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Characterization of radioactively contaminated sites for remediation purposes

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FOREWORD

Radioactive contamination of the environment, including soils and waters, has occurred in many parts of the world as a by-product (sometimes accidental) of nuclear activities such as defence related operations, power production, research, medical and industrial applications.

Characterization of the contaminated site is essential before embarking on a programme for its remediation and ultimate restoration. Reliable and suitable data must be obtained regarding the distribution and physical, chemical and nuclear properties of all radioactive contaminants. Characterization data is necessary for assessing the associated radiation risks and is used in support of the required engineering design and project planning for the environmental restoration. In addition, continuing characterization can provide information regarding efficiency of the cleanup methods and influence possible redirection of work efforts. Similarly, at the end of the remediation phase, characterization and ongoing monitoring can be used to demonstrate completion and success of the cleanup process.

Other technical aspects of environmental restoration are also being addressed by the IAEA. This report is complementary to others dealing with the technology available for environmental restoration, development of strategies for environmental restoration, and characterization/monitoring of contaminated sites following cleanup.

The suggested methodology represents a contribution attempting to solve the issue of pre-remediation characterization in a general manner. However, a number of difficulties might make this methodology unsuitable for general application across the diverse social, environmental and political systems in the IAEA Member States. This TECDOC covers the methodologies used to characterize radioactively contaminated sites for the purpose of remediating the potential sources of radiation exposure and assessing the hazards to human health and the environment.

The initial draft of this report was prepared at a consultants meeting held in Vienna in October 1995. D.E. Clark of the IAEA served as Scientific Secretary at this and subsequent meetings. An advisory group meeting was held in May 1996 to provide national contributions to the draft report. Finally, a consultants meeting reviewed and revised the text. The report received an internal review in the Waste Technology Section of the Division of Nuclear Fuel Cycle and Waste Technology, and was finalized for publication by Z. Dlouhy of the IAEA.

EDITORIAL NOTE

In preparing this publication for press, staff of the IAEA have made up the pages from the original manuscript(s). The views expressed do not necessarily reflect those of the IAEA, the governments of the nominating Member States or the nominating organizations.

Throughout the text names of Member States are retained as they were when the text was compiled.

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1. INTRODUCTION

1.1. BACKGROUND

In recent years, it has become widely recognized that there are a large number of sites in countries around the world which have become radioactively contaminated as a result of nuclear fuel cycle activities; nuclear weapons programmes; the use of radioisotopes in medicine, research, industry; accidents; and so on. For some sites, fairly localized contamination has occurred as a result of specific industrial processes or operations as, for example, contamination arising from radium luminescence plant operations or the burial or disposal of radioactive materials. For others, widespread contamination with radioactive materials has occurred following planned activities such as nuclear weapons testing or from accidental releases of radioactive materials (e.g. the Chernobyl reactor accident of April 1986). The presence of such radioactivity, be it natural man-enhanced or anthropogenic, may pose hazards to human health or the environment. Therefore, proper characterization and remediation may be a mandatory requirement in order to reduce the radiological hazards to acceptable levels.

This TECDOC provides general information on the characterization of radioactively contaminated sites for remediation purposes. Thus, it presents technical approaches (e.g. strategies, planning, sampling, radiation measurements, laboratory testing, etc.) used to determine the extent of contamination and its chemical/physical form, nuclear properties, distribution, etc., primarily to support the remediation (cleanup) of radioactive contamination. It may be considered as a complementary or companion publication to other IAEA publications in the field of environmental restoration. The pertinent activities in this field currently include development of environmental restoration strategy [1], technology for environmental restoration [2], technical options for groundwater remediation [3], and characterization/monitoring to assess the effectiveness of remediation [4].

Past IAEA efforts with respect to environmental restoration have largely focused on accident responses (including cleanup of large areas as a result of a nuclear accident) [5–10]. A number of other publications have been issued which discuss broad aspects of the characterization of radioactively contaminated sites; existing remediation technologies, as well as those under development; and associated environmental restoration activities [11–19].

1.2. OBJECTIVES AND SCOPE

This publication is to serve as a source of information on characterization methodologies and techniques that are generally applicable for determining the characteristics of radioactively contaminated sites, including the specific radionuclides, their chemical and physical forms, site properties, geological features, and migration pathways. The publication should give guidance on characterizations that are needed for different stages of the evaluation process and also suggests methodologies to carrying out the particular characterizations required. It provides guidance on structured approaches to be taken during site characterization studies. The suggested methodologies and approaches represent a contribution attempting to solve the issue of pre-remediation characterization in a general manner. However, a number of difficulties might make some of these methodologies and approaches unsuitable for general application across the diverse social, environmental and political systems in the IAEA Member States.

The main goal of this publication is to facilitate the planning and execution of the characterization phases of a possible remediation. By giving advice and by alerting the reader to

the wide variety of methods available, some of them not widely used, it is hoped that the publication can assist in allowing an efficient and appropriate characterization to be carried out.

This TECDOC does not provide guidance on dose/risk assessment methodologies, but discusses some likely characterization techniques which could provide information for that and other purpose. Furthermore, it does not seek to specify the levels of radioactivity, or dose or risk reduction that a remediation ought to achieve. Interim guidance on relevant radiation protection principles is given in a recent IAEA publication [20]. Reference to this subject is also to be found in the Safety Series publications of the IAEA being prepared under the RADWASS programme. In addition, such factors as economic, political, cultural, and other scientific information can all influence these matters.

1.3. STRUCTURE

Following the Introduction, Section 2 describes typical examples of sites encountered throughout the world and the main reasons for characterization. It briefly discusses the types of risk or problems associated with the various types of site which may be encountered and highlights the need to identify correctly the problems which remediation must address and gives some general advice and warnings.

Section 3 focuses on the role of characterization in remediation; it enlarges on the reasons why a site characterization may be undertaken, and on the factors which will influence what may be required of such a study.

Section 4 deals with the approaches (e.g. organizational structures and planning objectives) which may be useful or even required in a site characterization. It describes the various models for planning and the characterization tasks that are likely to be needed.

Section 5 deals with the range of characterization methodologies and techniques which may be used in a characterization. The application of each technique, its advantages and disadvantages are discussed in a general way.

Section 6 provides a short summary of the report and the principal conclusions drawn.

This publication is supplemented by a list of references and by two Appendices providing information on characterization costs and environmental transport of radionuclides. In the Annexes, examples of various national experience in characterization of contaminated sites are given.

2. RADIOACTIVELY CONTAMINATED SITES AND SOURCES OF CONTAMINATION

The use of radioactive materials for a variety of purposes has resulted in contamination of sites (i.e. land areas, including structures, soils, rocks, biota, surface and groundwaters, etc.) throughout the world. The radionuclides involved may have been produced for a variety of reasons, including scientific research, industry, medicine or warfare. Another possibility is that they are simply an unnatural concentration of the naturally occurring radioactive elements. The affected sites can range from small localized areas in urban environments to larger areas encompassing many tens or hundreds of square kilometers.

The source of the radioactive contamination may be from a known activity at the site and the radionuclides involved may be known. Records may give information about the radionuclides involved and their likely disposition and chemical state. Alternatively, a chance discovery may have revealed the presence of contamination and no other information is available. It might be that the site is populated and immediate steps must be taken to ensure no harm is done, or it could be that people are easily excluded and there is adequate time to undertake investigations.

These and other differences mean that each site must be treated as a unique situation taking into account its own particular circumstances. In general, all potentially contaminated sites will need an evaluation (characterization) based on the principles given herein. In minor cases of contamination, many steps can be treated summarily, but usually all will still have to be dealt with. This publication is intended to provide the reader with general insight on how to decide the most appropriate procedures and also to be alerted about potential problems.

2.1. SOURCES OF CONTAMINATION

Some examples of contamination that might be encountered are given below. The list is not exhaustive but is intended to show the wide range of problems that might be found; they can vary in extent from large land areas to relatively small areas such as a manufacturing facility.

(a) Nuclear power production and nuclear fuel cycle activities

The various stages of the nuclear fuel cycle and the operation and decommissioning of nuclear reactors all have the potential to create contaminated sites. The contamination may include mill tailings; spillage of ore end product at the mine and in transport; waste from enrichment and fuel fabrication operations; fission product and actinide waste streams from reprocessing of fuel elements; radioactive effluents from normal operations of nuclear power plants; wastes produced during decommissioning of reactors; and major releases under accident conditions. Annexes D (Canada) and H (United States of America) give examples of contamination from uranium ore and yellowcake handling. Annex F (Slovakia) gives an example of ^{137}Cs contamination of river banks following accidents at a nuclear power plant. Annex G (United Kingdom) describes contamination problems occurring on railway property due to rain runoff of fission product contaminants from fuel transportation containers.

(b) Production and use of radioactive substances for medical, research or industrial purposes

Radioactive materials have been used widely since their discovery for a variety of scientific, medical and industrial uses. In some cases, either through ignorance, carelessness, or accident (see (e) below), sites have been left contaminated with residues of the operations. Such sites include factories where radium was used in luminescent paint and thorium was used in thorium-coated gas mantles. Other locations where radionuclides have been handled have the potential for leaving contamination. Annex B (Belgium) discusses the widespread dispersal of radium contamination in the surrounds of a former radium extraction plant.

(c) Mining and chemical processing associated with U and Th impurities

Because uranium and thorium are present in many ores containing other useful minerals, the mining of these ores and the processing to recover materials such as copper, gold, niobium, coal and monazite will generally produce waste streams containing significant amounts of radioactivity. These have the potential to result in unacceptably contaminated sites. Annex C

(Brazil) discusses contamination from the processing of monazite ores. Annex E (Croatia) describes contamination issues arising from naturally occurring radioactive materials (NORM) found in coal slag piles, as by-products of the fertilizer industry, and in other forms.

(d) Military activities and the production, testing and use of nuclear weapons

The manufacture of nuclear weapons involves the handling, transport and storage of large quantities of radioactive materials. The testing of weapons may involve nuclear yield and the release of fission products and activation products, or may involve the deliberate dispersal of radioactive materials in the environment. Some military use is made of depleted uranium which may contain fission products if obtained from reprocessed fuel. All of these activities have, in the past, resulted in contaminated sites, many of very large areas. Annex A (Australia) discusses the widespread plutonium contamination resulting from the experimentation with nuclear weapons development.

(e) Major incidents

In the course of nuclear weapons production and transport, there have been several severe accidents resulting in considerable contamination [17, 20]. These include: Windscale Pile 1 (1957), Kyshtym (1957), Palomares (1966) and Thule (1968). The spread of contamination by accident or by human ignorance are illustrated by the cases of the Chernobyl reactor (1986) and Goiânia (1987).

Table I presents examples of typical radioactively contaminated sites found worldwide and provides details of expected radionuclide contaminants.

2.2. IDENTIFICATION OF PROBLEM

A contaminated site may pose a cause for concern for various reasons. In many cases, it is a proposed change of land use that triggers an evaluation of the radioactivity present. In the original context the radioactivity may have been considered un-noteworthy and subject to adequate controls but a change in circumstance, such as a factory becoming a residence or housing a child-care facility, may cause a drastic re-evaluation of what is considered an acceptable level of contamination. This example serves to show that it may not only be the mathematical risk of injury that is relevant in deciding what to do; the public perception of what is acceptable must also be taken into account.

When a long term problem is being addressed, full historical information may sometimes allow a very well planned characterization with no surprises. Alternatively, a sudden public outcry about an unknown site may demand immediate, independent measurements to achieve credible demonstration of acceptability.

The solution of the problem will generally be either in producing a credible demonstration that the present situation is acceptable, or in a remediation of some form that reduces the risk, in a documented way, to an acceptable level.

The proposed remediation must solve the problem in its widest sense. That is, it must address concerns of a non-technical nature as well as scientifically-based risks. The associated characterization must be tailored for the intended purpose and suit the needs of the final solution. For examples in meeting a regulatory limit the required quality assurance (QA) practices must

be followed or the characterization may not be acceptable. Thus, regulatory authorities should be involved in the planning from an early stage. In addressing public concerns, the formal or informal (but recorded) approval of proposed actions and end-points should be obtained.

TABLE I. TYPICAL RADIOACTIVELY CONTAMINATED SITES FOUND WORLDWIDE

TYPE OF CONTAMINATED SITE	RADIONUCLIDES PRESENT AND COMMENTS
Radium luminizing works	Radium-226 + Daughters. Radon (Rn-222) (+D) emanation from the underlying ground could present a significant inhalation hazard. This hazard would need to be assessed and engineered solutions adopted. Radium compounds may be soluble in water and if so, the ground water pathway could be significant.
Thorium gas mantel works	Thorium-232 + Daughters. Thoron (Rn-220) (+D) emanation from the underlying ground could present a significant inhalation hazard. This hazard would need to be assessed and engineered solutions adopted.
Phosphate fertilizer production	U-238 + Daughters are most significant contaminants, Th-232 + progeny
Nuclear weapons test site	Contaminants are mainly long-lived fission products and original fissile components of test weapons. Many species are only significant in the first year or so after the creation of this type of contamination. Some radionuclides have short lived daughters in secular equilibrium.
Coal ash/slag from coal-fired power stations	Levels typically low compared with U mining or nuclear accidents. Volumes may be very large. Radon emissions may be significant. Present containments may be rudimentary or non-existent. See Annex E for details.
Nuclear power plant sites and environs	Levels may vary from generally low but widespread to locally high specific activity. Off-site releases may be through gaseous , liquid or particulate routes. In most circumstances, fission or steel activation product, and tritium, would be expected to be the most significant isotopes.
Mining, milling and processing of copper (and other) ores having high U/Th impurities	Radionuclides of the U decay series may be found: for instance, in waste rock piles and slags. Sites may be contaminated especially by Pb/Po-210 particulate. Ores include copper, tin, silver, gold, niobium and monazite.
Nuclear weapon and fuel fabrication plant (uranium enrichment and lithium production) also fuel reprocessing plants	Contaminants are mainly from uranium and plutonium fuel and fission products (generally with half lives of at least 1 year).

2.3. MAJOR FACTORS IN SITE CHARACTERIZATION

Major factors in site characterization, to be taken into account, include:

(a) Characterization can be a large consumer of project resources. Mistakenly, its practical importance to solving the problem may not always be understood or appreciated. In some instances, the characterizations may be the "last word" measurements (e.g. for peripheral areas) and, as such, their credibility is vital.

(b) The amount of characterization should be proportionate to the extent of the likely remediation effort. Over-characterization can result in a disproportionate fraction of the budget being spent on measurements, leaving insufficient means to carry out acceptable remediation.

(c) Characterization should be adequate to allow a properly designed remediation; one that does not involve excessive amounts of unnecessary effort or environmental damage.

(d) Characterization efforts should be sufficient to demonstrate the existence of clean areas and to provide credible assurances that unremediated areas are safe.

(e) Characterizations should have a sufficiently broad focus that any other unknown contaminants are detected at a stage when they can be dealt with efficiently.

(f) The characterization, in the first instance, and the subsequent remediation should not make things worse by ill-advised first attempts that magnify or spread the problem. A guiding principle can be "first, do no harm".

Section 3 gives more details of these considerations and shows the relationship between the characterization and remediation activities.

3. THE ROLE OF CHARACTERIZATION IN REMEDIATION

3.1. THE NEED FOR CHARACTERIZATION STUDIES

Once the concern about a potentially contaminated site has reached some threshold level, a process of evaluation and assessment should be undertaken. The process nominally followed is indicated in Fig. 1 and may lead to a remediation of the site or to a "no further action" remediation. A central feature of the process is characterization. In this context, characterization refers to those investigations, specifically including measurements, undertaken to provide information and data about the contamination and affected site environment.

As shown in Fig. 1, the steps usually taken include:

- evaluation of the severity of the problem in terms of radionuclide concentration or dose levels to determine whether there is a need to remediate;
- evaluation of the remediation alternatives including the feasibility, cost, and risk reduction;
- design of the selected remediation option;
- implementation of the remediation option; and
- verification and/or monitoring of the remediation performance.

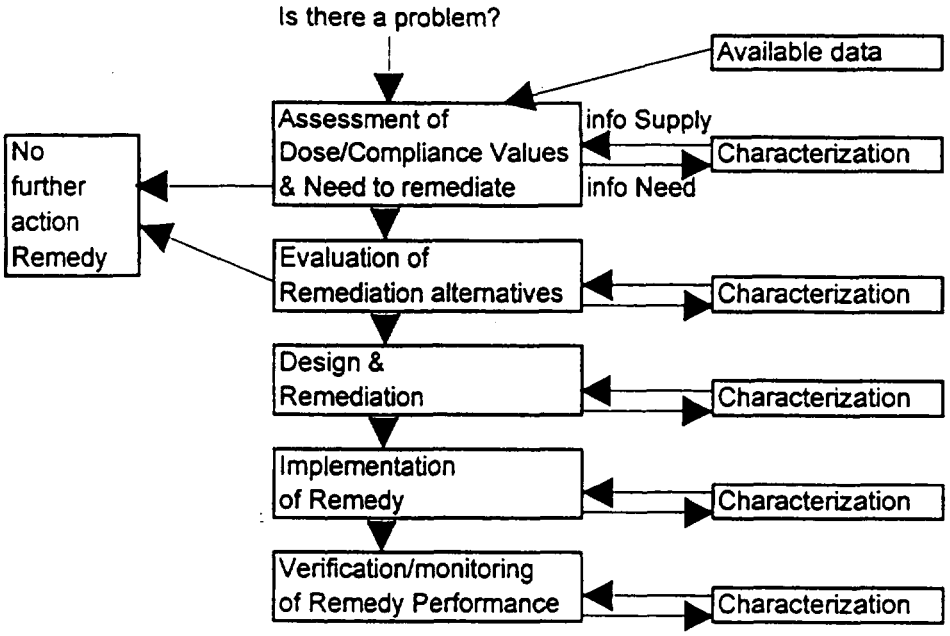

FIG. 1. Schematic diagram of the remediation process for radioactively contaminated sites indicating the role of characterization.

Characterization is a necessary prerequisite to provide critical information and data for each assessment/evaluation step in this process. Multiple characterization activities are common, with each characterization activity focused on gathering the information essential for the particular type of assessment or evaluation being conducted as indicated in the example in Table II.

While the general process of dealing with a potentially contaminated site shown in Fig. 1 is applicable to most problems, it may result in a range of characterization activities that vary widely in terms of scope, cost, and schedule. For example, a small "hot spot" of radioactively contaminated soil resulting from a recent small spill may be surveyed, hand shoveled up into a small container for proper disposal elsewhere, and the soil replaced with clean soil in a few hours. The steps in Fig. 1 would have been completed rapidly without much paperwork or calculation. The related characterization activities would have amounted to field survey instrument measurements of radiation prior to and after the hand shoveling.

Alternatively, the source of contamination may have been a leak of radioactive material that contaminated not only the surface soil in the immediate vicinity of the leak but also the subsurface soils and groundwater. Migration of the contaminant might now threaten the environment and population away from the leaking source. In this instance, the components of the assessment/evaluation process may be more complex and, consequently, the characterization activities may be more in number, more elaborate, and require months or years to complete.

TABLE II. ILLUSTRATIVE EXAMPLE OF SUCCESSIVE STEPS IN CHARACTERIZATION

STEP	ACTIONS TAKEN
Discovery of contamination	A routine radiological check of a disused site shows high levels of radioactivity in some areas. At this stage, results usually include only some count-rates from a radiation detector at several locations.
Confirmation and initial determination of scope	A grid survey using properly calibrated equipment is carried out. The property boundaries and exits of the natural drainage system are given special attention. This survey shows that dose limits for members of the public might be exceeded under some circumstances within the boundaries of the site. No evidence is found that contamination has moved off-site. The level of protection necessary for workers undertaking further characterizations is determined.
Primary characterization	If a significant problem exists, the primary site characterization survey is undertaken. The aim is to determine the exact spatial extent of the problem, including depth profiles, and to gather sufficient information to allow a full dose assessment for potential site occupiers and off-site populations. Any radiological threats to the environment from the existing situation or from possible remediation actions would be identified in such an assessment.
Secondary characterizations	If consideration of the results of the primary characterization led to a decision to undertake remediation, then a detailed characterization would be necessary to allow decisions to be made about the exact remediation method, and then on details of that action. At this stage, some characterizations will be needed to allow the full engineering design of the remediation. In addition, characterization of any waste stream may be necessary before the transport and disposal options for the waste are approved..
Remediation monitoring and quality control	As removal, treatment or fixation of the contamination proceeds, further monitoring and characterization continues to update remediation plans and to provide quality control information.
Verification and end-point dose assessment	Following the completion of remediation, a survey should be carried out to document the radiological status of the site and to allow a site for any future use.

3.2. CHARACTERIZATION TO SUPPORT EVALUATIONS AND ASSESSMENTS

3.2.1. Evaluation of contamination levels, radiation doses, and the need to remediate

The conceptual framework of radiological protection is stipulated in the IAEA Basic Safety Standards [21]. It recognizes two distinct situations: practices, which add radiation exposures, and intervention, which subtracts radiation exposures. Remedial actions taken to reduce actual or potential radiation exposures would constitute interventions.

The system of protection for intervention comprises two important principles:

(1) The proposed intervention should do more good than harm, i.e. the reduction in detriment resulting from the reduction in dose should be sufficient to justify the harm and costs of intervention.

(2) The form, scale and duration of the intervention should be optimized so that the net benefit of the reduction in dose, i.e. the benefit of the reduction in radiation detriment less than detriment associated with the intervention, should be maximized.

These measures may provide the basis for determining whether the contaminated site requires remediation.

Characterization actions may have to be taken in order to demonstrate compliance with a specified value of concentration in the contaminated media, corresponding in many cases to dose limit to a hypothetical group. Characterization results that indicate radioactivity levels well below regulatory limits may eliminate public concern and respond to political demands. See Ref. [20] for an example of criteria to be complied with.

In many cases, however, there may not exist specific regulatory levels for radionuclides in the media of interest or there are public and political concerns for human health and the environment not addressed directly by such compliance levels. In those cases, the assessment of radiation doses is undertaken.

The general types of information needed for the assessment include:

- nature and concentration of the contamination;
- potentially exposed population (workers, residents, etc.); and
- exposure pathways for radionuclides to reach the potential human receptors (e.g. exposure by inhalation, ingestion, external dose; the ingestion pathways may include exposure through drinking water or food chains).

There are several mathematical models to provide an estimate of the radiation risk to an individual or to the population [8, 22–26]. Such modelling may require comprehensive characterization of the contaminated site, including such parameters as the radionuclides present, their spatial distributions, and the expected site use and occupancy.

3.2.2. Evaluation of remediation alternatives

Evaluation of alternative remediation approaches, which involve consideration of the feasibility, cost, and potential dose reduction associated to each alternative, provides the basis for a decision about the remediation to be implemented. This decision, of course, may involve social, political, and institutional considerations as well as strictly technical matters. A brief discussion follows to indicate what types of information and data from the characterization studies may be required.

Although this report does not deal with site remediation per se, certain considerations relative to the remediation process should be noted here. As part of the characterization studies, possible options for remediation (if contemplated) will need to be considered so that appropriate information may be gathered.

3.2.2.1. Aims of remediations

Remediation strategies can cover a wide range of activities from characterization sufficient to demonstrate compliance with established criteria to complete removal of all contamination

down to some readily accepted endpoint. The potentially more expensive the remediation, the more characterization it is worth performing in order to allow the optimization of the remediation. Care will need to be exercised that the probability of finding any contamination is statistically quantifiable, appropriate and acceptable to interested parties.

It is important to identify the motivation behind the remediation. Once that is identified, appropriate characterizations can be carried out to allow the choice of a satisfactory or optimum remediation technique. Broad classes of possible remediation methods are discussed below with general guidelines as to the types of problem for which each would be appropriate and the likely characterizations needed.

3.2.2.2. Remediation options

Remediation options are generally identified in the early planning stages of a programme. A semiformal selection process may be followed by the decision makers in their approach to environmental remediation for a particular site. Newer results from the ongoing site characterization studies may be useful in evaluating options and in considering changes in the strategy for remediation.

Remediation by characterization: demonstration of "no problem"

In many circumstances, the problem will be a public perception, or a scientifically-based suspicion, that a site is contaminated and poses a risk to potential occupants or neighbors. A well-planned characterization of the site may be able to demonstrate, to the satisfaction of the public or authorities, that any risk is well within acceptable limits. It will be worthwhile considering exactly who needs to be convinced by the report before carrying out the measurements.

If the site was known to have been used for handling radioactive materials and the problem is one of public perception, then a survey would be necessary which has a reasonable probability of detecting significant contamination. "Significant contamination," in this context, would be that which, if discovered, would pose an unacceptable risk or one which would require further investigation. A knowledge of the process originally carried out at the site may help decide whether to look for wide-spread low levels of contamination or isolated hot-spots. It is likely, in this case, that the radionuclides used would be known.

The problem may have arisen from the discovery of some contamination at a site where no contamination was suspected. In this case, the survey will need to look for various possible radionuclides and may need to combine a detailed search for hot spots with a wide-area search.

Where public perception of risk is the issue, provision for public comment on and public education about the survey may be appropriate. Alternatively, regulatory authorities may need to be consulted to ensure that the survey meets their requirements.

Exclusion: avoidance of the problem

There are circumstances where the best remediation is to simply exclude the potentially exposed population from the site of the contamination. This is particularly true when the costs of remediation are too large, such as in the case of nuclear test sites. In cases where radionuclides involved are short-lived, temporary containment combined with exclusion may be a solution.

Costs associated with this strategy include the direct costs of fencing or surveillance to achieve the exclusion, and the indirect costs, represented by the restricted use of site or loss of value of affected land.

Containment: covering or fixation *in situ*

The contamination may sometimes be attached to valuable equipment or facilities and removal may be very difficult. If external exposure is not significant, for example in the case of a pure beta-emitter, fixing the contamination by paint or other substance may allow continued use of the equipment for its normal working life. It may be necessary to ensure that records of the contamination are kept to avoid exposures during eventual disposal of the equipment. Warning labels or engraving may be worthwhile. This form of treatment may be particularly appropriate when the potentially exposed population is limited and perhaps includes only specially trained personnel. An example would be surface contamination in a scientific laboratory.

Where contamination is expected to lead to significant external exposure, it may still be possible to render it safe by sufficient covering with strongly absorbing material, such as earth or concrete. Such treatment may have indirect costs associated with the changes in the site due to the covering.

Characterization would need to identify the radionuclides and show that containment would last sufficiently long to contain the contamination for the life-time of the radionuclide. Activity levels would need to be low enough so that the risk of containment failure was acceptable. When very long half-lives are involved, considerations of what is a reasonable period of institutional control must be dealt with.

Treatment: concentration or extraction

There are far too many possible treatments to be discussed in detail here. Whatever methods are being considered, it is likely that particular characterizations will be required to allow the method to be properly designed and carried out. This will require determination of the extent of contamination and may involve chemical, physical and other investigations.

Removal: bulk removal of contamination and medium

This form of remediation is, in some ways, the simplest, and may be the most effective, but it may be the most expensive. It will probably involve significant environmental impact at the site, the generation of a waste stream with environmental consequences elsewhere, and the transport of a hazardous material. Characterizations will address the extent of contamination and be needed to plan each of these aspects. Considerations of waste disposal will also be relevant in other contamination scenarios.

The presence of non-radioactive hazardous materials may affect the remediation alternatives selected for evaluation: the presence of organic or biological materials, or of chemically hazardous material, may preclude certain options for treatment and/or disposal or may dictate particular handling precautions or methodologies for radioassay.

In summary, remediation alternative evaluations will typically focus on remediation performance designed to:

(I) *Satisfy an end point criteria.* Actions have to be taken in order to allow a residual radioactivity leading to acceptable radiation risk for a specific endpoint scenario to release the site for restricted or unrestricted use.

(ii) *Optimize the dose reduction.* Actions are taken to obtain the greatest dose reduction for acceptable costs or to achieve a given dose reduction for the least possible cost.

(iii) *Avoid the spreading of contamination from the site.* In case of early intervention, actions are taken in order to prevent the transport of radioactive material inter and intra ecosystems, by several environmental mechanisms, such as resuspension, soil or groundwater migration and biological uptake.

(iv) *Avoid an unnecessary risk, either now or in the future.* Actions are taken to minimize or even eliminate the actual radiation risks and potential future risks when it is cost-effective to do so at the present time.

In all of the above evaluations, a cost–benefit analysis may be required to determine the optimum balance between expenditure and risk. Assessment will be required of the long-term benefit of remediation weighed against the short term risk associated with the remediation process, both in terms of exposure of workers and public and in the cost of non-nuclear risks (e.g. industrial accidents during soil removal and transport).

3.2.3. Design of remediation approaches

When a decision has been made among remediation alternatives, a detailed design of that alternative may be needed to achieve the expected performance of the remediation. Typically, the level of evaluation of the chosen remediation approach that occurred during the earlier evaluations of several approaches will not have been detailed enough to produce an engineering design. The activity of designing the remediation, whether it is a relatively simple excavation of soil for disposal elsewhere or a complex in situ soil treatment, usually will require additional information and data to support the design activities.

It is difficult to generalize what the data requirements for this characterization are because they will depend from the selected remediation technology and from site-specific conditions. However, in addition to more detailed information about the contamination nature, extent, and potential migration pathways, other information related to engineering and construction matters may be required.

3.2.4. Implementation of remediation activities

Characterization studies in support of the implementation of a remediation may be very limited, or they may be extensive and critical to the implementation process.

In cases where the contamination is easily detected in the field, the detailed delineation of soil contaminated at or above a specified cleanup level may be left (for cost purposes) until actual excavation is underway. (In other cases, it may be done in advance as part of the design process.) When delineation occurs concurrently with excavation, this characterization needs to be planned carefully to support the ongoing remediation as construction delays can prove costly.

Another characterization activity that may be required to support the implementation of a remediation is the assay of materials generated as waste during the remediation. Regulations on disposal of waste may dictate such characterization support as a critical component of the cleanup.

3.2.5. Verification and monitoring of remediation performance

An important part on any remediation is verifying that the desired performance of the remediation has been achieved. Such verification activities can range from simple radiation surveys to assure the source material has been removed to complex, long-term monitoring of groundwater to detect any plume movement. In any case, this activity usually contains many characterization elements and, almost certainly, its design depends on earlier characterization of the contamination and site environment [4].

4. APPROACHES TO CHARACTERIZATION

The general approach to characterization in support of remediation is to:

- define the objectives and strategies for the characterization;
- plan specific characterization investigative tasks including important associated activities such as quality assurance, data management, and health and safety;
- conduct the characterization;
- analyse and interpret the data; and
- report the results.

As the first step in the process, developing a good characterization strategy to meet the objectives of the programme is essential (as indicated in Section 2). Once initial planning is conducted, the office, field, and laboratory investigations can be undertaken to collect the necessary information to meet your study objectives. Upon completion of the investigation tasks, the collected data are reduced and integrated to develop a site conceptual model. A description of the investigation and its results are then typically documented in a report. This Section discusses all aspects of the approach. The methodologies and techniques required to perform the investigation tasks are discussed in Section 5.

4.1. DEFINING THE OBJECTIVES AND STRATEGIES FOR CHARACTERIZATION

There is great variability in the details of characterization approaches appropriate to specific problems and sites. Differences among sites due to the heterogeneous character of the natural environment and to the nature and history of contamination are enough to require different approaches. However, the variety of other important influences on the remediation problem definitely require that a characterization approach be designed to address all such issues. The guidance here focuses on the important elements that any strategy developed for characterization of a specific site should consider. It also addresses the value of flexibility and phasing of study components to allow revision of the strategy as new information becomes available.

Characterization data are an important element in making effective remediation decisions. Clear specifications of the objectives and strategies for the characterization are important. Often characterization activities are begun with only vague notions of these objectives and strategies, and the problems that have commonly resulted include:

- wrong variables were measured;
- some variables that were needed were not measured;
- wrong set of samples was taken;
- data are compromised by interfering factors;
- funds were wasted on unnecessarily sophisticated instrumentation and analytical techniques;
- realized accuracy and precision are inadequate;
- more samples were collected and analyzed than needed, and/or there are too few samples collected to answer the question; and
- methods were not approved by regulators.

Successful characterization activities typically are found where the strategy is consistent with the characterization objectives. This subsection discusses how these approaches can assist in determining the quality and quantity of data to be acquired and in establishing the phasing and timing of the characterization activities.

4.1.1. Quality and quantity of data

Several methods currently being implemented (under many names and variations) to avoid inadequate or insufficient characterization are systematic planning tools based on technically sound approaches. They focus the designers of characterization strategies and plans on determining the type, quantity, and quality of data that will be sufficient and appropriate for the data's intended use. Such methods have been shown to improve the effectiveness, efficiency, and defensibility of decisions in a resource-effective manner. The process is amenable to operations that are iterative and interactive. This provides important flexibility as remediation operations and detailed measurements will often reveal problems areas not foreseen in the original characterization design. A stepwise summary of elements common to this type of process is outlined below.

(1) **Identify the problem that the characterization is to address.** Decision makers, regulators, and other interested parties address the principal problems and identify related issues. During this step, the parties also gather and review any existing information about the contaminated site that might help address the problems and issues, narrow their focus, or raise new issues.

(2) **Determine the decision to be made.** Having identified the problem, decision makers, regulators, and other interested parties must determine the decision to be made to help solve

that problem. Characterization efforts typically are directed at a specific decision --for example, deciding whether the levels of contamination at the site exceed regulatory standards or deciding whether remediation of contaminated soil is better accomplished by in situ treatment or by excavation and removal.

(3) **Identify inputs to the decision**. This step is typically critical to defining the objectives of the characterization because often much of the information required as input to the decision process is unavailable and is sought through characterization studies. For example, the horizontal extent of the contamination at the ground surface may be known, but its subsurface extent may not be.

(4) **Define the study boundaries**. The boundaries include the spatial and temporal limitations needed to be considered when making a decision using the characterization data. The boundaries can have a profound effect on the characterization strategy. For example, specifying whether individual environmental pathways are to be included or excluded in the characterization may affect the scope of the study.

(5) **Develop a decision rule**. Outputs from the previous steps are integrated into a series of single statements describing the logical basis for choosing among alternatives during the characterization. For example, a characterization aimed at delineating the spatial extent of a particular radionuclide in soil by a combination of surface soil surveying and subsurface sampling might have the following decision rule: "if the value of surface survey measurements exceeds a given threshold, then subsurface samples will be collected at 1 and 2 meter depths". By developing such explicit "if ... , then..." decision rules, characterization decisions can be made consistently by all of those assisting in the characterization.

(6) **Specify limits on decision errors**. Limits on the decision errors are developed by those who will make the decision. This permits the establishment of appropriate limits of uncertainty for the data that are obtained.

(7) **Optimize data design.** Finally, the information from the previous steps and knowledge of the uncertainties associated with measurement techniques and instruments are used together with statistical techniques to create alternative data gathering (e.g. sampling and analysis) designs. The design designates the quantity and quality of the sampling necessary. For a characterization, the "optimum" design provides the best balance among the expected cost, ability to satisfy uncertainly objectives, the feasibility of the study, and less tangible factors.

Related data quality issues that are reviewed in the establishment of a measurement design include the following:

- Precision: a measure of the reproducibility of measurements.
- Accuracy: a measure of the bias in the result.
- Representativeness: a reference to the high degree to which a particular measurement or sample reflects the typical condition.
- Completeness: the percentage of measurements that are judged to be valid.
- Comparability: the confidence with which one set of data can be compared to another.

4.1.2. Phasing of site characterization

The timing of characterization activities is driven in many ways by the requirements of the assessments and evaluations supporting remediation. In most instances, several characterization studies will be conducted during the course of remediation. In some cases, individual characterization studies will be carried out in several phases.

Typically, characterization activities are phased by beginning with reconnaissance efforts to evaluate the potential of a problem and by advancing to more complex measurement designs to support detailed assessments. The measurements undertaken usually progress from the methods that are non-invasive to those that are invasive. In general, non-invasive technologies are relatively less expensive, reduce the potential of worker exposure to the contamination, reduce the quantity of secondary waste, are regulated under less stringent regulatory permit requirements, and tend to be quicker. Historically, the most overlooked types of non-invasive site investigation are geophysical methods which in some instances can provide very high quality information to support the development of a conceptual model of the subsurface geologic environment. Here non-invasive is defined as a surface method or remote sensing method, minimally invasive methods include direct push methods and use of existing wells, while invasive technologies require additional soil boring and well drilling water. An example of changes in measurement designs in phased characterization activities is presented in the following text.

Phase I: Phase I is a preliminary investigation consisting of a formal desk study and a site reconnaissance. This investigation provides the information needed to decide site-specific investigation objectives and procedures for the subsequent on-site investigation. Determining the health and safety and environmental protection requirements for on-site work is an important task of preliminary investigation.

Phase II: The goal of a Phase II non-intrusive exploratory investigation is to use existing data in conjunction with surface field techniques to synthesize a general conceptual model that will allow for further refinement of the site characterization programme, such as optimal placement and spacing of sampling points during the following characterization phases.

Phase III: The conceptual model developed as a result of Phase II should be verified and refined using minimally intrusive and/or intrusive tests. An example of this type of approach would be the use of the cone penetrometer, which can be configured with a variety of sensors to collect groundwater, soil gas or sediment samples, to measure in situ contaminant concentrations using spectroscopic methods, to make geophysical measurements, as well as measure traditional geotechnical parameters to determine lithologic properties of the subsurface.

Phase IV: In many cases, access to the subsurface for sample collection or monitoring well installation can only be obtained by drilling and/coring. Since drilling and management of drill cuttings at hazardous, mixed waste, and radioactive waste sites can be prohibitively expensive, the bulk of monitoring well installation should be completed after non-intrusive and minimally intrusive studies have been used to prepare a conceptual model that can be verified and refined by strategic placement of monitoring wells.

4.2. PLANNING THE CHARACTERIZATION

The planning of the characterization is based on the previous definition of the objectives and strategies. How detailed the planning is, will depend on the size of the project. In general

there may be a sampling and analysis plan for the surveys of the site. However, other aspects such as quality assurance, data management, health and safety aspects and the project management will need careful attention and should result in a specific plan.

4.2.1. Sampling and analysis plan

Based on the definition of the objectives and strategies for characterization, a Sampling and Analysis Plan is developed which provides the implementor of the characterization with the necessary detail to conduct the characterization investigations in the field and in the laboratory. This includes what surveys will be done, what samples will be taken, and how they will be collected and measured (e.g. sampling point, time of collection, depth of sampling, and other variables necessary to tie a measurement of a specific sampling location in time and space). In the case of accelerated or expedited characterization, the plan will emphasize the process by which specific sampling activities will be driven on the basis of results. Planning documentation should also define analytical procedures for field and laboratory measurements to determine detection limits, precision, and accuracy.

Sampling points should be located where the contamination is most expected to occur, and only a limited number of constituents should be analyzed. The sampling plan may include the possibility of taking decisions on extending or limiting the survey on the spot, depending on the results of the measurements in the field. A limited number of samples may be defined in the beginning, to verify correlation between survey data and sampling analysis, increasing the number of samples if the correlation is not as good as expected. For complex cases, mathematical techniques may be used to define a statistical distribution of sampling points to give the wanted precision of the distribution of contamination. If pursuing the identification and location of "hotspots" of contamination, field screening should be used to help initially locate the areas of contamination and the boundaries of contamination. Later phases of investigation will be required to refine the configuration of the contaminant boundaries. Attempting to prove that a site is clean can be difficult and invariably requires very extensive investigation. In this latter case, sampling should always be systematic, and testing for a wide range of contaminants may be involved.

In the development of a sampling and analysis plan, due consideration should be given to the potential spreading of contamination, accessibility of the site and sampling locations, and the pre-sampling approvals that may be required prior to the performance of field work. These considerations are briefly discussed below.

Spreading contamination. Characterization practices should be designed in such way as not to contribute to the further spread of contamination at the site, or off site. This is of particular concern when dealing with radioactive contamination. For example, contamination can be spread through uncontaminated aquifers as a result of poor drilling and well completion practices. Care should be exercised so that onsite workers do not inadvertently carry radioactive contamination off the site through inadequate decontamination processes.

Accessibility. During the planning process it is necessary to consider access logistics, including the ability to physically gain entry to the site, especially for any equipment that is brought in (e.g. drilling rigs, cone penetrometer trucks). It should also be considered whether there are any overhead or underground utilities which may impact the investigation. It may be necessary to limit access to a contaminated area to only specially trained site workers and to allow for a decontamination zone for equipment and personnel.

Jurisdictional concerns. Before initiating field work, it is essential to obtain any approvals necessary to access the area to be characterized. Authorization may be required from governmental or private parties. In addition, it may be necessary to obtain certain permits for digging, drilling, or installing any groundwater wells. A check list of requirements should be prepared to ensure preparedness.

4.2.2. Quality assurance plan

A quality assurance plan (QAP) defines how the integral quality of the environmental data collection process is controlled. The quality objectives are to achieve technically sufficient investigation work, at the right time and at the right price. The QAP will include the description of all relevant responsibilities/functions and a named person for each function. It will also include procedures and working instructions on all operations that may be involved in the final quality of the results. Site operations which should be subject to quality assurance and quality control procedures include monitoring, sample collection and handling and, where appropriate, drilling works and waste disposal. Environmental protection and health and safety aspects should be given highest priority.

For laboratory investigations, the quality assurance plan should define the acceptance limits of measured data in terms of sensitivity, reproducibility, detection limits and accuracy. Quality assurance/quality control protocols are employed to demonstrate the reliability and sample analysis data and compliance with documented sample handling, storage, preparation and test methods.

The quality assurance plan should guarantee that the results are accurate, precise, reproducible, reliable and timely.

4.2.3. Data management plan

As an integral part of the overall characterization plan, the data management plan should define the amount and nature of data to be collected. There will be a strong relation between the quality assurance plan and the data management programme because both aim at the end-product of the characterization data as input for assessment and decisions.

The definition of the databases that will be used in the characterization tasks is also a part of the data management plan. New data will have to be incorporated in the databases as soon as possible to allow quick interpretation and subsequent steering of the characterizations to be done. Determination of working formats facilitates this action. There can be a different form for each task, but a number of items will be common to all forms.

The further handling of the samples can be documented in the same way. A chain of custody record may be important in many cases. Data collection procedures and working methods should be standardized as much as appropriate for data evaluation and for future use of the data. The choice of archiving methods for data and samples should be made taking into consideration the future availability of relevant items (storage media, references for mapping coordinates, computer data format and backup system). Good practice in data management includes the identification of a person responsible for data collection. A quick evaluation of incoming data, can avoid long-term gathering of nonrelevant or faulty data. All changes to procedures should be approved by a responsible person and be logged for traceability.

4.2.4. Health and safety plan

The investigation of contaminated sites will involve site workers to close, and possible prolonged, contact with potentially hazardous materials. Therefore, health and safety should be a fundamental consideration in the design and the selection of investigation methods (e.g. use of minimally invasive techniques). Site characterization personnel should be suitably trained and equipped. Planning should prescribe detailed working procedures, including monitoring and dosimetric equipment, protection clothing, identification of access, etc.

4.2.5. Project management plan (cost and schedule)

Once the elements of the characterization planning have been completed, it is necessary to make cost estimates and construct a site characterization time schedule. Estimating costs and time schedules may require revisions in the site characterization strategy. Cost estimates should consider cost of materials, labour, travel, accommodations, and any subcontracting support. The scheduling should anticipate contingencies and decision points. A project manager should be assigned to ensure compliance with the characterization budget and schedule. Selected methodologies to reduce characterization costs are illustrated in Appendix I.

4.3. CHARACTERIZATION TASKS

4.3.1. Site description

It is necessary to have a sufficiently clear description of the site to allow for good planning and use of resources in developing a remediation strategy. The site description is normally constructed by making use of existing data and information, as well as conducting investigations, as needed and appropriate.

4.3.1.1. Historical investigation

The historical investigation is a part of the planning stage and is essential to obtain initial information on the nature and extent of the contamination. It should include gathering information on the site layout, operational characteristics, materials handled, measurement results, accidents, etc. Since previous data may be of different quality and reliability, verification of historical information is usually necessary.

In certain locations, it may be necessary to consider the concerns of native inhabitants, where sampling may occur within ancestral, or sacred grounds, before conducting digging, drilling operations, or other intrusive activities. Destruction of cultural or archeological resources should be avoided.

Example of sources for historical investigation may include:

- site and company records (drawings, production logs, disposal records, etc.);
- interviews (personnel, residents);
- maps;
- photographs (i.e. of operations, facilities);
- scientific and technical literature; and
- reports on former characterizations or remediations.

4.3.1.2. Present situation at the site

The description of the present situation of the site will give the information necessary to delineate the suspected area. It will give the opportunity to define a grid/coordinate system that can be used as a reference for the location of all subsequent surveys, measurements, and samples.

As an example, the description may include:

- maps in a scale with sufficient detail to have all relevant information, usually 1/10000 for detailed characterization, other scale for a larger area;
- plans for structures (factories, houses, sewers, roads, etc.);
- a photographic survey of the site and its surroundings; and
- data on the relevant authorities: police, local community, government agencies, waste management agency, and so on.

4.3.1.3. Flora and fauna investigation

Investigations of flora and fauna at, and in the vicinity of the site, are important not only for assessing any potential existing or future inherent damage, but also in assessing their potential impacts on humans through the food chain. In addition, both flora and fauna can be used to help delineate the nature and distribution of the contamination. Distressed vegetation often provides clues to contamination. Further, certain flora may be uptaking contamination, providing clues pertinent to bioremediation techniques (i.e. agricultural remediation techniques).

4.3.2. Identification of radioactive contamination

The goal of the radioactive contamination investigation is to provide identification of the radionuclides involved, to assess how much radioactivity is present, and to determine the horizontal and vertical (depth) distribution is in order to evaluate the radiation doses for all potential recipients.

As a first step, the exact location of the source, its present or past release rates and types of released substances, should be determined. Contamination may result from buried leaking drums, leaking underground piping, or leaking waste dumps. It is recommended to remove the source of the contamination before proceeding with any remediation of surrounding media.

The investigation of the radioactive contamination of the site has to provide the initial data to circumscribe the extent of a possible problem and to decide on any necessity of immediate remediation. Depending on the goals of the characterization, the limitations of time, budget, available staff and available laboratories and equipment, a choice of relevant items to be investigated initially has to be made.

Measurement of the background radiation is necessary as a reference for the characterization of the site. This can be done in some cases by measuring background radiation off-site, i.e. by choosing a site with similar characteristics as the one being investigated. In other cases, spectrometry may differentiate between natural activity and the investigated contamination.

In many cases, characterization planning will represent a phased process of an iterative character. At first there may be a coarse radiation measurement survey, in situ and not intrusive, with appropriate survey meters or passive dosimeters or activated charcoal detectors for radon, to define the boundary of the suspected site. Depending on the findings, subsequent measurements may include in situ gamma spectrometry, alpha and beta measurements and continuous radon monitoring or the placement of alpha-track radon detectors for a longer period. In the case of large contaminated areas, mobile or aerial gamma surveys may provide valuable information.

If decided so in the planning, a number of samples of different media (soil, water, air, biota, structures) will be taken for calibration of the surveys and for direct characterization of the radioactive contamination (activity, specific activity, isotopic composition). The number of samples of different kinds has to be determined in such a way that the needed information is covered.

The investigation of the horizontal- and depth- distribution of the contamination can give valuable information for the determination of the possible volumes of waste to be expected at cleanup, while spectrometry and sample measurements contribute to the knowledge of the characteristics of this radioactive waste.

Some possible types of radioactive contamination to be investigated are:

- contamination in a waste dump;
- soil contamination;
- water contamination;
- contamination of biota;
- contamination of structures (buildings, roads, pipes, etc.); and
- air contamination (aerosol, radon, and others).

4.3.2.3. Non-radioactive contaminants

Although the characterization of non-radioactive contamination is not included in the scope of this publication, it may be necessary to investigate some hazardous materials in the site to be characterized. Processes used in the past (or still continuing) may have led to disposal of heavy metals together with radioactive substances. Organics, toxic or corrosive chemicals may have been used in processes together with radioactive material. These hazardous materials are of consideration for disposal of waste after cleanup. Organic complexing agents may change the migration behaviour of the radioactive contamination. The presence of hazardous matters may affect the design of remediation actions and clearly need to be included in health and safety considerations for the characterization.

4.4. REPORTING

Comprehensive and accurate reporting of site characterization work is fundamentally important. Reports should describe the findings of the investigation and document the decisions made and actions taken at all stages of the investigation. Site investigation reports may be required for different purposes and to serve the needs of a number of different parties. Those reports should be mindful that they may be viewed by many different people, only a few of whom may have detailed technical knowledge. It is vital, therefore, that a high standard of presentation is achieved and that the results and their assessment are presented with clarity and precision.

5. CHARACTERIZATION METHODOLOGIES AND TECHNIQUES

5.1. INTRODUCTION

A sensible phased approach should be adhered to resulting in a number of characterization tasks being set up and followed. The objective of Section 5 is to provide information and references on a variety of techniques, instruments and methods which may be of use during a characterization study.

Selection of the appropriate measurement instrumentation and protocols for their use is a very important component of any characterization project. Not all methods or instruments, however, will be used in any single study, and indeed many smaller characterizations may be completed using only one or two items from this section. It must be emphasized that most site remediations are unique, and that an organization embarking upon a characterization will need to exercise careful thought as to the methods which best meet their objectives.

A wide range of site characterization techniques exist, some of which will require changes in methodology depending on the particular site and contaminant [27]. Direct on-site measurements of radiation levels may be a good default option when gamma emitters are contaminating the site and are present at the ground surface. When external gamma dose is of concern or when hot spots have to be identified, the series of techniques related to gamma radiation measurement are of major interest. Sample collection will be the prerequisite to most measurements of alpha and beta emitters. With few exceptions, sampling and laboratory analyses will be required for the determination of radioactive isotopes whose concentration in the environment may not be amenable to direct field measurement.

Sometimes, laboratory measurements may provide results with large variations between individual samples of the same population while in situ measurements may give more representative results. However, regulators may prescribe that the concentration of a given pollutant be obtained from laboratory measurements of samples.

The knowledge of radionuclide behaviour on site, however, may require studies of pathways involving a wide range of disciplines such as hydrogeology, chemistry and biology. Consideration regarding these studies is presented in Appendix II.

5.2. CHARACTERIZING RADIOACTIVE CONTAMINATION

The nature and amount of radionuclides present will probably need to address the full three dimensional distribution of radioactive contamination. There are many possible contamination scenarios, including:

- a superficial distribution of deposited activity;
- activity which has been deposited on the ground surface and which has migrated into the ground;
- activity which has been buried or covered (e.g. by ploughing or building operations);
- activity which is to a greater or lesser extent distributed through a substantial depth of soil (e.g. waste tips);

- activity which is deeply buried (e.g. due to leakage from underground storage tanks or pipelines which have carried active material);

- activity distributed as hot particles which are individually hazardous;

- activity which is uniform over large areas or volumes; and

- localized hot-spots.

Sometimes there may be *a priori* reasons to believe that the distribution is known, or can be established with little effort. In other cases, determination of the distribution will be a major part of the characterization. Gamma emitting nuclides found near to the surface may be amenable to measurement by non-invasive means, whereas deeply buried material will usually require more complex and costly methods.

The identity of the radionuclides present may be obvious from the history of the site, or may require detailed measurement. Even where the identity of the contaminating radionuclides is known, care may need to be taken that radioactive decay products may be present which are not in equilibrium with the parent.

Non-intrusive in situ techniques have a number of advantages over conventional soil sample collection and analysis. In situ measurements gather data in real-time that can be analyzed immediately for decision making purposes, while conducting the characterization survey, and may offer a substantial cost savings over conventional sampling and analysis. In addition, the area observed by the in situ detector may in fact give a more representative picture of the extent of contamination than conventional sampling and analysis. However, an advantage with taking soil samples is that there is the possibility of doing additional measurement (e.g. depth characterization of radionuclides, radiochemical analysis for alpha and beta emitters, and determination of physical or chemical form of the contaminant).

A combination of remote readings and collection of samples is often the preferred approach. It is generally cost effective to focus primarily on remote readings and collect sufficient samples to verify that the remote readings are providing the required information. A phased approach to characterization that optimizes data quality objectives versus the cost of acquisition and analysis should be considered. It is often practical to obtain an initial picture of the contamination with an in situ screening survey, and then to follow up with a boundary definition survey and a more detailed characterization of the site.

5.2.1. Field measurements

5.2.1.1. Techniques for determining measurement location

It is important to know the exact location whenever measurements are made in situ or samples are taken. This allows the data collected to be properly coordinated and analyzed. Reproducible knowledge of the coordinates of measurements may negate the need to repeat work in the future and may provide a standard of quality assurance which is more readily accepted by regulators. Consideration should be given to whether a locally defined frame of reference is acceptable (for instance measuring locations relative to buildings or roads) or whether (perhaps if extensive site demolition is envisaged) a more permanent frame of reference (e.g. latitude and longitude) will be required.

The positioning techniques vary in both sophistication and performance and have different fields of application. Traditional surveying techniques require trained personnel and can be slow. Modern technology has provided methods which can assist considerably in the characterization process.

Traditional survey techniques

Traditional survey methods can generally provide the accuracy required for site characterization methods and can give good standards of quality assurance provided that they can be linked to a series of well-defined and permanent reference objects. However, such measurements may be time consuming and labour intensive and may limit the speed at which measurements can be made in the field. Good measurements may be possible using only limited equipment (tape measures, surveyors wheels, etc.) but will require that personnel have adequate training.

Global positioning system (GPS)

The United States Department of Defense operates a satellite-based system of absolute positioning (known as the global positioning system, GPS) which allows a low-cost hand-held device to give locations anywhere in the world to an absolute accuracy of about 100 m. The signals from the satellite are usually deliberately perturbed to improve the accuracy. By obtaining correction signals from one or more base-stations at known locations, positioning over a large area, such as a city or even continent, to an accuracy down to as little as 1 m is possible, with accuracy relative to a local datum of as little as 1 cm. Measurements may be taken from moving vehicles, and the equipment can be used to navigate to a series of waypoints or along a predetermined path. Such a system, called Differential GPS (DGPS) can be operated entirely by the user, either in real time or by post-processed corrections. Alternatively, correction signals are provided by commercial organization for a fee. The corrections can be broadcast over a local radio network, multiplexed with other signals or transmitted over satellite links. The overall result is a portable system that can be carried by a person or fitted to a vehicle and can provide accurate locations. GPS or DGPS requires, however, a clear view of the sky and cannot be used inside buildings or under dense tree cover, and may suffer from inaccuracies caused by reflections when used close to buildings. Positional data obtained from these measurements will be repeatable to an absolute frame of reference and so are of special value where major site engineering operations which would otherwise destroy reference objects are likely to take place.

Microwave ranging systems

Various techniques are available for providing relative positions over distances of tens of kilometers using microwaves. By measuring the time delays for a transmitted signal to be returned from two or more transponders, locations accurate to a few meters can be obtained. Such techniques have been used, in the past, for aerial radiometric surveys. In many cases, DGPS would now be the preferred technique because it is absolute and does not require the accurate placing of transponders and a clear signal path to them.

Ultrasound ranging systems

For relatively small sites, such as a disused factory, inside or outside, and where good spatial resolution is required, positioning systems based on ultrasound time-of-flight measurements are available. Such systems can provide locations to better than 1 m over distances

of the order of 100 m. It is necessary to place and accurately locate several ultrasound transducers around the area to be surveyed.

Advanced surveying techniques: laser ranging

Modern surveying equipment includes fully automatic total stations which use a laser device to measure the range and angle from a base station to a prism located at a mobile survey point. The accuracy of this equipment is typically in the mm range over distances of up to several km. The laser ranging equipment will track the prism and so is of use in moving vehicles provided that a line of sight between base and prism survey point can be maintained. The equipment needs two or more reference objects to be available to establish the position of the station but otherwise can give results comparable to DGPS. Single-handed operation is often feasible and the equipment could be used for surveying large indoor areas as well as outdoor areas in the vicinity of buildings where DGPS may be unusable.

5.2.1.2. Gamma radiation measurement in the field

Field radiation measurements may include dose rate measurements using hand-held radiometers, gross measurements of beta or gamma activity or energy specific measurements of gamma radiation. The latter is of use if identification of the isotopes present is a requirement. It may help if measurements of a specific isotope are required against a background, possibly varying with time or position, of another radionuclide (such as naturally occurring radium or ^{40}K). Although simple instruments may be valuable in locating or delineating areas of high activity, at levels near to the natural background they must be used with care if statistical counting effects or local variations in background are not to be misinterpreted as variation in contaminant. It may be the case that integration times of simple instruments have to be set sufficiently long that spectrometric methods could give a more accurate result in less time. It must be emphasized that background radiation levels can vary rapidly, not only spatially but also with time due to changes in solar radiation or due to radon releases from the ground changing with atmospheric pressure.

Screening surveys are designed to delineate the areas of major concern. In situations involving widespread contamination with sufficient gamma radiation emissions, aerial surveys can be a cost effective method for rapidly delineating and quantifying such areas. Helicopters are used for low-level work where maximum sensitivity is required. Positioning is generally accomplished with commercial navigation systems (e.g. GPS) which feed indicators to guide the pilot accurately along preselected routes. Gamma radiation, flight path, altitude and meteorological data are fed into an inboard data acquisition system for real time or post-flight analysis. Gamma radiation data including spectral data overlaid on aerial photographs indicate the location of the contamination very accurately. For smaller scale work where aerial surveys are not practical, surveying data can be acquired with vehicle mounted detectors, manually pushed carts, backpack carried detectors, and manually held detectors. Vehicle mounted systems can be used for more precise delineation of large areas, provided that access is feasible. Since vehicle-borne systems can easily monitor at one spot for as long as required, traverse slowly and get close to the contamination, such systems generally provide good detection sensitivities and resolution of changing contamination.

In addition to air and vehicle borne gamma survey systems, systems can be mounted on a portable cart or placed in a backpack. This approach provides a low-cost means of gathering large amounts of data. If limited laboratory data are gathered to correlate gross gamma radiation data with concentrations of the contaminant, total count gamma radiation data may be sufficient for

characterization. If it is possible to analyze data in real time, an adaptive approach may be feasible so that the most appropriate areas are covered in most detail. As discussed in Section 4, this approach will require careful pre-planning of the objectives of the survey and adequate training and instruction of the staff carrying out the survey.

As an example, if there is a known area of contamination on a site, staff may be instructed not to survey in detail the area of most concentrated contamination, but to explore the outer limits of contamination so as to be able to delineate the extent of the contaminated area.

Figure 2 shows a typical in situ gamma spectrometry measurement with the detector placed at 1 meter above the soil surface. At this height, 85–90% of the gamma radiation detected is originating from a circle with radius of 10 meter from the detector. The typical in situ gamma spectrometer will, at a height of 1 meter, effectively detect radionuclides to a depth of up to 15–30 cm. The effective area observed by this detector (> ~300 m^2) may, in fact, give a more representative picture of contamination than conventional sampling and analysis.

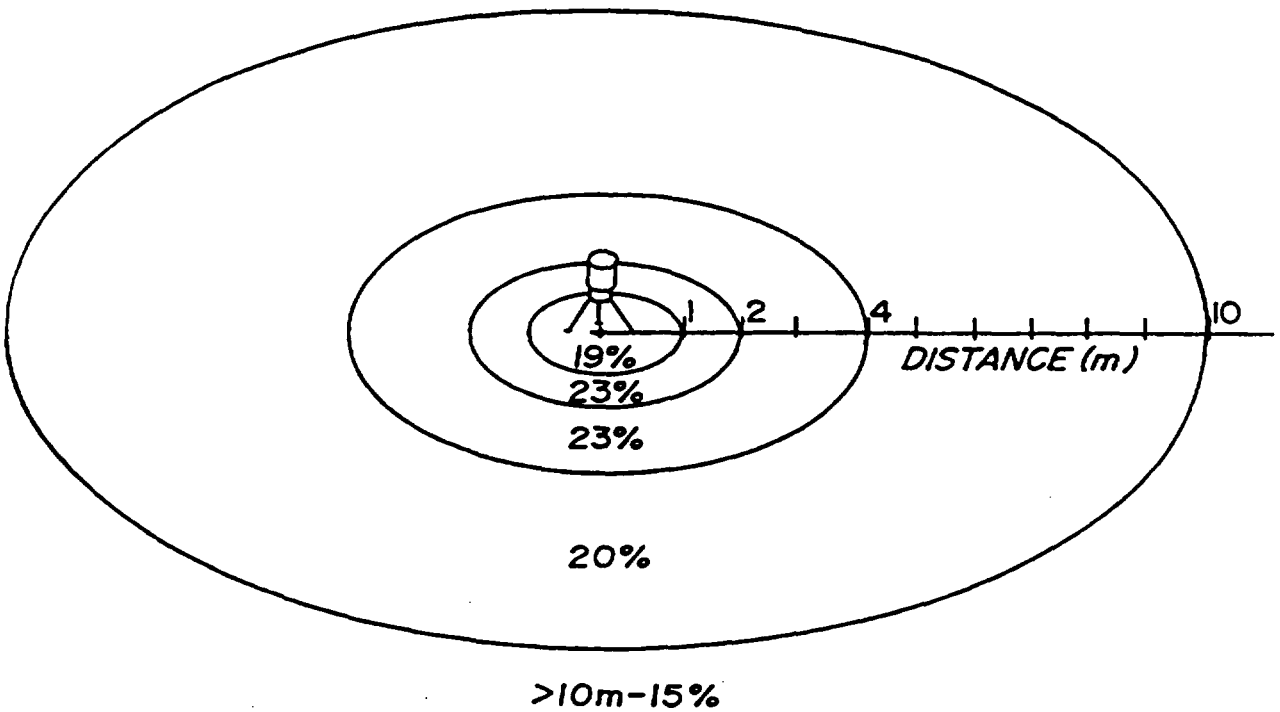

FIG. 2. Area observed by an in situ gamma ray spectrometer at 1 m above the ground. Depicted is the percent contribution to total 662 keV primary flux for a typical Cs-137 source distribution from past atmospheric weapons testing fallout.

As mentioned previously, in situ measurements usually cannot fully replace sample collection and analysis, but it may be used in conjunction with sampling and to reduce costs associated with characterization surveys. Figure 3 shows an estimated correlation between the various site characterization methodologies [28]. As can be seen, there is a significant unit cost difference in price between in situ spectrometry and laboratory soil sample analysis. This difference is mainly due to the additional time and effort required for soil sample processing and measurement. There is some variability in this cost estimate, which is dependent on the radionuclide measured and the local factors such as cost of labour and analysis.

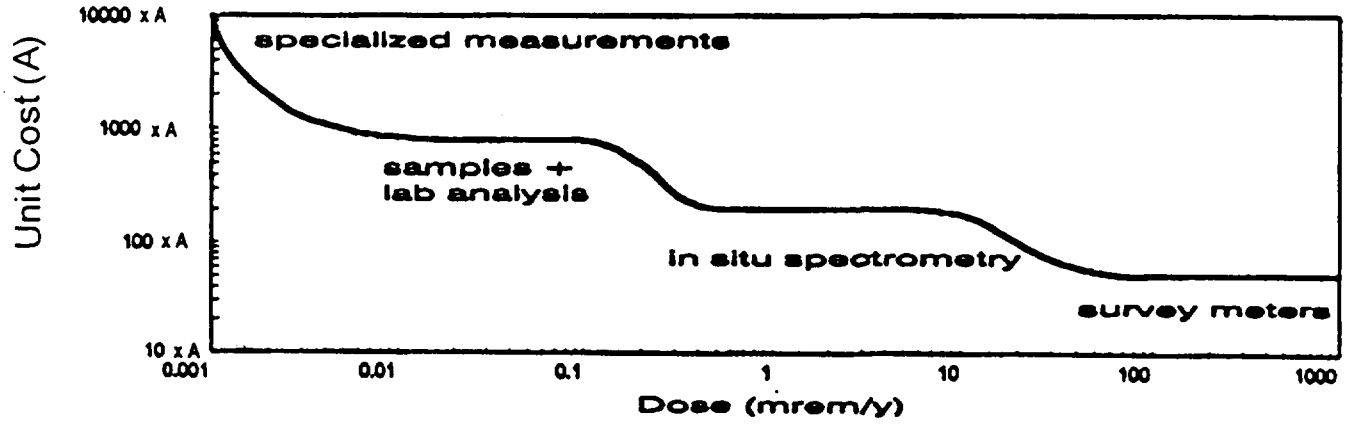

FIG. 3. General pattern of measurement methods and their costs as a function of level of radiation/radioactivity being measured. Unit cost A is equivalent to US $1.

5.2.1.3. Considerations for in situ spectrometry measurements

It should be mentioned first that there are limitations in using in situ spectrometry. Due to the nature of radionuclide transport through matter (soil and air) and to the attenuation of ionizing radiation, in situ spectrometry is, for the most part, limited to the measurement of gamma rays and some X ray emitters.

Regardless of which detector type is used, appropriate methods have been developed to obtain radionuclide soil concentrations levels using in situ gamma ray spectrometry [29, 30]. The ideal site for collecting a gamma spectrum is a large (20 m diameter or more) flat, open area with little or no natural or man-made obstruction. For standard measurements, the height of the detector above the ground is an important parameter. One meter is often chosen for reasons of convenience; the higher the detector the greater the area which contributes to the measurement (and the faster an area may be surveyed, although this is at the expense of lateral resolution). An example of an appropriate survey location is depicted below in Fig. 4. This photograph was obtained near ground zero at the Semipalatinsk nuclear test site during a site characterization investigation in July 1994 [31]. As shown in Fig. 4, a HPGe detector was used to obtain the gamma ray spectrum (the detector was mounted on the tripod which is furthest to the left of the survey personnel), and a Small Pressurized Ion Chamber for Environmental Radiation (SPICER) (which was mounted on the tripod just to the left of the survey personnel) was used to determine the total in-air dose rate from both cosmic and terrestrial gamma ray radiation [32].

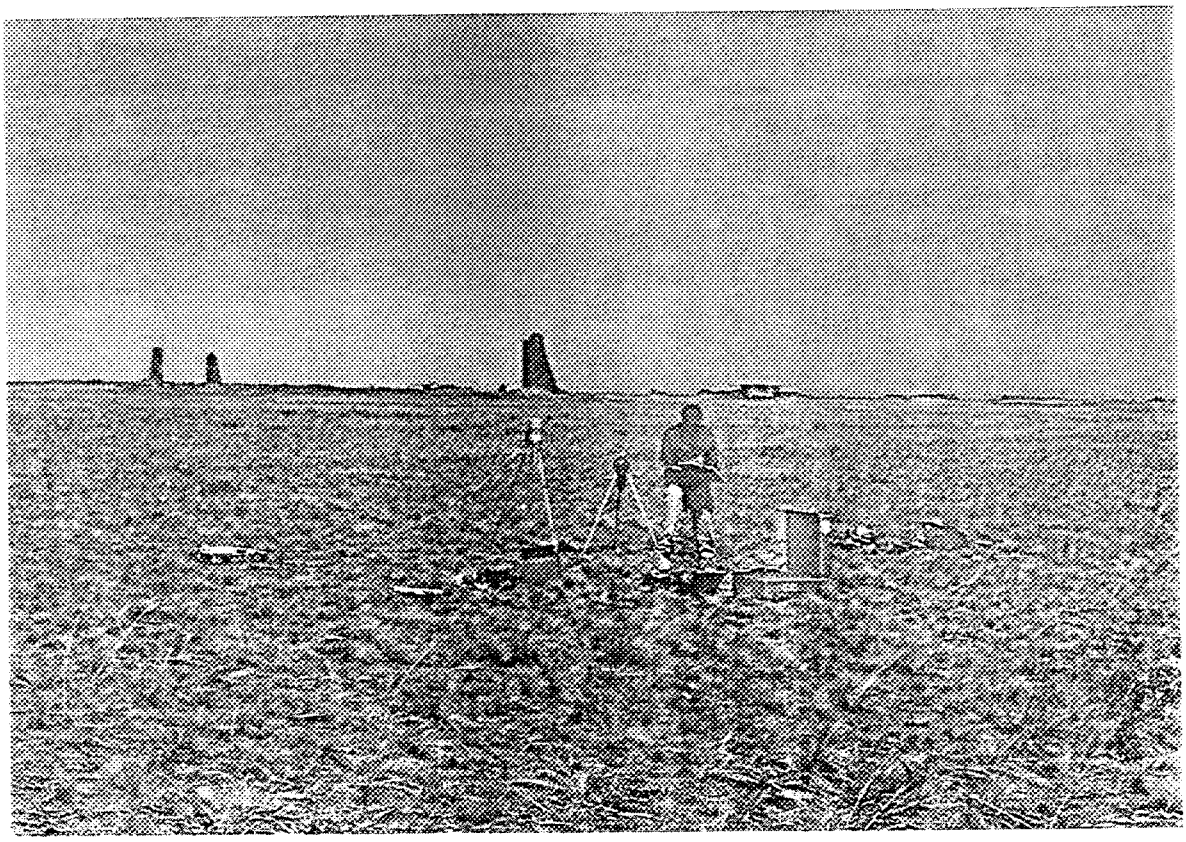

FIG. 4. Photograph of in situ characterization survey near ground zero at the former Soviet Union's Semipalatinsk Nuclear Test Site.

For undisturbed soils, the actual depth profile of the radionuclide of interest is highly dependent on whether it is present as a naturally occurring gamma ray emitter or it was released into the environment from anthropogenic sources and, if so, the time of the release, the mobility of the radionuclide in that specific environment, and the position of release (deposited on the surface, released from a buried pipe, etc.). Usually, naturally occurring emitters (e.g. ^{40}K, ^{238}U, ^{232}Th) are distributed approximately uniformly throughout the soil. Those that are present as the result of nuclear weapons testing fallout (e.g. ^{137}Cs) tend to be distributed with the activity decreasing exponentially with depth [33]. In the case of a very recent accidental airborne releases, the radionuclide probably would be distributed only on the soil surface.

In some cases, in situ spectrometry has been used to determine soil depth profiles directly using differential attenuation for those nuclides which emit two (or more) gamma rays, analysis of the scattered component of the radiation, or (with a lead shield) measurements of the angular incidence of the radiation [34, 35]. Demonstration of this technique was conducted at a former U.S. weapons production facility using a p-type germanium closed-end coaxial detector to determine the surface soil concentration of uranium [36]. The depth profile of ^{238}U was obtained

to a depth of 5–10 cm by observing the attenuation of 63 keV line with respect to the 93 keV line. Multi-line isotopes such as ^{134}Cs can be used to determine an approximate depth profile by virtue of the differential attenuation of gamma rays of different energies. This approach is limited by the small number of radioisotopes having the suitable spectra and which also are commonly found on contaminated sites.

5.2.1.4. Alpha and beta measurements in the field

Due to their properties, in situ detection of alpha and beta emitters is very difficult and often only qualitative.

In fact, very few portable detection systems exist for either of these type of emitters and they are often extremely radionuclide specific. One device that has been used extensively to assess plutonium concentration levels is the field investigation device using low energy radiation (FIDLER) detector [37]. In normal usage, the FIDLER does not detect plutonium, but rather the 60 keV X ray that is emitted from the decay of ^{241}Am, the progeny of ^{241}Pu, using a thin, large area, scintillation detector.

A system that has been recently developed and marketed enables the in situ detection of the beta ray emitter ^{90}Sr via its higher-energy beta ray emitting progeny ^{90}Y [39]. The detector consists of a series of stacked plastic scintillation fibres and has a minimal detectable activity (MDA) of 0.19 Bq/g (5 pCi/g). It also allows for the measurement of total gamma ray flux.

Portable, sealed large-area proportional counters also have been developed to assess transuranic elements via x ray emission [39]. Other systems currently under development include a optical technique for the gross detection of uranium by laser-induced fluorescence (LIF) [18] and a portable in situ Long-Range Alpha Detector (LRAD) for gross surface analysis of alpha particle emitters [40]. The LRAD system is based on detection of secondary ionization of air by the decay of alpha emitters.

5.2.1.5. Application of passive detectors

External dose can be assessed by using passive devices, which integrate the gamma radiation over the measurement period, such as thermoluminescence dosimeters (TLDs), the film badge and track etch. A device that possesses both active and passive qualities is the electronic dosimeter.

A typical TLD can detect radiation level approximately at 0.005 μGy/h but, at least one month exposure time is necessary. If all infrastructure for performing TLD measurements exist, it is an inexpensive methodology since the detector itself is of low cost (and re-useable). Its disadvantage is the long period of exposure required and special care is needed so as not to lose the TLD during the exposure time in an unguarded area. Film badges have similar properties but are more sensitive to the environment in which they are deployed (temperature, humidity) and require greater care and a larger facility for preparation and analysis.

For radon measurements, an in situ technique which has been widely used is exposure of a passive plastic detector to gas. After exposure for an extended period (usually weeks or months), the plastic is etched to reveal the tracks of alpha particles from the radon and daughters. Interpretation of the results into radon concentration and hence potential dose is not straightforward. These devices have been commonly used in some countries for measurement of

radon in domestic premises. They have the advantage of being cheap to manufacture and process, but there is a long interval between placing of the devices and being able to analyze them. They are thus not suitable for situations where rapid results are required or where conditions are changing on a short time scale.

5.2.2. Sampling procedures

Consideration must be given to the different sampling or measurement requirements of the different media present on a site. Radioactive contamination may be restricted to the soil layer; it may also behave differently in the vadose and saturated layers. Rock or clay layers may be impenetrable to activity or may bind activity. Where rocky inclusions are found in soil, they may be essentially free of activity. As an example, this may cause apparent discrepancies if soil sample measurements are compared with external gamma ray measurements on ground containing substantial amounts of rock. In this case, a soil sample would be unrepresentative of the average near-surface material.

If surface water is present it may contain activity; if activity is likely to have reached the saturated zone, it may also be relevant to sample groundwater. Sediments in lakes, rivers, estuaries and coastal sites may concentrate or fix activity.

Measurements in "secondary" media, i.e. other than those containing the main contaminant, may give useful information on the distribution or presence of activity. Biota which concentrate activity may help in detecting the presence of radionuclides which are otherwise buried or at low concentrations. Emanation of gases (radon, tritium) from buried sources can help determine the presence of activity. Most of these measurements should be regarded as qualitative rather than quantitative, but they prove valuable in early stages of characterization to identify areas for further investigation.

5.2.2.1. Sampling of gaseous products

Elements and compounds to be considered include radon, tritium, (possibly as water vapour), ^{14}C and other volatile compounds associated with radioactive materials. It has been shown that gas monitoring can be used to locate waste in the ground. Radon itself is a component that needs to be considered because it is of health concern in case it is present in building materials or in populated areas. The presence of above background concentrations of radon in air directly indicates that there is a source nearby of radium or its parent isotopes.

Methods commonly used for the sampling and analysis of radon include tracketch devices (mentioned in an earlier section), and air sampling through filter papers or charcoal packs followed by gross beta or gamma counting to detect radon and its radioactive decay products.

5.2.2.2. Sampling of flora and fauna

Some species of flora and fauna have the ability to concentrate naturally occurring or artificial radionuclides. Iodine, for example, is known to concentrate in certain algae and shellfish, while caesium can exhibit an enhanced uptake in plants like lichens, heather, fir and spruce, as well as mushrooms. It should be noted that in general, radionuclides have stable sister isotopes which are common in nature and are taken up to varying degrees by biota. Natural processes of plant or animal uptake have evolved which ought not to be affected by the nuclear

properties of the element. This results in a broad and mainly still uninvestigated field of promising use as bioindicators and, moreover, for bioremediation.

Some bioindicators have been identified, as shown in Table III, which does not claim to be exhaustive.

TABLE III. EXAMPLE BIOINDICATORS FOR SOME KEY RADIOELEMENTS

BIOINDICATOR	RADIOELEMENT
Algae, shellfish, peat deposits	Iodine
Heather	Caesium
Snail shell, fish bone	Strontium
Mushrooms, fir, spruce	Caesium
Mycorhiza plants	Caesium
Thyme	Caesium in Mediterranean regions
Lichen	Caesium in boreal ecosystems
Honey	Caesium
Milk	Caesium, strontium and iodine
Seaweed	Ruthenium, technicium
Sheep droppings	Caesium

5.2.2.3. Sampling of soil and subsurface access technologies

Where measurements of the activity distribution with depth are required, it will be necessary to provide subsurface access for instruments or sampling. At the simplest, this could involve digging a hole to carefully remove samples at different depths. More sophisticated approaches would use auger, penetrometer, or borehole technology to provide access to deeper strata.

Soil sampling, both above and below ground level, can provide essential information towards determining the accumulated amounts of contaminants which have been deposited on the ground. It is very important to ensure that the samples taken are seen to provide a realistic representation of both the perceived problem and the area (laterally and/or vertically) over which the contamination is anticipated to exist. For example, the depth at which a radionuclide has penetrated down into the soil is dependant on the age of the release and the mobility of the radionuclide in that particular environment.

Practical methodologies for soil sampling and criteria for methodology selection to assess radioactivity contamination have been compiled [41, 42]. Several of the more commonly used

sampling methods like coring, trenching, and core penetrometer testing (CPT) technology are described below.

Coring

While investigating contaminated areas one of the main objectives will be to ensure the acquisition of an undisturbed sample, preferably with a 100% recovery rate. When samples may be taken using coring equipment, caution must be taken that cross contamination of samples below more active strata does not take place. This can occur if activity is carried on the coring bit or if cutting fluids are used during the operation. The influence of cross-contamination on individual samples can be reduced if the outer layer of the core sample is carefully removed before analysis takes place.

Once a core has been recovered it is important to carefully cut open the liner and expose the undisturbed core on a work bench. This should then be photographed, logged and sampled at a constant frequency (0.5 m may suffice in short length cores, although it may be appropriate to analyze at closer intervals if, say, the contamination is believed to have leached downwards from the surface and is concentrated near to the top layer of soil) and, in addition, at any particular features of interest. It is often advisable to confirm the size of the required sample with the laboratory and ensure that a duplicate sample is taken.

Trenching

Trial pits and trenches are often used as a relatively cheap yet quick method of viewing and sampling the subsurface strata. Stratigraphic and structural changes can be seen more clearly than in cored material and samples are easy to obtain. The approximate maximum depth of 4 m is one of the disadvantages of trenching. Sample points at one-half meter intervals are normally sufficient for contaminant analysis, and once the sample has been obtained the procedures prior to laboratory analysis are similar to that for cores. When done with care, trenching can be used to obtain subsurface samples free of cross-contamination, but it is labour intensive and may be unacceptable for environmental or safety reasons. Trenching may generate unacceptable quantities of waste and may expose workers to both physical hazards from unstable ground formations as well as high levels of radiation from the exposed surface.

Cone Penetrometer or Direct-Push Technology

Cone penetrometer testing (CPT) or, more generally, direct-push technology provides an opportunity for subsurface measurement without coring or boring. It depends on hydraulically pushing a small-diameter instrumented probe from the ground surface downward. Depending on the soil conditions and size of the pushing device, the depth of penetration can reach tens of meters.

CPT probes include a variety of sensors to identify different contaminants. They are often used to screen contaminated areas for later placement of monitoring wells. Sensors for radioactivity are presently under development and in testing.

The primary advantages of direct push technology over boring are small disturbances, relatively rapid sampling, low cost, and no creation of waste. The limitations are requirements for site access for the truck-mounted device, resistance of some lithologies to penetration, and semi-quantitative nature of the measurements from present sensors.

5.2.2.4. *Sampling of water and sediments*

Water and sediments sampling is often a necessary prerequisite in the determination of radionuclide migration. Contaminants could be transported by water in solution on suspended particulates and sorbed into sediments. Lateral movement could be highlighted by analysing surface water and bottom sediment samples. Downward radionuclide migration through the vertical soil profile can be determined in core samples and groundwater samples. Sampling approaches are outlined, for example, in Ref. [42].

5.2.3. Laboratory measurements

Certain radionuclides cannot be effectively measured directly in the field, and in addition, samples are often required to confirm depth profiles. For such situations, samples must be collected and analyzed back at the laboratory. Typical environmental samples include soil (both surface samples and core samples), sediment, water, vegetation (and other biota) and air filter samples. The collected samples are sent to a laboratory and processed prior to analysis. The extent of processing is dependent on the radionuclides of interest and its concentration level in the sample. Some simple processing may be feasible in the field, and there have been a number of instances where mobile laboratories have been set up on site so as to reduce the delay between sample acquisition and results being known. This enhances the feasibility of an adaptive approach being taken to the characterization study.

Laboratory measurements may have the advantage of being conducted in a controlled environment with a full range of support services, but may it take longer before results are available due to the need to transport samples from site and to collate field and laboratory results in one place.

Increasingly, the difference between field and laboratory measurements is being eroded as equipment which formerly could only be used in a laboratory becomes available in more compact, transportable or rugged forms. Some organizations have set up mobile laboratories using a lorry trailer or transport container. These can be quite sophisticated, or may be areas where preliminary sample preparation and screening takes place.

A whole range of configurations exists. A simple solution with the benefit of high mobility could include a van with sampling equipment, limited sample preparation facility and a gamma-spectrometry system. A very sophisticated "transportable" solution could include a series of interconnected laboratory-containers, fully equipped for sampling, complete sample preparation, all appropriate measuring equipment comparable to the ones used in fixed laboratories, meteorologic instrumentation, a ventilation system with high efficiency filters, air conditioning and a decontamination facility. Anything in between these two options is also possible. The choice will depend on the characteristics of the site and on the type and size of the contamination, on the duration of the project, on available resources (staff and money), on the possibility to replace some sampling and laboratory measurements by in situ measurements and on the possibility of access to fixed laboratories.

The location of the mobile laboratory may be such that the contamination risk is minimized (upwind, not too close to removable contamination). If there is a risk of contamination of the laboratory, all normal procedures in radiation control are of application at the entrance. People and materials have to be measured on external contamination, a physical barrier (bench) will be

installed to put on overshoes, material for decontamination of people and surfaces will be available.

Direct communication between the mobile laboratory and the headquarters of the characterization/remediation project is essential. This can be done by radio, cellular telephone, or standard telephone and fax if a connection is available. Data communications (e.g. by Internet) can aid in the rapid collation of results from different locations.

Mobile laboratories exist in many countries in the context of nuclear emergency preparedness. These can be very useful in a characterization project, but they may need some adaptation for the specific problems of the site under investigation (special sampling and measuring equipment).

As an example, a typical mobile laboratory includes the following equipment:

- Electricity supply: generator 5 kVA and external connection possibility
- Water supply: container 100 L, collector for 250 L of waste water
- Refrigerator (60 L)
- Telescopic meteotower (6 m): wind speed and direction, temperature and precipitation sensors
- Fixed environmental gamma dose rate meter
- Portable alpha, beta, gamma and X ray measuring instruments
- Portable telescopic gamma detector for high dose rates
- Electronic personal dosimeters
- Thermoluminescent personal dosimeters
- High resolution gamma spectrometer
- Sampling equipment (air sampling, plastic containers, spade, scissors to cut grass, etc.)
- Protective clothing (masks)
- Decontamination box (soap, shampoo, complexant solution, nailbrush, scissors, safety razor, etc.)
- Personal computer (reporting, recording and transmission of data).

5.2.3.1. Direct radiation measurement

Many of the methods and instruments used in the laboratory are similar to those used in the field: dose meters or other detection instruments may be used, but the more controlled environment of the laboratory and the ability to precisely characterize background radiation may improve the accuracy of results.

Gamma spectrometers will usually be screened against background radiation to improve the minimum detectable activity of measurements. (The level of radon daughters inside a shield can be reduced further by allowing the liquid nitrogen Dewar usually used to cool high resolution gamma spectrometers to vent into the shield thereby purging room air and replacing it with clean gas.)

Techniques not readily usable in the field include measurement of alpha and beta radiation (usually involving some chemical preparation) by alpha spectrometry or liquid scintillation counting (LSC) [43].

5.2.3.2. *Radiochemical analysis*

When samples contain radioisotopes other than pure gamma emitters, some radiochemical preparation will usually be required [44–46]. This may be a simple distillation to separate tritium or a long and complex assay to separate actinides or strontium. For environmental samples, even if gamma emitters are being quantified, there may be advantages in terms of improved sensitivity and reduction of counting time if sample preparation can concentrate the radioactivity. The ashing of biological samples, for instance, will reduce the volume enabling a more sensitive counting geometry to be used. For complex preparations, the addition of yield tracers may be useful to quantify how much nuclide has been lost in the procedure.

Radiochemistry will generally be useful when it is necessary to separate a "hard to measure" radionuclide for analysis by alpha spectrometry or LSC, when an isotope is present at such low levels that it needs to be concentrated before counting is possible, or when it is necessary to separate an interfering radionuclide or one which overwhelms assay of another radionuclide of interest.

"Fingerprinting" techniques are often useful in reducing the number of samples which require complex chemical separations. "Fingerprinting" involves using measurements of easy to measure radionuclides (usually gamma emitters) to quantify harder to measure nuclides. This could involve measurements of ^{241}Am to determine actinide inventories, or use of measurements of ^{137}Cs to indicate ^{90}Sr levels.

Americum-241 is formed as a decay product of ^{241}Pu. Plutonium-241 itself (and other plutonium isotopes) is difficult to measure, but ^{241}Am is a low energy (59.5 keV) gamma emitter. Americium and plutonium (as well as other actinides) often behave in a similar manner in the environment (perhaps as insoluble particulates). Thus, if the ratio of ^{241}Am to other actinides can be established, a measurement of americium will serve to quantify the other actinides.

At least one measurement of the hard-to-measure radionuclide will usually be required, and it may be necessary to invest considerably more effort in order to establish the validity of the fingerprint.

Fingerprints may be used in several ways. First, a firm correlation based on readily justifiable scientific grounds may be established between the easy to measure radionuclide and the hard to measure isotopes (such as may be the case in the relationship between ^{241}Am and ^{241}Pu, or between ^{137}Cs and ^{135}Cs). Second, an empirical correlation may be made between two isotopes which may not be necessarily expected to behave in an identical manner: for instance, it may be established that ^{90}Sr levels can be linked to ^{137}Cs levels. Third, it may be possible to establish a bounding relationship between two isotopes. As a hypothetical example, if a correlation between ^{90}Sr and ^{137}Cs can be found at the surface of a site, and it can be shown that ^{137}Cs is less mobile than ^{90}Sr, measurements of ^{137}Cs in core samples could give an indication of the amount of ^{90}Sr in the subsoil.

5.2.3.3. *Autoradiography*

Autoradiography can provide information about the nature of the contamination, e.g. whether it is homogenous or particulate, or both. Moreover, the combination of different techniques has the potential for identifying the particulate contamination as being that of uranium, plutonium, or fission products. If the detected particles are sufficiently large in size (>20 μ),

further investigations can be made to elucidate the physical/chemical and isotopic characteristics of the particles.

Autoradiography is based on the simple blackening of a sensitive film. After the exposure to radiation, the film is developed and shows a blackening where it has been exposed. Commercially available X ray film (e.g. Kodak AG7) is suitable for the detection of fission products in samples obtained from different soil layers, ashed biota, air filters, etc. The limit of detection for a point source for film exposed overnight is typically equal to about 0.1 Bq of ^{137}Cs.

5.2.3.4. Activation analysis

Another method which can provide valuable information on samples taken from a contaminated site is based on activation by neutrons. The nuclear track detection techniques for detection of U and Pu particles are Neutron Induced Fission Track Analysis and Alpha Track Analysis. Both methods are commonly used and are well documented in the literature.

In Neutron Induced Fission Track Analysis, a subsample of soil, ashed biota or air filter is brought onto a sticky plastic layer spread until all the material has covered the sticking layer. Then the sample is covered with mica film an tightly attached to the Fission Track Detector. The detector is a polycarbonate foil. In this technique, the sample is irradiated with neutrons. A typical integrated flux of 10^9 n/s/cm^2 may be reached during the neutron irradiation; fissile isotopes, such as ^{235}U, ^{238}Pu, ^{239}Pu, ^{240}Pu, etc., will absorb neutrons and thereby generate two fission-product nuclides with a shared kinetic energy of ca. 100 MeV. In the case where the fission product atom penetrates the detector, it will cause radiation damage along its path. In order to make the track visible, the detector is etched (this results in an increased diameter of the track hole. In the detector materials that are used mainly for Neutron Induced Fission Track Analysis, alpha particles will not create tracks. After the detector has been developed, the distribution of fissile isotopes can be studied with a light microscope. The presence of particulate fissile-produced isotopes is observed in a cluster of tracks. With standardization of the method, an estimation can be made of the particle size and the mass of fissile isotopes which are present in the particle. For the instrumented readout of the detectors, automated microscopic systems with an image analyzer are being used.

For the detection of alpha-emitting particles, the use of a material for registering the alpha radiation is required. Such materials are commercially available and the processing of the detectors is comparable to the above described procedure for fission track analysis. With alpha track detection, plutonium is much more effectively detected (on a mass basis) than uranium because of the large differences in specific activities between them. Comparing the number of alpha tracks and fission tracks for a single particle can give an initial indication of the elemental composition of the particle.

5.2.3.5. Other measurements techniques

In cases where sites are contaminated with high concentrations of radioactive materials, gross alpha, beta and gamma measurements are able to produce a rough picture about the total inventory of contaminants and the extent of the contamination. If more detailed, qualitative and/or quantitative information is required, radiochemical methods will need to be applied. Due to the nature of radiochemical techniques, they allow only one or a small group of isotopes to be determined simultaneously. Depending on the method and detection system used, radiochemical methods may provide limited information about the elemental composition, especially in case of

uranium and the actinides. For radionuclides with half lives longer than about 200 years, nonradiometric techniques can be more sensitive than their radiometric counterparts.

Nonradiometric techniques to be considered are:

- Glow discharge mass spectrometry [47] is a technique that does not require dissolution of the sample and can measure in a described configuration a considerable number of elements in the periodic system with one measurement. The sensitivity reaches the parts per billion (ppb) level.

- Real time aerosol mass spectrometry [48] performs qualitative elemental and isotopic characterization from aerosols in air with sizes below 1 micrometer.

- Inductively coupled plasma mass spectrometry (ICPMS) [49, 50] can be used to detect trace levels of stable or long-lived radionuclides. In this context, it is particularly useful for assay of isotopes such as ^{36}Cl, ^{129}I and actinides.

- High performance ion liquid chromatography on-line with inductively coupled plasma mass spectrometry for qualitative and quantitative characterization of all uranium and actinide isotopes of interest in sample materials. In cases where only qualitative information is required, the uranium, plutonium and americium isotopic compositions can be determined in a single sample run.

- Many other standard chemical techniques may be found useful, particularly those aimed at determination of trace elements.

- X ray fluorescence [51] has been used for determination of many materials including uranium and plutonium, as well as for nonradioactive elements such as lead.

Nonradiometric techniques are not described in detail, but their possible application should be considered when it is necessary to determine a very long-lived radionuclide.

5.2.4. Instrumentation

5.2.4.1. Simple counting instruments

In case of a contamination involving gamma-emitting radionuclides, dose rate or contamination measurements may be suitable as a preliminary characterization or screening survey in the quantification or qualitative evaluation of contaminated areas. Dose rate meters will also, of course, be an adequate means of evaluating the external exposure. However, this technique does not allow identification of the radionuclide composition of the source.

Instruments that directly read out the radiation levels during the measurement are often referred to as active detectors or survey meters. Typical examples of active external dose rate or survey devices include the pressurized ionization chamber, the Geiger-Muller survey meter, portable proportional counters, and portable scintillators.

The active devices mentioned above can detect radiation levels down to about 1 uR/h (0.01 uGy/h) depending on measurement time or device time constant used. The sensitivity of each type of survey instrument (counts per second per Bq/kg) may differ considerably depending on the

detector size, the density of the detecting medium (gas, scintillator, etc.), and the nature of the radiation emitted by the radioisotope.

An important parameter to be considered in the choice of a detector is the time constant, i.e. the measurement time of the detector. The sensitivity of the detector should be matched to the radiation level and the required duration of measurement. When a short measurement time is the preferred option (e.g. in the monitoring of a remediation process), more heavy but sensitive equipment, such as scintillation detectors or large-area plastic scintillators that can be used in a gross counting mode, may be chosen.

For aerial or mobile surveys, the detector time constant is an important parameter that determines resolution of incremental changes in the radioactive contamination in a given land area. Typical time constants as used for survey measurements are 1, 10 and 100 seconds.

5.2.4.2. Gamma spectrometers

Gamma ray spectrometry is usually carried out with either scintillation detectors (usually thallium-activated sodium iodide crystals [NaI(Tl)] coupled to a photomultiplier tube] or high resolution germanium detectors. The NaI(Tl) detector has the advantage of being low priced, available in large sizes (giving good detector efficiencies), and operating at room temperature. Its main disadvantages are that it is low resolution (typically 80 keV FWHM at 1 MeV) and the photomultiplier is sensitive to temperature change, as well as to stray magnetic fields.

Measurements of various gamma ray emitting radionuclides can be accomplished using portable scintillation (e.g. NaI(Tl), ZnS(Ag)) or solid-state (e.g. HPGe) spectrometers. The selection of one type of detector over another generally depends on the spectral resolution required, the expertise and experience of the personnel conducting the survey, and the availability of resources for equipment purchase and maintenance. For instance, NaI(Tl) equipment costs approximately half as much as a HPGe solid state instrument. In addition, they are generally more robust than HPGe detectors and do not require cooling. However, the peak resolution is significantly inferior compared to these solid state detectors, and they are prone to energy drift or uncertainties due to temperature changes, instability in the high voltage supply, and the influence of external magnetic fields.

High purity germanium semiconductor detectors (HPGe) offer the advantage of having a very high resolution (less than 2 keV FWHM at 1 MeV) and being essentially unaffected by changes in ambient temperature or magnetic field. Their disadvantages are a relatively high price, a lower maximum efficiency than NaI(Tl) detectors, and the need to be maintained at a low temperature. Usually, HPGe detectors are cooled using liquid nitrogen supplies, but it is possible to used an electrically powered cryogenic cooler in place of the usual Dewar.

Both types of detector are somewhat fragile. More rugged scintillation spectrometers have been constructed using photodiodes as the light-sensing device, but these are currently restricted to the smaller sizes of detector. CdTe semiconductor spectrometers are available in small sizes, but they have significantly poorer resolution than HPGe detectors and are only available in small sizes. However, they offer the advantage over a HPGe detector of having a greater stopping power for equal-sized crystals and of operating at room temperature.

The assessment of measured spectra entails determining calibration factors for each observed total-absorption peak of interest. A proper calibration is used to convert the total

absorption peak count rate (counts per second, cps) into the corresponding activity per unit area or unit mass (Bq/m^2 or Bq/kg). The calibration for a certain radionuclide is, of course, site specific, and it will depend strongly on such local factors as the presence of other radionuclides and the radionuclide depth profile. Details concerning calibration can be found in the literature [52]. An example of calibrating the response of a NaI(Tl) detector with the geometry for the detection of both Cs-137 and Co-60 radiation is shown in Fig. 5.

The lower resolution detectors have the disadvantage of being less able to distinguish gamma energies which are closely separated (which may be important if it is necessary to separate a wanted spectral line from the background). The poorer resolution may also result in the time taken to reach the limit of quantification for an isotope at near background level being greater than for a high resolution detector of lower nominal efficiency. (HPGe detector efficiency is often specified relative to the arbitrary standard of a 2.76 cm by 2.76 cm NaI(Tl) detector at 1332 keV. HPGe detectors are available in relative efficiencies (RE) exceeding 100% on this scale, but at the time of writing the price increases rapidly above about 40% RE.)

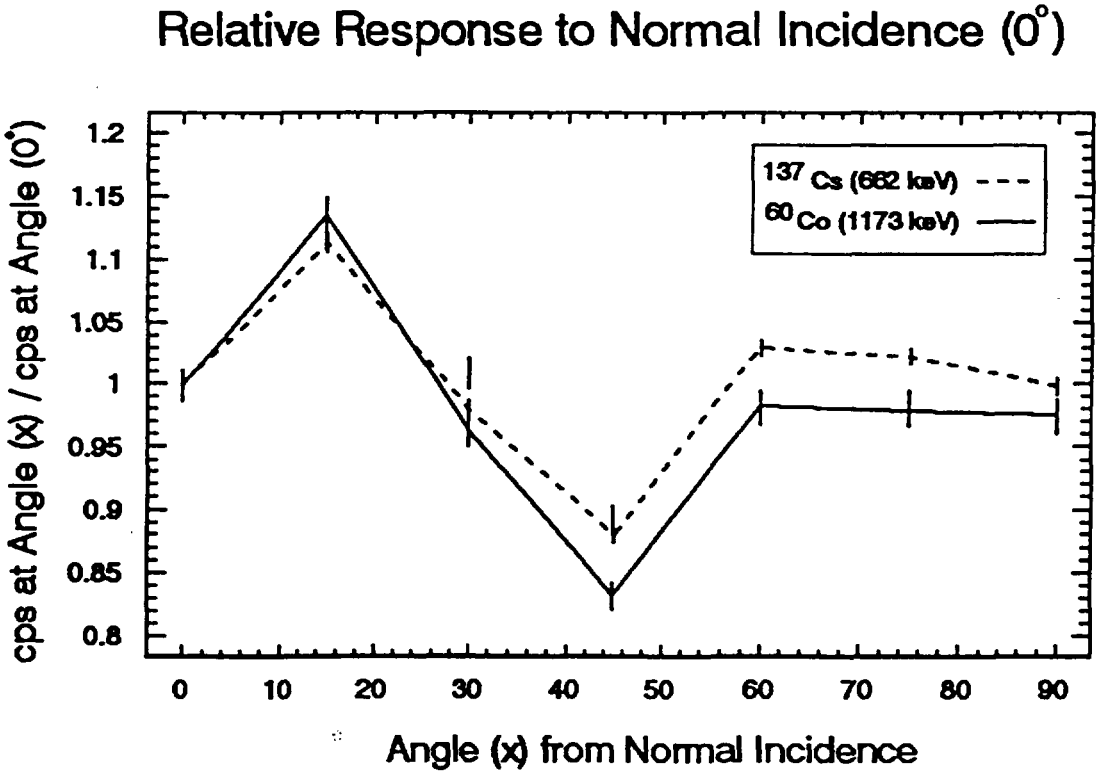

FIG. 5. Curves depicting the relative count rates for Cs-137 (Ba-137m) 662 keV and Co-60 1173 keV at angle, x, away from the detector face and at a fixed distance of 110 cm for a 5 by 5 cm NaI(Tl) detector.

Plastic scintillators have also been used for counting applications, either coupled to a photodiode or to a photomultiplier tube. These detectors have the advantage of being relatively inexpensive and rugged, and they are available in almost any (arbitrary) size and shape. However, their energy resolution is very poor, and they should really be considered as contamination meters, rather than as spectrometers. Thin, large-area, plastic scintillators have been effectively used in the field screening of samples where the detector-to-sample geometry can be optimized to give rapid results.

Both the NaI(Tl) and HPGe detectors must be connected to pulse-characteristic processing electronics and data handling equipment. Nowadays, this will frequently comprise either discrete nuclear interface modules (NIM) or a dedicated computer module, with a computer being used for data storage and analysis. For details of using a gamma ray spectrometer, the reader is referred to one of the standard texts on the subject, such as Refs [29, 53–55].

It is necessary to calibrate the detector for its efficiency for the range of energies over which it is required to carry out measurements, as well as for the geometries which are to be used.

In those cases where in situ gamma spectrometry is contemplated as a measurement tool, it will be necessary to decide whether or not collimation is to be used. The relative advantages of using an unshielded (uncollimated) or a collimated detector are summarized in Table IV.

TABLE IV. ADVANTAGES OF USING COLLIMATION WITH GAMMA SPECTROMETRY

Uncollimated detector	Advantages	Lighter, more portable, measurements on a flat site will collect information from a larger area
	Disadvantages	Interpretation of data is more complex. Background radiation from nearby radioactive facilities may give interferences. The distribution of activity may be non-uniform — this may lead to a single hot spot in view of the detector, dominating the gamma ray flux and yet not being properly quantified.
Collimated detector	Advantages	The area under examination by the detector can be well-defined. The interpretation of the data acquired may be simplified. The calibration procedure for the detector may be simpler -- it may be possible to approximate the gamma ray flux seen by the detector as being unidirectional or even approximating that from a point source. Any calculations required to account for shielding and source distributions (e.g. depth distributions in soil) may be more amenable to mathematical manipulation.
	Disadvantages	Heavier. Less portable. May need to account for shadowing effects in directions only partly screened from the detector by the collimator. Care needs to be taken that the degree of shielding from hot spots outside the field of view of the collimator is evaluated and sufficient. Care needs to be taken that the detector can be inserted into the collimator in a reproducible manner.

Whether or not a collimator is used, it is necessary to make an assumption about the distribution of the radioactivity. Often, the assumption will be that the activity is uniformly distributed across a surface (or is homogenous on the scale of length "seen" by the detector), with a known depth profile (usually determined separately by coring). The source distribution is often approximated as being distributed as an exponentially decreasing level of activity with depth, as a surface source, or as a uniform distribution. The assumption of a uniform distribution may be particularly useful if an approximate estimation of the specific activity (Bq/g or Bq/m^3) of an extended source is required. Care must be taken if the assumption is of a surface source, since the theoretical gamma flux from an infinite planar source would be limited only by the shielding afforded by passage through air. In practice, mixing of the source with the surface of the site or small undulations in the surface will greatly reduce the flux measured by the detector in comparison with a theoretical estimate.

A typical collimator used for these purposes would be a hollow lead cylinder of wall thickness 5–10 cm, which fits over the detector end-cap, extending forwards sufficiently to define the required angle of view and backwards sufficiently to shield the rear of the active detector volume.

In some circumstances, it might be useful to measure the radiation reaching the detector in situ by other than directly through the collimator aperture in order to evaluate how much of the detected radiation may be coming from the ground beneath the detector. This can be done by acquiring a spectrum with a plugged collimator (e.g. with a substantial lead plug inserted into the front of the collimator).

5.2.4.3. Alpha spectrometers

Alpha spectrometry is usually performed in the laboratory using a semiconductor charged-particle detector which is situated in a vacuum chamber. The inherent resolution of the alpha detector is likely to be of the order of 50 keV, but this is frequently limited in practice by energy loss in the source. Alpha particles have very high linear energy transfer (LET) values and so will lose a large fraction of their energy in traversing a few μ of sample matrix or a few mm of air. Sample preparation is critical for alpha spectrometry [56], and it often involves an elemental separation followed by electroplating onto a sample carrier.

5.2.4.4. Beta spectrometers

Beta spectrometry is usually carried out in the laboratory using a liquid scintillation counter (LSC) [57]. This method involves the dissolution or dispersal of a prepared aliquot of sample in a liquid scintillation "cocktail" which is then counted on a dedicated photomultiplier, usually incorporating a system for counting a series of samples on a carousel or similar device. The poor resolution of this system, coupled with the continuum nature of beta energies emitted by radionuclides, means that it is common to separate each element radiochemically before attempting to count it. LSC devices can also be used for alpha counting even in the presence of beta emitters, since the alpha emitter will be emitting particles at discrete energies in the 6–7 MeV range, well above the highest energy of the beta emitters in the sample.

5.3. ERRORS AND UNCERTAINTIES IN RESULTS

A detailed consideration of the treatment of errors and uncertainties is beyond the scope of this publication . However, when radiometric measurements are made, it is always important to maintain an awareness of uncertainties in the data and to take appropriate precautions so that the data which are obtained are adequate for their intended purpose. In this regard, pre-measurement consideration of the data quality objectives may be especially important.

5.3.1. Counting statistics

When counting samples, three limits of uncertainty are commonly quoted [58]. There is much confusion as to the meaning of these limits. The *limit of confidence* (also known as limit of decision) is defined as the amount of a radionuclide that would be need to be detected by a measurement in order to be confident that the identification is genuine. The *limit of detection* is the amount of radionuclide that one can be confident would be detected by a measurement. The *limit of quantification* (also known as limit of determination, and often referred to as minimum detectable activity (MDA)) is the amount of radionuclide that will have to be present in order to be confident that a measurement is adequate. Whenever quoting results and uncertainties of counting measurements on low-activity samples, it is important to assure that one has specified and adhered to a consistent standard of reporting.

Before making a radiometric determination, it will be necessary to decide what sensitivity (limit of detection or limit of quantification) is required and to design the measurement such that this can be achieved. Failure to do so may result in having to repeat the measurement or in drawing an unwarranted conclusion that a particular isotope is not present.

If a given radioisotope is present in sufficient quantity, it may be possible to terminate the measurement early once the results have reached the desired statistical accuracy. An adaptive approach here can save much effort and time. Care should be taken that an overly conservative measurement (i.e. with an overly low level of uncertainty) is not required. In many cases, the overall uncertainty in a radiation measurement result will be dominated by factors other than counting statistics (in particular, there is the large variability which is inherent in sampling).

5.3.2. Other sources of uncertainty

Counting errors are often not the limiting factor in the repeatability or accuracy of results. Whenever samples are taken from a heterogenous medium such as soil, there will usually be a large sample to sample variation. In general, the larger the sample size taken, the more statistically valid will be the result. Where gamma spectrometry is being undertaken, the use of a Marinelli beaker which surrounds the sensitive volume of the detector will give an optimum geometry in terms of sensitivity and in terms of maximizing the sample size. If this approach is taken, care should be taken that: 1) true coincidence summing does not adversely affect the results at a significant level and 2) the range of gamma rays in the sample medium is not much less than the thickness of the sample (otherwise, the detector will be sensitive to a much smaller volume of sample than might have been believed). The latter effect will be compensated adequately if the calibration standard used is similar in density to the sample density.

5.4. ADDITIONAL ASPECTS TO BE CONSIDERED

There are a number of aspects which impact on site operations during characterization or remediation which may need to be considered. Most of these fall outside the scope of this publication but are worthy of mention.

5.4.1. Hazardous materials

When measuring samples there should be an awareness that there may be non-radiological contaminants present. Samples gathered for radiological purposes may also be suitable for assay of non-radioactive hazardous materials, in which case there may be special requirements for sample handling and storage. Conversely, samples gathered for non-radiological purposes may be suitable for radiometric investigations; even if the sample is required to be maintained intact, a non-destructive gamma spectrometry measurement may be possible.

Hazardous materials may also impact on how samples gathered for radiometric purposes are handled. There may be health and safety implications, if the material is hazardous, and care may be necessary to ensure that radiochemical assays are not affected..

There may also be waste and regulatory considerations if hazardous materials are present. Mixed hazardous wastes may be considerably harder or more expensive to dispose of than wastes containing only radiological or common chemical contaminants.

5.4.2. Safety of workers

Although beyond the scope of this publication, there should be an awareness of health and safety considerations for workers during characterization (or remediation) operations. Issues to be addressed involve safety of workers, control of contamination and control of radiation exposure. A balance should be reached between the risk of injury to workers during site engineering operations and the overall risk of the hazard being treated.

5.4.3. Waste characterization and assay

Where waste is being generated, there is likely to be a requirement to characterize it and assay it. It is likely that measurements made for site characterization purposes will be relevant to waste characterization, if only in determining the isotopes that are present. During the site characterization process, it may be worth considering whether a small extension to the programme can prevent duplication of effort in the waste characterization programme. What is necessary for waste characterization and assay will depend on the regulatory regime and further considerations in this publication are inappropriate.

5.5. SUMMARY OF METHODOLOGIES AND TECHNIQUES FOR CHARACTERIZATION

The foregoing subsections have mentioned a number of techniques which may be of use during a site characterization. The methods and instruments actually deployed will depend not only on the nature of the site, but also on other constraints of availability of trained staff and budgetary restrictions. If time or effort restrictions are not a problem, then the most appropriate methods may be to use relatively simple instrumentation or to carry out a large number of sample

analyses. On the other hand, in situations where trained staff are expensive to deploy, a more automated solution may be more appropriate if it enables the job to be completed more economically.

Many field monitoring tasks can be carried out using simple hand-held instruments. The difficulty with use of these instruments is often that a long integration time is necessary in order to establish an accurate reading when measurements are required at environmental levels. It is often quicker to utilize an energy specific instrument such as a high resolution spectrometer. However, if time and effort are available, acceptable results may be achievable using a simple dose rate meter. Determination of the position of the measurement may equally well be achieved by use of an expensive GPS receiver, enabling the spectrometer operator to operate single-handedly in the field, or a more labour intensive traditional survey method may be used to give equally valid results.

The methods used in characterization of sites contaminated by radioactive materials are rapidly developing. The future is likely to see the development of more rugged spectrometers with higher efficiencies, and the availability of integrated counting and positioning equipment. There is much scope for the improvement of room temperature semiconductor detectors.

Techniques for invasive sampling and probing are likely to improve with the development of guidable drilling and coring equipment. There is much further work to be done on non-invasive geophysical techniques for the visualization of below ground structures.

In the field of environmental chemistry, there is still much to learn about the binding and migrational behaviour of radionuclides which will help greatly in assessment of the consequences of soil, rock, and groundwater contamination. The use of bioindicators is still at a very early stage, and there is also much potential for development of bioremediation technologies.

6. SUMMARY AND CONCLUSIONS

Sites contaminated with radioactive material occur in many places on a scale ranging from the very small (e.g. localized; individual building or structure) to national or multinational proportions. Some of these sites may be straightforward to deal with, but often a complex set of interrelated factors will need to be addressed including technical, economic, social, political, legal, and ethical issues.

Characterization methodologies will be required at more than one stage when a remediation is being undertaken. At each stage of remediation, from pre-remedial evaluation through to post-remedial verification, some characterization work is likely to be required.

If the costs of characterization are to be kept under control, it is strongly recommended that a planned approach be taken with clear objectives in mind, as well as consideration being given to whether the immediate objectives can be tailored to assist with the likely requirements of future stages of the overall project. In this context, costs may be regarded in the broadest sense of including such items as future use of the site plus human and environmental impacts.

It is equally important in controlling the costs of a project that a flexible approach be taken so that the programme of measurements can be modified in response to results as they are obtained.

The methodologies which may be deployed during a characterization study may utilize not only the obvious radiological methods but also a wide range of technologies from the fields of earth science, biology and chemistry. In many cases, the technologies used are identical to those used in conventional pollution and contaminated site studies, where there is a large body of expertise to be consulted.

The methods adopted during any study will depend not only on the technical nature of the problem but also on the constraints of cost and availability of trained staff. In many cases, as good a result will be possible using relatively simple equipment in an appropriate but labour-intensive manner, as may be obtained through a technologically sophisticated approach which may enable economies in labour costs.

There are many techniques, instruments, and methods being developed which will affect the way in which characterization work is carried out in the future. These trends and developments include:

- Improved methods for measurement of alpha and beta emitters;
- More rapid methods for radiochemical analysis;
- Improved detector technology, including more efficient detectors, more rugged detectors, the development of room temperature semiconductors;
- Use of bioindicators;
- Improved access technology, such as guideable sampling and boring equipment;
- More use of field measurements through miniaturization of equipment formerly considered as laboratory based, providing rapid availability of results enabling flexibility in planning of work.

There is a need for greater awareness, adoption of new or improved techniques and methodologies, and training in characterization issues. Also needed is a more effective transfer of technologies and training to developing countries and a propagation of methods to reduce the costs of characterization.

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Appendix I

SELECTED METHODOLOGIES FOR REDUCING SITE CHARACTERIZATION COSTS

Several methodologies are being employed to reduce the costs of site characterization While differing in details, these methodologies have some common features:

- Decision making processes that affect sampling are determined before going to the field, but actual sampling decisions of "where" and "how many" are made in the field in "real time" by experts on the basis of evolving sampling results (such sampling and analysis plans are known as "dynamic" or "flexible" sampling plans).

- Regulator approval for the "science-based" approach over the traditional step-by-step approach in which regulators approve each phase of sampling before it is undertaken.

- Use of a suite of non-invasive and minimally-invasive technologies and field screening supported, when possible, by high quality on-site sample analysis with smaller amounts of verification sample analysis in off-site laboratories.

- Technology for efficient management, visualization, and interpretation of data to facilitate on-site, "real time" decision making.

Summaries of the individual methodologies are given below.

A. Observational approach

The observational approach draws on tenets of geotechnical engineering in which it is accepted that the subsurface environment can never be reasonably sampled enough to create a conceptual model that contains no uncertainty. Geotechnical engineering deals with this uncertainty by designing subsurface building structures based on the "nominal" conditions and preparing contingency plans to handle the uncertainties should they be encountered in construction.

The application of this approach to remediation of contaminated site stresses accelerating characterization to determine only the nominal conditions needed for design of a specific remediation system and providing remedial contingency designs to be employed should nominal conditions not pertain. Applications of the observational approach have been made to both radiological and non-radiological contamination problems [I.1–I.3].

B. Streamlined approach for environmental restoration (SAFER)

The US Department of Energy created the so-called SAFER approach (streamlined approach for environmental restoration) which combined the bias for implementing remediation with accelerated characterization that relies heavily on the Data Quality Objective (DQO) approach. The results of applications [I.4, I.5] have been faster and less costly characterization (and potentially smaller total remediation costs).

C. Expedited site characterization (ESC)

Expedited site characterization (ESC) stresses taking a multi-disciplinary team of technical experts to the field to minimize the number of phases of characterization. The team members are very well versed in the site history, have an initial conceptual model of the site environment, are equipped with a suite of non-invasive and invasive technologies, and are prepared to carry out a dynamic sampling effort that may be adjusted daily as sampling results become available.

ESC has been particularly effective in accelerating and improving the characterization of the subsurface environment in cases of groundwater contamination. An appropriate combination of geological, geophysical, hydrogeological, and geochemical investigations is bought to bear concurrently as the study identifies and focuses on critical parameters [I.6–I.10].

D. Adaptive sampling and analysis

Adaptive sampling and analysis (or, as sometimes referred to as ASAP, for "adaptive sampling and analysis programme") exploits the opportunity for in- the-field decision making when field analytical and screening instrumentation can provide rapid results regarding contamination levels. The decision-making regarding sample location and number is facilitated by a decision support system [I.11] that uses the results of radiological or chemical analyses and other site information to estimate the extent of contamination. It also calculates the level of uncertainty associated with the estimate of extent. The system provides visualization of the data, contamination extent, and uncertainty. Just as important, it indicates where the next sampling should occur to have the greatest impact on reducing the uncertainty in the estimate of the extent of contamination. The system successively updates the prediction of new sampling locations after each set of new data is gathered and the estimate of contamination is refined.

In several cases of soil contamination, rapid rounds of iterative sampling guided by the adaptive sampling and analysis system have resulted in delineation of contamination with costs as low as 25–40% of the originally predicted sampling and analysis costs for a traditional uniform-grid sampling programme [I.12–I.16].

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Appendix II

CONSIDERATION OF RADIONUCLIDE ENVIRONMENTAL TRANSPORT

For a more detailed understanding of the behaviour of radioactive contamination in the nature, it is necessary to have good knowledge of the various environmental conditions influencing the fate of radionuclides in the biosphere, as well as of processes governing the radionuclide transport in the environment. In addition, information on human population as a potential receptor of radioactive contamination should also be known. This Appendix provides the relevant information in this context.

II.1. ADDITIONAL INVESTIGATIONS TO SUPPORT SITE CHARACTERIZATION

Geomorphology/topography

The geomorphologic investigation is conducted to develop an understanding of surficial features which influence the terrain stability and consequentially the integrity of the contaminated site itself. Namely, a series of slope processes like erosion (including landsliding, colluvial, and proluvial processes) may seriously threaten the contaminated site and promote the spread of contamination from the site into the environment.

Most of the data will be included in the site description. Information on the natural topography and man-made changes can be useful in this context.

Climatology/meteorology

Both the climate at a site and the particular weather conditions at the time of a release of radioactivity can be important determinants for the movement of radioactivity.

Meteorological parameters may determine air concentrations and deposition of airborne contamination on the ground and influence the soil-water balance. Statistical data on the climate will give information on likelihood of flooding, resuspension by wind erosion, risk of fire, and probability of relocation of contamination by melting snow.

In the case of airborne contamination, exact information, or even informed estimates about wind directions and speeds, at various heights at the time can assist greatly in finding the resulting contamination plumes. Depending on the settling time of the material released, the weather patterns on a local, regional or global scale may be important. Precipitation can greatly alter the pattern of deposition of airborne contamination. Other meteorological parameters, such as the presence of temperature inversions or turbulence, can affect the vertical mixing of the radioactive dispersion or cloud.

The longer term climate of a contaminated area will influence the movement of radioactivity into and across the ground. Maximum wind speeds will determine re-suspension of dusts and will therefore affect off-site migration and become a factor in dose assessment. The prevailing wind direction will affect which populations are exposed. The rainfall patterns will affect likely future land use and influence off-site migration. Extreme weather conditions may also affect their choice of characterization techniques that can be used.

The climate can influence the choice of measuring instruments (watertightness requirements; exposure to low or high temperatures; etc.), and the humidity and air pressure may influence some measurements. Parameters generally included are: temperature, precipitation, wind speed and direction, atmospheric stability, humidity conditions, and air (atmospheric) pressure.

Geology/geophysics

The geologic investigation is conducted to develop an understanding of the subsurface environment in which the radionuclides may be present. The geology may strongly control the behaviour of the radionuclides, and hence risk assessment and remediation design. Information generally to be collected during a geologic investigation are sought from the following areas, such as stratigraphy, lithology, mineralogy, geotechnics and geochemistry, and tectonics and seismicity.

Near surface sediments and features can be further characterized through the utilization of intrusive and non intrusive geophysical techniques. The acquisition of geophysical data can help to build up a stratigraphic and structural picture of the underlying strata, and therefore tie in information between known geological control points.

Because geophysical techniques are often able to access difficult terrain and can produce data values relatively quickly, such techniques provide a relatively inexpensive way of acquiring data.

Geophysical surveys need to be very carefully planned, with the correct technique and associated methodology selected for the very specific problems of a given site. It will often be important to combine a number of techniques in order to build up an accurate picture of the underlying problem or feature. Examples of the effectiveness of multiple approaches is demonstrated in Ref. [II.1].

A detailed discussion of the various geophysical techniques is beyond the scope of this publication, but examples of their application and limitation are highlighted in Table II.1.

Hydrogeology

Hydrogeological data are important because they describe conditions above (the vadose zone) and below the water table (the saturated zone). They can also be used to predict future concentrations and movement of the contaminants. Long-term monitoring of the contamination profile and groundwater conditions may be needed for a full understanding of the hydrogeological regime and its likely relevance to, and influence on, any remediation strategy. Parameters which may collected during such an investigation encompass: hydraulic head, flow direction and velocity, recharge/discharge points, hydraulic conductivity, hydrostratigraphy (aquifers/aquitards), and aquifer age and water properties (e.g. pH, conductivity, temperature).

Measurements of these parameters could prove to be expensive tasks. This is because there will often be requirements for involving drilling, placement of piezometers, pumping tests, and tracer tests. However, such studies may be necessary to understand local transport pathways. Long-term monitoring of groundwater flow and contaminant transport and model development

are useful for providing a sound understanding of the groundwater regimes and in the cases of risk assessment would be necessary.

TABLE II.1. SUMMARY OF COMMON GEOPHYSICAL TECHNIQUES

TECHNIQUE	APPLICATION	LIMITATIONS
Seismic	Geological structure, lateral and vertical extend of landfills and trenches	Unconsolidated ground
Resistivity	Contaminant plumes geological features	Bad contact of electrodes
Ground penetrating radar (GPR)	Buried objects, geological structure	Build up areas, microwaves
Electric logging	Sedimentological and stratigraphic boundaries	
Cone penetrometer tests (CPT)	Sedimentological boundaries and contaminants	Will not penetrate coarse sediments
Magnetics	Buried metallic objects, like drums and tanks	Background clutter
Electromagnetics	Buried objects, extent of landfills and trenches	Background clutter

Hydrology

The hydrologic investigation addresses the physical characteristics of surface water bodies that represent potential pathways. Surface water bodies may be natural (i.e. rivers and lakes) or may be man made (i.e. irrigation, dam reservoirs, waste ponds). Parameters and descriptions which may collected or developed during such an investigation include water flow rates, water volumes, circulation patterns (in lake), sediment descriptions, artificial sources, variabilities over time, etc. (e.g. seasonal variations), and flooding history.

It could be beneficial to sample water which is upstream of contaminated areas in order to acquire data about background values. Water samples could be sampled at outset or during a monitoring programme continuously to create time series data. Fine grained sediments situated at the localities of highest depositional rates are generally preferred for sample collection.

Pedology

Pedologic investigation gives information to understand the properties of the soil layer supporting the contaminated site. Any spread of contamination from the site will penetrate it. Pedologic investigation can identify characteristics of soil as natural barrier for radionuclide transport. These include the physical properties (grain size, drainage class, lithological sequence,

permeability, porosity, density, water content); and geochemical properties (leachability, leachate quality, elemental composition of the soil, pH, K_d).

II.2. POPULATION PARAMETERS

Demographic investigation

One of the objectives of the characterization is to give the inputs for dose assessment and to enable choices of remediation for dose reduction to people. The demographic investigation will also be used in the determination of a critical group. In order to pursue these objectives, it is useful to know the geographical distribution of people living in the vicinity of the suspected site now, and perhaps in the future. In addition, it is recommended to collect the following information:

- geographical distribution of settlements;
- population density; and
- habits (e.g. consumption, leisure, spatial mobility, workplace routine).

Land use investigation

The use of the land is of consideration for dose assessment and for cleanup levels. The present land use and decisions on the future destination of the land may influence the degree of detail needed in the characterization.

In the future, the land may be used for agricultural, industrial, residential or recreational purposes. It may be decided that no detailed characterization would be required if the present or planned land use imposes no unacceptable risk for the population. However one should keep in mind that it is very difficult to be sure that the land-use will not change in the future. For example, sediments lying on the riverbanks may not represent an immediate danger, but could be used in the future as fertilizer, spreading the contamination. The economical aspects of the use of the land are an element in the evaluation of remediation strategies.

Water use investigation

The purpose of this investigation is to identify the use of water which may be impacted by the potentially contaminated site. It should be determined whether certain water quality may be impacted by a contaminated site, and therefore, certain institutional constraints should be placed on its use (i.e. use of bottled water for drinking). The inventory should include location of groundwater drinking wells, location of seeps and springs; and nature of water use at present and in the future (i.e. agricultural, industrial, drinking, recreational, medicinal, etc.).

Intrusion hazard

In some cases where the contamination is well-contained and the option of no action is considered, an investigation of the intrusion hazard may be undertaken. This investigation into the future intrusion into the contamination may include description of natural barriers, description of man-made barriers, and possibility to add barriers (e.g. geological stability of the site).

II.3. CHARACTERIZING RADIONUCLIDE TRANSPORT AND MIGRATION

Once the presence of radioactivity and contamination is known, it may be necessary to discover how much of the radioactivity is moving to other sites and how far and how fast it is moving [II.2–II.4]. In many cases, the radionuclide will be attached or strongly associated with some medium, such as a soil, so the migration can only occur either through the bulk movement of this medium or through movement of the radionuclide into another medium, such as groundwater. Sometimes the decay product of the primary radionuclide is itself radioactive and perhaps more mobile. A commonly encountered example is the noble gas radon, ^{222}Rn, coming from the decay of ^{226}Ra. The migration of the radionuclide may occur through movement of the substrate to which the radioactive species is attached or even through movement of the pure radioactive isotope.

In addition to migration of the primary medium containing the contamination, the chemical species containing the radionuclide may be subject to physico-chemical processes that transfer the radioactivity to a more mobile medium, such as water. The overall migration of the radioactivity will then be governed by the process of transfer, perhaps solubility, as well as the movement of the new medium. The disciplines of hydrogeology, geochemistry, etc. can play a large part in characterizing this behaviour.

The role of water in transporting radionuclides from the primary medium at a contaminated site is often extremely important and the movement of this water, on the surface and underground, may need to be studied in detail in order to carry out a dose assessment [5]. This part of the characterization may be most difficult and most important part. Geophysical techniques may be used to allow the underground structures which control the flow of water to be understood.

Transport of radioactivity

There are many ways in which radioactive material can be transported from sites into the broader environment where they may impact on human health either by external exposure, ingestion or inhalation [II.6]. The routes which usually give most concern are transport as airborne particulates (especially in dry climatic conditions) or by water. A number of other transport mechanisms are mentioned in the following subsections.

(a) Transport by airborne particulates

Where the contaminant is present in surface layers of dry soil, the raising of dust, through wind or by human or animal activities, may be an important part of an exposure pathway. The dust may be directly inhaled, or it may redeposit onto plants or other area where human contact is possible. Evaluation of this process will require, in general, knowledge of the meteorological conditions, the soil type, and the types of soil where disturbing activities are likely to occur.

The details as to where the contamination is located within the substrate and how it is distributed can effect its migration, its contribution to an exposure pathway, and its measurement. For example, an alpha-emitter attached to large soil particles may be impossible to inhale, difficult to resuspend, and difficult to measure in situ. Alternatively, the same radionuclide present attached to fine dust particles may be easy to measure and readily inhalable.

The amount of a radionuclide present in the range of dust particles that are respirable can differ markedly from its concentration in the bulk medium. The concentration per unit mass of respirable dust can greatly exceed that in the soil as a whole. Its distribution amongst various sizes of dust particles can effect the resuspension of the radionuclide as well as the rate of resettling.

Resuspension and deposition processes play an important role in the dissemination of the radioactive material from a primary contaminated site. These mechanisms are influenced by the type of the contaminated surface, time period after contamination and local meteorological conditions, among others. Although these processes have shown to be more important as a local process than a long-range process, this will depend on whether the environment is an open field or a complex environment structure such as a forest or an urban area. In addition, depending on the radionuclide and the size of the associated particulate, the inhalation pathway could be an important contributor (even the most important, such as the case of alpha emitters) to the dose. Therefore, air sampling would have to be taken in order to 1) estimate the inhalation dose, and 2) to estimate the potential long distance transport and further deposition onto other surfaces.

Total air sampling can be performed, both at the site and downwind from it, in order to estimate the radioactivity in the air. However, impactor (or similar) measurements may be required for a definitive characterization, since the activity concentration can be measured as a function of the particle size. It is important to characterize inhalable and respirable fraction avoiding unnecessary remedial action. An outline of important considerations follows:

- Setups for air sampling.
- Locations considered for air sampling.
- Air sampling devices.
  - filtering systems
  - impactors
  - cyclones
  - centrifuges
- Ways to analyze the obtained samples.
  - analysis of the entire (total) sample
  - analysis of single particles.

(b) Transport by liquid flow

Hydrogeological data are important because they can provide information about ground water including aquifers and contaminant migration pathways. The sorption/desorption processes, strongly controlled by soil and sediment vadose zone characteristics, could result in contaminant retardation in that unsaturated zone, or alternately downward contaminant migration to the water table.

Water samples are usually taken from boreholes or monitoring wells. Multi-level well completions within the same borehole, or even borehole clusters, can be used to target the different strata within the geological sequence if so required. A discussion of groundwater

sampling is beyond the scope of this report. However, techniques and methodologies can be found in several sources such as Refs [II.7, II.8].

(c) Transport due to extreme events (fire, flood, storm, earthquake, volcanic eruptions)

Extreme geological and meteorological events have the capacity for massive relocation of a contaminant. While the probability of such events is usually small, when multiplied by the potential exposure to population, the effective dose, or potential dose, may require consideration.

(d) Transport by other transfer agents

The activities of humans and animals may, deliberately or unconsciously, assist in the movement of the radionuclide. Contamination may adhere to animals feet, dust may be raised by the passage of animals, or, as a worst-case, contaminated objects may be deliberately collected for various purposes.

(e) Gaseous transport (radon, tritium)

Where ^{226}Ra is the contaminant, inhalation of the highly mobile radon decay product, ^{222}Rn, may be an important exposure pathway. Tritium is rather rapidly incorporated in water molecules and will tend to move with water vapor.

II.4. CHARACTERIZATION OF PHYSICO-CHEMICAL FORM OF ACTIVITY

Various characteristics of the contamination can affect its migration rate and its ability to enter exposure pathways. Its physical and chemical form can have very large effects on the resulting exposure. The dose assessment may require many physical and chemical aspects to be measured.

(a) Food chain (uptake by crops and animals)

The dose assessment process will often require consideration of ingestion of the radionuclide through food intake. The uptake will depend on many factors and may require extensive investigation of the soil type, its nutrient content and the growing conditions of the plant. Direct ingestion of the plant by animals used for meat may also be important.

Biogeography may be an important element of a site characterization methodology when large areas with various soil occupation are contaminated, such as in the case of a nuclear accident. The vegetation cover is a determining element of the various landscapes which can be contaminated, i.e. pastures, forest, cultivated fields. These landscapes will have to be considered in different ways for remediation purpose. For instance, forest which usually can occupy up to 50% of a given contaminated territory and may also have a significant contribution to the risk for critical groups of population must be considered separately. The quantity of produced biomass which may be required to be classed as waste or treated as contaminated material may vary in large proportions as function of tree species, the age of the stand and the soil type. This kind of information can be obtained using the methodologies applied in the field of the biogeography which includes plant associations and plant succession concepts. These concepts will provide

insights on the stability of the ecosystem if it would be subjected to decontamination and on the fluxes of radionuclides within the ecosystem.

(b) Chemistry

Solubility of radioactive materials can affect migration in aqueous media and also alter the resulting exposure. The chemical form of the radionuclide can effect its uptake by plants and its exposure it enters the body.

Various methodologies could be used to characterize the interactions of the chemical species present in the soil solution including very common measurements such as pH, electroconductivity, the concentration of various major elements in the soil such as K, Ca, Mg and specific ions competing with selected radionuclides, such as NH_4^+ which has a high affinity for caesium binding sites on clay. Techniques such as atomic absorption spectrometry, colorimetry may allow one to carry out a relatively large range of determinations. They may, of course, be completed by such more sophisticated techniques as mass spectrometry or neutron activation which are quite expensive and not always widely available.

The competitive ion exchange from the soil matrix to the soil solution is very important parameter for understanding the mobility of radionuclides in the environment. The affinity of various soil types or minerals for a given radionuclide may vary greatly for different natural conditions. The distribution coefficient, K_d, which is the ratio of the activity in the soil to that in the solution which is in contact with the soil under fixed conditions, does not describe the number of chemical interactions driving the measured equilibrium. However, this coefficient has a large application in most of radioecological models and is a very commonly used parameter. Various methods of K_d measurements have been described in the literature [II.9], and new methods are being issued as more research and experience applying sorption and physico-chemical concepts to complex natural systems is accrued. The choice of a method will depend of the natural model which is intended to be examined. Both methods for water-saturated and non-saturated soil conditions are available.

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CASE HISTORIES OF NATIONAL EXPERIENCE

A total of eight annexes are included in this publication to illustrate some of the experiences which have been gained in the characterization of radioactively contaminated sites for remediation purposes. The applicable case histories of national experience are as follows:

ANNEX A. CASE HISTORY — AUSTRALIA

ANNEX B. CASE HISTORY — BELGIUM

ANNEX C. CASE HISTORY — BRAZIL

ANNEX D. CASE HISTORY — CANADA

ANNEX E. CASE HISTORY — CROATIA

ANNEX F. CASE HISTORY — SLOVAKIA

ANNEX G. CASE HISTORY — UNITED KINGDOM

ANNEX H. CASE HISTORY — UNITED STATES OF AMERICA

Annex A

CASE HISTORY — AUSTRALIA

THE FORMER BRITISH ATOMIC WEAPONS TEST SITE AT MARALINGA ON MAINLAND AUSTRALIA

A-1. INTRODUCTION

Between 1952 and 1963, several sites in Australia were used by the United Kingdom to test nuclear weapons [A-1]. At Maralinga, which is located in the remote outback of South Australia, about 200 km inland and roughly halfway across the Australian continent, seven atomic devices were exploded with yields ranging from 1 to 27 kilotons. The resulting radioactivity from those explosions was widely dispersed at the time, and that remaining on site has largely decayed and no longer presents any significant health risk. However, several hundred minor trials, involving the explosive dispersal of uranium, plutonium and other radionuclides, were also conducted at Maralinga and have resulted in contamination of large areas. In spite of a major cleanup exercise by the U.K. in 1967 [A-2], a significant quantity of plutonium remains on the ground today and constitutes a potential hazard to future inhabitants [A-3].

The most highly contaminated area, Taranaki, was the site of the highest yield mainland major trial (27 kiloton), but it was left relatively uncontaminated from that test because of the specialized site-preparation and because the device was exploded at a height of 300 m. Taranaki was therefore able to be used for the Vixen B series of minor trials which were conducted in 1960, 1961 and 1963 involving, in all, about 22 kg of plutonium in 12 single-point safety trials or hydronuclear experiments. These trials involved essentially no fission yield but produced vertical jets of molten or burning plutonium, extending hundreds of metres into the air, which were then dispersed by the wind. The plutonium fell to the ground at varying rates forming plumes of contamination to the west, north-west, north and north-east.

In 1964, a major decontamination exercise was carried out with the intention of leaving the site in a condition where only ongoing care and maintenance would be needed. Following a further cleanup in 1967, the site was formally closed and the whole area, encompassing the major and minor test sites, has been under varying degrees of surveillance and subject to entry restrictions since then. The presence of plutonium, both in burial pits and dispersed over large areas was known at the time and steps were taken in the two cleanups to reduce surface concentrations of plutonium by mixing the soil to depths of several centimetres in extreme cases,, and by covering with clean soil. In 1984, when comprehensive surveys were made in preparation for release of the land to its traditional owners, the true extent and significance of the plutonium contamination from the "minor" trials became apparent. The levels of plutonium in the soil exceeded, by approximately an order of magnitude, the values expected from the British surveys made at the time of the experiments and during the subsequent cleanups. In contrast, the residual fallout and neutron activation radioactivity from the major trials was of little significance.

Following these discoveries, it was necessary to re-evaluate the real risk posed to potential occupants of the area, and a Technical Assessment Group (TAG) was formed by the Australian Government in 1986 to supervise the gathering of, and to report on, the scientific information

required [A-4]. The Australian Radiation Laboratory (ARL) has been conducting field work at Maralinga, together with associated laboratory investigations, since the closing of the range in 1967, and has performed the inhalation hazard assessment [A-3] as part of the TAG programme of studies.

Control of the Maralinga site and responsibility for planning and carrying out its remediation lies with the Commonwealth Department of Primary Industries and Energy. Following the TAG studies and agreements between the UK and Australian governments regarding financial issues, a rehabilitation exercise has been planned and is expected to be completed during the years 1995–1999. The intention is to allow unrestricted access by the Maralinga Tjarutja people to most of the site, but with a restricted area, of several tens of square kilometres, in which camping and full-time occupancy would not be permitted. To accomplish this, the contaminated soil from approximately 2 km^2 will need to be removed and buried in trenches several metres below ground level. The largely unknown amounts of plutonium placed in burial pits during earlier decontamination will be further treated to make it very much more secure than it is today.

A-2. CHARACTERIZATION OF CONTAMINATED AREAS

Location and extent of contamination

The presence of the γ ray emitting ^{241}Am has allowed the surveying of contamination by hand-held scintillation detectors, ground-based high-resolution γ ray spectroscopy and by helicopter-based aerial survey making use of a large array of sodium-iodide detectors [A-4]. The latter provided a comprehensive survey of the area and shows activity levels down to about 1.5 kBq/m^2 of ^{241}Am. However, this type of survey suffers from poor spatial resolution, and, in areas near major trial sites, has difficulty distinguishing ^{241}Am from other radionuclides.

To obtain the fine detail necessary for guiding remediation, ground-based, *in situ* measurements using a high-resolution germanium detector have been made These measurements will be quite accurate in areas where the soil has not been disturbed, but, in areas where the soil has been mixed, the attenuation of the 60 keV γ ray in soil (half-value-layer is ~2 cm) will cause the level of contamination to be underestimated. Nevertheless, the measured value provides a very good indicator of the inhalation dose, because inhaled dust will come primarily from the same surface layer as do the observed γ rays. As mixing to greater depths is encountered, the expected inhalation dose per unit count-rate gradually fall, in spite of the increase in total activity per unit area.

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 rays that escape the surface, together with the enhancement factor and plutonium-to-americium ratio.

A 20% relative efficiency closed-end coaxial detector has been mounted on a boom carried by a four wheel drive light truck. This permits measurements to be made at a nominal height of

about 4 metres over a wide range of terrain. Measurements of 100–200 seconds duration are adequate for the 30–50 kBq/m^2 levels of ^{241}Am which are involved.

The depth profile of the plutonium has been measured by layered soil sampling and laboratory analysis. At Taranaki, in areas where no previous attempt at rehabilitation has occurred, the plutonium lies at the surface of the predominantly sandy soil, with typically 85% of the activity being found in the top 10 mm. It lies along plumes starting near the site of each trial and extending for tens or even hundreds of kilometres in the direction of the wind at the time. The plumes are well-defined but often contain plutonium of different compositions from different trials. Discrete particles containing plutonium have been found along the plumes more than 100 km from the test site.

In other areas, the previous cleanup operation has involved mixing of the surface layers with deeper layers of soil, and the plutonium is now distributed through the top 100–200 mm of soil. In highly contaminated areas, clean soil was introduced as a top cover which further complicates any survey work.

In total, several square kilometres of land are contaminated to levels exceeding 300 kBq/m^2 of ^{239}Pu, and plutonium is readily detectable over more than 100 km^2. Localized areas are 10 or 100 times more contaminated.

Radionuclide composition

The plutonium contamination, as determined largely by gamma ray analysis of soil samples and particles, contains the following isotopes: ^{238}Pu (88 a), ^{239}Pu (24 110 a), ^{240}Pu (6600 a) and ^{241}Pu (14 a). The minor trials involved negligible fission yield, so that the isotopic composition of the source material was unaffected by the explosion, however, radioactive decay has substantially removed the ^{241}Pu and has replaced it with ^{241}Am (433 a). In practice, the 59.5 keV γ ray from ^{241}Am serves as the most useful indicator of plutonium in soil once the ratio of plutonium to americium has been determined experimentally. This ratio and the isotopic composition of the plutonium varies from site to site, and even from one trial to another at the same site [A-5]. At present, the americium content is slowly increasing as the remaining ^{241}Pu decays. The ^{239}Pu typically contributes about 80% to the total γ activity, and ^{241}Am between 4% and 10%. Because the γ emitting nuclides all have similar dose intake conversion factors for inhalation, the exact isotopic composition is relatively unimportant. Other radionuclides were used in various trials but, apart from ^{235}U, these were largely short-lived and have since decayed away.

Physical form

Plutonium contamination is present in three forms:

fragments plutonium contaminated debris C pieces of steel, plastic, wires, lead, etc., visually identifiable when lying on the surface.

particles sub-millimetre pieces of soil and other material incorporating plutonium oxide; indistinguishable from soil on casual inspection.

dust very finely divided, and potentially inhalable, grains of plutonium oxide or contaminated soil.

The fragments and particles have been observed with activities of plutonium ranging from a few kBq to many MBq. In addition to the widely dispersed contamination, there are numerous burial pits containing large pieces of debris and equipment contaminated with largely unknown amounts of plutonium. It is fine dust which constitutes the main hazard through the inhalation pathway. However, some of the particles have been found to be very friable and could produce fine dust if disturbed.

Chemical properties

The particles show very low solubility in simulated lung fluid and quite variable solubility in 0.16M hydrochloric acid. The fact that the plutonium has remained very near the surface for 30 years after deposition highlights its lack of solubility in rainwater [A-6].

Biological properties

Inhalable fractions of dust samples were tested on guinea pigs and rats at the UK National Radiological Protection Board (NRPB) [A-7, A-8]. It was found that test with americium and plutonium gave similar results. Both nuclides had transfer rates lying between Class Y and Class W. An appropriate and conservative classification for plutonium from the minor trial sites was 75% Class Y and 25% Class W. Gut transfer factors and uptakes from contaminated wounds were also measured, and this allowed assessment of the less significant ingestion and wound contamination pathways.

Dust properties

The important distribution properties of dust for dose assessment purposes are the aerodynamic diameter of the inhalable fraction, which determines the behaviour of the dust inside the respiratory tract, and the activity of the various radionuclides within that fraction. Depending on the source of the contamination, its activity (concentration) within the inhalable fraction of the dust may be quite different from the activity (concentration) in the total dust sample and more different, still, from that in a bulk soil sample. Soils from several sites at Taranaki were sieved to determine mass and activity distributions. The 0–45 μm fraction had a much larger proportion of activity than its mass would suggest.

Artificial resuspension experiments were undertaken to determine the activity median aerodynamic diameter (AMAD) as well as the relationship between the radionuclide concentration in inhalable dust and that in the soil from which it came. A cascade impactor was used to separate fractions to allow their mass and activity to be measured and the AMAD to be estimated. Observed values of about 6 μm from artificially raised dust were found, falling to 4.8 μm after several minutes settling time. Consequently, an AMAD of 5 μm was used in the dose assessment.

The enhancement factor, defined as the ratio of activity concentration in the inhalable fraction to that in the bulk parent soil, was found to vary from 3.7 to 32.5 in soils from sites at Taranaki. An average of 6 was used for the outlying "plume" areas and higher values may be

appropriate for the inner, more contaminated, areas [A-3]. Total activities for naturally resuspended dust collected with high- and low-volume air samplers were also measured.

A-3. DOSE AND RISK ASSESSMENT

Annual doses were calculated for different locations from dust loadings, radionuclide content (including enhancement factors), dose intake conversion factors and breathing rates appropriate for the two regimes. Calculations show that doses well in excess of 100 mSv per year are possible if continuous occupancy were to occur in some localized regions. While very few areas represented a significant hazard to the casual visitor, doses in excess of 1 mSv per year would be possible over a very large area; where the ^{241}Am concentration might exceed about 0.6 kBq/m^3. Because of the narrow plume structure of the contamination, 100% occupancy of contaminated areas by the very mobile Aboriginal people is extremely unlikely. It is only in the case of very specific (and probably intentional) behaviours that doses near the threshold for deterministic effects, say 500 mSv could be incurred. However, the presence of plutonium in visually identifiable pieces of debris in MBq quantities means that deliberate collection of such objects is possible, and malicious misuse must be considered as a possibility.

A-4. REMEDIATION AND FUTURE USE

After considering the different options and taking into account the importance to the potential occupants of damage to the environment, the planned intervention is as follows.

It has been decided to remove entirely, the contaminated soil along with contaminated debris. Soil will be removed from areas in which:

- the average level of ^{241}Am over a hectare exceeds 40 kBq/m^2;
- particles and fragments exceeding 100 kBq ^{241}Am are present; or
- particles of 20 kBq ^{241}Am exceed a surface density of 0.1/m^2.

The contaminated soil and debris will be buried in trenches excavated close to the site and covered with a minimum of 5 m of clean fill. By limiting the activity of the remaining soil to below 40 kBq/m^2 of ^{241}Am, and by limiting the occupancy factors to those typical of hunting activities in a particular location, worst-case annual doses of less than 5 mSv could be confidently expected.

Finally, the large or unknown amounts of contaminated debris in pits are to be rendered practically inaccessible by an in situ vitrification treatment.

The cleanup criteria were guided by conservative principles and estimation of doses for realistic scenarios. These included the possibility of an Aboriginal group living for an entire year on the edge of the non-residential area in regions of the highest activity permitted outside it (approximately 3 kBq/m^2 of ^{241}Am). This could lead to an annual dose of 5 mSv. If, more realistically, the group spent its time randomly dispersed over the Maralinga lands outside the restricted area, or even randomly around its perimeter, the average activity levels (and hence the doses) would be confidently expected to fall by at least an order of magnitude.

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

- The production of large amounts of dust in locations of very high plutonium concentration could certainly lead to doses exceeding permitted occupational limits. In these areas, soil will be removed.

- The deliberate collection of contaminated fragments and particle could allow a person to amass considerable, and dangerous, quantities of plutonium. While the likelihood of this leading to significant exposure to many people is small, even the threat of such exposure would have widespread consequences. Areas containing highly active particles or large numbers of particles will be cleaned.

- The potentially large amounts of plutonium in the burial pits will be rendered practically inaccessible both to deliberate seekers and to environmental factors, by in situ vitrification or other treatment.

A-5. LESSONS LEARNED

In the immediate post-firing period, the extent of the plutonium contamination was measured by alpha-particle monitoring a few days after deposition. It was known in the USA, and ought to have been known to those carrying out the survey, that, even on a clean concrete surface, measurements made after only 3 days underestimated the amount of plutonium by a factor of about 10. For measurements over soil, a further factor of 2 or 3 could be expected.

During the various subsequent monitoring exercises, much reliance was placed on soil sampling and laboratory analyses. Unfortunately, while much of the plutonium is dispersed as a fine dust and is readily observed in soil samples, a large proportion occurs in tiny (<1 mm), highly active, particles of recondensed device materials spread over distances of many kilometres. Apart from the occasional "anomalously high" sample, the laboratory analysis of even one kilogram soil samples shows little evidence of this activity. There are, in addition, many visually observable, larger fragments and pieces of debris contaminated with up to grams of plutonium. Survey methods, including soil sampling and grid measurements, made with every care and attention to detail, had little chance of finding these. It was only when hand-held gamma ray detectors, adjusted to detect the 60 keV gamma ray from Am-241 accompanying the decay of plutonium, were used to scan the area continuously that the full extent of the contamination was found. The presence of an audible indicator on the instruments greatly aided the process of discovery.

At various stages, attempts were made to estimate the potential doses arising from the inhalation of plutonium from the site. Unfortunately, simplifying assumptions which were believed to give worst-case results led to an underestimation of the likely dose by a factor of at least 6. It was assumed that the concentration of plutonium in the inhaled dust would be, at worst, the same as that in the bulk soil samples. In fact, experiments by ARL during the TAG studies showed that, depending on the site, the average plutonium concentration in the inhalable fraction

of dust varies from about the same as in the bulk soil to as much as 20 times higher (defined as the *enhancement factor)* [A-3].

Finally, the mixing of soil and the introduction of clean soil, which was performed prior to the closing of the range have made the present cleanup more difficult than it otherwise would have been. In untreated areas, despite the passage of more than 30 years, most of the plutonium and associated americium resides within a few millimetres of the surface. This enables the contamination to be detected and allows it to be removed with a minimum cut (scraping up) of the soil.

An incomplete characterization of the site, the use of inappropriate monitoring techniques, and the military levels of security surrounding this contaminated site, all contributed to the underestimation of the problem.

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Annex B

CASE HISTORY — BELGIUM

RADIUM CONTAMINATION FROM A RADIUM PRODUCTION PLANT IN AN URBAN SETTING IN BELGIUM

B-1. INTRODUCTION

A case history is provided in this Annex of the Olen radium extraction site in Belgium and the contamination due to the radionuclides ^{226}Ra and ^{222}Rn. A factory producing copper and cobalt(not radioactive) is located in Olen. From the beginning of the 1920s and up to the 1960s, this factory also produced radium from the same ores. The company contributed to a large part of the world production of radium. Five dumping grounds in the vicinity of the factory were used for the dumping of radioactive and other waste. Occasionally, some waste material was also used as a layer on a limited number of roads. Liquid effluents were released in a brook (called "Bankloop") since 1922. The brook flows through the village, crosses a canal (Kempisch Kanaal) and flows into the Kleine Nete and finally into the river Nete.

At the end of the 1950s, measurements in the frame of a study to obtain the necessary licences for the start of the Belgian Nuclear Research Center, made it clear that the water and the sediments of the Kleine Nete and of the Bankloop were contaminated with radioisotopes. The banks of the Bankloop brook were also contaminated because the brook was cleaned regularly and the removed sediments were placed on the banks. The Bankloop regularly flooded the land located just before its confluence with the Kleine Nete as a result of heavy rain, contaminating the boggy soil. Because an agricultural organization wanted to make this land available for farming, it had acquired the property and had taken some measures to change the water management of the area. A road (Roerdompstraat) was constructed to gain access to the area. The part of the Bankloop between this road and the Kleine Nete was moved approximately 100 m to the west, leaving the "Old" Bankloop as standing water. A drainage of the land between the road and the Kleine Nete was constructed, reversing the normal east to west flow.

This was the situation in 1960, when a first study on the biological cycle of radium, applied to Olen site, was undertaken from 1961 to 1967, with a follow up through 1977. This study included aerial radiological survey, gamma survey at ground level, sampling of water, fish, vegetables, agricultural products, etc. The results were reported by Kirchman [B-1]. As a result of the study, a number of actions were recommended. Some of these actions were carried out, while others were not. The actions taken included the filling up of "Old" Bankloop and the application of deep ploughing to make pastures for dairy cows.

In 1989 and 1990, the population living near the dumping grounds became anxious as a result of reports in the media of observations of very high (localized) contamination in some parts of the village. The existing old data were mostly for the land near the Kleine Nete, and data were not sufficient in the context of a more stringent radiation protection approach. As a result, the federal ministry of public health and environment (DBIS/SPRI) decided that a more detailed assessment of the scattered contamination should be done by a mobile survey and a survey on foot of the most contaminated parts, including the dumping grounds and the Bankloop. The assessment programme also included an evaluation of the radon exposure in the dwellings of St Jozef Olen, the village near to the factory, and in open air above the dumping grounds, as well

as an evaluation of radium concentration in airborne dust, surface water, ground water, the food chain and in children's teeth.

B-2. RADIOLOGICAL CHARACTERIZATION OF CONTAMINATED AREAS

The programme was executed by SCK-CEN and the Institute for Hygiene and Epidemiology, with a follow up by DBIS/SPRI. The results of the study were published in 1993 [B-2].

B-2.1. Radon in dwellings

To evaluate the presence of radium under houses, radon measurements were made. An iterative approach was used. Measurements were performed in all of the 846 neighboring dwellings with a short term measuring technique: an activated charcoal collector was placed in the cellar or in a unventilated room. The dwellings with radon concentrations above 150 Bq/m^3 were investigated with charcoal collectors in the living areas. Then, alpha track detectors were installed and operated for several months in those which were still above 150 Bq/m^3 . In one of the living areas, the average radon concentration in 6 dwellings was above 150 Bq/m^3. The number of dwellings exceeding the investigation level is in agreement with the radon distribution of the region [B-3]. The dwellings were also inspected with portable gamma detectors.

B-2.2. Mobile survey

All the roads in the neighboring village (Sint-Jozef-Olen) and in some of the other suspected areas were monitored with a van equipped with two 4"x4" NaI detectors for the low dose rates and two 2"x2" NaI detectors for the high dose rates. The exact position of the van was continuously determined with a gyrocompass and sensors on the wheels. The data were automatically stored in a computer system. The results are shown in Fig. B-1.

B-2.3. Survey on foot

Radiation measurements were made for areas above the investigative level of 200 nSv/h for the mobile survey (that is, at 2 to 3 times normal background) with use of four automatic hand-carts which were especially designed for this purpose. Three carts were used to determine the nodal points of a grid. At every 5 m, the dose rate was measured in the middle of the road and at fixed distance both left and right of the middle. The fourth cart and a portable gamma detector were used to locate local maxima. Samples were taken regularly, mostly at a local maximum dose point, to determine the corresponding ^{226}Ra concentration.

Considering surface areas of 5 m by 2 m at each nodal point and taking the isolated points into account resulted in 5800 m^2 of roads having dose rates above 200 nSv/h and of those 1950 m^2 above 400 nSv/h.

B-2.4. D1 dumping ground

The dumping ground D1 has a surface of about 100 000 m^2. Despite the sometimes dense vegetation a grid was established with a nodal point at every 25 m. The dose rates at the nodal points and at some local maxima were measured with portable gamma detectors. For 153 out of

a total 197 nodal points, the dose rates were above 200 nSv/h. The observed maximum value and the mean value are 150 000 nSv/h and 2800 nSv/h, respectively. A local maximum of 10^6 nSv/h was found.

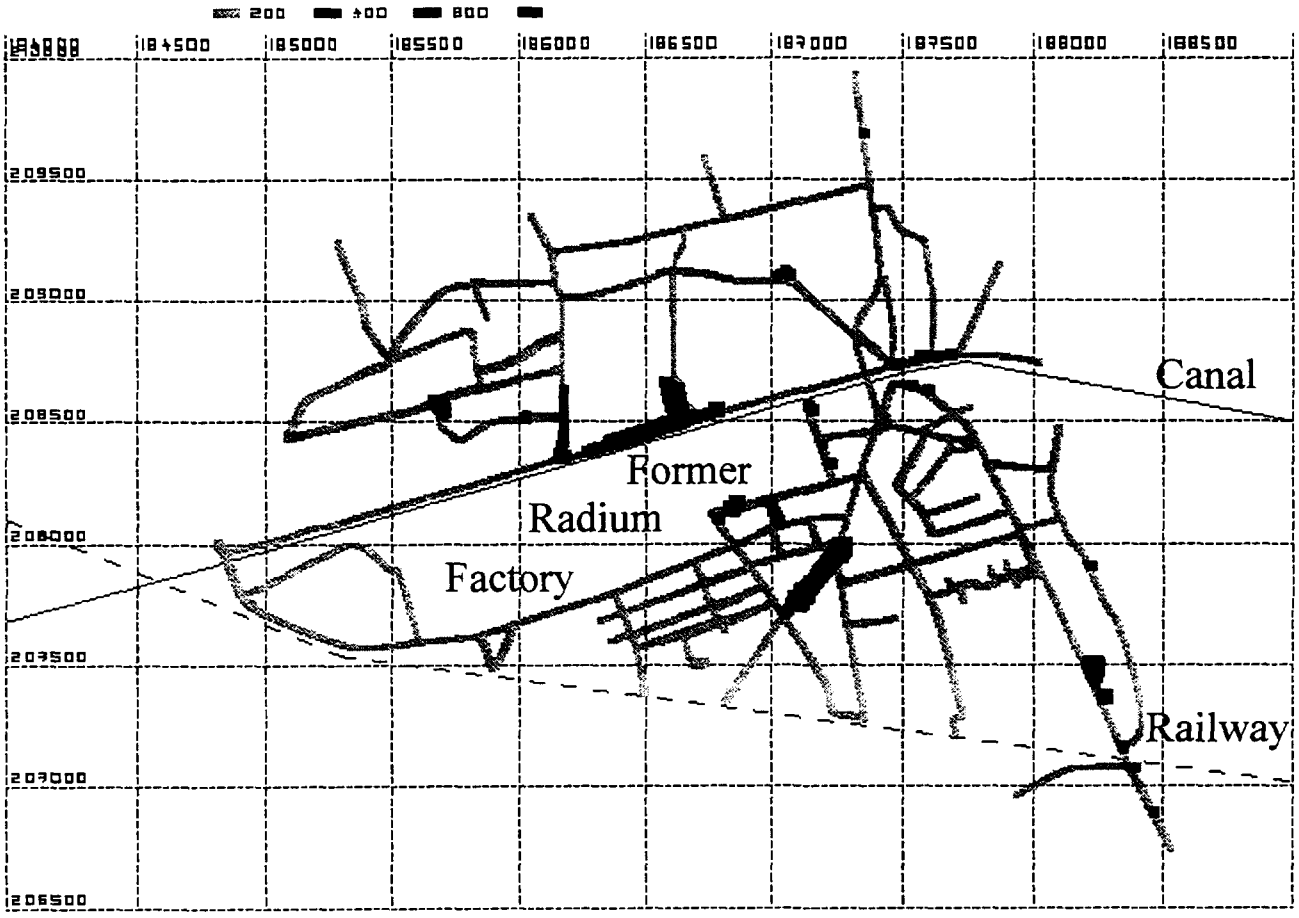

FIG. B-1. Dose rates recorded by mobile survey in the village of Sint-Jozef-Olen.

B-2.5. Bankloop brook

The liquid effluents of the former radium extraction plant were released into the brook Bankloop. The contamination was mapped out from the fence of the plant to the mouth into the river the Kleine Nete. The dose rates were measured along every 10 m in the middle of the Bankloop, at the shoreline and every 2 m along both banks, until the background value was attained. The Bankloop is 1,800 m long from the fence to its mouth. The first 600 m to the canal are a residential area and it then flows through an agricultural area. Thirty years ago, as a result of soil reclamation work, the last 420 m before the mouth was displaced. The pastures on the place of the former bed to the new bed were measured according to a grid with a nodal point every 10 m. The contaminated strip along the Bankloop is limited to about 10 m. It is mainly result of the dredged sediment disposed on the banks. In 47 sections, dose of 2000 nSv/h was measured, and in 17 sections doses of at least 5000 nSv/h were observed. The highest dose rate of a nodal point is 50 000 nSv/h registered at the crossing of the Bankloop with the canal. Nearby a local maximum of 100 000 nSv/h was found. The contaminated surface along the Bankloop is

estimated at 7000 m^2. The maximum value of the ^{226} Ra concentrations in 32 samples, mostly taken at a local maximum was 960 Bq/g

B-2.6. ^{226}Ra in airborne dust, surface water and ground water

The resuspension of radium by dust blown from the uncovered soils was investigated by pumping air through a filter and measuring the collected dust. Two sampling stations were set up. The first was placed at 300 m from the D1 dumping ground in the prevailing wind direction. The second was set up along the Bankloop at the backside of the municipal school. The stations functioned trouble-free during the six month measuring period. The filters were changed every week and measured by means of the Lucas technique [B-2–B-4]. The dose measured ranged between 0.09 and 1.66 μBq/m^3. They are comparable to the UNSCEAR [B-5] reference value of 0.5 μBq/m^3 for normal areas.

The migration of ^{226}Ra was investigated by taking samples of surface water close to the D1 dumping ground and samples of ground water from boreholes on the D1 dumping ground. The ^{226}Ra concentration was measured with the Lucas technique [B-4]. The ^{226}Ra concentration of the surface water ranged between 17 and 56 mBq/L and that of the ground water was between 5 and 18 mBq/L. These values are within the range of values for normal areas [B-6]. In conclusion, the radiation exposure of the population to airborne dust, surface water and ground water appears to be negligible.

B-2.7. ^{226}Ra in the food chain

The ^{226}Ra concentrations of a number of biological samples from contaminated areas were measured with the intention of determining the exposure of the food chain. The samples were analyzed by means of the Lucas technique [B-4]. Among other things 12 milk samples, 5 maize samples, 4 samples of chicken's eggs and 2 grass samples were taken.

The milk samples were taken from two farms with pastures and fields situated partly on grounds with an enhanced radium concentration. These grounds constitute less than 20% of the surface area of the farms. The average ^{226}Ra concentration of the milk, assuming a milk consumption of 120 L per year, corresponds to an intake of only 1.2 Bq. UNSCEAR [B-5] give the acceptable annual intake of ^{226}Ra at 22 Bq. The ^{226}Ra concentration of the milk is in agreement with the ^{226}Ra concentration of the silo maize [B-2] with the food-milk transfer factor of Halbert et al. [B-7].

According to the International Union of Radioecologists (IUR) database [B-8], the soil-to-plant concentration factor for grass on sandy soil is an order of magnitude greater than the corresponding value for corn. Thus, in contaminated areas, corn-feed is a preferable vegetation to grass-feed.

Along the Bankloop at areas with the highest dose rate (100 000 nSv/h), two grass samples were taken with a time difference of 4 months. These had ^{226}Ra concentrations of 103 and 10.6 Bq/g dry weigh, respectively. The radium concentration of the top soil is 230 Bq/g, confirming that the soil-to-plant transfer factor for grass on sandy soil is about 0.13 [B-8].

During the field investigation, no crops for direct human consumption were found on contaminated grounds. It is, however, not improbable that this could change in the future. According to the soil-to-plant transfer factors of the IUR [B-8], a concentration of 10 Bq/g in the root zone results in a concentration in the corn of 0.086 Bq/g (dry weight); in legumes, 0.046 Bq/g; in root crop, 0.051 Bq/g; in potatoes, 0.01 Bq/g; and in vegetables, 0.43 Bq/g. Assuming an annual consumption of corn of 60 kg dry weight results in an exposure of 1,140 μSv, 5 kg legumes, in 51 μSv; 5 kg root crops, in 56 μSv; 20 kg potatoes, in 44 μSv; and 5 kg vegetables, in 470 μSv. The production and consumption of suspect vegetables can be avoided. In modern society it is unlikely that someone lives purely or locally cultivated foodstuffs. Therefore, the dose to the critical groups will probably be below 1000 μSv per year.

B-2.8. Depth distribution in the D1 dumping ground

The D1 dumping ground is a landfill which is suspected to contain most of the contamination. The factory owner, who also owns these grounds, ordered a study to determine the depth distribution of the contamination in the D1 dumping ground for the evaluation of possible remediation scenarios. Drillings were performed to a depth of 3 m (the maximum depth of the original surface). An instrument to log the depth profile of the dose rate was developed. A watertight 2" x 2" NaI detector was used, together with a battery-operated rate meter and data logger (Fig. B-2). The detector was moved down in the bore-hole by means of a battery-operated motor. The depth position of the detector and the dose rate were measured by the data logger. Retrieval of the data was done off-line for further analysis and reporting. The results showed a very heterogeneous distribution in horizontal as well as in vertical directions.

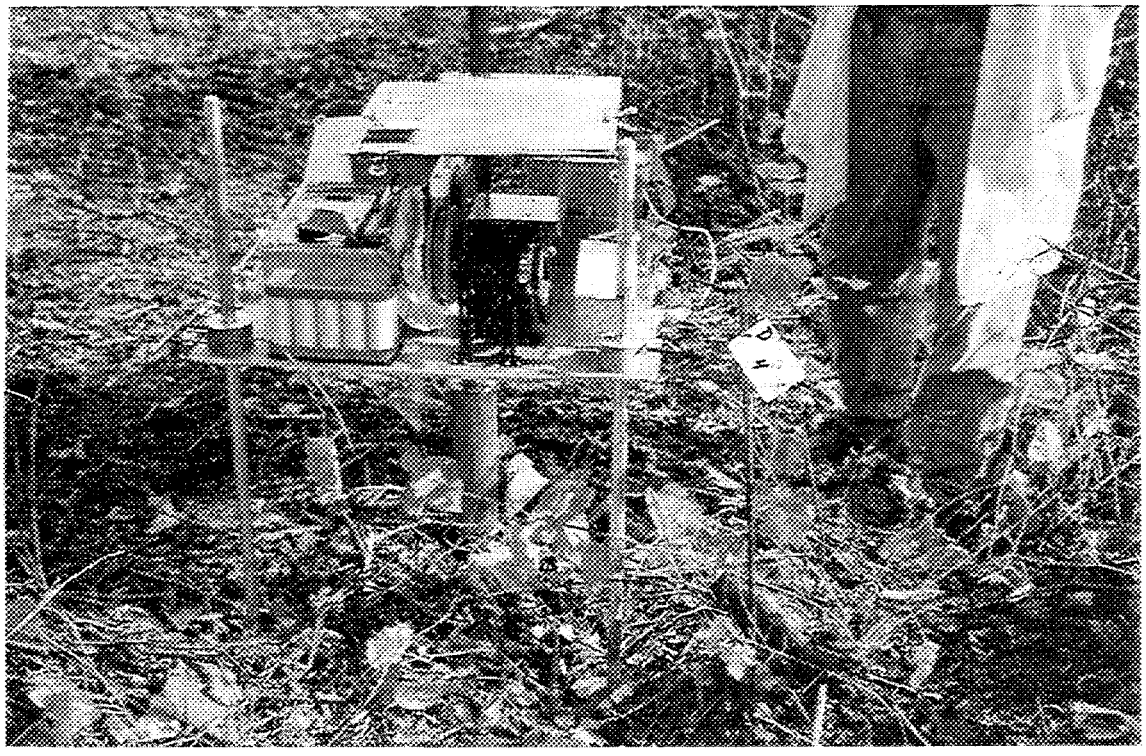

FIG. B-2. The field instrument used to log the depth profile of the dose rate in a borehole.

B-3. EVALUATION AND REMEDIATION

Government, factory, research institute (SCK), local government, NIRAS (federal nuclear waste agency), OVAM (regional non nuclear waste agency) and a number of other authorities are working together to define possible remediation strategies taking into account all relevant aspects (radiological evaluation, chemical and toxicological hazards, cost, public acceptance, public concerns, etc.). Because the largest volume and the highest activities are found on the D1 dumping ground, a "final" solution is being studied for this area by evaluation of a number of remediation and waste handling and storage scenarios. This scenarios include the "do nothing" option, several intermediate options and the "dig it all up and send it to the responsible authorities for handling". In the mean time it was decided to clean up the roads, during normal maintenance works, with health physics supervision, and to store the produced waste temporarily on the D1 dumping ground. Two roads were cleaned in this way down to a maximum dose rate of 200 nSv/h.

B-4. LESSONS LEARNED

- A remediation is linked to the context of its time. The remediation approach which was decided upon in the 1960s, was judged to deal with the situation. However, due to a changed perception of "danger" and to changed criteria, a new remedial effort became necessary in the 1990s.

- Not all of the recommendations of the 1960s were followed, e.g. planting trees on the most contaminated part of the region near the Kleine Nete. This demonstrates that a follow-up and checking of all remediation aspects is necessary.

- It is important to involve all of the relevant parties early in the decision process, in order to have a consensus between the major parties, even if this causes the decision making to be more difficult.

- Even if it can be proved that there is no real problem today, future developments, including human intrusion must be included in the evolution scenarios; responding to public perception may lead to the cleaning up to a lower level than necessary for dose considerations, etc.

- The phased approach to the characterization is generally very well suited to the resolution of contamination problems.

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Annex C

CASE HISTORY — BRAZIL

RADIOACTIVE CONTAMINATION IN BRAZIL: THE INTERLAGOS MILL SITE FOR THE PROCESSING OF MONAZITE [C-1]

C-1. INTRODUCTION

The Interlagos Mill (USIN) site is located on an industrial area near the downtown of the largest Brazilian city, Sao Paulo. Sao Paulo has around ten million inhabitants. The USIN site encompasses 60 000 m^2 in which there are three buildings. One of the three buildings, which was used for the operation of a lanthanides separation plant, was closed in 1991. The other two very rundown buildings were used for storing different monazite processing wastes. There were no records about what was deposited or removed from the site. Through characterization of the site, it was found that there was a radioactively contaminated area of ca. 4700 m^2, with an average depth of contamination of 60 cm and an estimated volume of contamination of 2500 m^3. The contaminated soil contained an average concentration of 4 Bq/g of ^{223}Ra and 3 Bq/g ^{226}Ra. The contaminated water contained an average concentration of 77 Bq/L ^{223}Ra and 18 Bq/L ^{226}Ra.

The characterization of the site began due to a complaint received by the Public Ministry that the site was being utilized as a radioactive deposit by the monazite processing plant [B-2]. Consequently, it was requested that the Brazilian National Commission of Nuclear Energy should survey the area. The Brazilian National Commission of Nuclear Energy was responsible for the characterization and for developing response plans for the environmental restoration.

C-2. STRATEGY UTILIZED FOR THE CHARACTERIZATION

The strategy which was utilized for the characterization of this site was to perform a thorough field and laboratory radioanalysis, as follows:.

Gamma Survey: The gamma radiation levels were measured using a scintillation detector positioned one meter above the ground surface on a 10 meter grid.

Surface and subsurface soil collection: Surface soil samples to a depth of three centimeters were collected at locations where an elevated level of gamma radiation was observed. Based on the surface soil results, points for collecting samples for a soil profile were chosen. These profile samples were collected with a 10 centimeter diameter boring ring to a depth of 4 meters. The samples were taken in 1-meter deep layers, mixed and then analysed (leading to a total of 30 soil profiles to be carried out). Additional 1-meter deep soil profiles were taken where the previous profile had shown ^{223}Ra concentrations higher than 200 Bq/kg on the 1 meter deep layer. They were then cut into 10 centimeter slices to be analysed.

Surface water and shallow qroundwater samplinq: Water samples from the creek located in front of the USIN site, as well as shallow groundwater samples, were collected and analysed. The surface water from a river into which the creek flows was also sampled and analysed.

Leachinq tests: In order to characterize the nature of the contaminants in the soil, including their bioavailability, leaching tests with ammonium acetate were performed.

Laboratory methods were used for determining the radionuclides present in the collected samples, as follows:

Soil: ^{226}Ra and^{228} Ra concentrations were determined by gamma spectrometry, using high resolution intrinsic germanium detectors. ^{238}U concentrations were determined by low-energy gamma spectrometry; and, in some of the samples, ^{232}Th (as thorium) was determined by spectrophotometry with Arsenazo III indicator.

Liquid samples: ^{226}Ra and ^{228}Ra were determined, following co-precipitation with barium sulfate, by alpha and beta counting.

C-3. DOSE AND RISK ASSESSMENT

The methodology for the dose and risk assessment calculations was the one described by Till and Moore [C-3]. The model is based on a scenario/exposure-pathway analysis in compliance with an annual dose limit. An allowable dose limit of 0.3 milliSievert/year was adopted, based on the Brazilian Radiological Protection Guide. Three different scenarios were used, taking into account several types of reutilization or future uses for the site.

Industrial use: For a continued industrial use of the site, the allowable residual levels of Th and ^{226}Ra were calculated to be 1,500 Bq/kg, and the estimated volume of soil to be removed of the site would be ca. of 1,300 m^3.

Intruder family scenario: The most conservative scenario would involve the unrestricted use of the site. For such scenario, the allowable residual levels of Th and ^{226}Ra were calculated to be ca. 200 Bq/kg. The estimated volume of soil to be removed from the site would be 2500 m^3.

Temporary repository or onsite storage of radioactive materials: For this scenario, in which the contaminated soil would remain at the site, albeit in a controlled mode, the allowable residual level would be ca. 60 000 Bq/kg, considering the dose limit for workers to be 10 mSv/a.

C-4. PLANNING OF REMEDIATION WORKS

The private company responsible for the USIN site was compelled by the authorities to close another one of its installations which is located in a very populous residential area of Sao Paulo. At that site, there was a considerable quantity of cake II, mesothorium cake and other by-products which had resulted from the chemical processing of monazite stored at this closed installation. Due to absence of a repository the State of Sao Paulo, and to the political difficulty of removing and relocating it in another state, the best option was deemed to be that of converting the USIN site to serve as a temporary repository of radioactive materials, that is, making it a controlled site.

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Annex D

CASE HISTORY — CANADA

URANIUM ORE CONTAMINATION OF A RAILHEAD IN NORTHERN CANADA

D-1. INTRODUCTION

This Annex presents a case history of the characterization and cleanup of soils which had been contaminated by the spillage of uranium ore and concentrates at a railhead in northern Alberta, Canada. The contamination was discontinuous and spread over some 45 hectares of industrial land. This paper describes the elements of this cleanup, with emphasis on characterizing the discontinuous contamination.

For much of this century until the 1960s Fort McMurray, Alberta was the northern limit of the railroad in northern Canada. Cargo heading north was transferred from rail to barge in Fort McMurray and, likewise, products from the north heading south were transferred from barge to rail. One such cargo was uranium ore from the Port Radium Mine at Great Bear Lake, Northwest Territories, which was transferred from barge to rail at one site in Fort McMurray in the 1930s and 1940s, and then at a second site from the 1940s to the 1960s. In 1992, soils contaminated with uranium ores and uranium concentrates were discovered on these industrial lands [D-1]. The discovery was made during an investigation of the historic water transportation route from Great Bear Lake to Fort McMurray.

The investigation was sponsored by the Low-Level Radioactive Waste Management Office (LLRWMO). The LLRWMO was established by the Canadian federal government in 1982 to resolve historic waste problems (those for which the original producer can no longer reasonably be held responsible and which are managed in a manner no longer considered acceptable) that are a federal responsibility, to ensure that a user-pay service is established for the disposal of low-level radioactive waste (LLRW) produced on an on-going basis, and to address public information needs concerning LLRW. In Canada, LLRW is defined as all radioactive waste except nuclear fuel waste and uranium mill tailings.

D-2. CHARACTERIZATION OF CONTAMINATED AREAS

It quickly became apparent during work to characterize the extent of contamination at the various sites, that a relatively small volume of original ores and concentrates had been spread through a much larger volume of native soils. In addition to the naturally radioactive elements uranium, radium and their radioactive decay products, the ores also contained other elements such as arsenic. A key element of the cleanup plan was then to segregate materials as follows:

- Category A material exceeded a uranium concentration of 500 ppm. It required a licence to possess under Atomic Energy Control Board (AECB) regulations.

- Category B material had a uranium concentration of less than 500 ppm but exceeded one or more of the cleanup criteria for uranium (30 ppm), arsenic (30 ppm) or Ra-226 (0.1 Bq/g). This was mainly contaminated soil, and was defined as and treated as industrial waste.

- Category C material, on average, did not exceed any of the cleanup criteria for uranium or radium, but may contain rocks with elevated amounts of one or more of these elements.

Preliminary investigations began by gathering information, reviewing records and contacting individuals with knowledge. An initial survey was then conducted. Gamma radiation readings and soil samples were taken at the surface and at a few selected test pits. Such surveys continued until the entire area affected by contamination had been defined. Leach tests were conducted on selected samples of contaminated soils to demonstrate their classification as industrial waste.

Preliminary investigations in Fort McMurray determined that some 45 ha of industrial land contained some amount of uranium ore contamination. The object was to establish the area requiring cleanup and to verify both that the cleanup was successful in the areas where cleanup was conducted and that no cleanup was required elsewhere on the properties. As part of the characterization of suspect properties in Fort McMurray, a representative 10% of the surface areas was surveyed using a large area gamma survey (LAGS) system [D-4, D-5]. The LAGS system (Fig. D-1), which consists of commercially available hardware and custom software, was developed by the LLRWMO to survey several hectare large tracts of land in a reproducible manner. Part of this LAGS analysis was conducted in trenches that were excavated in lifts that extended to the depth of disturbance. Data were analysed for discrete particles of uranium ore and areas identified by this method were investigated with an intensive hand-held instrument scan. This permitted characterization of the detailed nature of contaminating particles (volume, mass, composition) and their spatial density (number of contaminated particles per 100 m^2). Identified sources were removed and characterized for uranium content to determine whether the cleanup criteria for the project were exceeded.

D-3. REMEDIATION OF CONTAMINATED AREAS

The regulatory agency responsible for ensuring that the cleanup project was done properly was the Northern Lights Regional Health Authority. Alberta Health provided assistance and advice to the local organization and assisted with liaison with other provincial departments, including Alberta Environment which was responsible for confirming the waste classifications. In addition to these approvals, the project was subject to the requirements of the AECB, the nuclear regulatory agency in Canada and, since the LLRWMO is a federal organization, the requirements of the federal Environmental Assessment and Review process (EARP) [D-3]. Cleanup criteria were specifically established for radium, arsenic and uranium as follows [D-2]:

- The radium criterion was cleanup to levels within the normal background range for soils in Fort McMurray (up to 0.1 Bq/g). A study was carried out to measure the range of concentrations of radium, arsenic, and uranium which occur naturally in Fort McMurray.

- The arsenic criterion was cleanup to the "residential land use" level (30 ppm) as established by the Canadian Council of Ministers of the Environment (CCME). This is also the criterion for park land.

- The uranium criterion is the same level (30 ppm) as for arsenic. This is a conservative approach when the potential hazards of each element to human health and the environment are compared.

FIG. D-1. Large Area Gamma Survey (LAGS) System.

These cleanup criteria were developed for unrestricted land use in the future. They allow the land to be used for residential housing or parks, and are more stringent than the criteria required for industrial or commercial use of the land.

Following cleanup a comprehensive verification programme was carried out. The programme consisted of 100% surface LAGS survey both in areas which had been excavated as well as areas where no excavation had taken place. In areas where no excavation had taken place, test pits were excavated on a 20 m grid, the depth of disturbance limit identified, and a single sample taken of the disturbed material. Trenches were excavated in areas where an analysis of surface and test pit data indicated the likelihood of additional buried contaminated material. In excavated areas, soil samples were taken and portable gamma spectrometer readings were made.

As of 1995, approximately 30 hectares of industrial land in Fort McMurray had undergone characterization, cleanup and verification activities. Following successful completion of the verification programme, no restrictions on land use were required. Approximately 31 000 m^3 of contaminated material has been removed from the properties. The area requiring cleanup was

5.3 hectares. The cost of verification in Fort McMurray is approximately $13 000 per hectare or approximately $12/m^3 of cleaned up material, and is roughly equal for verifying that a cleanup is successful or verifying that no cleanup is required. The cost of cleanup, restoration and management of waste for this project is approximately $80/m^3. Since about 80% of the 30 hectares affected did not require cleanup, the savings compared to cleaning up the entire area affected by distributed waste may be $10 to $12 million.

D-4. CONCLUSIONS

A cleanup of soils contaminated with uranium ores and concentrates has been conducted in Fort McMurray, Alberta. Unnecessary cleanup work has been avoided and restrictions on land use due to possible radioactive contamination are not required on some 30 hectares of industrial land due to the selection and application of waste characterization techniques designed for wide spread, distributed contamination.

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Annex E

CASE HISTORY — CROATIA

E-1. INTRODUCTION

Radioactively contaminated sites in Croatia are generally not related to accident-generated contamination or to the uranium mining and milling industry, but rather they have resulted mainly from the dumping practices for naturally occurring radioactive materials (NORM) such as coal slag and ash, or phospho-gypsum residues/by-products in the phosphate fertilizer industry. Waste dumps of concern represent mostly a back-end of regular industrial production and cannot easily be shut down if the operation of respective facilities is expected to go on. Major waste dumps, presumed to be radioactively contaminated, have been controlled by authorized bodies and institutions in Croatia, but some uncertainties concerning final remediation actions still remained. The initiative taken by the IAEA in inaugurating a Technical Co-operation Project on Environmental Restoration in Central and Eastern Europe (RER/9/022) provided an impetus for the current efforts in Croatia to perform remediation at most of the contaminated sites. Unfortunately, there have been some aggravating factors (above all, a lack of finances and the need for reference facilities to maintain a continuous operation) which have retarded the full implementation of the programme.

The preliminary screening in Croatia has identified four NORM groups as follows: (1) dumps containing contaminated coal slag and ash; (2) dumps containing contaminated phosphates and phospho-gypsum from the fertilizer industry; (3) geothermal springs and gas/oil wells; and (4) sites containing natural radioactive "raw materials" (e.g. brickyards, ceramic factories, cement industry, etc.).

Facilities to which groups (1) and (2) usually refer are plants producing plastic materials (e.g. PVC), light-metal factories, iron works, cement factories, fertilizer plants, oil refineries, coking plants and coal-fired thermal power plants, but also old railway slag and ash-piles should be considered.

The programme performance co-ordination function has been assigned to the APO, the Hazardous Waste Management Agency, which serves as the national agency responsible for management of radioactive waste and other radioactive materials in Croatia. The main co-operating institutions are the Institute "Ruder Boskovic" and the "Institute for Medical Research and Occupational Health." Both of these institutes have radwaste storage facilities, where the wastes collected from nuclear applications in medicine, industry, agriculture and scientific researches can be temporarily held. The company EKOTEH, which is authorized to handle and transport radioactive materials, also participates in the programme. The regulatory framework for the programme performance is controlled by the Ministry of Health, the Ministry of Economy (Department of Energy) and the State Administration for Environmental Protection.

E-2. CHARACTERIZATION OF CONTAMINATED SITES

The sites of interest are being evaluated in accordance with the criteria indicated below. The evaluation is underway and will be used as basis for formulating proposals on further NORM dumping practices.

- site geology: lithology, hydrogeology and tectonics;
- site seismicity;
- meteorological features of the site: temperature, precipitation, winds, etc.;
- hydrology and water supply;
- demography: population density and distribution of settlements; occupational exposure of the on-site personnel;
- land use features of the areas neighboring to NORM dumps;
- NORM dumps characteristics: history of accumulation, quantities and radioactivity levels of dumped NORM; and
- transportation routes of NORM from sites of its origin to dump sites.

Descriptions are provided below for the highly prioritized sites which have resulted from the characterization phase.

E-2.1. The INA-VINIL plant in Kastel Sucurac

There are two coal slag and ash dumps with elevated radioactivity levels at the operating PVC factory INA-VINIL in Kastel Sucurac, located some 5 km north of the city of Split (population 230 000). The waste dumps are situated extremely close to the coastline of Kastela bay, so that contaminated material has been occasionally dumped even into the coastal waters. The site geology is characterized by Eocene flysch surrounded by carbonate lithology (limestone, dolomite). The original coal was transported to the site by the sea from the Rasa coal mine in Istria and from the bay of Boka Kotorska, as well as by railway from the adjacent north Dalmatian coal mines of Siritovci and Dubravice.

Slag and ash have remained after combustion of coal in the plant's energy-producing facility (fireroom) during past 40 years. One pile, with dimensions 100 m x 100 m x 1 m (i.e. 10 000 m^3) is closed off, having been covered by soil and PVC sheet. The other, smaller dump pile, which is presently in operation, contains up to 1000 m^3 of slag and ash, and is subjected to continuous monitoring by the Institute of Medical Research and Occupational Health. According to past measurements of radioactivity, concentrations of 18 600 Bq/kg of ^{238}U and 6200 Bq/kg ^{226}Ra have been identified at the older, now closed coal-slag and ash dump.

In order to obtain most recent, accurate and reliable data on actual radioactive contamination at the site, it is planned to carry out a sampling programme (using gamma-spectrometry, etc.) on both dump sites in a grid measuring 20 x 20 metres. Thermoluminescent dosimeters (TLDs) and Geiger-Mueller (GM) probes will also be set up at the site, and the radioactivities will be measured over a period of some 6 months.

A final decision on the possible need for, and the type and extent of, cleanup actions will be made after completion of the measurements plus other activities proposed in the project performance structure. Among the possible remediation methods being considered for application to the closed dump are (1) emplacement of a concrete Asaracophagus@ surrounding the NORM, and (2) displacement or removal of the dumped NORM.

E-2.2. Coal-fired power plant PLOMIN

A coal-slag/ash dump is located near to the coal-fired power plant PLOMIN site. The dump itself is situated in a dry valley, on Eocene flysch sediments surrounded by Cretaceous limestones and dolomites, approximately 1 km from the coast of the Plomin bay. There are about 15 000 inhabitants living within a 10 km radius around the dump. Covering approximately 120 000 square metres, some 700 000 cubic metres of slag and ash are estimated to be accumulated on the site so far. Slag and ash is accumulating continuously due to regular operation of the power plant (the average annual quantities of slag and ash may reach some 40 000 tonnes). The dump is partially covered by 1–2 m thick soil layer, is equipped with a drainage system (leading to a collecting tank), and, along with the power plant, is surrounded by the fence. Coal used for operation of the plant originates from nearby Tupljak, Ripenda and Koromacno mines. Ash removed from electro-filters in the power plant contains radioactivity levels of about 2600 Bq/kg ^{238}U and 2500 Bq/kg^{226} Ra. Determinations of radioactivity in the dumped slag and ash have yielded values of about 1600 Bq/kg ^{238}U and 1000 Bq/kg ^{226}Ra.

In order to identify the possible spread of radioactive pollution, the following sampling and measurements are being planned:

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

A radiological risk assessment of this NORM dump is currently under way. Among the possible remediation methods being considered are (1) covering up the entire dump with a layer of soil, (2) closing the dump, and (3) displacing or moving the dumped NORM material.

E-2.3."INA- PETROKEMIJA" phosphate fertilizer plant in Kutina

There are nearly 20 000 inhabitants living inside a 10 km radius around the INA-PETROKEMIJA plant. Essentially, there are two sites belonging to the industrial zone, where increased radiation levels are expected: (1) the in-door plant area itself (referring to phosphates as the raw material, and phosphate acids and fertilizers as the final products), and (2)

phospho-gypsum landfills lying ca. 5 km southward from the plant on the floodplain of Sava river. In fact, there are four pools having a total capacity of 20 million cubic metres, situated in alluvial sediments (gravel, sand, mud); some 3.5 million cubic metres of phospho-gypsum have been disposed of up to this time. Phospho-gypsum is transported in a water slurry from the plant to the 5 km distant storage pools through a special pipeline. Since the current practice of accumulating the phospho-gypsum mixed with water does not seem to be environmentally acceptable, the plan is, inter alia, to modify this technology to achieve a dry sedimentation of phospho-gypsum in the dump area. Additional modifications of the phospho-gypsum dumping practice, or the possible isolation or even removal of the dumps, will be evaluated in the future.

Albeit small, but increased levels of radiation could be also identified on agricultural lands where the fertilizers are being used. The basic difference in the perceived nature of contamination at these sites derives from the fact that ^{238}U is determined as a basic radioactive pollutant in the phosphate acids, mono-ammonium phosphate, and the final fertilizer product, while the ^{226}Ra remains in the phospho-gypsum. Thus, it was discovered through gamma-spectrometry that the specific activity of ^{226}Ra in both of the "Boucra" and "Morocco" phosphates, which are used in the plant, ranges up to about 1100–1500 Bq/kg. On the other hand, the specific activity of ^{238}U in the phosphate acids is about 1700 Bq/kg; in monoammonium phosphate, it is ca. 1800 Bq/kg; and in fertilizers it ranges up to about 800 Bq/kg. The observed peak values for phospho-gypsum samples taken from the dump sites are about 1200 Bq/kg ^{226}Ra.

Transportation routes for the imported phosphates (raw materials) from Adriatic ports of Sibenik and Rijeka are precisely known, and no significant contamination incidents along these routes have occurred.

The possible remediation actions under consideration include (1) on-site accumulation of dry phospo-gypsum, (2) use of concrete barriers surrounding the pools, (3) discontinuing the dumping practices, and (4) displacement or removal of the existing dumps.

E-3. PLANNING OF REMEDIATION WORKS

The general remediation programme structure consists of eight steps as they are presented below:

1. Sites suspected to be contaminated (~35 sites)
   (a) sites containing coal/ash piles — 15 sites;
   (b) sites containing phosphates and phospho-gypsum remaining from the phosphate fertilizer industry — 2 sites;
   (c) geothermal springs, oil and gas wells — 8 sites; and
   (d) sites containing naturally contaminated materials (cement, ceramics, bricks, etc.) — 10 sites.

2. Collection of available data on suspected sites
   (a) quantities of contaminated material;
   (b) composition of radionuclides contained in the material;
   (c) history of the contamination;
   (d) type and operational status of facilities generating contaminated materials;
   (e) lithology, hydrogeology, and seismo-tectonics of sites;

(f) vicinity to major urban or other densely populated areas; and
(g) identification of transportation routes of the material.

3. Identification and ranking of priorities:

Prioritization is based on measured (identified) NORM radioactivity. It was done with due regard to the following:
(a) protection measures applied at the NORM dump sites;
(b) vicinity to densely populated areas;
(c) land use practices in territories adjacent to NORM dumps;
(d) transportation routes of contaminated material; and
(e) geological and similar site characteristics.

4. Highly prioritized sites (3)
(a) INA-VINIL PVC plant in Kastel Sucurac.
(b) PLOMIN Coal-Fired Power Plant.
(c) INA-PETROKEMIJA Phosphate Fertilizer Plant in Kutina.

5. On-site inspection, detailed sampling and measurements
(a) radiochemical analyses;
(b) gama-spectrometry;
(c) identification of exposure doses (by TLDs);
(d) determination of Working Levels; and
(e) measurements of radon concentrations, etc.

6. Radiological risk assessment

7. Cost–benefit analysis for possible clean-up options

8. Decision making on physical cleanup options

E-4. CONCLUSIONS

As the ultimate goal of the remediation programme is a reasonable reduction of the currently existing radioactive contamination in the country and consequent reduction of possible harmful effects of contaminated sites to the environment and human health, it is reasonable to expect a considerably improved situation in Croatia. It remains clear that goals of the remediation programme cannot be realized in the short term, but through the programme = s efficient conduct and professionally performed actions, a successful conclusion should be attainable within a few years. Through this remediation programme of radioactively contaminated sites, not only is it expected that contaminated material can be efficiently "extracted" or isolated from the general environment, but there also will be an improvement in some processing techniques in industries or energy production. Thus, the associated generation of unreasonably high quantities of radioactively contaminated material is expected to be reduced. These results would represent a real benefit for the environment, and this should justify the efforts and expenses necessary for the programme realization.

Annex F

CASE HISTORY — SLOVAKIA

^{137}CS CONTAMINATED RIVER BANKS FROM NUCLEAR POWER PLANT (NPP) EFFLUENT AT THE BOHUNICE SITE IN SLOVAKIA

F-1. INTRODUCTION

The 18 km long river banks which represent a waste discharge route for the Bohunice NPP site, were contaminated by ^{137}Cs as a result of two incidents that occurred at the NPP-A1 in 1976 and 1977. In total, contamination exceeding 1 Bq/g of ^{137}Cs encompassed over 55 000 m^2 of land adjacent to the river. This Annex describes planning for restoration of these banks, with emphasis on characterization of the contaminated site.

The contamination of banks was identified in 1991 in connection with a flood control project in the nearby area. Soon afterwards, proper restoration action was requested by the competent hygienic authority from the operator of the Bohunice NPP, who was responsible for the bank contamination. A cleanup level given for this purpose by the authority was set up ad hoc on a very low level of 1 Bq/g of ^{137}Cs and a restoration project, including site characterization of the concerned part of the river, initiated by the NPP. The project included removal of 5000 m^3 of soil, and its disposal within the NPP site, into a subsurface concrete structure.

However, for various reasons it was necessary to postpone implementation of the restoration project. First, the extent of contamination was larger than expected previously. Secondly, the cleanup level set up by the authority appeared to be unacceptably low, considering the limited disposal capacity within the NPP site. Thus, a new restoration project has been developed which would issue from the latest site characterization studies and take into account all safety, social and political aspects of the site specific conditions.

F-2. SITE CHARACTERIZATION

Gamma radiation readings and soil samples for laboratory analysis were taken from the affected areas. The laboratory analysis has shown that ^{137}Cs is the dominant contaminant. Negligible amounts of other radionuclides, such as ^{60}Co, ^{134}Cs, ^{239}Pu, were found in the samples as well. Concentration of ^{90}Sr in soil, determined in 20 samples, was found to be from 50 to 100 times lower than that of ^{137}Cs. Nevertheless, its contribution to effective dose showed to be significant. Accessible areas close to the outer side of banks, were continuously scanned using the Vehicle Mounted Gamma Survey system (VMGS) [F-1]. The VMGS, which consists of a large shielded scintillation detector (NaI(Tl), 100 x 100 mm), commercially available nuclear instrument module (NIM) electronics, microcomputer and custom software, was developed in the VUJE research institute to survey the outer accessible side of the 18 km long affected banks and several hectares of the nearby land-fields and flood plain area. Contamination spread over 2000 m^2 alongside the Dudvah bank and a limited flood plain area of the Vah River and the former Dudvah River were discovered and evaluated by this method.

Consequently, the VMGS was mounted on the hydraulic arm of a tractor and tested also for hard-to-access steep Manivier channel banks. Inside the levees, hand held, slightly shielded, gamma survey meter with a large plastic scintillator (75 x 75 mm) was used for point

measurements within a 20 m line grid. Measuring time constant, τ = 10 s, was applied to reach sufficient sensibility of the detector. In critical places with the highest activities, a line grid with 10 m measuring step was used achieving about 15% covering of the surveyed bank surfaces. The shielding of the detector was important to achieve sufficiently close correlation between the activity concentration and the detector response above the contaminated bank strip with variable width. For the canal sections with contaminated spots, in addition to mentioned line grid survey, continual scanning with τ = 1 s to search for the most intense contaminated spots had been applied as well. Some of discovered spots were investigated in detail by scanning and soil sampling to determine its shape, size, specific activity and depth distribution of ^{137}Cs. No tight correlation was found between the detector response and specific activity of the spots. This was caused by its irregular shape and variable size, which in most cases were smaller than the circle area seen by the used detector (R_{dw} = 70 cm).

Finally, three groups of spots were selected according to typical average activities obtained by sample laboratory analysis. Sizes and linear densities of these contaminated spots were roughly estimated on the basis of obtained device reading. These data were then applied to make a conservative assessment of average bank strip contamination. More accurate evaluation of these discontinuities in bank contamination requires application of a more sophisticated measuring technique with demand for further development.

The detailed and comprehensive survey done between 1991 and 1994 shows that the top soil contamination on the banks widely varies from background level to 20 Bq/g for the Dudvah River and reaches 250 Bq/g of ^{137}Cs for spotted section on the channel banks. The contamination is spread over about 0.5 to 3 m wide strip on the lower part of the banks (on average. over 2.2 m) and the average level of ^{137}Cs in the top 10 cm soil layer reaches 6.3 Bq/g. Large spots of contamination with activities up to 10 Bq/g in a subsurface 5 to 20 cm layer of sediments were found in the Vah river flood plains in the places where the Dudvah River flew in the past before its reconstruction. A thick grid survey together with frequent sediment sampling and assumption of the ratio between the surface and more contaminated subsurface sediment layer were necessary to evaluate this type of contamination. The underlying gravel bed and the ground water were sampled and measured, too, but no significant contamination has been found.

Thus, the overall contaminated area with the activity level exceeding 1Bq/g of ^{137}Cs, is approximately 67 000 m^2 and the total volume of soil which has to be removed according to the first cleanup limit exceeds 13 000 m^3. However, volume distribution investigation of the contaminated soil has shown that removing only the mostly contaminated soil would result in a significant improvement in remediation of the contaminated site.

F-3. DOSE ASSESSMENTS

To determine plant/soil concentration ratios for ^{137}Cs and ^{90}Sr, a variety of grass samples were analyzed. Selected scenarios with authorized parameters (residential, agricultural use of land) were applied in the site assessment studies , as well as for development of proper acceptance and cleanup limits. Ingestion pathways using transfer factors for goats milk and meat and loamy soil according to [F-2] was also part of dose estimates. The calculated effective dose for a long-term exposure at the contaminated site does not exceed 0.35 mSv/a, whereas the use of contaminated soil may result in doses ranging from 2 to 3 mSv/a.

F-4. PLANNING OF REMEDIATION WORKS

Acceptance criteria and cleanup limits for the contaminated banks were derived on the basis of authorized principles and the most critical soil use scenario dose factors [F-3]. According to the accepted approach of the International Committee on Radiation Protection (ICRP), both the potential and the actual individual risk from the contaminated banks must not exceed an effective dose level of 1 mSv/a.

To meet this requirement, average ^{137}Cs activity levels are as follows:

- AL_{200}=6.0 Bq/g in 200 m^3 of soil, or over 300 m of the bank, or
- AL_{50} = 8.0 Bq/g in 50 m^3 of soil, or over 80 m of the bank.

In addition, specific ^{137}Cs activities are stipulated as follows:

- $AL_{.3}$ = 25 Bq/g for isolated small spots on the canal banks, and
- Al_{ws} = 4 Bq/g over 1 km of bank which, together with the first two concentration limits, were proposed as cleanup criteria for the contaminated banks.

To be in compliance with these criteria, it is necessary to remediate about 11 000 m^2 of contaminated area on the Dudvah River banks and 8000 m^2 on the Manivier canal banks. As engineered flat terraces prevail on the Dudvah River banks, according to the authorized principles, clean soil cover should be applied over 9500 m^2 of contaminated flat area [F-4]. As a result, the volume of soil to be removed and safely disposed of is about 1100 m^3.

For a comprehensive remediation of the entire contaminated site, VUJE research institute has been involved since 1993. One feature has been a lack of clear legislation or regulations in this field. Consequently, there is a basic need to develop some principles for evaluating the contaminated site and to justify the remedial measures, including the establishment of appropriate cleanup. This demand was realized by the VUJE Institute working in close co-operation with the Institute of Preventive and Clinical Medicine in Bratislava. The submitted proposals of the comprehensive remediation project was approved by the State Health Institute in Bratislava in early 1995. The proposed restoration project is planned to be implemented by the Bohunice NPP in 1996/1997.

F-5. CONCLUSIONS

Re-evaluation of a ^{137}Cs contaminated bank restoration project has been conducted for NPP Bohunice site on the basis of comprehensive and detailed site characterization. As there is no clear legislation in this subject area in Slovakia, principles for contaminated bank evaluation had to be defined and approved by the competent authorities. Site-specific cleanup criteria have been developed depending on the size of contaminated area, which are 6 or 8 Bq $^{137}Cs/g$ in soil. Due to the application of a consistent site characterization techniques and dose assessment method, in the new restoration project disposal of slightly contaminated can be avoided.

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Annex G

CASE HISTORY — UNITED KINGDOM

PRODUCT FISSION CONTAMINATION OF RAILWAY LAND BY RAINWATER RUNOFF FROM FUEL TRANSPORTATION FLASKS

G-1. INTRODUCTION

This Annex presents a site characterization and remediation study for contamination occurring at loading areas for nuclear fuel transport containers. A number of sites where irradiated fuel transport containers (flasks) had been loaded onto rail transport wagons were believed to have been contaminated by low levels of radioactivity. It is most likely that the activity had been washed from external contamination on the paint and bodies of the flasks. Most of the activity was very old in origin, coming from the early days of operation of civil nuclear power.

G-2. CHARACTERIZATION METHOD OF CHOICE

A staged approach was taken to the problem. The aim of the exercise was declared to be to remediate the site so that residual contamination arising from operations was below the exemption limit where authorization is required to dispose of radioactive waste under UK law. For the fission products of concern, this level is set as 0.4 Bq/g (see Ref. [G-1]). The approach taken for characterizing the site was to determine the extent of gamma emitting contamination by survey, whilst the hard-to-measure isotopes (Sr-90, etc.) by correlation with Cs-137 determined from a restricted number of samples from the site. The possible presence of non-gamma emitting nuclides meant that the easy to measure Cs-137 had to be quantified to levels considerably below 0.4 Bq/g to give a reasonable standard of assurance that all radioactivity present as a result of fuel transport operations had been removed to below the exemption limit.

The use of simple hand-held health physics instrumentation was rejected for determining the extent of the contamination as the low quantification limit required meant that discrimination against the naturally occurring background radiation from radium and K-40 would require long counting times and a very well characterized background. Variations in the background would lead to uncertainties in the results.

In situ gamma spectrometry using an uncollimated high purity germanium detector of about 30% relative efficiency, mounted 1 m above the ground on a tripod, was therefore used to determine the extent of the contamination. Initial results showed that the extent of contamination was essentially confined to a small area between the tracks close to where the flask loading operations had taken place or where wagons carrying flasks had stood while awaiting departure.

Recording the location of the detector was simple on these sites as positions could be determined relative to the rail tracks and sleepers. On each site, several hundred gamma spectra were acquired and analysed in real time for Cs-137.

A small number of samples dug from the site gave the variation of activity with depth. In some places, the usual decreasing profile of activity with depth was confused by movements of

track ballast during past maintenance operations. With some knowledge of the extent of the contamination and with the aid of simple shielding calculations, it became possible to quantify the amount of Cs-137 in the area under examination and so (with the aid of the fingerprint data) to determine the volume of material to be removed.

G-3. REMEDIATION OF CONTAMINATED SITE

After the initial survey, material was excavated to the depth which it had been calculated would enable clearance of the site to below the exemption limit. The material excavated was transferred to drums or half-height ISO containers (a standard freight transport container of approximately 6.1 x 2.4 x 1.3 m) for consignment as potential low level waste (LLW). As a check of the techniques used, assay of the waste material using a LLW drum monitor gave excellent agreement with assessments of the quantity of activity predicted by the in situ measurements. Following the remediation operations, a further survey using in situ gamma spectrometry confirmed that the objectives of the cleanup had been met.

G-4. CONCLUSIONS

The methods used demonstrated that in-situ gamma spectrometry is an effective tool for assessing in a short time large areas of land where contamination was expected to be near the surface. The counting times at each survey point were short due to the large volume of material "seen" by the detector. The effort involved was significantly less than would have been required to have achieved similar results from sampling and laboratory analysis. The methods used enabled us to determine the extent, vertically and laterally, of the material to be removed and so minimized the cost of consignment of material as low level waste.

REFERENCE TO ANNEX G

[1] The Radioactive Substances (Substances of Low Activity) Exemption Order 1986, Statutory Instrument 1986, No. 1002 (United Kingdom).

Annex H

CASE HISTORY — UNITED STATES OF AMERICA

ADAPTIVE SAMPLING AND ANALYSIS PROGRAMMES FOR SOILS CONTAMINATED WITH RADIONUCLIDES IN THE US DEPARTMENT OF ENERGY FORMERLY UTILIZED SITES REMEDIAL ACTION PROGRAM (FUSRAP): THE PAINESVILLE SITE, PAINESVILLE, OHIO

H-1. INTRODUCTION

During the 1940s, 1950s, and 1960s, work was performed at sites throughout the United States as part of the nation's early atomic energy programme. Some sites' activities can be traced back as far as World War II and the Manhattan Engineer District (MED); other sites were involved in peacetime activities under the Atomic Energy Commission (AEC). Both MED and AEC were predecessors of the current Department of Energy (DOE). Most sites that became contaminated during the early atomic energy programme were cleaned up under the guidelines in effect at the time. Because in most cases those cleanup guidelines were not as strict as today's, trace amounts of radioactive materials remained at some of the sites. Over the years, contamination was spread to other locations, either by demolition of buildings, intentional movement of materials, or by natural processes.

DOE began Formerly Utilized Site Remedial Action Programme (FUSRAP) in 1974 to study these sites and take appropriate cleanup action. When a site is thought to be contaminated, old records are reviewed and the site is surveyed. If contamination is found that is connected to MED or AEC activities, cleanup is authorized under FUSRAP. Some sites with industrial contamination similar to that produced by MED or AEC activities have also been added to FUSRAP by the United States Congress. Since starting FUSRAP, DOE has examined records or performed surveys on more than 400 sites. Most were not contaminated, but 46 sites in 14 states have been found to be contaminated with radioactivity that exceeds current cleanup guidelines. Limited cleanup began at some sites in 1979, and major remedial action has been underway since 1981. Cleanup has been completed at 23 of the sites; 19 others have been partially cleaned up. To date more than 175 vicinity properties, including homes, parks, and streams, have been cleaned.

This case study describes how various FUSRAP methodologies and technologies were applied to the Painesville, Ohio site.

The Painesville site is a hazardous waste site in Painesville, Ohio, that is being restored by the Department of Energy's FUSRAP programme. In the early 1940s, the Defense Plant Corporation constructed a magnesium production facility on property owned by the Diamond Magnesium Company. In 1952 and 1953, Diamond Magnesium received approximately 1450 tons of radioactively contaminated scrap steel from the Lake Ontario Storage Area. Steel was used to control chlorine emissions during the magnesium production process. At the time of the magnesium production work, about a third of the site's approximately 150 acres was covered by large buildings and rail lines. At present, some of the same buildings are in use; others have been removed and/or replaced with other buildings, storage tanks, overhead pipe-rack systems, and spill-retention areas. In the early 1960s, the General Services Administration sold the Diamond

Magnesium facility as commercial property to private concerns. Portions of the facility are still in operation. Regulatory oversight is being provided by the Ohio Environmental Protection Agency (OEPA).

H-2. SITE CHARACTERIZATION

At Painesville, DOE through FUSRAP was responsible for characterizing and remediating any radionuclide contamination present that was a likely result of MED or AEC supported activities. In addition, DOE was responsible for any chemical contamination commingled with the radionuclide contamination irrespective of the source of the chemical contamination. In the case of Painesville, a team consisting of FUSRAP staff, Science Applications International Corporation (SAIC), Bechtel National, Inc. (BNI) and Argonne National Laboratory (ANL) was tasked with the responsibility for planning and executing a site characterization programme.

In 1988, a preliminary site evaluation was performed by the Oak Ridge National Laboratory. This evaluation entailed performing a gamma walkover survey over selected portions of the site and collecting soil samples for radiological analysis. The radiological survey results have indicated radionuclide concentrations in excess of federal guidelines in both surface and subsurface soils. The primary contaminants of concern (COCS) were ^{238}U, ^{230}Th, and ^{226}Ra. The radionuclide contamination encountered was confined to soils. Insufficient information was collected during the preliminary site evaluation to determine whether ground water beneath the site was also affected. Soil activity levels up to 76 pCi/g, 310 pCi/g and 1500 pCi/g for ^{238}U, ^{230}Th and ^{226}Ra, respectively, were encountered.

In addition, because of the commercial operation of the site as a chemical production plant, elevated levels of metals, volatile organic compounds (VOCs), and semi-volatile organic compounds (SVOCS) potentially existed in the areas containing soils contaminated with radionuclides. The metals and chlorinated solvents which may have been used historically at the site were of particular concern since they have the potential for creating a Resource Conservation Recovery Act (RCRA) waste or mixed waste. The OEPA Emergency Response Online System had listed several spills or releases of VOCs and SVOCs at the site. Disposal costs for mixed waste contamination in the United States can range into the thousands of dollars per cubic meter.

The characterization activities at Painesville incorporated several innovative methodologies/technologies to expedite the characterization/remedial action process. These included:

- DOE's SAFER approach to hazardous waste remediation decision-making;
- Adaptive Sampling and Analysis Programme (ASAP) techniques;
- Field screening and field analytical methods for rapid data generation;
- World Wide Web technologies for data transmission, organization, and communication;
- Site specific risk assessments; and
- An Engineering Evaluation/Cost Assessment approach to regulatory requirements.

The characterization activities at the Painesville site were based on DOE's SAFER approach to environmental remediation. SAFER expedites characterization by explicitly stating the problem to be addressed, identifying the decisions that need to be made, identifying the types of data required to make those decisions, defining the spatial boundaries for data collection, developing decision rules based on the data to be collected, specifying the uncertainty that will be tolerated when making those decisions, and then optimizing sampling programme design to meet those decisions. Through these steps, SAFER keeps data collection focused on the decision to be made.

An adaptive sampling and analysis programme (ASAP) relies on field analytical methods and in-field decision-making to guide data collection. ASAP takes its name from the fact that data collection adapts, or changes, to reflect the information that is being collected. A key component to ASAP are dynamic sampling and analysis plans. Rather than specifying the numbers and locations of samples to be collected, dynamic sampling and analysis plans include the decision-making logic that will be used to determine where samples should be collected as the sampling programme progresses and when sampling can stop. The use of ASAPs can provide significant characterization costs savings by reducing analytical costs, limiting the number of samples that are collected, and bringing characterization to closure within one data collection programme.

ASAP programmes require field screening and field analytical methods to be effective. In the case of Painesville, a variety of technologies were used to expedite the availability of data. These included gamma sensors typically used during site walkovers combined with geographic positioning systems, on-site gamma spectroscopy capabilities for discrete sample analyses, down-hole gamma detectors, and quick-turn-around contracting with off-site laboratories for alpha spectroscopy, metals and VOC analyses.

For ASAP programmes to be effective, the proper decision-making support must be made available to technical staff in the field. In the case of sites such as Painesville where there are multiple stake-holders and potential decision-makers, it often becomes logistically impossible to have these key people at the site at all times. This is particularly true when key decision-makers are involved with multiple, concurrent projects that may not require their total attention all of the time, but do demand immediate attention at unpredictable points in the programme. For Painesville, World Wide Web techniques were used to move the site to the desktop of decision-makers, where ever they might be.

The generic DOE guidance for the cleanup of soils contaminated with radionuclides is based on what is known as the "5/15" rule --- the allowable level of contamination above background for surface soils is 5 pCi/g, and for subsurface soils 15 pCi/g. In practice this can lead to remedial actions that are more conservative than warranted by a site specific risk assessment. In the case of Painesville, the "5/15" rule was used to determine whether contamination existed above DOE's generic guidelines, but the evaluation of alternative remedial actions in the EE/CA were based on a site specific risk assessment based on the contaminants observed.

The ultimate product of this characterization was an engineering evaluation/cost assessment (EE/CA) that specifies and evaluates the remedial action alternatives based on the contamination encountered. Within the United States regulatory context, the EE/CA process provided an accelerated path towards site cleanup and closure, with the total elapsed time between initiation of characterization and completion of the selected remedial action remedy for Painesville

expected to be less than 12 months. This can be compared to the more traditional remedial investigation/feasibility study approach which can take years to simply select the best remedial action.

The dynamic sampling and analysis plan developed for Painesville two principal questions that had to be addressed: (1) whether the contaminants of concern (COCs) in the various media at the Painesville site posed a significant risk to human health and the environment; and (2) what remedial action alternatives would cost effectively minimize or eliminate public exposure to the site COCs. To address these two questions, the sampling and analysis plan specified a data collection programme that contained two distinct phases.

The first phase included these stages:

Stage 1: Review historical records, aerial photographs and drawings to gather information on historical operations, types and quantities of chemicals used, volumes of process waste generated, waste disposal practices, detailed process information and current site information such as ecological receptors present at the site.

Stage 2: Collect ambient air samples from selected areas of the site where contamination is thought most likely. This stage verified that there were no immediate health and safety risks for on-site workers.

Stage 3: Collect external gamma exposure rate measurements at selected locations across the site, including inside buildings focusing on areas where contamination is thought most likely. This stage verified that there were no immediate health and safety risks for on-site workers.

Stage 4: Conduct gamma walk-over surveys with bicron and fidler instruments using global positioning systems to map readings as the walk-overs proceed. Conduct building radiological surveys. This stage identified "hot spots" and was used to assist in defining the lateral extent of surficial contamination.

Stage 5: Perform an ecological receptor evaluation for the site, including a habitat survey and a pathway analysis to identify mechanisms by which COCs might affect flora or fauna.

Stage 6: Conduct surface geophysical surveying to identify potential fill areas that might contain buried wastes.

Stage 7: Collect surface water and sediment samples from drainages leaving the site to evaluate the migration of COCs off-site via these pathways.

Stage 8: Perform shallow soil sampling to define the depth and areal extent of shallow soil contamination based on the results from the walk-over surveys.

Stage 9: Complete any discretionary activities to resolve issues raised during the data collection efforts of the first eight stages.

Phase II activities included:

Stage 1: Conduct deep soil sampling in areas where shallow soil sampling failed to encounter the vertical limits of contamination.

Stage 2: Conduct direct push ground water sampling to determine if ground water has been affected by activities at the site, and if so the nature and extent of that contamination.

Stage 3: Install permanent ground water monitoring wells if the results from the direct push ground water sampling indicate significant ground water contamination.

Both Phase I and Phase II activities were completed with one characterization programme that took place between July and November 1996. Field trailers were established at the site that included ISDN connections for personal computers housed in the trailers. Via both these ISDN connections and standard modems, staff on site were able to directly upload, download and query data stored in an SQL (structured query language) database at Oak Ridge, Tennessee.

A Web home page was established for the characterization programme that included digital photographs taken at the site as work progressed, computer generated graphics that showed the progress and results from the radiological walk-over surveys as they progressed, data tables downloaded from the SQL database that showed sample analytical results for samples taken from soil bores installed at the site, an anonymous ftp site where data files could be placed and accessed by project personnel, and an E-mail list server that allowed home page users to mail questions and/or concerns about project data and progress to key project personnel. This home page was accessible both to technical staff at the site and to anyone else with access to a World Wide Web browser. The Web page and associated ftp site became the principal means for data transfer and communication as the project progressed.

The combined gamma sensor/GPS system was able to survey at the rate of approximately 1.5 hectares per day, generating approximately 3000 data points per acre. An on-site gamma spectroscopy laboratory provided 24 hour turn-around for soil samples collected at the site. Off-site laboratories provided 7 day turnaround for alpha spectroscopy (required for ^{230}Th) and metals NOC analyses. The walk-over survey provided 100% coverage for the site and successfully identified nine distinct areas with elevated radionuclide levels. These areas ranged from isolated "hot-spots" to approximately 0.4 hectares in size. Using discrete sample results along with the gamma walk-over data, the gamma walk-over data was converted into a probability map for the site, mapping the probability of exceeding DOE surface soil standards based on the gamma walk-over data. This probability map was updated with the discrete sampling results using combined Bayesian/geostatistical techniques. From the combined gamma walk-over data and discrete sample results, estimates of total areal extent could be derived.

The interiors of each of the areas with elevated radionuclide levels were sampled randomly to both support a site specific risk assessment and to determine whether chemical contaminants were also present. No significant chemical contamination was encountered that was concurrent with radionuclide contamination. The results from the site specific risk assessment, along with the volumetric contamination estimates derived from the gamma walk-over surveys and discrete

samples, were used to identify and evaluate various remedial action alternatives for the site as part of the EE/CA process.

H-3. LESSONS LEARNED

The Painesville site was the first FUSRAP site that attempted to integrate several innovative characterization methodologies and technologies to expedite the remedial action process. Several lessons can be learned from FUSRAP's experience at Painesville:

(1) The ASAP programme deployed at Painesville was successful in delineating contamination extent and in providing the data necessary to support the site specific risk assessment. Despite a fairly significant amount of logistical planning before the start of data collection, the rapid pace of characterization and the large amounts of data generated placed a heavy strain on the data management and analysis plan that was in place.

(2) The use of the World Wide Web site proved to be an excellent means for assembling, communicating and disseminating information generated throughout the course of the programme to key decision makers and affected parties.

(3) Site specific risk assessments provided DOE with a broader range of remedial action alternatives than would have been available if the generic radionuclide guidelines for soils had been strictly applied. Because the current and expected future use of the Painesville site is heavy industry, less restrictive cleanup goals derived from the site specific risk assessment could be applied.

(4) The EE/CA process provided a regulatory "fast track" option for characterizing and remediating the site. While an EE/CA is not always an appropriate path forward, in the case of Painesville, combined with the ASAP used for characterization, it reduced the remedial action time frame from years to months.

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Advisory Group Meeting

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