE CHARACTERISATION PROGRAM AT THE BELGIAN NUCLEAR RESEARCH CENTRE SCK·CEN

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ABSTRACT

Environmental restoration is of importance. In Belgium, there is primarily a need for restoring radioactively contaminated sites and the need for assessing the radiological impact of waste disposal or discharges. Participation in international programmes is necessary for keeping in touch with important developments. SCK·CEN has been performing site characterisation for more than twenty years within the framework of its statutory mission, but also within the framework of international programmes aiming to improve the safety of contaminated sites and the environment in general. The paper presents the most important activities which has been performed in the following fields during the period covered by the RCM.

1. RADIOLOGICAL CHARACTERISATION

1.1. Radiological characterisation of sites

Projects dealing with radiological characterisation of sites have been or are still carried out for the Belgian Federal Government and for the private industry:

- Radiological characterisation of the surroundings of a former Radium production plant; the co-contamination with heavy metals was characterised in cooperation with VITO (Flemish Institute for Research in Technology, regionalised former part of the SCK·CEN).
- Radiological characterisation of waste dumps of a Phosphate production plant.
- Radiological characterisation of the Belgian territory after the Chernobyl accident.
- Radiological survey programme of the Belgian territory.
- Monitoring and cleanup of roads in a site of power plants.

SCK·CEN developed the characterisation strategy for these projects, and developed or adapted the necessary instrumentation for the specific needs of each project.

1.2. In-plant characterisation

In-plant characterisation is a daily activity of the SCK·CEN.

Personal dosimetry (external exposure, surface contamination, internal exposure), measurements of the ambient radiation field, measurement of contamination (volumetric and superficial, of air, ...), monitoring of waste streams, etc. are routinely performed as management activities.

1.3. Characterisation of radioactive materials

The SCK·CEN performs radio-chemical analysis of numerous type of materials (liquid and solid) for private companies and institutional customers. Destructive and non destructive measurements of waste, nuclear fuel and other nuclear materials are daily performed by the Radiochemistry services. The SCK·CEN has been “accredited” by the BELTEST national board for the QA-system for several of these measurement techniques.

1.4. Non radiological characterisation.

Non radiological characterisation of sites is an important part of running the projects dealing with disposal of waste and site remediation.

Within the framework of assessing the Boom clay layer underneath its site as a potential host rock formation for a high level radioactive waste repository, SCK·CEN has performed the geological and hydrological characterisation of the region. The underground has been fully characterised in order to estimate or determine the migration of radionuclide from the geo to the biosphere. The studies include the mechanical, geological, hydro-geological, chemical etc. characterisation of the site.

For near surface repositories, the policy followed by SCK·CEN with respect to the quantification of important parameters is to determine best estimate values and uncertainty ranges from literature review or from inquiries at competent services. These studies are performed for NIRAS/ONDRAF.

In a study carried out on behalf of CEC-DGXI concerning a general guideline for the radiological optimisation of site restoration [1], important parameters and site characteristics in the radiological assessment procedures have been identified.

In a study carried out on behalf of CEC-DGXI concerning a general guideline for the radiological optimisation of site restoration , important parameters and site characteristics in the radiological assessment procedures have been identified. In this RESTRAT project [2] [3] working packages are devoted to the characterization of sample sites (Drigg repository, Neet-river, Ravenglass estuary, Ranstad) and to the dependence of the migration and transportation of contaminants on the composition and state (e.g. pH) of liquid and solid phases.

2. POLICY SUPPORT ON RADIATION PROTECTION

Belgium, as a member of the European Union, has the obligation to monitor the level of radioactivity in the air, water, and soil on its entire territory and to verify the discharge authorisations of nuclear installations. In this respect, the know-how of SCK·CEN in radiation protection and measurement techniques is placed at the service of the Federal authorities.

2.1. Objectives

- to support and advise the Belgian authorities on specific problems concerning existing and potential hazards from exposure to ionising radiation in normal and accidental situations;
- to provide training, education, and information regarding nuclear matters.

2.2. Programme

The programme in policy support on radiation protection answers requests from three customers:

- for the Federal Ministry of Public Health and Environment (DBIS/SPRI), an environmental monitoring programme of the Belgian territory, to investigate the impact of Belgian and neighbouring nuclear facilities, and the assessment of the radiological impact of the former radium production factory at Sint-Jozef-Olen;
- for NIRAS/ONDRAF, the review of the actual knowledge on the effects of low doses;
- for the Ministry of Internal Affairs and DBIS/SPRI, the support and expert advice on emergency planning and response.

2.3. Achievements

The radiological surveillance of the Belgian territory has been continued. We monitored the radioactivity of rainwater, airborne dust, water and sediments of the Scheldt and Nete, soil, milk, moss, aquatic plants, and fish.

SCK·CEN continues its participation as expert in the committee established to propose a restoration plan for the Olen site. The radiological impact of the D1 dumping ground [4] [5] [6] for different restoration scenarios was studied under contract with Union Minière.

Under contract with NIRAS/ONDRAF, SCK·CEN and AIB-Vinçotte Nuclear (AVN) synthesized the knowledge on the effects of low doses of ionising radiation. The resulting book is intended for persons interested in science, but not professionally occupied with the general subject. It tries to answer such questions as "what do we know?'", "what is still uncertain?", "what do the authorities do to protect us against radiation?".

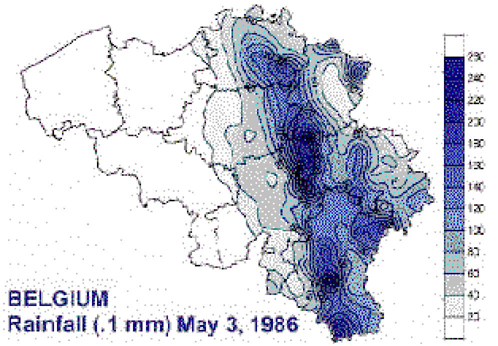

FIG. 1. Rainfall in Belgium on May 3, 1986.

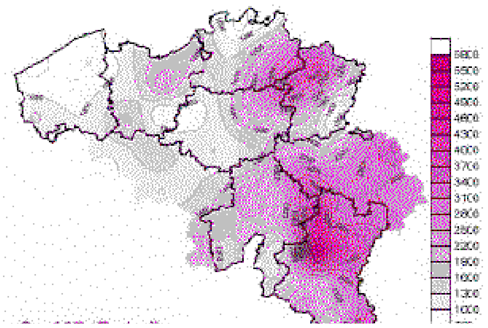

FIG. 2. Radiocaesium contamination in soil on May 3, 1986.

The in situ gamma spectrometry measurement technique for environmental radiation provides a powerful tool for the complete characterisation of the gamma radiation field at a given location. In 1995–1996, a decade after the Chernobyl accident, SCK·CEN and the Universiteit Gent have measured the residual radiocaesium concentration in the soil at more than sixty sites on the Belgian territory using in situ gamma spectrometry with low energy germanium detectors. Some level of correlation has been found between the geographic variations of the ^{137}Cs activity level in soil (Fig. 1) and the rainfall at the beginning of May 1986 (Fig. 2). Measurements performed during the period 1995–1996 are complementary to the ones performed in previous campaigns, shortly before and after the Chernobyl accident.

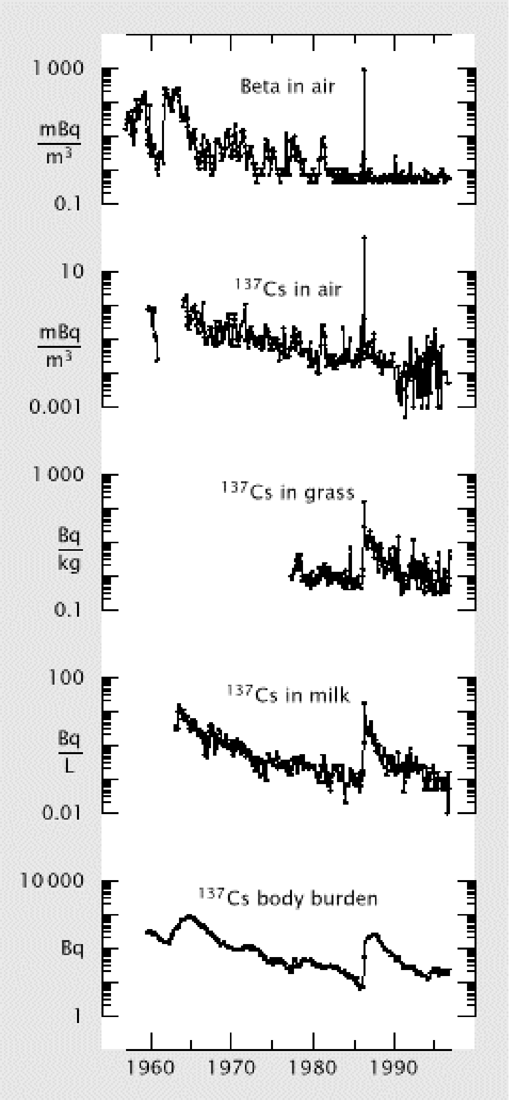

FIG. 3. Monthly average of (from top to bottom):

— *beta activity in airborne dust measured at SCK·CEN, Mol, Belgium;*
— *^{137}Cs activity in airborne dust at SCK·CEN;*
— *^{137}Cs activity in grass samples from a farm in the vicinity of Mol (Dessel, Belgium);*
^{137}Cs activity in milk samples from the same farm;
— *^{137}Cs body burden for a standard man of 70 kg.*

We summarised our systematic measurements on radiocaesium contamination in air, rain, grass, milk, and humans in Belgium from the late 1950s to present day. The average impact of atmospheric nuclear weapon tests on the human body burden in our region was shown to be more than six times that of the Chernobyl accident. The cumulated body burden from internal exposure to ^{137}Cs was 154 μSv between 1959 and 1986, and 25 μSv between 1986 and 1997 (Fig. 3).

In 1997, some additional in situ gamma spectrometry measurements on the Belgian territory showed a clearer geographical correlation between the ^{137}Cs activity level in soil and the rainfall on May 3, 1986 (Fig. 4). The ambient ^{137}Cs gamma ray field currently represents a dose of 0.6 to 7 μSv a year.

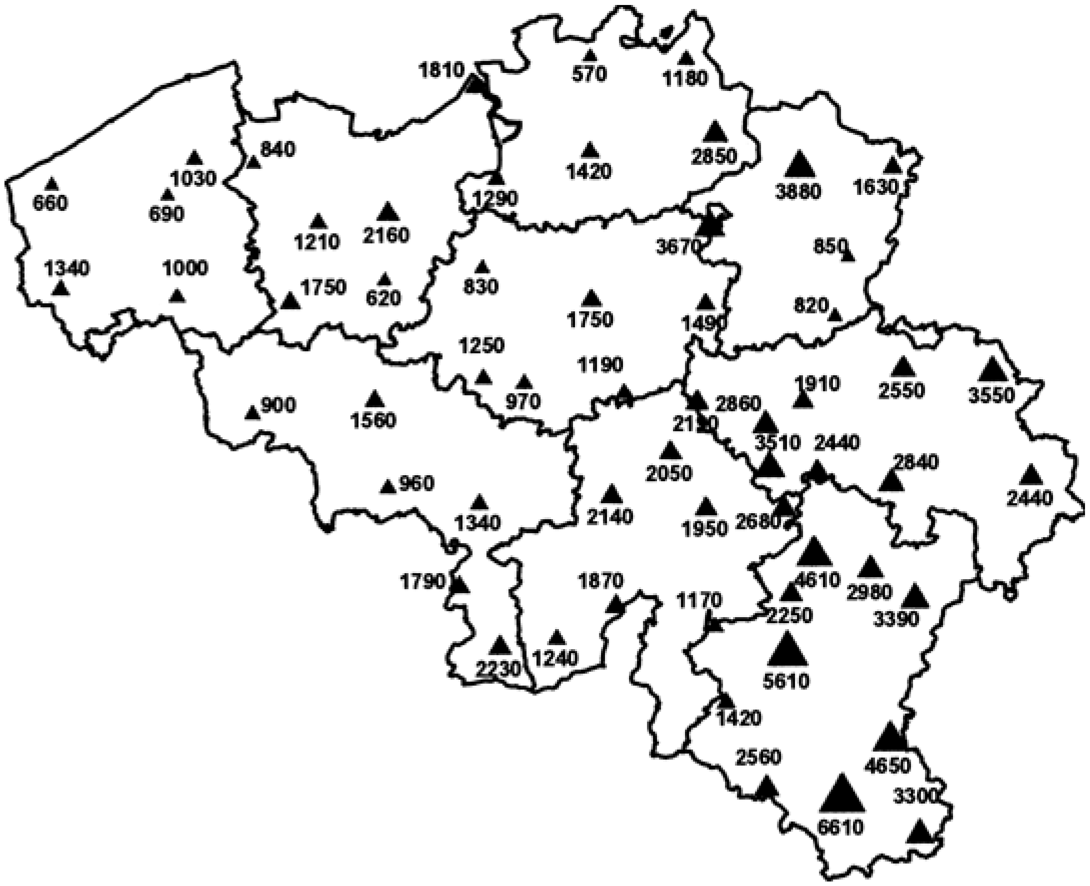

FIG. 4. ^{137}Cs soil contamination [Bq·m^{-2}] measured at different locations in Belgium. The measured activity level correlates well with the rainfall on May 3, 1986.

3. MIGRATION OF RADIONUCLIDES IN CONTAMINATED GEOMEDIA

The values of the model parameters for the migration of radionuclides and gases generated in deeply contaminated soils (e.g. after deep injection low liquid waste) can be determined by experiments on soil cores and by field tests. The models and the values of the model parameters are validated by large scale in situ experiments.

3.1. Objectives

- to understand the basic phenomena involved in the migration of radionuclides in the Boom Clay;
- to study the role of the Organic Matter (OM) in the migration of radionuclides (TRANCOM CLAY project) [7] [8];
- to develop a conceptual and mathematical migration model, and to determine the values of the model parameters by laboratory migration experiments on Boom Clay samples;
- to validate the migration model and the parameter values by large scale in situ migration experiments;
- to study the generation of hydrogen gas, due to anaerobic corrosion of metals, and its consecutive migration through the clay by diffusion and gas breakthrough (RESEAL, PROGRESS, and MEGAS projects) [9] [10] [23] [11];
- to test the validity of the gas migration model by large scale in situ helium injection experiments.

3.2. Programme

The programme, initially directed towards the migration of radionuclides in the Boom Clay formation, has been extended to the study of the generation and migration of gases, and to the study of the influence of the OM present in the clay water.

A similar method is followed for the three issues. First, the basic phenomena are studied; then a model is developed to help to design, simulate, and interpret the laboratory tests. Different types of experiments are performed for measuring the model parameters. These models and parameter values are used for performance assessment calculations and are validated by large scale in situ experiments.

Migration tests are carried out to study the diffusion of actinides, fission products, and nonretarded species, and the mobility of the dissolved OM in the interstitial clay water. Experiments with labelled OM allow to understand the role of the OM in the migration of radionuclides.

Large scale 3-D experiments with tritiated water and with ^{14}C-labelled bicarbonate, installed at the underground research facility, are in progress. They aim to confirm the anisotropy of the hydraulic parameters and to validate the migration model and the parameter values.

Two large scale in situ gas injection experiments are performed to validate the gas migration model. One large scale gas generation experiment is in preparation.

An existing shaft, lined with porous concrete blocks, is used as a macropermeameter to validate the values of the hydraulic parameters obtained from small scale tests.

3.3. Achievements

New migration experiments on clay cores were started for the following three radionuclides: ^{14}C, ^{241}Am, and ^{95}Zr.

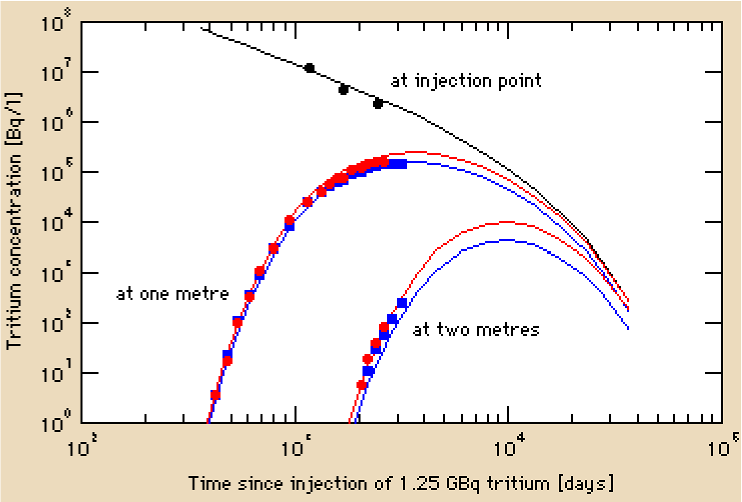

FIG. 5. After eight years, we still observe a good agreement between calculated (lines) and measured (symbols) tritium concentrations for the injection filter, for filters at a distance of one metre, and for filters at a distance of two metres.

The monitoring and sampling of the large scale in situ injection experiment with tritiated water, started in January 1988, was continued. After nine years, tritium has been detected at two metres from the injection point. The measured concentrations in the interstitial water agree very well with the results of the MICOF computer code simulation [12]. Figure 5 shows the good agreement between the experiment and the model calculation.

A large scale 3-D in situ migration experiment has been started in 1995. Tritiated water and ^{14}C-labelled bicarbonate have been injected. The first water samples from the filters at distances of one metre have been collected. Tritium is detected in three filters in the horizontal plane, but the ^{14}C level is below the detection limit.

To study the influence of temperature and radiations on the migration properties of radionuclides through Boom Clay, an in situ migration experiment with ^{241}Am and ^{99}Tc was performed in the surroundings of the CERBERUS demonstration test [13]. This experiment is now complete and interpretation is in progress.

Hydrogen gas reaction experiments with Boom Clay slurries showed the presence of methane and methane-forming bacteria, and also of sulfur-reducing bacteria.

In situ gas injection experiments indicate that, at breakthrough, the gas flows along preferential pathways alongside the injection piezometer (de-stressed zone). The breakthrough pressure is considerably lower than the value measured on confined clay cores. The local disturbance of the in situ stress field seems to govern the gas flow. After gas breakthrough, water has been injected to test the influence of the gas breakthrough on the local hydraulic parameters. The results indicate no change in the hydraulic parameters and no fracture flow.

The results after two years of an in situ macropermeameter test show that the values of the hydraulic parameters obtained from small scale tests on clay cores and on small in situ filters are reliable for large scale calculations.

A new method using an electric field for accelerating the migration of radionuclides has been developed (Fig 6) [15]. The first results are promising. We demonstrated and validated the applicability of the electrokinetic methods to determine diffusion coefficients in porous media (Fig. 7):

- diffusion coefficients can be obtained by independent calculation methods;
- the experimental time for diffusion studies is reduced markedly.

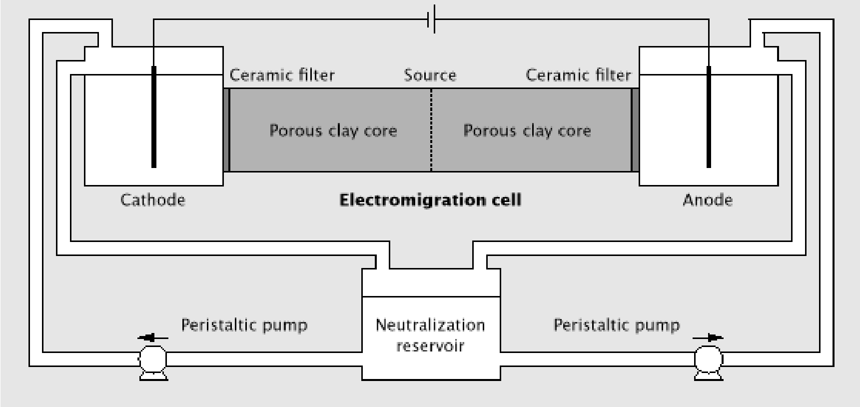

FIG. 6. Experimental set-up of the electrokinetic migration experiments.

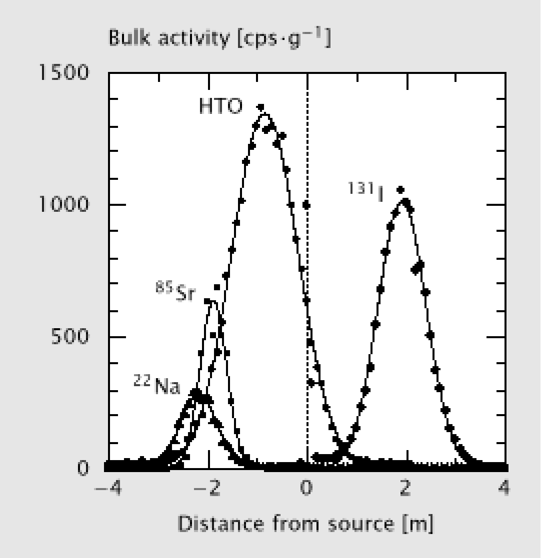

FIG. 7. Distribution profiles after electromigration of ^{85}*Sr (72 V·m*$^{-1}$*, 71 hrs),* ^{22}*Na (84 V·m*$^{-1}$*, 20 hrs), HTO (79 V·m*$^{-1}$*, 20 hrs),* ^{131}*I (81 V·m*$^{-1}$*, 20 hrs).*

In the framework of the collaborative EU/Russian Federation programme on radioactive waste management, the SCK·CEN evaluates the consequences of the injection of liquid high level waste in thin sand layers surrounded by thick clay layers in Siberia (Russia, CIS). SCK·CEN also contributed to an international project on the improvement of the safety of the radioactive waste management in the North West of Russia (Kola peninsula). The primary role of SCK·CEN is to transfer technology to the Russian partners (i.e. performance assessment methodologies including numerical models, parameter databases, etc.) and to assist them in performing safety studies for potential repositories in hard rock.

The use of networks of glass fibres for the monitoring of contaminated sites as well as for a post-remediation monitoring is being examined.

4. NON RADIOACTIVE CHARACTERISATION OF SITES

Since aquifers are essential components of the evolution of contaminated geomatrices due to their capacity for transportation of contaminants, accurate hydrogeological modelling is a critical element for assessing the long term behaviour of the environment.

4.1. Objectives

- to collect geological, piezometric, and hydraulic data required for studying the hydrogeological system in the Mol area;
- to develop a regional aquifer model for Northeastern Belgium;
- to test and validate computer codes used in the performance assessments for simulating water flow and transport of radionuclides in aquifers.

4.2. Programme

The hydrogeological studies sponsored by NIRAS/ONDRAF are piezometric measurements in the boreholes of SCK·CEN's hydrogeological network and the elaboration of a data-acquisition campaign aiming to collect data needed for the further development of the multilayer regional aquifer model of Northeastern Belgium. The data-acquisition campaign is complemented by an internal R&D project investigating the spatial variability of the hydraulic conductivity of the Boom Clay.

4.3. Achievements

For simulating the transport of radionuclides released from the host clay layer through the Neogene aquifer into the biosphere, an intermediate scale aquifer model is being developed using the Groundwater Modelling System (GMS) package [14]. The modelled area is roughly triangular in shape and limited by natural borders consisting of the rivers Kleine and Grote Nete and the water divide Scheldt-Meuse.

We started to develop a system of quality-assurance procedures for the following aspects of the performance assessment: collection of data for consequence calculations, verification of the software applied in the assessment, and keeping of complete records of the performed calculations.

Four boreholes have been foreseen as part of the hydrogeological data-acquisition campaign. The boreholes reach the top of the Ieper Clay. Piezometers are installed in the two aquifers underlying the Boom Clay, i.e., the Ruisbroek-Berg and Lede-Brussel aquifers. Geophysical loggings have been carried out in the three boreholes. Cores have been taken from the borehole located at Zoersel. The hydraulic conductivity of clay cores taken from boreholes at Zoersel, Mol, and Weelde are measured in our laboratory to study the variation with depth of the hydraulic conductivity of the Boom Clay, and to look for correlations with the litho-stratigraphy of the clay layer. In the framework of an internal R&D project we collected clay cores in clay pits and during the excavation of the second shaft on the SCK·CEN site. In situ measurements of the hydraulic conductivity of the Boom Clay at Doel were carried out. The results of these measurements together with those obtained from the measurements on clay cores taken during the data-acquisition campaign will allow us to study the spatial variability of the hydraulic conductivity of the Boom Clay on a regional scale.

In the framework of an internal R&D project, we collected clay cores in clay pits and determined their vertical and horizontal hydraulic conductivity. The results of these measurements, together with those obtained from measurements on clay cores taken during the data-acquisition campaign, will allow us to study the spatial variability of the hydraulic conductivity of the Boom Clay on a regional scale.

5. RADON IN THE INDOOR ENVIRONMENT

Radon progeny is responsible for by far the largest part of the human exposure to ionising radiation. The key to a correct assessment of the health risk ensuing is a better understanding of the deposition behaviour of this progeny in the respiratory tract. This deposition depends critically on the interactions between the radon progeny and airborne indoor aerosol.

5.1. Objectives

- to investigate the deposition of radon progeny in the human respiratory tract by means of direct measurements as a function of aerosol conditions;
- to assess retrospectively the radon concentrations in buildings by means of volume traps.

5.2. Programme

The programme foresees the direct measurement of deposited radon progeny in the human respiratory tract by means of the bronchial dosimeter, an instrument simulating the airflow in the respiratory tract. In a first stage, these measurements will take place in the well controlled environment of a reference chamber, and in a second stage, in real field conditions of actual dwellings. The retrospective assessment programme foresees the development of the volume trap technique and is carried out in the framework of the Radiation Protection Research Programme of the EC, more precisely by taking part in one shared cost action and two concerted actions.

5.3. Achievements

In 1996, we completed the design, construction, and testing of the radon reference chamber. Figure 8 shows a picture of part of the installation. It actually consists of three separate chambers. The first chamber is a large one, with a volume of 4.15 m^3 and a surface-to-volume ratio of 3.75/m. It is equipped with special outlets for connecting the bronchial dosimeter. Absolute filters have been installed for obtaining an aerosol-free environment, and suitable in and outlets have been foreseen for connecting aerosol producing and measuring equipment and radon measuring devices. The radon concentration can be varied up to values of 0.5 MBq/m^3, using three separate radon sources. We purchased an atomiser for producing a standard aerosol, and a condensation nucleus counter for counting the aerosol particles. Both are state of the art devices produced by TSI, the firm that sets the standards in these very specific applications. The second chamber is small, with a volume of 0.075 m^3, to be used for calibrations of radon detectors, experiments with the volume traps, or other smaller experiments. The third chamber has a volume of only 0.010 m^3 and will be used as a buffer volume. All chambers were extensively tested for leaks and were found to be leak free. Figure 9 shows the results of such a leak test for the large chamber. In a next stage, accurate calibrations should enable us to determine the response of the bronchial dosimeter in well-controlled conditions.

Within the framework of the Radiation Protection Research programme of the EC, we made further efforts to extend the applicability of our volume trap technique for assessing radon exposures in the past. More specifically, due to the great difficulties encountered in some areas to find spongy volume traps, we tried to extend the technique to other materials, such as wood or fibreboard. A simple, accurate and fast technique was developed for the purpose of testing all relevant characteristics of any candidate volume trap material. It allows within one week to assess the response of the material to radon exposure, and to calibrate it in terms of ^{210}Po activity to be expected per volume unit of volume trap material for a certain radon exposure. It also gives the necessary information on the speed of radon diffusion throughout the material. All of this can be achieved by a simple radon exposure during typically two days, followed by a monitored exhalation of the absorbed radon. There is no need for detailed knowledge of the structure of the material itself, neither for complex radon transport models.

Moreover, for testing a new volume trap material, there is no need to wait for the actual ingrowth of ^{210}Po, followed by chemical separation, which would take at least three to six months. The method was applied to several types of wood and fibre board. More careful numerical analysis however has shown that none of these materials are good volume traps, since they contain a too high and very variable natural ^{210}Po background. Unfortunately this is almost always the case for wood, thus making wood generally unsuitable as a volume trap material. It is possible that in the future other materials with little or no ^{210}Po background can be found for use as volume traps. Certain types of synthetic materials, such as polyethylene, might be deemed good volume trap candidates using this technique.

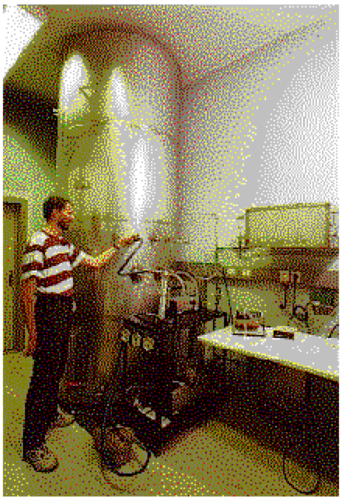

FIG. 8. The radon reference chamber. On the left is the large cylindrical radon chamber, connected to the three different measuring heads of the bronchial dosimeter. The smaller rectangular radon chamber is visible on the right. Fixed tubing interconnects all chambers and connects them to the three radon sources (bottom right).

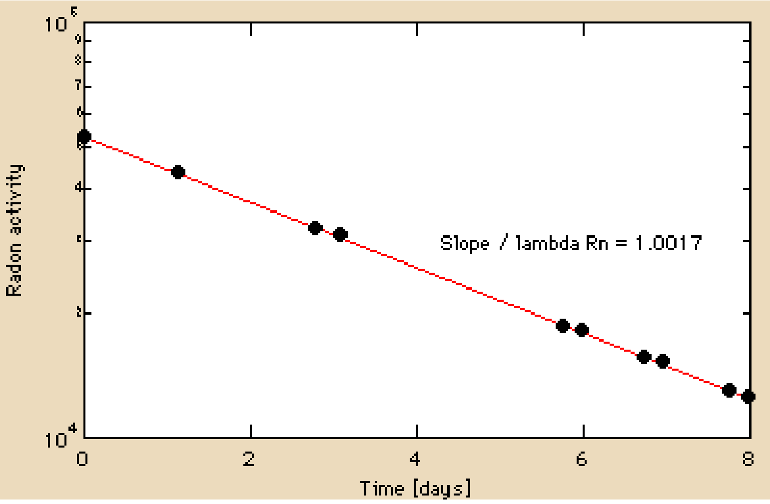

FIG. 9. Evolution of the radon concentration in the large chamber during one week following initial filling. The slope accurately matches the radon decay constant, showing the absence of leaks.

In 1998, a considerable amount of field work with volume traps has been carried out [16] [17] [18]. A decisive attempt was made to obtain them from interesting field locations, and to actually assess retrospective radon concentrations in dwellings from them. In total over two hundred volume traps were obtained from locations in Norway, Sweden, Germany, and Serbia. The ages of the retrieved volume traps varied between a minimum of two years up to as much as 60 years, with an average age between 15 and 20 years. Over 80% of them were polyester sponges from mattresses, chairs, cushions etc., of different sizes and shapes, densities and degree of cleanliness. The rest of them were mostly industrial wool or cotton, or even straw or seaweed. For the spongy samples, very reproducible and very good results were obtained. This was particularly the case for samples obtained from dwellings where a good radon history record was available. It was shown that very dirty sponges have to be treated more carefully, since in some cases radon decay products, coming from radon decay outside the volume trap, can penetrate to some extent into the volume trap, thus falsely increasing the resulting retro radon concentration. Figure 10 shows this phenomenon for a very dirty sponge retrieved from Serbia.

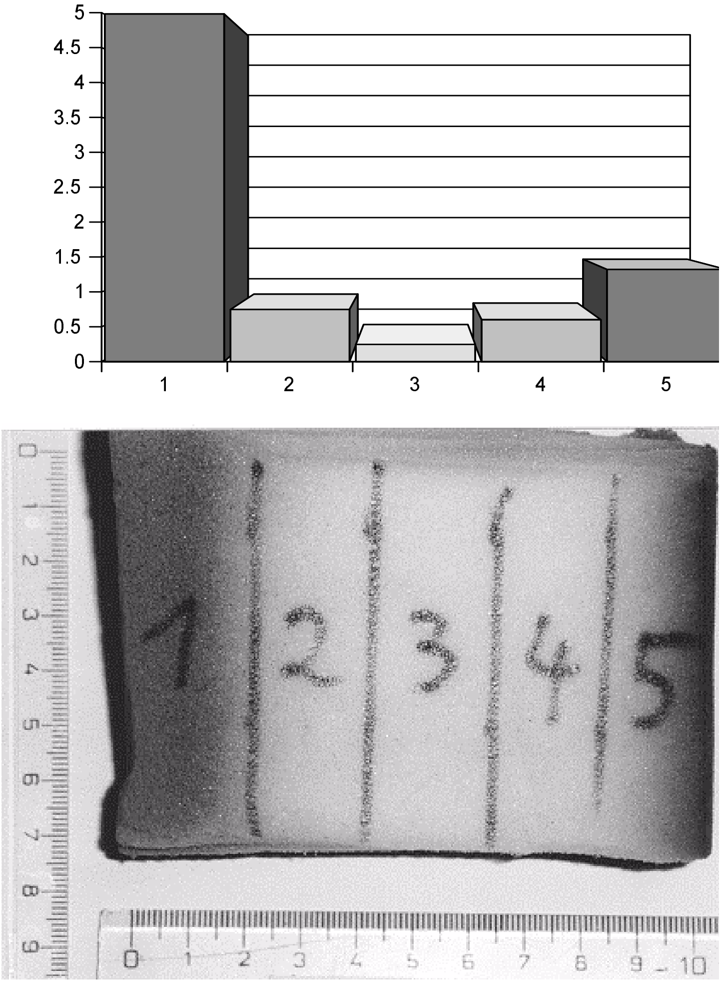

FIG. 10. Polyester spongy volume trap retrieved from a Serbian dwelling. The graph shows that only the central part which is free from dust yields a reliable retro radon.

6. CHARACTERISATION OF THE POPULATION (EPIDEMIOLOGY)

Radiation workers are studied to allow direct estimation of cancer risk in populations with long term exposure to low doses of ionising radiation. An international multicentre study, coordinated by the International Agency for Research on Cancer (IARC/WHO) in Lyon, will combine data on radiation workers from 14 countries.

The methodology used for this study could be applied to the population living on contaminated sites.

6.1. Objectives

The overall objective of our study is to study cancer mortality and morbidity in nuclear workers in Belgium, to document the feasibility of retrospective cohort studies in Belgium, and to participate in the IARC study.

Specific objectives were:

- to extend our study in the Mol-Dessel region to other nuclear facilities in Belgium;
- to complete data collection in the three participating facilities in Mol-Dessel.

6.2. Programme

We set up a retrospective cohort study (1953–1994) at five nuclear facilities in Belgium: SCK·CEN, Belgonucléaire, Belgoprocess, and the power plants of Doel and Tihange. The data will be pooled in the IARC study. Prospective follow-up of the present cohort study will be set up to monitor cancer mortality in these workers (cancer mortality registry).

6.3. Achievements

So far, we included in our study 4,859 workers from SCK·CEN, Belgonucléaire, and Belgoprocess. Electrabel NV agreed to participate, and data collection has already started in its power plants of Doel and Tihange. We added about 3,000 workers to the study in 1997 [19] [20] [21].

A joint proposal of IARC and four European countries entitled "Studies of Cancer Risk among Radiation Workers in the Nuclear Industry" has been accepted by the Nuclear Fission and Safety Programme of the EC (concerted action). The collaborators are Belgium, France, Germany, and the United Kingdom.

7. BELGIAN CONTAMINATED SITES

7.1. Objectives

- to evaluate the radiological impact of environmental contamination from natural occuring radioactive materials (NORMs) from inter alia the former radium industry (Olen), the phosphate industry, coal-fired power plants (fly-ash), the oil and gas exploration and production industry, the building material industry (phosphogypsum), and the metal recycling industry (contaminated metal scrap). The use of the recycled materials is also considered.

7.2. Programme

There are many contamination situations where the unintentional presence of natural radioactivity leads to a significant increase in the exposure of workers and members of the public. Important industries where these problems exist:

- relics from the former radium industry (environmental contamination, lost sources, disposal areas, etc.) — Union Minière Olen;
- phosphate industry (phosphogypsum and calciumfluoride disposal areas, liquid effluents, etc.) — Prayon, Kuhlman and Tessenderlo Chemie;
- fly-ash form coal-fired power plants (fly-ash disposal areas);
- the oil and gas exploration and production industry (NORM can be found in the forms of sludge, scale, production water and radon in natural gas);
- the building materials industry (phosphogypsum and fly-ash are widely utilized in Belgium);

— the metal recycling industry (most of the contamination in scrap metal is in the form of NORM).

Our activities in this field mainly consist of evaluating the radiological impact of these natural sources of ionising radiation on workers and members of the public on behalf of the authorities and the industries in question.

7.3. Achievements

7.3.1. Radium extraction plant

As a consequence of operating, from 1922 till 1969, of one of the world's largest radium extraction plants, radium has been dispersed in Sint-Jozef-Olen and Geel (Belgium). An extensive radiological characterisation programme identified several contaminated sites. The parties concerned agree increasingly in favour of a global restoration plan, not because of the current population exposure, but because most of the scenarios concerning the future use of these areas show enhanced exposure of future generations.

The site of Olen, around the Bankloop brook, provides also a test scenario for BIOMASS (BIOsphere Modelling and ASSessment methods), an IAEA Co-ordinated Research Programme (CRP), providing an international focal point in biospheric assessment modelling [22]. The two major themes of BIOMASS are indeed radioactive waste disposal and environmental releases, the latter further divided into dose reconstruction and remediation assessment. SCK•CEN is participating in the first theme on behalf of NIRAS/ONDRAF in the framework of its radiological impact assessments for radioactive waste disposal or discharges, and is leading the working group on remediation assessment in the second theme.

On behalf of the Belgian Federal Government (DBIS/SPRI), SCK•CEN wrote a compilation of all relevant information, so that the competent authorities can decide on the necessary licences to carry out the environmental restoration in Sint-Jozef-Olen and Geel. The waste volumes to be removed depend on the selected action levels (Table 1).

TABLE 1 SUMMARY OF THE PROPOSED ACTION LEVELS AND THE RESULTING (ROUNDED) WASTE VOLUMES. THE DUMPING GROUND CAN HAVE 0.4 μSV/H IF DUG DEEPER THAN ONE METRE AND THEN COVERED WITH CLEAN SOIL.

Contaminated site	**Action level (gamma dose rate, μSv/h)**	**Waste [1×10^3 m^3]**
Dumping ground	0.2	125
River	0.15	10
Farmland	not considered	30
Streets	0.2	1
Total		**136**

Our radiological assessment of the five options for restoration of the dumping ground was based on the following selection criteria: the cost of restoration, the surface area of the new storage facility, the collective dose of the intervention workers, and the individual dose for people who will be living above the storage facility after the period of institutional control has ended. The best solution turned out to be removing the waste to a nearby location (with the radioactive waste burried under the chemical one) and applying a multilayer cover. The cost for the global restoration plan is estimated to be around 370 Million Belgian Francs, of which

81% is for restoring the dumping ground and constructing the storage facility, 17% for cleaning up the river, and 1% each for the streets and for the builtup ground. The municipal authorities of Olen decided to clean up the Kapellekensstraat in 1998 and to dispose of radioactively contaminated soil onto the dumping grounds. The action level for the cleanup of the Kapellekensstraat was set at 0.2 μSv/h. 500 to 600 m³ of contaminated material, mostly slag, with a dose rate between 0.5 and 15 μSv/h was removed to the so-called D1 dumping ground. The radium concentrations of the slag were up to 21 000 Bq/kg. During the cleanup activities a point source of 0.542 g and with a total radium activity of 122 kBq was discovered.

This cleanup work was monitored by SCK•CEN.

7.4. Tessenderlo Chemie

For more than half a century, the liquid effluents of Tessenderlo Chemie containing enhanced concentrations of radium and heavy metals from phosphate production, are discharged into the Grote Laak, a small river in the South of the Campine region. This has caused a significant contamination of the river and its surroundings. On behalf of the Flemish authorities (AMINAL) SCK·CEN has performed a large scale gamma scanning of the river banks and the flooded zones in combination with an extended sampling programme. The purpose is to map the radium contamination but also the heavy metal contamination, using radium as tracer. The report is due at the beginning of 1999.

8. SUMMARY

The fist objective of site characterisation is to collect sufficient environmental data to make a decision whether a remedial action is necessary. Although a radiological characterisation of the site is essential, non radiological characterisation is as important to predict the evolution of the contamination and to decide “how clean the site must become”. There must be data collected, among others, on the anthropology, irrigation practices, meteorology, pedology, geology, hydrology, etc. of the site.

The activities of SCK·CEN covers most of these aspects. Characterisations methods have been developed and applied before and after remediation. In some cases, SCK·CEN participated also in the remediation actions.

8.1. Radiological characterisation

SCK·CEN has carried out, inter alia, the following radiological characterisations of contaminated sites for the federal government and for the private industry:

- Characterisation of the site of a former radium factory (Olen);
- Radiological characterisation of the water and banks of the Molse Nete river (Contamination is due to release of low level liquid waste);
- Radiological characterisation and follow-up of the Belgian territory after the Chernobyl accident;
- Radiological characterisation of the roads in the city of Geel;
- Radiological survey programme for the Belgian territory (DBIS/SPRI);
- Monitoring of entrance roads at the KC Doel nuclear power plant.

8.2. Non radiological characterisation

In the framework of remediation projects (Drigg repository, Neet-river, radium factory, etc.), SCK·CEN determine also parameters which are essential to know before starting the remediation work (K_d, pH, hydrological characteristics, organics, ...). SCK·CEN iniates also projects dealing with the bacterial characterisation of contaminated sites, as it is proven by long term monitoring of contaminated sites that the evolution of contaminations is influenced by microorganisms.

Further, SCK·CEN developed a elektro-kinetic method, accelerating the assessment of the migration radionuclides in soils, reducing the time needed by more than a factor of 100 and retaining a comparable accuracy.

In general, the experience of SCK·CEN in non radiological characterisation allows to predict the natural evolution of a contamination, and also to gather the parameters needed to assess remediation technology (transfer from soil to water, secondary effects of decontamination processes, etc.)

8.3. Other activities

SCK·CEN also has been, and remains, active in the collection of data (characterisation) needed to decide on remedial actions, e.g. epidemiology, evaluation of individual and global doses vs. remediation strategy (from “natural remediation” to “full restoration”, i.e. removal, treatment, conditioning, temporary storage and disposal of the radioactive or mixed contamination).

ACKNOWLEDGEMENTS

I should like to thank the IAEA for having organised this RCM on site characterisation techniques used in environmental restoration. I hope that the international collaboration initiated during this RCM will be continued in the future.

I thank also all my colleagues of the SCK·CEN who provided me the information necessary for my participation in the RCM and for the edition of the progress and final reports.

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RESTORATION OF AN URBAN SITE AFFECTED BY A MONAZITE PROCESSING PLANT

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ABSTRACT

For half a century the Santo Amaro Mill, USAM, processed monazite sand in order to isolate rare earth elements. At the beginning of its operation, the mill was located far from the centre of São Paulo city. However, over the years the city spread and engulfed the mill, which, together with economical and radiological problems, led to its being shutdown and later decommissioned. After the dismantling the equipment and the demolition of the buildings, a radiological survey was carried out, leading to the release of the site for unrestricted use. Based on a future residential occupation scenario complying with a dose limit of 1 mSv/y, it was concluded that a 0.65 Bq/g value of activity concentration of ^{228}Ra in the soil was the allowable residual limit of contamination for the site. The strategy to verify this requirement for site release comprised a preliminary soil survey by direct total gamma measurement with a sodium iodine scintillator detector, direct local gross alpha and beta counting of soil samples for monitoring during cleanup, and germanium detector gamma spectrometry of soil grab samples for the validation of monitoring methods.

1. INTRODUCTION

The Santo Amaro Mill (Usina de Santo Amaro, USAM), a branch of a State Company, was located in a densely populated residential district of São Paulo City, the largest city in Brazil (Fig. 1). In 1942, when it was built, the area was sparsely populated situated far from downtown São Paulo City. However, over the time the city embodied the mill. The main purpose of the plant was the separation of the heavy minerals zirconite, rutile, ilmenite, and the chemical processing of monazite ore. Due to public pressure, economic and radiological problems, USAM stopped its activities in 1992. The decommissioning started in 1994.

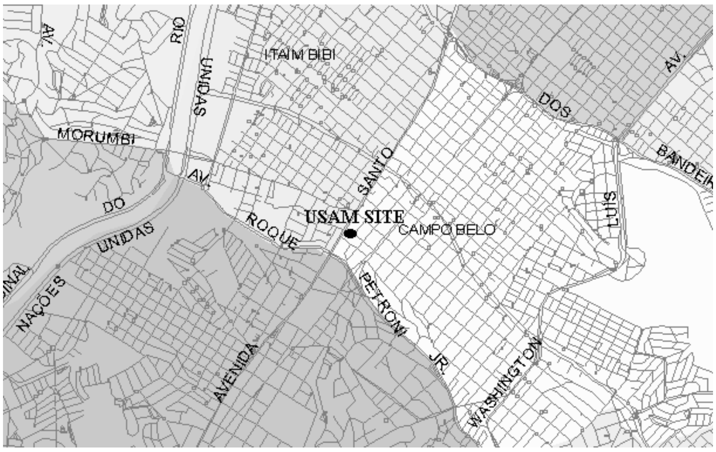

FIG. 1. Location of the USAM Site in the Campo Belo district of São Paulo City.

The products of the monazite chemical processing, based on sand alkaline digestion were lanthanide chloride solution and tri-sodium phosphate. From the consecutive steps of the monazite alkaline digestion, two main wastes resulted: a mixture of thorium and uranium hydroxides, named Cake II, and a barium radium sulphate, Ba(Ra)SO_4, named Mesothorium Cake [1]. It is known that besides the processing of mineral sands, some research concerning processing of other minerals, as well as the production of substances such as thorium nitrate were carried out at the mill. The presence of contaminants in the soil of the 16,000 m^2 site was suspected, since the radiation protection standards when the activities of the mill began were not as rigorous as today. Since there is a lack of information about the former activities in the mill and most records of its early history are lost, the presence of others contaminants was suspected, not just the wastes from the monazite processing. During the dismantling of the equipment and the demolition of the buildings, contamination had been found in some unexpected places.

The decommissioning was carried out in four stages: the intial stage included the suitable packaging and removal of remaining wastes from the plant, consisting of 532 tons of Cake II in 3250 barrels, and 73 tons of Mesothorium Cake in 745 barrels; the second stage was the decontamination and dismantling of the equipment; the third stage was decontamination of the floors and walls, followed by demolition of the buildings; and finally, the site radiological survey and its decontamination.

The Institute for Radiation Protection and Dosimetry (IRD), a branch of Brazilian Nuclear Energy Commission (CNEN), was responsible for the establishing of the criteria, the limits for residual radiation, and the measuring methods to be used during the cleanup process. The aim was for unrestricted release of the site. The company owning the site performed the decontamination, the progress of which was checked periodically by inspections and testing by IRD, with the aim to ensure that the requirements established were being complied with. The present report deals with the establishing of the limits, the conditions for the re-integration of the site into the city, and the research to find simpler and more economic methods of radiation measurements for monitoring of compliance with the release requirements of the area.

2. ALLOWABLE RESIDUAL LEVELS

The Brazilian Radiological Protection Guidelines established two limits that could be used in such situations. The limit for releasing solid waste to domestic landfills, and the primary dose limits for members of the public, which is 1 mSv/y. There are no generic limits for permissible remaining radionuclides in soil classified for unrestricted use of the land. To derive a limit, an approach was used, which was based on a scenario exposure pathway analysis in compliance with the annual dose limit of 1 mSv [2] [3] [4]. Considering the general local use of the land, the probable re-utilization of the site would be for residential or commercial high-rise buildings. A residential scenario was chosen as the most conservative one: A family living on the first floor was assumed. The thickness of concrete layer of the floor was 30 cm. The family had one child, which would stay indoors for around 5500 h/y and in the residential garden for about 700 h/y. The inhabitants did not consume water from the site, nor ingest any food grown on the site. The exposure pathways considered were direct exposure, inhalation of dust, and secondary ingestion of radioactive contamination. The pathway analysis indicated external exposure as the main exposure pathway, being responsible for ca. 80–90% of the total dose. The preliminary analysis of the samples by gamma spectrometry showed the thorium-series radionuclides to have the highest concentrations in soils. For this reason, the contribution of thorium-series radionuclides to the dose was higher than the uranium-series one, and it increased with the increasing of the ratio Th/U. As the values of the ratio $^{228}Ra/^{226}Ra$ in the soil ranged from 1 to 69, the contribution from thorium-series radionuclides to the dose reached values close to 100%. Hence, considering the equilibrium of Th-series, ^{228}Ra concentrations can be used as criteria for soil contamination. Considering the measured local background of ^{228}Ra soil concentration of 0.1 Bq/g, the allowable residual level (**ARL**) of ^{228}Ra was set to 0.65 Bq/g of soil.

3. RADIOLOGICAL CHARACTERIZATION OF THE SITE

The majority of data for this purpose were obtained by radiological survey using total gamma counting portable monitor and a sodium iodine “in situ” gamma spectrometer. Laboratory analyses of selected grab samples of soil were required for validation of analytical methods. In order to observe the established limit of soil contamination in a fast and economic way, the analytical methods that could be used were compared based on the available local monitoring equipment, a sodium iodine scintillation meter and gross alpha and beta counter.

The initial survey was performed by total gamma counting using a scintillation meter, SAPHIMO SPPE-2. The measurement was performed 0.5 meter above the ground surface and based on a 2 m x 2 m grid. In order to identify and determine the specific activity concentrations of radionuclides, soil samples were collected and analyzed using a germanium detector gamma spectrometer. These results allowed the conversion of the scintillation reading into specific concentration of radionuclides. In addition, total alpha and beta counts were determined in the samples in order to evaluate the adequacy of using this method for checking the monitoring during the clean up operation with a view to decide on further actions.

The soil grab samples were collected over 1 m^2 of soil surface and 3 cm of depth. Samples were split for the measurement by the two different methods: To perform gamma spectrometry, the soil samples were dried at room temperature, sieved to 2 mm-mesh and weighed before being analyzed. The ^{226}Ra and ^{228}Ra contents was determined by a high resolution intrinsic germanium detector a month later by the analysis of the line peaks of ^{214}Bi (609 keV) and ^{228}Ac (911 keV), respectively. For the analysis of beta and alpha emitters, the dried and sieved soil sample was placed on a stainless-steel plate. The alpha and beta radiation were determined in a gas flow proportional counter**.**

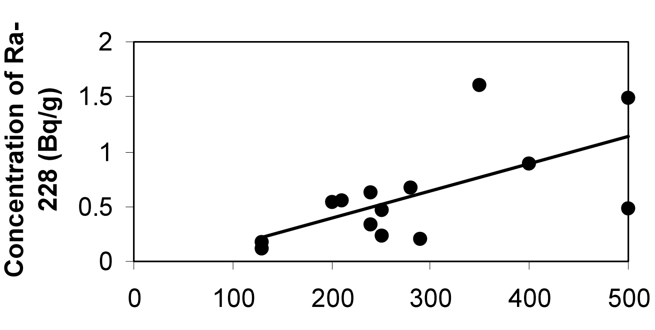

FIG. 2. Relationship between the scintillation readings and the ^{228}Ra activity concentrations.

For the correlation analyses of the measured data, only the values closest to the limit established (0.65 Bq/g) were considered. The correlation among the scintillometer reading (cps) and the ^{228}Ra activity concentrations determined by gamma spectrometry is shown in the Figure 2 and it can be represented by the expression:

$$^{228}\text{Ra [Bq/g]} = 0.0025 \text{ [cps]} - 0.1012,$$

with a correlation coefficient r=0.63 for n=14 and with a 95% confidence interval.

The value of 300 cps was chosen as a value representative for the limit (0.65 Bq/g of ^{228}Ra) for the initial cleanup of the site. The results of the scintillometric gamma survey of the site, converted to concentration activities, are shown in Figure 3. The map shows the contamination distribution in the area and it indicates the locations where cleanup was needed.

The gross alpha and beta counting showed a good correlation with the activity concentration of ^{228}Ra (r_{total}= 0.86, r_{beta}= 0.93). Based on these results the gross beta counting was chosen for the monitoring during clean up. Based on the original limit established for the site, an allowable limit level of 3.5 Bq/g of gross beta counting in the soil at the site was derived from Figure 4.

In deriving the ARL, secular equilibrium of the thorium decay-chain was supposed, in order to evaluate its actual state, the measured ^{228}Ra activity concentration in grab soil samples was compared to the thorium concentration determined on the basis of readings of a sodium iodine "geophysical *in situ*" gamma spectrometer (EXPLORANIUM-GR-320) performed around the soil sampling point. The results are shown in Figure 5. A correlation coefficient of r= 0.98 indicated a near equilibrium condition for the thorium series in the soil, and validated the assumptions which went into establishing the ARL, thus showing the suitability of the measurement methods adopted.

The preponderance of the thorium series radionuclides over the uranium series radionuclides was assumed for deriving the ARL of 0.65 Bq/g for ^{228}Ra, based on the initial soil collection and measured ^{228}Ra/^{226}Ra ratio. In Figure 6 the frequency distribution of the ^{228}Ra/^{226}Ra ratio measured in all grab soil samples is illustrated. In the 43 samples analyzed the ^{228}Ra/^{226}Ra concentration ratios ranged from 1 to 81, which shows that the predominant contaminants of the soils were nuclides from the thorium series. The data exhibit a log-normal distribution. The plot shows a sudden change of gradient around a cumulative frequency of 90%, indicating a bi-modal distribution of the data. One distribution has a geometric mean of 3.5, while the other has a geometric mean of 60. Besides to the presence of Cake II, which has an average value of around 60, the results pointed to the presence of Mesothorium Cake and/or to wastes of physical processing performed in the mill, which of value of average around 3.5 [5].

4. FINAL STRATEGY FOR DECONTAMINATION

In the areas where the values of the counting rate were higher than or equal to 300 cps, soil layers were removed until the counting reached values less than 300 cps. After that, samples of the local soil were taken to be analyzed by gross beta counting, in order to counter-check the monitoring of the cleanup action. In accordance with the Brazilian guidelines soils with concentrations higher than 30 Bq/g were sent to a repository site. Soils with concentrations lower than 30 Bq/g, but higher than 0,65 Bq/g of ^{228}Ra, higher than 300 cps or higher than 3.5

FIG. 3. Distribution of the contamination on the USAM site.

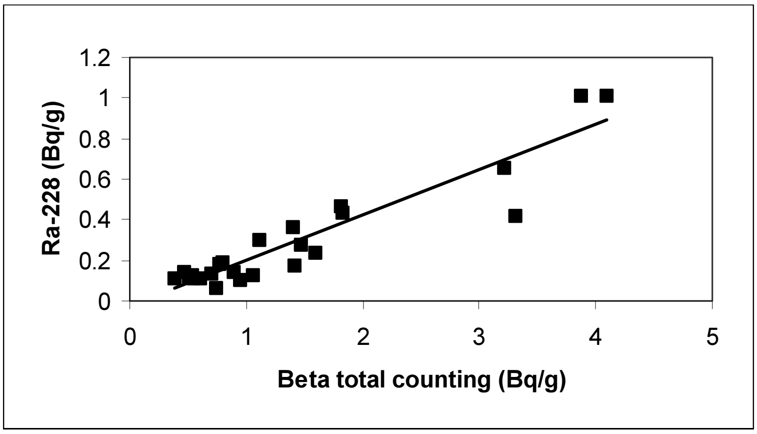

FIG. 4. Relationship between the concentration of ^{228}Ra and the gross beta counting.

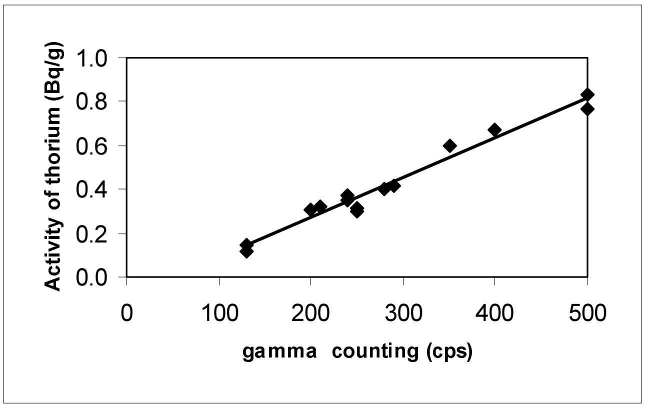

FIG. 5. Relationship between the activity of thorium by NaI and gamma counting by scintillator.

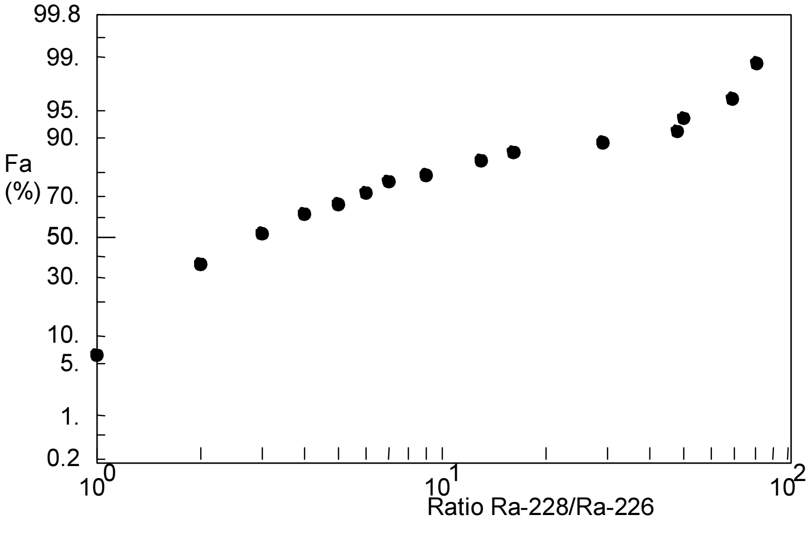

FIG. 6. Accumulated frequency of ^{228}Ra/^{226}Ra ratios in USAM soils.

Bq/g of total beta counting were sent to a municipal landfill. Soils with concentrations less than 0.65 Bq/g of ^{228}Ra, or 300 cps, or 3.5 Bq/g total beta counting respectively could remain in the site.

5. SUMMARY AND CONCLUSIONS

In December 1998 the State Company finished the soil cleanup actions and applied for the license to sell the site with a view to unrestricted use. At this time the last radiological survey was performed by IRD and soil samples were collected to be analyzed by gamma spectrometry in order to verify the compliance with the limits established. As no ^{228}Ra concentrations in the soil samples exceeding the established limit (0.65 Bq/g) were found, a license was granted by the Brazilian Nuclear Energy Commission on January 1999. About 2300 m^3 of soil were removed from the site, of which 60 m^3 was sent to a repository and 2240 m^3 to a municipal landfill. A picture of the site after the cleanup actions is given in figure 7. The proceedures established for on-site radiation measurements resulted in a cost reduction and the speed-up of the analyses. The total cost of the decommissioning of the USAM mill was estimated to be around 20% of the sale-off value of the site. Considering the location and the size of the site, as well as the public pressure involved, the dose limit of the of 1 mSv/y was considered a defendable limit.

FIG. 7. The USAM site after the cleanup action.

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CORRELATION OF GAMMA SPECTROMETER MEASUREMENTS AT SURFACE WITH CONCENTRATIONS AND DISTRIBUTIONS OF SUBSURFACE RADIUM CONTAMINATION: DEVELOPMENT, VERIFICATION AND APPLICATION OF METHODOLOGY

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ABSTRACT

This work is a step forward in the investigation of data gathering principles and analysis tools for improved estimates of subsurface radium contamination concentrations and distributions using surface gamma radiation spectra. Techniques to solve the inverse problem of estimating surface gamma radiation spectra given a fully known subsurface radium distribution have been investigated and applied with success. These techniques fell into three broad categories: empirical (using laboratory and field data), analytical (using mathematical derivations of relationships), and computer simulation (using Monte-Carlo photon transport simulation methods). Methods of analyzing surface spectra to estimate certain source parameters have been studied. The most fully developed methods are those involving the ratio of the areas of two peaks of differing energy from the same radionuclide to determine the source depth. For a point source of radium and its progeny, these techniques are able to reliably estimate the source depth from a single gamma radiation spectrum taken at the surface directly above the source. The only significant uncertainties in this case are the soil density and uncertainties introduced as a result of counting statistics. Further work remains to fully achieve the goals of the larger project: to develop a comprehensive suite of tools for the improved interpretation of surface gamma radiation spectra from subsurface distributions of radium contaminated soil.

1. INTRODUCTION

1.1. Background

Radium contaminated sites exist in many countries from the radium industries that operated during the first half of this century under a less stringent regulatory climate than exists today. In Canada, radium contamination dates back to 1933 when a radium refinery began operation in Port Hope, Ontario. The refinery operated for about 20 years, refining ores that were mined at Port Radium, some 3300 km away. During this time, wastes from the refinery were treated no differently from any other industrial wastes. Transporting the ore from the mine to the refinery included a 2200 km water transportation route through lakes rivers and portages, followed by approximately 3000 km by rail. During the operation of the refinery, waste products were used as fill materials around the town of Port Hope as the town expanded. The radium produced in Port Hope was used at several dial-painting operations for the manufacture of radioluminescent products. Many of these products, such as aircraft instruments, are still in use today and are still maintained.

The radium contamination legacy left by the Canadian radium industry is summarised in Table I.

When characterizing radium contaminated sites, a primary objective is to determine the spatial distribution and activity levels of the contaminant with a minimum expenditure. It is generally cost effective to use field gamma radiation measurements as much as possible for the characterizations, because such non-intrusive measurements can be made with little effort and relatively inexpensive equipment. The gamma radiation emissions from radium and its decay products are such that background concentrations are readily measurable with simple hand-held equipment.

The nature of radium contamination is such that interpretation of data can be complicated. The distribution of radium contamination in soil varies from site to site depending on the transport processes that have occurred. Natural transport processes tend to produce a continuous or homogeneous distribution. Human activities, such as earth moving, often result in a discontinuous, heterogeneous distribution presenting special problems for characterization and cleanup. The volume of suspect soil can be orders of magnitude larger than the volume of discrete particles and pockets of contaminated soil that are present.

In Canada, the Low-Level Radioactive Waste Management Office (LLRWMO) manages historic radioactive wastes that are a federal responsibility. The LLRWMO has developed and refined methods to characterize and verify the success of cleanups of soil with heterogeneous uranium and radium contamination. A combination of surface and sub-surface measurements are collected and statistically analyzed to determine the cleanup limits.

Waste characterization surveys conducted by the LLRWMO proceed generally as follows. First, the surface gamma radiation profile is established. The primary method used for surface characterization is the Large Area Gamma Survey System (LAGS) that has been developed by the LLRWMO with assistance contracted from SENES Consultants Limited. Gamma radiation data are collected continuously along specified lines (generally one-half metre or one metre lines) of pre-established blocks and read along with location data into a computer. Data are analyzed and the potential locations of discrete anomalies or areas of suspected distributed contamination are identified for follow-up investigation by a combination of manual detailed surveys and computer analysis. The manual survey consists of gathering surface and sub-surface spectral data and collecting and analyzing soil samples.

1.2. Purpose and scope of work

The primary purpose of this project is to improve the interpretation of surface gamma spectroscopy measurements with respect to determining the activity level and spatial distribution of subsurface radium.

This project has focused on the characterization of radium in the near surface (upper meter of soil) uranium series radionuclide contamination, particularly ^{226}Ra and its progeny. Discrete sources have been considered in detail, and distributed sources have been considered as an extension of the simpler point-source problem. Measurement techniques investigated were limited to surface gamma radiation spectrometry using sodium iodide field spectrometers.

The rationale for development of the techniques described here are examined first. A literature review was then performed to identify similar previous work. Predictive tools were developed using information obtained through the literature review and newly-developed techniques.

These tools were developed using computer simulation, mathematical modelling, and some experimental and field data.

2. RATIONALE FOR DEVELOPMENT

2.1. General context

The Low-Level Radioactive Waste Management Office (LLRWMO) is operated by Atomic Energy of Canada Limited (AECL) through a cost recovery agreement with Natural Resources Canada, the federal department which provides the funding and establishes national policy for LLRW management. Part of the mandate of the LLRWMO is to resolve historic radioactive waste problems that are a federal responsibility. Historic radioactive wastes are LLRW for which the original owner can no longer be held responsible and which are managed in a manner no longer considered acceptable. The wastes generally stem from spillage of ores and ore concentrates during transport from mine sites to refining facilities during the period when these activities were conducted by the federal government, and from contaminated materials originating from the use of radium in luminous dials.

The current owners of property with historic wastes are not licensed by the Atomic Energy Control Board (AECB) to possess these materials. Once the federal government accepts responsibility for the LLRW, the LLRWMO acts as the owner of the materials. The LLRWMO remediates properties by removing the contaminated materials to storage or disposal. Where contaminant concentrations are such that a license is required, the materials are stored in a facility operated by the LLRWMO and licensed by the AECB. Where contaminant concentrations are not of licensable activity, materials may be disposed of at a local site or held in interim storage until a permanent site is established.

Two examples of LLRWMO projects that made use of characterization techniques being refined under this coordinated research programme are given in the following section.

2.2. Fort McMurray Historic Uranium Cleanup Project

2.2.1. Project overview

Beginning in the 1930s, uranium ore and ore concentrates were shipped by barge from the Port Radium mine in Canada’s Northwest Territories along a 2200 km water transportation route to the barge-to-rail transfer point in Fort McMurray, Alberta [1]. From Fort McMurray, the ore was transported by rail to its final destination in Port Hope, Ontario, for refining. From the 1930s to the 1960s, approximately 30 hectares of riverside property in Fort McMurray was used for the unloading of barges and loading of railway waggons. Incidental spillage and tracking during the transfer of uranium bearing materials was the cause of the contamination of these properties. In the summer of 1992, during investigations of transfer points along the water transportation route, elevated levels of radioactivity were discovered on these riverside properties at Fort McMurray, Alberta [2]. The management of materials discovered at transfer points along the water transportation route are within the LLRWMO mandate.

2.2.2. Pre-remedial activities

Data from a local background study, site characterizations, and an examination of human and environmental toxicity were used to develop cleanup criteria for the project in consultation with the technical working group [3]. The contaminants of concern at the site, associated with the uranium ore, were uranium, arsenic, and radium. An arsenic criterion of 30 ppm was adopted from the recommendations of the Canadian Council of Ministers of the Environment [4]. A comparison of arsenic and uranium toxicity indicated that uranium represented a hazard less than that of arsenic, so 30 ppm was established as a conservative cleanup criterion for uranium. The most restrictive criterion for the project was the ^{226}Ra criterion of 0.1 Bq/g, the upper end of the range of background values found in Fort McMurray. The regulator approved the cleanup criteria before remedial activities took place at the sites.

The nature of contamination at the Fort McMurray sites fell into three categories [5]:

Category A: materials exceeding a uranium concentration of 500 ppm (mainly uranium ore) and therefore requiring a license from the AECB;

Category B: soil heavily populated with fragments of uranium ore with average concentrations greater than 30 ppm and less than 500 ppm uranium; and

Category C: material that does not exceed the cleanup criteria but contained occasional rocks with elevated contaminant concentrations.

2.2.3. Contaminants characteristics

Contaminants attributable to the spillage of pitchblende ore at the Fort McMurray sites have been identified as natural uranium and uranium series decay products, notably radium, and arsenic. Analysis of pitchblende ore fragments and low-grade uranium bearing rock samples, typical of those found on the Fort McMurray sites, was conducted to determine the range of uranium, radium and arsenic concentrations in these materials. Ore analysis summary information is provided in Table II.

Appropriate categorization of materials is confounded by soil samples that may miss discrete particles and surface gamma radiation readings that cannot easily distinguish between concentrated and disperse sources. To compensate for these shortcomings, efforts in materials characterization are intensive, requiring area compositing of soil samples and gridded gamma radiation surveys be conducted with a significant spatial density of measurements. While initial delineation surveys to estimate material volumes and plan and execute remedial works are intensive, the methods to verify compliance with cleanup criteria are suitable more rigorous.

2.2.4. Verification strategy

A Verification Plan was prepared for the project. The verification work was designed to measure the amounts of residual contaminants, specifically uranium, arsenic and radium, throughout the site, and to detect and react to any areas containing contaminant concentrations in excess of the cleanup criteria. Statistical considerations were incorporated into the Verification Plan so that reported results could be compared directly to project criteria. The Verification Plan contained detail of soil sampling frequency, sample compositing, averaging areas and volumes, and prescribed methods. All project verification activities were conducted on the site at the completion of remedial activities but prior to backfilling excavated areas. The regulator approved the Verification Plan prior to the commencement of site verification activities.

Verification sampling systematically sampled both property areas where remedial work was conducted as well as areas where no remedial work took place.

In areas where cleanup had taken place, individual surface soil samples were collected on a 5 m grid pattern and sets of 6 contiguous samples were composited for analysis. At the sample locations, portable gamma ray spectrometer measurements were taken. Sample analysis values exceeding the project criteria triggered additional remedial work in the sample area.

In areas of sites where no cleanup work had taken place, test pits were installed on a 20 m grid pattern and samples collected from the surface down to native, undisturbed soil. Sample results exceeding the project criteria triggered remedial work in the vicinity of the test pit. Trenches were installed in the non-remedial areas by removing 15 cm layers of material and conducting gamma radiation surveys over the exposed surface.

Over the site areas, both remedial and non-remedial gamma radiation surveys were conducted with a computer-assisted radiation survey system developed by the LLRWMO. The system collected gamma radiation readings on a 1 m line spacing and the data were used to identify the locations of discrete pieces of uranium ore which were subsequently recovered and characterized. Based on analyses and categorization of the recovered pieces of rock, an estimate of contaminant concentrations was determined over the survey area and compared to the project criteria.

As an example of the level of effort required to conduct a successful verification programme to satisfy regulators that the cleanup criteria described above were met, Table III summarizes salient points for a cleanup conducted on one property in Fort McMurray.

2.2.5. Potential applications

With regard to the spilled uranium ore cleanup work in Fort McMurray, Alberta, Canada, the interpretive spectral tools discussed in this paper would have two primary applications:

- determination of source type (i.e. disperse, discrete, distributed); and,
- categorization of soils (i.e. Category A, B or C).

The primary objective of the verification programme conducted in Fort McMurray was to insure no Category A or B materials remained on the site following cleanup activities. The level of effort required to conduct the verification programme described above was significant. The turn around time of analysis results directly impacted the project schedule and budget. Where areas were found to exceed project criteria, additional excavation work was conducted and the programme re-applied to the area.

The consequence of classifying material into a category higher than appropriate, i.e. Category B as Category A or Category C as Category B, is the increased expense for disposal and the wasted effort associated with excavation and verification activities. Mistakenly classifying material into a lower category would result in materials exceeding project criteria remaining on the site.

The application of interpretive tools for spectral analysis would facilitate better and more rapid categorical classification of *in situ* materials. Simple, traditional spectral analysis of gamma ray spectra collected in the field were not useful in determining compliance with cleanup criteria. Mean values based on *in situ* spectroscopy did not correlate well with the corresponding composite soil sample analysis results. As a result, *in situ* data could not be relied on to make immediate determinations of criteria compliance.

The ultimate application of a fully developed set of interpretive spectral tools would see the need for soil sample analysis significantly reduced with soil samples analyzed only for quality control purposes. A reliance on *in situ* measurements would eliminate the turn around time required for analysis results and so limit the impact on project budget and schedule.

Improved categorization of soil materials and identification of source type would reduce costs due to over-excavation and conservative errors used to determine category.

2.3. Malvern remedial project

2.3.1. Project overview

Historic low-level radioactive waste on residential properties in the City of Scarborough, Ontario, Canada, was discovered in 1980. This contamination resulted from a small radium incineration and processing operation on a farm in this area during the 1940s [6]. Development of the farm into a residential area during the early 1970s resulted in the spread of the radium-contaminated materials throughout the development. During the 1980s, proposals to remediate the contaminated properties by removal and relocation of the radioactively contaminated soils were deferred because of unavailability of a storage site.

Detailed characterization studies were conducted in 1992 through 1994. Cleanup of the sites began in the spring of 1995. Contaminated soils were excavated from the subject properties and segregated with the use of a soil sorting plant. LLRW was segregated, containerized and shipped to a licensed storage facility; mildly contaminated material was stored in an interim storage mound, and clean excavated soil was used as cover material for the interim storage mound.

The contamination existed in two forms, as bulk volumes of contaminated soils and, as discrete, radium contaminated “artifacts” found in otherwise clean soil.

2.3.2. Pre-remedial activities

Criteria for the project were developed [7]. The cleanup action level for bulk soil contamination corresponded to the 98th percentile of naturally occurring radium concentrations in the metropolitan Toronto area, 0.073 Bq/g [8]. Samples collected were to represent areas no greater than 10 m^2 and 0.50 m in depth. For discrete particles, a risk-based assessment was performed using the most conservative parameters. The goal was to identify and remove any discrete artifact with an activity greater than 0.15 MBq (4 μCi). As the contaminated artifacts were known to exist primarily in the topsoil layer, detection instruments were required to detect a source of this activity buried beneath 10 cm of soil.

A computer-assisted, large area gamma radiation survey system was developed by the LLRWMO to perform surveys of properties with data collected electronically with an on-board computer [9]. Radiation measurements were collected on a frequency of 4 readings per square meter. The intense survey coverage resulted in a greater than 90% probability of detecting a 0.15 MBq source buried at 10 cm.

Prior to the cleanup project and development of the soil sorting conveyor system, a pilot project was conducted to test the technical realities of the development of a sorting system. The primary goal of the sorting system was to:

- detect and segregate bulk soil volumes with a specific activity greater than 3.7 Bq/g;
- detect and segregate radium contaminated artifacts with an activity greater than 0.15 MBq; and,
- to determine the average activity of batches of soil for calculation of the radioactive inventory [10].

A detailed investigation of all residences in the effected community was conducted. Gamma radiation surveys were conducted on over 450 residential properties using the computer-assisted radiation survey technology developed by the LLRWMO. The survey programme detected areas of bulk radium contamination on 25 properties not identified during previous survey campaigns, bringing the total number of properties requiring cleanup to 75.

The level of survey detail during initial investigative work is summarized in Table IV.

2.3.3. Verification strategy

A Verification Plan [11] for the work was prepared. The work described included systematic soil sampling, and spectral measurements within the excavation area were designed so that composite samples would represent the area a house may be built on, while the individual measurements could be compared to the recommended averaging area of 10 m^2. The verification programme was also designed to detect contaminated materials in areas on the property outside the excavation area, in the event that they had been overlooked during delineation surveys.

On completion of the removal of contaminated materials from each property, a series of measurements were made within the excavation area and, outside the excavation area in the undisturbed soils. All areas of the property were surveyed with the computer-assisted survey system, and where radiometric anomalies were identified in the survey data, a series a measurements were collected with hand held survey meters. Gamma radiation measurements greater than the upper limit of normal for the property were targeted for further remediation, as they may be indicative of bulk contamination or discrete artifacts.

Within the excavation area, soil samples were collected on a 3 m grid spacing and composites of 16 contiguous samples were prepared, analyzed by gamma spectroscopy, and compared to the project criterion for ^{226}Ra. At each sample location, surface spectral data were collected *in situ* with a portable gamma-ray spectrometer. Values exceeding the project criterion for ^{226}Ra measured either *in situ* or by laboratory analyses triggered additional characterization and further remedial work.

Outside the excavation area, a series of boreholes were installed around the excavation perimeter and spectral data gathered *in situ* on 10 cm depth increments with a borehole probe. Where values exceeded the project criterion for ^{226}Ra, excavations were expanded during additional remedial work.

The application of the verification programme on a cleanup property typically required a level of detail summarized Table V.

2.3.4. Potential applications

It was the Malvern Remedial Project that instigated the LLRWMO involvement in this programme of research. Intensive, intrusive and expensive characterization work on 450 residential properties involved the collection of thousands of soil samples and rigorous gamma radiation surveys to identify areas of remedial work, remove hundreds of discrete radium contaminated artifacts, and deem properties to meet cleanup criteria.

The first level of survey, an automated gamma radiation survey conducted on a 0.5 m line spacing, was used to identify anomalous readings for further investigation. Contaminated materials took generally two forms, either as discrete pieces of radium contaminated plastic tubing or as areas of discontinuous, heterogeneous soil contamination. The identification, delineation and recovery of these materials was confounded by the mix of contaminated particles with clean soils and the thin layer of overburden material that may cover the contamination. To discern the source of the anomalous radiation reading, several simple *in situ* spectral measurements were taken to estimate mean activities for areas, and several soil samples were taken and analyzed to compare to cleanup criteria. Comparison of *in situ* and soil sample data indicated that *in situ* data were not reliable enough to determine compliance with cleanup criteria. Recovery of discrete particles was time consuming, as exact locations were difficult to determine, and an effort was made to minimize damage done to the lawns of property owners.

The post-cleanup verification programme described above was problematic because of its reliance on soil sample analyses. The project conducted required relocation of homeowners while excavation work was conducted, so delays due to sample turn around time and re-cleanups of failing areas had significant impacts on project schedule and budget.

Interpretive spectral tools would have found their usefulness in identifying the type of source (i.e. discrete particles, disperse contaminated soils, heterogeneous distributed materials) and reliably quantifying activities for immediate comparison to cleanup criteria.

2.3.5. Project summary

At project completion, all 75 properties met the cleanup criterion for ^{226}Ra established for the project. Approximately 33 m^3 of soil and several hundred radium-contaminated artifacts were removed as LLRW to a licensed storage facility [12]. Material volumes of 9077 m^3 exceeding the project cleanup criterion for radium, but not considered licensable, were stored in an interim storage mound, to await future disposal. Materials excavated but that were found not to be contaminated were used as top cover for the interim storage mound.

2.4. Highfields farm remediation

2.4.1. Site description

The Highfields Farm is located northwest of Toronto, Ontario, Canada. The majority of the property has been used as farmland for either animal grazing or crop production. Many of the fields have now overgrown by natural processes or by the planting of coniferous wood lots. The only area of active farming, other than for grazing purposes, is the field, which includes an area north of the barn near the southeast farmhouse residence.

A portion of the Highfields Farm property was used as a radium recovery operation similar to the one described earlier for the Malvern Remedial Project. A limited cleanup was completed in the early 1980's. Activities involved the use of radium for industrial and medicinal purposes. Some activities involving the use of radium were carried out on the property preceding the regulations governing the possession, use and disposal of radium. This led to the spread of radium contaminated materials at the property.

2.4.2. Summary of survey results

Gamma radiation surveys were conducted of the approximate 100 hectare Highfields Farm property. A few individual data points were found to exceed 0.60 μSv/h (100 μR/h), measured within 15 cm of the ground. The areas of greatest effect were approximately 2-4 m^2.

2.4.3. Cleanup objectives

The objective of the cleanup was to remove radium-contaminated soils with concentrations greater than 3.7 Bq/g radium. Possession of such materials, considered low-level radioactive waste (LLRW), requires a license issued by the Atomic Energy Control Board (AECB). To achieve this objective, areas where the gamma radiation exposure rate exceeded 0.60 μSv/h (100 μR/h) measured at a height of 0.15 m above the ground were remediated. On completion of the remedial work, gamma exposure measurements were not to exceed 0.60 μSv/h (100 μR/h), measured at a height of 1 m from the excavation surface.

2.4.4. Remedial work

Excavation of contaminated soils was conducted in April 1998. Materials were excavated with a mini-excavator, placing soil directly into steel drums. When each drum was full, its lid was secured and, once removed from the work area, characterization measurements were taken on the drum to estimate radium content.

3. DEVELOPMENT OF PREDICTIVE TOOLS

3.1. Overview

The overall objective of this project is to be able to provide tools that would allow the user to estimate subsurface distributions of radium contamination based on spectra collected at the surface. The general approach taken was to first deal with the inverse problem of predicting surface spectra or important features of surface spectra based on a known subsurface distribution of radium contamination.

Three different approaches to predicting spectra based on subsurface radium contamination distributions were taken. The first approach discussed below is a review of empirical data, collected both under "laboratory" conditions and within the context of field projects. The second approach is to consider analytical (mathematical) methods of predicting the behaviour of particular spectral features. The third is to make use of computer simulations to generate surface spectra based on a modeled subsurface radium contamination distribution.

The primary advantage of the empirical approach is that it makes use of actual field data. The downside is that the information on subsurface radium distributions, although quite extensive, will still be somewhat imprecise. As well, the data will be limited to those situations that have been encountered in LLRWMO fieldwork and thoroughly studied, or those situations that can be fairly easily constructed in practice. The empirical methods and results are discussed more fully in Section 5.2.

The analytical approach has several advantages. The results (and uncertainty limits) will be extremely precise, and the dependence on all of the relevant variables will be well known. Also, the path from problem to solution will be very explicit. However, there are also several disadvantages. Only the simpler relationships will be considered due to theoretical and practical limitations. Required assumptions and simplifications will restrict the application of this method and may introduce some uncertainty into the results. The analytical methods and results are discussed more fully in Section 5.3.

Computer simulation of surface gamma spectra was accomplished using Monte Carlo photon transport simulation. The strengths of this method are its precision and flexibility. Source distributions can be defined exactly, and in whatever configuration is desired. The primary weakness of this method are that there may be some deviation from real-world spectra due to the limitations of the simulation model, and that a large amount of computing effort is required to produce useful results. The simulation methods and results are discussed more fully in Section 5.4.

This reason for using this multiple method approach is that it should be very robust against methodological errors since the results of each approach will provide cross-checks on the results of the others. As well, each of the three approaches brings a different set of strengths (and weaknesses). Table VI shows a brief summary comparing the advantages and disadvantages inherent in each of these approaches.

3.2. Empirical data

3.2.1. Laboratory data

Two sets of laboratory data were gathered. The earliest laboratory investigations made use of a large graduated cylinder filled with dry sand to various depths and with a small radium source placed on top of the sand. This cylinder was then placed directly on top of a HPGe gamma spectrometer and spectra were gathered. The empirical data referred to in Fig. 1 were from this experimental arrangement.

The most significant laboratory data gathered for this project made use of a small radium source buried in a well-controlled volume of soil. Topsoil was placed and compacted into an approximately 1.0 m x 1.2 m x 0.8 m deep steel container. The density (2.06 g cm^{-3}) and moisture content (12.6%) of this soil were determined. A small radium source was buried at various depths below the center of the surface of this “soil cube”. Figure 2 shows spectra measured by an Exploranium GR-256 3”×3” NaI(Tl) field gamma spectrometer due to a small radium source buried at depths of 0, 5, 10, 15, 20, and 30 cm.

The areas of the 609 keV and 2204 keV peaks of ^{214}Bi were calculated for each of the spectra from 0 to 20 cm. Figure 3 shows these peak areas as points fitted to an exponential curve. Also shown in this figure is the relationship between the ratio of the peak areas to the source depth, along with a fitted exponential curve. The curve is a very good fit, with an R^2 value of 0.996. Under the conditions of this experiment, the ratio decreases by a factor of two for approximately every 10 cm of increased source depth. The change in peak ratio with source

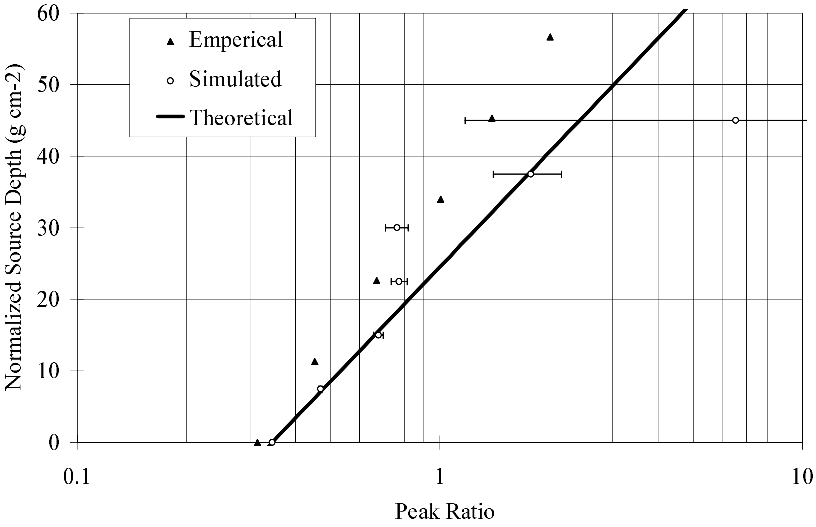


FIG. 1. Normalized Source Depth vs. 1765/609 keV Peak Ratio

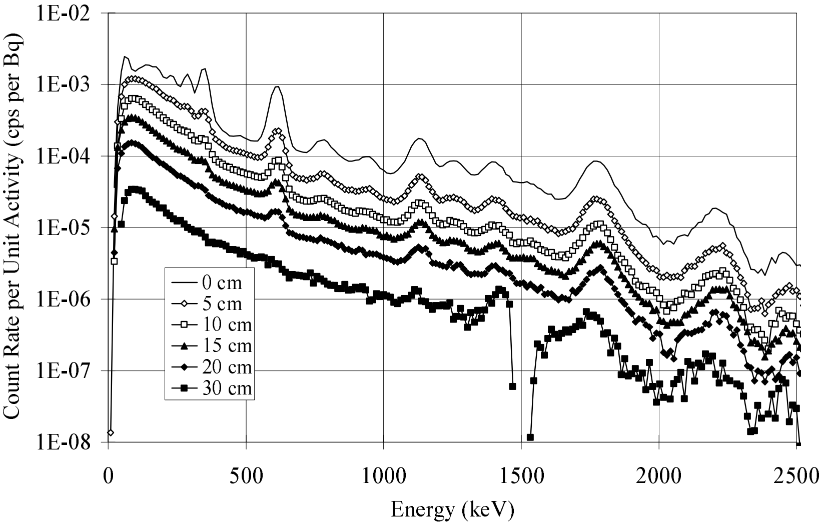


FIG. 2. Normalized Energy Spectra by Soil Depth for a Point ^{226}Ra Source

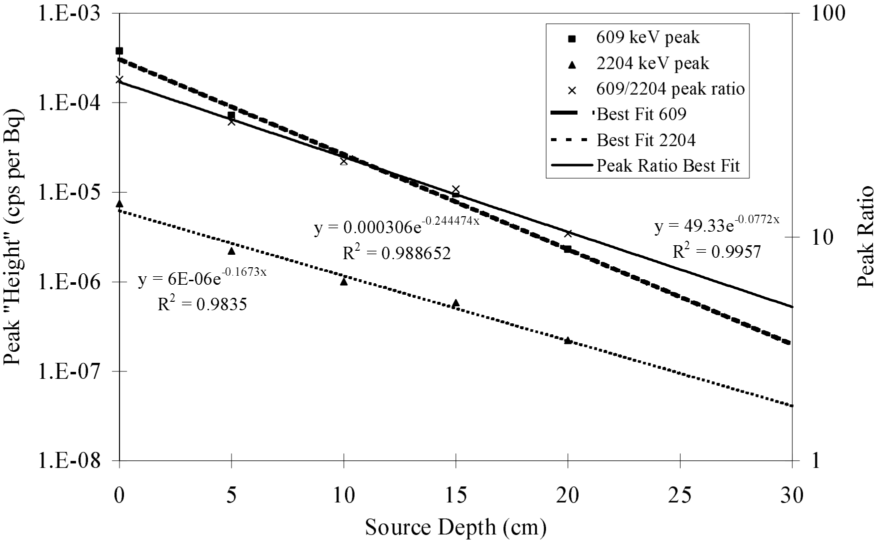


FIG. 3. Peak Heights and Height Ratios by Depth (609 and 2204 keV Peaks)

depth is sufficiently large that, given reasonable counting statistics, the uncertainty in the depth of the discrete source as estimated using this peak ratio would be acceptable.

3.2.2. Remedial project data

3.2.2.1. Malvern Remedial Project

During the Malvern Remedial Project, which pre-dated the present experimental work, surface gamma spectroscopy measurements were taken in both background and contaminated areas. However, very little effort went into determining the actual distributions of the subsurface radium contamination in detail. Nevertheless, some simple observations were made.

Since the contamination was found primarily within the top 15 cm of the soil, it was expected that the ratio of 2.1 MeV to 1.1 MeV photons would be slightly lower than for infinitely uniform contamination. The ratio between 2.1 and 1.1 MeV peaks was highly variable but, overall, the ratio was statistically significantly lower in the contaminated properties than in the background properties, consistent with expectations.

Similarly, it was expected that the ratio of 1.7 MeV to 1.1 MeV photons would also be slightly lower than for uniform conditions. However, contrary to expectations, the ratio between 1.7 and 1.1 MeV peaks was, overall, significantly higher in the contaminated properties.

There are several problems with this data set that might explain why these observations are somewhat contradictory. Although the instrument type was the same for all measurements, the actual instrument used to take measurements differed for the contaminated and background properties. The soil type differed from property to property, but was probably more consistent for the contaminated properties than the background properties due to their relative physical proximity to one another. Overall, the analyses of these data are inconclusive. However, it is possible that further analysis might produce useful results.

3.2.2.2. Highfields Farm remediation

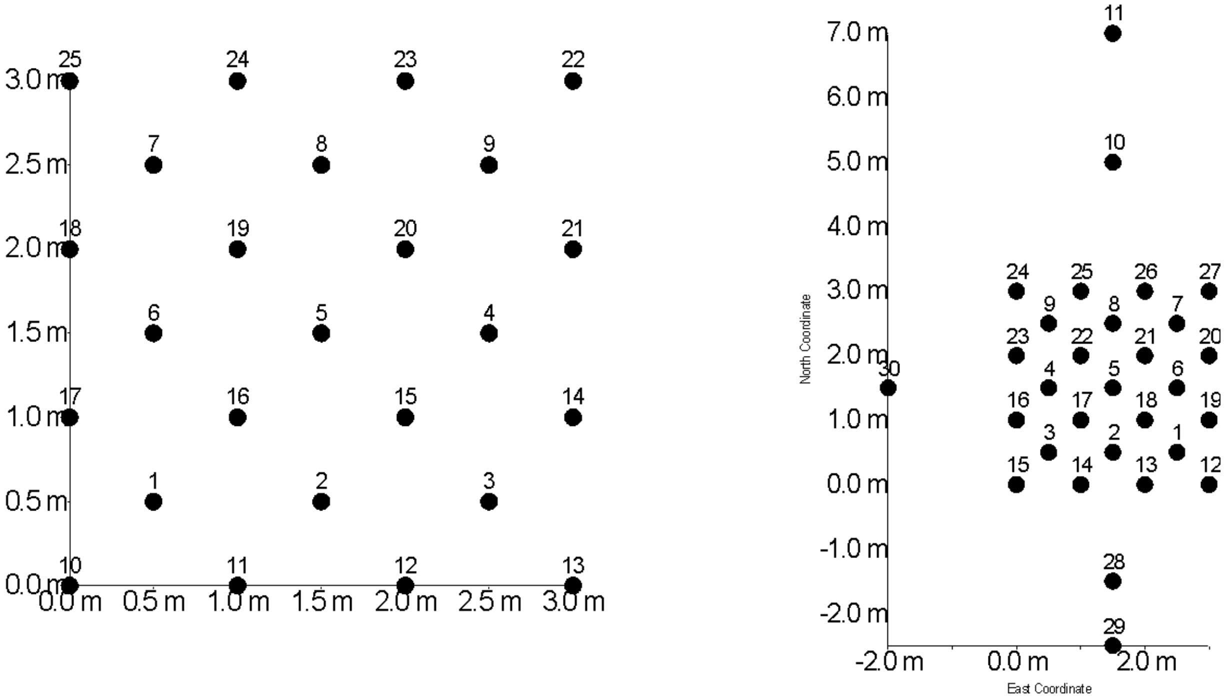
The excavation of radium contaminated soils was conducted at two small areas at the site, labeled excavation area X and T. Prior to excavation a series of gamma ray spectra were collected with the portable GR-256 spectrometer. The detector was placed on contact with the ground surface at grid locations established over the excavation area. Once all surface spectra were taken, soil samples were collected at the spectral measurement points on 10 cm increments to a total depth of 50 cm. Soil samples were also collected at several other grid locations over the excavation area. Soil samples were analyzed in the PHFSO laboratory by HPGe gamma spectroscopy to determine ^{226}Ra activity (based on ^{214}Bi activity).

3.2.2.3. Area T specifics

A total of 14 surface spectral were collected in this excavation area (see Fig. 4a). Spectra were collected at locations 1 through 9 with the Cs-137 energy stabilization source in use and at locations 2,4,5,6, and 8 without the use of Cs-137. Soil samples were collected at locations 1 through 25 on 10 cm increments to a total depth of 50 cm at each location.

3.2.2.4. *Area X specifics*

A total of 26 surface spectral were collected in this excavation area, prior to excavation (see Fig. 4b). Spectra were collected at locations 1 through 9 with the ^{137}Cs energy stabilization source in use and at locations 2, 4, 5, 6, and 8 without the use of ^{137}Cs. At some locations, multiple spectra were collected. Additionally, surface spectra were collected at locations 10, 11, 28, 29, and 30, outside of the excavation area. Surface readings were retaken at locations 10 and 11 after the excavation was complete. Soil samples were collected at locations 1 through 25 on 10 cm increments to a total depth of 50 cm at each location.



(a) (b)

FIG. 4. Area T Data Locations (a), and Area X Data Locations (b)

3.2.2.5. *Data analysis*

Figures 5a and 5b show the median concentration by depth with distance (m) from the center of contamination for each of the two areas. Figure 5b, which is for area X, shows that ^{226}Ra concentrations at the center of the contaminated area near the surface (0-10 and 10-20 cm horizons) were about 7.4 Bq/g while concentrations below 20 cm were about 25 times higher or about 185 Bq/g. Away from the center of contamination, the concentration profile is substantially different: the higher concentrations are present at the surface (above 20 cm) and the concentrations are lower below the surface. The profiles at area T shown in Fig. 5a are similar although somewhat less pronounced.

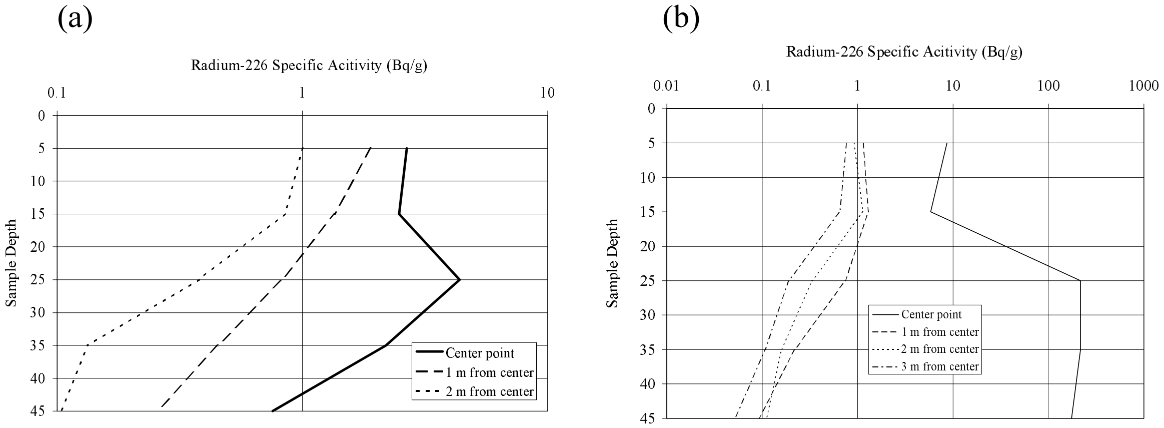


FIG. 5. Radium-226 Concentration Depth Profiles at Various Distances from the Centre of the Contaminated Area: (a) Area T, (b) Area X.

The near-surface concentrations decrease with distance from the center of contamination and the sub-surface concentrations show a decreasing gradient with depth that is most likely due to vertical transport through a combination of leaching, soil cracking and vegetation processes. The negative exponential model has been applied often to describe this pattern for surface contamination. Typical background for this area is about 0.037 Bq/g ^{226}Ra and these levels are approached at depth for location 3 m from the center of contamination.

The contamination was located in an agricultural field that suggests the following plausible interpretation of the contamination pattern. A trench (or hole) was excavated and filled with contaminated material. Agricultural tillage of the field spread the contaminated soil located within the tillage zone (about 20 cm) horizontally from where the contaminated material was placed. Concentrations decrease with distance largely due to this dispersion process. A secondary dispersion process occurred in the vertical direction as ^{226}Ra spread out from the center was transported downward through a combination of physical and chemical processes.

This area was interesting since it contained elevated ^{226}Ra levels located under 20 cm of contaminated material. It is of interest to see if this material impacts the surface spectra from *in situ* gamma spectroscopy.

3.2.3. Discussion of Peak-to-Peak Ratio Theory

Consider a point source of ^{226}Ra buried in soil. It is possible to take a spectral measurement directly above the point source by locating the point of maximum gamma radiation rate on contact with the surface, and taking the measurement at that location.

It is possible to determine the depth of the source given this single spectrum even if the source activity is unknown. Consider two gamma energies E_1 and E_2 from the decay of a single radionuclide. Each of these gamma energies has particular gamma ray intensities, say P_1 and P_2. Now, let $\mu_0(E,S)$ be the mean free path for photons of energy E in material with composition S of density $\rho = 1.0$ g cm^{-3}. For any other density, the mean free path $\mu(E,S) = \mu_0(E,S) / \rho$. The attenuation factor, λ, is given by:

$$\lambda = \frac{1}{\mu} = \frac{\rho}{\mu_0(E,S))} \qquad (1)$$

Assume the point source is buried a distance d under the surface of the ground. The flux, ϕ, of full energy photons at a height h from the ground surface (total distance from the source is $r = d + h$) will be proportional to the radionuclide activity, A, gamma ray intensity, P, inversely proportional to the square of the total distance (r), and proportional to $e^{-\lambda d}$, ie:

$$\phi_i \propto A_i \times P_i \times \frac{1}{r^2} \times e^{-\lambda d} = \frac{P_i e^{\frac{-d\rho}{\mu\,(E,S))}}}{(d+h)^2} \qquad (2)$$

Now consider the flux ratio ϕ_1/ϕ_2, where ϕ_i is the flux of photons of a given gamma energy from a point source. When $A_1 = A_2$, such as when the radionuclide is the same in both cases, or when the radionuclides producing these particular gamma radiations are in equilibrium, this ratio can be given as:

$$\frac{\phi_1}{\phi_2} = \frac{P_1}{P_2} \times e^{-d\rho\frac{\mu_1-\mu_2}{\mu_1\times\mu_2}} \qquad (3)$$

where $\mu_i = \mu(E_i,S)$. Rearranging this equation yields an expression for the normalized source depth (depth times scatterer density):

$$d\rho = \frac{\mu_1\times\mu_2}{\mu_1-\mu_2}\ln\left(\frac{\phi_1}{\phi_2}\right) + \frac{\mu_1\times\mu_2}{\mu_1-\mu_2}\ln\left(\frac{P_1}{P_2}\right) \qquad (4)$$

which is a simple logarithmic equation of the form $y = A \ln(x) + B$. It is interesting to note that the expression for $d\rho$ does not depend on h, the height of the detector above the ground. The only variable (once a pair of energy lines is chosen) is the flux ratio. However, to obtain the absolute depth, d, one must know the soil density ρ.

Figure 1 shows how this analytical solution can be applied to predict the ratio of peak areas depending on the normalized source depth. Peak energies of 0.609 MeV and 1.765 MeV, corresponding to two of the most significant ^{214}Bi peaks, were selected. Mean free paths for these two energies were chosen that correspond to scattering through dry sand. Note that it is normalized source depth, and not absolute source depth, which is plotted on the vertical axis. If the soil density were known, then it would simply be a matter of dividing the normalized depth by the density to determine the absolute source depth.

For this particular application, the ratio of 0.609 MeV and 1.765 MeV peak areas seems to be a potentially useful indicator of point source depth. The logarithm of the ratio varies linearly with depth (as seen in Fig. 1), and the change is significant over the depth range of interest. At the surface, the ratio is approximately 0.34, while at a depth of only 10 cm (a normalized depth of 15 g cm^{-2} assuming a soil density of 1.5 g cm^{-3}) the ratio has approximately doubled to 0.66. The ratio continues to approximately double for every additional 10 cm increase in depth.

3.3. Photon transport simulation

3.3.1. General approach

The general approach taken was to divide the problem of simulating surface spectra from subsurface radium contamination distributions into a number of smaller problems. One simplification was to look only at point sources, recognizing that a more general source distribution could be constructed by integrating the point source results over space, or similarly by modeling a more general distribution by a number of point sources. A second simplification was to model a small number of discrete energies rather than the entire spectrum of interest for each point source with the intent to then to generate interpolated results to derive a spectrum from a monoenergetic source of any energy. Adding several monoenergetic source spectra together would then generate a full spectrum. A third simplification was to disregard energy resolution effects during the Monte Carlo simulations. It was decided that it would be simpler to include these effects at the end of the process, once the absorbed energy spectra had been produced.

This general approach is summarized in Fig. 6. On the left in the boxes are each of the "products", and on the right are the processes performed to produce the products.

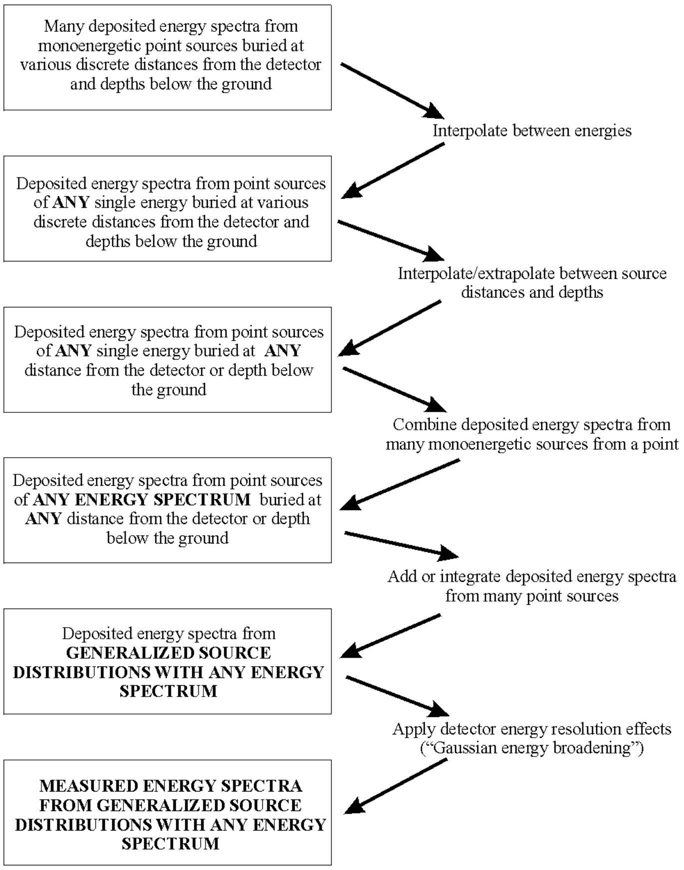


FIG. 6. General Approach to Photon Transport Simulation

3.3.2. The Monte Carlo method of photon transport simulation

Monte Carlo photon transport simulations were performed to determine detector response to mono-energetic point sources. The conditions of the simulation can be completely understood and controlled exactly, which is very difficult if not impossible using empirical methods. For example, the moisture content of the soil can be specified exactly, and will not vary throughout the soil volume or with time. As well, the photon transport simulation methods can be applied to situations that might be difficult to reproduce experimentally. For instance, it is a very simple matter to adjust the simulated soil composition to determine the effects of an increased proportion of heavier elements. The amount can be adjusted exactly and to any level.

Monte Carlo photon transport simulations also have some disadvantages over experimental techniques. The results obtained may not be exactly representative of the real world, potentially due both to simplifications in the computer model, and to errors in the input to or algorithms within the simulation. Discrepancies due to implementation errors are generally easy to detect because the results are often wildly incorrect. However, discrepancies due to computer model simplifications may be subtle and are therefore more difficult to detect. In order to guard against both sources of error, the results of the simulation are compared to experimental data for validation.

Monte Carlo photon transport simulation methods [13] [14] were used to develop detection efficiencies based on a given source spectrum and detection geometry. These methods trace the paths of individual photons from the source to the detector, simulating the physical interactions that take place along the way.

3.3.2.1. Problem geometry and physical characteristics

The basic geometry of the problem reproduces the field conditions. The problem space is defined within a column of radius 500 cm and length 1000 cm, beyond which is "void". A plane divides the upper and lower halves of the problem space into "air" and "soil" regions. The detector, modeled after the Exploranium GR-256, is a sodium iodide crystal shaped into a cylinder of length 7.62 cm and radius 3.81 cm. It is surrounded by a 1.5 cm thickness of foam rubber, all contained within a 0.2 cm thick aluminum housing. The center of the crystal is 15 cm from the surface of the soil, and the crystal is oriented such that its axis is perpendicular to the normal to the soil surface. Fig. 7 shows the basic geometry of the problem.

The dry fraction of the soil was assumed to consist of the ten most abundant elements in the earth's crust [15]. A moisture content of 12.6% by mass was assumed to be made up of pure water. This moisture content corresponds to that of the soil used to collect the experimental data. Table VII shows the simulated composition of the soil. Two densities were simulated. First, a density of 2.06 g cm^{-3} was chosen to match that measured for the soil used in the laboratory test. A density of 1.6 g cm^{-3} was also used to represent more typical in-situ soil.

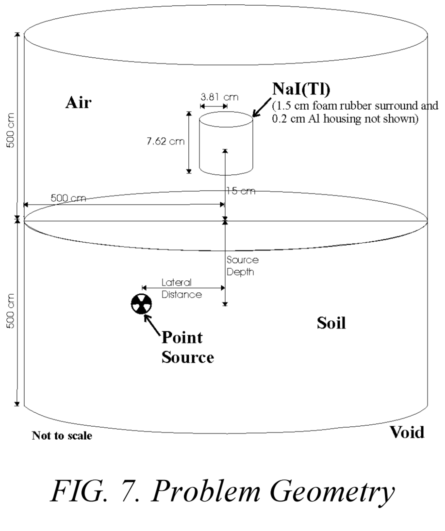


FIG. 7. Problem Geometry

It is important to note that soil composition can have a significant effect on the results. Although outside the scope of this work, Federal Guidance Report 12 examined the effects of different selections of soil compositions on the air kerma from sources in the soil [16]. At energies above a few hundred keV, the differences are relatively small. However, at energies between 10 and 100 keV, the difference can be as large as a factor of five or six. The "earth's crust" model examined in Federal Guidance Report 12 was similar to the composition used in this study, and was about half way between the two extremes in air kerma ratios for varying soil compositions.

The air was modeled as having a density of 0.00122 g/cm^3, and being composed of 76% nitrogen and 24% oxygen by mass. This is a simplified version of the "US Standard Atmosphere" taken at sea level [15].

The composition of the thallium activated sodium iodide (NaI(Tl)) crystal was taken to be 84.4% iodine, 15.3% sodium, and 0.3% (10^{-3} mole fraction) thallium by mass, with a density of 3.67 g cm^{-3}, as described in [17].

The 1.5 cm thick layer of foam rubber padding was assumed to be composed of long polymer chains which would mean that there would be approximately two hydrogen atoms for every carbon atom. The result is 86% by mass of carbon, and 14% by mass of hydrogen.

The 0.2 cm thick detector housing was assumed to be composed completely of aluminum with a density of 2.7 g cm^{-3}.

Point sources were simulated buried at various depths in the soil and at various lateral distances between the center of the detector and the point source.

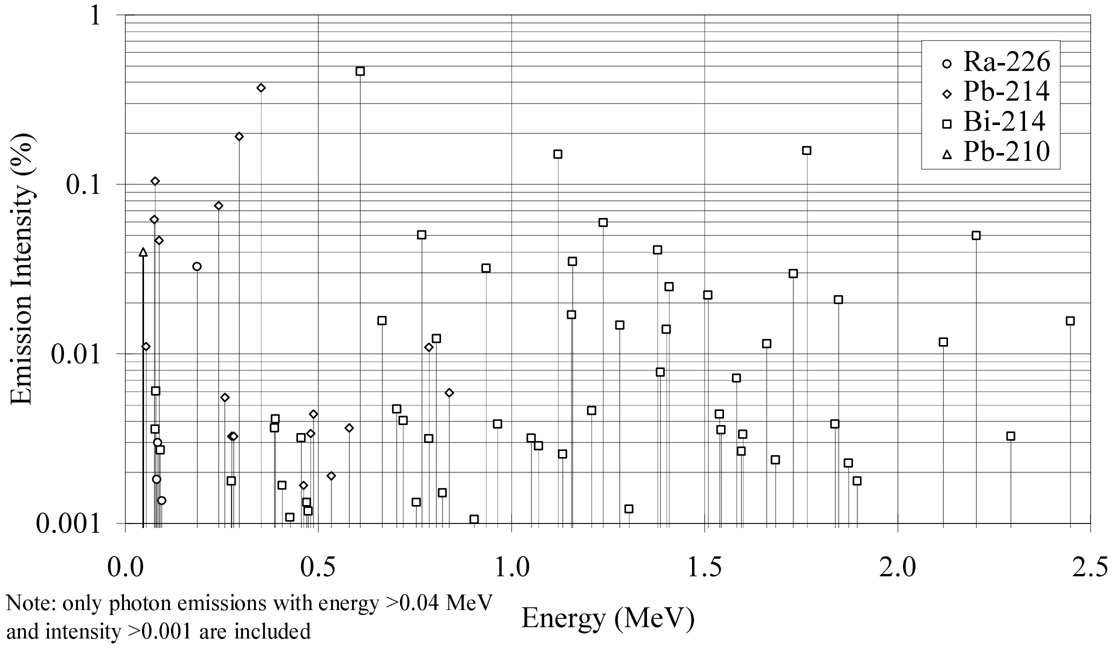
3.3.2.2. Radionuclide data

The radionuclides of primary interest are ^{226}Ra and its progeny. These are shown in Table VIII. Other naturally occurring radionuclides are also of interest, since they will influence the gamma radiation spectra collected in the field. The remainder of the uranium series radionuclides (of which ^{226}Ra is the final part), the actinium series radionuclides, the thorium series radionuclides, and potassium-40 are also primordial radionuclides which are naturally present in soil in sufficient quantities as to be easily measured by gamma spectroscopy. Caesium-137, a fission product, is another gamma radiation emitter that is occasionally found in surface soils. However, unless the specific activity of ^{137}Cs in soil is particularly high as result of a specific event (such as the Chernobyl incident), it is generally not a concern when interpreting environmental gamma radiation spectra.

Of the radium series radionuclides in Table VIII only four emit gamma radiation of any significance for the purposes of this study: ^{226}Ra, ^{214}Pb, ^{214}Bi, and ^{210}Pb. All other gamma radiations have a gamma ray intensity of less than 0.1%. Table IX shows the significant gamma decay energies and intensities for the three radionuclides of interest in energy ascending order. Gamma rays having an intensity of less than 0.1% or energies less than 0.05 MeV are omitted. These are also shown graphically in Fig.8.

An important feature of the ^{226}Ra decay series is the presence of ^{222}Rn. This radionuclide appears in the form of a noble gas with a half-life of 3.8 days, and can therefore be transported significant distances from its original site. This phenomenon introduces a major confounding factor, since the significant gamma radiation emitters in the ^{226}Ra decay series (other than

^{226}Ra itself, which contributes only about 2% of the gamma photons) are all radioactive progeny of ^{222}Rn.



Note: only photon emissions with energy >0.04 MeV and intensity >0.001 are included

FIG. 8. Radium Plus Progeny Emission Spectrum

For localized sources, it is often the case that the emanated ^{222}Rn diffuses far enough so that its progeny are diluted to a point where their contribution is essentially indistinguishable from background. In this case, it is convenient to define an equivalent radium (eRa) activity. A source with a given eRa activity is such that it produces the same photon flux as a source of the same activity of ^{226}Ra in equilibrium with its progeny. If the radon emanation coefficient (the fraction of radon produced which leaves the source) is ξ, and all emanated radon is assumed lost, then the eRa activity, A_{eRa}, can be expressed as $A_{eRa} = A_{Ra}(1- \xi)$, where A_{Ra} is the ^{226}Ra activity.

Another useful concept is the equilibrium ratio which refers to the ratio of A_{eRa} / A_{Ra}. Since ^{214}Pb and ^{214}Bi are essentially always in equilibrium with each other, the equilibrium ratio is also equivalent to the activity ratios ^{214}Pb / ^{226}Ra or ^{214}Bi / ^{226}Ra.

In general the equilibrium ratio is less than one (ie. some radon escapes and is lost). However, equilibrium ratios greater than one are possible, if a nearby source of radon is contributing significantly to the radon progeny concentration at the location of the source under consideration. The simplifying assumption of complete equilibrium will be used throughout this study, with the understanding that the results can be applied to non-equilibrium cases as well by interpreting the results in terms of ^{226}eRa activity instead of ^{226}Ra activity.

3.3.2.3. Simulation of monoenergetic point sources

The photon transport simulation was performed using a commercially available code developed at Los Alamos National Laboratory called MCNP (Monte Carlo N Particle), version 4B [18]. MCNP is a general-purpose code that can be used for the Monte Carlo simulation of neutron, photon and/or electron transport. For photon transport, the code takes into account coherent (Thompson) scattering; incoherent (Compton) scattering; photoelectric absorption with the possibility of zero, one or two fluorescent emissions; and pair production absorption with local emission of annihilation radiation. Electron transport is not simulated in the model used: all energy transferred to electrons is assumed to be deposited locally, although bremsstrahlung photons are generated using a thick-target Bremsstrahlung model.

The first general-purpose Monte Carlo code developed at Los Alamos was written in 1963. Several other similar codes were developed there over the next 15 years. In 1977, after having merged several of these codes together, the first version of MCNP appeared. However, it was not until 1983 that MCNP was first distributed internationally. The current version of MCNP (version 4B) has about 3000 active users at about 200 installations worldwide. The source code is written to comply with the ANSI FORTRAN 77 standard, and compiled to run on many different platforms. The installation used for this project is licensed to Atomic Energy of Canada Limited (AECL) Chalk River Laboratories, and runs under the UNIX operating system. The simulations were run on a Silicon Graphics workstation located at Chalk River Laboratories.

Monoenergetic point sources of five discrete energies (0.5, 1.0, 1.5, 2.0 and 2.5 MeV) were simulated at various lateral distances from the detector center ranging from 0 to 200 cm, and buried at various depths from 0 to 100 cm. Fig. 9 shows all source locations simulated.

MCNP is controlled primarily through the use of input files, which specify most user-supplied problem parameters. Although hundreds of different input files were required to develop the detection efficiencies in this report, they all followed the same general form. The result of the Monte Carlo photon transport simulations was a large number of deposited energy spectra from monoenergetic point sources at various discrete distances from the detector and buried at discrete depths in the soil. Spectra that included energy resolution effects were also generated, but not used further in this work.

Figure 10 shows an example of spectra from a monoenergetic (2.5 MeV) point source buried 5 cm in soil at a lateral distance of 5 cm from the center of the detector. For comparison, both the deposited energy spectrum and the spectrum that takes into account detector resolution effects are shown. Of particular note is the full energy peak at 2.5 MeV, the single and double

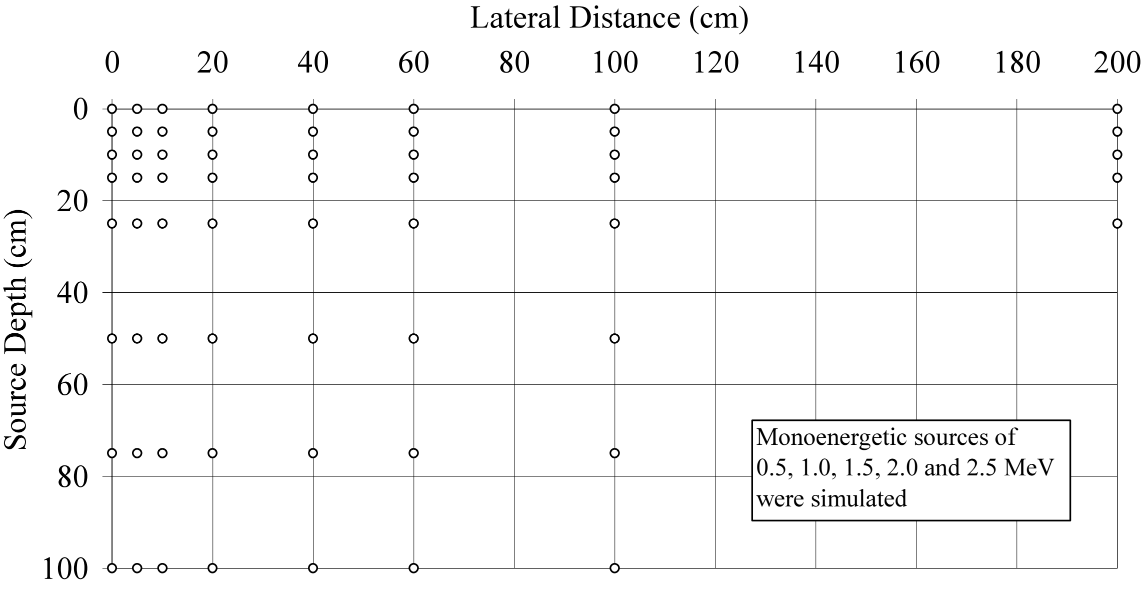


FIG. 9. Simulated Source Locations

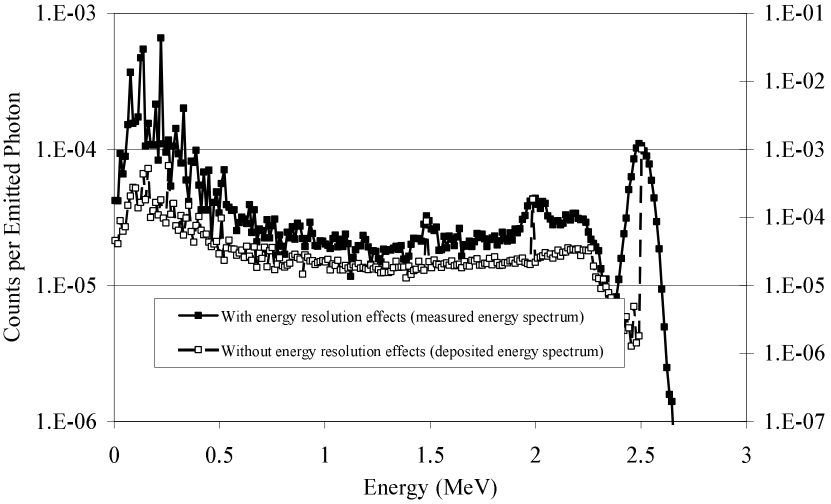


FIG. 10. Examples of "Deposited" and "Measured" Energy Spectra (2.5 MeV source buried 5 cm, 5 cm laterally from detector center)

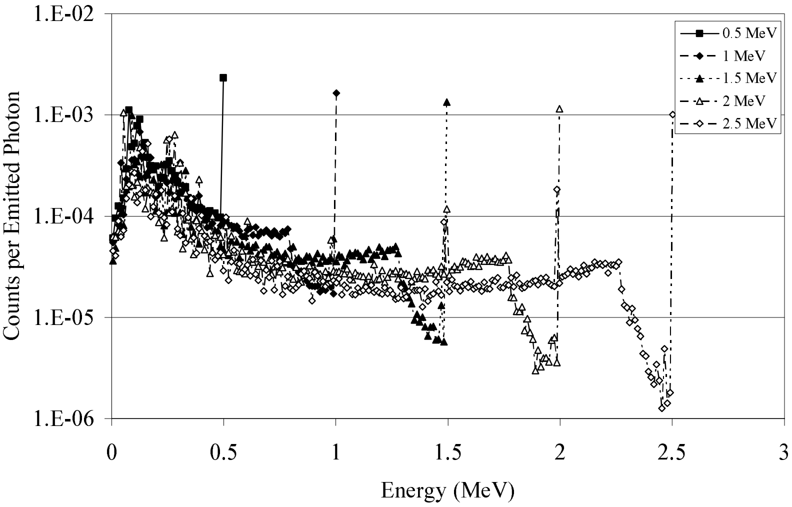


FIG. 11. Examples of Deposited Energy Spectra From Monoenergetic Point Sources (source buried 5 cm, 5 cm laterally from detector center)

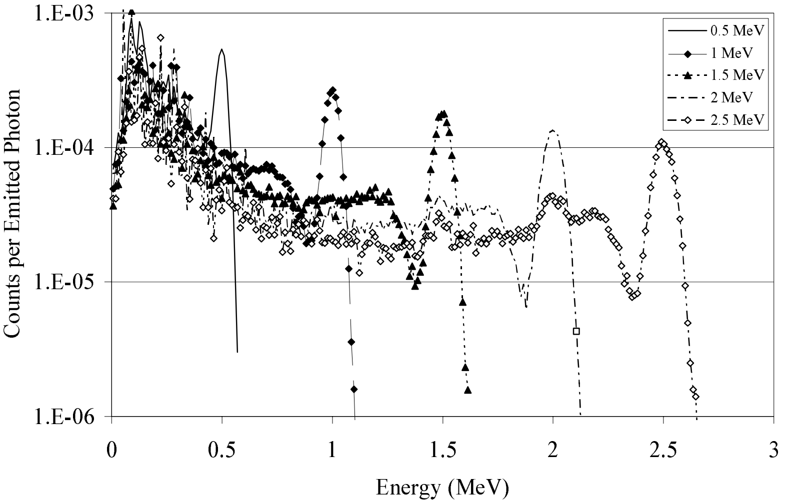


FIG. 12. Examples of Deposited Energy Spectra from Monoenergetic Point Sources (source buried 5 cm, 5 cm laterally from detector center)

escape peaks at 1.989 MeV and 1.478 MeV, and what appears (at least in the deposited energy spectrum) to be the pair production peak at 0.511 MeV.

Figures 11 and 12 show a series of spectra from a point source buried 5 cm in soil at a lateral distance of 5 cm from the center of the detector. The five spectra shown in each figure is from a monoenergetic source of each of the simulated energies (0.5, 1.0, 1.5, 2.0 and 2.5 MeV). Figure 11 is of deposited energy spectra, and so the peaks are at discrete energies. Figure 12 is of "measured" energy spectra, so the peaks have a distinct width that reflects the energy resolution of the simulated detection system. Note in Fig. 13 that the full-width at half-maximum (FWHM) of the full energy peaks increases with energy as expected.

3.3.2.4. Detector energy resolution

The energy resolution of a NaI(Tl) spectrometer cause a "blurring" of the deposited energy spectrum. Although there are many origins of resolution loss, the primary source in most NaI(Tl) spectroscopy applications is a result of statistical spreads resulting from the finite number of electrons generated per event at the initial photocathode in the photomultiplier tube. Take, for example, the case of 1 MeV of electron energy deposited in an NaI(Tl) crystal by a gamma ray photon. For this type of detector, the scintillation efficiency is approximately 12% and the scintillation photon energy is approximately 3 eV. Therefore, approximately 120 keV is converted into visible light, which translates to approximately 40,000 scintillation photons. Allowing for some light-loss, perhaps 30,000 of these photons reach the crystal-phototube interface. Assuming an average quantum efficiency of the photocathode over the scintillation spectrum is 20%, then 6,000 photoelectrons are produced. This is the stage in the signal chain where the number of information carriers is at a minimum, and therefore the expected variability is at a maximum. If we further assume that Poisson statistics hold, then the standard deviation on the mean of 6,000 photoelectrons is 77 (the square root of 6,000), or about 1.3%

Energy resolution is formally defined as the full width at half maximum (FWHM), in percent of peak energy, of the peak. For a Gaussian distribution, the FWHM is 2.35 times the standard deviation, so the energy resolution at 1 MeV in the example given above would be 3.0%.

The energy resolution of the simulated detection system (the Exploranium GR-256) was determined empirically using the four most prominent peaks from a spectrum of ^{226}Ra plus progeny: 0.609 MeV, 1.12 MeV, 1.765 MeV and 2.204 MeV. The results are shown graphically in Fig. 13. The energy resolution curve, $R(E)$, was fitted to an equation of the form:

$$R(E) = \frac{FWHM(E)}{2.35} = \frac{A + B \times \sqrt{E + C \times E^2}}{2.35} \tag{5}$$

where A, B and C are numerical constants selected in the fitting process. This particular form was chosen since the numerator of this equation, which describes the standard deviation of the Gaussian peaks, is the form accepted by MCNP for its "Gaussian Energy Broadening" function. Through iterative fitting, it was found that simplifying the equation slightly by setting A= 0 did not significantly effect the "goodness of fit" to the data. The result was B=°0.0731 and C= -0.0636 MeV^{-1}. The smooth curve in Fig. 13 represents this fitted equation using these parameters.

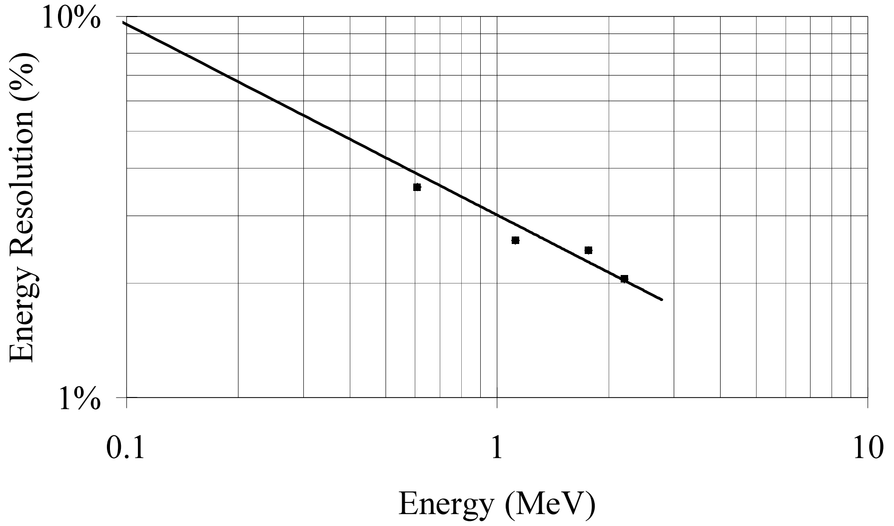


FIG. 13. GR-256 Energy Resolution

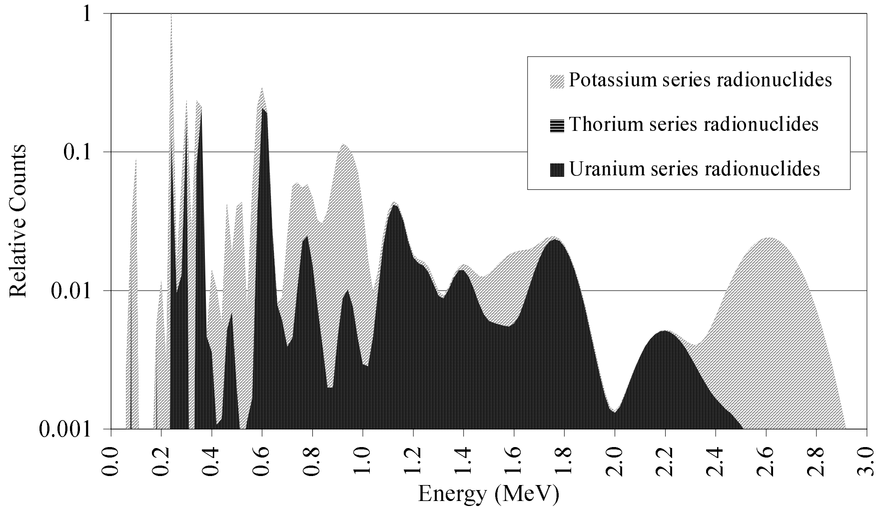


FIG. 14. Relative Naturally Occuring Radionuclide Gamma Radiation Emission Spectra Convoluted with Detector Resolution Effects
(K:U:Th Specific Activity Ratio of 15:1:1)

Detector resolution effects could have been incorporated in the MCNP simulation. In fact, the MCNP simulations of monoenergetic point sources produced two spectra each: one with and one without resolution effects. To simplify the analysis of the data, it was decided to use the non-broadened spectra in the following analysis, and then apply the energy broadening effect at the end of the process.

To demonstrate the resolution effects on the measured spectrum in the absence of other convoluting factors such as scattering, and detection efficiency, Fig. 14 shows an example gamma radiation emission spectrum for naturally occurring radionuclides to which the effects of detector resolution have been applied. The energy resolution used is the same as described above for the GR-256. Note the narrower peaks in the low energy range and the significant increase in peak width as energy increases. Unfortunately, in real spectra scattering effects tend to overwhelm the low end of the spectrum to make these relatively narrow peaks unusable. As well, due to the relative ease with which lower energy photons are attenuated, far fewer of the low energy emitted photons reach the detector.

3.3.3. *Model fitting*

The MNCP simulation produced the energy spectra absorbed by the NaI crystal for a matrix of depths, distances and monoenergetic sources. Statistical models were fit to these spectra to produce general relationships relating adsorbed spectra to a continuous function of emitted energy, depth, and distance. These models facilitate estimation of spectra (interpolation) between simulated locations for discrete (point) sources and allow integration across volume sources.

The models comprised terms relating to the physical distance between source and the crystal, the distance through soil, the distance through soil and the depth in the soil. Modifying terms incorporating the energy of the absorbed photons were included in the model. Three separate models were fit to the simulated data depending on the region in the spectra; specifically, the uncollided peak, the continuum 350 keV or lower than the emitted energy and the area between the emitted energy and the continuum more than 350 keV lower than the emitted photons.

The Marquardt algorithm, implemented under PROC/NLIN of the SAS software system, was used to fit the logarithms of the simulated counts to the models. The model fitting was restricted to energy levels of 1000 keV or larger for two reasons: first, the GR-256 uses a Cs-137 source for energy calibration during the *in situ* measurements and this source makes simulation of the spectra difficult for energies less than about 800 keV. Second, there are no major peaks from the uranium (^{226}Ra) series between the ^{137}Cs energy and 1,000 keV.

The initial model fitting fit the data reasonably well; however, it is felt that the fitted model could be improved substantially with further work.

3.3.4. *Estimated spectra*

The models were used to the estimate spectra based on the source geometry and activity. The definition of source activity includes not only the activities present but the energy, and abundance, for each emitted photon. There a large number of photons released in the natural radionuclide series; hence, only those that contribute significantly to the spectra were modeled. A cut-off of 0.1% abundance was used.

Absorbed energy spectra are numerically integrated over the source volume for each emitted photon using the fitted models and then the gaussian energy broadening correction is applied. The contributions are summed over the entire spectrum to provide the observed spectra in the GR-256 device.

Figure 15 compares two spectra. The simulated background spectrum is for typical natural background radionuclide levels with 0.037 Bq/g for the natural uranium and thorium series and 0.6 Bq/g level for ^{40}K. The second spectrum in this figure is an actual measured spectrum for soil with approximately these specific activities. Energies below 1 MeV are not shown since the measured spectrum in this energy region is significantly influenced by a small ^{137}Cs source within the detector housing used to stabilize the spectrum. The contribution to the simulated from this source is not taken into account, so comparisons of these spectra at energies below approximately 1 MeV are not possible. Although the shapes of the spectra are very similar, it is clear that there is a shift in energy between the two that is more pronounced at higher energies than at lower energies. Stretching of spectra measured in the field due to temperature and other effects on the electronics is not an uncommon phenomenon.

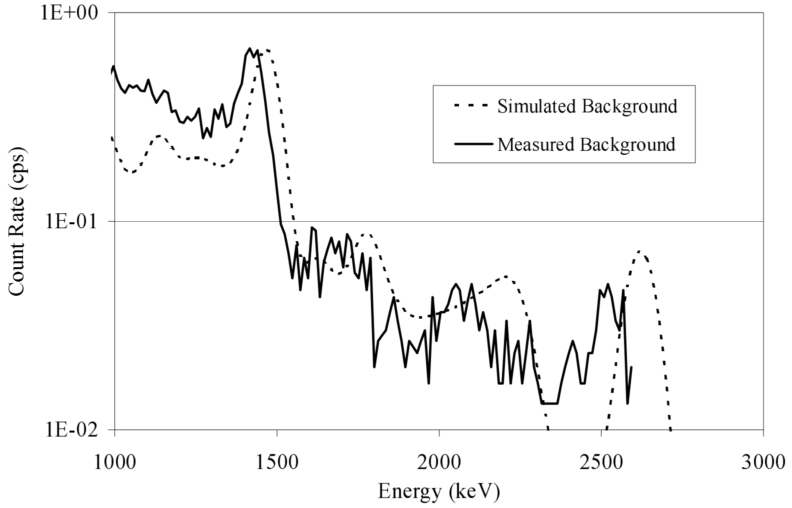


FIG. 15. Comparison of Simulated and Measured Background Spectra

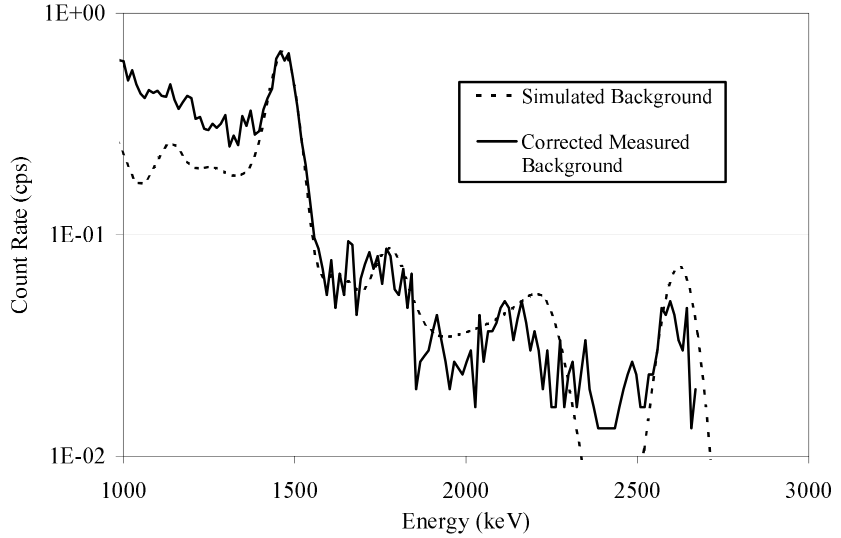


FIG. 16. Comparison of Simulated and Energy Calibration Corrected Measured Background Spectra

Figure 16 compares the same two spectra as Fig. 15, except that the energies associated with each channel in the measured background spectrum have been multiplied by a constant factor to compensate for the observed stretch in the measured spectrum.

Figure 17 compares measured and simulated spectra from point sources at various depths. The measured spectra are the same as those shown in Fig. 2 and discussed in section 4.2.1. of this report. The measured and simulated spectra shown in Figs. 16 and 17 compare fairly well. The full energy peaks line up very well, although the match for the continuum regions is not as good. Although this is by no means definitive, it does appear that it is possible to make use of the full energy peak areas from the simulated spectra to reliably estimate the real-world full energy peak areas. As well, it is clear that some additional work remains before the continuum regions of the simulated spectra can be used.

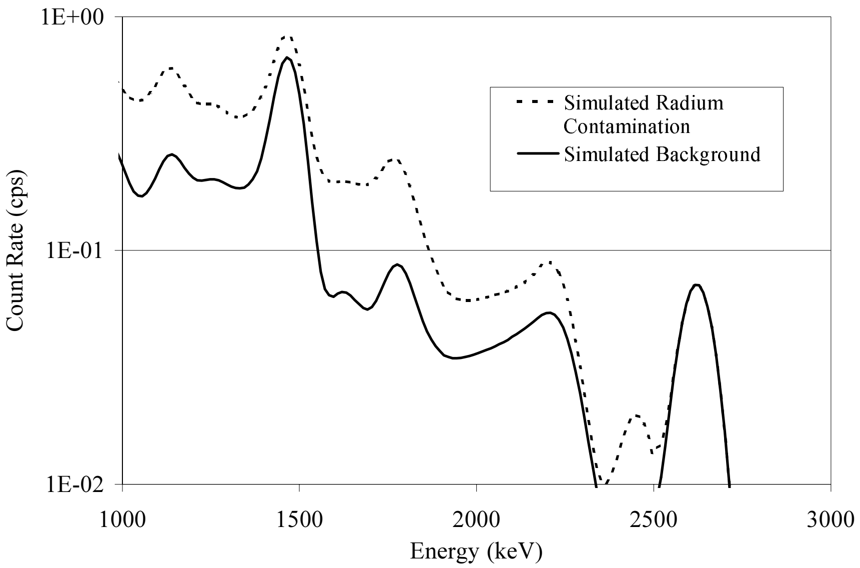


FIG. 17. Comparison of Simulated Spectra from Background and Radium Contaminated Soil

3.4. Review of potentially useful spectral features

Peak-to-peak ratios are potentially the most useful spectral feature for determining source depth. With some depth information, the determination of the activity of the source should be improved over current techniques. Other spectral features, such as peak-to-trough ratios, may also prove useful. A full analysis of these, and possibly other, spectral features should be a primary focus of future work.

4. SUMMARY AND CONCLUSIONS

4.1. Summary of results

Techniques to solve the inverse problem of estimating surface gamma radiation spectra given a fully known subsurface radium distribution have been investigated and applied with success. These techniques fell into three broad categories: empirical (using laboratory and field data), analytical (using mathematical derivations of relationships), and computer simulation (using Monte-Carlo photon transport simulation methods). Methods of analyzing surface spectra to estimate certain source parameters have been studied. The most fully developed methods are those involving the ratio of the areas of two peaks of differing energy from the same radionuclide to determine the source depth.

4.2. Conclusions

The work performed within this Coordinated Research Project is a significant step forward in the investigation of data gathering principles and analysis tools for improved estimates of subsurface radium contamination concentrations and distributions using surface gamma radiation spectra. For a point source of radium and its progeny, peak ratio techniques are able to reliably estimate the source depth from a single gamma radiation spectrum taken at the surface directly above the source. The only significant uncertainties in this case are the soil density and uncertainties introduced as a result of counting statistics.

Estimates of source depth may be useful themselves in some specific circumstances. In general, a more important use of the estimated or effective source depth is its use in improving the estimate of source activity.

Further work is required to fully achieve the goals of the larger project: to develop a comprehensive suite of tools for the improved interpretation of surface gamma radiation spectra from subsurface distributions of radium contaminated soil.

4.3. Future work

Further work remains to fully achieve the goals of the larger project: to develop a comprehensive suite of tools for the improved interpretation of surface gamma radiation spectra from subsurface distributions of radium contaminated soil. This work might first focus on improving the simulation of the continuum portion of the spectra. Additional work in quantifying the peak-to-peak ratio method for estimating source depth would likely be a primary focus. With well quantified tools for estimating source depth, estimates of source activities from surface spectra can be improved significantly.

TABLE I. RADIUM CONTAMINATION FROM THE CANADIAN RADIUM INDUSTRY.

Source of radium contamination	cause	inventory	status
Water Transportation Route	Spillage of ore during transport	60,000 m^3 of contaminated soil at 14 areas over 2200 km of transportation network.	Approximately 31,000 m^3 of soil contaminated with uranium ore has been cleaned up. Estimated doses are less than 1 mSv/a for remaining sites. Some properties have been redeveloped.
Port Hope area	Radium contaminated material from the refinery	1,300,000 m^3 of contaminated soil, harbour sediments and refinery residues	Approximately three-quarters of this material is in two waste management facilities, with the remainder at many smaller sites within the Town of Port Hope. The area communities are currently discussing the final disposition of all of the material.
Former radium dial painting operations	Contamination of work areas followed by a reuse of the buildings	Five buildings and properties	All buildings have undergone partial cleanups and are acceptable for occupancy. Doses are estimated at less than 1 mSv/a. Some 200 m^3 of contaminated building materials has been generated. More than 1000 m^3 of contaminated soil and building materials remains.
Instrument Repair shops	Working with luminescent dials without contamination control.	Several hundred shops with varying amounts of radium contamination.	Investigations and/or cleanups have taken place at about 50 shops. Generally, 1 to 10 m^3 of mildly contaminated waste is generated at a location.

TABLE II. ORE ANALYSIS

Statistic	Pitchblende fragments			Low-grade uranium bearing rock		
	Uranium [%]	**Arsenic** [%]	**^{226}Ra** [kBq/g]	**Uranium** [%]	**Arsenic** [%]	**^{226}Ra** [kBq/g]
Mean	36	1.1	86.5	4.8	0.20	8.65
Minimum	12	0.03	33.5	0.05	0.0014	0.078
Maximum	53	13	155	20	0.94	30.3
No. of samples	22	22	22	11	11	11

TABLE III. FORT McMURRAY PROPERTY SUMMARY INFORMATION

Item	Number
Property Area	99,000 m^2
Excavation Area	25,950 m^2
Excavation volume	22,000 m^3
Individual soil samples and spectral readings	1,405
Composite soil samples	232
Gamma Radiation readings	
Test Pits	177
Verification Iterations	496

TABLE IV. MALVERN REMEDIAL PROJECT EXTENDED SURVEY SUMMARY INFORMATION

Item	**Number**
Number of properties	450
Typical property area	600 m^2
Number of gamma measurements/property	2400
Number of soil samples/property	81
Spectral readings/property	40

TABLE V. MALVERN REMEDIAL PROJECT VERIFICATION SURVEY SUMMARY INFORMATION

Item	**Number**
Number of properties	75
Typical property area	600 m^2
Typical excavation area	400 m^2
Number of gamma radiation readings	2400
Number of individual soil samples	72
Number of spectral readings	72
Number of iterations of verification programme	2

TABLE VI. STRENGTHS AND WEAKNESSES OF EACH APPROACH

Approach	**Strengths**	**Weaknesses**
Empirical	Results are based directly on field data	Data available are limited to those encountered during LLRWMO projects and experiments
		Dependencies on independent variables may be difficult to resolve due to the lack of control over the source and soil parameters in project data
	A large database of relevant data already exists	Surface spectrum measurement uncertainties will introduce significant uncertainty in the results, particularly under low geometric efficiency situations
		Incomplete knowledge of subsurface radium distributions will introduce significant uncertainty in the results
Analytical	Exact results can be obtained	Only practical for the simplest of scenarios
	Dependence on all significant variables can be considered independently	Results require verification using field data
	Results directly relate spectral features with source parameters	
Simulation	Complex scenarios can be considered, and reasonably precise results obtained	The problem of predicting spectral features from source parameters must be solved before working on inferring source parameters from spectral features
	Scenarios not often found during project work or easily reproduced experimentally can be considered	Significant PC computing power and time are required
		Low geometric efficiency scenarios may require prohibitive amounts of computing time
	Radium distributions can be specified exactly	Results require verification using field data

TABLE VII. SIMULATED SOIL COMPOSITION

Element	**Mass fraction**
Oxygen	0.527
Silicon	0.246
Aluminum	0.073
Iron	0.049
Calcium	0.036
Sodium	0.021
Magnesium	0.020
Potassium	0.018
Titanium	0.005
Hydrogen	0.005

TABLE VIII. ^{226}Ra AND ITS DECAY PROGENY[a]

Radionuclide	**Half-life**	**Decay mode**[b]
^{226}Ra	1600 y	α
^{222}Rn	3.823 d	α
^{218}Po	3.05 m	α
^{214}Pb	26.8 m	β
^{214}Bi	19.9 m	β
^{214}Po	164 μs	α
^{210}Pb	22.3 y	β
^{210}Bi	5.01 d	β
^{210}Po	138.4 d	α
^{206}Pb	stable	n/a

[a] Low-probability branchings (in all cases less than 0.02%) have been omitted

[b] The primary decay mode

TABLE IX. GAMMA RADIATION EMISSIONS FROM ^{226}Ra AND ITS PROGENY[a]

Radionuclide	Energy [MeV]	Intensity [%]	Radionuclide	Energy [MeV]	Intensity [%]
^{226}Ra	0.084	0.30	^{214}Bi	1.408	2.49
^{226}Ra	0.186	3.28	^{214}Bi	1.509	2.22
^{214}Bi	0.077	0.36	^{214}Bi	1.539	0.44
^{214}Bi	0.079	0.60	^{214}Bi	1.543	0.36
^{214}Bi	0.090	0.27	^{214}Bi	1.583	0.72
^{214}Bi	0.274	0.18	^{214}Bi	1.595	0.27
^{214}Bi	0.387	0.37	^{214}Bi	1.599	0.34
^{214}Bi	0.389	0.41	^{214}Bi	1.661	1.15
^{214}Bi	0.406	0.17	^{214}Bi	1.684	0.24
^{214}Bi	0.427	0.11	^{214}Bi	1.730	2.97
^{214}Bi	0.455	0.32	^{214}Bi	1.765	15.80
^{214}Bi	0.470	0.13	^{214}Bi	1.838	0.38
^{214}Bi	0.474	0.12	^{214}Bi	1.847	2.09
^{214}Bi	0.609	46.30	^{214}Bi	1.873	0.23
^{214}Bi	0.665	1.57	^{214}Bi	1.896	0.18
^{214}Bi	0.703	0.47	^{214}Bi	2.119	1.17
^{214}Bi	0.720	0.40	^{214}Bi	2.204	4.98
^{214}Bi	0.753	0.13	^{214}Bi	2.293	0.33
^{214}Bi	0.768	5.04	^{214}Bi	2.448	1.56
^{214}Bi	0.786	0.32	^{214}Pb	0.053	1.11
^{214}Bi	0.806	1.23	^{214}Pb	0.075	6.21
^{214}Bi	0.821	0.15	^{214}Pb	0.077	10.46
^{214}Bi	0.904	0.11	^{214}Pb	0.087	4.67
^{214}Bi	0.934	3.21	^{214}Pb	0.242	7.50
^{214}Bi	0.964	0.38	^{214}Pb	0.258	0.55
^{214}Bi	1.052	0.32	^{214}Pb	0.275	0.33
^{214}Bi	1.070	0.29	^{214}Pb	0.281	0.33
^{214}Bi	1.120	15.10	^{214}Pb	0.295	19.20
^{214}Bi	1.133	0.26	^{214}Pb	0.352	37.10
^{214}Bi	1.155	1.70	^{214}Pb	0.462	0.17
^{214}Bi	1.158	3.51	^{214}Pb	0.480	0.34
^{214}Bi	1.208	0.46	^{214}Pb	0.487	0.44
^{214}Bi	1.238	5.94	^{214}Pb	0.534	0.19
^{214}Bi	1.281	1.48	^{214}Pb	0.580	0.37
^{214}Bi	1.304	0.12	^{214}Pb	0.786	1.10
^{214}Bi	1.378	4.11	^{214}Pb	0.839	0.59
^{214}Bi	1.386	0.78	^{210}Pb	0.047	4.00
^{214}Bi	1.401	1.39			

[a] Gamma radiation emissions with energy < 0.05 MeV or intensity < 0.1% have been omitted

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CHARACTERIZATION OF ENVIRONMENTAL RADIOACTIVITY IN THE INFLUENCE ZONE OF IFIN-HH BUCHAREST, MAGURELE

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ABSTRACT

As a consequence of 40 years of nuclear related activities on the Bucharest Magurele site, the environment surrounding the area of the institute was significantly influenced. The main facilities acting as sources of radioactive pollution are inter alia the VVR-S research reactor, the U-120 cyclotron, the radioisotope production centre, the radioactive waste treatment plant, and spent fuel storage facility.

In the framework of the IAEA Co-ordinated Research Project entitled “Site characterization techniques used in environmental restoration activities”, under research contract No.8735, methods for the characterization of radioactive contamination in the institute influence zone have been developed. The attention focused on the routes involved in radioactive waste and spent fuel management. Based on the environmental monitoring programme developed for the nuclear units on the site, the monitoring network was improved in area Reactor – Spent Fuel Storage – RWTP – railway station for shipping the drums to the national repository Baita-Bihor, an area with elevated radiological risks.

Additionally, the work has been extended to the temporary radioactive waste storage at the former Magurele military fort, which is now partially decommissioned.

The paper presents techniques, methods and instrumentation used for the radiological characterization of the site Magurele and its influence area, during the period of 1995 – 1999, as well as the respective quality assurance (QA) procedures. The results of this radiological survey presented here will be used to define the environmental restoration programme of the zone.

In addition the system for the information of the public developed in this project is discussed.

1. INTRODUCTION

Magurele Centre near Bucharest was the first nuclear site to be developed in Romania, starting in 1957 with the commissioning of the VVR-S research reactor and the U-120 cyclotron. Based on these installations a number of research and application programmes have been developed. These activities had some radiological impact on the Magurele area through radioactive emissions, as well as through the disposal of liquid and solid radioactive wastes.

Most significantly the wastes arose as by-product of the radioisotope preparation. The radioisotope production begun in hot cells and labs located in the underground level of the reactor building, at. In seventies this radiochemical production was transferred to a new dedicated building on the same site. The site was also upgraded with a new radioactive waste treatment plant (RWTP).

Before the treatment in the Magurele RWTP commenced, the wastes originating from the reactor operation and radiochemical labs, as well as in other applications developed on the site, were stored in an old military fort near the Institute of Atomic Physics. The institute was also in charge to collect all the radioactive wastes from country.

In 1977, after the serious floods of the Magurele fort, the wastes were transferred to the RWTP storage area and partially treated.

In the 1980s, the national repository Baita-Bihor, located in the Western Carpathians mountains, was legally commissioned. This repository is situated in an old Uranium mine and is designed to receive 20,000 standard 200 l-drums. The radioactive wastes are transported from the RWTP Magurele to the national repository Baita-Bihor by train. The site is provided with a special area for unloading of drums from the railway wagons.

Taking into account the growing public concern, the increased attention paid by international organizations [1] [2] [3], as well as the development of the national legislation on environmental protection and safe operation of nuclear installations, environmental monitoring in the vicinity of nuclear facilities is performed according to the following legal framework:

- Law of environmental protection [4];
- Law of safe operation of nuclear installations [5];
- Rules, instructions and legislative documents (regulations) [6];
- The environmental concept of IFIN-HH (the holder of the license) [7];
- Practices developed in different nuclear units and QA procedures.

This research programme for characterizing the environmental radioactivity in the influence area of IFIN-HH Bucharest Magurele, has been developed as a part of the Coordinated Research Programme of the International of Atomic Energy Agency entitled “Site Characterization Techniques Used in Environmental Restoration Activities”.

2. PRESENT STATUS OF THE SITE

2.1. Geographical characterisation

2.1.1. Site location

The general plan of the zone given in Figure 1, and shows the geographical location of Magurele in the capital of the country. The site is situated on the Romanian Plain, between the rivers Sabar and Dambovita about 3 km to the SW. The city-center of Bucharest is some 8 km away. To the SSW of the site the Ciorogarla, Sabar and Arges rivers flow.

2.1.2. Geomorphology

The site belongs structurally to the Romanian Plain, forming a plateau at an altitude of 75–78 m above sea level, is situated on the left bank of Ciorogarla river, about 1500 m away from it. The site is generally flat, with little relief and thus allowing easy dispersion of released contaminants.

2.1.3. Surface waters

The catchment basin of Ciorogarla river has a surface area of about 116 km^2 and is entirely located within the Plain. The mean altitude of this catchment basin is 98 m above sea level.
The river has its origin in Brezoaiele. The minor waterway is 35–40 m wide and 3.5–4 m deep. Certain sections of the larger rivers are prone to flooding, in particular in the Plain.

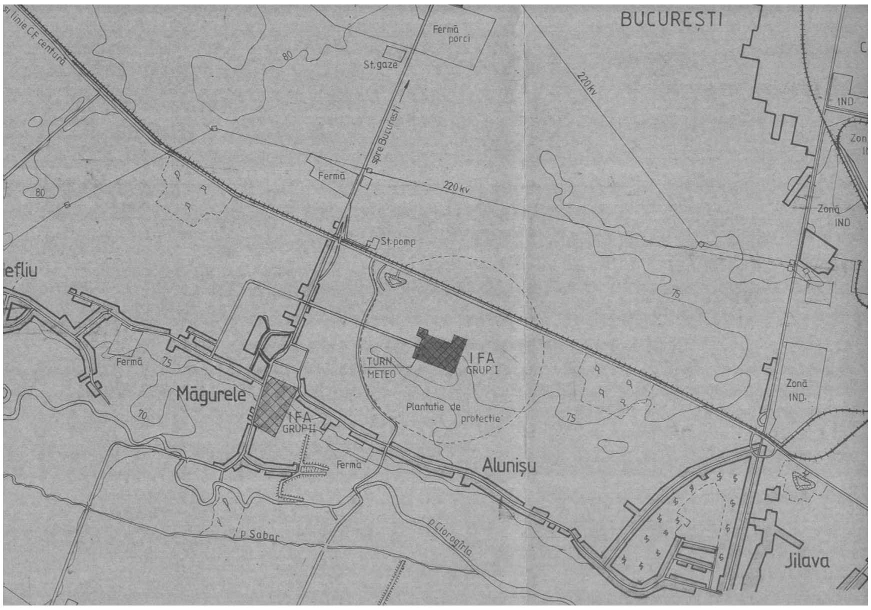


FIG. 1. General map of the area.

The Marguele site being on a high plateau above the river, it should be beyond the reach of the floods. It is worth noting that during severe winters, the river can freeze entirely and the flow then is considered zero.

2.1.4. Groundwaters

The first aquifer is found at 7 m in depth and is formed by sands and gravel layer 3–5 m in thickness. The aquifer is recharged by infiltration from the whole terrace, which extends to the East and up the river Dambovita. To the West, the aquifer discharges in springs at the bottom of slopes to the lower terrace of the river Sabar. The deeper groundwater drains through gravel layers into the same river. The groundwaters are used as drinking water for the neighborhood villages. The water table in the aquifer is 4 m above the river Sabar level and, therefore, the influence of the variations in river level upon the groundwater level is relatively limited. Variations in the groundwater levels are due to seasonal rainfalls and amount to about 2 m.

2.1.5. Climatic conditions

The IFIN-HH site, being located in the central part of the Romanian Plain, is influenced by the large masses of continental air, which are characterized by extreme differences between the maximum and minimum values of the same meteorological parameter. The current frequency distribution of dominant winds is presented in Table I.

The annual quantity of precipitation in its various forms sums up to about 500 mm in this area. The highest average monthly precipitation (80 mm) is falling in June, but it may also occur in May and August. However, during the summer, particularly in June, heavy local downfalls may occur, which may exceed 250 mm for a single event. The lowest average monthly quantities are falling in February (30 mm). The maximum annual quantity of precipitation may exceed 800 mm during years with intense cyclonic activity. In winter, the maximum monthly precipitation may exceed 100 mm.

Annual average temperatures are in the range of 10–11 °C, the long term annual average temperature being 10.4 °C. The variations of the average monthly air temperatures from month to month are the ones characteristic of continental areas in the temperate region. The maximum value recorded in July exceeds 22 ºC, while the minimum in January is below -4 ºC). The monthly average values of the daily maximum and minimum temperatures include external momentary values, constituting important characteristics of the thermic regime. The monthly average maximum temperatures may reach its highest values (above 35 ºC) in July and August, and its lowest one (below -10 ºC) in January.

2.1.6. Land use

The Magurele District covers an area of 43.39 km^2, and land use is distributed as indicated in Table II.

It is clear from Table II that the largest surface area is taken up by agricultural land. However, the cultivated land is located outside the sanitary protection area, which is defined by a radius of 800 m around the Reactor. This area is surrounded and planted outside the young forest limits, the access in this area being strictly controlled.

2.1.7. The distribution of population and activities in the IFIN-HH area

Within a radius of 1 km, there is no inhabited land, the only area which reaches this limit is Magurele being situated to the West of unit I. The inhabitants of the western area, one of the most densly populated areas, are distributed between several villages within a 6 km radius: Magurele, Varteju, Bulgaru and Bragadiru, which have a total of 7242 inhabitants. In the SE area, the population of about 8020 is distributed between two villages: Jilava and Odaile. The closest limit of Bucharest city is about 4 km away to the NE, in wind direction from the SW. The part of town being located to the North, is not situated in the direction of dominant winds. The main activity of the inhabitants living in the neighbouring villages is agriculture, particularly the cultivation of vegetables. At present, IFIN-HH has some 2200 non-residential employees, and there are also a number of high school and college students [8].

2.2. Radiological status of the site

IFIN-HH Bucharest is the legal owner of nuclear site, which comprises the following nuclear installations:

- the nuclear reactor VVR-S Bucharest;
- the RP01-zero power reactor (PUB);
- the subcritical assembly HELEN (UB);
- the neutron generator TEXAS;
- the cyclotron U-120;
- the TANDEM accelerator;
- the radioactive waste treatment plant (RWTP;
- the radioisotopes production center (RPC); and
- the nuclear medicine center (NMC).

The estimated maximum emissions to the atmosphere from these installations are between 0.1 and 1000 Ci/year. For the liquid effluents, the major contribution arises from the RPC, the nuclear reactor (including wet storage of spent fuel) and the RWTP. The solid wastes arise from in particular the RPC, the research reactor VVR-S Bucharest, the radioisotopes applications laboratories and some other nuclear units in the country.

In the IPNE influence zone nuclear activities have been ongoing for about 40 years, beginning with the startup of the research reactor VVR-S and the radioisotope production laboratory. This activity inherently led to environmental contamination by the release of the gaseous products to atmosphere and also due to the manipulation of liquid and solid radioactive materials.

Within the influence area of IPNE there is also the old Magurele Fort used as temporary storage for radioactive wastes. The storage in the fort was partially decommissioned in the seventies, but there are no records on the actual decommissioning activities.

Systematic measurements were performed in the area affected by the research reactor decommissioning in order to provide a reference status for site. This environmental survey will continue during the research reactor decommissioning and RWTP modernizing, taking into account possible impacts on the environment of these activities. Another important aspect in this context is the long term monitoring of decommissioned nuclear installations.

The permanent shut down of the reactor at the end of 1997 led to a significant decrease of contamination from the gaseous radioactive discharges. At presently, the estimated radioactivity of the inventory due the activation is about 1900 Ci, distributed over 150 t of different materials [8]. The contaminated inventory is contained in about 50 t of various materials, e.g. experimental installations, tubes, pipes, etc. The radioactive inventory of VVR-S research reactor (calculated as of 31.12.1999) is summarized in Table III.

The major radioactive inventory is provided by spent fuel stored in a deposit far away from reactor, on the site [9]. This deposit has 4 ponds, with 240 fuel assembly spaces. The radioactive inventory from spent fuel is estimated to be 106900 Ci as of 31 December 1999. At present there are 152 fuel assemblies of the EK-10 and 74 of S-36 type in this wet storage. The claddings of the fuel are affected by the fast development of corrosion. The content of ^{137}Cs in pond water is around 40000 Bq/l. The radioactive inventory in the spent fuel from research reactor, also calculated for 31.12.1999 is given in Table IV.

In the yard of the RWTP, there is a significant amount of historical wastes stored in drums (more then 800) in an advanced state of corrosion, being a serious source of radioactive contamination of the site. For example, RWTP treated up to 1997 the following wastes:

—	LLW liquids	25000 m^3
—	MLW liquids	2 m^3
—	LLW solids	2000 m^3
—	MLW solids	2 m^3
—	Total	5300 standard 200 l drums were sent to Baita-Bihor

Another problem, in terms of contamination, is connected with the railway platform for dispatching of the drums to the national repository Baita-Bihor. Preliminary measurements in the fort Magurele area indicate also a spreading of radioactive contamination due to wood wastes from the Scrovistea site stored near the facility, and also due partial decommissioning of this storage site following the floods in 1977. The Radioisotopes Production Centre reduced its activity, but there are still many sources of contamination resulting from its operation.

3. ENVIRONMENTAL MONITORING PRACTICES

3.1. Site monitoring

IFIN-HH has developed an environmental monitoring concept [7]. Nineteen monitoring points for the sampling of vegetation, soil and waters were established. IFIN-HH also established the sampling frequency and the methods applied, including their respective detection limits.

Significant amounts of radioactive wastes (in both, liquid and solid forms) are result from the routine nuclear activities on the Magurele site. All nuclear activities are grouped into nuclear units. Their monitoring is performed using the above mentioned environmental concepts [7] and subject to various QA procedures:

ACPL 0812 –	Monitoring of total β and γ radioactivity of the environmental factors for nuclear units of IFIN-HH;
ACPL 0821 –	Monitoring of total β and γ radioactivity of the liquid effluents (released by the nuclear units of IFIN-HH);
ACPL 0813 –	Monitoring of total β and γ radioactivity of atmospheric aerosols and fallout.

The monitoring places and the sampling procedures are defined by internal documents. These sampling locations cover a large area, taking into account the meteorological factors and nuclear activities. On Figure 2 the sampling points are marked, a description of which, the type material sampled and the compass sector from the site are given in Table V.

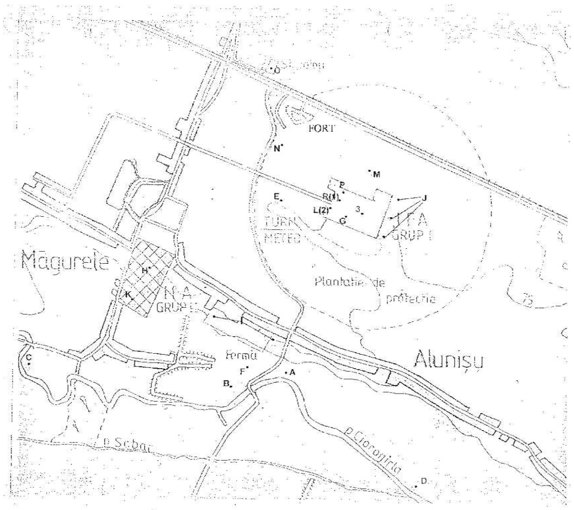


FIG. 2. IFIN-HH and the influence zone.

The sampling frequency depends on the characteristic average time of residence along an exposure route for each nuclide. Total β and γ measurements are the methods used for the characterization of the radioactivity and its influence area around IFIN-HH. Table VI indicates the sampling frequency for different contaminants.

3.2. Monitoring of the reactor area

Taking into account the future development of the nuclear activities in the IFIN-HH zone [10], including the decommissioning of the VVR-S research reactor, modernizing of RWTP, the decontamination of the old fort site, there is an absolute necessity for an improvement of the environmental monitoring system outlined above. As part of the nuclear activities large amounts of liquid and solid radioactive wastes are manipulated, resulting potentially in airborne radioactive contamination in the IFIN-HH influence zone. The required radiological site characterization must become more detailed by the increasing of the number of sampling points and the use of new measuring techniques.

To this end, in 1996 nine new sampling points were established, which are listed in Table VII. In 1997 we considered four additional points in the RWTP yard. In 1998 the surveyed area was enlarged in order to cover the waste routes up to railway facility used for the dispatching of drums to the National Repository Baita-Bihor. In Figure 3 all sampling points are indicated (with the exception of those at the railway facility) [11] [12].

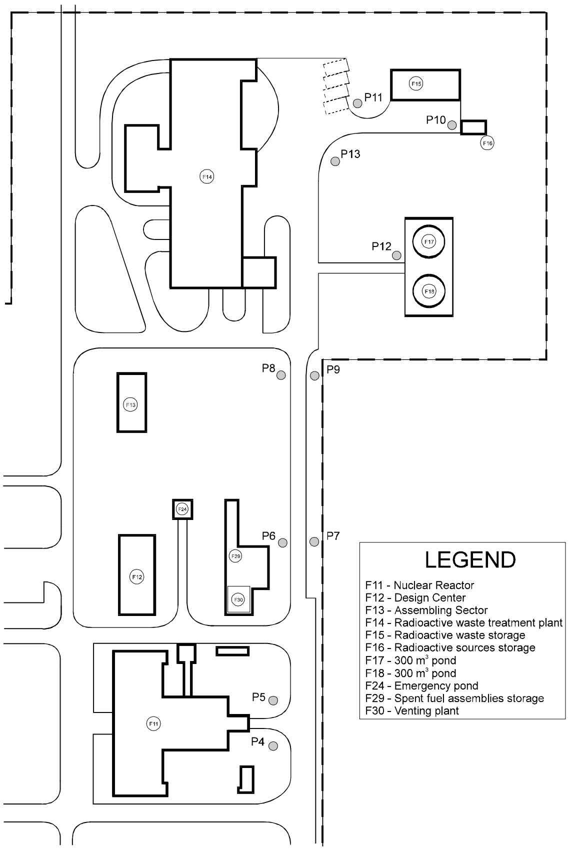


Figure 2

FIG. 3. Sampling locations.

Eighty one sampling points were established in the area around the reactor spent fuel storage for investigating its radiological status. At 48 of these points, including the access way to the reactor and the radioactive waste treatment plant (RWTP), the doses and contamination have been measured. Some sampling points were set up in the area of the pond for storing possibly contaminated waters.

In Figures 4 and 5 the measuring points for dose rates and contamination, on the asphalt covered roads, as well as on the vegetation of the green verges along the wastes transportation routes, are indicated.

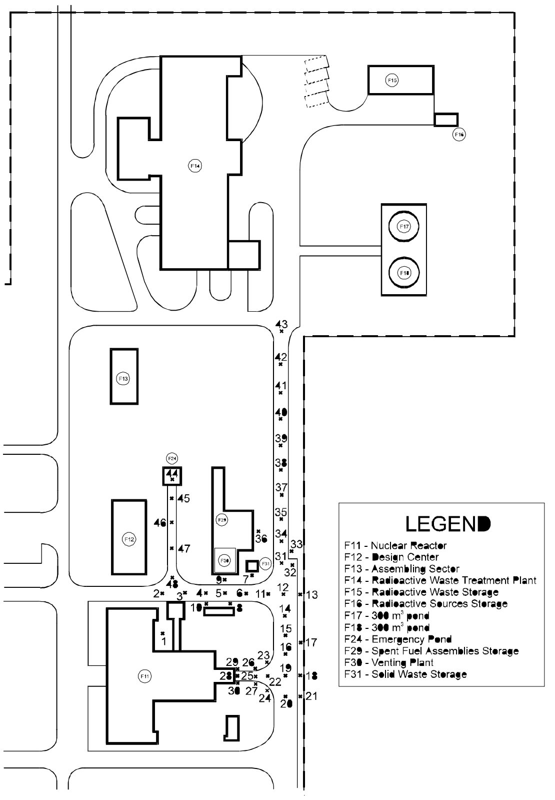


FIG. 4. Locations for dose rate and contamination measurements.

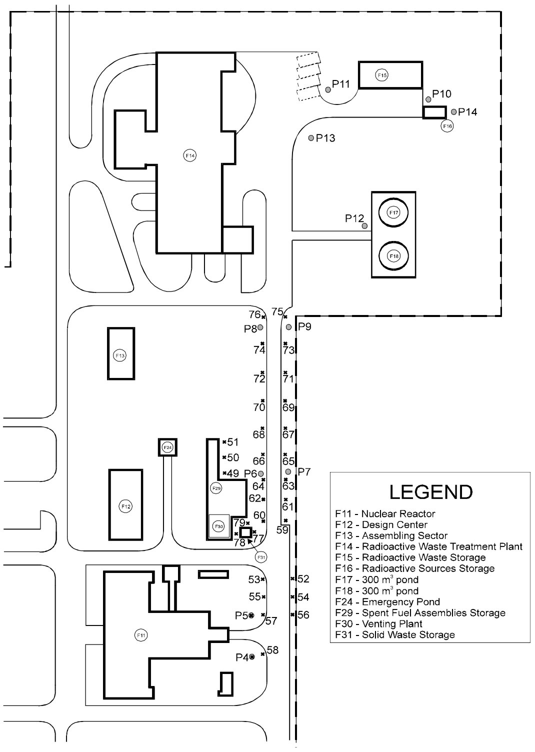


FIG. 5. Locations for dose rate measurements.

3.3. Monitoring Magurele Fort

The Magurele Fort, located near the Institute, was used in the 1960s as a temporary storage for radioactive sources and wastes. In 1974, a few tonnes of trees contaminated with ^{60}Co from biological research, performed in sixties at Scrovistea, were stored near the Fort, outdoors and without weather protection. In 1975, it was first tried to remove the radioactive materials from the Fort, which had accumulated due to flooding; but the operation was not completed. The windows were safely closed by concrete walls. In 1979, it appears that the physical barriers had been destroyed, but they were rebuilt. In 1998 the Regulatory Body (CNCAN) inspected the area. The physical barriers were, again, broken and then the decision for ecological restoration of the zone was taken. In this context, the institute performed some preliminary measurements for radiological characterization of the site [13] [14].

In early spring 1998 doses have been measured in one room of the fort, where radioactive wastes had been stored. Doses have also been measured at a number of points in the near neighborhood of the fort and at the storage area of the contaminated wood. Samples were taken from different depths and analyzed using γ-spectrometry in order to identify the contaminants.

In summer 1998 a new measurement was performed in the contaminated wood storage area. The measurements included dose rates and the determination of contaminants on a 2 m-grid, covering about 72 m^2. At the central point of the grid *in situ* γ-spectrometry measurements were performed. Another γ-spectrum was determined in front (outside) of the fort gate. The distribution of soil radioactivity was determined in depths up to 35 cm.

4. METHODS

4.1. Instrumentation

In situ measurements were performed using portable instrumentation (type SMART 200), with the associated detectors, and detailed in Table VIII. The high resolution γ-spectrometry measurements were performed using a NOMAD spectrometer, with absolute calibration. The main characteristics of the spectrometer are given in Table IX.

In accordance with the Romanian national legislation referring to metrology, all the instrumentation, which was obtained through generous assistance by the IAEA in the framework of the Technical Assistance Projects ROM/9/017 and ROM/9/017 EXTENDED, must be certified by National Bureau of Legal Metrology. Also, the personnel involved in standardizing measurements is to be authorized by above mentioned organization. The Investigator's laboratory is authorized to perform standardizing measurements for radioactive soils, vegetation and waters. The metrological calibration of α-, β- and γ-dosimeters is also to be performed by the National Institute of Metrology Bucharest.

QA procedures for sampling, calibration of instrumentation, measuring and reporting (bulletins) were developed.

4.2. Calibration of the spectrometer

In the following paragraph are presented the calibration results for the NOMAD installation working with the following parameters:

HV (+) = 2300 V;
Gain 0.8752x40=35.00;
Shaping time = LONG;
Preamplifier type = Resistive feedback;
Automatic Pole Zero Adjustment;
Digital Spectrum Stabilizer;
ADC Conversion Gain = 4096;
Digital Offset = Automatic PZC setting up was performed.

The energy calibration

An ^{152}Eu point source has been used for the determining of the function of energy vs. channel with the following geometries given in Figure 6.

Maximum nonlinearity

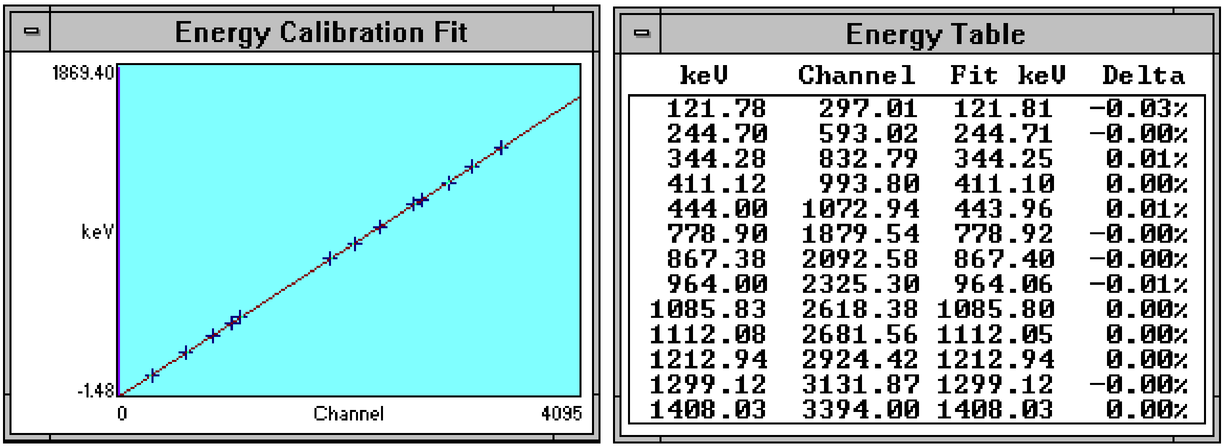
0,025% for the range (20...1800) keV
Linear calibration constant: 0,415 keV/channel; ±0,2% for P^*=99,73%, in the range (35...1600) keV.

Measured energy resolution

0,62 keV for 59,54 ke V ^{241}Am (0,96 keV from fit)
1,39 keV for 661,62 keV ^{137}Cs (1,392 keV from fit)
1,82 keV for 1331,52 keV ^{60}Co (1,841 keV from fit)

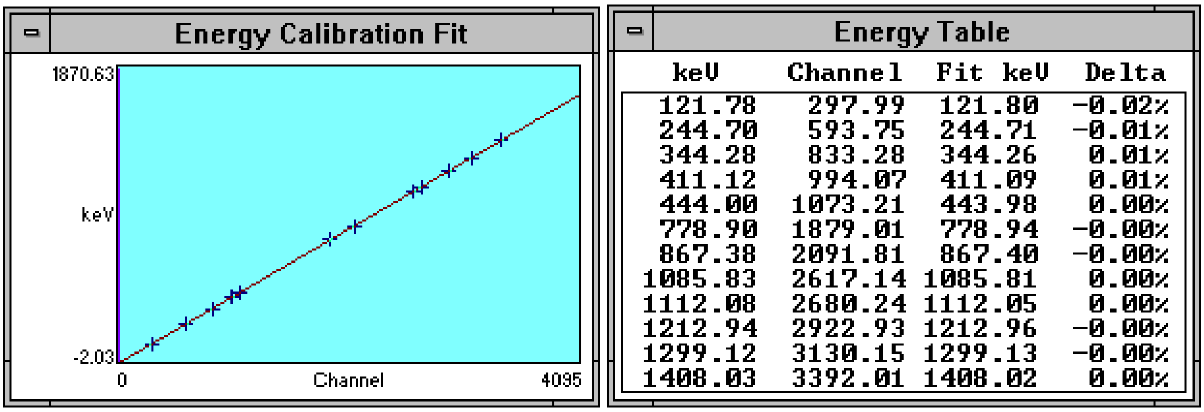
See also Figure 7.

A) point source



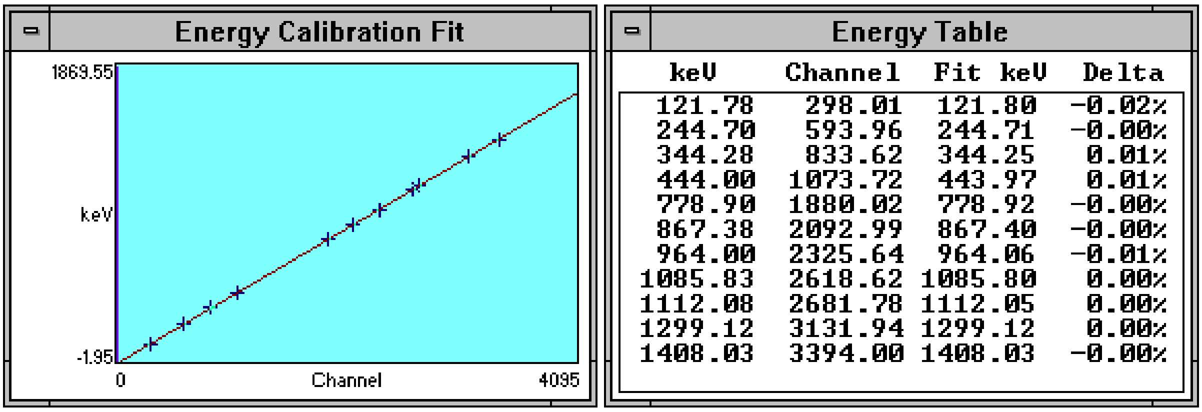
keV	Channel	Fit keV	Delta
121.78	297.01	121.81	-0.03%
244.70	593.02	244.71	-0.00%
344.28	832.79	344.25	0.01%
411.12	993.80	411.10	0.00%
444.00	1072.94	443.96	0.01%
778.90	1879.54	778.92	-0.00%
867.38	2092.58	867.40	-0.00%
964.00	2325.30	964.06	-0.01%
1085.83	2618.38	1085.80	0.00%
1112.08	2681.56	1112.05	0.00%
1212.94	2924.42	1212.94	0.00%
1299.12	3131.87	1299.12	-0.00%
1408.03	3394.00	1408.03	0.00%

B) volume source



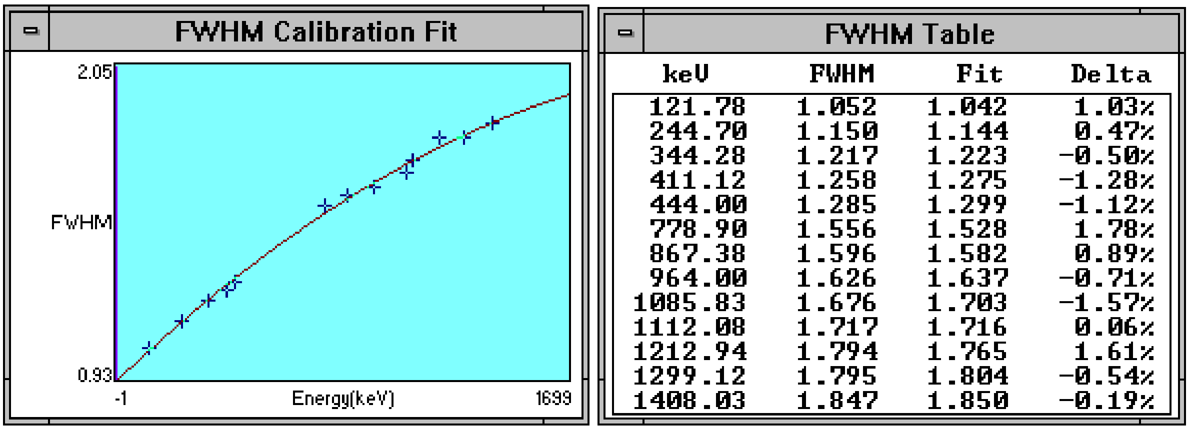
keV	Channel	Fit keV	Delta
121.78	297.99	121.80	-0.02%
244.70	593.75	244.71	-0.01%
344.28	833.28	344.26	0.01%
411.12	994.07	411.09	0.01%
444.00	1073.21	443.98	0.00%
778.90	1879.01	778.94	-0.00%
867.38	2091.81	867.40	-0.00%
1085.83	2617.14	1085.81	0.00%
1112.08	2680.24	1112.05	0.00%
1212.94	2922.93	1212.96	-0.00%
1299.12	3130.15	1299.13	-0.00%
1408.03	3392.01	1408.02	0.00%

C) Marinelli source



keV	Channel	Fit keV	Delta
121.78	298.01	121.80	-0.02%
244.70	593.96	244.71	-0.00%
344.28	833.62	344.25	0.01%
444.00	1073.72	443.97	0.01%
778.90	1880.02	778.92	-0.00%
867.38	2092.99	867.40	-0.00%
964.00	2325.64	964.06	-0.01%
1085.83	2618.62	1085.80	0.00%
1112.08	2681.78	1112.05	0.00%
1299.12	3131.94	1299.12	0.00%
1408.03	3394.00	1408.03	-0.00%

FIG. 6. Calibration of nomad: energy vs. channel for (a) point, (b) volume, and (c) marinelli sources.



keV	FWHM	Fit	Delta
121.78	1.052	1.042	1.03%
244.70	1.150	1.144	0.47%
344.28	1.217	1.223	-0.50%
411.12	1.258	1.275	-1.28%
444.00	1.285	1.299	-1.12%
778.90	1.556	1.528	1.78%
867.38	1.596	1.582	0.89%
964.00	1.626	1.637	-0.71%
1085.83	1.676	1.703	-1.57%
1112.08	1.717	1.716	0.06%
1212.94	1.794	1.765	1.61%
1299.12	1.795	1.804	-0.54%
1408.03	1.847	1.850	-0.19%

FIG. 7. Calibration of nomad: measured energy resolution.

Time stability of the peak position

The time stability was determined over the duration of 8 hours by 1 hour repeated measurements at the energies of 121.7 keV and 1408.03 keV. The spectrometer was run with a peak stabilizer, the shifting stability being lower than the tolerances accepted by the analyzing program.

Minimum detectable activity

This parameter is defined by the relationship MDA= $3{\cdot}S_{background}/Eff$, where MDA is the minimum detectable activity, $S_{background}$ the error in background counting and Eff the efficiency. The values determined are given in Table X.

Efficiency calibration

Efficiency calibrations for various geometries are illustrated in Figure 8.

Errors

NOMAD Calibration errors are given in Table XI.

Time stability

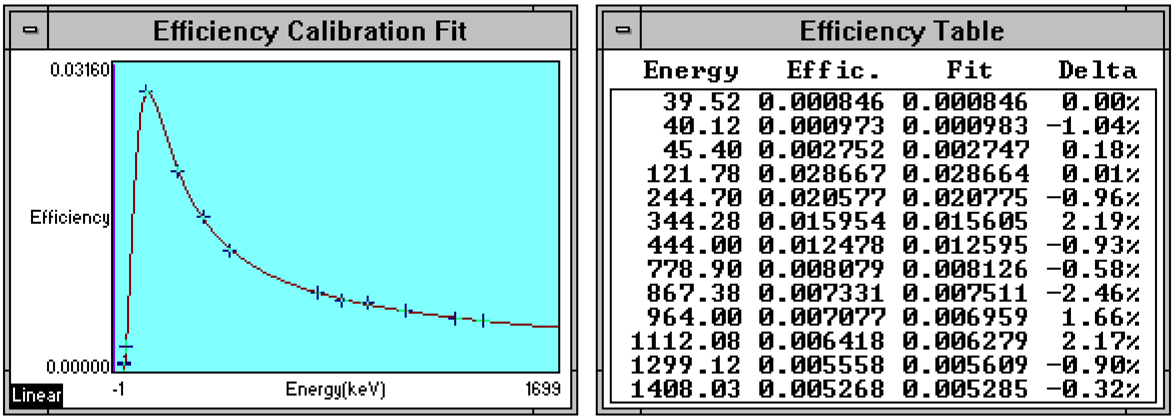
The time stability of the NOMAD calibration is illustrated in Figure 9.

4.3. Calibration for *in situ* measurements

For the determination of gamma-spectrometer calibration factors to be used for *in situ* measurement a circular geometry (1 m radius) was chosen. The ring was divided into 24 equal sectors. The reference sources used were the following:

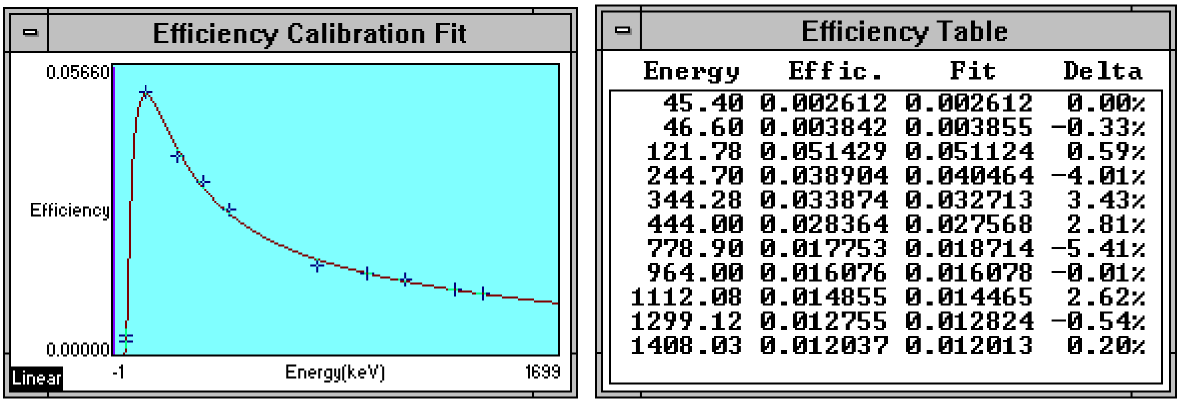
^{152}Eu with an activity of 370.0 kBq
^{60}Co with an activity of 306.9 kBq
^{137}Cs with an activity of 255.7 kBq
^{241}Am with an activity of 338.1 kBq
all as of 1 July 1985.

A) point sources



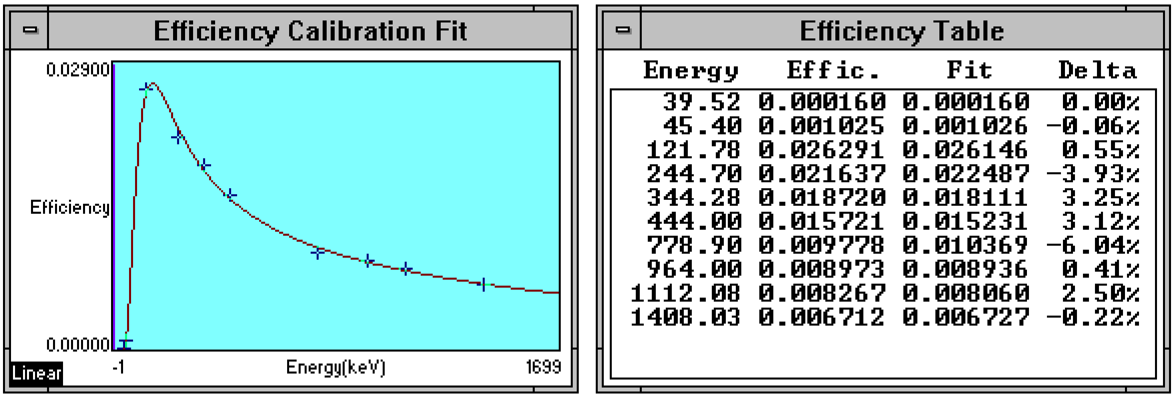
Energy	Effic.	Fit	Delta
39.52	0.000846	0.000846	0.00%
40.12	0.000973	0.000983	-1.04%
45.40	0.002752	0.002747	0.18%
121.78	0.028667	0.028664	0.01%
244.70	0.020577	0.020775	-0.96%
344.28	0.015954	0.015605	2.19%
444.00	0.012478	0.012595	-0.93%
778.90	0.008079	0.008126	-0.58%
867.38	0.007331	0.007511	-2.46%
964.00	0.007077	0.006959	1.66%
1112.08	0.006418	0.006279	2.17%
1299.12	0.005558	0.005609	-0.90%
1408.03	0.005268	0.005285	-0.32%

B) volume sources (of cylindrical shape)



Energy	Effic.	Fit	Delta
45.40	0.002612	0.002612	0.00%
46.60	0.003842	0.003855	-0.33%
121.78	0.051429	0.051124	0.59%
244.70	0.038904	0.040464	-4.01%
344.28	0.033874	0.032713	3.43%
444.00	0.028364	0.027568	2.81%
778.90	0.017753	0.018714	-5.41%
964.00	0.016076	0.016078	-0.01%
1112.08	0.014855	0.014465	2.62%
1299.12	0.012755	0.012824	-0.54%
1408.03	0.012037	0.012013	0.20%

C) volume Marinelli sources



Energy	Effic.	Fit	Delta
39.52	0.000160	0.000160	0.00%
45.40	0.001025	0.001026	-0.06%
121.78	0.026291	0.026146	0.55%
244.70	0.021637	0.022487	-3.93%
344.28	0.018720	0.018111	3.25%
444.00	0.015721	0.015231	3.12%
778.90	0.009778	0.010369	-6.04%
964.00	0.008973	0.008936	0.41%
1112.08	0.008267	0.008060	2.50%
1408.03	0.006712	0.006727	-0.22%

FIG. 8. Calibration of nomad: efficiency calibration for (a) point, (b) volume, and (c) volume marinelli sources.

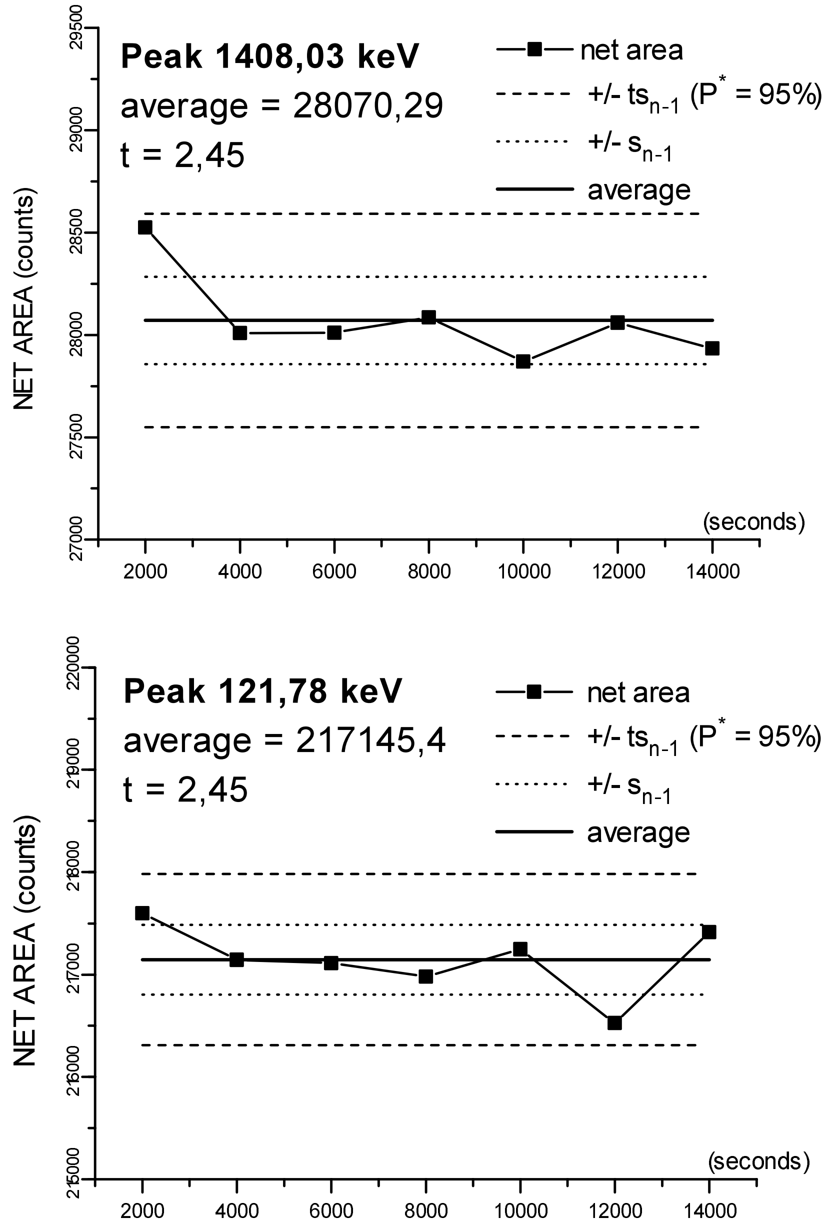


FIG. 9. Calibration of nomad: Time stability.

A field calibration was performed by a combined method (experimental and theoretical) as recommended by Helfer and Miller [15]. Field calibrations differ from those for laboratory sample counting in that no spiked samples are used; rather, a combination of experimental and theoretical methods are applied. The procedure can be summed up as follows:

The basic calibration equation for *in situ* spectrometry can be expressed in terms of the peak count rate N, the activity or inventory in the soil A, and the uncollided flux Φ as:

$$N_f/A = (N_f/N_0)(N_0/\Phi)\,(\Phi/A) \qquad (1)$$

where the fundamental calibration parameters are expressed in ratios as follows:

N_f/A is the total-absorption peak count rate (cpm) in the spectrum at the energy of a particular nuclide γ transition per unit inventory ($Bq \cdot m^{-2}$) or concentration ($Bq°g^{-1}$) of that nuclide in the soil,

N_f/N_0 is the angular correction factor of the detector at that energy for a given source distribution in the soil,

N_0/Φ is the peak count rate (cpm) per unit uncollided flux ($\gamma \cdot cm^{-2} \cdot s^{-1}$) for a parallel beam of γ rays of the same energy that is incident-normal to the detector face, and

Φ/A is the total uncollided flux ($\gamma\ cm^{-2} \cdot s^{-1}$) at that energy arriving at the detector per unit inventory or concentration of the nuclide in the soil.

In order to obtain the calibration factor (N_f/A) for a particular nuclide, the three quantities (N_f/N_0, N_0/Φ and Φ/A) are determined separately. The first two terms are detector dependent and can be determined experimentally, while the latter can be calculated on a purely theoretical basis.

The value of Φ/A at a particular energy is obtained by counting a γ-emitting point source of known strength, placed at a distance of at least 1m from the detector face to simulate a parallel beam of normally incident radiation.

The value of N_f/N_0 for a particular energy and source distribution is calculated from:

$$N_f / N_0 = \frac{\int R(\theta)\Phi(\theta)d\theta}{\int \Phi(\theta)d\theta}, \tag{2}$$

where $R(\theta)$ is the peak count rate for γ-rays of energy E at angle θ, relative to the peak count rate at $\theta = 0^\circ$ (normal incidence) and $\Phi(\theta)$ is the γ-ray flux at energy E at angle θ.

5. RADIOLOGICAL CHARACTERIZATION

5.1. Reactor area — RWTP — spent fuel storage — platform

The techniques developed in the framework of this project have been applied to the radiological characterization of the reactor area, the spent fuel storage facility, the RWTP, and the railway platform for dispatching of drums for transport to Baita-Bihor national repository.

The following types of measurements have been performed:

- measurements of radioactivity on the soil and vegetation samples, collected in the period 1996–1997, in places indicated in Figure 6;
- measurements of doses, and β- and γ-contamination on the waste transport routes, as well as at the spent fuel manipulation facility;
- *in situ* spectrometry in order to determine the radioactivity distribution in the soil [16] [17].

The samples of soil and vegetation have been collected in autumn 1996, at the points P1 to P9. The samples have been processed and measured by the procedures presented in the previous progress reports [11] [12]. The results are given in Table XII. The measurements show a ^{137}Cs contamination at locations P6 and P7, and in the neighbourhood of the spent fuel storage, the soil radioactivity there being two times higher than in other areas of the site. This contamination appeared due to the diffusion of fission products from ponds (e.g. through cracks in the lining). The volatile fission products are dispersed outside of the building.

The measurements in the water from the spent fuel storage ponds showed during the same period the presence of 40.000 Bq/l of ^{137}Cs. The decision that the venting in the spent fuel storage was to be stopped (for cost reasons) led to an increase of radioactivity in the soil near the facility. Other measuring places show a radiation levels comparable to those at control points P1 and P2 (see Figure 6) far away from the reactor.

The RWTP yard was included into the monitoring area in 1997. The soil and vegetation samples were taken at points P1 to P13. The results are given in Table XIII for the vegetation and soil samples respectively. A decrease of Cs presence can be observed for the spent fuel storage area, which is due the use of the venting. A serious contamination with Cs (about 1800 Bq/kg) was identified in the RWTP yard (point P11). At points P11 and P13 a significant contamination with ^{60}Co (up to 6000 Bq/kg) was found. In this area ^{241}Am at a level of 1240 Bq/kg is also present.

The vegetation samples, taken in 1997, identified the presence of ^{137}Cs at points P8 and P9, located near the access gate to the RWTP. In the historical storage areas for wastes the vegetation is contaminated with ^{60}Co, the results of measurements being in good agreement with those on the soil samples [18].

In March 1998 soil samples for 9 points in reactor area were collected. The results are given in Table XIV. There are no significant values, except for points P6 and P7 (near the spent fuel pond).

During the summer of 1998 samples have been collected in a wider area, determined on the basis of results obtained in 1997. Additionally, the platform for dispatching the drums to the Baita-Bihor national repository was included into the sampling network.

The results from measurements on the soils are presented in Table XV. The presence of Cs at point P10 indicates a contamination in the RWTP area. The results for points P15, P16, P17 located in the area of drum transfer into wagons show a 5 times increase of Cs contamination compared to average values on the site. In 1998 a reduction of Co contamination in the RWTP yard could be observed, which is due the removal of same historic wastes in this area. Point P10 remains a “hot spot” (1000 Bq/kg).

In Table XVI the results of measurements on the vegetation collected in the same places are presented. One can be observe an ^{131}I contamination at all points for this period, which was due the radioactive iodine production in the CPR. ^{60}Co is present in the RWTP yard, the results being similar to those for the soils [19].

In 1999, the soil samples have been taken at all points previously established. These results are presented in Table XVII. Examining the results, one can observe that the radioactivity in the reactor area was not significantly modified. But in the RWTP zones have arisen with high contamination, particularly at points P11, P13 and P14. In this area old drums containing historical wastes have been recovered. The drums were in an advanced state of corrosion, allowing the spread of contaminants. At point P11 the contamination with ^{60}Co is high: 22.000 Bq/kg. Also, the values determined for ^{137}Cs and ^{241}Am are very high, 2600 and 1800 Bq/kg respectively. For point P13, the radioactive inventory is 1400 Bq/kg ^{60}Co, 3600 Bq/kg ^{241}Am, and 1400 Bq/kg^{137}Cs. At sampling point P14, near the reactor filter storage, the Co and Cs content also is significant.

In Table XVIII the results of dose and β-, and γ-contamination measurements on the asphalt surfacing along the waste and spent fuel transport routes are presented. Table XIX presents the results of dose rate measurements performed on the vegetation along the green verges of the roads. The measuring points are indicated in Figures 4 and 5.

Examining the results from the measurements, one can see that the maximum value of contamination is about 0.19 Bq/cm^2, the minimum value being 0.05 Bq/cm^2. The point of maximum contamination is No.1, corresponding to the access into Experimental Building, near Reactor.

Referring to dose values, these are relatively low in this area. The minimum value was been determined for point No.14, on the access way to the reactor ramp: 0.02 μSv/h. The point with the highest dose value is point No.75, near the RWTP gate and close to the access way: 0,3°μSv/h.

5.2. Magurele Fort area

The characterization of the radioactive contamination in the temporary waste storage Magurele Fort began in 1998. Preliminary results have been reported in June 1998 in a progress report [12]. The radiological assessment of the area was performed by the measurement of doses, β-, and γ-contaminations, γ-spectra, and the depth distribution of contamination.

The dose rates in some places outside of the Magurele Fort were in the range of 0.9 – 4 mR/h.

Sample were taken at three locations outside the facility: at one point at the main gate, and at two points in the contaminated wood storage area. At all three points samples were taken to depths of up to 9 cm. Table XX are summarizes the results of these measurements. A ^{60}Co-contamination is present in all samples. At point P3M, on the access way, there is an additional contamination by ^{137}Cs.

In July 1998, a new survey of contaminants in soil was performed in the neighborhood of the Fort. The results are presented in Table XXI. The main contaminant is ^{60}Co. The maximum depth of sampling was 35 cm, where ^{60}Co was found. The measuring grid for doses and contamination is outlined in Table XXII. The maximum β-, and γ-contamination is 5 Bq/cm^2. The maximum dose at the same location is $4.8 \cdot 10^{-3}$ mSv/h (point 1C).

In situ γ-spectrometry was been performed at the point 0C, together with soil sampling, and at the second point at the main gate of the Fort.

The γ-flux (per square centimeter and per second) was determined using the method described above [15]. The measured values of efficiency (*vs.* solid angle) have been used to determine the required parameters for calculations. The results are presented in Table XXIII as a function of the relaxation factors AS mentioned in the table. The γ-flux values for ^{60}Co, at the first point, are in the range of 7.5 to 39 particles s^{-1}cm^{-2}, depending on relaxation parameters. Comparing these values with those obtained in the reference [20], measured in 1993 for three point in the same area, one observe a good agreement. The values reported in reference [20] are in the range of 25 to 56 particles $s^{-1}cm^{-2}$.

6. OTHER ACTIVITIES

QA system. For environmental monitoring of the Magurele site and its influence zone, the QA system of the Institute was extended to the work performed under this research contract.

Public acceptance. Some activities to increase public acceptance [21] have been developed during the course of this IAEA CRP project. Based on the results obtained from the radiological characterization of the Magurele Fort area a public information sheet was produced. Another one was prepared referring to the decommissioning of the VVR-S research reactor and its environmental impact. A TV presentation on spent fuel management was also prepared, in order to explain the associated problems to the public.

International intercomparison exercises. Based on the practices developed under this project, an intercomparison between the IFIN-HH Magurele and VKTA Rossendorf (Germany) sites is in progress.

7. CONCLUSIONS

(1) The work performed in the framework of the coordinated research programme led to the development of specific methods for the investigation of radioactive contamination in the influence zone of IFIN-HH. The activities under this contract are complementary to the current monitoring programme at the site, which focused on nuclear fuel handling and waste disposal areas.

(2) By comparing the radiological characterization results obtained before and after the permanent shut down of the reactor, one can observe that the operation of this facility did not contribute significantly to the environmental contamination. The radioactive release during the operation was less than 1000 Ci per year. The most important part of the inventory released were gaseous effluents with short half-lives.

(3) The zone including reactor area and spent fuel storage site is not highly contaminated. Cs appeared in soil and vegetation as a result of the diffusion when the ventilation in storage was shut down.

(4) The RWTP area is highly contaminated at places, due to the past storage and processing of wastes. The soil and vegetation show different degrees of contamination, depending on the management of the wastes within the plant and outside. Hot spots with Cs, Am and Co contamination were found.

(5) The Magurele Fort and its surroundings showed a level of radioactive contamination requiring a site restoration programme.

Text cont. on page 126.

TABLE I. ANNUAL WIND FREQUENCY DISTRIBUTION AT THE MAGURELE SITE

Direction	Annual average wind frequency (%)
NE	22.3
E	14.6
SW	14.8
W	14.6

TABLE II. LAND USE IN THE MAGURELE DISTRICT

Land use categories	Surface area [km^2]	%
Agricultural (cereals, vineyards, orchards, fruits, pasture-lands)	36.57	84.0
Forest, water surfaces, entertainment facilities, roads	5.34	12.5
Dwellings	1.48	3.5
Total village land	43.39	100.0

TABLE III. CALCULATED RADIOACTIVE INVENTORY OF THE VVR-S RESEARCH REACTOR AS OF 31.12.1999

Activated material	Activity	Mass
Aluminium	37 Ci	3670 kg
Steel	1622 Ci	84250 kg
Concrete	229 Ci	60250 kg
Total	1900 Ci	152000 kg
Main isotopes ^{60}Co, ^{63}Ni		
Contaminated material		**Mass/Volume**
Primary circuit		47000 kg
Hot cells		3130 kg
Reactor Hall		up to 30 tones of different materials
Contaminated concrete		20-50 m^3
Secondary wastes		**Volume**
Liquids		120 m^3
Ion exchange resins		1-3 m^3

TABLE IV. RADIOACTIVE INVENTORY OF SPENT FUEL FROM THE MAGURELE RESEARCH REACTOR AS OF 31.12.1999.

Fissile materials in spent fuel

Nuclide	**Mass**
^{235}U	10.5 kg
^{238}U	178.0 kg
^{239}Pu	0.251 kg
^{240}Pu	0.0989 kg
^{241}Pu	0.0032 kg

Radioactivity

Fuel Assembly	**Total Activity [Ci]**	**^{90}Sr - ^{90}Y [Ci]**	**^{137}Cs - ^{137m}Ba [Ci]**	**^{85}Kr [Ci]**	**^{3}He [Ci]**
EK-10	$5.20 \cdot 10^4$	$7.14 \cdot 10^3$	$7.67 \cdot 10^3$	447	16
C-36	$5.49 \cdot 10^4$	$8.21 \cdot 10^3$	$8.08 \cdot 10^3$	789	24
Total	**$10.69 \cdot 10^4$**				

TABLE V. MONITORING POINTS AND MATERIALS SAMPLED

Point	**Description of place**	**Sample**	**Sector**
A	1st channel of Reactor	- water - sediment - vegetation - soil	S
B	2nd channel IFA	- water	S
C	Ciorogirla river - upstream	- surface water - sediment	SSW
D	Ciorogirla river - downstream	- surface water	ESE
E	Used domestic water plant	- water - sediment	WSW
F	ICAB plant	- water - sediment	S
G	1st group IFIN-HH	- drinking water	WSW
H	2nd group IFA	- drinking water	
I	Village fountains	- water piping in 1st channel - Reactor (ground water) - water piping in 2nd channel - IFA (ground water)	ESE ESE
J	Surveillance bore holes	ground water in the Reactor area	
K	IFA park	- soil - vegetation - sediment	WSW
L	Helen subcritical assembly	- soil - vegetation	WSW
	Bucharest-Titan (background)	- soil - vegetation	ENE
M	Forest	- soil - vegetation	WSW
	Village (channels area)	- soil - vegetables - cereals - fruits - milk	S
N	HV supply	- soil - vegetation	WNW
O	Pumping station	- soil - vegetation	NW
P	Mechanical workshop	- soil - vegetation	N
R	Lab. No. 8	- air	NW

TABLE VI. MONITORING FREQUENCY

Nuclide	$T_{1/2}$	Sample	Average residence time	Average time for exposure route	Frequency
Tritium	12.3 years	air	minutes	minutes	continuously
		vegetables	3 months	3 months	quarterly
		drinking water	5 days	5 days	weekly
		surface water	1 day	1 day	daily
		milk	5 days	5 days	weekly
Noble gases	2 days	air	minutes	minutes	continuously
^{131}I	8 days	air	minutes	minutes	continuously
		milk	5 days	4 days	weekly
^{134}Cs	2 years	milk	5 days	5 days	weekly
		air	minutes	minutes	continuously
		surface wastes	2–4 weeks	4 weeks	monthly
		drinkable water	2 months	2 months	bimonthly
		sediment	2 months	2 months	bimonthly
^{90}Sr	30 years	milk	5 days	3 days	weekly

TABLE VII. SAMPLING LOCATIONS

Point	Sampling point
P1	Nuclear Medicine Lab
P2	Cyclotron building (left side)
P3	Reactor plateau (center)
P4	Reactor auto-gate (left)
P5	Reactor auto-gate (right)
P6	Spent fuel storage (left)
P7	Spent fuel storage (right)
P8	Reactor - RWTP route (5 m right from plant gate)
P9	Reactor - RWTP route (5 m left from plant gate)
P10	Waste pond (underground)
P11	Surface waste storage
P12	300 m^3 ponds
P13	Heating plant
P14	Reactor filter pond
P15	Electrical power supply (behind RPC)
P16	Railway platform (third label ?)
P17	Railway platform (first label ?)

TABLE VIII. SMART RADIATION MONITOR SRM-200, DETECTORS AND ACCESSORIES

MODEL NO.	**TYPE OF MEASUREMENT**	**USEFUL RANGE**
AC-3	Alpha Contamination	Background to 33,000 counts/s
HP-210	Beta-gamma Contamination, 2 inch window	Background to 66,000 counts/s
HP-260	Beta-gamma Contamination	Background to 66,000 counts/s
HP-270	Gamma Exposure or Exposure Rate	Background to 2500 mR/h
HP-290	Gamma Exposure or Exposure Rate	0.005 to 50 R/h
LEG-1	Low Energy Gamma or X Ray 20 to 70 keV	Background to 33,000 counts/s
SPA-3	High Sensitivity Gamma 40 keV to 1 MeV	Background to 25,000 counts/s
SPA-6	Medium Sensitivity Gamma 40 keV to 1 MeV	Background to 25,000 counts/s

TABLE IX. NOMAD-PLUS CHARACTERISTICS

Dimensions			
Crystal Diameter	58.6 mm		
Crystal Length	79.4 mm		
End Cap to Crystal	3 mm		
Absorbing Layers — Aluminum	1.27 mm		
- Inactive Germanium	0.70 mm		
High Voltage Bias			
Recommended Operation Bias, POSITIVE	2300 V		
Performance Specifications (measured at a nominal rate of 1000 counts/s)	**Warranted**	**Measured**	**Amplifier Time Constant**
Resolution (FWHM) at 1.33 MeV, ^{60}Co	2.00 keV	1.73 keV	6 µs
Peak-to-Compton Ratio, ^{60}Co	58	68.8	6 µs
Relative Efficiency at 1.33 MeV, ^{60}Co	40 %	42.5 %	6 µs
Peak Shape (FWTM/FWHM), ^{60}Co	1.98	1.87	6 µs
Peak Shape (FWFM/FWHM), ^{60}Co	2.98	2.54	6 µs
Resolution (FWHM) at 122 keV, ^{57}Co	1100 eV	766 eV	

TABLE X. CALIBRATION OF NOMAD: MINIMUM DETECTABLE ACTIVITY.

Nuclid	E(keV)	Yield	MDA (s^{-1})	MDA (Bq)
^{241}Am	59.54	0.359	0.435	56.45
^{137}Cs	661.66	0.8521	0.151	8.25
^{60}Co	1173.24	0.999	0.184	8.56
	1332.5	0.999	0.212	9.88
^{152}Eu	121.78	0.284	0.14	23.07
	344.28	0.266	0.129	22.63
	778.9	0.1297	0.132	47.36
	1408.03	0.208	0.155	34.78

TABLE XI. CALIBRATION OF NOMAD: ERRORS

Source type	Nuclide	Source label	Activity [Bq]	Uncertainty [%]	Measured activity [Bq]	σ (%)	ε (%)
point	^{152}Eu	7-713	7026	±3	7038	±1.91	+0.2
	^{152}Eu	7-669	5188	±3	5162	±1.99	-0.5
	^{137}Cs	6-675	57759	±3	60597	±1.91	+4.9
	^{60}Co	7-510	2625	±1.5	2599	±1.91	-1.0
volume	^{152}Eu	7-660	6500	±3	6579	±2	+1.2
	^{152}Eu	7-644	3500	±5	3631	±2	+3.7
	^{137}Cs	8-309	2695	±5	2764	±2	+2.6
	^{60}Co	8-310	1910	±5	1952	±2	+2.2

TABLE XII. SPECIFIC ACTIVITY [Bq/kg] OF SELECTED ISOTOPES IN VEGETATION (TOP) AND SOIL (BOTTOM) SAMPLES TAKEN IN 1996.

Sample	**96veg1**	**96veg2**	**96veg3**	**96veg4**	**96veg5**	**96veg6**	**96veg7**	**96veg8**	**96veg9**
Mass [g]/ Isotope	61.18	54.75	60.85	56.25	52.55	56.89	60.99	61.00	60.88
^{7}Be	227.88	446.08	193.96	202.66	400.54	136.26	127.60	130.52	258.52
^{40}K	310.80	241.54	805.10	599.06	137.58	802.78	1152.30	905.86	320.68
^{134}Cs	26.18	40.58	22.34	25.76	24.00	15.38	13.68	14.76	13.22
^{137}Cs	6.04	10.86	3.96	4.56	3.60	7.32	13.12	6.46	7.44
^{212}Pb	0.00	27.38	0.00	3.62	10.18	0.00	0.00	0.00	0.98
^{214}Pb	28.40	70.46	19.52	16.44	57.50	45.14	32.70	42.04	52.92
^{131}I	5.64	6.20	4.52	7.60	0.00	0.00	0.00	6.50	3.86
^{214}Bi	1.18	1.02	0.00	0.00	0.98	0.84	0.88	1.20	1.28
^{228}Ac	1.18	12.98	59.50	1.32	6.88	0.00	0.00	0.00	2.06
TOTAL	**607.30**	**857.10**	**1108.90**	**861.02**	**641.26**	**1007.72**	**1340.28**	**1107.34**	**660.96**

Sample	**96sol1**	**96sol2**	**96sol3**	**96sol4**	**96sol5**	**96sol6**	**96sol7**	**96sol8**	**96sol9**
Mass [g]/ Isotope	216.13	215.52	196.50	206.40	195.11	195.00	193.05	203.35	193.62
^{40}K	830.78	822.32	930.60	916.98	903.38	862.44	756.56	914.16	815.04
^{134}Cs	7.22	6.04	6.72	7.94	6.76	12.54	9.74	7.86	6.00
^{137}Cs	200.10	150.36	195.04	244.60	169.88	493.36	322.54	214.84	182.96
^{212}Pb	54.16	57.90	67.92	63.78	74.06	60.04	40.17	63.48	56.22
^{214}Pb	53.52	50.40	57.74	59.50	60.54	51.38	51.60	60.36	48.30
^{212}Bi	33.84	29.00	40.54	31.54	36.26	29.56	18.54	38.62	33.66
^{214}Bi	40.02	34.40	39.46	40.34	43.02	35.60	33.14	43.80	30.00
^{226}Ra	167.08	149.74	210.72	186.76	213.98	195.74	166.66	198.44	185.68
^{235}U	122.80	141.60	152.16	111.92	194.46	167.58	88.38	143.10	161.46
^{208}Tl	18.88	17.72	19.64	18.02	20.92	18.88	10.46	20.46	17.04
^{228}Ac	52.46	41.92	50.06	48.14	52.60	44.28	25.34	48.78	40.60
TOTAL	**1580.86**	**1501.42**	**1770.60**	**1729.52**	**1775.86**	**1971.40**	**1523.13**	**1753.90**	**1576.96**

TABLE XIII. SPECIFIC ACTIVITY (Bq/kg) OF SELECTED ISOTOPES IN VEGETATION (TOP) AND SOIL (BOTTOM) SAMPLES TAKEN IN 1997.

Sample	**97 veg1**	**97 veg2**	**97 veg3**	**97 veg4**	**97 veg5**	**97 veg6**	**97 veg7**	**97 veg8**	**97 veg9**	**97 veg10**	**97 veg11**	**97 veg12**	**97 veg13**
Mass [g] Isotope	84.750	89.300	81.200	53.800	63.370	77.420	67.800	62.550	61.600	89.090	110.280	100.600	62.410
^{7}Be	26.754	24.721	0.000	34.063	30.753	32.625	52.956	0.000	19.824	162.513	179.906	149.605	146.678
^{22}Na	0.000	0.000	0.000	0.000	0.000	2.301	0.000	0.000	0.000	0.000	0.000	2.549	0.000
^{40}Ka	979.00	1246.84	1246.96	611.13	743.72	1118.66	622.08	1859.23	1707.14	727.30	679.15	1033.5	525.55
^{241}Am	0.000	0.000	0.000	14.978	6.714	0.000	0.000	0.000	0.000	0.000	56.118	0.000	23.174
^{60}Co	0.000	0.000	0.000	0.000	0.000	0.000	0.000	14.510	14.120	232.236	184.295	8.118	66.631
^{134}Cs	0.000	0.000	0.000	0.000	0.000	0.000	0.000	9.589	13.116	5.982	0.000	0.000	3.457
^{137}Cs	0.000	0.000	0.000	0.000	0.000	0.000	0.000	603.894	871.847	10.749	4.932	0.000	22.277
^{208}Tl	0.000	0.000	0.000	0.000	3.968	48.838	39.398	92.608	90.957	48.425	53.746	50.785	41.971
^{212}Pb	20.605	21.061	18.982	14.539	14.136	15.257	14.985	145.428	146.419	19.445	46.265	17.003	0.000
^{214}Pb	0.000	0.000	0.000	0.000	14.835	0.000	9.994	39.655	46.404	0.000	0.000	0.000	0.000
^{212}Bi	0.000	0.000	0.000	0.000	0.000	0.000	0.000	64.927	70.710	0.000	0.000	0.000	0.000
^{214}Bi	12.894	0.000	0.000	7.553	20.350	9.912	0.000	34.441	50.492	16.826	7.811	0.000	0.000
^{228}Ac	9.005	0.000	0.000	0.000	0.000	0.000	0.000	110.923	101.523	14.136	35.351	0.000	0.000
TOTAL	**1048.26**	**1292.63**	**1265.94**	**682.26**	**834.47**	**1227.59**	**739.41**	**2975.21**	**3132.55**	**1237.62**	**1247.58**	**1261.56**	**829.73**

Sample	**97 sol1**	**97 sol2**	**97 sol3**	**97 sol4**	**97 sol5**	**97 sol6**	**97 sol7**	**97 sol8**	**97 sol9**	**97 sol10**	**97 sol11**	**97 sol12**	**97 sol13**
Mass [g]/ Isotope	219.470	215.100	220.020	225.280	224.870	217.000	223.630	213.400	213.050	233.000	236.500	219.220	198.600
^{7}Be	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	7.655	0.000	51.907	0.000	0.000
^{22}Na	0.000	0.000	0.000	0.000	0.000	1.048	0.000	0.000	0.000	0.000	0.000	0.000	1.536
^{40}Ka	779.286	753.360	780.790	787.260	813.004	759.147	710.940	717.957	659.224	748.667	485.716	707.432	575.341
^{241}Am	0.000	0.000	0.000	0.000	0.000	0.000	13.036	0.000	0.000	0.000	1240.35	20.094	146.257
^{60}Co	1.909	0.000	0.000	3.809	3.991	0.000	4.374	5.604	5.454	179.127	5868.23	30.546	836.652
^{65}Zn	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	26.227	0.000	0.000
^{134}Cs	2.201	2.802	2.912	2.766	2.629	3.413	3.960	3.704	5.067	3.759	419.697	4.780	29.325
^{137}Cs	158.108	203.978	188.140	177.031	182.872	211.915	278.002	233.198	336.670	242.211	1800.93	203.850	457.192
^{208}Tl	36.789	35.950	39.363	38.725	39.532	38.178	32.943	35.763	35.126	35.946	192.564	38.894	34.109
^{212}Pb	65.503	61.070	68.656	64.578	67.567	59.443	50.203	56.158	56.541	61.621	602.041	68.811	53.871
^{214}Pb	34.451	35.517	18.413	27.557	24.204	16.772	24.304	15.314	17.920	18.659	202.966	41.828	88.213
^{212}Bi	31.608	25.963	34.816	36.442	35.841	29.530	24.541	25.074	27.307	27.958	335.558	44.129	30.100
^{214}Bi	35.016	31.991	22.226	29.936	28.441	22.217	26.505	13.300	19.497	21.256	184.554	34.615	84.526
^{228}Ac	45.564	45.446	51.556	52.139	60.049	48.777	37.800	42.835	39.204	52.732	498.747	52.891	39.673
TOTAL	**1190.44**	**1196.08**	**1206.87**	**1220.24**	**1258.13**	**1190.44**	**1206.61**	**1148.91**	**1209.66**	**1391.94**	**11909.5**	**1247.87**	**2376.79**

TABLE XIV. SPECIFIC ACTIVITY (Bq/kg) FOR SELECTED ISOTOPES IN SOIL SAMPLES TAKEN 1998 IN THE REACTOR AREA.

Sample	**98sol1**	**98sol2**	**98sol3**	**98sol4**	**98sol5**	**98sol6**	**98sol7**	**98sol8**	**98sol9**
Mass [g] Isotope	195.15	186.30	175.40	193.95	182.50	185.70	178.10	193.85	168.80
^{40}K	701.97	585.88	697.15	607.89	2512.33	683.25	503.77	631.73	666.41
^{51}Cr	0.00	0.00	0.00	0.00	0.00	12.39	0.00	0.00	0.00
^{54}Mn	0.00	0.00	0.00	0.00	2.58	0.00	0.00	0.00	0.00
^{60}Co	0.00	1.87	2.26	4.44	61.67	0.00	7.20	3.27	18.85
^{65}Zn	0.00	0.00	0.00	0.00	4.93	0.00	0.00	0.00	0.00
^{69}Zn	0.00	1.22	1.90	1.27	0.00	0.00	0.00	0.00	0.00
^{134}Cs	0.00	2.23	4.22	2.76	5.08	3.74	5.43	1.73	4.94
^{137}Cs	129.39	152.82	287.27	193.16	187.63	267.98	379.89	108.88	370.95
^{208}Tl	19.09	16.77	19.13	12.32	45.69	17.07	12.10	14.07	16.94
^{212}Pb	60.68	55.89	64.26	38.04	121.82	54.02	36.59	44.24	52.32
^{214}Pb	40.46	41.06	44.47	35.71	127.23	45.93	33.82	33.94	31.65
^{212}Bi	29.54	30.53	44.13	21.53	93.68	34.87	18.65	25.50	22.17
^{214}Bi	37.96	37.82	38.71	31.57	132.29	44.32	29.10	31.01	25.68
^{224}Ra	79.12	53.43	63.24	0.00	104.48	63.28	38.88	34.00	65.59
^{226}Ra	64.90	80.83	93.59	97.67	204.83	47.88	45.87	58.66	94.37
^{228}Ac	54.02	46.61	55.92	32.26	131.15	46.14	32.06	39.88	43.05
^{235}U	0.00	0.00	0.00	0.00	2.21	0.00	0.00	0.00	0.00
TOTAL	**1217.14**	**1106.95**	**1416.26**	**1078.65**	**3737.59**	**1320.88**	**1143.37**	**1026.91**	**1412.92**

TABLE XV. SPECIFIC ACTIVITY (Bq/kg) FOR SELECTED ISOTOPES IN SOIL SAMPLES TAKEN IN 1998.

Sample	**298sol1**	**298sol2**	**298sol3**	**298sol4**	**298sol5**	**298sol6**	**298sol7**	**298sol8**	**298sol9**
Mass (g)/ Isotope	177.10	183.50	181.30	202.80	194.15	186.05	186.90	177.20	190.60
^{40}K	665.95	708.72	708.00	732.89	762.55	479.46	691.65	608.41	629.80
^{241}Am	0.00	0.00	16.79	0.00	0.00	0.00	0.00	0.00	0.00
^{60}Co	0.00	0.00	0.00	0.00	4.02	0.00	0.00	3.99	0.00
^{69}Zn	0.00	0.00	0.00	0.00	0.00	14.24	0.00	0.00	0.00
^{134}Cs	2.22	0.00	0.00	0.00	0.00	0.00	2.57	0.00	0.00
^{137}Cs	252.06	184.51	195.76	104.64	131.47	262.82	281.42	211.35	62.42
^{208}Tl	17.23	19.49	18.38	17.91	18.12	10.11	16.75	13.69	18.41
^{212}Pb	59.57	65.72	63.61	63.13	66.68	40.00	57.46	47.61	55.83
^{214}Pb	12.73	23.99	7.17	16.10	16.14	0.00	13.34	9.42	9.14
^{212}Bi	32.43	40.80	43.73	33.57	35.25	21.24	33.00	25.96	36.41
^{214}Bi	15.36	21.54	9.28	13.41	13.66	0.00	12.28	6.17	8.47
^{224}Ra	63.62	63.32	51.16	60.01	66.35	0.00	66.61	58.02	57.38
^{226}Ra	95.08	65.81	86.12	111.66	90.60	0.00	114.55	36.66	54.53
^{228}Ac	50.06	57.49	54.92	49.02	54.03	35.38	49.98	43.22	47.34
^{235}U	0.00	0.00	0.00	0.00	0.00	0.00	0.00	2.61	0.00
TOTAL	**1266.31**	**1251.39**	**1254.91**	**1202.34**	**1258.86**	**863.25**	**1339.61**	**1067.13**	**979.73**

Sample	**298sol10**	**298sol11**	**298sol12**	**298sol13**	**298sol14**	**298sol15**	**298sol16**	**298sol17**	**298sol18**
Mass (g)/ Isotope	174.85	182.80	200.75	189.55	184.90	189.00	224.68	218.45	208.80
^{7}Be	0.00	0.00	0.00	0.00	0.00	11.69	18.86	0.00	13.18
^{40}K	612.24	605.69	638.31	692.53	684.59	396.82	279.17	294.47	421.09
^{241}Am	0.00	135.96	0.00	0.00	0.00	0.00	23.09	0.00	613.84
^{60}Co	1023.05	1551.75	5.12	17.21	38.98	0.00	826.73	141.73	579.93
^{65}Zn	0.00	10.39	0.00	0.00	0.00	0.00	0.00	0.00	0.00
^{69}Zn	0.00	0.00	0.00	0.00	0.76	1.61	0.00	0.00	0.00
^{134}Cs	81.65	34.41	3.07	3.70	1.51	5.81	127.80	15.62	89.53
^{137}Cs	497.37	239.38	247.07	201.32	107.58	610.74	1086.66	1079.97	895.45
^{208}Tl	13.83	22.35	15.08	18.24	17.60	7.99	5.97	4.32	7.76
^{212}Pb	49.80	82.80	57.29	61.32	59.61	22.24	21.72	17.70	23.89
^{214}Pb	143.63	28.36	10.46	152.29	29.38	0.00	11.97	0.00	0.00
^{207}Bi	0.00	0.00	0.00	0.00	0.00	0.00	2.42	0.00	0.00
^{212}Bi	35.09	48.13	35.10	39.51	32.55	13.26	18.77	9.83	26.42
^{214}Bi	129.04	26.11	9.82	130.69	28.19	0.00	12.09	0.00	0.00
^{224}Ra	0.00	78.34	65.81	0.00	59.62	0.00	0.00	0.00	0.00
^{226}Ra	291.84	240.26	87.28	248.59	126.44	81.58	430.15	112.53	0.00
^{228}Ac	47.92	64.01	48.64	53.61	49.23	21.13	12.29	19.02	20.80
^{110m}Ag	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	8.87
^{235}U	0.00	0.00	0.00	0.00	0.00	0.00	19.73	3.89	0.00
TOTAL	**2925.45**	**3167.94**	**1223.05**	**1619.03**	**1236.07**	**1161.18**	**2878.55**	**1699.09**	**2687.59**

TABLE XVI. SPECIFIC ACTIVITY (Bq/kg) FOR SELECTED ISOTOPES IN VEGETATION SAMPLES TAKEN IN 1998.

Sample	**98veg1**	**98veg2**	**98veg3**	**98veg4**	**98veg5**	**98veg6**	**98veg7**	**98veg8**	**98veg9**
Mass (g)/ Isotope	67.02	75.30	73.70	59.05	48.77	62.78	61.08	61.22	68.62
^{7}Be	123.98	117.18	142.23	392.92	356.88	219.59	110.40	162.11	196.08
^{40}K	628.11	410.31	482.25	664.84	443.53	590.68	532.91	675.30	624.88
^{60}Co	0.00	0.00	0.00	0.00	0.00	4.87	0.00	2.28	0.00
^{69}Zn	2.04	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
^{131}J	111.07	138.79	14.98	3.55	7.71	20.75	19.72	25.53	14.05
^{137}Cs	0.00	0.00	0.00	0.00	0.00	8.68	0.00	13.01	0.00
^{224}Ra	0.00	0.00	88.53	0.00	0.00	0.00	0.00	0.00	0.00
^{228}Ac	0.00	0.00	14.22	0.00	0.00	0.00	0.00	0.00	0.00
^{235}U	6.37	7.26	15.76	0.00	0.00	6.52	0.00	0.00	0.00
TOTAL	**871.58**	**673.54**	**757.96**	**1061.32**	**808.12**	**851.10**	**663.03**	**878.24**	**835.01**

Sample	98veg10	98veg11	98veg12	98veg13	98veg14	98veg15	98veg16	98veg17	
Mass (g)/ Isotope	66.97	84.50	81.15	64.50	61.12	63.82	62.45	56.45	
^{7}Be	78.38	79.92	65.37	118.33	110.08	165.83	127.93	144.76	
^{40}K	550.34	659.91	831.69	675.27	1889.89	795.93	806.58	1435.77	
^{51}Cr	0.00	0.00	0.00	0.00	0.00	0.00	31.40	0.00	
^{60}Co	38.20	88.25	0.00	6.73	46.01	0.00	14.65	0.00	
^{69}Zn	0.00	0.00	0.00	0.00	2.12	0.00	0.00	0.00	
^{131}J	113.47	65.64	40.57	52.32	47.11	18.69	8.68	11.85	
^{134}CS	0.00	0.00	0.00	3.16	0.00	0.00	0.00	0.00	
^{137}Cs	31.91	0.00	0.00	3.43	22.33	12.52	40.90	38.08	
^{214}Pb	0.00	0.00	0.00	21.97	113.40	0.00	0.00	0.00	
^{214}Bi	0.00	0.00	0.00	22.80	114.09	0.00	0.00	0.00	
^{226}Ra	0.00	0.00	0.00	0.00	192.06	0.00	0.00	0.00	
^{228}Ac	0.00	16.88	0.00	0.00	0.00	0.00	19.65	0.00	
TOTAL	**812.30**	**910.59**	**937.64**	**904.00**	**2537.08**	**992.97**	**1049.79**	**1630.46**	

TABLE XVII. SPECIFIC ACTIVITY (Bq/kg) FOR SELECTED ISOTOPES IN SOIL SAMPLES TAKEN IN 1999.

Sample	**99sol1**	**99sol2**	**99sol3**	**99sol4**	**99sol5**	**99sol6**	**99sol7**	**99sol8**	**99sol9**
Mass [g]/ Isotope	166.50	171.90	161.98	206.35	188.23	180.40	173.20	178.50	154.00
^{7}Be	49.50	45.97	25.09	0.00	0.00	0.00	46.42	11.27	0.00
^{40}K	692.07	685.95	693.27	778.74	848.89	582.89	694.05	688.65	637.78
^{134}Cs	0.00	0.00	0.00	0.00	0.00	3.22	0.00	0.00	0.00
^{137}Cs	162.11	186.85	234.11	195.58	196.67	357.70	210.95	79.60	234.42
^{208}Tl	18.18	18.17	18.65	21.29	21.94	13.05	18.22	19.03	15.35
^{212}Pb	63.29	69.81	67.28	69.71	73.75	46.66	63.02	62.49	51.84
^{214}Pb	20.67	23.98	43.13	32.13	53.27	32.81	55.32	46.76	35.04
^{212}Bi	35.71	34.40	36.99	40.08	46.17	25.54	33.76	34.50	34.25
^{214}Bi	14.58	19.08	41.74	29.63	51.10	27.81	50.86	40.34	29.89
^{224}Ra	109.14	68.56	80.88	78.47	82.70	67.81	49.86	77.11	65.33
^{226}Ra	96.92	76.62	127.25	120.07	148.04	59.24	107.80	161.02	124.89
^{228}Ac	50.88	55.54	54.56	61.17	64.13	37.11	48.35	50.96	38.30
TOTAL	**1313.05**	**1284.95**	**1422.94**	**1426.88**	**1586.66**	**1253.85**	**1378.62**	**1271.72**	**1267.09**

Sample	**99sol10**	**99sol11**	**99sol12**	**99sol13**	**99sol14**	**99sol15**	**99sol16**	**99sol17**	
Mass [g]/ Isotope	173.60	186.00	130.85	160.80	151.60	153.90	199.30	170.70	
^{40}K	596.60	369.90	706.80	424.10	300.01	419.02	380.82	343.12	
^{60}Co	92.87	22668.20	45.53	1390.32	337.66	3.84	0.00	0.00	
^{241}Am	0.00	1841.30	41.81	3576.96	50.97	0.00	0.00	0.00	
^{134}Cs	0.00	148.26	0.00	31.48	7.66	0.00	0.00	3.19	
^{137}Cs	280.06	2645.97	171.49	1435.37	725.17	325.46	56.74	439.75	
^{208}Tl	16.27	65.07	20.59	43.47	5.78	10.32	7.75	6.04	
^{212}Pb	54.55	238.20	64.64	149.11	23.59	34.14	20.16	23.66	
^{214}Pb	46.70	265.75	61.23	377.02	20.25	21.40	18.94	23.18	
^{212}Bi	36.05	141.29	38.93	86.65	0.00	22.29	0.00	12.59	
^{214}Bi	40.82	232.74	51.81	327.32	16.39	22.01	17.10	23.01	
^{224}Ra	56.75	278.23	56.82	108.92	49.40	0.00	0.00	36.33	
^{226}Ra	89.75	0.00	129.47	822.29	0.00	55.95	0.00	39.54	
^{228}Ac	45.26	174.61	55.70	128.51	20.56	31.07	19.54	17.74	
^{235}U	0.00	210.15	0.00	0.00	2.84	0.00	0.00	0.00	
TOTAL	**1355.69**	**29279.67**	**1444.82**	**8901.53**	**1560.27**	**945.50**	**521.05**	**968.16**	

TABLE XVIII. RESULTS OF DOSE AND β-, AND γ-CONTAMINATION MEASUREMENTS ON THE ASPHALT SURFACING ALONG THE WASTE AND SPENT FUEL TRANSPORT ROUTES

Point	HP 210L	HP 270	Contamination	Dose rate	
No.	β+γ [cnt/min]	γ [mR/min]	[Bq/cm^2]	[mR/h]	[mSv/h]
1	66.70	$1.62\cdot10^{-4}$	0.1859	0.0097	$8.52\cdot10^{-5}$
2	28.60	$2.92\cdot10^{-4}$	0.0797	0.0175	$1.54\cdot10^{-4}$
3	24.10	$2.92\cdot10^{-4}$	0.0672	0.0175	$1.54\cdot10^{-4}$
4	31.10	$9.75\cdot10^{-5}$	0.0867	0.0059	$5.13\cdot10^{-5}$
5	31.10	$2.76\cdot10^{-4}$	0.0867	0.0166	$1.45\cdot10^{-4}$
6	25.10	$2.27\cdot10^{-4}$	0.0700	0.0136	$1.19\cdot10^{-4}$
7	43.60	$2.44\cdot10^{-4}$	0.1215	0.0146	$1.28\cdot10^{-4}$
8	25.10	$2.27\cdot10^{-4}$	0.0700	0.0136	$1.19\cdot10^{-4}$
9	25.60	$2.44\cdot10^{-4}$	0.0714	0.0146	$1.28\cdot10^{-4}$
10	31.60	$2.76\cdot10^{-4}$	0.0881	0.0166	$1.45\cdot10^{-4}$
11	28.10	$8.12\cdot10^{-5}$	0.0783	0.0049	$4.27\cdot10^{-5}$
12	28.10	$2.76\cdot10^{-4}$	0.0783	0.0166	$1.45\cdot10^{-4}$
13	28.60	$4.87\cdot10^{-5}$	0.0797	0.0029	$2.56\cdot10^{-5}$
14	28.10	$3.25\cdot10^{-5}$	0.0783	0.0020	$1.71\cdot10^{-5}$
15	23.60	$1.79\cdot10^{-4}$	0.0658	0.0107	$9.42\cdot10^{-5}$
16	26.60	$2.27\cdot10^{-4}$	0.0741	0.0136	$1.19\cdot10^{-4}$
17	54.60	$1.95\cdot10^{-4}$	0.1522	0.0117	$1.03\cdot10^{-4}$
18	25.10	$2.44\cdot10^{-4}$	0.0700	0.0146	$1.28\cdot10^{-4}$
19	28.60	$2.60\cdot10^{-4}$	0.0797	0.0156	$1.37\cdot10^{-4}$
20	24.60	$1.95\cdot10^{-4}$	0.0686	0.0117	$1.03\cdot10^{-4}$
21	57.60	$1.14\cdot10^{-4}$	0.1606	0.0068	$6.00\cdot10^{-5}$
22	25.60	$1.95\cdot10^{-4}$	0.0714	0.0117	$1.03\cdot10^{-4}$
23	23.60	$1.79\cdot10^{-4}$	0.0658	0.0107	$9.42\cdot10^{-5}$
24	19.50	$1.79\cdot10^{-4}$	0.0544	0.0107	$9.42\cdot10^{-5}$
25	26.10	$2.92\cdot10^{-4}$	0.0728	0.0175	$1.54\cdot10^{-4}$
26	31.60	$3.41\cdot10^{-4}$	0.0881	0.0205	$1.79\cdot10^{-4}$
27	29.60	$1.79\cdot10^{-4}$	0.0825	0.0107	$9.42\cdot10^{-5}$
28	45.60	$4.06\cdot10^{-4}$	0.1271	0.0244	$2.14\cdot10^{-4}$
29	48.10	$2.92\cdot10^{-4}$	0.1341	0.0175	$1.54\cdot10^{-4}$
30	52.60	$2.27\cdot10^{-4}$	0.1466	0.0136	$1.19\cdot10^{-4}$
31	27.10	$2.27\cdot10^{-4}$	0.0755	0.0136	$1.19\cdot10^{-4}$
32	29.10	$1.95\cdot10^{-4}$	0.0811	0.0117	$1.03\cdot10^{-4}$
33	43.60	$1.14\cdot10^{-4}$	0.1215	0.0068	$6.00\cdot10^{-5}$
34	17.50	$2.27\cdot10^{-4}$	0.0488	0.0136	$1.19\cdot10^{-4}$
35	27.60	$2.44\cdot10^{-4}$	0.0769	0.0146	$1.28\cdot10^{-4}$
36	29.60	$2.44\cdot10^{-4}$	0.0825	0.0146	$1.28\cdot10^{-4}$
37	31.60	$1.79\cdot10^{-4}$	0.0881	0.0107	$9.42\cdot10^{-5}$
38	22.10	$3.25\cdot10^{-4}$	0.0616	0.0195	$1.71\cdot10^{-4}$
39	27.10	$2.44\cdot10^{-4}$	0.0755	0.0146	$1.28\cdot10^{-4}$
40	27.10	$2.76\cdot10^{-4}$	0.0755	0.0166	$1.45\cdot10^{-4}$
41	32.60	$3.57\cdot10^{-4}$	0.0909	0.0214	$1.88\cdot10^{-4}$
42	26.10	$3.09\cdot10^{-4}$	0.0728	0.0185	$1.63\cdot10^{-4}$
43	22.60	$3.57\cdot10^{-4}$	0.0630	0.0214	$1.88\cdot10^{-4}$
44	39.60	$2.92\cdot10^{-4}$	0.1104	0.0175	$1.54\cdot10^{-4}$
45	32.60	$1.79\cdot10^{-4}$	0.0909	0.0107	$9.42\cdot10^{-5}$
46	32.10	$2.60\cdot10^{-4}$	0.0895	0.0156	$1.37\cdot10^{-4}$
47	25.60	$1.30\cdot10^{-4}$	0.0714	0.0078	$6.84\cdot10^{-5}$
48	35.60	$2.76\cdot10^{-4}$	0.0992	0.0166	$1.45\cdot10^{-4}$

TABLE XIX. RESULTS OF DOSE AND γ-CONTAMINATION MEASUREMENTS ON THE GREEN VERGE ALONG THE WASTE AND SPENT FUEL TRANSPORT ROUTES

Point No.	**HP 270 γ [mR/min]**	**Dose rate [mR/h]**	**[mSv/h]**
49	$1.95 \cdot 10^{-4}$	0.0117	$1.03 \cdot 10^{-4}$
50	$2.76 \cdot 10^{-4}$	0.0166	$1.45 \cdot 10^{-4}$
51	$3.76 \cdot 10^{-4}$	0.0226	$1.98 \cdot 10^{-4}$
52	$1.79 \cdot 10^{-4}$	0.0107	$9.42 \cdot 10^{-5}$
53	$3.09 \cdot 10^{-4}$	0.0185	$1.63 \cdot 10^{-4}$
54	$1.95 \cdot 10^{-4}$	0.0117	$1.03 \cdot 10^{-4}$
55	$2.11 \cdot 10^{-4}$	0.0127	$1.11 \cdot 10^{-4}$
56	$1.79 \cdot 10^{-4}$	0.0107	$9.42 \cdot 10^{-5}$
57	$2.27 \cdot 10^{-4}$	0.0136	$1.19 \cdot 10^{-4}$
58	$2.92 \cdot 10^{-4}$	0.0175	$1.54 \cdot 10^{-4}$
59	$2.44 \cdot 10^{-4}$	0.0146	$1.28 \cdot 10^{-4}$
60	$3.41 \cdot 10^{-4}$	0.0205	$1.79 \cdot 10^{-4}$
61	$2.76 \cdot 10^{-4}$	0.0166	$1.45 \cdot 10^{-4}$
62	$2.44 \cdot 10^{-4}$	0.0146	$1.28 \cdot 10^{-4}$
63	$2.76 \cdot 10^{-4}$	0.0166	$1.45 \cdot 10^{-4}$
64	$3.09 \cdot 10^{-4}$	0.0185	$1.63 \cdot 10^{-4}$
65	$3.90 \cdot 10^{-4}$	0.0234	$2.^{-5} \cdot 10^{-4}$
66	$2.92 \cdot 10^{-4}$	0.0175	$1.54 \cdot 10^{-4}$
67	$3.09 \cdot 10^{-4}$	0.0185	$1.63 \cdot 10^{-4}$
68	$4.06 \cdot 10^{-4}$	0.0244	$2.14 \cdot 10^{-4}$
69	$2.76 \cdot 10^{-4}$	0.0166	$1.45 \cdot 10^{-4}$
70	$3.25 \cdot 10^{-4}$	0.0195	$1.71 \cdot 10^{-4}$
71	$1.79 \cdot 10^{-4}$	0.0107	$9.42 \cdot 10^{-5}$
72	$2.92 \cdot 10^{-4}$	0.0175	$1.54 \cdot 10^{-4}$
73	$3.25 \cdot 10^{-4}$	0.0195	$1.71 \cdot 10^{-4}$
74	$2.11 \cdot 10^{-4}$	0.0127	$1.11 \cdot 10^{-4}$
75	$5.85 \cdot 10^{-4}$	0.0351	$3.08 \cdot 10^{-4}$
76	$3.25 \cdot 10^{-4}$	0.0195	$1.71 \cdot 10^{-4}$
77	$2.11 \cdot 10^{-4}$	0.0127	$1.11 \cdot 10^{-4}$
78	$2.11 \cdot 10^{-4}$	0.0127	$1.11 \cdot 10^{-4}$
79	$3.74 \cdot 10^{-4}$	0.0224	$1.97 \cdot 10^{-4}$
P4	$2.11 \cdot 10^{-4}$	0.0127	$1.11 \cdot 10^{-4}$
P5	$1.46 \cdot 10^{-4}$	0.0088	$7.68 \cdot 10^{-5}$

TABLE XX. SPECIFIC ACTIVITY (Bq/kg) FOR SELECTED ISOTOPES IN SOIL SAMPLES TAKEN IN 1998 OUTSIDE OF THE MAGURELE FORT.

Sample	**98fortp1**	**98fortp2**	**98fortp3**	**98fortp4**	**98fortp5**	**98fortp6**	**98fortp7**	**98fortp8**	**98fortp9**
Depth [cm]	P1 (0-2)	P1 (2-4)	P1 (4-6)	P1 (6-8)	P2 (0-4)	P2 (4-8)	P3 (0-3)	P3 (3-6)	P3 (6-9)
Mass [g] **Isotope**	178.85	208.00	210.55	221.55	199.05	210.35	168.45	213.05	216.20
^{40}K	550.99	600.00	628.97	574.68	602.76	434.43	398.36	480.17	401.98
^{60}Co	2687.62	8527.88	7160.29	4086.62	4036.22	385.21	1355.24	662.38	477.06
^{65}Zn	11.11	17.92	25.89	9.59	0.00	0.00	0.00	0.00	0.00
^{69}Zn	2.83	0.00	0.00	0.00	0.00	1.68	0.00	0.00	0.00
^{137}Cs	186.03	203.06	86.39	62.51	124.06	50.25	2404.27	1098.15	583.16
^{208}Tl	9.55	12.21	11.67	10.83	13.87	7.26	17.84	13.74	12.86
^{212}Tl	43.69	51.56	46.07	43.39	54.94	28.29	58.99	47.25	44.36
^{214}Pb	11.21	36.13	30.97	41.45	47.28	42.77	57.86	48.73	40.88
^{212}Bi	0.00	0.00	0.00	19.35	0	13.74	37.82	28.58	26.53
^{214}Bi	8.82	22.91	21.06	36.73	53.22	34.44	50.85	49.97	35.86
^{224}Ra	79.45	97.16	81.40	61.35	0.00	0.00	0.00	0.00	0.00
^{226}Ra	0.00	0.00	104.78	53.22	0.00	0.00	89.16	77.41	49.52
^{228}Ac	29.82	46.39	41.63	38.11	0.00	23.23	50.91	41.28	40.77
^{235}U	3.59	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
TOTAL	**3624.71**	**9615.22**	**8239.12**	**5037.83**	**4932.35**	**1021.30**	**4521.31**	**2547.66**	**1712.98**

TABLE XXI. SPECIFIC ACTIVITY (Bq/kg) FOR SELECTED ISOTOPES IN SOIL SAMPLES TAKEN IN 1998 AT THE MAGURELE FORT.

Sample	**98foso1**	**98foso2**	**98foso3**	**98foso4**	**98foso5**	**98foso6**	**98foso7**	**98foso8**
Depth [cm]	0-2	2-4	4-6	6-8	8-10	10-12	12-14	14-16
Mass [g]/ Isotope	157.40	173.25	189.50	191.15	218.75	220.50	210.72	225.00
^{7}Be	22.96	0.00	0.00	0.00	0.00	0.00	0.00	0.00
^{40}K	678.65	639.94	657.36	738.90	705.97	723.90	669.70	670.27
^{60}Co	2061.82	3109.78	1920.42	1028.88	366.20	139.13	95.71	67.88
^{134}Cs	4.74	6.90	0.00	0.00	0.00	0.00	0.00	0.00
^{137}Cs	361.84	275.88	109.16	57.20	28.80	18.15	16.51	17.03
^{208}Tl	14.59	15.72	17.72	17.66	15.18	14.43	14.36	12.82
^{212}Pb	52.79	57.26	57.46	57.52	49.43	47.08	47.11	45.60
^{214}Pb	32.15	32.05	30.68	36.40	30.56	29.42	22.26	35.55
^{212}Bi	28.26	31.12	42.26	31.56	24.59	30.07	28.47	29.74
^{214}Bi	30.69	31.70	25.33	36.12	29.90	31.88	21.45	31.72
^{224}Ra	61.64	65.25	63.44	75.55	57.94	49.50	50.92	34.43
^{226}Ra	63.78	48.61	0.00	104.10	89.38	77.51	78.05	82.73
^{228}Ac	45.86	47.38	52.05	48.69	42.18	41.77	40.44	38.23
TOTAL	**3459.76**	**4361.59**	**2975.88**	**2232.57**	**1440.13**	**1202.85**	**1084.98**	**1066.00**
Sample	**98foso9**	**98foso10**	**98foso11**	**98foso12**	**98foso13**	**98foso14**	**98foso15**	
Depth [cm]	16-18 cm	18-20 cm	20-22 cm	22-25 cm	25-28 cm	28-31 cm	31-35 cm	
Mass [g]/ Isotope	207.45	204.30	191.00	228.60	231.85	214.70	172.35	
^{40}K	664.88	572.10	483.79	500.48	524.00	571.77	607.72	
^{60}Co	65.10	92.92	91.07	28.93	16.34	27.25	74.64	
^{137}Cs	16.92	44.65	36.98	12.11	4.82	2.68	5.54	
^{208}Tl	11.69	10.00	8.57	8.11	7.75	9.17	10.61	
^{212}Pb	39.30	37.56	29.39	23.46	25.44	31.10	45.15	
^{214}Pb	33.71	22.31	19.62	11.80	13.32	22.16	29.93	
^{212}Bi	25.73	23.96	16.70	14.55	12.86	16.20	21.76	
^{214}Bi	29.98	20.10	22.13	14.01	13.50	20.55	27.51	
^{224}Ra	41.88	37.73	29.55	0.00	0.00	0.00	64.11	
^{226}RA	40.06	70.60	0.00	0.00	0.00	0.00	66.71	
^{228}Ac	38.22	31.74	23.67	22.12	23.73	26.84	35.89	
TOTAL	**1007.48**	**963.67**	**761.47**	**635.57**	**641.78**	**727.73**	**989.57**	

TABLE XXII. RESULTS OF DOSE AND β-. AND γ-CONTAMINATION MEASUREMENTS AT THE MAGURELE FORT.

Point No.		**HP 210L** β+γ **(cnt/min)**	**HP 270** γ **(mR/min)**	**Contamination (Bq/cm^2)**	**Dose rate (mR/h)**	**(mSv/h)**
-4	A	29.10	$4.10\cdot10^{-4}$	0.0811	0.0246	$2.16\cdot10^{-4}$
	B	711.00	$7.55\cdot10^{-3}$	1.9819	0.4530	$3.97\cdot10^{-3}$
	C	252.00	$2.57\cdot10^{-3}$	0.7024	0.1542	$1.35\cdot10^{-3}$
	D	78.20	$1.80\cdot10^{-3}$	0.2180	0.1080	$9.47\cdot10^{-4}$
-3	A	38.60	$3.41\cdot10^{-4}$	0.1076	0.0205	$1.79\cdot10^{-4}$
	B	185.00	$1.66\cdot10^{-3}$	0.5157	0.0996	$8.73\cdot10^{-4}$
	C	219.00	$1.79\cdot10^{-3}$	0.6105	0.1074	$9.42\cdot10^{-4}$
	D	33.10	$3.90\cdot10^{-4}$	0.0923	0.0234	$2.05\cdot10^{-4}$
-2	A	35.10	$6.01\cdot10^{-4}$	0.0978	0.0361	$3.16\cdot10^{-4}$
	B	42.60	$6.82\cdot10^{-4}$	0.1187	0.0409	$3.59\cdot10^{-4}$
	C	42.10	$6.17\cdot10^{-4}$	0.1174	0.0370	$3.25\cdot10^{-4}$
	D	35.60	$5.20\cdot10^{-4}$	0.0992	0.0312	$2.74\cdot10^{-4}$
-1	A	37.10	$3.41\cdot10^{-3}$	0.1034	0.2046	$1.79\cdot10^{-3}$
	B	72.70	$6.50\cdot10^{-4}$	0.2026	0.0390	$3.42\cdot10^{-4}$
	C	48.60	$6.66\cdot10^{-4}$	0.1355	0.0400	$3.50\cdot10^{-4}$
	D	36.60	0.0	0.1020	0.0000	0.0
0	A	38.10	$5.85\cdot10^{-4}$	0.1062	0.0351	$3.08\cdot10^{-4}$
	B	270.00	$3.14\cdot10^{-3}$	0.7526	0.1884	$1.65\cdot10^{-3}$
	C	417.00	$6.04\cdot10^{-3}$	1.1624	0.3624	$3.18\cdot10^{-3}$
	D	46.60	$7.64\cdot10^{-4}$	0.1299	0.0458	$4.02\cdot10^{-4}$
1	A	31.60	$5.20\cdot10^{-4}$	0.0881	0.0312	$2.74\cdot10^{-4}$
	B	79.20	$1.93\cdot10^{-3}$	0.2208	0.1158	$1.02\cdot10^{-3}$
	C	1810.00	$9.16\cdot10^{-3}$	5.0453	0.5496	$4.82\cdot10^{-3}$
	D	37.60	$9.10\cdot10^{-4}$	0.1048	0.0546	$4.79\cdot10^{-4}$
2	A	31.60	$4.06\cdot10^{-4}$	0.0881	0.0244	$2.14\cdot10^{-4}$
	B	87.20	$1.27\cdot10^{-3}$	0.2431	0.0762	$6.68\cdot10^{-4}$
	C	56.60	$9.26\cdot10^{-4}$	0.1578	0.0556	$4.87\cdot10^{-4}$
	D	27.60	$4.06\cdot10^{-4}$	0.0769	0.0244	$2.14\cdot10^{-4}$
3	A	35.10	$3.41\cdot10^{-4}$	0.0978	0.0205	$1.79\cdot10^{-4}$
	B	39.10	$5.20\cdot10^{-4}$	0.1090	0.0312	$2.74\cdot10^{-4}$
	C	33.60	$5.69\cdot10^{-4}$	0.0937	0.0341	$2.99\cdot10^{-4}$
	D	28.10	$4.55\cdot10^{-4}$	0.0783	0.0273	$2.39\cdot10^{-4}$
4	A	43.60	$4.55\cdot10^{-4}$	0.1215	0.0273	$2.39\cdot10^{-4}$
	B	130.00	$1.82\cdot10^{-3}$	0.3624	0.1092	$9.58\cdot10^{-4}$
	C	41.60	$1.30\cdot10^{-3}$	0.1160	0.0780	$6.84\cdot10^{-4}$
	D	58.10	$8.29\cdot10^{-4}$	0.1620	0.0497	$4.36\cdot10^{-4}$

TABLE XXIII. RESULTS FROM *IN SITU* MEASUREMENTS AT MAGURELE FORT.

Point 1	Φ_γ [$s^{-1}cm^{-2}$]			
Relaxation factor	^{137}Cs (661.62 keV)	^{60}Co (1173.23 keV)	^{60}Co (1332.51 keV)	^{40}K (1460.75 keV)
$6.25 \cdot 10^{-2}$	1.5595	39.6257	38.6556	1.0018
$2.06 \cdot 10^{-1}$	0.7617	20.1946	19.6043	0.5556
$3.12 \cdot 10^{-1}$	0.6145	16.6602	16.2193	0.4189
$6.25 \cdot 10^{-1}$	0.4324	12.1438	12.0179	0.3138
$6.25 \cdot 10^{0}$	0.2636	7.5346	7.4594	0.1948

Point 2	Φ_γ [$s^{-1}cm^{-2}$]			
Relaxation factor	^{137}Cs (661.62 keV)	^{60}Co (1173.23 keV)	^{60}Co (1332.51 keV)	^{40}K (1460.75 keV)
$6.25 \cdot 10^{-2}$	2.1196	0.5609	0.5582	1.1694
$2.06 \cdot 10^{-1}$	1.0353	0.2858	0.2831	0.5901
$3.12 \cdot 10^{-1}$	0.8352	0.2358	0.2342	0.4889
$6.25 \cdot 10^{-1}$	0.5877	0.1719	0.1735	0.3663
$6.25 \cdot 10^{0}$	0.3583	0.1066	0.1077	0.2274

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SITE CHARACTERIZATION TECHNIQUES USED IN RESTORATION OF AGRICULTURAL AREAS ON THE TERRITORY OF THE RUSSIAN FEDERATION CONTAMINATED AFTER THE ACCIDENT AT THE CHERNOBYL NPP

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ABSTRACT

The experience gained in the aftermath after the heavy radiation accidents shows the need for improvement in site characterisation techniques and methodology in order to provide a link between site characterisation and the selection of restoration strategies. This paper gives an overview of the site characterisation techniques used in restoration of contaminated agricultural lands on the territory of the Russian Federation. The approach used for site characterisation and selection of restoration strategies is described. The main site specific factors influencing the choice of restoration options are identified. Data on the efficiency of major restoration measures used for the remediation of agricultural lands after the Chernobyl Nuclear Power Plant and the Kyshtym accidents are given. A description of the decision support system FORCON, designed to simplify the selection of restoration options, is presented.

1. INTRODUCTION

The accident at the Chernobyl Nuclear Power Plant (NPP), both in times of released activity and the area subjected to contamination, was the most serious ever to have occurred in the history of nuclear energy. Those countries most seriously affected include the republics of Russia, Belarus and the Ukraine, which are estimated to have received about 70% of the fallout [1]. As a result in the first time in history vast areas were heavily contaminated, and a large proportion of them require large scale restoration.

In this context, agricultural ecosystems are among those environments of considerable interest. It has been shown that in many cases the contribution from internal irradiation is comparable to that of external irradiation. Thus, in the first year after the accident at the Chernobyl NPP the contribution of internal irradiation to the total equivalent effective radiation dose for the population of the Confederation of Independent States (CIS) countries amounted to 45% (that of external to 52%), and for the life time dose these values were 39% and 60%, respectively [2]. In regions with high mobility of radionuclides in the soil, where the contribution of internal irradiation predominates, the restoration of agricultural lands should have priority in restoration policy. On the contrary, in such areas where external irradiation exceeds considerably the internal one, attention can be paid to the more costly options based on the decontamination of the territory.

The experience gained in the aftermath of the heavy radiation accidents shows that, in the case of large scale contamination, the limitation of internal radiation doses to people living in areas subject radioactive contamination by means of restoration of agricultural lands is economically more realistic, than to decrease dose burdens from external irradiation. Therefore, the problems connected with the optimal restoration strategies of agricultural land subject to radioactive contamination were of crucial importance from the point of view of providing safe living conditions for the population in the regions affected after the Chernobyl and Kyshtym accidents. A necessary component in the decision making for the selection of optimal restoration options is the site characterisation of agricultural areas and the evaluation of factors controlling the accumulation of radionuclides in agricultural products.

The objectives of this paper are to present the analysis of the site specific factors considered in selecting the remediation options in regions contaminated after the accident at the ChNPP and to give an overview of site characterisation techniques used in restoration of agricultural areas on the territory of the Russian Federation heavily contaminated by the radiation accidents.

2. APPROACH USED FOR THE SELECTION OF OPTIMAL RESTORATION OPTIONS FOR REMEDIATION OF AGRICULTURAL LANDS

The general objectives of the agricultural land restoration can be formulated as follows:

- To provide opportunities for the production of agricultural products which can be used without any restrictions;
- To provide radiation safety standards for people involved in agricultural production.

Formally, justification of restoration strategies in the agricultural production could be presented in several steps:

(1) Site characterisation of contaminated agricultural lands;

(2) Evaluation of the need for restoration;

(3) Assessment of the effectiveness of different restoration options under particular conditions;

(4) Comparative analysis and ranking of the restoration options;

(5) Justification of the most effective restoration measures with taking into account costs, availability of resources, possible limitations and socio-economic impacts.

The general logic of making decisions concerning the application of restoration options on agricultural lands in the contaminated regions of the Russian Federation is outlined in Figure 1. It is clear that all steps concerning justification of the restoration options depend directly or indirectly on the quality of the site characterisation. So, evaluation of the necessity for restoration, of the effectiveness of the restoration options, and of their ranking has to take into account many site specific factors, which control the mobility of radionuclides in agro-ecosystems, and the significance of different pathways for irradiation of the population.

The specific peculiarity of agricultural areas as a subject for remediation is the complexity of criteria used for the evaluation of restoration options. So, on the one hand, agro-ecosystems are important contributors to the collective dose for the population living outside of the contaminated area (imported dose). On the other hand, very often they determine the dose of irradiation the rural population involved in the food production receives. These result are different criteria for the restoration of agricultural land used for producing exported and locally consumed products.

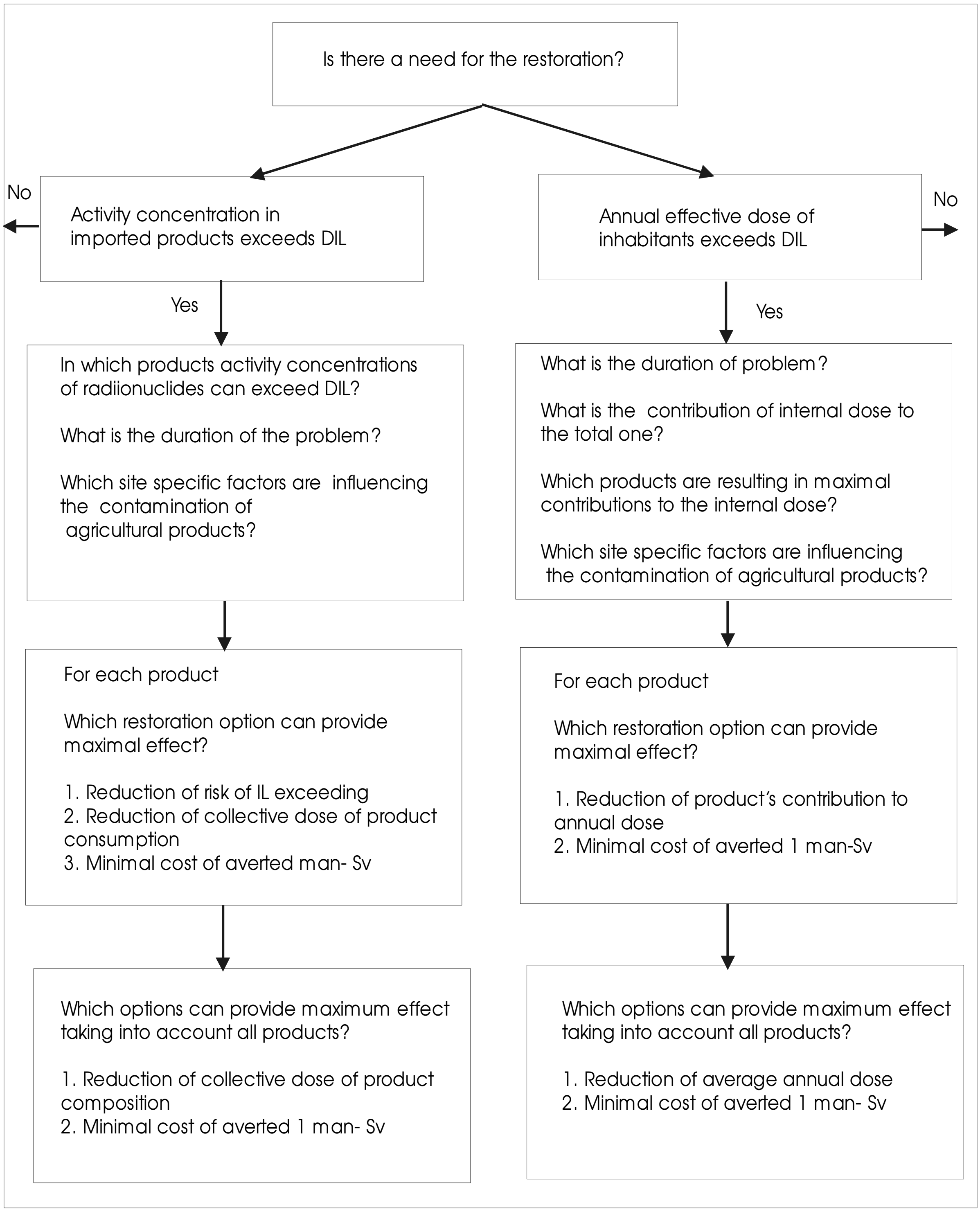

FIG. 1. The conceptual scheme of decision making on the application of restoration measures for agricultural land.

In the first instance the main criterion for determining the necessity for restoration is the exceeding of appropriate non-accidental Intervention Levels for radionuclides concentrations in agricultural products, which have been adopted for each product and each radionuclide. In the second instance, the criterion for determining the necessity for restoration is the average annual effective dose received by inhabitants of settlements located on the contaminated territory. The Federal law adopted in 1996 by the Russian Federation restricts the additional irradiation of the population by 1 mSv/y, which is now applied as Intervention Level [3]. The main difference between these criteria is that the first one limits the production of agricultural products in zones, which require the restoration, and the second one is aimed at lowering annual doses to the local population to less than 1 mSv/y.

Another point which should be considered when justifying the approach selected from the range of available restoration options, are the criteria for the selecting these options. It is clear that, at the stage of identifying optimal restoration strategies, using the criteria allows to make a reasonable choice. Among such criteria the following need to be considered:

- Reduction of risk from exceeding of Derived Intervention Levels (DIL_S);
- Reduction of collective dose from product consumption;
- Cost of averted dose of 1 man-Sv.

The first of the above criteria is the most important, when the concentrations of radionuclides in agricultural products are above DIL_S over a large territory.

It allows to evaluate the feasibility of obtaining foodstuffs that meet existing standards, as well as to determine the period of time for which restoration measures need to be applied to zones according to their level of contamination. The remaining criteria are of secondary importance and are used for selecting the most effective options. Thus comparable effects in terms of decreasing the radionuclide concentrations in products can be achieved by applying a wide range of different restoration measures. In order to select the most rational ones, additional criteria and data should be employed. In this case one should use more general criteria which take into account the dose reduction resulting from the application of these activities and their respective costs.

The cost of one man-Sievert being averted as a result of applying restoration measures application is often used as an integral indicator. The ICRP Publication no. 37 [4] considers measures justified, when the cost of reducing the collective dose by 1 man-Sv resulting from their application is within the range of 10–20 thousand US$. In this case the upper limit can be treated as a criterion for the most developed and the lower limit for the less developed countries.

3. SITE CHARACTERIZATION OF AGRICULTURAL LANDS FOLLOWING RADIOACTIVE CONTAMINATION

The importance of agricultural ecosystems as a source of irradiation for the population depends on site-specific features of the contamination, which directly highlights the need for adequate site characterisation. Among specific characteristics of agricultural land contamination determining the need for restoration measures the following can be considered:

- Degree of contamination of the agricultural lands by long lived radionuclides;
- Mobility of long lived radionuclides in soil;
- Land use.

As was noted earlier the goal of such characterisation is to provide the information needed for decision making on the restoration of contaminated lands. The parameters listed above are considered as the most important for this type of site characterisation. Owing to this fact three monitoring programs in agriculture were carried out in the Russian Federation after the heavy radiation accidents.

The objective of the first monitoring programme was to provide a characterisation of agricultural lands at the scale of individual fields in production. Identification of such site-specific characteristics as a degree of contamination by long lived radionuclides, soil properties and land use were included into this programme. Three surveys were performed between 1986 and 1996.

The second programme carried out on an annual basis at the level of each collective farm or settlement and concentrated on the estimation of current levels of radionuclide concentrations in agricultural products.

The aim of the third program was to estimate mobility of radionuclide in agroecosystems as a function of soil properties, type of land use and other factors. Estimations of the decrease in the contamination of typical plants and soils for each type of land use were also included. These more comprehensive studies were organised on stationary plots, where observations were carried out on a regular basis.

A system of distributed data bases was created for the collation of all these data in order to allow the implementation of a decision support systems for the selection of appropriate restoration strategies as is described below. On the whole, this approach allowed to establish clear links between site characterisation and optimisation of restoration policies.

3.1. Levels of contamination of agricultural lands by long lived radionuclides

The levels of contamination by long lived radionuclides are one of the most important criterion for different restrictions on agricultural activities on contaminated lands.

In the case of the Kyshtym accident, the reference radionuclide was ^{90}Sr, determining the long term hazard of contamination of the environment. The agricultural lands with a contamination level above 74 kBq m^{-2} were excluded from economic use [5].

^{137}Cs is the basic dose-forming radionuclide in the zone of the Chernobyl NPP accident (except for the period during and immediately after the accident when short lived and intermediate term lived radionuclides played an important role). Only in the part of the 30-km zone around the Chernobyl NPP, where economic activities had to be discontinued, and in a small zone beyond it, ^{90}Sr is of some importance. Therefore, the evaluation of the radiological consequences of the accidental releases from the Chernobyl NPP, as well as the planning and implementation of restoration measures, are based on information on the ^{137}Cs levels in the environment and the trends of its concentration changes in agricultural products [6].

In the first period after the accident at the ChNPP, the decisions concerning the organisation of agricultural production on contaminated territories were mainly based on the level of contamination of agricultural lands by ^{137}Cs. Thus, agricultural land with a level of contamination above 1480 kBq m^{-2} were excluded from the economic use [7]. However, at the next stage, different restrictions and recommendations for the organisation of agricultural production, taking into account the mobility of radionuclides in the soil and the land use, were introduced for agricultural lands with contamination above 185 kBq m^{-2}.

3.2. Mobility of radionuclides along the agricultural food chain

The mobility of radionuclides along the agricultural food chain, and initially through the soil, is another important factor determining consequences of radioactive contamination and the need for the application of restoration measures. The mobility of radionuclides in agroecosystems depends mainly on two factors, i.e. on the soil properties and on the physical and chemical properties of the fallout itself.

3.2.1. Soil properties

The relevance of internal irradiation is particularly high in regions where low fertility soils are widespread (soils poor in nutrients and humus, with acidic pH, and of light sand or sandy loam composition). In such regions radionuclides bio-availability is high, and in consequence, transfer rates through the soil-plant system are higher than in heavier, more fertile soils, resulting in increased levels of radionuclides in agricultural products [8]. Such are the characteristic bio-geochemical conditions of the main zone with the highest contamination levels after the accident at the Chernobyl NPP. The zone includes the Polesyes area, straddling Belarus, Russia and Ukraine, where light sandy and sandy loamy soddy-podzolic and hydromorphous peat soils are widespread. In this region an increased mobility of ^{137}Cs (as well as that of ^{90}Sr) in the soil-plant system was noted as early as the 1960's following global fallout after atmospheric nuclear weapon tests [8]. According to the data in Balonov [9], on soddy-podzolic soils in the Polesye region, the contribution from internal irradiation to the effective equivalent dose after the Chernobyl NPP accident amounted to 90%, whereas on fertile heavy chernozem soils in the same region it did not exceed 10%.

Therefore, considerable effort was applied to ranking of the soils according to the mobility of radionuclides. The technique, which was applied for characterisation of soils, is based on the estimation of aggregated transfer factors (TFs) calculated as the concentration (Bq kg^{-1}) in plant per deposition density (kBq m^{-2}).

There are approximately 2,000 permanent measurement stations in Russia, where samples of soils and agricultural produce have been collected for radioactivity measurement since the mid-1970's. Of these, just over 200 are located in the Bryansk, Kaluga, Orel and Tula regions, which were subject to the most intense radioactive contamination following the Chernobyl accident. The main results from this monitoring programme are already available [10] [11]. For the purposes of soil characterisation, only those sites, which were subject to deposition following the Chernobyl accident, and to which no restoration measures were applied, have been included in the evaluation.

The Chernobyl accident resulted in the contamination of soils with widely varying soil characteristics. The main soil types in the contaminated zone are soddy-podzolic, soddy-gleyed and soddy soils of different mechanical composition, grey forest soils and chernozems of different subtypes (leached, podzolized etc.), peaty soils and flooded meadows. For the purpose of this study, soils have been divided into four groups according to their ability to retain ^{137}Cs. One group consists of peaty soils for which the highest ^{137}Cs transfer coefficients are normally observed. Mineral soils were divided into three groups according to their mechanical composition using the N.A. Kachinsky-classification routinely applied in Russia [12]. The main characteristics of the different soil types are presented in Table I.

The results show a clear difference between the transfer of ^{137}Cs to different crops and between soils respectively. As reported elsewhere [13] [14] [15] [16], the highest transfer rates are observed on peaty soils and on mineral soils. The rate of uptake decreases with increasing clay content. In terms of crops, the highest ^{137}Cs transfer is to annual grasses, followed by perennial grasses, maize and finally fodder beet. The data for all crops studied have been merged in order to derive a relationship between the uptake of ^{137}Cs from the different soil groups (Table II). Ratios have been calculated separately for each year and each crop and the geometric mean was subsequently calculated. The observed ratios show good agreement with those recently reported by the International Atomic Energy Agency for temperate climates [17].

3.2.2. Land use

Taking into account specific features of technologies four types of land use (natural, cultivated pastures, and arable lands used for silage, and cereal or potato production) can be considered. Annual grasses, perennial grasses, maize and cereal crops are typical plants for these types of the land use.

As was already noted, there are clear differences in accumulation of radionuclides between these plants. Data on the uptake of ^{137}Cs by plants show that the respective differences can be as great as a factor of 100 dependent on soil properties. With respect to selected groups of plants and within the period of 1991–1994 the highest TFs for ^{137}Cs occured to natural grasses (the geometric mean of TFs amounts to $15.6\ 10^{-3}\ m^2\ kg^{-1}$), followed by perennial grasses ($6.1 \cdot 10^{-3}$), maize ($1.7 \cdot 10^{-3}$) and finally cereals, potatoes and fodder beet ($0.1 \cdot 10^{-3}$). This shows that the contamination of agricultural produce and, hence, the restoration requirements, depend on the specific combination of soil type and plant grown, i.e. they depend on the type of land use. On the other hand, the DILs for different agricultural products can also be different and, therefore, can influence the restrictions on agricultural land use.

The data from the monitoring programme mentioned earlier were used for estimating the differences in the levels of soil contamination, which would result in exceeding DILs in various products for different types of land use (Figure 2). These levels of soil contamination calculated as a ratio of DILs to TFs for reference plants are considered as the levels determining the need of restoration and can be easily applied in practice.

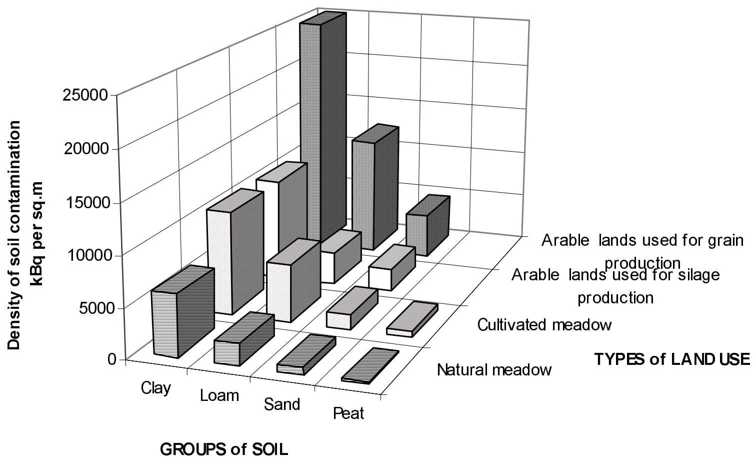

FIG. 2. Levels of soil contamination by ^{137}cs which would result in exceeding dils in the various products at different types of land use and soil groups.

The data presented show that differences between ^{137}Cs accumulation in plants typical for each type of land use result in different levels of residual contamination still restricting the use of agricultural produce. The influence of soil properties on the need for restoration can be also seen from the data given in Figure 2. It is thus possible to decide whether an area with a defined land use and soil type requires restoration, or whether a change in land use might be more appropriate. Such options might include the conversion of arable land into pasture, or handing over agricultural land to forestry.

3.2.3. Fallout properties

One of the peculiarities of the Chernobyl NPP accident was the duration of the radionuclide release into the environment [18] resulting in overlapping of radioactive trails and formation of zones with different physico-chemical compositions of the fallout [19] [20]. The major part of ^{137}Cs was deposited in the form of easily soluble finely dispersed aerosols, and some of it was included in coarsely dispersed particles and in fuel particles.

The fact that the ^{90}Sr and ^{137}Cs deposition after the Chernobyl NPP accident occured in two main forms — fuel particles from the destroyed reactor core, and condensed ones — resulted in two radionuclide fluxes into the agro-ecosystems, which were rather dynamic in the course of time after the accident and depended on the distance from the ChNPP. In the area where the condensed form of ^{90}Sr and ^{137}Cs dominated, the contaminants were characterised by high mobility and consequently elevated availability for plant uptake. To the contrary, ^{90}Sr and ^{137}Cs released into the environment as fuel particles were less available for root uptake at least during first few years after the accident. This fact did not correspond to the common behaviour of ^{137}Cs and ^{90}Sr of global origin, as well as to the data from experimental investigations in the accident area in the South Urals (Kyshtym, 1957).

The presence of ^{137}Cs in form of particles in the soils results in the development of two simultaneous, but opposing processes, i.e. an increase with time of the plant "available" amount due to the destruction of fuel particles, and a decrease in its "mobility" due to the fixation of ^{137}Cs by the organic and clay-mineral soil fractions.

Results derived from studies carried out in the vicinity of the ChNPP [21] show that the mechanism of plant uptake of fuel particle-derived radionuclides is of a complicated nature. For instance, in 1986 the ^{137}Cs plant uptake decreases with increasing fuel component in the fallout, due to a high proportion of it in fallout being bound in fuel particles during this period and, thus, not available for plant uptake. Subsequently, as a result of destruction of fuel particles and leaching of ^{137}Cs, the character of this dependence changes, and in the 3rd year after the fallout, plant uptake of radionuclides is an inverse function of the fraction of the fuel component, i.e. an increase of ^{137}Cs availability in the zones with high concentration of fuel particles in the soil can be observed (Figure 3).

Five to 7 years after the deposition, the ^{137}Cs plant availability in zones with different fallout characteristics equal out, which is due to the decreasing amount of radionuclides present in the form of fuel particles, and to caesium fixation in soil. It should also be noted that the degree of influence of fallout properties on the dynamics of ^{137}Cs-TFs to meadow vegetation depends also on soil characteristics. In the case of hydromorphic soils, the influence of fuel particles is more pronounced than on automorphic ones because of the higher mobility of ^{137}Cs in hydromorphic soils [21].

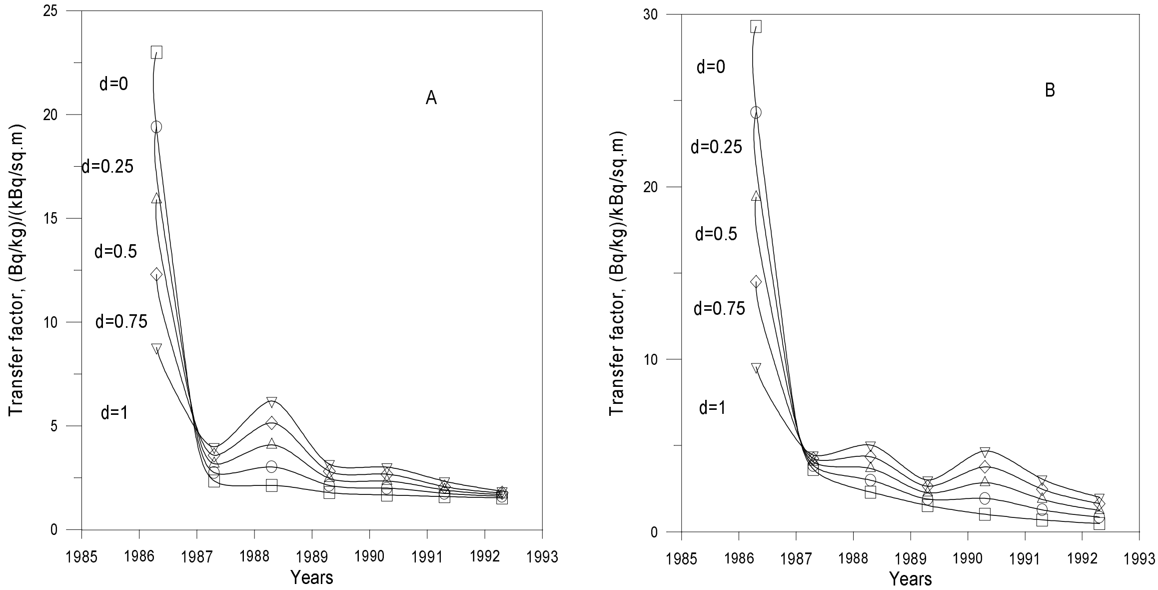

FIG. 3. Variation with time in the dynamics of ^{137}cs transfer factors (tfs) to meadow plants for different proportions (d) of fuel component in the fallout [21];
(a) automorphic soils; (b) hydromorphic soils.

4. FACTORS GOVERNING THE DECREASE OF ^{137}CS CONCENTRATIONS IN PLANTS

Assessment of the dynamics of internal doses during the post-accident period for the purpose of implementing restoration measures should include the estimation of decreases of radionuclide concentrations in agricultural produce with time elapsed after a single (accidental) release into the environment. This decrease is due to the fact that radionuclides transferred to the environment are gradually fixed by natural sorbents (soils, bottom sediments in water ecosystems, etc.) and become less biologically available for inclusion into the migration chain, in particular, into the soil-plant system. Therefore, effective half lives of ^{137}Cs in plants and residence times of ^{137}Cs in soil are among the most important parameters used for assessing the need for making adjustments to the restoration measures in the course of time after the contamination event.

4.1. Effective half lives of ^{137}Cs in plants

For comparative analysis of radionuclide bioavailability in food chains it is reasonable to use the half-life of the decrease of radionuclide levels in the various compartments of agricultural or natural ecosystems. Since radionuclide transfer is governed to a considerable extent by ecological factors, such parameters are usually defined as ecological half-life (T_{ec}).

According to the definition, ecological half-lives are equal to the period of time when content of radionuclides in some definite compartment of the trophic chain is decreased by half, due to all other factors, but except for radioactive decay. Effective half-lives (T_{eff}) have a similar definition, however in this case radioactive decay is taken into account.

Effective half-lives of ^{137}Cs TFs to plants are calculated on the basis of information derived from the monitoring programmes for the periods 1987 to 1990 and 1990–1994 mentioned earlier, and are given in Table III. For the initial years after the accident, the calculated values generally lie between one and four years, with most of the values ($R^2 > 0.9$) being less than two years.

The effective half-lives calculated for the period 1987 to 1994 are generally longer than those for the period up to 1990, thereby indicating that from 1990 onwards the rate of reduction in the uptake of ^{137}Cs by vegetation and fodder crops is reduced.

The databank of the International Union of Radioecologists has been used to estimate an effective half-life for ^{137}Cs uptake by agricultural crops, which was between four and eight years, although the inclusion of some early data from the former Soviet Union may have biased these calculations towards the lower end of the true values [22]. A different estimate is provided by the studies following the nuclear weapons testing, which suggest a half-life of eight to 15 years [15], although this value may not be directly comparable with the Chernobyl situation due to the different periods of deposition of the fallout. Clearly, a longer period of observation is required before the effective half-live can be applied to the current situation.

4.2. Mean residence times of ^{137}Cs in soil

This parameter requires the study of vertical distributions of radionuclides in soil profiles for different periods after the contamination event, as a basis for the validation and parametrization of the model used for the evaluation. Standard techniques of soil sampling and measurements are used at the first stage [8], allowing the determination of radionuclides concentrations in soil as a function of time and depth.

Validation of the model used for the residence-times calculations is a more complicated procedure. It has been shown elsewhere [23] that mean residence times calculated on the basis of simple compartimental or convection-diffusion models can not be used for the long term prediction of radionuclide transfer in soil, as they do not take into account the changing the mobility of radionuclides with time.

Therefore, to estimate the half-times of radionuclides in the top layer the more complicated model describing the variety of mobile ^{137}Cs fraction in soil as a result of sorption of radionuclides by soil and leaching the radionuclides from the fuel matrix as well as their vertical transfer was used [24].

The mean residence times in undisturbed soils of meadow of different types are shown in Table IV as an example of the results derived from this study.

The results presented indicate that the role of self-clearance for mineral soil is negligible. On the contrary, for wet meadows or peat land it can be an important factor, which effects a decrease in the intensity of ^{137}Cs transfer into food chains.

5. EFFECTIVENESS OF THE RESTORATION MEASURES

Restoration measure in agriculture can be classed into two groups: the restoration measures of the first group are based on the mechanical, physical or chemical treatment of soils, effecting either the removal of radionuclides from the soil or their fixation in some stable form, thus restricting their mobility in the environment. Normally they include [24]:

(1) Methods, which are based on the removal with or without replacement of the most contaminated top layer of the surface soil, with subsequent disposal of it at specially allocated places (excavating, planing, scraping, etc.);

(2) Methods for stabilization *in situ* without relocation of contaminated material, e.g. by cement stabilization, freeze crystallization or vitrification by thermal treatment, or the relocation of contaminants into deeper strata on site, e.g. by plowing-under, turning of flagstones, resurfacing roads,

(3) Methods for removal of contamination from soil *ex situ* or on site, e.g. physical separation techniques (screening, gravity and magnetic separation, vacuum swiping etc.), soil washing techniques (solvent extraction using water or solutions containing surfactants, chelating agents, acids or bases), or electrokinetic techniques.

The respective advantages and disadvantages of these methods are discussed elsewhere [24]. It is obvious that many of these techniques can only be applied to limited areas, owing to high unit costs, a general disturbance of the environment, and adverse impacts on important soil properties. Therefore, they had only limited application after large scale radiation accidents.

In general restoration measures in agriculture can be classed into three groups: organisational, agrotechnical, and agrochemical restoration methods.

Organisational methods in principal concern changes in land use. This may include increasing the area of land allocated to crops having a low accumulation rate for radionuclides. Other changes in land use are the conversion of arable land into pasture, or handing over agricultural land to forestry.

Agrotechnical restoration methods include deep ploughing with turning under the upper layer (on high fertility soils), and deep or superficial improvement of pastures. Many studies have been carried out on the effectiveness of root uptake reduction by meadow grass stands as a function of the various amelioration techniques. The techniques can be placed into two categories: deep and surface improvements. In addition, many combinations with ameliorants (lime, organic fertilizers and mineral fertilizers) which are potentially useful have been evaluated experimentally. Some restoration methods, however, have limited applicability; deep ploughing, for instance, is not applicable to soils with a thin humus layer. The advantage of these restoration methods is that they can be easily implemented as part of the normal agricultural practice.

Agrochemical options include liming of acid soils, application of increased doses of mineral fertilisers, addition of natural sorbents (different kinds of clay minerals) and use of organic fertilisers. The objective of applying mineral fertilisers to contaminated soils is to modify the ratio of the main plant nutrients. Liming is also one of the most important restoration methods that have been used to reduce caesium contamination levels in plants.

In the case of the Kyshtym accident, for the first time in the world experience with large scale restoration operations was gathered. Introduction of a complex of restoration programme for agricultural land in the area of the Eastern Urals Radioactive Trail (EURT) was iniated in spring, 1958. In the head part of EURT, the most contaminated zone, the soil cover consists of fertile soils, leached chernozems [8]. By a special soil treatment with burial of the upper layer containing radionuclides to the depths of 60 and 80 cm [8] [25]. In the EURT area, a radical amelioration of meadows and pastures was carried out, with the objective of providing a safe feeding basis for farm animals, that resulted in a 2- to 5-fold decrease of ^{90}Sr contents in plants used for fodder (Table V). Costly methods such as the removal and disposal of the contaminated upper 5–10 cm of the soil (scrapping) made it possible to achieve a 5- to 15-fold decrease of ^{90}Sr-transfer to the crops; however, this method was not applied widely [5] [8]

[25]. In essence an area of only 6200 hectares was subject to decontamination, using mainly the method of deep ploughing, thus bringing the upper radionuclide-containing layer to a depth of 50 cm.

In the of Kyshtym accident zone, the effect of mineral and organic fertilisers was tested, as well as that of liming the acid soils, with the objective of lowering the ^{90}Sr uptake by plants. On the average, liming resulted in a 10–30% decrease of ^{90}Sr-transfer to plants on the soils of the EURT, which belong to light acid type. Other options for lowering ^{90}Sr content in plant products were considered, such as cultivating crop varieties characterised by low accumulation of this radionuclide.

The Chernobyl accident provided a new example for a large scale application of such restoration actions. The experience from the previous accident was applied, taking into account the specific characteristics of the contaminated regions. A summary of the data on effectiveness of respective restoration measures based on information from both the Kyshtym and Chernobyl NPP accidents is given in Table V.

6. SITE CHARACTERISATION AND DECISION MAKING ON RESTORATION OF AGRICULTURAL LAND

Following the large scale radioactive contamination of agricultural land, the two main questions of importance in decisions making concerning restoration are:

— Which is the proportion of agricultural land requiring the restoration?
— When will the restoration measures need to be implemented?

As was discussed earlier, the answers to both of these questions are directly based on the site characterisation. The decisions on restoration of contaminated agricultural lands depend on several factors, which are to be defined on the basis of site characterisation activity. Among the factors which were distinguished were the levels of contamination by reference radionuclides, and their respective mobility in soil.

It is also obvious that the need for restoration, as well as the effectiveness of the methods applied, judging on the basis of criteria such as averted doses (for instance the cost of 1 man-Sv averted), is decreasing with the time elapsed after the contamination event.. Therefore, the amount of agricultural land where restoration is needed is also decreasing in time. The rates of this decrease depends on type of land use and respective soil properties. The phenomenon of change in the amount of in agricultural land in need of restoration is an important element in the long term policy for the organisation of agricultural production on the contaminated territories.

The criteria and classification of factors controlling the accumulation of ^{137}Cs in plants described earlier simplify the estimation of threshold contamination levels for each type of land use above which a specific restoration method can be considered as justified. It should be noted that due to the progressive decrease of radionuclides concentrations in soil and plants these levels are shifting towards higher values. This is illustrated in Figure 4 by the respective contamination levels calculated on the basis of different criteria for natural pastures on sandy soils as a function of time lapsed after the contamination event. The following criteria were considered in these calculations:

— The DIL for milk in regions the most affected after the Ch NPP accident is 370 Bq l^{-1};

— The cost of 1 man-Sv averted by radical improvement of natural meadows (the cost of implementing this measure is 150 USD per ha), calculated for the levels recommended by ICRP [4], are between 10,000 and 20,000 USD).

Figure 4 plots the initial concentration of ^{137}Cs-contamination versus the time elapsed before restoration and is parametrized by the dose criterion. Concentrations above the respective thresholds would justify restoration measures. It can be seen that with the increase in time elapsed since the concentration event, the initial concentration for which restoration would be justified increases. In other words, waiting for prolonged periods reduces the need and justification for restoration.

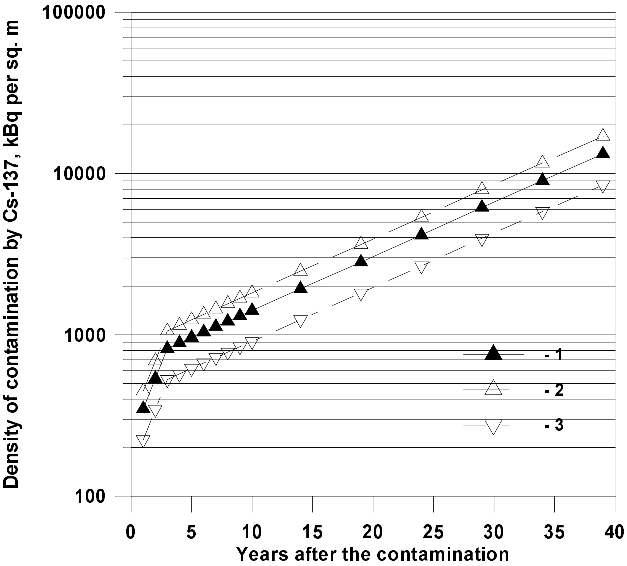

FIG. 4. Threshold levels of ^{137}Cs contamination in natural pastures as a function of time for which restoration measures are justified [27]. 1 and 2 threshold based on exceeding given permissible levels of ^{137}Cs in milk (370 Bq l^{-1}); 2 and 3 threshold based on the cost of averting 1 man-Sv (10 and 20 thousand USD, respectively)

A comparison of two criteria, lowering of the contamination levels of ^{137}Cs in milk below DIL$_S$, and lowering the collective dose by 1 man-Sv, shows that the contamination levels at which restoration is justified are 2–2.5 times higher for the second criterion than for the first. This indicates that the application of restoration measures can be considered justified for several years even after permissible levels of ^{137}Cs content in milk have been achieved, i.e. until both criteria have been met.

Figure 5 shows that inherent differences between soils concerning the decrease of mobility of radionuclides and its transfer outside of root containing layer should also influence the priority setting for large scale restoration policies of agricultural lands. In this example ^{137}Cs concentrations in milk were used as the criterion.

The data presented in this figure can be treated as threshold-levels of soil contamination for intervention after radioactive fallout. In the first year after the accident, the application of restoration measures on meadows with peaty soils is expedient for ^{137}Cs contamination above 110 kBq m^{-2} in the collective farms.

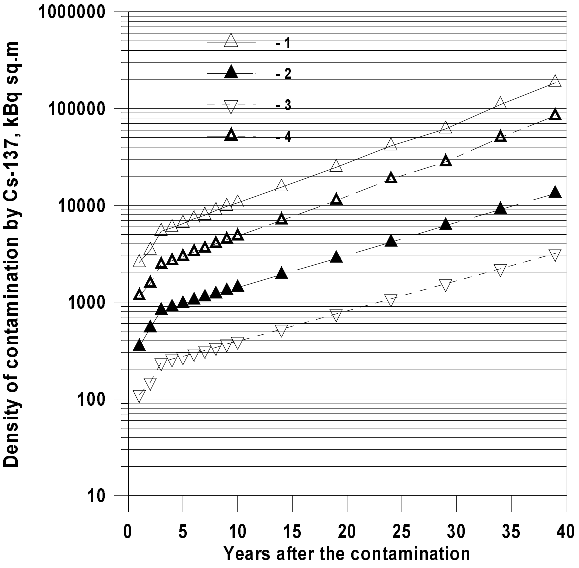

FIG. 5. Threshold levels of^{137}cs contamination in natural pastures as a function of time for which restoration measures are justified. (1), (2), (3) and (4) refer to clay, sand, peat and loam soils respectively [27].

For sandy and heavy loamy soils these values are 350, 1150 and 2500 kBq m^{-2}, respectively. It can be seen from these data that restoration measures are only justified within 30 years following the contamination of natural meadows on peaty soils, within 20 years for sandy soils, and within the first five years on loam. Analogous results were derived for other types of land use.

Data on contamination levels, land use, and soil properties became available in electronic format from 1987 on. Using this information allowed to assess which areas of agricultural lands required long term restoration measures, and to set priorities for their implementation taking into account site specific characteristics.

7. APPLICATION OF DECISION SUPPORT SYSTEMS FOR THE SELECTION OF OPTIMAL RESTORATION STRATEGIES

It is clear from the discussion above that many site specific factors could influence the need for restoration of agricultural lands. Spatial and temporal variability of these factors make the analysis of options which could be applied a rather complicated task. This shows the need for using a flexible decision support system (DSS), capable of providing practical advice on restoration strategies, and taking into account site specific features of the contaminated agricultural lands. The general objectives of using a DSS in restoration planning are first to provide a link between site characterisation and the decision making on restoration options, and secondly to provide the means for a comparative analysis of the effectiveness of different restoration options, taking into account site specific characteristics of the contaminated land.

The decision support system FORCON (Figure 6) was specially designed to simplify the selection of restoration options.

This computer-based system is intended for widespread use by agricultural specialists, who are required to give advice, taking into account the local features of contaminated agricultural lands.

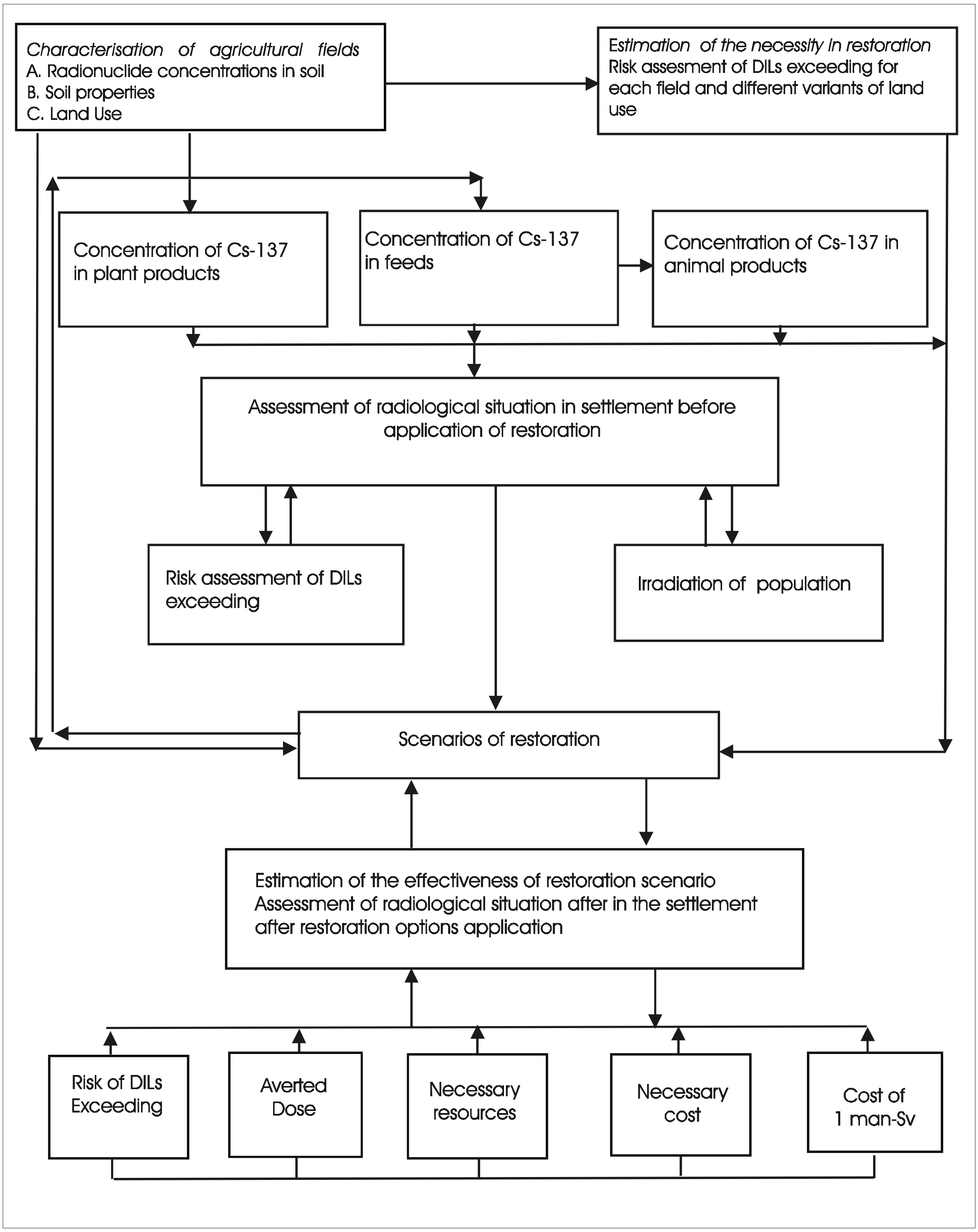

FIG. 6. Schematic representation of the 'FORCON' decision support system design.

The basic steps performed by the system are:

(1) Analysis of the existing radio-ecological situation on the basis of site characterisations of agricultural lands and the identification of those agricultural products that require some form of intervention;

(2) Assessment of the likely effectiveness of various restoration options taking into account prevailing conditions;

(3) Identification of the most appropriate restoration strategy.

The need for intervention and the development of a restoration strategy is based on either comparisons with Derived Intervention Levels (DILs), evaluation of the dose averted, or economic and other ancillary factors.

For an evaluation of the existing situation (without application of restoration measures) and the identification of the most appropriate strategy, the following information in the framework of the system is used: information on contamination levels on agricultural lands, type of land use, and soil characteristics. Within the FORCON, this information is represented on a map.

An analysis of this information constitutes the first stage in the selection of potential restoration options that are applicable. The second stage consists of an evaluation of contamination levels in crops on each field, and of the radionuclide contents in all types of other agricultural products from the settlement. Data on the quantities of each product are given in the form of tables, which include the proportion of each product in which DILs are exceeded.

To enable the user to select the most appropriate restoration strategy, the system provides the following information:

— maps which outline the possibilities for growing alternative products that would be below the relevant DIL for each year after the contamination. This analysis takes into account both, the contamination levels in individual fields. and soil characteristics;
— estimates for the radio-ecological effectiveness of different measures applied to each separate field in terms of lowering the activity concentrations in the product;
— the probability that a proportion of each product will exceed the DIL in the form of tables and graphics.

For each scenario of restoration chosen by the user, the following values are calculated: radionuclide concentrations in all of the foodstuffs produced; for animal products, data are presented separately for the periods spent on pasture and indoors; yields of each product; resources required (materials, transport, labour etc.) for the implementation of a given restoration option and the associated costs; the probability of the DIL being exceeded in each type of foodstuff after the restoration measures have been applied; averted dose; and averted dose per unit of costs. The user, therefore, has a comprehensive data sets on which to base his decisions, where radiological considerations may be constrained by other factors, such as finance or the availability of resources.

The restoration measures considered within the FORCON system are classified into four groups: organisational, agrotechnical, agrochemical, and restoration measures in animal husbandry.

Organisational restoration measures in principal concern changes in land use. This includes increasing the area of land allocated to crops characterized by a low accumulation of radionuclides, and in the case of areas of high contamination the abandoning of land from agricultural production. Within the current FORCON system, other changes in land use that can be considered are the substitution of current crops by hay, grain, potatoes, or pasture.

Agrotechnical restoration measures considered include deep ploughing with turning under of the upper layer (on high fertility soils), and deep or superficial amelioration of pastures.

Agrochemical restoration measures include liming of acidic soils, application of increased doses of K and P-K fertilizers, addition of natural sorbents (different kinds of clay minerals), and the use of organic fertilizers.

Decisions on the application of restoration measures in contaminated areas are based on predictions of the radionuclide content of foodstuffs. Many existing approaches are based on site specific data, with single values being allocated to each parameter. However, many parameters have an inherent variability, for instance because of non-uniform contaminant deposition, differences between animals in a single herd, or variations in soil properties. The approach taken within the FORCON system enables the radiological situation to be assessed in probabilistic terms, by taking into account information about observed variations in contaminant deposition and inherent variability in parameters that describe the radionuclide migration through the food chain. More detailed descriptions of the models and the approaches for dose calculations are given elsewhere [28].

To assess uncertainties in predicted values Monte-Carlo simulations are usually employed in radio-ecological models. However, this method is time consuming from the computational point of view, thus making it unsuitable for the interactive approach stipulated for systems such as FORCON. The procedure used in the FORCON combines deterministic and probabilistic simulations, the latter being based on observed distributions of parameter values whenever possible. The characteristics of these observed distributions are assumed to remain constant over time. Thus, for each year under consideration the best-estimate values for each parameter are calculated using dynamic models; the same probabilistic distribution is then applied to each individual value.

A model to estimate the cost which is associated with each restoration scenario is also included. It takes into account any changes in productivity which may result from the imposition of any given restoration option.

All the parameters required for the calculation, analysis and presentation of information are contained in the database. The database also includes supporting data for the cartographical representation of information. The database of the FORCON system consists of four groups of data needed for the assessment of the efficiency of restoration options, including parameters for the transfer and accumulation of radionuclides in various food chains, data on restoration efficiency by method, associated costs, and resources required to implement the method, and inquire into radiological information that have been derived from the assessment of data within JSP1 [29]. To date the system’s database contains information on the efficiency of more than 300 variants of agricultural restoration methods. However, the use of FORCON is confined at present to radio-caesium, since most of the information used in the development of the system has come from areas affected by the Chernobyl accident. In the future the system should be enlarged with corresponding information for other potentially important radionuclides, such as ^{90}Sr.

8. CONCLUSIONS

Following a number of serious radiation accidents a wide range of different restoration options have been developed and adopted for use in the case of radioactive contamination of agricultural land. In the ten years since the accident, various measures have been implemented and a vast amount of data on their effectiveness has been generated, together with information on ancillary factors, such as the required resources and costs. These measures vary considerably by their effectiveness, cost and practicability in actual situations. Their effectiveness depends on many factors, including soil and climatic conditions, or specific features of the agricultural production management. Owing to this the implementation of these restoration measures on the basis of general expert estimations often results in inadequate decisions. The results presented in this paper underlined the importance of the role of site characterisation in the selection of restoration strategies and demonstrated the need for implementing a flexible decision support system, which is capable to organise a comprehensive set of site specific information, and which provides a practical tool for advising on agricultural land restoration.

TABLE I. MAIN CHARACTERISTICS OF SOIL ACCORDING TO THE CLASSIFICATION USED IN THE STUDY.

Soil groups	**Soil types, as classified in Russia [12]**	**pH_{KCl}**	**% Humus**	**Cation exchange capacity[a]**	**Clay content[b]**
Sandy, Loamy sand	Soddy-podzolic; Soddy-gleyed; Soddy; light grey forest	3.5–6.5	0.5–3.0	3.0–15.0	<20%
Light loam, Medium loam	Soddy-podzolic; Soddy, grey and dark grey forest; Leached chernozem; podzolized chernozem	4.0–6.0	2.0–6.5	5.0–25.0	20–40%
Heavy loam, Clay	Dark grey forest; leached chernozem; podzolized chernozem; typical chernozem; usual chernozem	5.0–8.0	3.5–10.0	20.0–70.0	>40%
Peat	Peaty; peaty-bogged; peaty-gleyed	3.0–5.0	5.0–30.0	20.0–200.0	

[a] measured as meq per 100 g soil; [b]particle sizes <0.001mm

TABLE II. RATIO OF TRANSFER FACTORS OF ^{137}Cs FOR DIFFERENT SOIL GROUPS

	Ratio	**95% confidence interval.**	**Range**
PEAT : SAND	3.6	3.0–4.4	2.0–8.0
PEAT : LOAM	10.5	7.5–14.7	3.3–37.5
PEAT : CLAY	27.0	20.5–35.5	8.3–48.7
SAND : LOAM	2.4	1.9–2.9	0.8–6.0
SAND : CLAY	5.2	4.1–6.6	1.6–14.3
LOAM : CLAY	2.2	1.8–2.6	0.4–4.4

TABLE III. ENVIRONMENTAL HALF-LIVES FOR THE UPTAKE OF ^{137}Cs BY AGRICULTURAL PLANTS IN 1987–1990 AND 1990–1994.

	1987–1990			1990–1994		
Soil type	T_e	$Tf_1(0)$	R^2	T_e	$Tf_2(0)$	R^2
			Barley			
Sand	1.3	0.43	0.99	6.2	0.14	0.65
Loam	1.9	0.21	0.98	6.7	0.09	0.71
Clay	1.4	0.17	0.98	3.8	0.07	0.87
			Maize			
Sand	2.4	6.3	0.99	7.5	2.9	0.86
Loam	1.5	2.1	0.76	16.1	1.1	(0.21)*
Clay	1.9	1.3	0.98	17.3	0.53	(0.47)
			Potatoes			
Sand	1.2	0.57	0.88	7.5	0.12	0.99
Loam	2.4	0.14	0.98	8.5	0.1	(0.20)
Clay	2.9	0.07	0.95	5.0	0.04	0.64
			Beet-roots			
Sand	2.9	0.32	0.92	5.2	0.29	0.83
Loam	2.6	0.25	0.82	5.9	0.12	0.98
Clay	2.9	0.18	0.72	7.2	0.15	0.96
			Annual grasses			
Sand	1.6	29.0	0.99	15.4	18.3	0.87
Loam	1.3	7.0	0.99	4.7	2.3	(0.37)
Clay	1.3	3.6	0.99	4.9	1.4	0.56
Peat	1.8	83.4	0.84	10.9	29.6	0.72
			Perennial grasses			
Sand	2.3	11.8	0.99	4.8	3.5	0.91
Loam	2.5	11.7	0.95	4.6	0.6	0.60
Clay	2.5	1.9	0.95	10.2	0.65	0.66
Peat	2.6	23.4	0.99	21.0	9.9	(0.21)

TABLE IV. HALF-LIVES OF RADIONUCLIDES IN VARIOUS TYPES OF MEADOW SOIL [24]

		T_{ec} [years]		T_{eff}
Meadow type	**Soil type**	**Mode**	**95% CI**	**[years]**
Dry meadow	Sand	1870	1380– 2515	29.5
Dry meadow	Loamy sand	2030	1690–2400	29.6
Dry meadow	Heavy loam	1970	1260–3070	29.5
Lowland (wet) meadow	Light loam	530	403–692	28.4
Lowland (wet) meadow	Peat	280	200–387	27.1
Flood plain meadow	Loamy sand	440	220–890	28.1
Flood plain meadow	Peat	60	49–73	20.0
Transient peat land	Peat	17.4	14.2–21.3	11.0
Low peat land	Peat	26	19–35	13.9

TABLE V. EFFECTIVENESS OF THE MOST WIDESPREAD AGRICULTURAL RESTORATION MEASURES [8]

Restoration options	**Soil type**	**Reduction factor in plants (times)**	
		^{90}Sr	^{137}Cs
Ploughing	Mineral Organic	1.6–4.0	1.8–2.5 2.0–3.2
Ploughing with turning over upper layer	Mineral Organic	3.7–4.0	2.8–5.6 3.3–6.9
Liming	Mineral Organic	1.5–3.4 3.0–3.2	1.6–2.5 1.5–5.0
Application of organic fertilisers	Mineral	1.8–4.6	1.1–3.1
Application of mineral fertilisers	Mineral Organic	1.1–2.8 1.2–2.8	0.7–2.2 1.1–2.5
Radical improvement, including ploughing, standard dose of fertiliser, re-seed	Mineral Organic	1.6–3.5 2.5–5.5	1.8–3.3 3.1–3.9
Surface improvement, including rotary cultivation, standard dose of fertiliser, re-seed	Mineral Organic	1.1–2.5 1.5–4.1	1.3–1.9 1.4–2.2

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MEASURING TECHNIQUES FOR THE CHARACTERISATION OF ^{137}Cs CONTAMINATED RIVER BANKS, SLOVAKIA

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ABSTRACT

In 1990, ^{137}Cs contaminated areas were discovered along 19 km of the banks of the rivers receiving discharged waste waters from the Bohunice NPP. The contamination was the result of uncontrolled releases of drainage water from the radioactive waste treatment unit and two accidents on the CO_2 cooled and heavy water moderated NPP-A1 unit in 1976 and 1977. In total, more than 67.000 m^2 of bank surfaces (the lower and middle parts of their slopes) have been found to be contaminated with ^{137}Cs at levels exceeding 1 kBq/kg of soil. This paper describes the planning for restoration of these river banks with emphasis on site characterization techniques used for these purposes. The general strategy and application of monitoring techniques for the purposes of characterisation of contaminated river banks is the main topic of the work performed in the context of the IAEA CRP on ”Site Characterisation Techniques for Environmental Restoration”. First results in developing new and more sophisticated measuring techniques, needed for the characterization of discontinuous and spot-contamination found on parts of the banks are given. The technique described is based on a dose rate measurement approach, using ratio of two detector readings at two different distances above ground and Monte-Carlo simulations of the plastic scintillation detector responses. The method will also be useful, for the final, post-remediation survey of residual spots of ^{137}Cs contamination.

1. INTRODUCTION

The present work was carried under the Research Agreement No 8731/OR between the IAEA and VUJE Institute Trnava, Slovakia. According to this agreement, the subject and long term goal of the VUJE Institute participation concerns to the developing and innovation of monitoring technology necessary for radiological survey and characterisation of contaminated sites resulting from nuclear accident.

The research topic received baseline funding and support within the framework of the national project no. PE1024 ”Innovation of the monitoring systems and procedures”, and carried out in the time period between 06/1996 to 12/1998. Experiences gained from performing tasks related to this project were further utilised in the context of the IAEA project on ”Site characterisation for remediation purposes” [1] and on ”Post-restoration compliance monitoring” [2].

Project PE1024 was focused partly on large-area monitoring and the monitoring plan optimisation, and partly on developing and testing new methods using Monte-Carlo detector response simulations to ^{137}Cs contaminated circle soil spots. The latter method is intended for use in the context of remediation and post-restoration monitoring, and for characterisation purposes on spot-contaminated river banks. Detailed description of results obtained during the project are described in two VUJE reports [3] [4].

The objective of this annex is to describe the general strategy of monitoring and characterisation techniques as applied to the contaminated banks of Manivier canal-Dudváh river system. The description relates, in general, the planning, performing and assessing of the site characterisation and radiological surveys with subsequent validation of compliance with the derived cleanup criteria. In addition, the recent results achieved in the field of Monte-Carlo simulation of the response of a plastic scintillation detector, and the determination of ^{137}Cs content in small circular spots of contaminated soil, including field verification examples are given. The tested plastic scintillation detector with a 76x76 mm size was shown to being sensitive enough to apply it not only as the main measuring device during clean-up, but for post restoration monitoring purposes as well.

Detailed descriptions of radiological environmental pathway studies and dose assessments for the contaminated banks, including derived clean-up levels can be found in the companion references [1] [5] [6] [7].

2. BACKGROUND INFORMATION AND DESCRIPTION OF THE SITE CONTAMINATION

2.1. Characteristics of the site

The 19 km long banks of the Bohunice NPP waste water recipient has been identified as contaminated by ^{137}Cs as a result of two accidents on the CO_2-cooled and heavy water-moderated NPP-A1 unit in 1976 and 1977 respectively. In total, more than 55,000 m^2 of river banks (their lower and central parts) have been found as being contaminated at levels exceeding 1 Bq^{137}Cs/g of soil. Until 1992, NPP waste water had been routed through the 5 km long, concrete paved Manivier canal to the smaller rated Dudváh River ($Q_{average}$= 1.8 m^3/s) which joines the Váh River (Q_a= 150 m^3/s) further 13 km downstream, 90 km above River Váh's confluence with the Danube River (see Figure 1).

In the period between 1976 and 1978, when both accidents happened, a flood control project on Dudváh River had just been being implemented over a length of 8 km upstream of its mouth. In the following 5 km upstream river section affected by NPP, the flood control conditions are insufficient and has, hitherto, caused permanent concern of the public. The affected recipient water flows directly through, or passes close by, four villages with approximately 5,000 inhabitants. The contaminated banks have, however, unrestricted access for some 11,000 residents living in a 3.5 km wide strip alongside the river.

2.2. Types of contamination on the river banks

We can distinguish between a primary and a secondary contamination of the banks; the latter arose as a result of improper handling of the highly contaminated river bed sediments and mud. A number of types of river bank contamination were identified at the Bohunice site:

(1) In the upper part of the flow, bank surfaces were primarily contaminated during the accidental releases which coincided with water level being elevated by by-passing during construction works the flow some 8 km upstream from the mouth of Dudváh River.

(2) At the mouth, contrary to expectations, both river embankments along a 3 km stretch of the river, which were constructed as part of the widening works, were found to be highly contaminated too. It is assumed that highly contaminated bottom sediments from the by-passed river bed were used as cover material in constructing the embankments.

(3) Along a 2 km section of the canal banks, just downstream from the NPP discharge point, frequent and highly intense spots of ^{137}Cs contamination have been discovered on the central part of the banks. The spots probably result from past improper clean-up operations on the paved canal bed. During this clean-up operation, highly contaminated mud discharged from the contaminated NPP-A1 sewage system after flooding in 1980, was manually excavated from the paved canal bed and scattered over the central part of the embankment surfaces.

(4) Along an 1 km section of the present Váh river flood-plain, which was occupied by Dudváh River before its channelling, large spots (5 to 10 m radius) with inversely depth distributed contamination were found as a result of naturally settled, highly contaminated sediments from the Dudváh river bed where it deepened or widened.

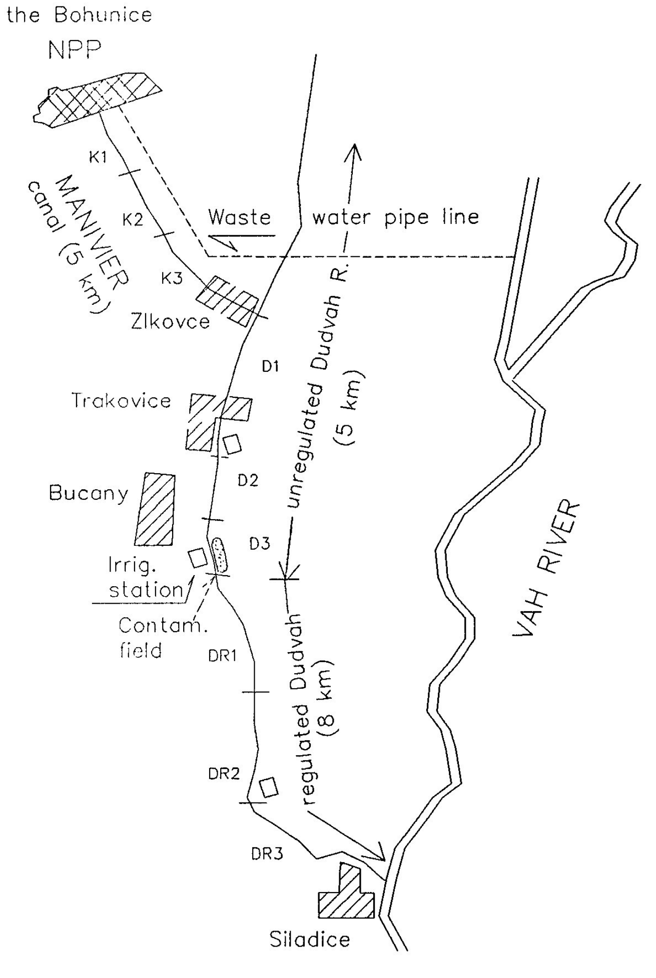

FIG. 1. Scheme of the river-reservoir system at the Bohunice NPP site

2.3. Initial remedial responses after discovery of the problem

The contamination of the river banks and its significance was discovered in 1991 in connection with the preparatory work for the implementation of the above mentioned flood control project on the still not-engineered part of the river. As a result, the project implementation was stopped during its licensing procedure by the responsible health authority. Soon after, proper restoration action was requested from the operator of the Bohunice NPP, who was considered responsible for the contamination. A clean-up level was prescribed for this purpose by the authority, and ad hoc set on the rather low level of 1 kBq^{137}Cs /kg in the soil.

In 1992, a bank restoration project including site characterization for the affected part of the river was initiated by the NPP with a projected disposal capacity of 5,000 m^3 for removed soil. It was planned to dispose the soil into a subsurface concrete structure on the NPP site, which was considered to be the option most acceptable to the nearby public. During the ongoing monitoring exercises, other parts of the river banks were found to be affected as well. Therefore, a comprehensive post-emergency radiological survey was deemed necessary to include all potentially affected river banks and the adjacent land.

Initially, a ground based screening survey was conducted on the flood plain of the Dudváh (18 km) and Váh rivers (25 km), including the Kralova Reservoir in order to identify contaminated locations in the area. After completion of the monitoring exercises, it was recognized that the 1 Bq ^{137}Cs g^{-1} value was too low and inappropriate for use as clean-up criterion. It emerged that the previous restoration project planning needed to be re-considered with emphasis on the complexity of the proposed cleanup measures, including alternative remedial technologies (fencing, clean covering, trenching), cost-analysis and the development of criteria for justifying cleanup.

The VUJE Research Institute has been involved since 1993 in addressing in a comprehensive way the contamination problems described above. A typical problem in the context of these efforts was the absence of clear legislation in the field. Therefore, the primary requirement was to develop some principles for evaluating the justified scale of clean-up measures. The first priority were the development of appropriate clean-up criteria in order to obtain public confidence and to facilitate authorization of the reviewed final environmental restoration plan. This task was realized in close co-operation with the relevant health authorities and experts. The proposals submitted for comprehensive remediation measures at the site were supported by the State Health Institute in Bratislava in the beginning of 1995 and are described in more detail in references [5] [7].

Owing to recent the changes in economic and political circumstances, and the not too critical radiological impacts, restoration activities are again under review. The main question now is the justification and feasibility of the remedial measures planned. Other aspects, including the selection of an appropriate disposal site and burial technology, investments required, and the issue of public acceptance are now under review. It appears that the implementation of flood control measures in the upper part of River Dudváh will be the main driver in the future development of proper remedial activities and priority sections along River Dudváh will be set accordingly.

3. MEASURING AND SAMPLING TECHNIQUES EMPLOYED

3.1. Soil and grass sampling including laboratory analyses

Gamma radiation readings from a hand-held plastic scintillation detectors have been used for biased sampling of the contaminated soil layers at locations selected for being typical of different levels of contamination. For laboratory analyses, sliced bulk soil samples from well defined depths and of defined thickness were taken from the embankment slopes inside and outside the levees along the river.

The depth distribution study gave meaningful results only, if the reproducibility of the sampling depth and location was ensured by a special soil sampling technique. For this purposes, a special shovel-sampling device with a 200 cm^2 cross-section was developed in-house, which allows depth-oriented soil layer sampling. The shovel is fitted with special lateral ”wings”, which move in an U-shaped metal frame as to well reproduce the sampling depth. It is shown ”in action” in Figure 2. In places, where contamination levels are higher and sampling depth accuracy is not so critical, standard cylindrical augers have been in use as well.

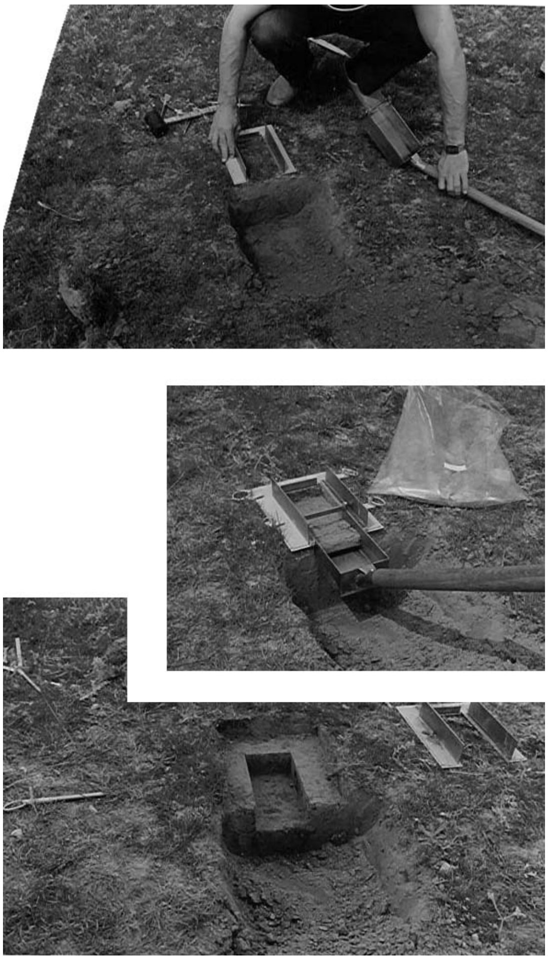

FIG. 2. Sampling of soil layers by special shovel with wing and U-shaped frame.

Laboratory analyses were performed using a standard HPGe gamma-spectrometry system consisting of two High purity Germanium (HPGe) detectors with about 35% relative efficiency and 2.0 keV resolution (1332 keV), and modular spectrometry electronics. These analyses showed that ^{137}Cs is the dominant contaminant at the site. Only negligible concentrations of other gamma emitting radionuclides, such as ^{60}Co and ^{134}Cs, were found in the samples.

On selected samples, radiochemical analysis and determination of ^{90}Sr and ^{239}Pu were carried out in addition. The respective results are reported in [8]. The main aim of these analyses was to determine the absolute and relative content of some very toxic radionuclides with either alpha or pure beta particle emissions in comparison to the well determinable ^{137}Cs activity concentrations in the samples. The ^{90}Sr content in the soil, estimated on the basis of a set of about 20 samples analysed, has been found to be 50 to 100 times lower than the ^{137}Cs activity concentration. Nevertheless, its contribution to the effective dose showed to be not insignificant. The ^{239}Pu concentration has been found to be 5 to 7 thousands time lower than the ^{137}Cs levels in the samples and, therefore, could be completely neglected for the dose assessment purposes.

In addition to the soil samples, grass samples from the most contaminated parts of the banks were analysed in order to estimate soil-to-plant concentration ratios for ^{137}Cs and ^{90}Sr. A 0.5 x 0.5 m frame was used for sampling the grass on the bank. Some of the grass samples were rinsed in water, the water was analysed, and the results were compared with results from the rinsed grass. The results so obtained show that the grass contamination is mostly caused by re-suspension of the soil particles due to rain splashing.

3.2. Mobile, vehicle borne techniques for screening and activity concentration measurements

For accessible locations on the inland slopes of levees, continuous scanning by a Vehicle Mounted Gamma Survey System (VMGS) was employed [8]. The VMGS (Fig. 3.) consists of a large, 100×100 mm, shielded scintillation NaI(Tl)-detector (sodium iodide activated with thallium), commercially available rack and modular electronics, and a micro-computer with custom software. The NaI(Tl) detector is inserted into a flexible car holder containing also a 5°cm thick led cylindrical shielding The system was developed in the VUJE Institute as part of a national project. The rack for electronic moduls containing the high voltage (HV) and amplifier/ counter modules are supplied by a 12 V standard car battery and a current converter designed specially for this purpose. This equipment was used for surveys of the 18 km long Dudvah River banks, several hectars of land fields nearby the Dudvah River, and areas on the Vah River flood plane as well.

The NaI(Tl) gamma counting system was mounted on the hydraulic arm of a tractor in order to test the monitoring system in difficult-to-access steep bank conditions of the Manivier canal, as is shown in Figure 4.

3.3. Hand held scanning and discrete measurements on steep banks

On the slope of the levees facing the river, a hand held, lightly shielded gamma survey meter with a large plastic scintillator (76×76 mm), as shown in Figure 5, was used for point measurements on a 20 m line grid. A measuring time constant of τ= 10 s was used to reach a sufficient sensitivity of the detector. At critical locations, where activities were highest, a 10 m grid was used, thus covering about 15% of the embankment surface area. The application of detector shielding was intended to achieve sufficiently close correlation between the soil ^{137}Cs activity concentrations and the detector responses above narrow contaminated strips on the bank of variable width between 0.5 to 2.5 m. Measuring the narrowest strips caused some problems at times because its width could be narrower than the effective field of view of the detector used. Thus, the detector response strongly depends on the width of the strip measured.

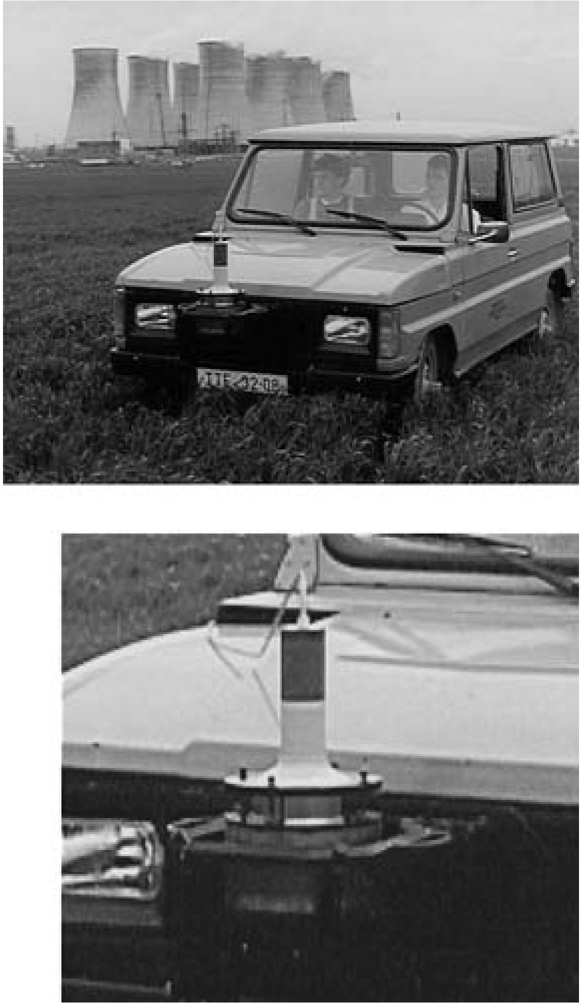

FIG. 3. Vehicle-borne NaI(Tl) gross gamma counting survey and mapping system.

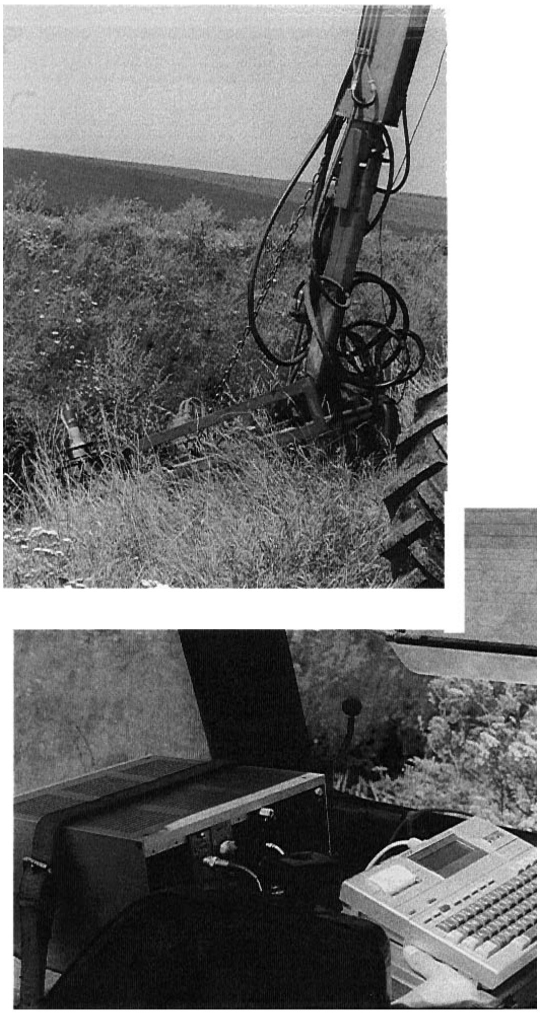

FIG. 4. Tractor hydraulic arm-borne gross gamma-survey system for difficult-to-access sites (e.g. steep river banks).

FIG. 5. Hand held plastic scintillation (76×76 mm) detector with a light 2 cm lead shielding for hand-held scanning and discrete-spot measurements.

For the canal sections with contaminated spots, in addition to grid survey described above, continuous scanning with τ= 1 *s* to search for the most highly contaminated spots was employed. Some of the spots discovered were examined in detail by scanning and soil sampling to determine their size, shape, specific activity, and the depth distribution of ^{137}Cs. There was no close correlation found between the detector response and the specific activity of the spots. This was found to be due to their not-regular shape and variable size, which in most cases was smaller than the circular area ”seen” by the detector (R_{dw}= 70 cm).

3.4. Assessment of small spot contamination on the river banks

Finally, three groups of spots were selected based on to the typical average activities determined by laboratory analysis of the samples taken. Sizes and linear densities of the contaminated spots were roughly estimated on the basis of the readings obtained from the monitoring devices. These data were then applied to make conservative assessments of the average contamination of the affected bank section (only on its lower part).

More accurate evaluation of these discontinuous bank contaminations requires the application of more sophisticated measuring techniques/methods, which require furthers scientific development and field verification (see Chapter 6).

3.5. Summary of radiological survey results

3.5.1. 3.6.1. Extent of contamination

The detailed survey carried out between 1991 and 1994 shows that the top soil contamination on the banks varies widely from background levels (0.01 kBq/kg), to 20 kBq/kg of ^{137}Cs (around 4 MBq/m^2) for River Dudváh, and reaches 250 kBq/kg for spot contaminations on the canal banks near the NPP discharge point. The contamination is spread over a strip along the lower part of the banks approximately 0.5 to 3 m wide (on average over 2.2 m), and the average level of ^{137}Cs in the top 10 cm of the soil layer reaches 6.3 kBq/kg. This is equal to about 1.1 MBq /m^2 of ^{137}Cs surface activity, if one reckons a soil density of 1.3 kg/m^3 and that 75% of the total activity are located in top 10 cm of the soil profile. A summary of the monitoring results on selected river bank sections (indicated in Fig.1) as were predetermined on the basis of prevailing conditions is given in Table I.

Thus overall, contaminated area identified, with an activity level exceeding 1 kBq/kg of ^{137}Cs, comprises approximately 67,000 m^2. The volume of soil to be removed according to the first clean-up limit exceeds 13,000 m^3. However, the assessment of the volume distribution of the soil contamination has shown that removing the mostly contaminated soil only would result in a significant improvement in the radiological situation at the contaminated site. Details concerning the relevant distribution of the contamination in space on the embankment surfaces are described for instance in reference [8].

TABLE I. CONTAMINATED AREAS AND ^{137}Cs ACTIVITY CONCENTRATIONS ON THE AFFECTED BANKS NEAR BOHUNICE NPP.
(see Fig. 1 for a map of the respective river and canal sections)

Contam. section	**S,> 1** [m^2]	**A_S** [kBq/kg]	**S,> 8** [m^2]	**A_S** [kBq/kg]	**A-resid** [kBq/kg]	**Note**
K1	10000	6.7	2000	9.5	4.9	strip, 1-1.5m
K2	5730	16.2	5730	16.2	3.3	strip, 0-2.5m
K3	9725	2	0		2	(near a village)
D1	1200	1.8	0.0	0.0	1.8	
D2	1500	3.5	0.0	0.0	3.5	(arable field)
D3	3725	4.7	1400	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	10.2	0.9	clean soil cover
VPK1	4760	2.1	10	8.7	2.1	S< min area
VK35	2530	2.8	25	9.2	2.8	S< min area
VK6	4260	1.8	0		1.8	
SUM	**67420**	**-**	**18615**	**-**	**-**	

S,>1, S,>8 are areas with activity concentrations higher than 1 and 8 kBq/kg respectively, A_S is the average activity concentration at area S,>, A-resid is the estimated residual activity concentration at the indicated bank section after removal of the soil from areas with contamination higher than 8 kBq/kg.

4. SITE CHARACTERISATION THROUGH DOSE ASSESSEMENT

4.1. Radiological risks definition for the site

The planned flood control project at the Dudváh headwater, which would involve the removal large volumes of the contaminated soil from the river banks, and their spread over the adjacent environment poses a most serious threat to the nearby population. In a worst case scenario, it can be envisaged that the contaminated soil be uncontrollably relocated and used as a in-fill or be spread on garden soil around residential houses. This type of radiation risk could also arise, but with smaller amounts of contaminated soil being involved, from maintenance or other construction works on the bank. It can be considered the most critical exposure pathway, and would also arise at contaminated river bank sections with satisfactory flood control conditions at present.

4.2. Exposure pathway analysis and its radiological characteristics

Typical use and occupancy scenarios were selected and analysed in close co-operation with the health authorities to evaluate actual and potential risks from the contamination on the banks. Scenarios with legally accepted parameter values were employed for the final evaluation of the contaminated site. The two types of scenarios were considered: the use of the banks for recreation (e.g. fishing), and second the potential use of contaminated soil itself (after its removal to residential and garden plots), have been shown to be the most significant. In addition to external radiation exposure, the ingestion pathway, using transfer factors from goat's milk and meat, and the loamy soil according to [6] [10] for both ^{137}Cs and ^{90}Sr, were part of the dose estimations performed. The radiological characterisation resulting from the selected and authorised occupancy and "soil use" scenarios is summarised in Table II.

This table indicates that the most critical exposure pathway is the potential use of contaminated soil. Table II further indicates that the effective dose from a stay on the bank does not exceed 0.35°mSv/y, while the potential risk from the use of contaminated soil after its relocation to a fictive residential garden results in much higher effective dose levels of up to about 2 to 3°mSv/y.

The annual collective dose from the scenario "stay on the banks", corresponding to a land use which is not too-intense, is very low, at maximum at the level of about 100 to 200 man mSv.

TABLE II. E FACTORS (DF) RELATED TO 1 kBq/kg OF ^{137}Cs IN SOIL AND DERIVED INTERVENTION LEVELS (DILs) FOR SELECTED SCENARIOS. DOS

SCENARIO	**T_0 [y]**	**geom.f t_{exp} [h/y]**	**INGESTION [relative unit]**	**DF [mSv/y]**	**DIL(1,T_0) [kBq/kg]**
stay on banks	0	300x1.4 g= 0.54	0.4 milk+meat	0.035	28.6
stay on contam. 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

Relative unity of ingestion dose = 0.04 mSv/y from consumption of potato 110 kg + root veg. 55kg + leaved veg. 55kg,
g = (dose rate used /Bq.g^{-1}):(0.118 microSv.h^{-1}/Bq.g^{-1}) (geom. f. against a half-indefinite ^{137}Cs source),
ve - vegetable, po - potato, mi - milk, me – meat, DIL - derived intervention level
DIL(l,T_0) = (1 (mSv/y) / DF).exp(λT_0), where:
T_0 time from which the likelyhood of applied scenario is considered as non-zero, .λ= ^{137}Cs decay constant.

4.3. Acceptance and cleanup level derivation

The same principles as described above were applied for the development of site specific clean-up limits as well. Acceptance criteria and clean-up levels for the contaminated banks were derived on the basis of authorized principles [11] and the most critical soil use scenario dose factors (see Table II). If in accordance to the accepted restoration approach, both the potential and the actual individual risk from the contaminated banks must not exceed an effective dose level of 1 mSv/y. The following average ^{137}Cs activity levels for the banks correspond to this requirement:

- AL_{200} = 6.0 kBq/kg on 800 m^2 of surface area, or over a 300 m distance along the bank (use of 200 m^3 of soil), or
- AL_{50} = 8.0 kBq/kg on 200 m^2 of surface area, or over a 80 m distance (50 m^3) along the bank.

In addition, the following ^{137}Cs activity concentrations were proposed in conjuction with the first two concentration limits as cleanup criteria for selected parts of the contaminated banks:

- $AL_{.1}$ = 25 kBq/kg at isolated small spots on the canal banks, (ca. 1 m^2) and
- AL_{ws} = 4 kBq/kg on 2500 m^2 of surface area or over 1 km of the bank for installing warning signs.

4.3.1. Characterisation by verification of contamination compliance with the cleanup levels

To be in compliance with these criteria, it is expected that it will be necessary to remediate about 11,000 m^2 of the contaminated area on the Dudváh River banks, and 8,000 m^2 on the Manivier canal banks. As engineered flat terraces prevail on the Dudváh River banks, it will be sufficient to apply a clean soil cover over 9500 m^2 of the contaminated area, according to the authorised principles [12]. As a result, the volume of soil to be removed and disposed of safely amounts to about 1,100 m^3. However, selection of this remediation option for the flat terraces can not be considered the final decision, as it will be affected by flood control engineering requirements and public acceptance of restoration measures at this particular section as well.

4.3.2. Activity distribution of ^{137}Cs contamination at the site

The volume distributions of the activity concentrations in the soil on the banks are important characteristics of the site. It can be expected to be a non-linear distribution with a shift to values in the lower activity region of the curve. Concentration data for the site are available, as indicated in Figure 6, and are given in more detail in [12]. They were obtained on the basis of detailed mapping measurements of individual bank sections. Using these data distribution curves, one can demonstrate that clean-up measures, applied even only to the small part of the banks which is most contaminated (with 6 to 7 kBq/kg of ^{137}Cs), would lead to a significant improvement of the radiological situation at the site.

5. MONTE CARLO (MC) SIMULATIONS FOR PLASTIC SCINTILLATION DETECTOR RESPONSE ABOVE DIFFERENT CIRCULAR SLAB SOURCES

More accurate evaluation of the above described ^{137}Cs-contaminated spots along the Manivier canal bank requires the application of more sophisticated measuring and evaluation techniques. As can be shown that one single standard measurement reading is insufficient to determine the activity concentration in a circular slab source with unknown radius. This is, because a detector response above a circular soil source, in general, depends on three independent parameters: 1. the activity concentration A_m, 2. the circle radius that describes the surface area of the contaminated spot, and 3. the thickness of the contaminated slab (the last two parameters combined give the total amount of contaminated soil, M, at the location).

Field sampling and measurements for ^{137}Cs have shown that a typical depth distribution can be well represented in effect by a homogeneous slab source having a thickness of 10 cm. Thus the remaining two parameters have to be assessed by more then one measurements, using for instance the detector responses at several (at minimum at two) pre-defined measuring positions, or for different shielding of the detector (e.g. ratio of unshielded- to shielded detector response).

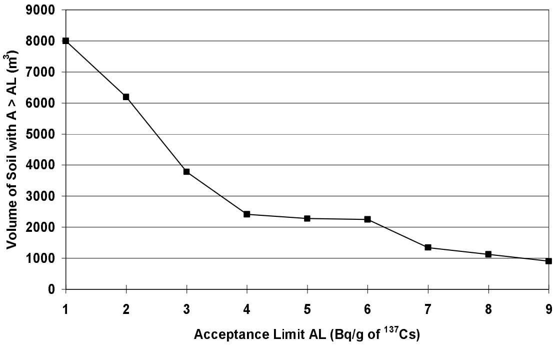

FIG. 6. Volume of soil to be removed from the contaminated sections of the River Dudvah and the Manivier Canal banks as a function of the Acceptance Level AL.

For simple measurement techniques such as standard dose-rate meters (i.e. not for spectrometric systems), scattered photons play an important rôle for the detection of gamma-radiation. Therefore, knowledge of the complex photon transport through the source, the source-to-detector space, and the detector media, are required to unambiguously solve the given problem.

In this context, Monte-Carlo (MC) simulation of selected (sufficiently sensitive) detector responses (from the NB3201 detector with 76×76 mm plastic scintillator) have been carried out by a joint efforts between the VUJE institute and the Comenius University's Nuclear Physics Department. The GEANT Monte-Carlo code from CERN [13] for homogeneous 10 cm circular slab sources geometries was employed for these purposes. The main objective of the work was to investigate and

- to find the dependence of detector response on the radius of a circular slab sources for two different measurement distances above ground, and for shielded and un-shielded detector respectively, and
- to develop appropriate measuring procedures for estimating the effective radius of the measured ^{137}Cs spot in soil, including its total activity or activity concentration.

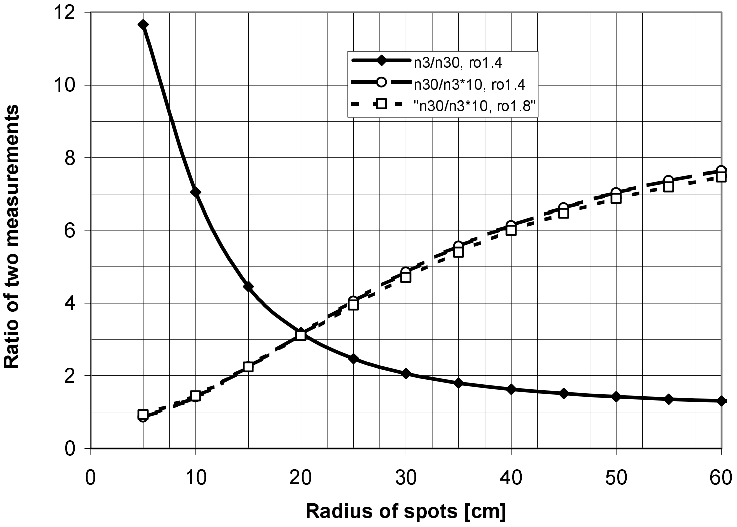

FIG. 7. Monte Carlo simulation results for the ratio of two measurments positioned at 3 and 30 cm above the centre of a ^{137}Cs spot by NB 3201 plastic scintillator.

5.1. Monte-Carlo simulation results and their use for ^{137}Cs circular spots determination

The procedure of MC simulations is described in more detail in [4]. A first preliminary simulation for simplified detector arrangement, consisting of a 76 × 76 mm plastic scintillator only, has been carried out in 1997. Typical simulation results for the measurement ratio for two selected heights above ground and sensitivities of the detector are shown in Figures 6 and 7, respectively.

Figure 7 shows a strong dependence on the circular spot radius for the calculated ratios between measurement at the heights 3 cm and 30 cm above ground in the center of spot. At the same time, it can be seen that this parameter allows to evaluate the effective diameter of a circular source. The measuring heights above ground tested were selected based on an evaluation of available data for un-scattered photons. The uncertainty of this type of assessment may be generally large, but it allowed to distinguish the different sizes of ^{137}Cs spots reliably. Therefore, it will form the basis for a more accurate characterization of existing spot-like contamination on the river banks.

The MC simulations in 1998 were focused on achieving as realistic detector response simulations as possible. The sensitive material and the construction, that is a small CsI(Tl) detector and a metallic Fe-ring used for detector NB3201 energy and angle dependence compensation, were considered in the simulation model. Lightly shielded (2 cm of lead) and unshielded detector responses for the two distances (3 and 30 cm) above the centre of circular soil slabs of 10 cm thickness with different radii from 5 to 90 cm have been calculated by MC simulation. Simulations for circles of up to 150 cm radius were performed for the unshielded detector in order to study the angular distribution of the total photon fluxes above a homogeneous soil slab ^{137}Cs source of 10 cm thickness.

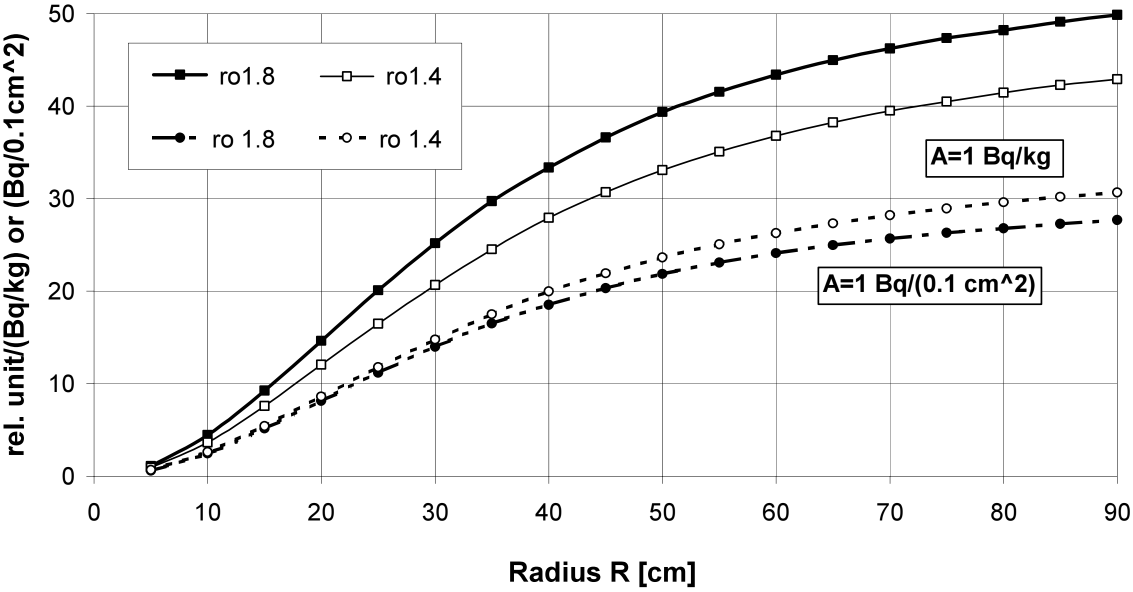

FIG. 8. Monte Carlo simulated relative sensitivity of the NB 3201 plastic scintillator detector to contaminated circular ^{137}Cs spots with various radii and two soil densities.

The above calculations were also performed for different soil densities in order to study their effect on the detector sensitivity. These results are shown in Figure 8, where a slight dependence of detector response sensitivity on the soil density is observable. The graphs in Figure 8 also demonstrates that the density effect is somewhat smaller in the case of surface activity determinations as opposed to the mass activity determinations.

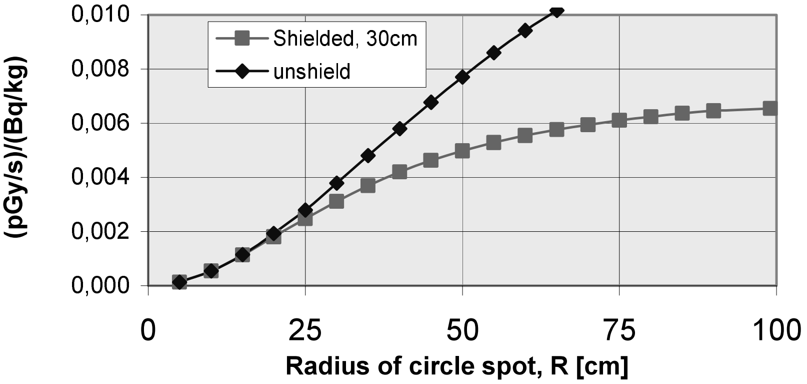

FIG. 9. Monte Carlo simulation results for the sensitivity of shielded and unshielded NB 3201 plastic scintillator detector 30 cm above the centre of a ^{137}Cs spots depending on the spot radius.

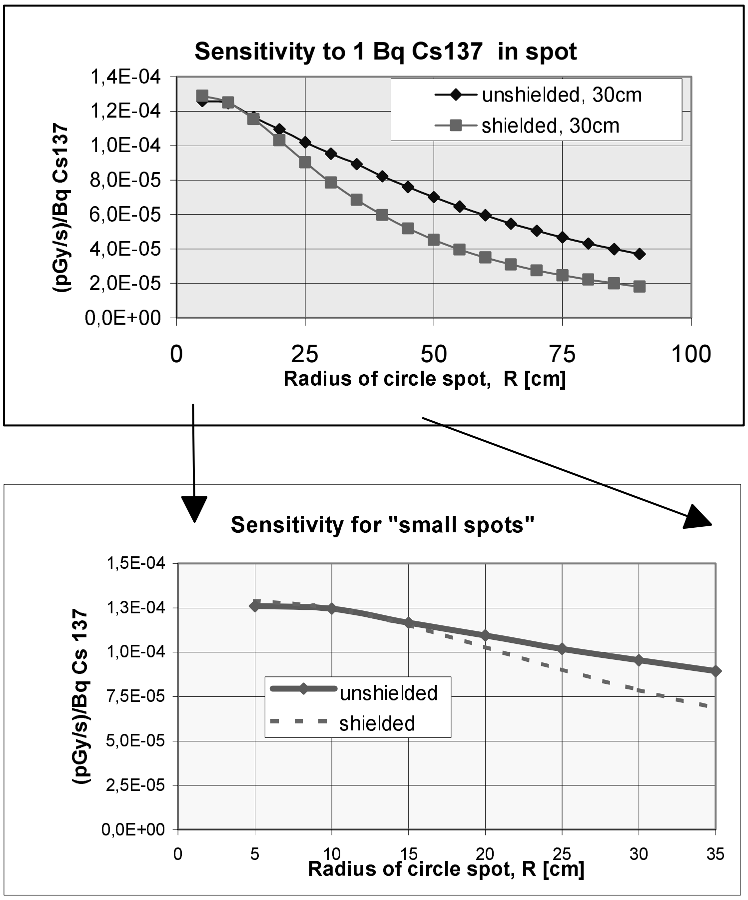

FIG. 10. Monte Carlo simulation results for the sensitivity of shielded and unshielded NB 3201 plastic scintillator detector 30 cm above the centre of a ^{137}Cs spots normalised to the total activity over the spot with different radii.

Figure 9 shows the calculated (MC) sensitivity of a shielded and an unshielded detector respectively, positioned at 30 cm measuring height above the spot centre, as a function of the spot radius. The detector dose response is given in pGy/s to a total of 1 Bq/kg ^{137}Cs in soil in circular spot. As can be concluded from this graph, for larger spots of contamination, it would be feasible to use a shielded detector and the dose rate-to-activity concentration conversion factor (C_M). The sensitivity factor for the shielded detector shows only slight source radius dependence above a radius of 60 to70 cm. This is, because the spot surface area effectively seen by the shielded detector increases rather slowly above this radius, in contrary to the unshielded detector, for which the effective field of view is much larger.

The two graphs in Figure 10 show the same simulated detector sensitivities as in Figure 9, but, which are normalised to the total contamination in the spot, that is the dose response to an activity of 1 Bq ^{137}Cs distributed over contaminated slabs of varying radius. For smaller circular spots, e.g. with radii smaller than about 30 to 40 cm (i.e. a surface area of less than 0.3 to 0.4 m^2), the detector sensitivity to the total activity in the spot shows only slight dependence on the circular spot size. This means that the detector dose-rate-to-total-activity conversion factor of the unshielded detector (C_T) at the selected measuring height (e.g. 30 cm) above the centre of the spot gives the most reliable estimate of the total activity in the spot, as is shown in the lower part of Figure 10. For mass activity concentration assessment in the spot, the spot radius estimation may be used according to Figure 7, as is shown in Table IV. According to these simulation results, it appears that the activity distribution within the contaminated slab is becoming less important as the radius of the contaminated spot decreases to sufficiently low values, i.e. the relative spatial resolution of the detector configuration decreases.

The proposed method for ^{137}Cs field monitoring is based on dose rate measurement technique and exploring the ratio of two measurement readings for different measuring height above the centre of contaminated spot. The method is currently under final development and verification in real field conditions.

5.2. Proposed measuring procedures for estimating ^{137}Cs spot contamination

The conclusion from the MC simulation results and the discussion in the previous chapter is that the proposed method exploring the ratio of two dose rate measurements at different heights above the identified centre of contaminated spot is appropriate for determination of the gamma contamination in regular more or less circular spots. The influence of shape un-regularities to accuracy of mentioned above determination would be subject of further investigations. Imagination about significance of the mentioned problem gives the data for detail mapping above two selected spot surfaces in Table III and its expression in integral central dose rate data in Table IV. The protocol of measurements for regular spots can be summarised as follows:

- Identify locations with enhanced (above background) detector response;
- Identify the centre of contaminated spot by scanning at 3 cm distance above ground with a shielded detector and determine its maximum dose - D3;
- Determine the maximum dose at 30 cm above ground for the same, i.e. the shielded detector- D3O;
- Calculate the ratio of net detector signal (e.g. above the background) P3/30 = (D3-B3) / (D30-B30), where B is the background dose response of the detector used;

— Referring to the shielded detector response-graphs from the Monte Carlo simulations, determine the effective radius “R” of a circular spot and classify its size (detector specific): small ($R < 30$ cm), medium (30 cm $< R <$ 60 cm), and large ($R > 60$ cm);

— Referring to the net detector response at 30 cm above ground (D30 – B30) for an unshielded detector and the respective MC results, determine the total and/or specific activity A at the spot. It depends on the estimated size of spots:

(1) for small spots - preferably use dose-to-total activity conversion factor $C_T(R)$ [(pGy/s)/Bq], determine total activity A_T and recalculate activity concentration $Am= A_T/V\rho$;

(2) for large spots use MC values for dose-to-activity concentration conversion factor C_M [(pGy/s)/(kBq/kg)] and determine directly Am (as it is usual, the direct determination of the total activity is not the goal of the measurement),

(3) for medium spots the action is similar as in the case of small spots, only be aware that the result is strongly affected by the estimated effective radius of spot; therefore in addition, check the spot size and shape more carefully by scanning with the shielded detector at 3 cm above the ground.

5.3. Field testing of the procedure

Some limited field testing of the developed protocol was performed within the framework of national sub-project PE 1024. Detailed mapping measurements above the surface of identified spots were carried out directly on the contaminated Manivier canal bank in order to assess the dose rate and the areal distribution of the ^{137}Cs contamination within the spots. Typical results are given in Table III. Azimuts according to the “hour-hand” system are used in Table III to indicate the survey lines starting from the centre of an previously identified spot. As can be seen in Table III, spot No 2 is of aproximately circular shape with a radius of about 20 cm. A different case represents spot No. 1, which is, according to survey results, of irregular shape and elongated in direction ”6” hours.

Examples for the application of the protocol developed are given in Table IV for the two spots mentioned. The results from dose rate measurement at 3 and 30 cm height above the centre of the spot and the parameters estimated from the MC simulation curves are presented in Table IV. The last two rows of Table IV contain a comparison of two estimates of activity concentrations in the two spots. One estimate, A_m in Table IV, is based on the protocol developed for in situ dose rate measurements; the other, A_m-lab, is based on standard laboratory determinations on samples from taken from the centre of the spots (circular slab 10 cm thick and with a 20 cm radius) identified by in situ measurements and scanning (see Table III), and on semiconductor gamma spectrometry analyses of the homogenised soil sample. As it is shown in Table IV, the activity concentrations are underestimated by about 15% in the case of the irregular shaped spot No 1.

The actual surface distribution of the contamination over the spot surface area, i.e. divergence from the circular shape, of course, can influence the activity concentration estimations (see results in Table IV), however more detailed investigation of this effect was beyond the scope of this CRP.

TABLE III. DETAILED MAPPING RESULTS IN READING UNIT OF THE DETECTOR [pGy/s] FOR THE MANIVIER CANAL BANK AS A FUNCTION OF THE DISTANCE R [cm] FROM THE CENTRE OF THE CONTAMINATED SPOT
(shielded detector nb3201 at 3 cm above the contaminated spot).

Spot No. 1	**r**						
Direction	**centre**	**10cm**	**20cm**	**30cm**	**40cm**	**50cm**	**60cm**
2	297	159	111	70	41	33	
3	297	255	159	134	105	68	70
4	297	266	211	139	105	74	55
6	297	259	222	195	151	150	120
8	297	196	124	76	49	43	37
9	297	210	130	92	68	64	57
10	297	191	101	57	49	33	
12	297	220	162	86	55	42	31

Spot No. 2	**r**						
direction	**centre**	**10cm**	**20cm**	**30cm**	**40cm**	**50cm**	**60cm**
1.5	336	197	83	60			
3.0	336	274	100	60	109	110	126
4.5	336	195	115	70			
6.0	336	216	114	90	70	55	47
7.5	336	232	138	90			
9.0	336	242	75	72	50	37	
10.5	336	219	130	97			
12.0	336	206	101	68	46	33	

Note: Directions – azimuts according to hour hand system, e.g. 3 hr - east.

TABLE IV. EXAMPLES OF DETERMINATION OF ^{137}Cs CONTAMINATED SPOTS ON MANIVIER CANAL (cf. TABLE III) ACCORDING TO THE PROPOSED PROTOCOL (SHIELDED PLASTIC SCINTILLATION DETECTOR).

	Spot No.1	**Spot No. 2**
DP30 [pGy/s]	110	101
DPB30 [pGy/s]	21	21
N30 [pGy/s]	89	80
DP3[pGy/s]	297	336
DPB3[pGy/s]	24	27
N3[pGy/s]	273	309
P= N3/N30	3,06	3,9
R [cm]	24	20
C_T(R) [pGy/s/kBq]	0.103	0.109
A_T(N30/C_T) [kBq]	870	730
M (R) [kg]	25.3	17.6
A_M=A_T /M [kBq/kg]	34.4	41.5
A_M-lab. m. [kBq/kg]	39.2±3	40.0±2

Abbreviations:
DP3, DP30= dose rate at 3, 30 cm; DPB3, DPB30 = background dose rate;
N= net signal DP-DPB; C_T= dose rate to total activity (in spot) sensitivity factor;
A_T, A_M= total and mass activity respectively, M= V(R)×ρ the mass of soil in the spot
A_M-lab= activity concentration of the soil sampled and determined by laboratory semiconductor gamma spectrometry

6. SUMMARY AND CONCLUSIONS

A re-evaluation of a restoration project for a river bank contaminated by ^{137}Cs was conducted at the Bohunice NPP site on the basis of a comprehensive and detailed site characterisation. Cleanup levels as the average activity concentrations 6 or 8 kBq/kg of ^{137}Cs in the soil were developed taking into account the site specific conditions in the bank use and the continuously contaminated bank area. Owing to the application of consistent site characterisation techniques and dose assessment methods, excessive removal and disposal of soil will be avoided during the new bank restoration project, which is now under preparation.

Monte Carlo simulations were used to propose more sophisticated monitoring techniques for remediation monitoring purposes on the spot-contaminated Manivier canal banks. The proposed method is based on sophisticated measuring procedures using the ratio of two readings for two different (3 cm and 30 cm) measuring distances above the contaminated spot. The proposed measuring protocol was found to be appropriate for the determination of small circular contaminated spots.

The CRP in particular helpt to address the problems associated with the

- identification of discrete, spot or strip, contaminations on a larger site;
- development of strategies for the identification of discontinuous areas of contamination;
- selection of sensors sufficiently sensitive;
- the evaluation of detector responses as a function of the shape and distribution of a contamination;
- verification by laboratory and field testing of the methods developed.

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EXPERIENCE WITH ENVIRONMENTAL BEHAVIOUR OF Pu AT PALOMARES, SPAIN

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ABSTRACT

In 1966 as a consequence of an accident involving two US military aircraft near Palomares two weapons were dropped and their plutonium contents partially scattered. The objective of this paper is to present the most important activities carried out at Palomares in order to characterise the plutonium contaminated site, and to gain further knowledge of the plutonium behaviour in that area.

The behaviour of plutonium and americium associated with the soil for more than 30 years has been studied. These studies included chemical and mineralogical investigations of plutonium in the different fractions of the soil, lung solubility of plutonium associated with the breathable fraction, and statistical estimations of the inventory of plutonium remaining in the top first 45 cm of the soil at the most contaminated area in which remediation work was carried out. Other studies focused on the determination of the isotopic relations of plutonium and americium in the surface level.

The results show a large variation in plutonium particle sizes in the soil from Palomares. Theoretical calculations estimate that the plutonium remaining in the area could still amount to 16% of the total contents in two bombs. Sequential extraction shows that the highest mobility stage of plutonium represents 0.2% of the total extracted, the remainder (93%) being in the residual fraction. The particles show very low solubility in a simulated lung fluid, and the enhancement factor, defined as the ratio of activity concentration in the inhalable fraction to that in the bulk parent soil was found to be 0.26.

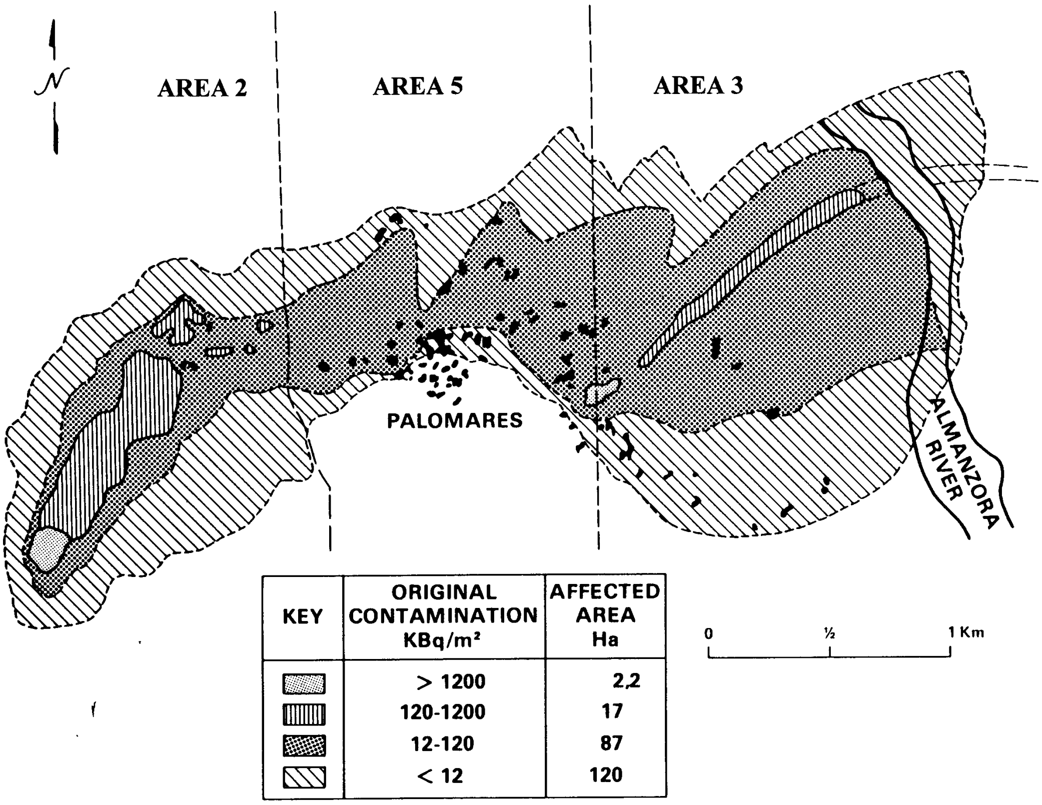
1. INTRODUCTION: PURPOSE AND SCOPE

The purpose of this contribution is to present the most important aspects of the behaviour of the Pu contamination in the Palomares area (Almería, Spain), which occurred in 1966 as a consequence of an accident of two US military aircraft during a in-flight refuelling operation.

Of the four thermonuclear bombs which were dropped by a B-52 bomber, two encountered problems with their parachutes and fell to the ground unchecked, where part of their fissile material contents was dispersed. As result, 226 ha of urban, agricultural and waste land were contaminated with different levels of activity (Fig. 1).

Immediately after the accident, and before the remedial actions were taken, alpha surface contamination levels were measured with proportional counter PAC-15 alpha detectors. A so-called “zero” line was defined and some study plots were established, from which soil and vegetation samples were taken periodically for radioactivity analysis.

The decontamination and remediation work of the zone consisted of removing layers of soil, as well as of highly contaminated vegetation [1]. Once these tasks were completed, the resulting wastes were sent to the USA in drums.



KEY	ORIGINAL CONTAMINATION KBq/m²	AFFECTED AREA Ha
▩	> 1200	2,2
▥	120-1200	17
▒	12-120	87
▨	< 12	120

FIG. 1. Map of the palomares ares showing levesl of contamination.

The temporal storage and loading of the drums were carried out in one of the contaminated areas, which according to its natural characteristics (hills with scarce vegetation) was considered as the most suitable; this zone, as a result, has higher residual contamination levels than the remainder.

An environmental and personal surveillance programme was established after the cleanup work was completed [2].

A range of human activities since the time of the accident have significantly altered the landscape at Palomares. The intention of the changes was to increase the total area available for cultivation, to store more water for crop irrigation, and to conserve moisture by reducing evaporation. These activities have increased re-suspension and re-distribution of plutonium and americium [2].

Greenhouses were constructed to allow crop production all the year round. Although these plastic enclosures have no floors, they prevent any removal of residual Pu contamination from the soil by wind action and re-suspension. However, because the structures retain moisture and have no floors there exists the potential of a change in the Pu soil chemistry with increasing time.

In 1986 a ‘co-operative’ reservoir was constructed to hold 151,600 m^3 of water for irrigation. A further pond was constructed in 1988, bringing the total storage capacity to about 347,000 m^3. These reservoirs will now provide water for the farmers, if allowed, to extend farming down to and beyond Impact Point 2 (Fig. 1). Also, the topography of the land in Area 2 was significantly altered as the land owners redistributed soil taken from hillsides to increase the amount of level land available for cultivation.

Since 1986, the environmental behaviour of Pu and Am, associated with the soil then for 20 years, has been studied [3]. The first goal of our group is to study and to characterise the contamination, including the chemical and mineralogical associations of Pu with the different fractions of the soil. Another goal is to investigate lung solubility of Pu associated with the breathable fractions.

A new statistical calculation of the inventory of Pu remaining in the top 45 cm of the soil in Area 2 was made using Pu data from radiochemical analyses. Other studies dealing with the determination of the isotopic relations of Pu in the surface level are being performed.

2. SOIL CHARACTERIZATION

2.1. Soil particle-size and activity measurements

Size distributions of particle-bound Pu in Palomares soils studied by several techniques show a large variation. Iranzo et al. [3] showed gross radioautographic evidence of the large variation in Pu particle-sizes in soil from Palomares.

Soil samples taken in 1981 from a depth of 15 cm at several study plots were measured for ^{241}Am and Pu. Each sample was treated by maceration and ultrasonic separation and then divided into eight size fractions: Φ= 0-5, 5-10, 10-20, 20-40, 40-63, 63-250, 250-1000, and >1,000 µm. Fifteen-gram samples were measured for ^{241}Am using low energy photon spectrometry with intrinsic germanium detectors, and for Pu using alpha spectrometry [3].

The $^{239+240}$Pu/^{241}Am ratio was almost constant with a large (0.87) coefficient of correlation within each size fraction, except for the 5-3B plot sample. The activity mass concentration for both Am and Pu was highest in the 63-250 µm fraction except for plot 5-3B, where the maximum activity mass concentration occurred in the 0-5 µm fraction [2].

An association between specific mineral components of the soil and concentrations of Pu and ^{241}Am was found using autoradiographic analyses. The main concentrations of alpha tracks were found in fragments of iron oxy-hydroxide and, to a lesser extent, in carbonate (dolomite) and muscovite-illite fragments. The potential leaching of Pu, estimated from concentrations measured in the maceration-elution water, was in the range of 2×10^{-4} to 21×10^{-4} % of the total soil Pu. The larger values were associated with the most oxidized and least alkaline soils. Of particular interest is that the soil in plot 5-3B, which was not contaminated by the original accident, showed 57% of the sample activity in the <5 μm fraction.

The particle-size distribution of soils collected in November 1986 close to Impact Point 2 was reported for various soil fractions [4]. The samples were subject to maceration and ultrasonic dispersion. From each sample, eight size fractions were obtained with the following intervals: 0-5, 5-20, 20-40, 40-63, 63-125, 125-250, 250-1,000, and >1,000 μm diameter. The silt-size particles (5-63 μm), and the sand-size particles (≥63 μm) accounted for 55% and 36% respectively of all the particles; the remaining 9% of the particles were less than 5 μm diameter. Twenty percent of the soil particles were 10 μm or less in diameter; these 5 and 10°μm diameter size ranges are generally thought to be the fraction retained in the alveolar lung after inhalation. The actual deposition value is density dependent, being smaller for higher density particles.

The Pu contents of the eight particle-size fractions were reported for these soil samples [4]. The total Pu activity in the silt fraction (5-63 μm), and in the sand fraction (≥63 μm) accounted for 63% and 35% respectively of all the Pu; the remaining 2% of the activity were associated with the <5 μm fraction. The average Pu concentration in the total sample was 1,944 $Bq·g^{-1}$. The concentration values range from 254 to 6,389 $Bq·g^{-1}$ (Table I). The observed bi-modal distribution shows peaks at 3,370 $Bq·g^{-1}$ and 6,389 $Bq·g^{-1}$ for the particle size fractions 20-40 μm and 125-250 μm respectively. These size ranges represent 42 and 15% of the total Pu in the soil sample.

The 125-250 μm size fraction is of particular interest, as it accounts for 15.39% of the total Pu, yet only 4.60% of the mass of the soil sample (Table II). Because of the high Pu concentration (6,389 $Bq·g^{-1}$), the Pu concentration was studied as a function of soil density and magnetism for components of this fraction. More Pu was found in the more magnetic classes due to the Pu compounds being associated with Fe minerals having strong magnetic properties. Pu bound to Fe minerals or compounds will have higher magnetic susceptibility and lower density, so these physical properties can be used to isolate particulate Pu and, therefore, 'hot' Pu particles. The highest activity levels (65%) were found in the class of larger density. Three density ranges were chosen; <2.89, 2.89 to 3.3 and >3.3 $g·cm^{-3}$. For the lowest and intermediate densities, six magnetic fractions were chosen, and four were chosen for the highest density fraction. Pu concentration increased with increasing density; 1,711, 4,707 and 116,379 $Bq·g^{-1}$ for the lowest, intermediate and highest densities, respectively. The total Pu was distributed as follows: 29, 5, and 65% for the lowest, intermediate and highest densities, respectively (Table III). In general the Pu concentration increased with magnetic class in the lowest and intermediate densities. In the highest density fraction, the maximum Pu concentration of 502,471 $Bq·g^{-1}$ was found for the lowest magnetic susceptibility (probably particulate Pu). Most of the Pu in that fraction corresponds to the particulate form.

The chemical and mineralogical composition of the soil was also studied. The soil is of siliceous-carbonic nature; its main chemical components, accounting for 87% of the total mass, are SiO_2, Al_2O_3, CaO and CO_3. There is a general tendency for quartz and feldspars to decrease, and carbonates and phyllosilicates to increase as particle-size decreases. As shown in Table IV, dominant minerals in the gross soil sample are quartz (39%), carbonates (37%) and plagioclase (13%); carbonates, including calcite (30%) and dolomite (7%), and there is a small amount of gypsum (<0.5%). The following account for 94% of the minerals in the <5°µm diameter particles: calcite (58%), plagioclase (11%), quartz (10%), mica (8%) and chlorite (7%) [4]. The 63-125 µm particles are high in quartz (47.39%) and calcite (27.42%).

2.2. Geochemical associations of plutonium in soil

In order to gain further insight into the geochemical associations of Pu in the soil, a sequential leaching method was applied to a contaminated soil from Palomares. The method employed is based on McLaren and Crawford [5], as modified by Cook et al. [6], and by the work of the authors [7], see Table V.

The procedure was carried out in seven consecutive stages, using two parallel samples, each weighing five grams. The soil activity for $^{239+240}$Pu is 1,727±180 Bq·gr^{-1}, with a ^{239}Pu/^{238} Pu ratio of around 54 (1992).

The results obtained for $^{239+240}$Pu in the different eluates are shown in Table VI. The most mobile fraction of $^{239+240}$Pu, which is made up of the *soluble*, *exchangeable* and *inorganically adsorbed* fractions, constitutes only 0.2% of the total $^{239+240}$Pu extracted. Associated with organic matter are 6% of the total $^{239+240}$Pu , and the remaining approximately 93% are associated with sesquioxides or are in the two residual fractions (sesquioxide-bound Pu amount to 34% and the residual part to 59%). The ^{238}Pu distribution does not differ substantially from that of ^{239}Pu, except for the *soluble* and *exchangeable* fractions, where the ^{238}Pu percentage is around three times the ^{239} Pu percentage, which brings about a variation in the ^{239} Pu/^{238}Pu ratio.

2.3. Lung solubility

The purpose of the experiments performed is to study the pulmonary biokinetics (by means of simulation) of plutonium and americium associated with contaminated soil. The method used takes into account different size-fractions of the soil: <5 µm (respirable fraction), 20-40°µm, 125-250 µm and total soil. To simulate the pulmonary system, small amounts of each dust sample were placed in a tube-shaped, semi-permeable membrane closed at both ends. Each membrane was placed in a plastic bottle with 400 ml of pH 7.3 physiological solution. Each bottle was kept at 37 ºC and was shaken for half an hour every three hours, so as to simulate the physiological conditions of the lung. Each week the membranes were transferred into another bottle with fresh physiological solution, and the solutions were analysed using radiochemical techniques.

The chemical composition of physiological solution used contained inorganic salts (Na, Ca, NH_4, Cl, SO_4, PO_4), and amino acids (glycine, Na_3Cit).

The first stage of the experiment lasted for 3 months. During this period it was observed that the amount leached did not exceeded 0.5% of the total plutonium contained in the sample. Another simulation experiment was performed with two more aliquots of the fraction <5 µm. During the first weeks of this experiment similar percentages of ^{239} Pu+ ^{240}Pu and ^{241}Am were dissolved, but the percentage of ^{238}Pu was several times higher. The total amount of $^{239+240}$Pu and ^{241}Am extracted during the first 140 days were less than 0.58% and 0.32% respectively of the total $^{239+240}$Pu and ^{241}Am contained in the respirable fraction of soil. For ^{238}Pu the values were about 2%.

Six months after the start of the experiment, the physiological solution used in one of the samples was replaced by the specific reagents for sequential leaching, in order to establish the geochemical associations of the radionuclides not extracted in the pulmonary simulation leaching experiment. The method employed is based on that of McLaren and Crawford [5]; ragents and fractions extracted are shown in Table VII.

The sequential leaching of the two samples (taken 6 months and 1 year after the start of the pulmonary simulation experiment) with acetic acid and sodium pyrophosphate showed that the *specifically adsorbed* and *organically* bound fractions had been reduced, or have even disappeared after one year into the pulmonary leaching experiment. This result indicates that the radionuclides in *exchangeable*, *specifically adsorbed* and *organically* bound forms are the most readily leached by physiological solution. These fractions represent less than 4% of the total activity for $^{239+240}$Pu and 8% for ^{238}Pu in the total soil sample.

It is important to underline that in the pulmonary simulation experiments with the respirable fraction sample (employing 3 aliquots), the ^{238}Pu extraction, especially during the first four weeks, was higher than the $^{239+240}$ Pu extraction obtained. This phenomenon was not observed in simulation experiments with the other soil fractions. The water solubility of ^{238}Pu, when associated with relatively large plutonium oxide particles (>1 µm), would be expected to be greater than the solubility of $^{239+240}$Pu oxide particles of similar size, as a result of crystal damage and radiolysis arising from the greater (by approximately a factor of 270) specific activity of the ^{238}Pu.

During the first six weeks the percentage of the activity leached in the respirable fraction experiment, is similar for both $^{239+240}$Pu, and ^{241}Am. The ^{238}Pu activity percentage leached is about 4 times higher than that of $^{239+240}$Pu; the difference being more pronounced during the first three weeks.

The results obtained from pulmonary leaching experiments performed with the 20-40 µm and the 125-250 µm granulometric fractions, as well as the total soil show, in all cases, a similar behaviour of the plutonium isotopes. However, in the experiment performed with the respirable fraction, the most soluble form was ^{238}Pu. The different behaviour of the breathable fraction of the Palomares soil in the simulation experiments can be explained by its different mineralogical composition (see Table VIII). Correlation factors between the plutonium activity concentration in each granulometric fraction and the abundance of the minerals identified in each fraction were calculated for selected samples of the Palomares area and are shown in Table IX. These factors show a correlation between the plutonium activity concentration and the abundance of minerals such as chlorite and illite-muscovite. Both of them are phyllosilicates, i.e. are formed by sheets 14 and 10 Å thick respectively, with inter-layer cations like K, Na, Fe, Mg. Plutonium and americium are mainly sorbed to the surface of these minerals, but a certain amount of these radionuclides can also be occluded or trapped into the inter-layer space, judging from the results of the extraction step with the HF-HNO_3 mixture.

3. INVENTORY OF Pu

3.1. Inventory of Pu estimates for zone 2-0

Re-surveying of Pu contamination in certain areas around Palomares has been initiated with the objective to provide estimates of the residual plutonium and americium in the area. Pu and Am depth distributions in Area 2 are the result of the decontamination activities carried out by American troops, rather than caused by downward migration or accumulation of silts. Soil collected in other zones was buried in two big pits in this area and then was covered with the material previously excavated.

Thirty-seven sampling points in the most contaminated zone were selected for soil sampling to a depth of 45 cm. At each point samples were taken at five depth intervals (0-5, 5-15, 15-25, 25-35, and 35-45 cm). The samples were analysed by α-spectrometry for Pu, and by low energy photon spectroscopy for ^{241}Am. The concentrations of Pu and ^{241}Am were not distributed homogeneously. Both radionuclides were found down to a depth of 45 cm in the valley, while on the hillsides the concentration of Pu and ^{241}Am below the first 5 cm was at least one order of magnitude less than that in the valley. An average Pu concentration of about 82 $\text{kBq}\cdot\text{kg}^{-1}$ was found, with a wide variation between minimum to maximum. The average ^{241}Am concentration was about 13.4 $\text{kBq}\cdot\text{kg}^{-1}$, with broad range of values around the mean. For both Pu and Am, the highest concentrations were found near the original Impact Point 2. A gross estimate of the plutonium content of the 6.5 ha site was 6×10^{11} Bq (about 17 Ci) [8].

A recent study of a selected 5.25 ha site in Area 2 suggests an inventory of 2.86×10^{12} Bq (77°Ci) Pu to a depth of 45 cm [9]. The data are from surveys conducted in 1986. At 37 locations Pu samples were taken at six depth intervals down to 45 cm (Fig. 2). Various statistical procedures were employed to calculate the volume, mass and Pu concentration of each soil layer (0-5, 5-15, 15-25, 25-35, and 35-45 cm), i.e for the entire 5.25 ha to a depth of 45 cm. The relative soil density was found to be 1.6 $\text{g}\cdot\text{cm}^{-3}$. The 2.86×10^{12} Bq (77 Ci) inventory equals to a mass of about 1.3 kg Pu. Although the area covered is somewhat limited, it is thought to represent the most contaminated land within Area 2. Calculations indicate that the removal of the top 5 cm of the soil, containing 1.45×10^{12} Bq Pu, and weighing about 4.1×10^{3} metric tons, would remove 51% of the total inventory. Figure 2 shows the location of the study area and the residual Pu concentrations in the soil.

This 2.86×10^{12} Bq inventory equals to about 33% of the Pu in a nuclear weapon assumed to contain 4 kg Pu. However, we have to keep in mind that soil contaminated with Pu from the weapons both in Areas 2 and 3 was tranferred to Area 2, which is within the 5.25 ha studied, for packaging in 200 l steel drums.

Estimates of total Pu released from weapons 2 and 3 are very uncertain. The amount of Pu originally in weapons 2 and 3 is classified information and we do not know how much Pu was removed from Spain for disposal in the U.S. Neither do we know with any level of confidence the amount of Pu remaining after the initial remediation in 1966. We do know that, based on the relatively crude measurements made using the PAC 15 detectors, Pu concentrations below 1.2×10^{3} $\text{Bq}\cdot\text{m}^{-2}$ were ploughed into the soil to reduce original surface concentrations.

3.2. Plutonium vs. americium

The isotopic composition of Pu in the weapons-grade Pu has an activity ratio for ^{241}Pu *vs.* $^{239+240}$Pu of at least 5. Taking into account that ^{241}Am derives from the decay of ^{241}Pu, the activity concentration of Am is expected to increase with time. In order to estimate the correlation between Pu and Am, 45 samples from the area studied have been used, all of them with activity values for both radionuclides much higher than the LID. These data are plotted in Fig. 3. Pu and Am are not homogeneously distributed, as shown int the figure, though the highest concentration in both cases are located at the same points.

The linear correlation of $^{239+240}$Pu *vs.* ^{241}Am is shown in Fig. 4, the correlation coefficient being better than 87%.

4. SUMMARY AND CONCLUSIONS

During the present CRP a range of investigations were undertaken in the area of Palomares:

- Characterization of the biological behaviour of Pu bound to Palomares soil during more than 30 years;
- Estimation of pulmonar absorption parameters and inhalation dose factors;
- Calculation of a local pulmonar model for the Palomares area;
- Assessment of the Pu remaining in the upper 45 cm of the soil in area 2-0 following the decontamination activities (collection and disposal of contaminated soil) carried out in on agricultural lands. Previous investigations provided the essential foundation for these studies, giving us a valuable insight into and knowledge of the evolution of the contamination.

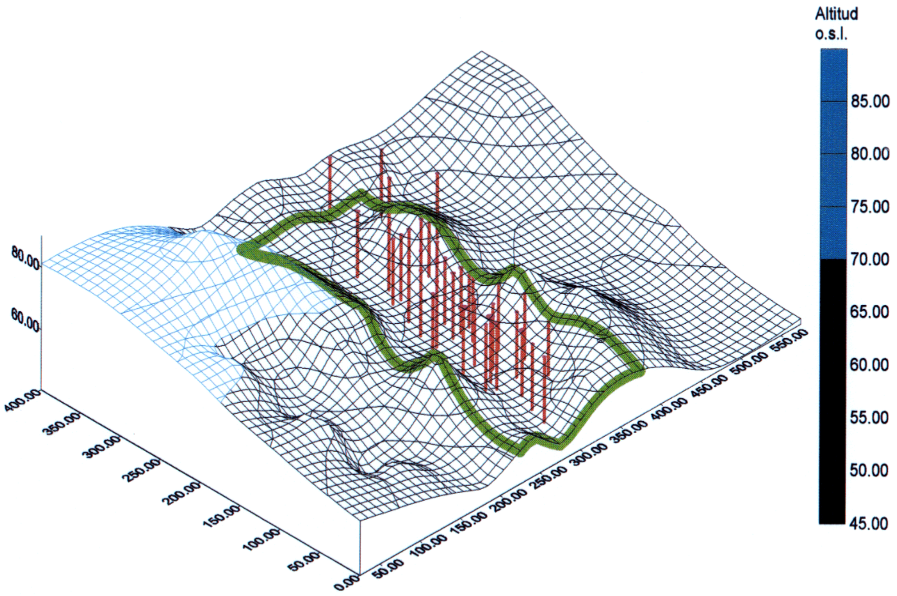


FIG. 2. Location of boreholes.

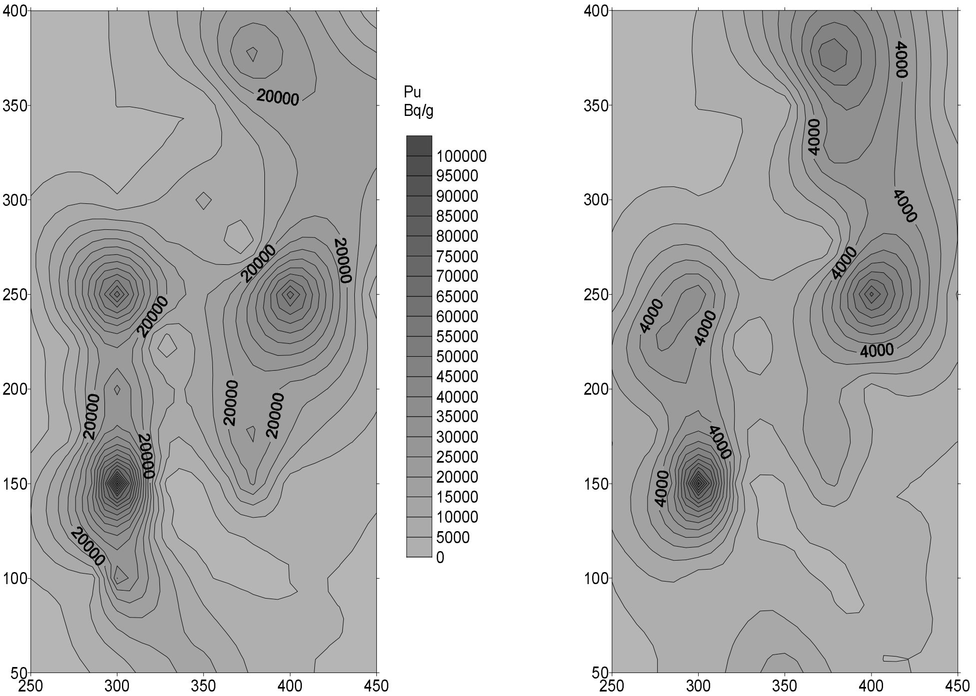


FIG. 3. Countour maps of the Pu and Am distribution.

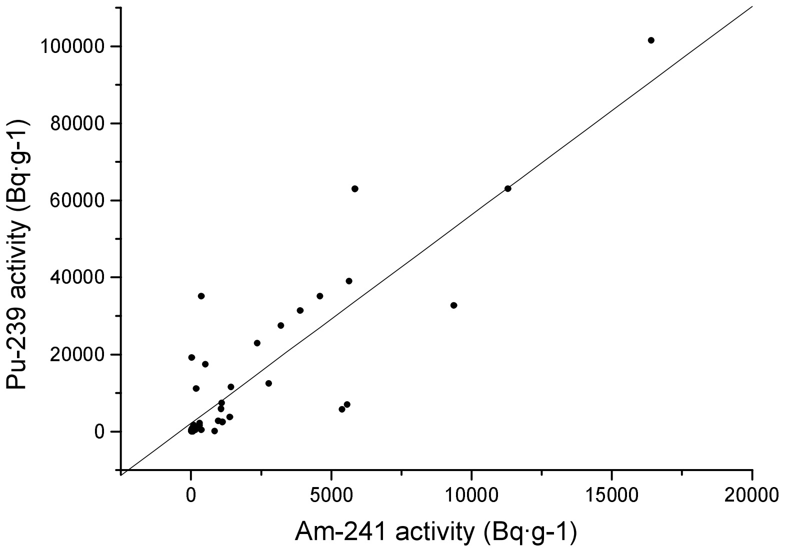


FIG. 4. Linear correlation of Pu activity vs. Am activity in soil samples.

Future work will focus on obtaining isotopic relations of Pu and Pu/Am in different types of samples in order to be able to determine possible similarities or differences in the behaviour of these radionuclides.

5. ACKNOWLEDGEMENTS

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TABLE I. PLUTONIUM ACTIVITY IN THE PARTICLE SIZE FRACTIONS OF SOIL

Size class [μm]	Abundance [%]	Pu [Bq·g^{-1}]	Pu Abundance [%]
Silts			
5	8.78	504±14	2.32
5-10	11.40	1333±79	7.96
10-20	8.77	1056±67	4.85
20-40	23.73	3370±210	41.37
40-63	10.77	1504±82	8.48
Sands			
63-125	13.79	2183±99	15.76
125-250	4.60	6389±315	15.39
250-1,000	2.87	904±42	1.36
>1,000	15.19	2544±9	2.02

TABLE II. PLUTONIUM DISTRIBUTION IN RELATION TO DENSITY AND MAGNETISM OF COMPONTS IN THE 125-250 μm GRANULOMETRIC FRACTION

Densimetric Class [g/cm^{-3}]	Magnetic class [× 10^{-6} cgsem]	Abundance [%]	^{239}Pu + ^{240}Pu	
			[Bq/g]	Abundance [%]
ρ <2.89	2.02	74.89	59.4 ± 9.1	0.84
	2.30	5.34	579.0 ± 87.8	0.58
	5.18	6.64	435.0 ± 65.2	0.54
	10.60	2.18	7741 ± 1161	3.18
	20.70	0.64	12211 ± 1856	1.47
	>32.40	1.32	91364 ± 13887	22.72
2.89< ρ< 3.3	2.30	1.14	244 ± 37	0.05
	5.80	0.33	1639 ± 249	0.10
	8.10	1.21	868 ± 130	0.20
	14.40	1.80	973 ± 146	0.33
	32.40	1.43	3295 ±501	0.89
	129.00	0.10	199564 ± 26340	3.76
3.3 <ρ	5.80	0.21	502471 ± 75371	19.88
	14.40	0.32	287720 ± 43158	17.34
	32.40	1.89	29967 ± 4495	10.67
	>129.00	0.56	16532 ± 25129	17.44

TABLE III. PLUTONIUM ACTIVITY DISTRIBUTION AS A FUNCTION OF DENSITY FOR 125-250 MICRON DIAMETER SOIL PARTICLES.

Density [g·cm^{-3}]	**Weight abundance [%]**	**Pu [Bq·g^{-1}]**	**Pu [%]**
<2.89	91.0	1,711	29.3
2.89-3.3	6.0	4,707	5.3
>3.3	3.0	116,379	65.4

TABLE IV. MINERALOGICAL COMPOSITION OF THE TOTAL SAMPLE AND SELECTED SIZE FRACTIONS

Mineral	**% of total Sample**	**125-259 µm**	**63-125 µm**	**5-10 µm**	**ß5 µm**
Quartz	38.61	59.75	47.39	12.45	10.28
Carbonates	37.55[a]	9.03	29.71	53.09	62.22
Plagioclase	13.46	15.69	12.29	15.66	10.74
Others [b]	10.38	15.53	10.61	18.80	16.76

[a] Calcite (30.09%), Dolomite (6.97%) and Gypsum (0.49%)

[b] Mica, Chlorite and Orthoclase

TABLE V. APPLIED SEQUENTIAL LEACHING PROCEDURE.

Fraction obtained	**Reagent**	**Volume/soil [ml·g^{-1}]**
Water soluble	Distilled water	20
Exchangeable	0.005M $CaCl_2$	20
Specifically adsorbed	0.05M Acetic acid	20
Organically bound	0.01M $Na_4P_2O_7$	100
Oxide bound	0.175M $(CO_2NH_4)_2$/ 0.1M$(CO_2H_2)_2$	75
Residual	7.8M HNO_3	70
Residual (strongly bound)	HF + HNO_3	70

TABLE VI. DISTRIBUTION OF PLUTONIUM IN THE VARIOUS FRACTIONS OF THE SEQUENTIAL LEACHING PROCEDURE

Fraction obtained	**^{239}Pu+^{240}Pu [%]**	**^{238}Pu [%]**	**^{239}Pu/^{238}Pu**
Water soluble	0.0036	0.01	19
Exchangeable	0.0054	0.013	22
Specifically adsorbed	0.2	0.18	55
Organically bound	6.0	7.8	50
Oxide bound	34.3	33.9	55
Residual	31.7	30	58
Residual (strongly bound)	27.8	28.1	54

TABLE VII. SEQUENTIAL LEACHING PROCEDURE FOR LUNG SOLUBILITY SIMULATION

Fraction extracted	Reagent
Specifically adsorbed	0.05M Acetic acid
Organically bound	0.01M $Na_4 P_2 O_7$
Oxide bound	0.175M $(CO_2NH_4)_2$/0.1M $(CO_2H_2)_2$
Residual	7.8M HNO_3
Residual (strongly bound)	HF + HNO_3

TABLE VIII. MINERALOGICAL COMPOSITION OF SIZE FRACTIONS

Size [µm]	Chlorite d(Å)-14	Illite moscovite d(Å)-10	Quartz d(Å)-4.24	Calcite d(Å)-3.03	Dolomite d(Å)-2.88
< 5 µm	67.39	46.48	6.65	100.00	5.42
$5 < \Phi > 10$	58.70	72.24	11.97	76.78	10.54
$10 < \Phi > 20$	58.70	59.62	21.51	61.30	7.53
$20 < \Phi > 40$	84.78	97.18	55.43	65.33	24.90
$40 < \Phi > 63$	62.22	100.00	58.09	39.00	100.00
$63 < \Phi > 250$	80.43	93.43	84.92	49.54	54.81
$250 < \Phi > 1000$	41.30	33.80	100.00	73.99	19.58

TABLE IX. MULTIPLICATIVE COEFFICIENTS OF CORRELATION BETWEEN THE ACTIVITY CONCENTRATION OF $^{239}Pu+^{240}Pu$ AND SELECTED MINERALS.

Sample	Clorite	Moscovite	Quarz	Calcite	Dolomite
1	0.82	0.90	-0.03	-0.56	0.45
2	0.85	0.90	0.31	-0.49	0.50
3	0.87	0.88	0.03	-0.55	0.50
4	0.97	0.74	-0.08	-0.15	0.21
5	0.75	0.41	-0.13	0.03	-0.07

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CHARACTERISATION AND RESTORATION OF CONTAMINATED LAND ON THE DOUNREAY AND HARWELL SITES OF UKAEA

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ABSTRACT

The historical mission of the United Kingdom Atomic Energy Authority (UKAEA) was to develop nuclear power in the UK. The UKAEA was formed in 1954 but work actually began at the Atomic Energy Research Establishment (AERE) Harwell in 1946 where the first experimental reactors were built. Additional sites were established at Windscale, Winfrith and Dounreay among others as the needs of the programme developed. Following the successful development of nuclear power, UKAEA's mission is now the restoration of its nuclear sites. The final stages of decommissioning require confidence that the land on which the facilities stand is clean enough to allow unrestricted public access. Characterisation of the ground is therefore an important factor in the final clearance stage.

This paper summarises progress on some of the work carried out within UKAEA during the period of the IAEA research collaboration at Harwell and Dounreay. There is continued interest in defining the level of background radioactivity on sites, clean up of known liabilities in disposal pits at the Harwell site and characterisation on the Dounreay site of areas where the land quality is suspect. In some cases it may be feasible to clean up the soil by various techniques such soil washing, and the final part of this paper describes developments concerned with soil treatment.

1. SITE CHARACTERISATION PROGRAMME AT HARWELL

A wide range of work has been carried out at Harwell in support of the Nuclear Power programme, from work involving highly active beta/gamma materials (typically fission product type inventories) to chemical processes where specific radionuclides were studied (such as pure beta emitters like strontium-90 or alpha materials including plutonium and americium isotopes).

The Harwell site management plan envisages the progressive release of areas of the site for commercial or residential development. In some cases this involves release of land from regulatory control, where the regulatory standards that have to be satisfied to achieve this end point are as yet unclear. Characterisation of the land is needed to prove that land which is believed to be clean is actually clean in practice, and also in order to determine the extent of remediation required if the land proves not to be clean.

1.1. Generic site characterisation developments

Although some areas of the site are believed to have no history of radioactive work, the clean status of the land needs to be demonstrated. The experience gained over the past two years has shown that reliance on a surface radiation survey, or a programme of core sampling, may not be sufficient unless coverage is total. Total coverage by core sampling is not always economic and it is therefore important that sampling is targeted well. This requires a combination of approaches to be used, in order to gather as much supporting information on the status of the land to identify any areas where specific investigation may be needed.

A series of factors are now considered in determining the approach to, and extent of any characterisation programme required, either as part of a remediation programme or simply in demonstrating land to be clean. These factors are discussed below, noting that there is a degree of interdependence and overlap between them.

- Consideration of end point usage;
- Derivation of required clean up targets;
- Likelihood of contamination present;
- Pre-characterisation investigations;
- Knowledge of background radioactivity present;
- Characterisation regime;
- Remediation programme;
- Final survey clearance against clean up targets.

1.1.1. Consideration of end point usage

The planned end point may be residential redevelopment, commercial redevelopment, agricultural use or simply making the land accessible as an open area. The extent of characterisation that is required will be dependent on the scale of the remediation planned which is turn dependent on the planned end usage.

1.1.2. Derivation of cleanup levels

Different clean up levels will be appropriate dependent on the proposed end use of the land. This will also depend on whether the clean up is regarded as a part of a process of intervention or part of a process of release from existing control.

Clean up targets are based on the results of modelling for specific end points, for a range of pathways, eg food consumption, contamination of groundwater, direct radiation from contamination present in the ground or from inhalation pathways as a result of resuspension of materials from the ground. Any characterisation regime will have to be capable of demonstrating that the derived clean up levels have been satisfied.

1.1.3. Likelihood of contamination being present

The scale of the characterisation programme that is required will depend greatly on the perceived likelihood of contamination being present. In some cases, it may be well established that the land has had no previous use and is therefore expected to be clean and the scale of characterisation required may be relatively small. In other cases it may be readily apparent that contamination has arisen. Other areas may lie somewhere in between in that a degree of uncertainty exists over whether contamination is present or not.

This can best be evaluated from a knowledge of the previous use of the land, often based on a review of the historical records, drawings and maps etc to establish whether there are any recorded features which require investigation. Typical features might be pipes, bunkers, old foundations etc which are not readily accessible from the surface. If these are observed, then analytical effort can be targeted in these areas.

1.1.4. Additional pre-characterisation investigations

Experience shows that information on records and drawings is often only partially correct, or ambiguous, and unexpected features may also come to light during remediation. It is better that these are encountered prior to remediation, and further information may be gained from non intrusive techniques which do not break up the ground but allow features below the ground to be identified. Useful visual information is often obtained from aerial photographs of sites. Additional probing techniques are also available such as ground penetrating radar, electromagnetic imaging systems, ultrasonic techniques etc.

1.1.5. Knowledge of background radioactivity levels

Account must be taken of the levels of naturally occurring radioactivity in the soil, since this may form a significant contribution to the total activity present. This is especially true in the case of a clean site where, it is necessary to know the level of background in order to demonstrate that no enhanced activity is present. Knowledge of the background levels of radioactivity present is necessary since this defines the ultimate clean up target that can be achieved, and it may be that clean up targets are set at some level above background.

1.1.6. Targeting characterisation effort

The aim of the characterisation programme will be to define the area where contamination is present and to quantify its extent. It will also need to be able to meet the requirements of the clean up criteria which in some cases may be very stringent.

Sampling will be targeted to where the likelihood of contamination is greatest, and will also have to provide general coverage to provide confidence that no significant areas of contamination have been missed.

The programme devised will take account of all the factors mentioned under the previous headings to determine the limits of detection set for the analysis of samples in terms of concentrations of radioactivity in the soil, and for the measurements of doserates in situ. Three types of situation might be encountered.

1.1.6.1. Widespread contamination from a wind blown source

In this situation, contamination is expected to be fairly smoothly distributed and detectable at the surface. Assuming there are beta gamma emitting radionuclides present, a simple walk over survey to detect any enhanced gamma radiation should confirm the extent of contamination, and regular spaced core sampling should be sufficient to determine the concentrations of contamination across the whole field of survey. This should be sufficient to define the area for remediation and give a reasonable prediction of the quantities of waste that will be removed. In many situations this would require a degree of preparation of the site to clear vegetation away. Where the contamination is very extensive, then vehicle mounted or even airborne mounted detection systems may be used to carry out the radiation survey.

If a variety of radionuclides are present incorporating beta gamma emitters and alpha emitters, it will be necessary to try to establish fingerprints based on known or likely combinations of radionuclides to infer the presence of alpha and pure beta emitters present. If there are no grounds for establishing a fingerprint, then greater reliance will have to be placed on core sampling, being more extensive in coverage and intensive in analytical effort.. Establishing fingerprints may help to characterise the presence of alpha and pure beta emitters by reference to known ratios of gamma emitting radionuclides. If not, more extensive analytical effort will have to be targeted on measurement of individual alpha and beta emitters.

1.1.6.2. Isolated spills

These will most likely be present at the surface and concentrated as hot spots making them relatively simple to detect if gamma emitters are present. A preliminary walk over survey with a gamma radiation sensitive device is usually sufficient to identify any localised enhancements of activity that might require more intense core sampling to characterise the extent of contamination. If pure beta emitters are present, then detection will be harder, but a comprehensive walk over survey could be successful if sufficient contamination remains in the near surface and a suitable detector is used.

1.1.6.3. Contamination associated with discrete features

These may be present at surface or below ground. If gamma emitters are present, the walk over survey should again define areas for targeting core samples if the contamination is near to surface. If below ground (certainly greater than 1 m), then gamma emitters will not be detectable at surface. Targeting of effort relies on information gained from pre-characterisation investigations using non intrusive techniques, aerial photographs, maps of drain runs etc. Characterisation will rely heavily on the results of coring, or will have to be supplemented by further characterisation at the time of remediation. Measurements of groundwater may provide helpful information. Use of known fingerprints may again help to characterise the presence of non gamma emitting nuclides, but if no gamma emitting radionuclides are present, then surface monitoring is virtually useless and reliance must be placed on targeted coring.

1.1.7. Characterisation during restoration programme and post clearance

At this stage, further characterisation will also be required in order to confirm the extent of contamination as the restoration takes place, unless the full extent of contamination was established during the pre characterisation.

Assuming the full scale remediation is successfully completed, this still needs to be confirmed by a stage of post remediation characterisation. This characterisation needs to be thorough enough to demonstrate that the end points have been met and in particular that final concentrations are now consistent with background, or if not, within the cleanup targets that were set for the specified end point.

1.2. Experience to date with combined techniques

1.2.1. Chemical burial pits

An example of where some of these techniques have been used involves an area of land that has been used in the past for the disposal of chemically contaminated materials containing traces of radioactivity. A remediation programme is already in progress which involves the characterisation of wastes from 6 chemical disposal pits and five other pits containing beryllium contaminated wastes.

Characterisation work on the chemical and beryllium pits has used a combination of ground penetrating radar, various electromagnetic geophysical methods, surface metal detection and conventional core sampling. The use of ground penetrating radar has helped confirm the boundaries of the pits, and by mapping of the geophysical features within the pits, sample locations have been chosen which avoid hard objects which might damage the sampling equipment. Samples were then taken using conventional coring equipment.

During the period of the collaborative research programme, techniques have vastly improved for the spatial accuracy of measuring changes in doserates across wide areas. This has been achieved using the combination of sensitive sodium iodide detectors coupled to data logging equipment together with accurate position mapping provided by satellite global positioning systems (GPS). These have allowed rapid and very accurate identification of very localised contamination associated with near surface regions outside of the known burial pits which can also be remediated.

1.2.2. Catapult pit

Further investigations have also been carried out at Harwell on an old feature of the site called the catapult pit. The catapult pit was originally a second world war feature which was developed for the launching of aircraft, but was never used. The feature was just below ground level and was known to have been filled in during the early operational years of the UKAEA on the site. However, there was little knowledge of what if anything was disposed of in the pit. No radioactivity was detectable at surface, and previous excavations had also revealed no radioactivity.

The feature was investigated with a variety of non intrusive techniques including electro-magnetic imaging, ground penetrating radar and simple surface radiation measurements. These techniques successfully revealed the presence of materials below the ground surface and the shape of the feature was clearly identifiable. Electromagnetic imaging provided the clearest pictures.

However, when further intrusions using mechanical digging equipment were carried out, some radioactivity above background levels was readily detectable using simple hand held contamination and gamma radiation meters. The material encountered turned out to be natural uranium ore. This case highlights the problem of trying to determine levels of activity if buried below about 1m of ground since very comprehensive radiological surveys were also undertaken using accurate GPS systems at the ground surface, but these revealed no evidence of increased radioactivity above background level.

The site has also undergone radon gas sniffing and borehole monitoring to look for radioactivity in nearby groundwater. Radon gas sniffing has revealed enhancements in the region where the uranium ore was discovered, which was promising but not conclusive. It may mean that the radioactivity is confined to the region where the initial discovery was made, or it may mean that the gas has been more able to escape from the region where the ground has already been disturbed.

These observations taken together underline the need to use a variety of techniques in pre-characterisation before carrying out the restoration, and the need to be able to carry out on-line characterisation as the restoration is carried out. Most importantly, it demonstrates the need to develop characterisation techniques which are able to measure below ground radioactivity.

One possibility is the use of thermoluminscent dosimeters (TLDs) to measure accumulated dose if practicable ways could be found to position them below ground in suitable arrays. These devices are small and robust, can provide good statistical measurements but have to be left in situ and then removed for readout. There has been no development work of this type at Harwell to date.

1.3. Background soil activity

Work is currently being carried out to assess the background level for environmental radioactivity as a reference for comparison of clean soils with suspect soils from different areas across the site. The aim is to ensure that end point criteria can be demonstrated for the case of releasing parts of the site from regulatory control. This has involved taking of core samples from extensive areas across the site, particularly in regions where the land is regarded with most confidence to be clean.

One advantage of taking samples back to the laboratory for analysis is that detection limits can be lower than for field based measurements and more accurate data can generally be obtained. However, experience has shown that analytical methods have to be very carefully controlled. Even within the same analytical laboratory, spurious systematic differences between two sets of measurements have occurred which were initially attributed to possible differences in soil types. On further investigation these were found to be due to changes in methods applied by the analytical laboratory, where the calibrations applied to the later series of samples had been changed. Re-assessment using the same calibration standards enabled the differences between the data sets to be understood.

Top soil samples across the Harwell site have shown variations from 150 to 850 Bq/kg_{dry} alpha total activity (mean 490 Bq/kg), and beta between 120 and 760 Bq/kg (mean 420 Bq/kg). This variation is large compared with potential clean up targets for certain radionuclides and these measurements are therefore of limited value unless nuclide specific data for naturally occurring radionuclides are also obtained. It has therefore been concluded that reliance can not be placed on any method based on total alpha and beta measurements, and other techniques must be used in addition. Another problem with the gross alpha and beta method is the very small volume of material sampled (of the order of gram quantities).

Greater reliance is placed on gamma spectroscopy which can be carried out on more representative samples (1 kg rather than gram quantities). However, there may be instances where there are pure beta or alpha emitting radionuclides present which will not be detected by gamma ray methods. A combination of the two techniques is almost always required.

The aim is therefore to develop sampling and analysis protocols for soil assessment programmes. This will cover sample collection and preparation requirements. The preferred method is total digestion of the whole sample, not allowing sieving of small stones. This prevents bias of the results towards the finer, most likely contaminated fractions which could result in concentration measurements being too high. Also, preferred regimes for sample bulking are being considered. If the samples are properly prepared and controlled, then bulking of individual samples to make larger samples should give better results than a series of individual samples with less rigorous analysis.

2. DOUNREAY WORK PROGRAMME

The Dounreay site in Scotland was established for the development of the fast reactor programme in the UK. Site operations began with the 250 MW Dounreay Fast Reactor (DFR) with its own fuel cycle operations to separate out plutonium for return to the reactor. A prototype reactor (PFR) followed, and a Materials Testing Reactor (MTR), also operated on the Dounreay site for a number of years. These reactors have all now closed, and UKAEA activities on the Dounreay site are now concentrated on waste management, decommissioning and environmental restoration.

2.1. Management and control programme for contaminated ground

Ongoing characterisation of on- and off-site contamination is being carried out at Dounreay as part of the site's management and control programme for contaminated ground.

Although many investigation programmes were carried out in the past, the programme that has been developed over the last three years is a more co-ordinated approach. This programme was developed after an extensive surface survey of contamination on the site.

The approach involves:

- Characterisation of the contamination (radionuclides, location, area, depth, indication of potential sources, potential pathways/migration);
- Sentencing and assessment;
- Actions and prioritisation of actions – which include:
  - Further characterisation which can involve invasive core sampling and characterisation of the contamination with depth;
  - Monitoring (or additional monitoring);
  - Containment measures;
  - Restoration.

In prioritising the actions required to deal with the contamination a number of factors are considered. These include:

- health risk to workers and to the public;
- potential to migrate (particularly off-site);
- radioactive decay over the period of institutional control;
- regulatory position;
- site development needs (e.g. particularly where contaminated ground has to be excavated as part of a site construction project).

2.2. Characterisation and remediation

2.2.1. Surface contamination

Surface contamination is ascertained by "traditional" monitoring techniques using beta-gamma handheld probes, and as at Harwell, systems using GPS linked to detector systems.

Much of the surface contamination on the site consists of small spots of elevated levels of contamination – much of which is from diffuse or dispersed sources. When found, the first measure taken with these areas is to screen them off using physical barriers. As most of the contamination in these areas is gamma emitting, it can be readily located during subsequent investigation and is then removed. Once removed, the barriered area is monitored over a period of 6–12 months to ensure no subsequent recontamination. If after this period there is no evidence of re-contamination, the barrier is removed. In those cases where recontamination occurs, the barrier remains in place and the area is further investigated to reassess potential sources and any need for containment measures.

Initially 61 areas of contaminated ground were barriered off which totalled an area of 20,400°m^2. After a year of phased remediation works 42 areas (13,978 m^2) have been surface remediated, monitored and have had the barriers removed. The majority of the remaining barriered areas have contamination which extends to depth and a programme of further investigation and assessment is in place to deal with it.

2.2.2. Contamination at depth

Not surprisingly, most of the contamination found at depth is associated with old drainage systems. In many cases this contamination is found to be localised to the drainage system with very little lateral migration. This is mainly along the path of disturbed ground which surrounds the drain.

2.2.3. Characterisation of off-site radioactivity by aerial survey

To obtain a more comprehensive understanding of the radiological distribution and to augment the conventional sampling programme, an aerial radiological survey was undertaken in September 1998. The survey covered an area of approximately 130 km^2 of ground around, but not over-flying the Dounreay site.

The survey employed a gamma spectrometer utilising an array of sodium iodide detectors mounted within a Squirrel helicopter. Flying height was 90 metres along flight lines spaced at 50 metres within 2 km of Dounreay and 100 metres 2 km beyond — amounting to a total of 1682 line km flown. The aircraft was positioned using a Global Positioning System (GPS) with in-flight differential correction which provided aircraft position accuracy to ±5 m. The purpose of the survey was to establish the distribution of ^{137}Cs, ^{241}Am and ^{60}Co and in addition the distribution of natural nuclides of potassium (K), uranium (U) and thorium (Th).

The instrumentation was calibrated by hovering at different altitudes over a large flat field approximately 2 km east of Dounreay. Soil samples were taken from the field and analysed, as found, for concentrations of K, U, Th and Cs which were then used to calibrate the spectrometer. The activity levels are derived from data received by the detectors, averaged over the resolution area and based on the wet weight samples taken from the top 20 cm of soil in the calibration field.

Distribution maps of the concentrations of the radionuclides were produced. (In the cases of uranium and thorium which do not emit gamma radiation, concentrations were obtained by monitoring for daughter products, ^{214}Bi and ^{208}Tl respectively).

The aerial survey showed no measurable concentrations of ^{241}Am or ^{60}Co. The concentrations of K, U and Th were found to be at natural levels expected from the rock and soil types covered by the survey. Elevated levels of ^{137}Cs above the general background of 30–50 Bq/kg were found in a few areas, notably in a valley 6–8 km Southeast of the site where a value of 397 Bq/kg was recorded. The pattern of the distribution indicates that this was almost certainly an accumulation of activity arising from nuclear weapons testing and predominantly the Chernobyl incident.

Along the coastline (maximum recorded level 130 Bq/kg), elevated activity levels are probably due to site discharges via a sea to land transfer mechanism. In addition, elevated levels to the south and east of the site (maximum recorded level 125 Bq/kg) also probably arise from site discharges to the air.

It was concluded from the aerial survey that the activity levels found around Dounreay were consistent with known levels previously established through the environmental sampling programme and are well below levels which represent significant risks to people or to the environment.

2.3. Dounreay particles

A particular problem which exists on the Dounreay site and in the marine environment adjacent to the site is the presence of small particles (~1 mm in dimensions) of irradiated Materials Testing Reactor fuel.

Regular (fortnightly) surveys of the foreshore using conventional hand held gamma radiation survey instruments have been carried out every two weeks since 1984 following the discovery of a radioactive particle on the Dounreay foreshore in 1983. Since then, particles have been recovered from the Dounreay foreshore at an average rate of around one per month. Since 1984, three such particles have also been found on the nearby public beach.

In terms of the total volumes of sand either at Dounreay or any of the public beaches, the number of particles (which are typically the size of a grain of sand) is extremely small with the estimated probability of any member of the public coming into contact being correspondingly low. Nevertheless, the problem is taken very seriously both by UKAEA and by the Regulators.

A programme of work is thus in place to:

- assess and characterise the particles in terms of their potential and behaviour in the environment;
- model and predict their movement and erosion with time;
- assess the associated risks particularly with time;
- frequently monitor the foreshore and beaches.

In recent times, the monitoring programme has been intensified with the introduction of vehicle mounted gamma radiation detection instruments using GPS tracking.

As part of the characterisation programme, detailed offshore investigations have been undertaken. As well as obtaining more detailed information on seabed topography, and current and wave movements, seabed radiological surveys have been undertaken. This has involved using specially adapted gamma detectors – one instrument tracking the seabed whilst being towed from behind a boat, and another being operated on the seabed with divers. The former instrument enabled large areas of the seabed to be surveyed so that anomalies could be followed up by the divers. 35 particles were recovered in 1997 and a further 89 particles in 1998. This intensified surveying programme has demonstrated the presence of a significant population of particles in an area spreading to the north east from the location of the low level liquid effluent outfall. Better estimates of the number in this area await further detailed studies.

In addition, preliminary results using the towed gamma detector in deeper water indicate that particle populations in those areas are very much lower than in the shallow waters.

Background anomalies, in the form of high counts, identified by the towed system have also been investigated by divers and demonstrated to arise from naturally occurring radioactivity.

Completion of statistical analysis of the data from the towed detector will give more detailed estimates of the numbers of particles in the offshore sediments.

3. SOIL TREATMENT TECHNIQUES

3.1. Soil washing tests

As part of its supporting R and D activities, UKAEA is undertaking a number studies to ascertian the potential of particular remediation technologies to the treatment of contaminated ground on its sites.

One particular technique being studied is volume reduction by soil washing. This programme of work is being conducted at laboratory scale and also at pilot scale using the UKAEA Soil Characterisation Pilot Plant facility based at Harwell.

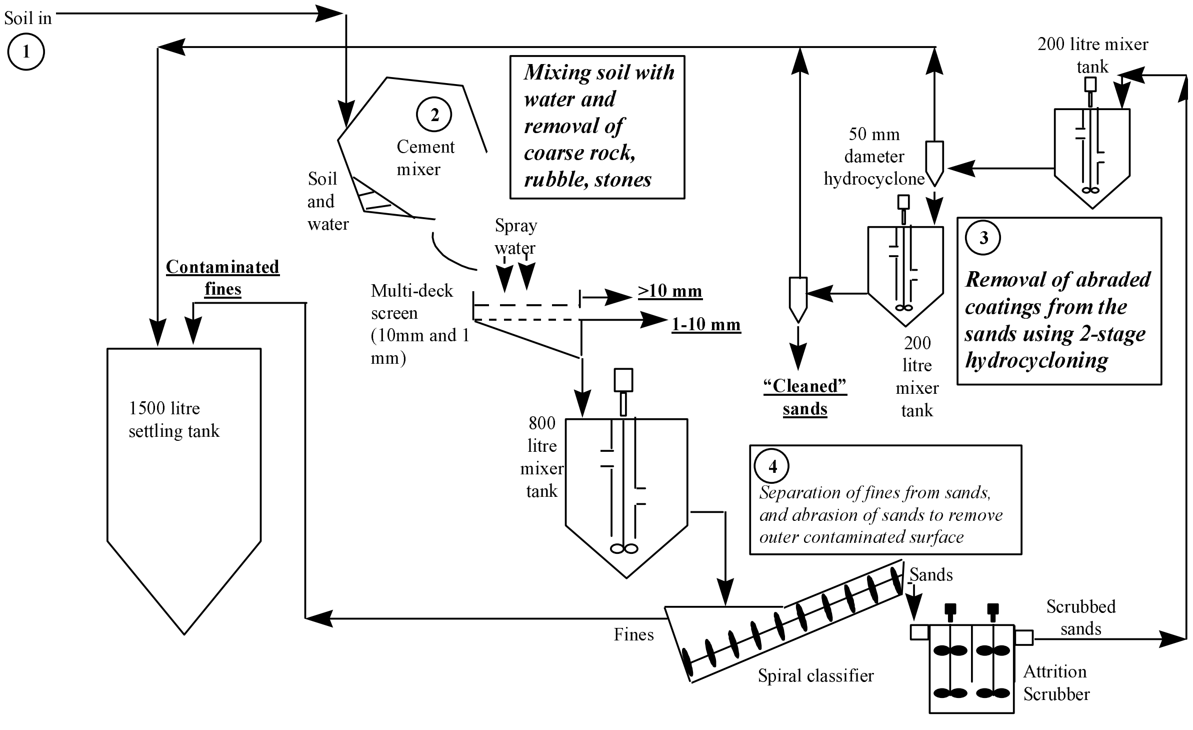

FIG. 1. Pilot scale soil washing circuit.

The type of soil washing being considered is one based on particle separation using equipment from the mineral processing industry. The principle of the processes used is to remove those particles which contain elevated levels of contamination from those that are contaminant depleted using differences in grain size, settling velocity, abrasion and surface chemical properties. Low Level Waste disposal is very expensive. Soil washing means that only those fractions which have elevated levels of contamination require LLW disposal, instead of disposing of all the soil. The contaminant-depleted fractions can then either be released for more economic disposal, or if contamination levels are low enough, reused. Not all soils behave in the same manner, therefore the tests have been carried out to determine applicability.

Although called “soil washing” and being a “wet” process, the type of soil washing used transfers very little of the contamination into the water (confirmed during both laboratory and pilot scale tests). Initially laboratory tests were conducted on nine soils from three UKAEA sites. Following these tests, an 800 kg sample of soil from one of the sites was treated at pilot scale using the flow diagram illustrated in Figure 1.

3.2. Results from laboratory tests

Results from the tests showed that in nearly every soil examined, there is a marked elevation in the contamination in the finest fractions compared to the coarser fractions. There was one exception to this. In that particular soil, the distribution of the radionuclides with size was erratic and this was suspected to be related to the morphology and mineralogy of the soil – it being weathered chalk.

To assess the degree of contaminant-depletion in the coarser fractions, a comparison was made of the ratio of the contamination in the original material to that in the coarse fractions. This ratio is termed the “Decontamination Factor”. No one “cut-off” size was used for the coarse fractions, rather a criteria based on the coarse fractions being 70% of the mass of the total soil and thus the fines being 30% of the mass of the total soil. Results are as follows:

TABLE I. DECONTAMINATION FACTORS FOR LABORATORY TEST SAMPLES

Nuclide	DF	No. of soils analysed
^{137}Cs	1 – 7	9
^{90}Sr	1– 3	6
^{238}Pu	1 – 2	3
^{239}Pu	1 – 5	5
^{241}Pu	1 – 5	6
^{241}Am	1 – 5	3
^{60}Co	4	1

Other than the poor preferential distribution of contaminants to the chalk-rich soil, there is no other obvious trend which accounts for the variability of the DF with soil type, site type or level of contamination in the original material.

One of the soils examined had a very high proportion of material less than 0.038 mm (61% by mass). Particles >2.0 mm in this soil showed a very marked depletion in ^{137}Cs contamination in particular. However the proportion of material in this fraction is only 4% by weight making the volume reduction potential of this material poor.

Two soil samples were attrition scrubbed. From the limited attrition scrubbing tests carried out, it was apparent that this additional process was effective in removing further contamination from the sand and silt fractions. Thus, for the two soils that were attrition scrubbed, there was a further 50% reduction in the levels of ^{137}Cs, ^{90}Sr, ^{241}Pu and ^{60}Co in the sand and silt fractions.

3.3. Pilot scale tests

Pilot scale testwork was undertaken on 800 kg of soil from one of the sites. The circuit used for the tests involved:

- mixing the soil with water to make a slurry using a cement mixer;
- sizing the particles in the slurry at 1 mm and 10 mm using a vibratory screen;
- removing the fines from the <1 mm material using a spiral classifier;
- attrition scrubbing the spiral classifier coarse product to abrade contaminants from the outer surfaces;
- classifying the resulting scrubber product by hydrocycloning to separate the scrubbed coarse product from the removed coatings. (The removed coatings then being fine particles).

The material used was from a remedial action on the site where the restoration target was 4°Bq/g ^{137}Cs. Tests were carried out on material >4 Bq/g ^{137}Cs. Because the activity in the test material received was lower than originally envisaged, it was relatively easy to reach the target just by removal of the coarse material greater than 1 mm. This was 73% of the mass relative to the original feed material (based on dry matter) and had a ^{137}Cs content of 3.04 Bq/g. With spiral classification, attrition scrubbing and hydrocycloning, 86% of the mass (based on dry matter) was recovered as a clean product with a ^{137}Cs content of 3.09 Bq/g.

By recalculating the activity balance around the circuit relative to the fractions <10 mm, it is possible to assess the effects of processing a finer soil. Thus from a feed material (<10 mm) of 6.50 Bq/g ^{137}Cs, processing by screening, classifying, scrubbing and hydrocycloning would produce a clean fraction which is 70% of the mass (based on dry matter) which has a ^{137}Cs content of 2.89 Bq/g, and would therefore meet the 4 Bq/g target criterion.

Further recalculation of the activity balance shows that for an even finer soil, say equivalent to the <1 mm fractions of the soil, then from a feed material of 9.38 Bq/g ^{137}Cs, a contaminant depleted product could be produced which is 47% of the mass (based on dry matter) and with a ^{137}Cs content of 3.35 Bq/g.

On work carried out so far, the economic viability of soil washing in support of site restoration depends significantly on the volume of soil involved, and may beneficial if volumes of 2000 – 5000 m^3 are involved, provided the soil characteristics are favourable. One factor that could therefore become important as part of the characterisation process is the physical and chemical nature of the soil, since remediation options could be more favourable for certain types of soil.

One additional observation from the pilot scale operations was that the concentration of the contamination measured in the field analysis was three times greater than that calculated for the feed material derived by analysing the products and carrying out an activity balance. The discrepancy between the field based measurement and that carried out on a large mass of soil in the pilot plant has again highlighted the problems in assessing activity concentrations in large volumes based on measurements made from small samples taken in the field.

Further studies are continuing on bulk quantities of soil which will involve the pilot equipment for establishing a more reliable soil assay, particularly with respect to the correlation of field assay methods to laboratory analysis. Further studies are also being carried out for treatment on soils >40 Bq/g.

4. CONCLUSIONS

Work has been carried out within the Harwell and Dounreay sites of UKAEA towards the restoration of contaminated land and for release of areas currently under regulatory control. Work continues in developing a multi disciplinary approach to characterisation. The main area where advances are most urgently required is in the development of techniques which allow detection of below ground radiation.

Progress has been made in determining the presence of natural radioactivity for the purpose of identifying background levels. Tight controls need to be placed on the analytical methods used in characterising the contamination against natural background levels, particularly where release from regulatory control is the final endpoint.

The development of soil treatment techniques may assist in the eventual clean up options for some types of contaminants, and is also assisting in the understanding of how some characterisation methods may overestimate contamination concentrations.

The advances made in the use of accurate radiation mapping using satellite global positioning systems has been the major development during the period of the CRP. The refined use of these techniques has enabled accurate pinpointing of contamination, providing greater confidence in site restoration, and savings in terms of minimising the volumes of wastes segregated for disposal. The development of these techniques has been the most important area of collaboration from the CRP.

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IDENTIFICATION AND CHARACTERIZATION OF RADIOACTIVELY CONTAMINATED SITES IN UKRAINE AND PLANNING FOR ENVIRONMENTAL RESTORATION ACTIVITIES

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ABSTRACT

In the Pridniprovsk-Krivoy Rog region uranium, titanium, iron and manganese ores were mined and milled beginning in the 1950s. These activities have caused radioactive contamination of the environment at some sites. In recent times intensive works concerning the surveying of contaminated areas and substantiating the need for remediation have been initiated. The research methodologies applied and the results from radiation surveys are presented for the site of the first uranium mine in the Ukraine, for tailings originating from the Pridniprovsk Chemical Plant (PChP), for the recultivated dump-site of the former "O"-mine, as well as for the wastes, raw materials and production of the Nicopol Ferro-Alloy Plant. The planning procedure for the remediation activities at the town of Zhovty Vody is described.

1. INTRODUCTION

The regions where mining and milling enterprises are located are often considered as locations with technogene impacts on the environment. They are regions of potential pollution by natural radionuclides (NRN) due to the accumulation as industrial wastes on the ground surface, their involvement in the milling processes, and their utilization in some comercial applications. The relevant radionuclides such as uranium-238, thorium-230, radium-226, polonium-210, lead-210, protactinium-231 are classified as the most hazardous pollutants.

From this point of view, the Pridneprovsk-Krivoy Rog region is the most affected in Ukraine. There is a number of large mining and milling enterprises located in the region (Fig. 1):

- VostGOK Mining and Milling Enterprise;
- Industrial Corporation“Alexandria Coal”;
- Verchnedniprovsky Mining and Metallurgical Enterprise;
- Industrial Corporation “Krivbussruda” —;
- Industrial Corporation “Pridniprovsky Chemical Plant”;
- Large Mining & Milling Enterprises (InGOK, SevGOK, CGOK, NKGOK, UGOK).

The VostGOK M&M Enterprise has been active in uranium ore mining and processing for more than 40 years. As a result of its activity, large territories (about 14 km^2) were affected and have been contaminated. The dust from dumps and dry beaches of tailings ponds creates an additional pollution of atmosphere with radon and long lived uranium- and thorium-series radionuclides.

At present, mining for uranium ores is going on in the Kirovograd region of Ukraine at the Smolino and Ingoolsky Mines respectively.

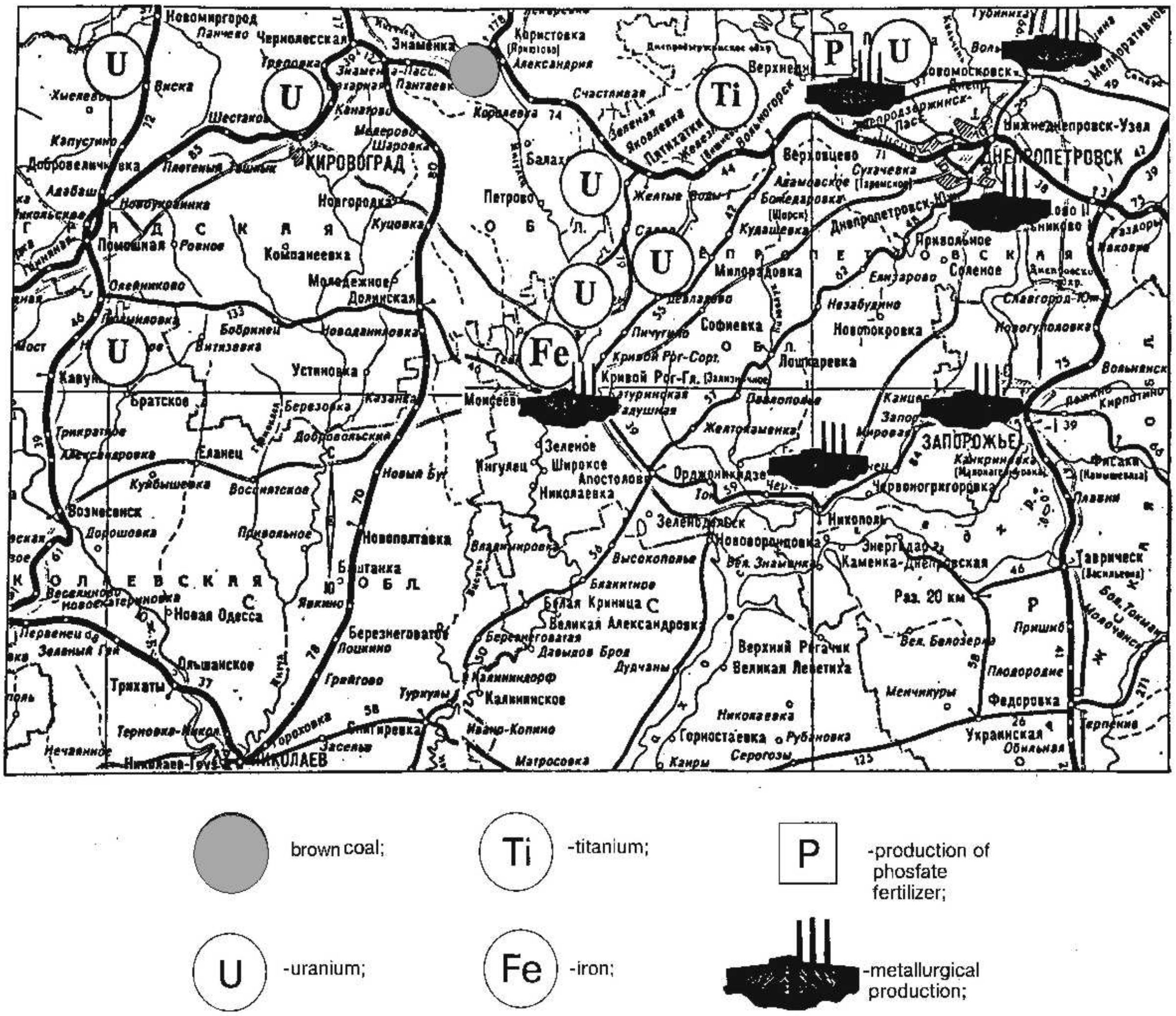

FIG. 1. Mining and ore processing in the region.

In the town of Zhovty Vody the uranium mining in the Novaya Mine has ceased, and the mine is used for the extraction of iron ores only. Uranium ores from Smolino and Ingoolsky Mines are still being processed in a hydrometallurgical plant in Zhovty Vody.

In 1950s and 1960s the mining of uranium ores took place in the Pervomayskaya Mine in the city of Krivoy Rog. After the uranium deposit were exhausted, the activities in the Mine switched to the development of iron ore body. At present, this Mine is also going to be closed.

Besides the traditional mining method, in situ leaching of uranium was employed in Ukraine at the Devladovo and Bratsky sites. They have been closed down and the site have been recultivated.

Uranium ores coming from both Ukraine itself and from abroad were processed between 1947 and 1991 at Pridniprovsky Chemical Plant (PChP) in the city of Dniprodzierzhinsk. When the PChP ceased to operate, many spots with radioactive contamination and the tailings storage facility remained at its former site. At present the plant has been switched to new products (zirconium and gold). The tailings storage facility is being investigated in detail.

2. REGULATORY BASIS

One of the basic documents regulating the work and the activities concerning the decontamination of radioactively contaminated sites is the “Sanitary Regulation for the Liquidation, Conservation and Conversion of Radioactive Ores Mining and Milling Enterprises” [1], established in 1991. Over the last few years new Ukrainian laws have been passed, regulating radiation safety at plants, in urban areas, as well as in the environment in general. These include:

- “On Nuclear Energy Utilization and Radiation Safety” [2];
- “On Human Protection against Ionizing Radiation Impact [3];
- “On Radioactive Waste Management” [4];
- “On Uranium Ore Mining and Milling” [5].

According to [3], an individual dose limit for the population in general must not exceed 1°mSv of effective exposure dose per year. Therefore, the average effective doses for humans belonging to the critical group must not exceed the established dose limits independent of the conditions and the way of the dose received.

In 1998, the new "Radiation Safety Standard of Ukraine" (NRBU-97) [6] has been put into force. It was drawn up on the basis of recommendations of both, the International Commission for Radiation Protection (ICRP) [7] [8] and International Safety Standards [9]. In this connection a transitional period is in force now in the Ukraine, during which relevant implementation regulations are updated and new one are developed.

3. METHODS OF CHARACTERIZATION AND INSTRUMENTATION

To characterize contaminated territories and radioactive wastes, a combination of field and laboratory methods was used.

The field method includes the following:

- dosimetric surveys;
- determination of α- and β-radioactive contamination on surfaces;
- emanation surveys;
- radon emission survey (measuring radon flux density from surface);
- measuring radon and its equivalent equilibrium volume activity, both in the atmosphere and inside dwellings.

For the gamma-radiation dose rate measurement occupational dosimeters and field radiometers were used.

Geiger-Müller counters were used as detectors in occupational dosimeters, and scintillation NaI (Tl) crystals were used as detectors in radiometers. The energy dependence of the dosimeters did not exceed ±2.5% within an energy range between 0.05 and 3 MeV. For the field radiometer this value was up to 60%. For this reason the radiometer was used only for detecting zones with abnormal radiation, and was not used for dose estimation. Both devices were calibrated at a special gauging organization in Zhovty Vody.

Surface contamination was determined with the aid of occupational dosimeters containing alpha and beta detecting removable units.

The measuring range for alpha-radiation was from 0.04 to 400 $Bq\cdot cm^{-2}$, and for beta-radiation from 0.04 to 4000 $Bq\cdot cm^{-2}$.

Emanation surveys were carried out with alpha-active gas field radiometers. Soil air sampling was carried out by drawing the air into a chamber of 50 cm^3 capacity from a depth of 0.6 m. Analyses took place 3 hours after sampling.

The density of ^{222}Rn flux from the ground surface was measured by gas sorption onto activated charcoal [10]. Special cartridges with activated charcoal discs were placed on the surface to be studied and exposed to it. After exposure, the charcoal was analyzed by gamma-spectrometry. The minimum measurable value of ^{222}Rn flux was 2 $mBq\cdot m^{-2}\cdot s^{-1}$.

For measuring the volumetric activity of ^{222}Rn in the air of houses a classic scintillation method (with a Lucas cell) was used [11]. The device consists of set of scintillation chambers, each with volume of 500 cm^3. The air was sampled by pumping it through a series of inter connected aerosol filters, a dryer and the chamber. Aerosol sampling was performed through the filter. Aerosol filters were made of ultra-thin perchlorovinyl fibres. The decay coefficient of these filters is 95% for Rn progeny. Markov's method was implemented using a low level alpha-counter and a portable air pump with a throughput of 20 l/min [12]. The alpha-counter detector includes ZnS plates and a photo-multiplier. The efficiency of the total alpha-irradiation registration by the detectors was equal to 43%:

$$\text{Efficiency of alpha irradiation registration} = \frac{\text{Count rate of standard source } [s^{-1}]}{\text{Activity of standard source [Bq]}}$$

The background reading of the counter did not exceed eight counts per hour. The detection limit for ^{222}Rn was 18.0 $Bq\cdot m^{-3}$ with a relative error of ±30%. In some cases, measurements were carried out with an "Alpha GUARD" (Germany) radon-monitor. Calibration of the devices for ^{222}Rn and its measurement of its progeny was performed with a standard Radon-chamber of 18 m^3 volume, which was located a special gauging organization in the town of Zhovty Vody.

Methods using field gamma-spectrometry with HPGe-detectors are still not widely used in Ukraine because of lack of domestically produced instruments, and of uncertainties connected with interpretation of results obtained by this method.

The following laboratory methods were used to determine the natural radionuclides (NRN) content in radioactive wastes, contaminated soils, raw materials and industrial wastes:

— radiometric measurements of total alpha-activity in samples;
— radiochemical analysis of NRN content;
— low-background gamma-spectrometric analysis of NRN content.

Radiometric measurement of total alpha-activity in environment samples were used for fast assessment of a site for alpha-emitting nuclides contamination. Two modifications of this method were available:

— measurement of sample alpha-activity in "thick" layer;
— measurement in scintillator layer.

Measurements of ^{238}U concentrations in soil and wastes were carried out in the laboratory using radiochemical methods with photometric detection.

The contents of ^{226}Ra, ^{232}Th, ^{40}K were determined by a gamma-spectrometric method with low-background gamma-spectrometer. The content of ^{210}Po was determined by a radiochemical method with radiometric detection using a low-background alpha-counter. Concentration of ^{210}Pb were determined by both methods: a radiochemical one with radiometric detection using a low-background beta-counter, and a spectrometric one with low-background gamma-spectrometer using a high-purity germanium detector. The relative errors of measurements (at the 95% confidence interval) were ± 20% for the radiochemical methods, and did not exceed ± 25% for the gamma-spectrometric and radiometric methods.

4. RADIATION SURVEY EXPERIENCE

4.1. Radiation survey of the "Pervomayskaya" Mine area

The first uranium ore mining enterprises in Ukraine was the "Pervomayskaya" Mine. Uranium ore mining began in the late 1940s at one of its sites ("2/6"). The mining activities were extended by opening the mines "Obyedinennaya" and "Severnaya-Ventilation" of the same enterprise. In the late 1960s, uranium ore mining ceased, but at "Obyedinennaya" mining for iron ore continued. In early 1998 the enterprise also ceased to mine the iron ore and transferred into liquidation.

Over the past 30 years all surface buildings of the mine "2/6" have become unused and derelict, as is the case for almost all main and auxiliary buildings and installations of the "Severnaya-Ventilation" mine (Fig. 2). The total area of the mining enterprise territory to be rehabilitated is 68.5 ha. The enterprise activities have resulted in contamination of environment, particularly from the following: scattered ore; natural radionuclides leaching from the ores; uncontrolled utilization of wastes for other purposes; dispersion of dusts etc. In the course of time, the ores and soil have been intermingled thus increasing the concentration of natural radionuclides in soils.

FIG. 2. Derelict ore processing building at the "Severnaya-Ventilation" Mine.

In order to assess the sites' contamination, a comprehensive range of characterization methods have been applied. Initially, dosimetric gamma-surveys were carried out, which helped to reveal and to contour in general occurrences of contamination. Then followed a radiometric determination of surface alpha- and beta-contamination. The same method was used to distinguish between contaminated and non-contaminated mine equipment, as well as construction materials. Radon emanation and radon emission surveys have allowed to detect contamination sources located at shallow depths (0.4 ÷ 0.5 m), which had not been identified with gamma-surveying.

Laboratory methods have shown the total alpha-activity of the mixture ore and soil ranges from 813 Bq/kg to 4.06×10^5 Bq/kg, radium-226 ranges from 110 to 5.35×10^4 Bq/kg (see Table I). At soil spots with radioactive contamination one can notice a considerable amount of radon exhalation to atmosphere, as well as increased radon levels in the soil air (Table II). Exhalations exceeded background values by 1.5–140 times over contaminated spots, while the radon concentrations in the soil air at abnormal spots are 40–815 times higher than background values.

Applying both field and laboratory methods for the characterization of affected areas has allowed to determine the contamination parameters and to develop respective recommendation for the remediation of the land.

TABLE I. NATURAL RADIONUCLIDE CONTENTS IN SAMPLES OF ORE AND SOIL DUST, SILT, AND DETRITAL SOIL MIXTURES TAKEN AT THE "OBYEDINENNAYA", "2/6", AND "SEVERNAYA-VENTILATION" MINE SITES

Sample number	**Sampling place and sample type**	**Natural radionuclide contents [Bq/kg]**			**Total specific alpha-activity [Bq/kg]**
		^{232}Th	^{226}Ra	^{40}K	
	"2/6" Mine				
1	Ore loading site, ore and soil mixture	10	239	244	6340±610
2	Ore loading site, ore sample	–	53500	–	$(4.06\pm0.05)\cdot10^5$
4	Ore loading site, ore and soil mixture	14	705	307	7190±690
	"Obyedinennaya" Mine				
5	Abnormal spot 1/1, ore and soil mixture	18	753	240	6750±610
	"Severnaya-Ventilation" Mine				
17	Left sump. Depth 0.5 m. Silt	28	110	494	813±190
22	Dust from gallery #7	25	2412	1441	$(2,47\pm0.08)\cdot10^4$
26	Abnormal spot 3. Depth 0.1÷0.3 m. Detrital soil	25	601	542	6811±850
39	Sample of background soil	15	20	169	588±176

TABLE II. EMANATION AND EXHALATION MEASUREMENTS ON THE TERRITORIES OF "2/6" AND "NORTHERN VENTILATION" MINES

Sampling point	**Sampling location**	**Radon exhalation [mBq·m^2·s^{-1}]**	**Radon concentration at a depth of 0.5 m [kBq·m^{-3}]**
"2/6" Mine			
12	Abnormal spot 1	355	50.5
13	Abnormal spot 1	459	48,4
14	Abnormal spot 2	1850	459
15	Boundary of abnormal spot 1	93	–
16	Abnormal spot 1	801	–
17	Abnormal spot 2	4050	–
18	Abnormal spot 3	614	–
"Northern Ventilation" Mine			
5	Temporary storage of uranium ore mining wastes	860	960
8	Temporary storage of uranium ore mining wastes	169	–
21	Left sump of mine cleaning water system	43	0.18
19	Background value	28	1,18

4.2. Wastes, raw materials and production of the Nicopol Ferro-Alloy Plant

Steels are alloys of iron with manganese, chromium, silicon etc. One possible raw material for the ferroalloys production are ores which already are naturally enriched with the oxides of the alloying constituents and processed ore concentrates. In some cases these may also contain increased concentrations of natural occurring radioactive materials, for the genesis of many ore deposits resulted in radioactive elements being present as accessories to the main elements being mined.

The Nikopol Ferroalloy Plant (NFP) utilizes raw materials delivered from a range of deposits in the Ukraine, Russia and other countries. In terms of total amount, the production share of ferroalloys with manganese is around 50%, and the share of those with silicon is around 30°%. Table III shows laboratory analysis results for samples of both raw materials and products of the NFP. The data presented give evidence of high contents of natural radionuclides in rutiles, zircons and baddeleyites. In the remainder of the raw materials the natural radionuclides content is at background values levels. Concentrations of all radionuclides are almost equal to background values in products, except for flux AN-65-U. The increased ^{226}Ra concentration in the flux AN-65-U is explained by utilizing rutile and zircon for its preparation.

Solid production wastes (casting slag, granulated slag) containing ^{226}Ra, ^{232}Th, and ^{40}K can be used with restrictions for the production of constructing materials. Slimes, i.e. wastes from de-dusting and gas scrubbing systems, are stored in a slime sump.

TABLE III. NATURAL RADIONUCLIDES CONTENT [Bq/Kg] IN RAW MATERIALS, PRODUCTS AND WASTES OF THE NIKOPOL FERROALLOY PLANT.

Type of sample	**^{238}U**	**^{226}Ra**	**^{232}Th**	**^{40}K**	**^{210}Pb**	**^{210}Po**
Raw materials						
Rutile (Ukraine)	169±62	266±32	27±6	70±9	620±100	450±50
Zircon (Australia)	590±120	694±190	1150±420	600±80	4580±310	8240±400
Zirkon (New Zealand)	3110±800	9990±200	1100±400	500±30	6800±800	8941±400
Baddeleyite concentrate (Russia)	2870±370	117-±130	307±130	-	1080±140	1150±60
Manganese ore (Ukraine)	87±30	78±28	8±3	179±90	-	188±50
Boron slag	2,5±2	12±6	7±2	180±82	1074±300	73±30
Products						
Flux AN-65-U	-	374±70	80±30	20±02	-	61±25
Flux AN-60	-	51±18	33±12	149±46	-	39±13
Flux AN-348-V	-	48±10	16±5	159±46	-	51±20
Flux AN-47	-	224±10	42±6	111±30	210±60	189±82
Flux AN-67-A	22±8	48±8	15±4	61±15	450±92	20±4
Wastes						
Casting Slag (silicomanganese)	-	194±28	35±12	607±128	-	-
Granulated slag (silicomanganese)	-	135±42	25±10	477±122	-	-
Granulated slag (ferromanganese silicon)	-	161±30	30±10	617±130	-	-
Slime (slime storage 1)	24±8	65±40	24±6	1206±300	6011±200	3200±1300
Slime (slime storage 2)	-	66±32	7±2	1250±300	2100±650	1420±400
Slag of sintering shop 1	205±68	154±42	26±8	513±160	-	191±32

The investigation of the slime storage gamma signals have shown no considerable increases over background values. To the contrary, laboratory analyses have identified high total alpha-activities of the slime samples. More detailed laboratory investigations, using radiochemistry techniques and low-background spectrometry with a HPGe-detector have shown that the slime is rich in both ^{210}Pb and ^{210}Po (Table III). A considerable shift of equilibrium between uranium decay-chain radionuclides in the slime bears witness to the enrichment of trans-radon chain elements in the wastes due to the technical process (involving high temperature treatment) compared to their background concentrations in raw materials. Thus, the method of choice for detecting and characterizing radioactive contamination at such facilities is to measure total alpha-activity in the samples, rather than gamma-dosimetric surveying.

According to the standards under development [13], slime are considered as slightly radioactive wastes. In this connection it will be clamed special requirement during recultivation activities.

4.3. Preliminary results of radiation surveys on the capped tailings storage of Pridniprovsk Chemical Plant.

There are capped tailings on the site of the PChP with an surface area of about two hectares. They have been created during the 1950s and do not conform to the present standards and regulations for radioactive wastes management. The tailings resulting from uranium ores processing have a thickness of 12.5 to 15.5 meters and are covered with loess loam layer. About 770,000 tones of tailings with a total activity exceeding 0.2 PBq are stored here. The tailings surface is covered with turf, but the slopes might still be subject to erosion.

Gamma-radiation dose rates on the site of the tailings impoundment fluctuate between 0.14 to 7.29 μSv/hour. Along the perimeter of the tailings impoundment contaminated spots have been found, where gamma-radiation dose rates reach a value of 15.76 μSv/hour. The ^{226}Ra content in the wastes varies from 789 to 1.72×10^6 Bq/kg (Table IV). The concentrations of ^{226}Ra in surface water samples taken from locations in the vicinities of tailings impoundment site range from 24 to 99 mBq·dm^{-3}, while in the Dnjepr-river and in groundwater they range from 8.5 to 18.5 mBq·dm^{-3}. Values of ^{222}Rn flux from the surface of tailings impoundment, where covered by a protective layer, is around 0.32 to 1.5 Bq·m^{-2}·s^{-1}, while radon exhalation background levels are only around 9 to 18 mBq·m^{-2}·s^{-1}.

On the basis of the results from the completed research, recommendations have been developed concerning the improvement of the tailings capping.

TABLE IV. CONTENT OF NATURAL RADIONUCLIDES IN WASTES OF PChP

Sample number	**Concentration [Bq/kg]**	
	^{226}Ra	**^{232}Th**
342	39330	-
358	27230	5169
324	3888	-
84	2738	54
38	789	25
189	1.72×10^6	-
53	69970	-
330	1.32×10^5	-
157	92080	-

4.4. "Olkhovskaya" mine recultivated territory radiation survey

At the end of the 1990s one of the first projects for restoration of contaminated land was realized at the former uranium ore mine "Olkhovskaya" site [14]. The work included the sorting of the waste rock on the site and the sanitation of contaminated land. From the waste rocks sorted, such parts containing rich ores were processed at the hydrometallurgical plant, the remainder was removed to "Novaya" mine. After rehabilitation post-remediation monitoring was performed, including:

- dosimetric surveys;
- emanation surveys;
- radon emission surveys;
- soil sampling and analysis for natural radionuclide contents.

In general, measured values of gamma-radiation equivalent doses rate on the remediated land is within 0.1 to 0.22 µSv/hour, which is in the range of natural background radiation of the region.

Emanation surveys were conducted for determining and contouring residual contamination occurring at depth and not detected by gamma-surveys. Minimal values of soil radon content were observed in the middle of the site. This can be explained by the blanketing of the soil under "Olkhovskaya" mine waste rock. On the major part of the site, the average radon concentration in the soil was within 10-20×10^3 Bq m^{-3}, which conforms to the background values of the regions. Maximum values of soil radon content were observed at southern part of the site, where the concentrations exceeded 25–50 times background values. Abnormalities determined, in general, are caused by the deposition and accumulation of radionuclides in topographical depressions (ditches, hollows etc.).

At the most conspicuous points of the site under survey, the radon exhalation from soil was measured. Five measurements at three points gave a background value of 15–25 $mBq{\cdot}m^{-2}{\cdot}s^{-1}$. In the centre of abnormal point 1, determined during the emanation survey, the radon exhalation reaches 553 $mBq{\cdot}m^{-2}{\cdot}s^{-1}$. The same situation is observed at abnormal point 2.

To specify contamination parameters at the abnormal points, boreholes have been drilled and soil samples have been taken. Results of radiation surveys at abnormal Points 1 and 2 are shown in Table V. At abnormal Point 1 soil contamination extends down to a depth of four meters, and is caused by leachates penetrating into the ground.

TABLE V. RESULTS OF RADIATION SURVEYS

No. of abnormal spot	Radon exhalation [$mBq{\cdot}m^{-2}{\cdot}s^{-1}$]	Rn conc. in soil [$kBq{\cdot}m^{-3}$]	Sampling depth [m]	Soil description	Concentration in soil [$Bq{\cdot}kg^{-1}$]		
					^{238}U	^{226}Ra	^{232}Th
Abnormal 1	553	579	0.0 - 0.25	Grey poured soil	37	34	38
			0.25 - 0.5	Grey poured soil	40	37	45
			0.5 - 0.75	Grey poured soil	45	56	57
			0.75 - 1.0	Grey poured soil	45	61	46
			1.0 - 1.5	Black loam	626	568	57
			1.5 - 2.0	Light-brown loam	248	146	35
			2.5 - 2.8	Light-brown loam	422	208	40
			3.5 - 4.0	Pale-yellow loam	434	42	33
			6.5 - 6.8	Brown loam	26	37	30
Abnormal 2	313	136	0.0 - 0.2	Poured loam	103	79	71
			0.2 - 1.4	Rocks soil	868	735	58
			1.4 - 1.6	Buried soil	75	35	32
			2.0 - 2.5	Light-brown loam	47	38	37
			3.5 - 4.0	Brown loam	33	34	33
			5.3 - 6.0	Brown loam	40	33	30

At abnormal Point 2 no significant contamination of the soil by natural radionuclides was noticed. The elevated values of radon content in the soil were caused by an one-and-half meter layer of waste rock. Following the survey results, some additional remediation measures at the contaminated spots were implemented.

4.5. Radioactive situation in the town Zhovty Vody

In the town of Zhovty Vody (Fig. 3), as in many of similar such settlements (for instance as in Grand Junction, USA), in the first studies of industrial zones, the harmful influences of low active wastes have being underestimated [15]. It has led to uncontrolled use of the dumped rocks for commercial activities of the population. This in turn led to serious contamination of some parts of the town. In the 1980s decontamination was organized for some parts of the town, but it appears not to have been sufficient; at present the average gamma- background on the area of the town is:

- for 88% of the area between 0.16 and 0.4 µSv/hour;
- for 8% of the area between 0.4 and 1.5 µSv/hour;
- for 3% of the area between 1.5 and 2.5 µSv/hour;
- for 1% of the area above 2.5 µSv/hour.

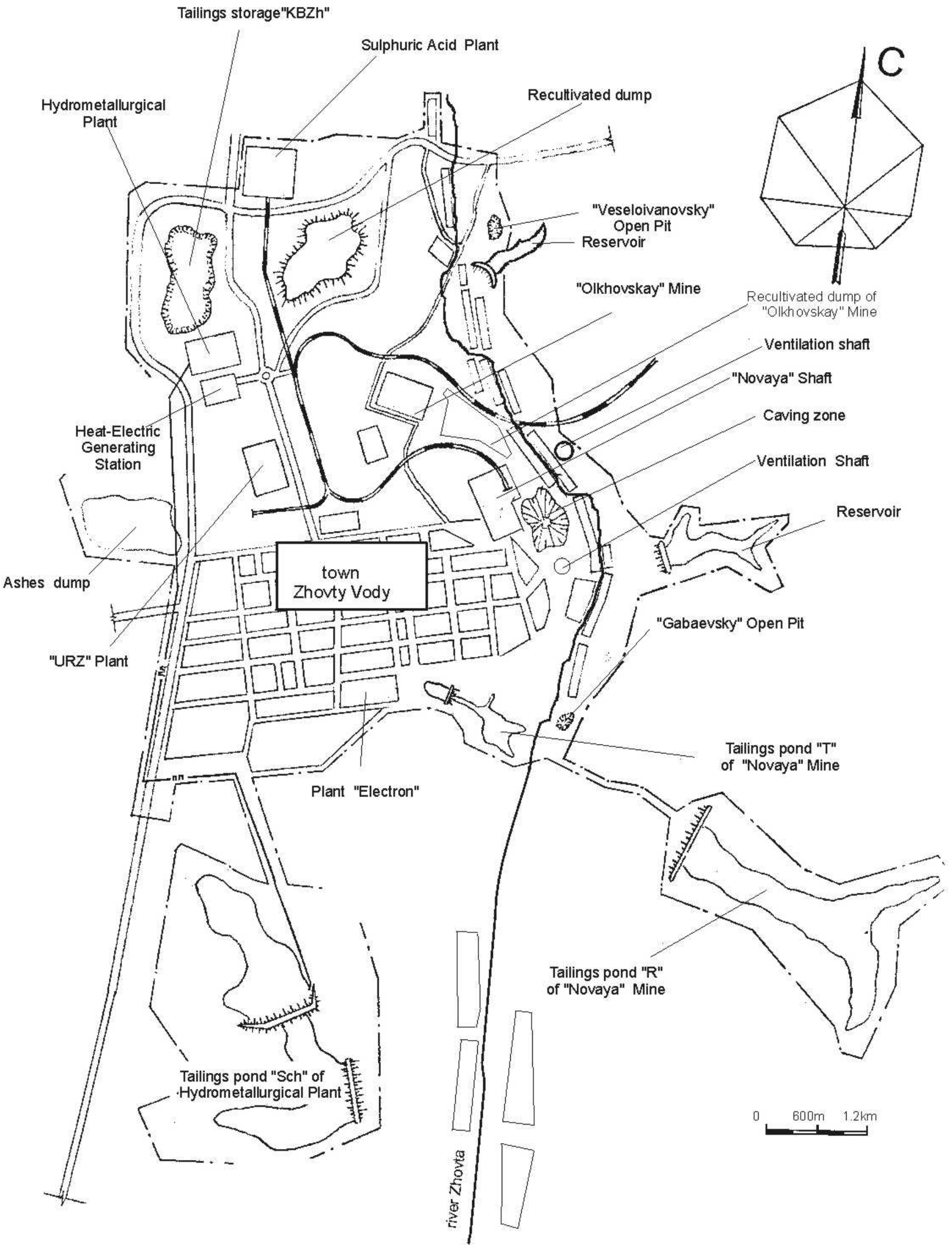

FIG. 3. Location of objects of potential ecological hazard in Zhovty Vody.

In radiation surveys more than 5,400 abnormal locations and places, where the gamma-radiation exceeds 1.2 µSv/hour, have being detected. At 460 locations the gamma dose rate exceeds 10 µSv/hour, and at another 57 locations it is higher than 30 µSv/hour. All these irregularities are related to rocks which contain elevated levels of natural radionuclides.

The abnormalities according to their type are distributed as follows:

- in house foundations — 1334 (= 25% of the total number of observed) abnormalities;
- roads and squares surface coverings — 2440 (= 44%) abnormalities;
- isolated spots — 1253 (= 23%) abnormalities
- sites with undefined localization — 430 (= 8%) abnormalities.

In 1994, two thousand measurements of the radon-gas equivalent equilibrium concentrations (EEC) in the living spaces of the town were performed. For 14% of all houses the EEC of radon was 100 to 200 Bq/m^3, for another 19% it was between 200 and 1000 Bq/m^3, and for 3.5% in excess of 1000 Bq/m^3.

5. THE PLANNING OF REMEDIATION WORK (USING THE EXAMPLE OF ZHOVTY VODY TOWN)

5.1. Choice of method

An important element of the planning for remediation work at a contaminated site is the choice of the optimum strategy of the implementation of the work. According to standards in force at present [6], both radiation safety and protection against occupational exposure are built on following principles:

- any practice involving the exposure of people must not take place if its benefit for the people being exposed and for society as a whole is less than harm (principle of justification);
- exposure dose levels from all types of practice must not exceed established dose limits (principle of non-exceedance);
- individual dose levels or the number of individuals being exposed, regardless of the source of irradiation, must be as low as can reasonably be achieved, taking into account economical and social factors (principle of optimization).

The above principles were used when planning the cleanup measures for radioactive contamination on the territory of Zhovty Vody town.

5.2. Determination of norms for urban residual contamination.

Norms for permissible urban residual contamination are determined by starting from the exposure of critical population groups according to following general steps:

- external exposure;
- exposure from Rn and it daughters by inhalation;
- exposure from long lived natural radionuclides in dust taken up by organisms.

No indigestion path for long lived natural radionuclides was considered because of its insignificance under the conditions at Zhovty Vody. Critical groups include the following three categories of population living on land with radioactive contamination:

- infants;
- children under 17 years of age;
- adults.

A background value of 20 µR/hour is accepted as gamma dose rate on the town's territory. An average value of 60 µR/hour is accepted as gamma dose rate on contaminated sites.

For the purpose of assessing the impact of doses on the general population caused by radioactive site contamination three values of gamma dose rates (30, 40, 60 µR/hour) are defined according to international standards [9] [16]. The results of the estimations are shown in Table VI. Taking a 30 µR/hour residual gamma dose rate from a contaminated site, the dose limit for member of the public will not be exceeded when a recommended dose margin coefficient of 2 is used [13]. By this, a certain dose safety margin for the impact from the contamination on the town area (Hydrometallurgical Plant, "Novaya" Mine, "Sch"-tailings storage) is provided.

The above accepted criteria (≤30 µR/hour) are in accordance with existing national and international ones:

- the ^{226}Ra concentration in soil shall not exceed 150 Bq/kg, which conforms to the criterion of a specific alpha-activity in soil equal 1200 Bq/kg, which is accepted for remediation by way of afforestation;
- the average Rn volume concentration over a given site shall not exceed 18.5 Bq/m^3.

TABLE VI. ANNUAL EFFECTIVE DOSES FOR CRITICAL GROUPS OF THE POPULATION AS CAUSED BY RADIOACTIVE SITE CONTAMINATION AND BASED ON THREE DIFFERENT GAMMA DOSE RATES.

Population category	**Exposure paths**	**Annual effective doses [mSv/year] for gamma dose rates [µR/hour] of**		
		30	**40**	**60**
Infant	External ionizing irradiation	0.142	0.284	0.568
	Radon and its decay products	0.107	0.232	0.357
	Re-suspended dusts	$1.7\cdot10^{-5}$	$2.1\cdot10^{-5}$	$2.9\cdot10^{-5}$
	Total	0.249	0.516	0.925
Children less then 17 years old	External ionizing irradiation	0.122	0.244	0.488
	Radon and its decay products	0.107	0.232	0.357
	Re-suspended dusts	$4.2\cdot10^{-5}$	$5.2\cdot10^{-5}$	$7.2\cdot10^{-5}$
	Total	0.229	0.476	0.845
Adults	External ionizing irradiation	0.113	0.226	0.452
	Radon and its decay products	0.107	0.232	0.357
	Re-suspended dusts	$3.9\cdot10^{-5}$	$4.7\cdot10^{-5}$	$6.7\cdot10^{-5}$
	Total	0.23	0.478	0.809

5.3. Estimation of the efficacy of remediation measures

The exposure doses to the population from radioactive site contaminations are estimated assuming the following conditions [16]:

- a dose coefficient, which is the ratio of effective dose rate and absorbed dose rate in air, of 0.74 for adults, 0.8 for children, and of 0.93 for infants;
- staying outdoors for 20% of the total time.

In Table VII the results of estimations for the effective dose from the soil on the town population are given.

TABLE VII. EFFECTIVE DOSE ON POPULATION (μSv/year) FROM DIFFERENT LEVELS OF EXTERNAL GAMMA DOSE RATE AT THE AREA.

Category of population	**Exposure dose rate [μR/hour]**				
	20	**30**	**40**	**50**	**60**
Infants	284	426	568	710	852
Children less than 17 years old	244	366	488	610	732
Adults	226	339	452	565	678

The average weighted population distribution in the town is as follows:

— infants	— 570 individuals;
— children less then 17 years old	— 11200 individuals;
— adults	— 48230 individuals;
— total	— 60000 individuals.

The weighted average dose E_a^n on every category of population is calculated according to Equation 1:

$$E_a^n = \frac{E_b^n \times (S_b - S_r) + E_r^n \times S_r}{S_b} \qquad (1)$$

where

- — E_b^n is the effective dose for category n of population residing on town land with a background value of gamma dose rate of 20 μR/hour;
- — E_r^n is the effective dose for category n of population residing on contaminated land with an average gamma dose rate as 60 μR/hour;
- — S_b is the total area of non builtup urban are in ha, i.e. $S_{b.}$ = 643 ha;
- — S_r is total area of radioact ive contaminated sites in ha, i.e. S_r = 51,3 ha;

From the above calculations it has been estimated, that the weighted average effective doses are:

— for infants	— 330 μSv/year;
— for children before 17 years old	— 282 μ Sv/year;
— for adults	— 262 μSv/year.

The results of estimating the collective dose for the town's population under different remediation dose rate targets are shown in Table VIII.

As the calculations show, in case of remediation down to the background values of gamma-radiation exposure dose rates, a decrease (from 15.982 man Sv) of the collective effective dose of 2.188 man Sv will be achieved.

In case of partial decontamination to 30 μSv/year of an average gamma-radiation exposure dose rate, the decrease will be 1.631 man Sv, and accordingly for 40 μSv/year it will be 1.079 man Sv.

TABLE VIII. COLLECTIVE EFFECTIVE DOSE FOR THE POPULATION FROM EXTERNAL GAMMA-IRRADIATION [man Sv]

Population category	**Annual dose [mSv/year] for average of gamma dose rates [μR/hour] of**			
	Background	**30**	**40**	**60**
Infant	0.162	0.175	0.183	0.188
Children less than 17 years old	2.732	2.842	2.950	3.158
Adults	10.9	11.334	11.770	12.636
Total	**13.794**	**14.351**	**14.903**	**15.982**

In addition to the exposure from gamma-radiation sources, a contribution to the total dose to the population is received from Rn and its daughters, as well as by re-suspended dusts. Table IX gives values for collective effective doses assuming background values for gamma dose rate and residual value of gamma dose rate from contaminated land of 30, 40 and 60 μR/hour respectively.

TABLE IX. COLLECTIVE EFFECTIVE DOSE TO TOWN POPULATION FROM ALL SOURCES ON LANDS WITH RADIOACTIVE CONTAMINATION [man Sv].

Sources of impact to population	**At background γ-dose rate**	**At average residual value of γ-dose rate [μR/hour]**		
		30	**40**	**60**
External exposure	13.794	14.351	14.903	15.982
Rn and its daughters	4.255	6.134	7.112	8.718
Re-suspended dust	-	0.002	0.0037	0.004
Total	**18.049**	**20.487**	**22.0187**	**24.704**

Remediation measures should result in a reduction of the dose for a long period of time. Radiation detriment V_c can be expressed in terms of cost [17] according to:

$$V_C = \alpha \cdot S_E, \tag{2}$$

where α is the cost of exposure (cost of collective dose unit), a used in radiation protection; in the present case α =10,200 USD/ man Sv $^($the exchange rate for Ukrainian Hrivna to US dollar was at the time of this work 1 USD= 2 Hrivna); S_E is prevented collective effective dose in man Sv.

The cost of implementing the remediation measures will be less than the cost incurred from the radiation damage. As a result, the expenditure on the remediation measures is expected to give a net “profit” for a long time period. The ICRP recommends [17] time periods of 70 years for children and 50 years for adults to use in assessments. Table X gives calculated values for radiation detriments, prevented as a result of implementing remediation measures, for different scenarios.

The calculations show that conducting remediation measures prevents radiation detriments for one year from 27394 to 67861 USD/year, depending on the assumed/targeted residual gamma-dose rate.

TABLE X. MONETARIAN VALUE [USD] OF PREVENTED RADIATION DETRIMENTS FOR DIFFERENT RESIDUAL GAMMA DOSE RATES AND DIFFERENT PERIODS OF AMORTISATION.

Critical Group	**Reduction of γ-dose rate to background**		**Reduction of γ-dose rate to 30 μR/hour**		**Reduction of γ-dose rate to 40 μR/hour**	
	for 1 year	**for whole period of optimization**	**for 1 year**	**for whole period of optimization**	**for 1 year**	**for whole period of optimization**
Infants	697	48790	383	26775	206	14455
Children under 17 years of age	12857	900025	8151	570605	5185	362915
Adults	54307	2715350	34471	1723550	22003	1100150
Total	**67861**	**3664165**	**43005**	**2320930**	**27394**	**1477520**

5.4. Estimation of optimum dose reduction to population

Optimization of radiation protection is performed in the cases, when individual doses are less than the dose limits for members of the public (1 mSv/year). The calculations for selecting a strategy for optimizing the implementation are based on [6]:

$$R < \frac{V_c - X}{\alpha}, \qquad (3)$$

where X is the expenditures [in USD] for carrying out the remediation measures, and R is the collective risk of stochastic effects arising from the exposure:

$$R = r_E \cdot S_E, \qquad (4)$$

where r_E is the risk coefficient for a cancerous decease arising with mortal as well as not mortal outcome and for serious hereditary effects arising. For a population r_E is 7.3×10^{-2} Sv^{-1}.

Calculated values for the optimization of relevant remediation measures are presented in Table XI.

The optimization calculations performed show that the maximum return of investments should be achieved when carrying out the remediation measures until obtaining a residual gamma-irradiation rate of 30 μR/hour. Remediation of the land down to background values is not efficient.

6. CONCLUSIONS

When investigating contaminated land, it is important to select an optimum range of methods allowing a detailed characterization of the site.

The main methods for characterizing contaminated sites are:

- dosimetric surveys;
- radiometric fast measurements of total alpha-activity in sample;

- emanation surveys;
- radon emission survey (measuring the density of radon flux from surface);
- measuring radon and its equivalent equilibrium volume concentration in houses and in the ambient atmosphere;
- soil sampling and analysis for concentrations of natural radionuclides.

TABLE XI. CALCULATION RESULTS CONCERNING OPTIMIZATION OF REHABILITATION MEASURES CONDUCTING UNDER DIFFERENT SCENARIOS

	γ-dose rate reduced to background	**γ-dose rate reduced to 30 μR/hour**	**γ-dose rate reduced to 40 μR/hour**
Prevented detriment V_c, [USD]	3664165	2320930	1477520
Calculated value of expenditure for rehabilitation measures X [USD]	3950000	1100200	569720
Calculated index $(V_c - X)/\alpha$	-28	120	89
Collective risk R	26.23	16.61	10.58

As the experience from applying these methods shows, no single one can be used separately for characterizing the contaminated land. In any given case it is necessary to apply a certain set of methods, depending on the extend of expected contamination and radionuclides involved.

According to results from this work it is expedient to perform efficacy analyses before and during implementation of remediation measures.

The analyses were performed in the context of planning for remedial actions on the contaminated areas in the town of Zhovty Vody. From these it was concluded that it would be efficient to clean the land down to a residual gamma dose rate of 30 μR/hour. Under those conditions the maximum effect will be obtained from the remediation activities without exceeding dose limits for the population.

Participation in the CRP made it possible to learn from international approaches to the identification and characterization of radioactively contaminated sites. The experience gained will be used both for the design of a new regulatory basis for the remediation of the contaminated sites and for carrying out remediation measures themselves.

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SITE CHARACTERIZATION TECHNIQUES USED IN ENVIRONMENTAL REMEDIATION ACTIVITIES

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ABSTRACT

As a result of decades of nuclear energy research, weapons production, as well as ongoing operations, a significant amount of radioactive contamination has occurred throughout the United States Department of Energy (DOE) complex. DOE facility are in the process of assessing and potentially remediating various sites according to the regulations imposed by a Federal Facility Agreement and Consent order (FFA/CO) between DOE, the state in which the facility is located, and the U.S. Environmental Protection Agency (EPA). In support of these active site remediation efforts, the DOE has devoted considerable resources towards the development of innovative site characterization techniques that support environmental restoration activities. These resources and efforts have focused on various aspects of this complex problem. Research and technology development conducted at the Idaho National Engineering and Environmental Laboratory (INEEL) has resulted in the ability and state-of-the-art equipment required to obtain real-time, densely spaced, *in situ* characterization data (i.e. detection, speciation, and location) of various radionuclides and contaminants. The Remedial Action Monitoring System (RAMS), developed by the INEEL, consists of enhanced sensor technology, measurement modeling and interpretation techniques, and a suite of deployment platforms which can be interchanged to directly support remedial cleanup and site verification operations. *In situ* characterization techniques have advanced to the point where they are being actively deployed in support of remedial operations. The INEEL has deployed its system at various DOE and international sites. The deployment of *in situ* characterization systems during environmental restoration operations has shown that this approach results in several significant benefits versus conventional sampling techniques. A flexible characterization system permits rapid modification to satisfy physical site conditions, available site resources, and cleanup requirements. The rapid and precise collection of *in situ* measurements can reduce operational costs by reducing the volume of material requiring remediation. This approach can also reduce uncertainty by obtaining a “total-area field screen” versus a representative subset of hand-collected lab-analyzed samples. It can lower on-site health risks by reducing human exposure to contamination through reduction of hand samples during the field sampling process. And, *in situ measurements* can improve management decisions by providing real-time, densely spaced, repeatable, characterization data.

1. INTRODUCTION

As a result of decades of nuclear energy research, weapons production, as well as ongoing operations, a significant amount of radioactive contamination has occurred throughout the United States Department of Energy (DOE) complex. Recent estimates project the DOE environmental problems to involve more than 5700 separate groundwater plumes, more than 600 billion gallons of contaminated groundwater, over 200 million cubic meters of contaminated soils, and numerous landfills containing more than 3 million cubic meters of buried waste.

This DOE radioactive waste and associated contamination represents unique remediation challenges. Because approximately half of all DOE waste was disposed of before 1970, many of the landfills are commingled with various types of waste. As a result of these past disposal practices, much of the waste throughout the DOE complex is contaminated with both hazardous and radioactive materials. As such, this waste continues to pose a threat to surface and groundwater. The DOE sites where this waste is predominantly located are the Hanford Reservation, the Savannah River Site, the Idaho National Engineering and Environmental Laboratory (INEEL), the Los Alamos National Laboratory (LANL), the Oak Ridge National Laboratory (ORNL), the Nevada Test Site (NTS) and the Rocky Flats Plant (RFP). Figures 1 and 2 show early disposal practices at two DOE facilities, the Oak Ridge National Laboratory and the Idaho National Engineering and Environmental Laboratory, respectively.

FIG. 1. Early waste disposal at the Oak Ridge National Laboratory.

FIG. 2. Early waste disposal at the Idaho National Engineering and Environmental Laboratory.

1.1. Environmental restoration strategy

With the passage of new environmental laws and regulations, DOE waste disposal sites are now being evaluated and if necessary remediated. Each DOE facility must be remediated according to the regulations imposed by a Federal Facility Agreement and Consent order (FFA/CO) between DOE, the state in which the facility is located, and the U.S. Environmental Protection Agency (EPA).

Certain sites fall under the regulations of the Comprehensive Environmental Response, Compensation, and Liability Act (CERCLA). Under this act, DOE conducts remedial investigations and feasibility studies. These remedial investigations characterize conditions at the site to identify the sources and extent of contamination. The feasibility study evaluates specific alternatives for cleaning up the site. Alternative remedial solutions are evaluated using specific CERCLA evaluation criteria. Finally, the selected remedial action is then documented in a Record of Decision which is subsequently implemented to achieve site restoration. Figure 3 illustrates the process to be followed to select an appropriate remediation solution for CERCLA sites.

1.2. Innovative site characterization techniques

In support of these active site remediation efforts, the DOE has devoted considerable resources on the development of innovative site characterization techniques that support environmental restoration activities. These resources and efforts have focused on various aspects of this complex problem.

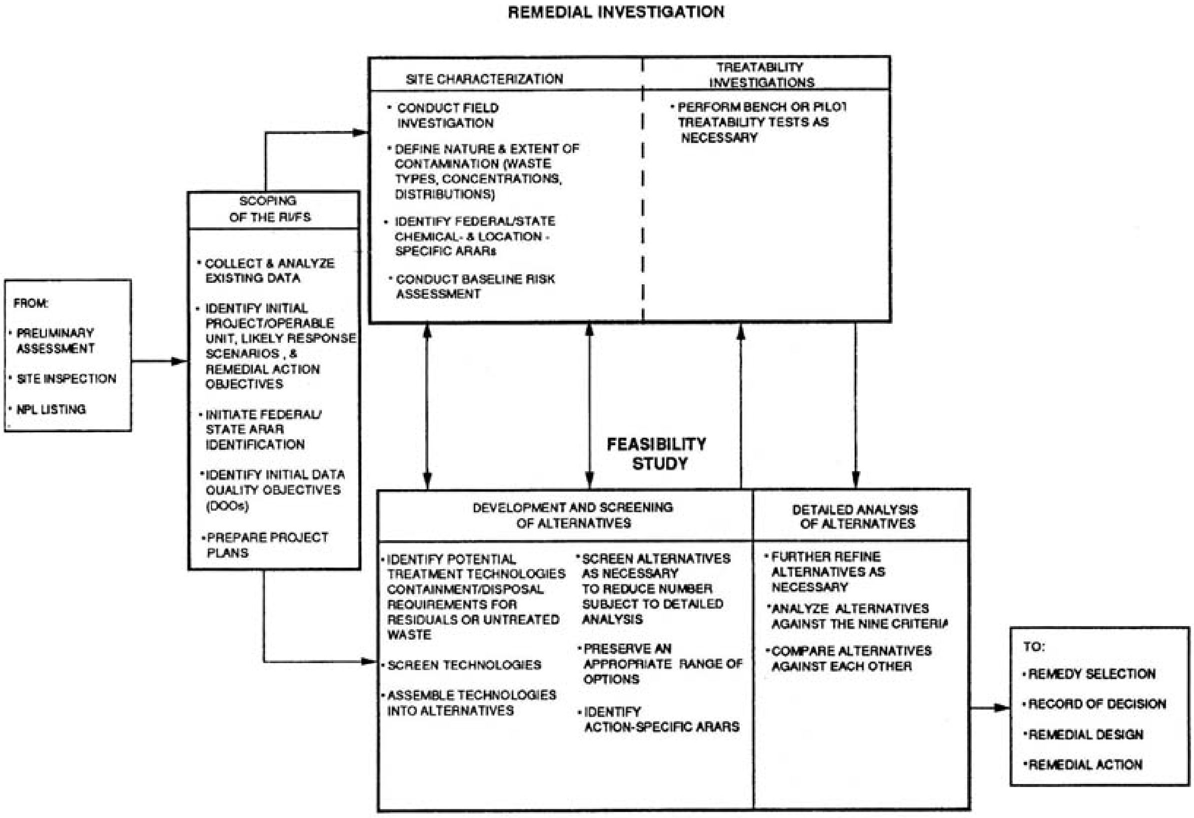

FIG. 3. USEPA CERCLA Remedial Investigation/Feasibility Study process [1].

INEEL research and associated technology development has resulted in the ability and state of the art equipment required to obtain real time, densely spaced, *in situ* characterization data (i.e. detection, speciation, and location) of various radionuclides and contaminants. The Remedial Action Monitoring System (RAMS) [2], developed by the INEEL, consists of enhanced sensor technology, measurement modeling and interpretation techniques, and a suite of deployment platforms which can be interchanged to directly support remedial cleanup and site verification operations. The RAMS is an *in situ* radioactive and hazardous waste monitoring and data management system that is applied during radioactive and hazardous waste cleanup operations. The RAMS consists of three major components:

— deployment platforms,
— characterization sensors, and
— the data management system.

The integration of these subsystems is flexible to produce an optimal *in situ* measurement and data management system to satisfy site specific applications where immediate, high quality field characterization information is required.

1.3. INEEL deployment platforms

Several deployment platforms have been developed and used by the INEEL to support the collection of *in situ* radiological measurements. The applicability of these platforms is dependent on site specific conditions.

1.3.1. Excavator mounted system

The Excavator Mounted System (EMS) is a specialized platform to which a variety of radiation sensors can be attached and used to investigate areas of contamination by methodically scanning the surface during data collection (see Figure 4). The system is used primarily in uneven terrain, for discreet delineation between clean and contaminated soil, mapping the lateral extent of contamination, and where layers of soil are excavated in controlled “lifts”. The platform possesses automated functions that enhance data quality by maintaining constant vertical platform position during scanning operation. This is accomplished by using mechanical dampeners on each of the platform’s axis of travel. A terrain following system maintains a constant, vertical sensor offset from the surface under investigation. The EMS is normally deployed from the arm of most commercially available tracked excavators.

FIG. 4. INEEL excavator mounted system (EMS) in situ characterization at DOE Mound.

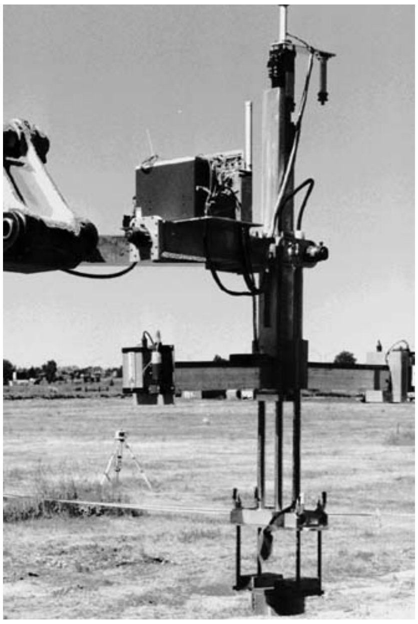

FIG. 5. INEEL excavator mounted system (EMS).

With a characterization sensor attached to the base of the sensor mount plate, the platform scans a surface, acquiring data and logging the exact position of each data point. This sequence can be repeated for successive vertical excavations. Information collected is converted real time into contamination maps. Site managers use these maps when making critical decisions regarding the next step in the remediation process.

This system has been implemented by Lockheed Martin Idaho Technologies Co. (LMITCO) at DOE sites in the States of Idaho, Ohio, and South Carolina. At the INEEL, RAMS has been used to map caesium-137 contaminated soils. In Ohio, RAMS was used on two separate remediations. First, an abandoned buried tank was retrieved as well as the surrounding thorium-232 and actinium-227 contaminated soil [3]. Second, RAMS was used to map plutonium-238 hotspots in a canal bed (see Figure 5). Finally, at the DOE Savannah River site, RAMS was used to characterize a caesium-137 soil site during its remediation [4]. Most recently the EMS was deployed at various sites within the United Kingdom to map potentially contaminated soils.

1.3.2. Surface area mapper (SAM)

INEEL's Surface Area Mapper (SAM) is a wheeled sensor platform that is easily deployed for broad area, surface, radiation characterization (see Figure 6). The SAM platform is designed such that radiation sensors are positioned above an open "window" at the base of the platform allowing high-energy particles to pass unimpeded into the open face of the sensor.

FIG. 6. INEEL's Surface Area Mapper (SAM).

1.4. Characterization sensors [5]

SAM is integrated with an automatic global positioning system (GPS), enabling the precise location and instant display of *in situ* data at a remote workstation. SAM workstation monitors the ongoing data collection process, displaying processed information on a monitor for easy viewing by decision makers. SAM can be used to collect precise characterization data even in more difficult terrain. Its configuration allows access to moderate variations in ground surface topography. And since it is fitted with a wide push bar, two people can easily maneuver it.

INEEL's RAMS makes use of both "enhanced" commercially available characterization sensors, as well as "INEEL-developed" characterization sensors. Table I lists potentially applicable sensors versus contaminants of interest. Of these, the INEEL has extensively modeled three sensors, useful for the detection of low level gamma and alpha radiation[5]. These sensors are a Plastic Scintillator for detecting moderate to strong gamma emitting radioactive contaminants, a Germanium-Spectrometer for speciation of radioactive contaminants, and a six-crystal array Calcium Fluoride (CaF_2) detector for detection of the L X ray component of primarily alpha emitting radionuclides such as ^{238}Pu.

1.4.1. Plastic scintillator detector (PSD)

The RAMS gross count detector consists of a 30 ×30 × 3.81 cm (12" × 12" × 1.5") plastic scintillator with an adjustable discriminator with the pulses above a discriminator level stored in a scaler. Gross counts are scaled and transmitted at 1second intervals. The detector and pulse processing circuitry are mounted as an integral assembly in an enclosed aluminum case. Shielding of 5-cm (2") thick by 10-cm (4") high lead bricks surround the plastic scintillator along its four sides. The top of the detector is unshielded. The sensitive front face of the scintillator is about 4.4 cm. (1.7") above the bottom of the lead brick shield.

This arrangement moderately restricts the viewing angle of the detector and provides some protection from background gamma rays. This entire assembly weighs about 105 kg (230lbs.).

TABLE I. POTENTIALLY APPLICABLE SENSORS/DETECTORS FOR VARIOUS CONTAMINANTS OF CONCERN.

Radionuclide	**Potential Sensor/Detector**
^{137}Cs, ^{108m}Ag, ^{232}Th, ^{60}Co	Plastic Scintillator, Ge semiconductor, NaI Scintillator
^{90}Sr, ^{90}Y	Xenon proportional counter, Plastic Scintillator
^{14}C, ^{99}Tc	No in situ techniques
^{241}Am	Plastic Scintillator, Ge semiconductor, NaI Scintillator
^{238}Pu, ^{239}Pu, ^{244}Cm, ^{233}U	Calcium fluoride detectors
^{238}U	Xenon proportional counter, Ge semiconductor
^{235}U	Plastic Scintillator, Ge semiconductor, NaI Scintillator

The field of view is approximately 120 cm × 120 cm (4 ft. × 4 ft) at the normal 15-cm (6") height above the soil surface. Any pulse above the lower level discriminator registers a count, therefore, the plastic scintillator detector's large size allows it to gather data quickly as it scans the ground. However, because of its poor energy resolution, this detector cannot differentiate among radionuclides.

1.4.2. Gamma ray spectrometer

In situ gamma ray spectrometer measurements or grab samples are routinely used to determine radionuclide compositions. The RAMS gamma ray spectrometer assembly incorporates a High Purity Germanium (HPGe) semiconductor detector in an all-attitude dewar. The HPGe detector is an n-type Ge detector 5.0 cm diameter and 2.0 cm thick. The detector is shielded by a bismuth shield/collimator, and is recessed 7.9 cm from the front of the collimator. The shield/collimator wall thickness is 4.44 cm (1.75") and attenuates 662 keV gamma radiation by a factor of 20. This configuration restricts the viewing angle of the detector to a defined solid angle provides some protection from background gamma rays. When shielded the detector has a field of view of about 60 cm diameter with the bottom of the collimator 15 cm (6") above the ground. The complete detector assembly with detector/ electronics, shield/collimator and support frame weighs approximately 50 kg (110 lbs). The spectrometer electronics consist of a Canberra Industries portable multi-channel analyzer (MCA). The MCA is equipped with the INEEL-designed dual-energy, low power pulser and processing electronics. Acquisition control, data transmission and storage uses software written by the INEEL.

1.4.3. Calcium fluoride detector

The RAMS Calcium Fluoride detector consists of a six-detector rectangular 2x3 array. The detector thickness is 0.152 cm. Which supports good counting efficiency for the Pu L X rays and at the same time, minimizes the efficiency for the higher energy 60-keV x rays and γ rays. The detector array is used in the scanning mode with a scanning rate of less than 15 cm/s for *in situ* measurements. The energies of the pulses from the L X rays and 60-keV γ rays emitted from a ^{241}Am source from each detector are aligned by adjusting the high voltage on each photomultiplier tube so that the gains are matched. This combined signal is fed into two single-channel analyzers (SCA) and counters that record the L X ray emitted by the Pu isotopes and the 59-keV γ rays emitted by ^{241}Am, respectively.

INEEL's approach is to couple the most appropriate characterization sensor(s) with the most appropriate deployment platform. INEEL has been the provider as well as the integrator of a variety of characterization sensors. Therefore, additional commercial and developmental characterization sensors can easily be integrated with the previously describe deployment platforms and the following data management system.

1.4.4. Real time data management system (RTMS)

The INEEL Real time Data Management System (RTMS) is a computer based program application that integrates hardware, software, sensor output, positioning information, and data analysis functions [6]. The RTMS is independent of any one hardware or sensor system and can be programmed to interact with the autonomous attributes of virtually any hardware and sensor combination. The RTMS can, therefore, be integrated with a site's existing monitoring hardware and sensors, thus utilizing available assets. For example, the RTMS is being utilized for site characterization at the DOE Fernald site [6].

The RTMS performs the integration of 3-dimensional positioning, real time data display, processing, transfer, and archiving of *in situ* data collected during remediation activities at the site. The system consolidates and automates many costly *in situ* measurement-related functions formerly performed separately and manually. RTMS is designed to enhance both large area as well as smaller more detailed and discreet hot-spot field screening functions.

The RTMS provides real time, on-site value to site managers and remediation operators in three primary areas.

- Health and safety of workers and remediation personnel,
- Segregation of contaminated and clean materials, and
- Verification of remediation operations.

2. IN SITU FIELD APPLICATIONS

In situ characterization techniques have advanced to the point where they are considered a valuable asset to remediation operations. The INEEL is actively deploying its systems in the field during remedial operations. This integration not only supports the remedial managers but it assists in furthering the development of these techniques. Two implementation examples include a RAMS deployment at the INEEL Waste Area Group 5 (WAG-5) site, and the DOE Mound Ohio-Erie canal remediation.

The INEEL WAG-5 operation involved the characterization of a ^{137}Cs contaminated area which was the result of a 1961 accident. Approximately 82,000 *in situ* measurements were obtained during this operation. This information was used to generate distribution maps of the ^{137}Cs contamination (see Figure 8). This information is being used to determine appropriate remediation plans. For example, this characterization could potentially reduce the volume of contaminated soils requiring treatment by up to 50 %.

The second example, which illustrates the value of *in situ* characterization, involves the DOE Mound site. Figure 9 presents a side-by-side comparison showing contoured ^{238}Pu data from

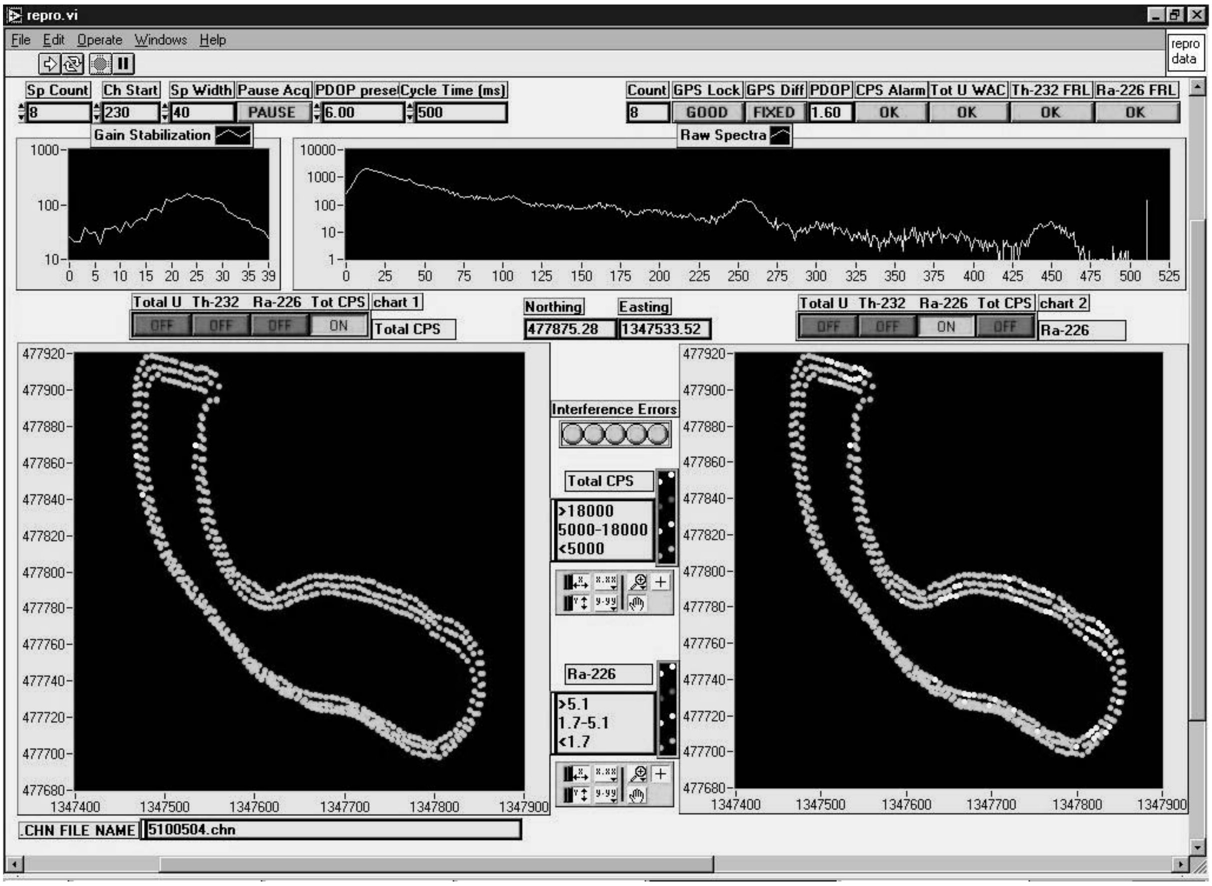

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FIG. 7. The RTMS integrated display screen used to manage the data flow process.

A) 30 hand collected samples, which took over 2 hours to collect; and B) 21,916 *in situ* measurements collected at the same location in only 91 minutes. The color gradation in both plots represent activity level as a function of the count rate. Hand collected sample points in panel (A) are superimposed on the panel (B) plot. The area of investigation involved approximately 672m^2.

3. INTERNET RESOURCES

The INEEL has had over 25 years of experience in the modeling and application of *in situ* measurements. Over this time period, a significant amount of information has been generated. An important reference for this information is now located on the internet and is available to interested parties. The INEEL Gamma Ray Spectrometry Center has recently established an internet Homepage to better disseminate its information as well as provide easy links to other related sites. This site provides online reference material such as the Gamma Ray Spectrum Catalogue, Scintillation Spectrometry, 2nd Edition; and the Gamma Ray Spectrum Catalogue, Ge and Si Detector Spectra, 4th Edition. The site also provides links to numerous resource sites for nuclear data, reference materials, professional societies, and other related links. The INEEL Gamma Ray Spectrometry Center homepage is located at *www.id.inel.gov/gamma/*

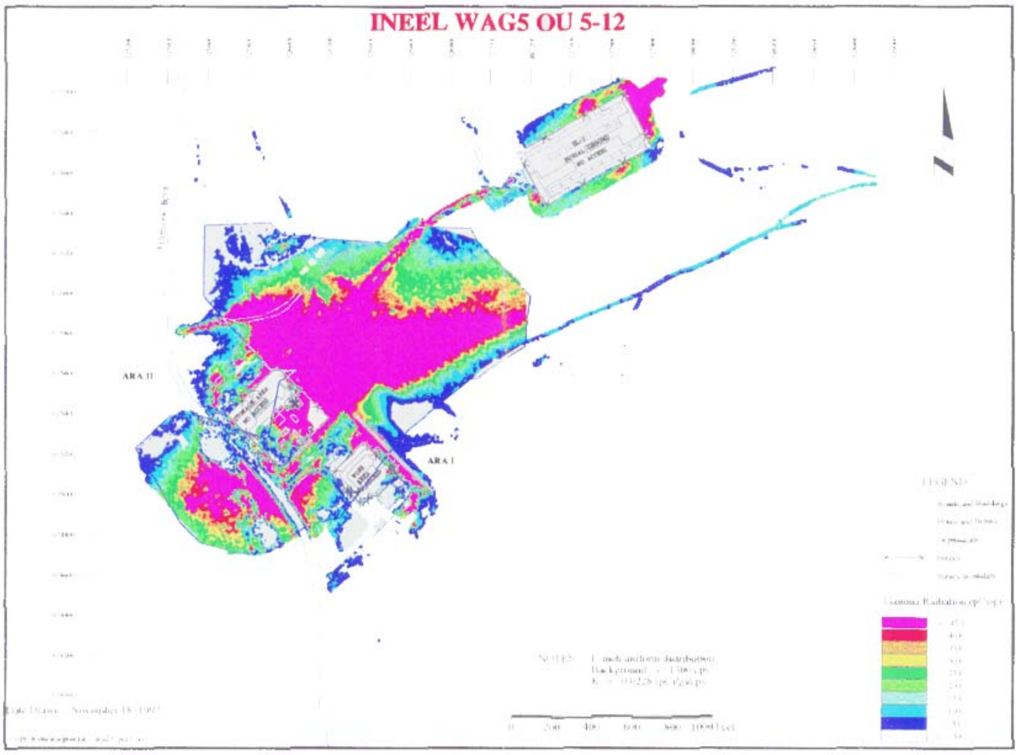

FIG. 8. INEEL WAG 5-12 137Cs distribution map.

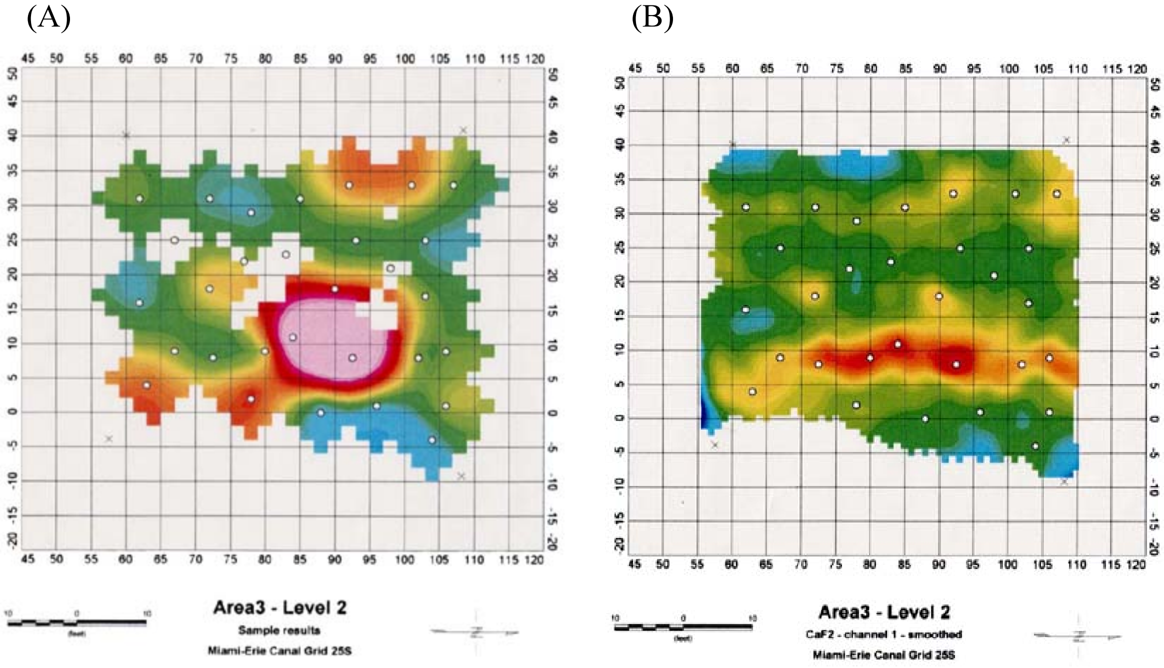

FIG. 9. Contoured 238Pu data from A) 30 hand collected samples and B) 21,916 in situ measurements.

4. SUMMARY

Innovative remediation technologies offer tremendous opportunities for cost savings, risk reductions, and operational efficiency. Implementation of *in situ* characterization systems during environmental restoration operations has shown that this approach results in several significant benefits versus conventional sampling techniques. For example, a flexible characterization system permits rapid modification to satisfy physical site conditions, available site resources, and cleanup requirements. As such, the rapid and precise collection of *in situ* measurements can:

- **reduce operational costs** by reducing the volume of material being remediated (i.e. more effectively delineating clean from contaminated material),
- **reduce uncertainty** by obtaining a “total-area field screen” versus a representative subset of hand-collected lab-analyzed samples,
- **lower on-site health risks** by reducing human exposure to contamination through reduction, and in some cases, elimination of hand samples during the field sampling process, and
- **improve management decisions** by providing real-time, densely spaced, repeatable, characterization data.

The INEEL promotes the development of innovative deployment, characterization, and interpretation techniques. Through on-going research, regulatory acceptance is being pursued to ensure that advanced techniques are available, for DOE and other customers, to achieve remediation goals and schedules. Regulator awareness and acceptance is being accomplished by integrating research into active environmental and waste management operations.

5. ACKNOWLEDGEMENTS

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