



# ***Technical options for the remediation of contaminated groundwater***



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## FOREWORD

Throughout the world, many countries have experienced problems associated with pollution of the environment. Nuclear fuel cycle, medicine, industry, weapons production and testing, research and development activities, as well as accidents, poor past practices, etc. have produced a large array of radioactively contaminated facilities and sites. Structures, biota, soils, rocks, and both surface and groundwaters have become contaminated with radionuclides and other associated contaminants, a condition that raises serious concern due to potential health effects to the exposed human populations and the environment.

In response to the needs of its Member States in dealing with the problems of radioactive contamination in the environment, the International Atomic Energy Agency (IAEA) has established an environmental restoration project. The principal aspects of current IAEA efforts in this area include (1) gathering information and data, performing analyses, and publishing technical summaries, and other reports on key technical aspects of environmental restoration; (2) conducting a Co-ordinated Research Project in Environmental Restoration; and (3) providing direct technical assistance to Member States through technical co-operation programmes. The transfer of technologies to Member States in need of applicable methodologies and techniques for the remediation of contaminated groundwater is an objective of this IAEA project.

This TECDOC focuses on the technical options for remediation of radioactively contaminated groundwaters, as well as on the planning and management options to accomplish environmental restoration of this valued resource. Most of the described technologies are applicable for the remediation of radioactive contaminated groundwater as well as for remediation of non radioactive contaminated groundwater. As used here, the term "remediation" includes decontamination of groundwater and other media, stabilization or isolation of contamination, together with the disposition of wastes arising from the cleanup. The worldwide experience and trends in the remediation of groundwater contamination have been surveyed and are summarized in the report.

This TECDOC was produced as a result of a series of consultancies and an Advisory Group meeting in 1997 and 1998. The IAEA officer responsible for this publication was D. Stritzke of the Division of Nuclear Fuel Cycle and Waste Technology.

It is intended that this report will serve as an important source of information to Member States who are faced with the problem of radioactively contaminated groundwater.

## *EDITORIAL NOTE*

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## CONTENTS

1. BACKGROUND .....	1
2. OBJECTIVE .....	2
3. SCOPE .....	2
4. NATURE AND EXTENT OF THE PROBLEM .....	3
4.1. Groundwater interactions .....	3
4.2. Groundwater contamination scenarios .....	4
4.2.1. Contaminant sources .....	4
4.2.2. Source control and soil remediation .....	5
4.2.3. Conceptual models .....	5
4.3. Exposure pathways .....	7
4.4. Hazard assessment .....	7
4.5. Contaminants of concern .....	9
4.5.1. Radioactive contaminants .....	9
4.5.2. Non-radioactive contaminants .....	12
5. A GENERAL APPROACH TO THE MANAGEMENT AND SELECTION OF REMEDIAL ACTIONS .....	13
5.1. Scoping analysis .....	14
5.2. Early decisions regarding further action .....	15
5.2.1. No further action needed .....	15
5.2.2. Further monitoring of contaminant plume is required .....	15
5.2.3. Insufficient data exist to make a decision .....	15
5.2.4. Remedial action is required .....	15
5.3. Public involvement .....	16
5.4. Establishment of remediation goals .....	16
5.5. Site characterization .....	16
5.5.1. Data collection objectives .....	17
5.5.2. Identification of uncertainties .....	17
5.5.3. Data quality objectives .....	17
5.5.4. Data analysis .....	18
5.6. Development and screening alternatives .....	18
5.7. Institutional controls .....	18
5.8. Analysis and design of preferred alternatives .....	19
5.9. Implementation action and performance assessment .....	19
6. MODELLING AND ASSESSMENT .....	20
6.1. The application of modelling in the groundwater remedial process .....	21
6.2. Stepwise implementation of the modelling process .....	22
6.3. Modelling techniques and approaches .....	23
6.4. Parameter uncertainties .....	25
6.5. Cost-benefit analysis and data worthiness .....	26
6.6. Limitations of groundwater modelling .....	26

7. TECHNOLOGIES FOR GROUNDWATER REMEDIATION .....	27
7.1. In situ remediation .....	29
7.1.1. Containment .....	29
7.1.2. Treatment technologies .....	33
7.1.3. Contaminant flushing .....	36
7.2. Ex situ remediation technologies .....	37
7.2.1. Collection and immobilization .....	40
7.2.2. Collection and treatment technologies .....	40
7.2.3. Process integration .....	43
8. SUMMARY AND CONCLUSIONS .....	43
REFERENCES .....	45
ANNEXES: CASE HISTORIES OF NATIONAL EXPERIENCE .....	49
ANNEX I: UNITED STATES OF AMERICA — THE UMTRA PROJECT .....	54
ANNEX II: REMEDIATION OF RADIONUCLIDE CONTAMINATED GROUNDWATER IN CANADA .....	56
ANNEX III: CLEANING OF RADIONUCLIDES FROM UNDERGROUND WATER AT SITES CONTAMINATED BY URANIUM MINING AND MILLING ACTIVITIES IN THE RUSSIAN FEDERATION .....	69
ANNEX IV: UKRAINE (CHERNOBYL EXCLUSION ZONE) .....	75
ANNEX V: PROGNOSIS OF GROUNDWATER CONTAMINATION CAUSED BY FLOODING OF URANIUM MINES IN EASTERN GERMANY .....	94
ANNEX VI: RADIOACTIVELY CONTAMINATED LAND AREAS AND WATERS IN BULGARIA .....	105
ANNEX VII: NATIONAL EXPERIENCE ON GROUNDWATER CONTAMINATION AND REMEDIATION ASSOCIATED WITH URANIUM MINING AND MILLING IN THE CZECH REPUBLIC WITH CASE EXAMPLE FROM THE NORTH BOHEMIAN CRETACEOUS AREA .....	110
GLOSSARY .....	125
CONTRIBUTORS TO DRAFTING AND REVIEW .....	129

## 1. BACKGROUND

Although many cases of radioactively contaminated groundwater have occurred throughout the world, few successful experiences in remediating the problems are identifiable at this time. Many countries are in the early process of identifying their potential groundwater contamination problems, or studying a known problem, while others are attempting remediation. Before a remediation strategy can be implemented, the objectives of the remediation programme should be identified and the potential problem should be investigated, at least, in sufficient detail to understand the magnitude of the problem and implications of the proposed remedial actions.

At present, many innovative technologies are being evaluated for their usefulness to remediate radioactively contaminated groundwater, as well as passive and no-action alternatives. Conventional extraction and treatment processes have often proven to be very expensive and slow with respect to remediating the contaminated groundwater back to pre-contamination conditions. One should be aware that most of the described groundwater remediation technologies in this report are convenient for radioactive as well as for non radioactive contaminants.

The report discusses important aspects of the nature and extent of the problem, a general approach to the management and selection of remedial actions, application of modelling, technologies for groundwater remediation, national experiences, and selected case histories representing various countries. In this report, it is assumed that the contaminant problem has been investigated in sufficient detail and the initial characterization has been completed.

Radioactive contamination of the environment has occurred in many parts of the world due to, inter alia, accidents; past practices in radioactive waste management and other activities in the nuclear fuel cycle; spills; nuclear weapons testing; and the mining, milling, and processing of radioactive ores. Because of the potential impacts of the contaminated media (e.g. groundwater, surface water, and soil) on the food chain and the overall health and safety of human populations and the environment, there is considerable interest in remediating the contamination problem to the extent that the site can be available for continued or alternative use. In compliance with its mandate to promote the safe and peaceful use of atomic energy, the International Atomic Energy Agency (IAEA) has an important role to play in assisting its Member States to plan management options, utilize available technologies, and seek solutions to these environmental concerns. Thus, the IAEA has initiated a project to focus on the environmental restoration of radioactively contaminated sites.

This report will directly complement and support other IAEA activities in the area of environmental restoration including: technologies for land remediation; characterization of radioactively contaminated sites; formulation of strategies for environmental restoration; and post restoration monitoring of decommissioned sites to ensure compliance with cleanup criteria. References [1–5] are IAEA publications which are related to this TECDOC.

One important task in this project is concerned with the planning and management options for the cleanup and remediation of radioactively contaminated groundwaters (the subject of this TECDOC). Apparently, in the past, this subject has found somewhat limited attention and was not addressed before in a cohesive manner and on a broad international level. With increasing public awareness of the limitations of groundwater resources, however, the need for protection and remediation of groundwater is gaining increased importance.

## **2. OBJECTIVE**

The objective of preparing and publishing this TECDOC on planning and management options for the remediation of radioactively contaminated groundwaters is to present an overview and provide guidance on the state of the art of available methodologies and technologies to remediate contaminated groundwaters and thereby effectively reduce the radiological impacts. The worldwide experience and trends in the remediation of groundwater contamination have been surveyed and summarized in the report. The report is intended to address the concerns of technical managers, regulators and competent authorities, and other parties involved in decision making and conducting of groundwater remediation activities.

## **3. SCOPE**

This report provides a description of the nature and extent of problems related to radioactive groundwater contamination by outlining the environmental impacts, the sources of contamination and the contaminants of concern radionuclides and their associated contaminants – the main exposure pathways and transport processes and the assessment of risks associated with contaminated groundwater. The main emphasis of this report is on methodologies used in groundwater remediation and available technologies. The methodology section outlines the importance of an initial scoping analysis including the evaluation of uncertainties of the available data and the necessity for defining clear objectives for data collection. This is then followed by a comprehensive site characterization, setting of goals and developing alternatives which will be analysed in detail.

Technological methods for remediating contaminated groundwater are described in terms of providing an overview which does not claim to be complete. Although substantial research is currently being carried out in this field, only a few innovative methods (see Section 5) have been developed to a state which could define these methods as proven technology. Also, it needs to be stressed that the applicability of groundwater remediation technologies is dependent on the site specific conditions.

Available technologies are grouped generally into in situ methods aiming at a containment of the contaminants in place and engineered treatment methods involving an alteration of groundwater flow, quantity and/or quality to achieve compliance with set goals. Groundwater remediation by natural flushing allows the natural groundwater movement and geochemical processes to decrease the contaminant concentrations to acceptable levels over a specified period of time. This method is increasingly accepted in areas where the use of groundwater can be temporarily restricted or engineered cleanup methods do not offer particular advantage over the natural processes.

The application of technological methods for remediating contaminated groundwaters has to be considered in conjunction with management options such as diversion and development of alternative water sources. The experience with groundwater contamination accrued in IAEA Member States is concentrated in those countries with active uranium mining and milling facilities and nuclear energy programmes. This experience is reported in the Annexes, which include case studies. It includes aspects of site specific configuration of radioactive contamination, social and political factors, legal and regulatory factors and constraints, economical and technical considerations and socioeconomic criteria, including the importance of public acceptance and public involvement in the decision making process of remediation of radioactive groundwater contamination.

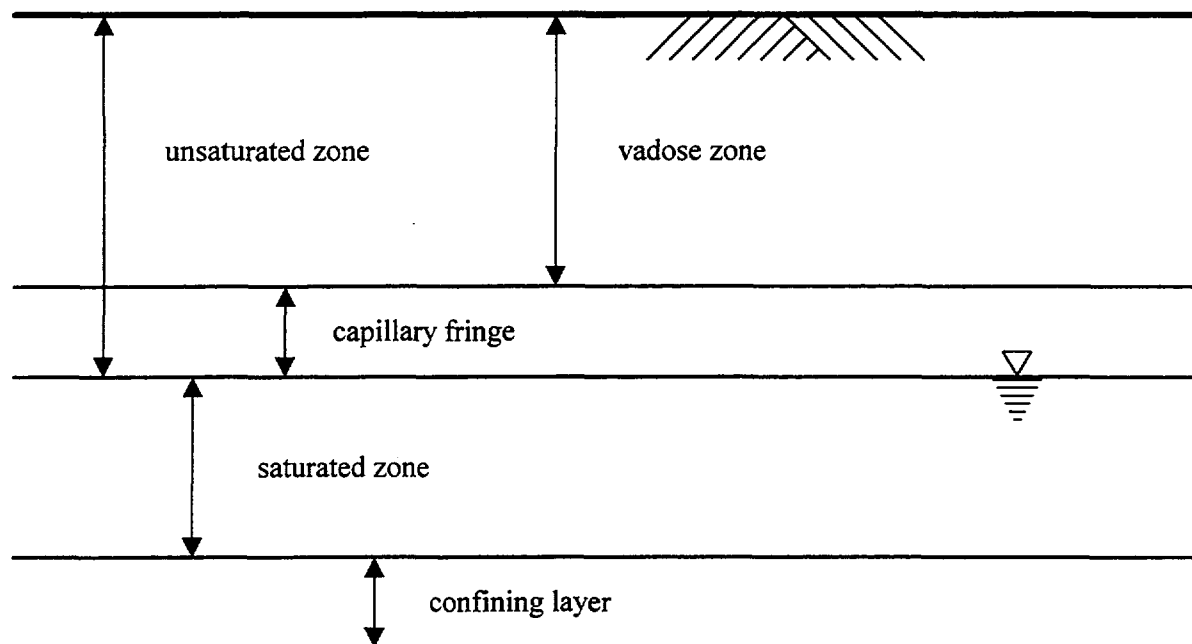


FIG. 1. Generalized Overview of the Subsurface Environment

In summary, this report provides an outline of problems associated with contaminated groundwater and a description of technical approaches and pertinent experience including national contributions and illustrative examples. While it is understood that groundwater as an integral part of the hydrologic cycle is very closely related to surface waters and the meteorologic conditions, it was not within the scope of this report to deal specifically with surface water contamination although many technologies are applicable to both groundwater and surface water. Furthermore, the interrelation between the various elements of contaminated soil, rocks, and groundwater have been elaborated or are only marginally important.

#### 4. NATURE AND EXTENT OF THE PROBLEM

For the purposes of this report, groundwater will be considered as water in the subsurface, in both the unsaturated and saturated zone (see Fig. 1). Any programme intended to assess and remediate contaminated groundwater must begin with the development of a structured conceptual model that embodies geology, hydrogeology, toxicology, radiology, and affected populations.

##### 4.1. GROUNDWATER INTERACTIONS

Groundwaters exist as an integral part of the larger hydrologic cycle of a given region. Interactions between groundwater and surface water bodies (recharge and discharge zones) provide one of the major pathways through which groundwater contaminants interact with humans and the wider terrestrial environment. These interactions can be beneficial by diluting the contaminated groundwater. This can be a major factor in the reduction of the impact of groundwater contamination. Alternatively, contamination may become concentrated in bottom sediments through precipitation and sorption processes, or may be taken up and accumulated in plants and animals. It is also possible that contaminants will be transported some distance from

the point of discharge only to become deposited elsewhere. This deposition usually occurs at an interface of some kind, such as when suspended particulates are deposited when a river flows into a static water body (e.g. a lake). Precipitation and coagulation can, for example, occur at the saline interface within estuaries. Changes in water chemistry can occur downstream in a river system or where two rivers meet. One example would be the anaerobic conditions which may occur when a polluted river flows into a clean river or where effluents such as sewage are discharged into a water course. These changes in chemistry could potentially result in the precipitation of uranium due to redox changes. All of these processes and others (which may be similar, although not described here) can influence the manner in which contaminants interact with man and the environment. As such, these potentially operative processes would be all be considered to a lesser or greater extent in the biospheric modelling section(s) of a risk assessment.

## **4.2. GROUNDWATER CONTAMINATION SCENARIOS**

Examination of various groundwater contamination scenarios is important for assessing the magnitude of the problem and changes expected to occur over time, and for selecting applicable remediation technologies and strategic approaches.

### **4.2.1. Contaminant sources**

Groundwater contamination can arise from:

- (1) the discharge of liquid wastes into infiltration pits or ponds, or into aquifers using injection wells;
- (2) inadvertent releases, spills or leaks of liquid wastes;
- (3) the leaching of contaminants from radioactive material deposited on exposed surfaces by fallout;
- (4) wastes stored or disposed of in surface or subsurface facilities; or
- (5) alterations of groundwater characteristics which may mobilize materials or species previously retained as solid phases or sorbed to the aquifer matrix.

In the first two cases above the liquids are likely to be aqueous. In cases where the liquids are partially or completely composed of immiscible fluids, multiphase flow will have to be considered in assessing the overall extent of the environmental problem. In the case of solid waste sources, contamination with radionuclides can be direct (the contaminants are leached from the stored solid materials or escape from incompletely contained or immobilized liquids) or indirect (contaminants are mobilized by chemical processes in the leachate). Indirect radionuclide mobilization can occur as a result of reactions in the waste that change pH and/or electrical conductivity, increase the concentration of complexing agents or competing ions, or provide colloid forming materials in pore fluids in the waste.

The alteration of groundwater flow through activities such as injection, dewatering, or the construction of barriers to inhibit flow may result in changes in aquifer chemistry. This may

mobilize materials that were previously immobilized as solid phases or by sorption to the aquifer matrix.

#### 4.2.2. Source control and soil remediation

A significant element for the success of any remediation strategy is to decouple/cut-off the source term from the groundwater pathway. In some contamination scenarios, the source may have only occurred over a short time period, such as a one time leak. However, other scenarios may involve continued contaminant source contribution, such as the seepage from an uranium mill tailings or mine debris pile. In scenarios with continued source term contribution to the groundwater pathway, one of the first remedial actions is to remove or decouple the contaminant source. The cleanup of a site will be extended indefinitely if the source to the groundwater is not fully stopped.

#### 4.2.3. Conceptual models

Conceptual models are basic elements of any remediation programme for environmental contamination. Such models may be relatively simple or they could be of a complex mathematical nature, depending on the particular situation at hand. Typically, there will be useful models of important aspects for describing and predicting the movement of contamination and its subsequent effects, such as the geologic, hydrologic, geochemical aspects, etc. These are briefly discussed below.

**Geology:** The conceptual model may be used as the basis for a mathematical model that can be used for treatment system design and performance prediction. The process of constructing a conceptual model usually begins with an understanding of the site's geology. The geological model provides an early indication of the spatial distribution of rock and/or soil properties which are important for describing the site.

**Physical hydrogeology:** Having established a geologic framework, the saturated hydraulic properties of the geologic units must be defined. Figure 1 provides a generalized overview of the subsurface environment.

The intrinsic properties of geologic formations and hydrogeology that influence water movement and contaminant transport are

- (1) permeability,
- (2) effective porosity, and
- (3) hydraulic gradient.

Permeability describes the ability of a material to transmit a fluid, and it is a function of the properties of both the material and the fluid. In the case of water as the fluid of interest, permeability is referred to as hydraulic conductivity, and this is the term that will be applied for this discussion (see below).

The effective porosity describes the space (available volume fraction) involved in fluid transport. In materials that are fractured, porosity is divided into primary (space in the matrix) and secondary (space in the fracture network). For unconsolidated sediments, the effective porosity normally ranges from about 10% to 50%, with lower values applying to materials with a wide range of particle sizes and high degrees of compaction and high porosities in loose, equigranular, sediments. The primary porosity in rock ranges from about 35% for poorly cemented sedimentary

rocks too less than 1% in crystalline rock. Rocks, and some cohesive sediments as well, may also possess a fracture porosity; this is generally less than 10%.

As with porosity, the hydraulic conductivity in fractured materials can be both primary and secondary. Hydraulic conductivities in geologic materials may range more than ten orders of magnitude, and these variations can occur over spatial dimensions of metres or even centimetres.

In addition to specifying the spatial distribution of hydrogeological properties, the distribution of hydraulic heads must be known to determine hydraulic gradients, flow directions, and hence velocities. The boundaries of the flow system should be defined as part of the development of the conceptual model. When contamination is at shallow depth, surface hydrology must be described to provide information on rates of water recharge to the subsurface, locations and rates of groundwater discharge to surface, and in any other features that represent boundaries for the groundwater flow system (e.g. lakes that represent regions of constant or specified water level or head).

Past and existing water use in the study area must be determined. Data from existing water wells and discharge points should be reviewed as part of the information compiled during development of the geologic framework and of the aquifer hydraulics in the study area, and groundwater use will frequently be a key driver or constraint on the remediation programme. Surface water use must also be considered, however, when the effects of the groundwater contamination and remedial actions on residents and ecosystems are being evaluated, and, in extreme cases, surface water additions or extractions may directly affect groundwater conditions.

**Geochemistry:** Information on the distribution of contaminant concentrations in the subsurface is required to evaluate the needs and the design of a groundwater remediation project. In addition, physico-chemical data on other subsurface components are required if a remediation plan is to be fully developed. Information on the natural quality of surface and groundwaters (i.e. background) is needed to evaluate the risks posed by the contamination and to establish remediation targets. Background water quality data should be obtained from samples collected from the aquifer that contains the contamination, and sampling should be undertaken at unaffected locations. Attention should be paid to the geochemistry of the contaminant(s) in the groundwater, both for predicting mobilities and for designing any remediation options.

The requirements for geochemical characterization are not limited to the aqueous phase. Because solid phase interactions are common geochemical processes, the mineralogy of the soil or rock of the system can be an important determinant of the groundwater interactions. Geochemical processes that influence radionuclide behaviour include:

- dissolution;
- precipitation and/or co-precipitation;
- sorption (e.g. ion exchange, chemisorption); and
- biologically-mediated reactions [6, 7].

The extent to which such processes require definition will depend on the remediation options being considered, but the initial assessment of the problem should include some definition of solid solution geochemical processes.

## **Subsurface mobility**

The subsurface mobility of relatively abundant and long lived isotopes will frequently be a key determinant in the need for groundwater remediation to remove them from active transport. Because of the relatively short history of anthropogenic uses and generation of radionuclides, the need for groundwater remediation will usually be restricted to elements that move at velocities that are a significant fraction of the velocity of water movement in the system of interest or concern. Interactions with the phases in the subsurface may reduce the rate of contaminant transport; and because most geologic materials have surfaces that possess a net negative charge, contaminants in cationic form are frequently observed to interact with solid surfaces, at least to some degree. Highly mobile radionuclides are most often those that form neutral or negatively charged species in the subsurface environment, although it should be stressed that specific chemisorptive processes may result in the retardation of even anionic species. The chemical forms that radionuclides may assume in the subsurface environment will be influenced by their form in the waste source, by other materials leached or released from the waste source, and by the geochemistry of downgradient (i.e. downstream) soils and groundwaters. To date, however, experience in most countries and geologic settings has shown that only a few radionuclides have a high probability for significant subsurface mobility; that there are many radionuclides that are only rarely transported rapidly enough to represent candidates for groundwater treatment. The major exception to the above is tritium, which is without exception highly mobile.

### **4.3. EXPOSURE PATHWAYS**

The exposure pathways are the routes by which the radioactive contaminants come into contact with the receptors, i.e. the affected population. Typical exposure pathways which are associated with groundwater contamination include the following:

- direct consumption of contaminated groundwater taken from a well or spring;
- irrigation of agricultural land with contaminated groundwater and the subsequent consumption of food products;
- contamination of surface water resources by groundwater discharge and either consumption, irrigation or recreational use of the contaminated surface water.

### **4.4. HAZARD ASSESSMENT**

The health effects of ionizing radiation are beyond the scope of this document. However, the following brief discussion is presented simply as a means of introducing some basic concepts of hazard assessment to the subject of groundwater remediation.

One criterion needed to determine the importance of groundwater contamination is the hazard that the radioactivity represents to humans or to the environment. Some aspects of this determination are relatively straightforward, such as the hazard stemming from the human consumption of contaminated water, and there may be prescriptive standards for the allowable contaminant concentrations. Health Canada, for example, recommends maximum acceptable concentrations (MACs) for drinking water that are derived from assumptions about the amount of water consumed (e.g. 730 litres per year for an adult) and from certain dose conversion factors (DCFs) which are based on dosimetric and metabolic models for converting unit quantities of activity into unit doses for individual radionuclides. In the case of the Canadian guidelines, the

DCFs are drawn from the National Radiation Protection Board (NRPB). Assessments of the human hazards represented by intake through food pathways, or by external exposure, are not as straightforward, and in many cases may be more significant than those stemming from direct ingestion. Pathways models, and an understanding of the routes of contaminant transport between the source and the receptor (i.e. the receiving population), may be required to make such determinations.

In the past, a standard assumption has been that the protection of human health will ensure the protection of non-human biota, but in recent years this assumption has been challenged by a number of regulatory agencies. There is, however, no well developed protocol for addressing this question. For the purposes of this report, the protection of human health will be assumed to be the objective of remedial actions and, accordingly, the basis for evaluating the options for remediation.

Although the risks to a critical group or population will be the ultimate base for deciding which radionuclides are of greatest concern, there are three criteria that can be used to provide some guidance as to which isotopes are likely to be of concern for groundwater remediation.

These criteria are as follows:

- (1) quantity present (mass or activity),
- (2) half-life, and
- (3) subsurface mobility.

The relative importance of any particular radionuclide in a groundwater contamination situation will generally depend upon these three criteria.

### **Quantity present (mass or activity)**

In spite of their radio toxicity in many cases radionuclides that are minor components in the contaminant source will not be key elements in a groundwater contaminant plume.

### **Half-life**

There will be some cases where the rates of subsurface transport are high, and the distance between a waste source and a point of potential human exposure is very short. In many cases, however, where radionuclides with half-lives of less than several months will decay to concentrations below concern either within the contaminant source itself or along the downgradient transport path (unsaturated or saturated).

### **Subsurface mobility**

The subsurface mobility for a radionuclide of concern at a given site is an important factor in assessing the need for remediation, determining the remedial options to be applied, and so on. A discussion of subsurface mobility was given in section 4.2. In section 4.5.1 below, some brief comments are provided on a number of isotopes that have frequently shown appreciable mobility during subsurface transport.

## 4.5. CONTAMINANTS OF CONCERN

For the most part, the contaminants of concern are radionuclides, although other elements and hazardous materials may be present in the groundwater and subsurface environment which should also be considered in the assessment of remedial options hazards to affected populations, and so on. It is important that every groundwater remediation project be approached in a system sense; i.e. characterization, selection of unit operations, and monitoring must be considered not only for the radionuclides to be removed from the subsurface, but rather all aspects of the entire system. For example, when considering using ion exchange for radionuclide absorption, consideration must be given to all the other substances in the extracted groundwater and how they might consume or foul the ion exchange resin.

### 4.5.1. Radioactive contaminants

The radioactive contaminants associated with a given contamination problem may be quite site specific. The classes of radionuclides that may be encountered include fission products, activation products, uranium and other naturally occurring radionuclides, other man-made radioisotopes, transuranics, tritium, and so on.

#### 4.5.1.1. Moderately and appreciably mobile radionuclides

A number of commonly encountered radionuclides normally exhibit moderate or appreciable mobility for the underground environmental conditions. These include the radioisotopes of strontium, uranium, hydrogen, carbon, cobalt, and iodine.

**Strontium-90:** This abundant fission product has been identified as a key radionuclide in groundwaters that have been contaminated by accidental releases of, for example, nuclear wastes containing fission products, to surface waters or from waste management/disposal areas. Substantial subsurface migration of  $^{90}\text{Sr}$  has been observed at Chernobyl, Hanford, Oak Ridge, and Chalk River, as well as at other sites where fission products have escaped into subsurface systems. Strontium is a member of the alkaline earth family and is normally present in aqueous solutions as a divalent cation. As such, one would anticipate that it would interact with geologic solids, exhibiting sorptive behaviour, and at all of the sites listed above this has indeed occurred with  $^{90}\text{Sr}$ . Sorption occurs to only a limited degree, however, and much of the transfer to aquifer solids is reversible. Thus, strontium has frequently exhibited rates of subsurface movement that are a few per cent of the velocity of the transporting water.

**Uranium:** Uranium ( $^{235}\text{U}$  and  $^{238}\text{U}$ ) contamination of groundwater is most commonly associated with uranium mine and mill tailings, and with wastes arising from fuel refining operations. Uranium is a polyvalent element and, under reducing conditions, its solubility is usually extremely low, being controlled by the precipitation of  $\text{UO}_2$ . Under oxidizing conditions, and where carbonate species are abundant in solution, the anionic uranyl carbonate species, which normally undergoes little interaction with solid phases, may form.

Consequently, uranium migration can be limited in some circumstances and it can be significant in others. It is therefore important to assess the ambient geochemistry when assessing uranium mobility.

TABLE I. OVERVIEW OF THE RELATIVE MOBILITY OF VARIOUS RADIONUCLIDE SPECIES

Species	Half-life [years]	Relative mobility		Note
		Unsaturated zone	Saturated zone	
H <sup>3</sup>	12.3	f	f	
C <sup>14</sup>	5730	f	f	
Co <sup>60</sup>	5.3	m	f	
Sr <sup>90</sup>	29.1	m	m	
Zr <sup>93</sup>	1.5E6	s	s	
Nb <sup>94</sup>	2.03E4	s	s	
Ru <sup>106</sup>	1.02	f	f	
I <sup>129</sup>	1.6E7	f	f	as I <sup>-</sup> or IO <sub>4</sub> <sup>-</sup>
Cs <sup>137</sup>	30.2	s	f	forms a complex in humus
Ra <sup>226</sup>	1.6E3	s	s	cases of groundwater contamination are very rare
Th <sup>232</sup>	1.4E10	s	s	
U <sup>235</sup>	7.03E8	m, s	m, s	mobility depends on the chemical valence
Np <sup>237</sup>	2.1E6	s	s	
U <sup>238</sup>	4.5E9	m, s	m, s	mobility depends on the chemical valence
Pu <sup>238</sup>	87.7	s	s	
Pu <sup>239</sup>	2.4E4	s	s	
Pu <sup>240</sup>	6.56E3	s	s	
Pu <sup>241</sup>	14.3	s	s	
Am <sup>241</sup>	432.2	s	s	

s = slow      m = medium    f = fast.

**Tritium:** Although tritium may be present in wastes that are spilled or disposed of on surface or in the subsurface in a variety of chemical forms, tritiated water is by far the most common compound. Because the tritium is present as water, it is transported in the subsurface at the same rate as all other water in the system. In combination with its high mobility, the half-life of tritium (12.4 years) is long enough that transport over appreciable distances can occur. In heavy water moderated reactors, tritium is a very abundant isotope, and it would be a key radionuclide in any fusion power systems.

**Carbon-14:** The most common mode of  $^{14}\text{C}$  production is by an n-p reaction on  $^{14}\text{N}$ , although it is also a low yield fission product and can also be produced by activation of  $^{13}\text{C}$  or  $^{17}\text{O}$ . One of the most common sources of  $^{14}\text{C}$  in radioactive wastes stems from its use as a tracer for a wide variety of studies of biological systems and of organic compounds. The common inorganic forms of  $^{14}\text{C}$  in radioactive wastes are as carbonates; in acidic media, it can be lost to the atmosphere as  $^{14}\text{CO}_2$ , but in neutral to alkaline systems, the ionic forms of  $\text{H}^{14}\text{CO}_3^-$  or  $^{14}\text{CO}_3^{2-}$  will dominate. These anionic species normally exhibit little in the way of chemical interactions with geologic materials, and they tend to migrate at groundwater velocity, although there has been laboratory evidence for some isotopic exchange with solid carbonates. When present in radio labelled organic compounds, as found in certain waste sources, the radiocarbon may also be transported at, or nearly at, groundwater velocities; this can be result of subsurface degradation which may convert such compounds to inorganic forms (e.g. mobile carbonates).

**Cobalt-60:** Although cobalt is a transition metal and would normally be expected to be present as a cation in aqueous solutions (and hence interact appreciably with geologic solids), experience at Chalk River [see Annex II] has shown that an appreciable fraction of the  $^{60}\text{Co}$  released from an aqueous wastewater infiltration pit is present as an anionic complex. The evidence also indicated that this is due to presence of an organic complexing agent that is naturally present in the aquifer. This complexed  $^{60}\text{Co}$  is subsequently transported at rates that approach or equal those of water movement through the subsurface. High  $^{60}\text{Co}$  mobility was also observed downgradient of a solid waste management area at Chalk River; for that case, there is no information of the  $^{60}\text{Co}$  form. Anionically complexed cobalt was also observed in experimental studies following tracer injections into a sand aquifer in Great Britain. Rapid movement of  $^{60}\text{Co}$  has not been observed in the surficial sands at Chernobyl, however, so cobalt complexation does not appear to be a universal phenomenon.

**Iodine:** In most cases, iodine can be expected to be present as an anionic species (iodide or iodate) and hence to be highly mobile. The half-lives of many of the common radioactive isotopes are too short to be groundwater contaminants of concern. Iodine-129 can be of potential concern, but its volatility during spent fuel reprocessing operations (which are the source of most of this isotope) has meant that it has either been dispersed to the atmosphere or, in more modern facilities, it has been collected by stack gas filtration and the spent filters have been stored with care.

#### *4.5.1.2. Other radionuclides*

In general, any other radionuclides that form anionic species under aquifer conditions can be expected to migrate at velocities approaching those of the water movement in the underground system. Ruthenium and antimony isotopes frequently exhibit rapid migration, or at least do so for a fraction of their inventory; but the half-lives of the common radioisotopes of these elements is one year or less, making them less likely to be of substantial concern.

**"Immobile" elements:** The quotation marks are intended to stress the qualified nature of this list, because there are almost always some cases where chemical conditions are so extreme that at least limited transport has been observed.

**Thorium and radium:** Among the naturally occurring radioisotopes, cases of groundwater contamination by radium and thorium are very rare. Normally, these elements are present in the form of insoluble compounds. Extremely acidic conditions have been known to mobilize them,

but reductions to concentrations likely to result in satisfactory water quality can be achieved by remediating (neutralizing) the groundwater's acidity.

**Caesium:** Among anthropogenic radioisotopes, one of the best examples of an element that very rarely exhibits appreciable subsurface mobility is caesium. This is evident by the prolonged surface retention of  $^{137}\text{Cs}$  deposited from atmospheric weapons tests over 30 years ago, and by the continued contamination of surface soils by radio caesium released during the Chernobyl incident. A section in Annex IV notes one study that showed  $^{137}\text{Cs}$  was rapidly fixed by clay minerals and organic matter present in soil and, as a result, only about 1–10% remained in a mobile form.

**Niobium and Zirconium:** Long-lived radioisotopes of these two elements are generated by neutron activation of reactor components. Because they are most common in materials that were designed to be inherently very resistant to corrosion, information on their behaviour in the subsurface is limited. Rapid and strong sorption to a wide variety of geologic materials appears to be the rule rather than the exception, however.

**Plutonium:** Apart from evidence for a very limited colloid formation under some geochemical conditions, the subsurface (and general environmental) mobility of plutonium appears to be extremely limited due to its normally being present in the form of highly insoluble compounds. The high toxicity assigned to plutonium, however, offsets the reduced risks due to the low mobility and makes it a high risk whenever it exists in the subsurface.

**Indeterminant elements:** The comments on mobility given above are intended to be used as guidelines rather than as absolute criteria. Certainly, for elements other than those listed, mobility should be assessed on a case by case basis.

#### 4.5.2. Non-radioactive contaminants

Wastes containing both radioactive materials and other hazardous agents are sometimes referred to as mixed wastes (e.g. as in the United States of America) when the different constituents are regulated by different national authorities, etc. Mixtures of radioactive and hazardous material wastes are commonly encountered, especially in older storage or disposal facilities, where the potential difficulties arising from the co-disposal of radioactive and hazardous materials was not widely recognized in the past. In some cases, radioactive and chemically or biologically hazardous materials arose jointly from a process (e.g. where organic solvents or mercury were part of spent fuel reprocessing operations). Suspect wastes (e.g. solvents and lubricants from machine shops handling radioactive materials or components) were frequently added to radioactive waste streams on the basis that radioactive contamination might have been present. In other cases, hazardous materials may have been added to radioactive waste streams simply because this was a convenient means of disposing of such wastes. Regardless of the reasons for their presence, there are a number of cases where it is the non-radioactive contaminants that have become the primary target for groundwater remediation programmes. The presence of such contaminants can have influence on behaviour of radionuclides (e.g. organics may tend to mobilize otherwise immobile radionuclides in the underground environment). Groundwater contamination with trichloroethene (TCE) at the Idaho National Engineering and Environmental Laboratory (INEEL) is a good example of a case where the non-radioactive contaminant has been the target of groundwater remediation.

Uranium mine and mill tailings present another case where co-contaminants may dominate the remediation requirements and also, importantly, control the radionuclide mobilities. Many uranium ores contain substantial quantities of sulphides that have become waste materials from the milling process. The finely divided sulphide minerals, when in contact with atmospheric oxygen in unsaturated tailings, rapidly generate large quantities of soluble sulphates, ferrous iron, and hydrogen ions. The resultant conditions of low pH and low redox potential in the high ionic strength solutions resulting from the sulphide oxidation can effectively mobilize residual uranium, radium, and in some cases even thorium from the mill tailings. Treatments to raise the pH and reduce the sulphate and iron concentrations in tailings leachate will generally provide at least partial, and possibly an adequate or acceptable level of removal of the dissolved radionuclides into a non-dissolved phase, effectively removing the radionuclides from the advective groundwater flow.

Other contaminants that may occur in groundwater could be by-products of mineral processing. Common constituents such as nitrates, sulfates, and ammonia are often “identified with” radionuclides and may require remediation as part of an environmental restoration programme.

## **5. A GENERAL APPROACH TO THE MANAGEMENT AND SELECTION OF REMEDIAL ACTIONS**

In this section, a general approach for the management and selection of remedial options for radioactively contaminated groundwaters is discussed. The methodology consists of a phased strategy to allow for the most cost effective and environmentally sound remedial approach. Its application allows all of the decisions and choices made during the management and selection process to be clearly seen and examined. This is an essential part of the process, and it can be particularly important, for example, when communicating with affected parties (e.g. members of the public) and regulators.

The initial discovery of radioactive contamination in the groundwater system and the decision to begin site investigation can result from various factors. For example, a site operator may become aware that the groundwater is contaminated and then must decide what action to take to prevent it from leaving the site boundary. Another possibility is that the problem may be discovered through epidemiological studies identifying health problems arising from the utilization of contaminated groundwater (though such a case has never occurred for radioactively contaminated groundwater). In the former case, there might be ample time to plan a complex strategy, whereas in the second case an immediate action would obviously be required. In situations where immediate action is indicated (e.g. to prevent health risks), it should be stressed that hasty decisions regarding remediation may not always be most appropriate. A more satisfactory approach might be to alleviate the health risk by institutional control (e.g. providing alternative water supplies); this would then allow time for a more structured approach to making decisions regarding the remedial action.

A phased approach can be particularly useful to allow for the most cost effective and environmentally sound disposition of a contaminated site. The phased approach will generally consist of the following elements:

- assessment of the existing information and data (scoping analysis);
- initial planning and decision making to consider what further action is required;
- selection of site characterization or monitoring requirements;
- assessment of remediation technologies for appropriate application to the problems at hand; and
- selection of remediation strategy to be employed.

## 5.1. SCOPING ANALYSIS

The logical approach to assessing a contaminated site is to identify the source, the hydrogeologic setting, and the potential receptors (i.e. the affected population) by the following:

- compile, review and analyse existing data and information;
- identify the contamination and its source;
- describe the hydrogeological system, develop a useful conceptual model; and
- identify the potential affected population and their points of contact with the contaminated groundwater

This conceptual understanding is based on site history, background information, previous investigations, known and suspected sources of contamination, processes used which generated the waste, routes of migration, and potential human and environmental receptors.

The history and background of the site should be evaluated to determine if any previous activities took place there that could potentially impact decisions made concerning characterization or remediation of the site. Such considerations could include previous industrial, commercial, agricultural or military uses.

A literature search or interviews with persons with historical knowledge should be performed to acquire a knowledge base on how the site became contaminated, the period of time which the contamination was released to the environment, release mechanisms, the types and quantities of contamination, and so on.

The existing geologic and hydrologic data for the site must be evaluated to help determine the fate and transport of the contaminants. Information regarding geologic formations and hydrologic parameters may be obtained through the description of sediment samples collected during drilling of production wells, irrigation wells or any other soil borings that may have taken place at the site. It should be recognized that the quality assurance of data collected in this manner may be suspect and therefore conclusions based on the data should be treated with caution.

At this stage, some modelling may take place. The complexity of the modelling should reflect the quality and quantity of site data available. This modelling may include groundwater

flow representations and contaminant transport simulations in association with risk assessment. As a first pass, relatively simple calculations of radiation dose and risk to individuals and populations may be made using assumptions that are conservative, resulting in estimates for dose and risk that are maximums.

## **5.2. EARLY DECISIONS REGARDING FURTHER ACTION**

After all or most of the existing data and information on the contaminated site have been collected and analysed, a determination for further action should be made. The alternatives to be considered may include: (1) no further action is required; (2) no further action is required other than to further monitor the contaminant plume; (3) more data are needed to make a decision; or (4) a remedial action should be undertaken.

### **5.2.1. No further action needed**

A decision of no further action can be made if it is determined that there is no contamination or that the extent of the contamination is below an acceptable risk level and below the regulatory requirements of concentration or radiological dose.

### **5.2.2. Further monitoring of contaminant plume is required**

Although no further action (e.g. remedial action) may be required, it might still be necessary or advisable to continue to monitor the site to ensure that the initial assessment of the situation is correct. For example, this could be outcome when it appears that natural processes such as dispersion and radioactive decay would result in the contamination having no significant impact on the receptors (i.e. affected population). Continued monitoring would allow the assumptions regarding movement of the groundwater contaminant to be routinely checked. In addition, continued monitoring could provide comforting reassurance to affected parties such as the local population.

### **5.2.3. Insufficient data exist to make a decision**

Following the assessment of existing data and information, it could develop that there are insufficient data to make an informed decision regarding the possibility or advisability of remedial action. Under such a circumstance, it is common that a site characterization programme be implemented to fill the identified gaps in information and data. If there is a decision to collect additional data, the data collection objectives should be clearly identified and used in designing the site characterization programme.

### **5.2.4. Remedial action is required**

In some cases, there will be wholly sufficient data and information regarding a site and the groundwater contamination problem to conclude that remedial action is required. In such a case, the strategy will advance to the technologies evaluation and remedial design phases.

### 5.3. PUBLIC INVOLVEMENT

One factor to consider when evaluating technologies or screening for remedial alternatives is involvement by affected parties and the general public. The public's perception of risk due to radiation exposure may be substantial enough to warrant a more stringent remedial goal for a contaminant in groundwater. It is important to involve the public and all affected parties in the decision making process.

### 5.4. ESTABLISHMENT OF REMEDIATION GOALS

Preliminary remediation goals are site specific. The initial remediation objectives should be established on the basis of the nature and extent of the contamination, the water resources that are currently or potentially threatened, and the potential for human and environmental exposure. These quantitative goals should define the extent of cleanup that is required to satisfy the established objectives. They include the required cleanup levels and the restoration time frame. A comprehensive discussion of radiation protection principles as applicable to restoration of contaminated sites is given in [6]. Clean up levels of contaminants are typically based on either drinking water standards or on excess lifetime cancer risk levels.

Past practices around the world have used extremely conservative scenarios for determining the risks of ionizing radiation to human health. As a result, remedial activities have been extremely costly. Recently, a philosophy of using more realistic risk scenarios appears to be becoming acceptable. In some cases, remediation has been avoided altogether, with only the cost of monitoring remaining. This strategy has reduced the cost while continuing to adequately protect human health. It is recommended that when selecting and analysing the risk scenarios, the expected land use, impacts on affected parties and environment, and the future groundwater needs should all be evaluated. A realistic scenario can then be developed which would allow for a more cost effective remediation while still ensuring the safety of the public. Obviously, the effectiveness and reliability of institutional controls may affect these decisions.

Risk assessment methods may be used, coupled with regulatory requirements, to determine achievable goals. The beneficial use of an aquifer must also be considered. Water which does not meet the required standards for domestic use may still be useful for agricultural or industrial purposes.

Finally, the potential effects on environmental receptors such as plant and animal species at or near the site may also affect the remediation goals.

### 5.5. SITE CHARACTERIZATION

Site characterization activities should take place if more data are needed to evaluate risks associated with the contaminated site or to understand the parameters necessary for selecting an appropriate remedial technology. The approach to characterizing the site is to select data collection objectives with an understanding of the associated uncertainties.

### **5.5.1. Data collection objectives**

A site specific data collection strategy should be organized to provide sufficient data to formulate a conceptual model of the contaminated site. The data collection activities should focus on understanding of the following:

- source term;
- geology (i.e. formations, grain size, plasticity, moisture content, density, mineralogy);
- hydrogeology, aquifer properties;
- geochemistry;
- nature and extent of contaminant plume; and
- exposure pathways.

### **5.5.2. Identification of uncertainties**

Inherent uncertainties are often encountered in characterizing contaminated sites. Many of these uncertainties arise from the necessity of characterizing the inherent heterogeneity of the aquifer with a limited number of sample points. Aquifer heterogeneity should be considered when developing a strategy for site characterization.

One approach to identifying and addressing aquifer system uncertainties is based on using the preliminary site conceptual model to identify the remedial strategy with the highest probability of success. At this stage of the decision making process, the probability of success is based on “most probable site conditions.” Acknowledging that site conditions have inherent uncertainties, reasonable variations from the “most probable conditions” are identified early, and contingency remedial action strategy alternatives are not ruled out.

To better plan site characterization activities, sensitivity analysis is often used for defining the importance of parameter input to predicted cost and remedial action performance. Data worthiness (e.g. adequacy or worth) evaluations are also becoming more popular for decision makers in their understanding of the relationship between uncertainty and sensitivity of site conditions, and remedial cost and performance. The observational method is an effective and economical means to manage uncertainties associated with remediating contaminated groundwater.

### **5.5.3. Data quality objectives**

Using the DQO process will help to ensure that when a data collection endeavour has been completed it will have accomplished two goals:

- Provided sufficient data to make required decisions within a reasonable uncertainty
- Collected only the minimum amount of necessary data.

The DQO Process embodies both of these two main goals and it is difficult to separate which is the more important or which drives the other. For example, the DQO process will strive to provide the least expensive data collection scheme, but not at the price of providing answers that have too much uncertainty.

DQOs are intended to ensure that the data generated during site characterization activities are adequate to support management decisions. A clear definition of the objectives and the method by which decisions will be made must be established early in the scoping process. DQOs are determined based on the end uses of the data to be collected. The level of detail and data quality needed will vary based on the intended use of the data. DQOs should be reviewed throughout the characterization activity and adjusted based on new available information as appropriate.

#### **5.5.4. Data analysis**

All of the data collected during the scoping and characterization phases of the project should be analysed with the results formally documented. This activity should be co-ordinated with the risk assessment and modelling personnel to provide for a more efficient use of the data. All decisions should be documented with an explanation of the logic used to arrive at the given conclusion. This includes decisions made as a result of scoping, establishment of preliminary remediation goals, data collection objectives, data quality objectives and screening, and the selection of the remediation technologies.

### **5.6. DEVELOPMENT AND SCREENING ALTERNATIVES**

This section will describe in general terms how remedial alternatives are developed. A few of the guiding principles for developing alternatives include technical practicability, cost/benefit analysis, schedule for implementing and completing the remedial action.

The nature of the source, the size of the plume, and the transmissivity of the aquifer also will directly affect the effectiveness of the remediation whether it be an in situ or ex situ treatment. Most groundwater technologies currently available are expensive to implement and take long periods of time to complete. Continued research is ongoing world wide to develop new techniques for in situ and ex situ remediation. A general list and description of these technologies can be found in Section 5 of this report. Care should be taken to evaluate the success or failure of the technologies which have been developed and to compare the site specific characteristics against the test site to determine the viability at a particular site. Critical parameters of the technology being evaluated should be identified for comparing the viability of success at each site. For example, a technology may work quite well at a site with alluvial sands, but not at all at a site with fractured rock.

Based on the analysis performed on the site characterization data, a list of alternatives and technologies can now be compiled. A screening process should determine if an active remediation is required or if a passive alternative (institutional controls, no action, monitoring, etc.) is desired. If an active remediation option is chosen, a detailed analysis of technologies should be performed.

### **5.7. INSTITUTIONAL CONTROLS**

Institutional controls may be implemented to reduce or eliminate potential threat of exposure to human health. The following kinds of institutional controls have been established in some countries and may be considered to prevent exposure to contaminated groundwater:

- Regulatory restrictions on construction and use of private water wells, such as well construction permits and water quality certifications;
- Acquisition of property by the government from private entities;
- Exercise of regulatory and police powers by governments, such as zoning and issuance of administrative orders;
- Restrictions on property transactions, including negative covenants and easements;
- Non-enforceable controls, such as well use advisories and deed notices;
- Relocation of affected populations (in extreme cases).

The effectiveness and reliability of these controls should be evaluated when determining whether rapid remediation is warranted. If there is adequate certainty that institutional controls will be effective and reliable, there is more flexibility to select a response action that has a longer restoration time frame or a determination that no remedial action is required [10].

## 5.8. ANALYSIS AND DESIGN OF PREFERRED ALTERNATIVES

During the detailed analysis, remedial alternatives that have been retained from the alternative development phase are analysed against a number of evaluation criteria. The purpose of the detailed analysis is to compare alternatives so that the remedy that offers the most favourable balance among a set of criteria can be selected. The analysis of a remedial action for groundwater can be made on the basis of the following evaluation criteria:

- Overall protection of human health and the environment;
- Compliance with applicable regulations;
- Long term effectiveness and permanence;
- Reduction of toxicity, mobility, or volume;
- Short term effectiveness;
- Implement ability;
- Cost;
- Community or government acceptance;
- Final disposal of residues.

Other criteria may also be established based on site specific conditions. A narrative discussion and summary table should be prepared for each part of the detailed analysis to provide a historical paper documenting the decision process.

## 5.9. IMPLEMENTATION ACTION AND PERFORMANCE ASSESSMENT

Performance evaluations of the full scale remedial action, based on monitoring data, are conducted periodically to compare actual performance to expected performance. The performance monitoring should be designed to provide information as such, but not limited to the following:

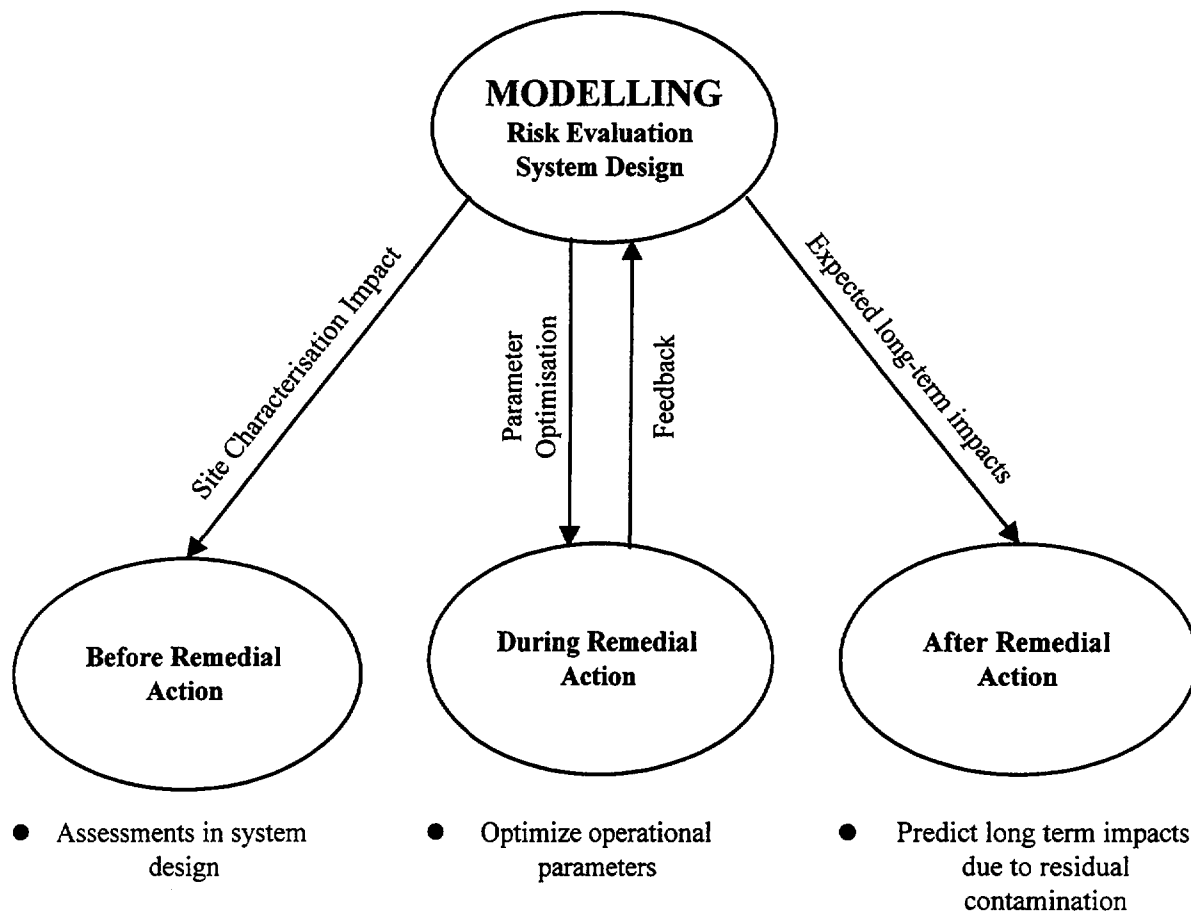
- Horizontal and vertical extent of the plume and contaminant concentration gradients, including a mass balance calculation;
- Rate and direction of contaminant migration;
- Changes in contaminant concentrations or distribution over time;
- Rates of contaminant mass removal and transition from advective removal to diffusion rate limited removal;
- Effects of hydrological events, such as above average rainfall, on contaminant mass removal and changes to groundwater flow;
- Calibration of model based on actual results and effects of changes of operational parameters to model predictions;
- Effects on regional groundwater levels and the resulting impacts;
- Effects of reducing or limiting surface recharge (if applicable);
- Effects of re-injection (if applicable);
- Effects of any modifications to the original remedial action; and
- Other environmental effects of remedial action, such as saltwater intrusion, land subsidence, and effects on wetlands or other sensitive habitats.

The frequency and duration of performance evaluations should be determined by site specific conditions. Conducting performance evaluations and modifying remedial actions is part of a flexible approach to attaining remedial action goals. Decisions should be verified or modified during remediation to improve a remedy's performance and ensure protection of human health and the environment.

The performance assessment may provide information that can be used to determine whether the remediation goals are being met, have been achieved or, in some cases, are technically impracticable to achieve in a reasonable time.

## **6. MODELLING AND ASSESSMENT**

This section discusses the role of modelling in groundwater remediation process and gives an overview of the general principles of model application to remedial analyses (see Figure 2). This includes the application of modelling to evaluate the associated risks before, during and after the implementation of a remedial action.



*FIG. 2. General principles of model application to remedial analysis and design.*

## 6.1. THE APPLICATION OF MODELLING IN THE GROUNDWATER REMEDIAL PROCESS

The overall objective of modelling is to provide the basis for making well-founded decisions on possible groundwater remedial actions. It is generally used to complement other decision making processes. Modelling can be used to develop and support the following:

- understanding of the role and behaviour of the hydrologic system;
- understanding of the groundwater pathway(s);
- assessment of contaminant transport and geochemical processes;
- evaluation of health risks, with and without corrective actions;
- evaluation of remediation techniques, including their effectiveness and cost benefits; and
- evaluation and prediction of post remediation or long term results.

Figure 2 shows general principles of model application to remedial analyses and design.

In addition, modelling can be used as a management tool to organize and prioritize data collection; to analyse results and make predictions; and to assist analysts in the improvement of their understanding of the factors controlling groundwater flow and transport [9, 10].

The important application of modelling is to assess long-term transport and fate of contaminants of concern in hydrogeological environment, and to predict concentrations of contaminants at exposure points, in order to evaluate health-risk bases for remedial actions. Once contaminant concentrations at receptors (i.e. in contact with the affected population) are assessed, the next step is to calculate doses/risks from exposure to contaminated water. This may be accomplished using relevant risk assessment methodologies, ranging in complexity from simple concentration-dose conversion factors to more sophisticated approaches.

Other applications include: evaluating the expected performance of remedial actions; elucidating the control of specific processes on groundwater systems and contaminant behaviour (sensitivity analysis); and the indirect estimation of hydrogeological and geochemical parameters using historical observation data (so called inverse modelling).

## 6.2. STEPWISE IMPLEMENTATION OF THE MODELLING PROCESS

Modelling should be seen as an evolving, iterative process which reflects the development and understanding of the site, and is flexible enough to continuously incorporate new data. Modelling of the groundwater pathway may typically involve several important steps [9, 11], as follows:

- clear definition of modelling objectives;
- development of conceptual model(s) of the hydrogeological system;
- compiling/assembling of hydrogeological and geochemical data (this, in itself, may involve a simplified level of modelling, e.g. the determination of hydraulic conductivity from aquifer pumping tests would typically involve ‘type curve’ matching);
- formulation of mathematical model(s) of groundwater flow and contaminant transport processes;
- selection or development of an appropriate analytical/numerical model(s);
- calibrating model(s) using field observations and data;
- applying the model(s) in a predictive manner; and
- comparing predictions against observations.

The above list implicitly assume feedback loops: many of the steps have to be repeated as new information and data are collected.

It is important to clearly define the objectives of the modelling process. These objectives will reflect the ultimate goal of remediation, but may reflect intermediate goals as well.

A conceptual model is a hypothesis or representation as to how a system or process operates. Before a meaningful model can be developed, a sufficient understanding of the site is required. The physical processes controlling groundwater flow and transport must be identified. This step relies largely on professional judgment. Therefore, it is important that the analyst has a good understanding of the basic hydrogeologic, physical, and geochemical processes [9, 10, 12]. The mathematical model describes relationships between parameters and the governing processes. The selection of a numerical model should encompass both the conceptual model and the corresponding mathematical description of the system. The types of software used (to embody the mathematical description) generally reflect the objectives of the modelling, the available data, the experience of the modeller, and the available computational facilities. Relatively simple models may be used in the early or planning phase of remedial design. As more data become available through the site characterization phase, and as a better understanding of the hydrogeological system is developed, somewhat more sophisticated, data intensive models can be utilized, as appropriate. A wide range of public domain and commercially available computer codes varying in complexity is currently available to model users [12, 13].

Parameters used in numerical models may be derived from a combination of site specific data, relevant published literature, historical information, and expert judgment. The predictive capacity of the models are crucially dependent on adequate input parameters. The general practice is to refine estimates of uncertain parameters for the purpose of the model calibration phase to match observed (i.e. actual) data as they are obtained. It is important to build confidence in the parameter estimates, to the degree possible, using data from laboratory and field studies. This empirical information is crucial for calibrating and refining the model and making the model a useful tool for use in remediation system design and performance optimization.

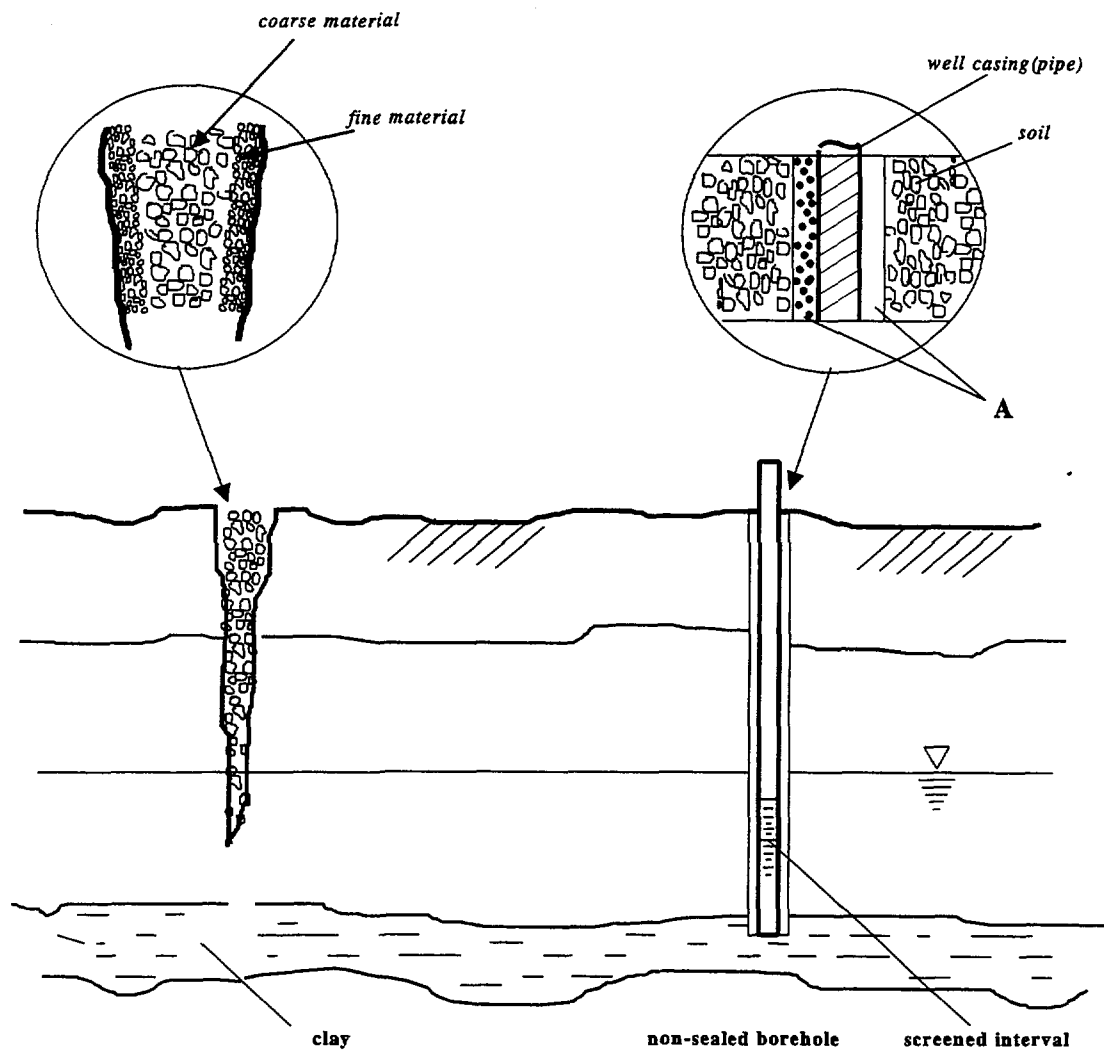
Rapid advances in modelling techniques and computing power have resulted in more sophisticated models and complex approaches to the evaluation of the groundwater pathway. It is important that the model assumptions, input parameters and the modelling results are carefully and systematically documented, both for quality assurance purposes and for clear presentation to decision makers and other interested parties [14].

### 6.3. MODELLING TECHNIQUES AND APPROACHES

The selection of the modelling approach to a given contamination problem should reflect both the objectives and particular phase of the assessment and remediation process. Two general groundwater modelling approaches [9] can be adopted, as follows:

- seek analytical solutions; or
- seek numerical solutions.

Analytical solutions are useful in the preliminary assessment of the hydrogeological system in the absence of significant amounts of data. The advantages of analytical solutions (i.e. solutions described by explicit analytical formulas) are simplicity and computational efficacy. The general shortcoming of analytical models is their simplistic representation of the hydrogeological system (e.g. rather simple assumptions of homogeneity of subsurface environment, steady state flow, one-dimensional transport, etc. may be used). Because of the screening application to which model predictions may be fit, and the fairly simple input data requirements, the analytical approach is often most suitable in the scoping phase of remedial



**A:** Annulus of borehole containing coarse material or no material. Much higher hydraulic conductivity than surrounding soils, resulting in a preferential pathway for contaminant flow.

*FIG. 3. Graphical representation of a clastic dyke (not to scale) and a non-sealed borehole.*

assessment. There are two major types of numerical modelling methods, that is, the finite difference method and the finite element method. Both methods are powerful modelling techniques, used to solve groundwater flow and contaminant transport problems in complex flow geometries. The finite difference method is more conceptually straightforward and physically based; however, the finite element method has proven to have greater flexibility in treatment of complex geometry.

For modelling to be used with confidence at the detailed assessment phase of remedial analysis requires significant quantities of site specific data. There are a number of methods used to model groundwater flow and contaminant transport. When interpreting the groundwater flow path, particle tracking methods are often used; these can give useful information concerning the travel time to receptors (i.e. the affected population) and the effectiveness of a hydraulic containment scheme [15]. Advanced modelling approaches are based on combining solute transport codes with geochemical thermodynamic models for predicting the speciation of contaminants. However, this is still an area of active research and not really a well established

modelling technique. Significant progress has been made in modelling of two and three dimensional saturated and unsaturated flow in porous and fractured geological media. More efficient numerical techniques and significant advances in computing power has opened up opportunities to increase the complexity of modelling; however, this complexity must be justified and underpinned by appropriate

Off the shelf groundwater flow and contaminant transport software will usually incorporate the processes of advection, diffusion, dispersion, equilibrium sorption, and radioactive decay. These may be steady state or transient. Pertinent modelling areas of active research include the flow in fractured media; multiphase flow; multi-species flow with chemical interactions; kinetically limited sorption/de-sorption processes; colloidal transport and the facilitated transport of complexes. Assessment of these process may require development of research-level models and software, and generally requires a high level of scientific expertise of the modeller [11].

#### 6.4. PARAMETER UNCERTAINTIES

Uncertainties are quite inherent in hydrogeological systems. They are present in the definition and nature of geological boundaries of the site, hydrogeological and geochemical parameters, and the spatial distribution of contaminants in the subsurface, etc. Parameter uncertainties may have profound impact on simulation results, and on remedial analysis as a whole. Therefore, uncertainties require a careful treatment in groundwater remedial modelling studies.

There are a number of approaches for dealing with uncertainty in hydrogeological analysis [9, 16], including the following:

- conservative approach;
- deterministic simulation with sensitivity analysis; and
- geo-statistic simulation.

A conservative approach attempts to set bounds on input model parameters, to establish bounds on output results, rather than realistically evaluate the behaviour of the simulated system. An example of this is the so called "worst case" scenario, in which the input parameters are assigned extreme values to estimate the maximum possible contaminant concentrations at receptors (i.e. exposure points for the affected population). Conservative analyses may be justified in the scoping phase of the remedial design. However, much more caution is required in detailed remedial assessment phase because unrealistically conservative impact assessment may result in unnecessarily high cleanup costs. It is recommended, that remedial assessments utilize more sophisticated techniques that properly address the issues of uncertainty.

The deterministic approach uses a base-case simulation (model) with a set of "realistic" or "best guess" parametric values. This is complemented by the application of sensitivity analysis in which the uncertainties in the input parameters can be accounted for in a systematic fashion. Sensitivity analysis can be used to determine which of the model parameters have the greatest impact on the performance of remedial actions. The results of a sensitivity study can be used to guide the site characterization activities, including prioritization of the data collection process..

Geo-statistical methods can embody the uncertainty in the input parameters in terms of probability distribution functions. These uncertainties can be propagated through the

hydrogeological simulation process. A frequently used approach to uncertainty propagation is Monte Carlo technique. This approach requires a large number of models to be simulated from sampled input parameters. The results of Monte Carlo simulations provide confidence intervals for the possible outcomes of remediation. Hence, this can provide an estimate of the probability that a given remedial action meets the design targets.

## 6.5. COST-BENEFIT ANALYSIS AND DATA WORTHINESS

A formalized methodology has been developed during the last decade for choosing preferred ('best') management alternatives for hydrogeological projects with due consideration for the various uncertainties. This approach appears to hold promise for remedial projects involving the cleanup of contaminated groundwater [16–19]. The decision of remediation alternatives is based on economic analysis, taking into account the costs and benefits of each alternative, and associated risks. The risk in this case is defined to be the *probability of remedial design failure* multiplied by the *monetary consequences of failure*. The probability of remedial design failure arises due to uncertainties in the expected performance of the groundwater remediation alternatives. In hydrogeological applications, such uncertainties and risks are often relatively high.

The cost benefit methodology involves the coupling of three separate models: (1) a decision model based on a risk–cost–benefit objective function, (2) a hydrogeological simulation model, and (3) a parameter uncertainty model. This can be carried out in a Bayesian framework in which additional site characterization data and remedial system performance data can be incorporated. An example of the framework of hydrogeological decision analysis methodology is given in [16]. The risk–cost–benefit analysis has been successfully applied recently to a number of “real-world” problems of radioactively contaminated groundwater in need of remediation [20, 21].

A feature of this methodology is the ability to assess the worthiness or adequacy of proposed site characterization and data collection programmes prior to their actual implementation. The issue is of particular importance in view of the high costs of data collection at groundwater contamination sites, which may not be cost effective. The value of obtaining additional data (data worthiness or adequacy) may be assessed by comparing the cost of additional data collection versus the expected value of risk reduction that would be provided by the further effort. The concepts and generic examples of data worthiness (or “data worth”) analysis are presented in [19, 22].

The attractiveness of the risk-cost benefit analysis methodology is that it enables decision-makers to see a coherent “big picture” at complex groundwater contamination sites by integrating economical considerations, technical aspects and uncertain site conditions. It provides a methodology of documenting the reasoning behind remedial decisions, and also can be an important tool for communication [21].

## 6.6. LIMITATIONS OF GROUNDWATER MODELLING

Limitations of groundwater modelling are due particularly to the complexity of the hydrogeological environment and to a lack of understanding of important physical and chemical processes that can influence contaminant transport in the subsurface (e.g. transport by colloidal

forms, etc.). Significant modelling difficulties can arise due to the heterogeneity of physical and geochemical properties of natural rocks and soils (which may result in preferential flow and transport processes). It is often impossible to characterize geological heterogeneity on a field scale with a degree of detail needed for adequate modelling [23].

For example, a recent extensive modelling effort to assess the long term fate and transport of radionuclides in the 200 area plateau of the Hanford Site determined that there was uncertainty in the analysis from the conceptual and numerical work performed for the vadose zone. It was concluded that, although many factors were considered and accounted for, the model results were limited because the model does not include preferential pathways such as clastic dykes and unsealed well bores (see Figure 3) [24].

In addition, long term predictions may be quite uncertain due to possible future changes in stresses on hydrogeological system as a result of natural or anthropogenic factors (e.g. climate changes; changes induced by industrial activities; etc.). Historical changes in the hydrogeological system are often not accurately known, which makes it difficult to obtain a reliable calibration of the groundwater model.

Groundwater modelling can be most effectively used if it is 'fit for purpose' or 'tailored to need'. In the early phases of site characterization and remediation design evaluation, the models are generally simple and the expectation of their predictive capacity is low. As the conceptual model and parameters are further developed, confidence in the modelling results will improve. As a consequence, the uncertainties in the modelling can be better addressed and more properly estimated. The model assumptions and predictions need to be continuously checked and refined using observed results (i.e. actual data and measures of remedial system performance). It is desirable that groundwater model predictions are always accompanied by some indication of their reliability.

## **7. TECHNOLOGIES FOR GROUNDWATER REMEDIATION**

There is a vast array of technologies currently available for the remediation of contaminated surface and groundwater. Many of these techniques were not, however, developed with radioactive contamination in mind. This section will outline and briefly describe those technologies which are directly applicable to the remediation of radioactively contaminated groundwater. Specifically, the review given here will concentrate on those technologies which are able to remove radioactive contaminants from groundwater or retard their migration. Consideration will be given to other contaminants, such as organic compound and heavy metals, where they might interfere with the efficacy of a specific technology, or where there is the opportunity for technology transfer. To make this review as comprehensive as possible, and to some extent allow for future developments, those technologies still at the research and development stage will be discussed along side those at pilot and field scale. Where possible and if available, the current stage of a technology's application will be indicated in the text with reference to specific site applications.

The basic nature of radioactive contaminants makes them unique when compared to other contaminants. Firstly, their radioactivity (i.e. radiation hazards) means that they may be considered a risk sometimes at concentrations lower than toxic elements (e.g. arsenic or heavy metals) would be. However, unlike these elements, radioactive contaminants are subject to

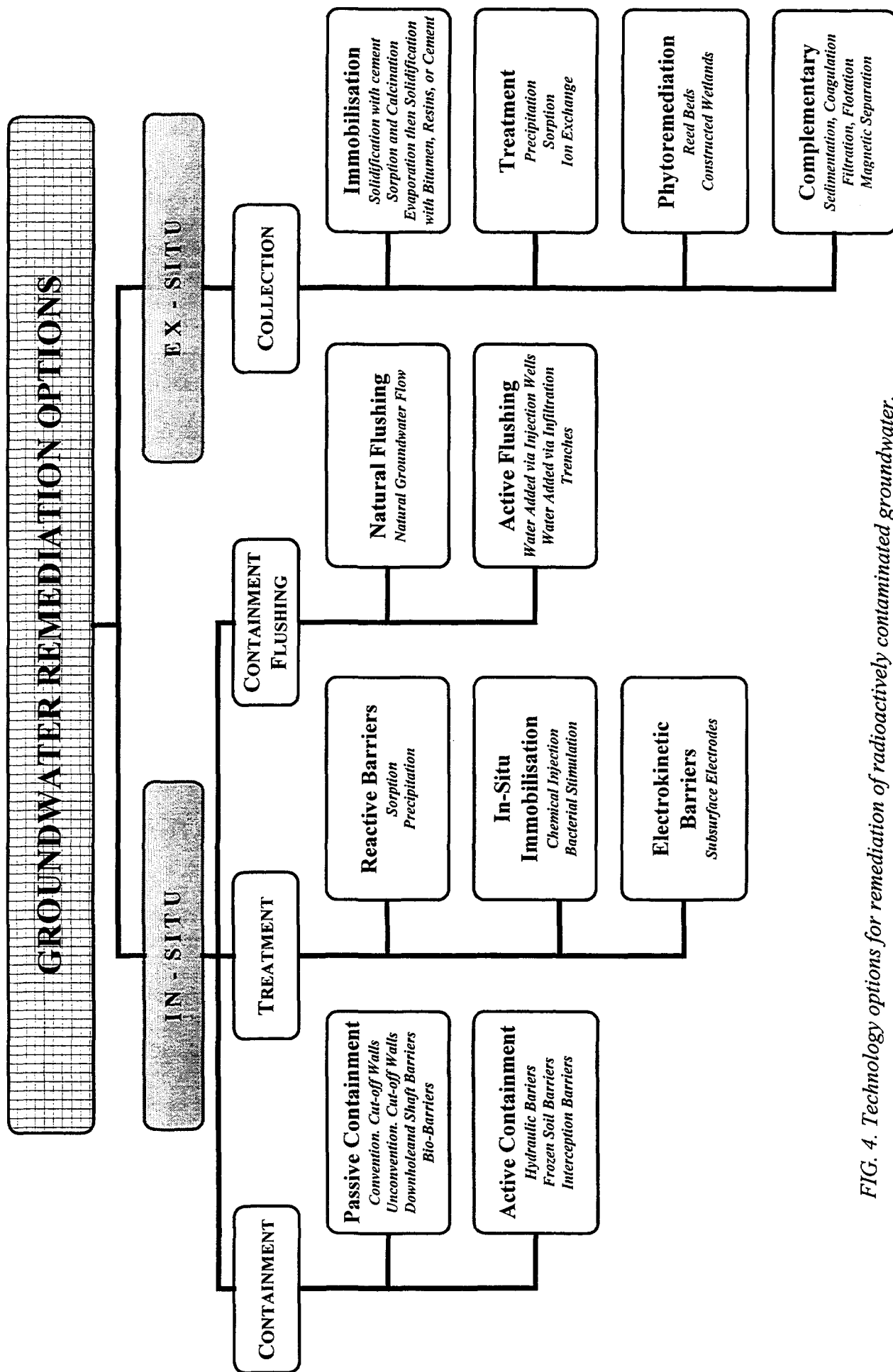


FIG. 4. Technology options for remediation of radioactively contaminated groundwater.

radioactive decay, meaning that the impact or potential threat of the contaminant is reduced with time. The time scale over which this will occur depends, of course, on the half-life of the contaminant in question. Since half-lives vary from seconds to millions of years, the extent to which decay assists remediation operations depends strictly on the contaminant(s) in question. However, unlike organic contaminants, the decay of radioactive contaminants cannot be influenced by the remediation technology. These decay properties impact upon the efficacy of any particular remediation operation and will be central to the selection of any technology option.

There are many way of classifying remediation technologies, but they generally fall into two main groups, that is, they are applied either in situ or ex situ. In situ remediation takes place within the soil/rock/water media, whereas ex situ techniques rely on the removal of the contaminated materials (e.g. groundwater) prior to treatment. Both of these categories can be further subdivided in a variety of ways until we reach the specific technologies available. Regardless of the specific details of the technologies employed in the remediation of radioactive contaminated groundwater, all of them can be simplified as representing two different approaches. Either the contaminant itself is removed from the groundwater (we could refer to this as groundwater “cleanup”), or the movement of the groundwater is altered or retarded. The breakdown of technology options is outlined in Figure 4, and for the purposes of this review starts with the in situ, ex situ classification.

## 7.1. IN SITU REMEDIATION

As mentioned above, in situ remediation occurs within the soil/rock/water media, with either the contaminant being removed from the groundwater (decontamination or “cleanup” of the groundwater), or the movement of the groundwater retarded/altered.

### 7.1.1. Containment

Containment is a method of groundwater remediation in which the movement of contaminated groundwater is retarded; the contained groundwater system can be defined as being either active or passive. An actively contained groundwater system, as defined here, is one which requires active (i.e. ongoing) operation and maintenance to remain effective. Passive systems, on the other hand, require little or no operational input once installed, but monitoring is essential. Containment systems are generally not contaminant specific since they target the groundwater (i.e. the medium) rather than the contaminant. Consequently, many containment technologies are generally transferable from one contaminant to another. When any containment remedial action is applied, there is always a long term requirement for monitoring to ensure that there is no contaminant breakthrough. Since the long term performance of such systems is difficult to predict, they are best applied to short lived radionuclides or used in combination with other remediation technologies.

#### 7.1.1.1. Active containment systems

The nature of active containment systems is such that their long term operation is generally neither practical nor economic. Therefore, they are seldom suited to the treatment of long lived radioactive contaminants where the system would have to be operated and maintained over long

periods of time (e.g. over many years, such as decades, etc.). The active containment approach is best applied as an interim measure as, for example, while the contamination problem is being further investigated or where it may be necessary to isolate a region of groundwater while another remediation technology is being applied.

### ***Hydraulic containment***

Hydraulic containment relies on the modification of the distribution or flow of groundwater around a region of contamination to prevent further migration of the radionuclides. Hydraulic containment can be achieved by the injection of clean (i.e. uncontaminated) water at strategic points within a contaminated aquifer such as at the leading edge of the affected body. Injection can be achieved either by pumping or, if the contamination is near the surface, through the use of dykes or channels filled with clean water. This injection changes the local groundwater flow pattern and thus prevents the contaminant from migrating in its original direction. To accomplish this, clean water needs to be available either from groundwater or surface water sources. The successful application of this approach requires a good understanding of the local hydrogeology so that the impact upon contaminant migration can be accurately predicted, and to ensure that the injection wells are properly placed. Key economic issues surrounding hydraulic containment are the long term operation and maintenance of injection and extraction wells. Hydraulic containment is a proven technology with demonstrated full scale applications at an in situ uranium mining site in the Czech Republic (see Annex VII).

### ***Frozen soil barriers***

In recent years, research has been conducted on the application of frozen soil barriers for the containment of contaminated groundwater [25]. This technology requires the installation of refrigeration tubes into/around a contamination plume. Although this technology has been tested at full scale by the US Department of Energy [43], it was only tested on a clean (i.e. uncontaminated) site. The results indicated that the barrier had low hydraulic conductivity and performed quite well when tested using  $^{137}\text{Cs}$  as the radiotracer. However, significant concern has been expressed regarding uniformly thick wall formation and also contaminant migration through the barrier over the long term.

### ***Interception barriers***

These barriers are constructed by the excavation of a dyke or channel to intercept contaminated groundwater flow. The interception barriers can either surround a contaminated region or they can be placed directly in the flow path of the groundwater. Intercepted water can be removed for treatment, stored in storage ponds, or re-injected in a region which poses less of an environmental or radiation threat. This method has been applied routinely at full scale for uranium contaminated groundwater arising from uranium milling operations in the Russian Federation. Dykes constructed to a depth of 25 to 30 metres have been in operation for this purpose.

#### ***7.1.1.2. Passive containment systems***

Passive containment involves the installation of a permanent, vertical barrier to intercept or contain contaminated groundwater. These barriers do not require any active operation once they have been installed, but monitoring of their performance is essential, and maintenance of the

barriers, if it is possible, may be required for some time in order to ensure satisfactory long term performance. Such barriers have been in use for many years in the construction industry to prevent groundwater seepage into underground structures and, as such, they represent a tried and tested technology. These barriers need to be extended into low permeability layers beneath the contaminated region to prevent migration under the barrier. The low permeability layer(s) could be either a natural clay layer or bed rock; and, alternatively, it is now possible to install horizontal barriers if no suitable natural formations are present. If it is desirable to achieve the complete isolation of a contaminant region, the vertical barriers can be emplaced in combination with a low permeability cap. This cap is required to prevent rain water ingress into the isolated region. If a cap is not employed, there is a danger of infiltration raising the water level within the isolated region until it can flow over the top of the vertical barriers, thereby releasing contaminant into the environment. Also, an increase in the hydraulic head will occur if infiltration is allowed and no groundwater pumping is performed. The increased head can cause leakage at rates not predicated and these leaks can have velocities much higher than the typical groundwater velocity in the area. Also, because of the additional head, leakage can occur through strata that normally allow no flow (e.g. low permeability silt lense).

### ***Conventional cut-off walls***

Cut-off walls are vertical barriers installed to prevent the horizontal migration of groundwater. There are a large number of construction methods and materials in current use for this approach. Typical materials for cut-off walls include concrete, cement based grouts, clays such as bentonite, and cement/bentonite combinations. The clay, bentonite, has been extensively used in the construction of cut-off walls due to its swelling properties on contact with water. A review of the application of clay as a barrier material to contain mobilized radionuclides has been provided in [26]. One of the main issues surrounding the long term performance of cut-off walls is their response to changes in their physical and chemical environment, e.g. dehydration, changes in pH, etc. can occur.

Cut-off walls can be classified into two basic types: displacement barriers and excavated barriers. Displacement barriers are constructed by forcing the barriers' material into the ground without any associated excavation. Examples of displacement barriers include steel sheet piling, membrane walls, vibrating beam walls and jet grouted walls. Material such as steel sheeting is not suitable for the containment of radioactive contaminants due to its short operational life span. Vibrating beam walls are constructed by displacing soil using a vibrating beam and the filling the resulting void, for example, with bentonite. Jet grouting barriers are constructed by drilling a borehole in which grout is injected by at high pressure jet through a horizontally rotating nozzle. The nozzle is raised slowly to form a grouted column, and a wall is formed by drilling and jetting a series of inter linking columns. The main problems encountered with displacement barriers arise if the joints are not impermeable or if buried debris and large rocks, etc. are able by their presence to prevent the successful installation of the barrier. Excavated barriers are constructed by removing soil material and replacing it with the barrier material. The commonest approach is the slurry wall where the excavations take place beneath a slurry consisting, for example, of a bentonite water mixture. The presence of the slurry prevents the walls of the excavation from collapsing. Once the required depth has been achieved, the slurry is displaced by the injection of the final construction material into the bottom of the excavated trench. Slurry walls are a tried and tested technology with many full scale examples around the world. The main advantage of slurry walls is the fact that there can be a high degree of confidence that the barrier will be continuous along its length and depth. The main disadvantage is that slurry walls may deteriorate with time,

and they may be susceptible to chemical attack by some contaminants such as acids, bases, salty solutions such as sea water, and alcohols.

### ***Unconventional cut-off walls***

Conventional cut-off walls are typically designed to have as low a permeability as possible. However, there are specific applications where it is important to allow groundwater flow to occur through a barrier. The aim in this case is to slow down, rather than completely retard, the flow of water. This approach is commonly used in uranium mines where it is necessary to block tunnels and/or connections among working regions where the levels of contamination may be quite different. It is important that these structural barriers retain their permeability during any flooding of the mines, so as help control the environmental pollution impacts. The barriers are typically constructed of cement grouting with the addition of fly ash; the exact formulations are purposely made to provide a significant permeability. A further refinement is to incorporate sorbing materials which will remove radioactive contaminants as the groundwater passes through the barrier. These barriers are only intended or designed to be effective during those time periods when mine flooding operations are occurring.

### ***Downhole and shaft barriers***

In a number of sites worldwide, uranium mining operations have left unplugged drill holes and mine workings, permitting contaminated water (including groundwaters) to mix with previously uncontaminated aquifers. Prior to mining, the aquifers would have been separated by aquitards. Mining operations can destroy the natural separation of aquifers through the drilling of boreholes and sinking of shafts. During mining operations, contamination of the overlying aquifer would usually be prevented by the cone of depression generated by dewatering activities. Problems then occur once the mining operations cease since dewatering is no longer carried out and re-saturation occurs. To prevent the contamination of a clean (uncontaminated) aquifer by the rising level of contaminated waters, it is necessary to close off the connecting shafts and bore holes. To accomplish this, barriers are placed in the shafts and bore holes at a point where the aquitard has been breached. These barriers are generally constructed with a cement based grout containing fly ash.

### ***Bio barriers***

The use of bio barriers is a rather innovative technology which uses the growth of bacteria to block the pores in a geological formation and, in so doing, forms a low permeable barrier. This technology was first developed for the oil industry where it was used to block high permeability regions in oil bearing strata to prevent short circuiting where water injection is used to force the oil out of oil bearing strata [27]. The approach employs nutrient poor bacteria which due to their small size are able to penetrate the pore spaces in the host geology. These bacteria are activated by the introduction of a substrate on which the bacteria can readily grow. During their growth period, the bacteria serve to block the pores through which the groundwater flows by both their own cells and the production of a slime material which is a by-product of their growth. One of the advantages of this approach is that the introduced bacteria will flow with the groundwater and naturally become attached to the geology (soil, rocks, etc.) through which groundwater is flowing. As the system becomes blocked, the groundwater will flow through the remaining unblocked regions, and carrying the bacterial cells along with it. These bacteria will then attach and grow in these regions, too, thus further reducing the unblocked portions of the region. This will

continue until essentially all the pore spaces are blocked, provided that the nutrients continue to be applied and available for the bacteria. The extent to which this technology has been applied at a field scale is difficult to assess due to the commercial (i.e. proprietary) nature of the oil industry. Applications to radionuclide contamination problems have been evaluated at a pilot scale and bio barriers appear to be a promising technology for the future. Bench scale demonstrations have shown 96–99% reductions in contaminant fluxes were achieved in cases of strontium and caesium contamination [28]. One of the major issues associated with this approach has to do with how long such a barrier will remain effective. However, experts in the field have suggested that because the pore spaces become blocked, nutrients become locked inside, the microbial system may actually be self sustaining for fairly significant periods of time. Another concern of utilizing bio barriers for remediation of radiologically contaminated groundwater is the adverse effect of the radiation on the bacterial population (although this would be unusual and would require very high levels of radiation).

### **7.1.2. Treatment technologies**

*In situ* treatment technologies act by immobilizing the radioactive contaminant within the contaminated region and thereby separating the contaminant from the groundwater. *In situ* treatments are often used in conjunction with containment technologies. With only one notable exception, *in situ* treatment operates by immobilizing radioactive contaminants by either precipitation or sorption. The exception is electrokinetics, where an electric field is employed to concentrate contaminants around electrodes inserted into the contaminated region. Although *in situ* treatments immobilize the contaminants within the contaminated region, with some technologies it is possible to recover the contaminant once the need for immobilization is over or periodically throughout the remediation.

#### *7.1.2.1. Reactive barriers*

Reactive barriers are water permeable barriers which are installed across the flow path of the contaminant plume allowing the plume to flow through whilst the contaminant is either precipitated or sorbed within the barrier (see Table II). There are a variety of chemical active agents which can be included in a reactive barriers, and these may serve to remove the contaminants in a variety of ways. Sorption barriers have been developed which employ organic materials such as peat, zeolites, and metal oxides of titanium and ferric iron. Precipitation barriers generally operate by either changing the pH or the redox potential of the groundwater. Materials such as hydrated lime and calcite (limestone) have been applied as pH modifiers, and elemental iron has been used to reduce the redox potential of groundwater, thus causing the precipitation of contaminants such as uranium.

In general, reactive barriers require the excavation of material and its replacement with the treatment composition. A variation on the reactive barrier concept is the funnel and gate system. In this system, impermeable barriers are used to divert or channel contaminated groundwater towards a reactive section of the barrier. This has the effect of concentrating the treatment process in a defined region. Since reactive barriers are essentially chemically dominated systems, their effectiveness is dependent on the chemical nature of the plume. Factors such as pH, redox potential, concentration, and the nature of multiple contaminants may all impact the effectiveness of reactive barriers. Likewise, reactive barriers may themselves impact the chemistry of the aquifer in which they are operating. This is particularly the case with precipitation barriers, and especially those which catalyse a change in the redox state of a contaminant in order to cause

TABLE II. REACTIVE BARRIERS TECHNOLOGIES

PROCESS	MATERIAL	STATUS	CONTAMINANTS	REFERENCE
Sorption	Zeolite	–	–	April (1996) [29]
	Clinoptilolite	Bench scale	Sr-90	Cantrell (1996) [30]
	Clinoptilolite	Bench scale	Sr-90	US DOE (1996a) [31]
	Clinoptilolite	Bench scale	Sr-90	Killey (1997) [32]
	Peat	“	Uranium mill tailing leachates	NA
	Titanium Oxide	“	“	NA
	Ferric Oxyhydroxide	“	“	NA
Precipitation	Lime	“	“	NA
	Calcium Carbonate	“	“	NA
	Phosphate	“	“	NA
	Modified clay	Pilot scale/Field Scale	Uranium, Technetium	Fruchter et al. (1996) [33]
	Elemental Iron		“	Dwyer et al. (1996) [34]

precipitation to occur. These barriers may result in a step change in chemical properties of the groundwater after it interacts with the barrier. The extent to which these changes could be detrimental will, of course, depend on the specific situation, but these potential changes should be taken into account when a reactive barrier system is being considered. Since reactive barriers are dependent on chemical properties for their performance, their effectiveness may degrade with time, precipitation barriers may become blocked and sorption barriers may become exhausted. Some reactive barrier designs allow for the replacement of the active component to overcome these problems. In general, no single reactive barrier technology will be applicable to all radioactively contaminated sites, because no two contamination situations will be the same. This means that reactive barriers need to be designed on a case by case basis, and the cost of this research and development phase may be significant.

Field demonstrations have been carried out with this technology to investigate its performance for radionuclide remediation [35]. These studies have proven the validity of the concept, they have demonstrated that such systems can function using fairly inexpensive materials, and, also, that the technology can be applied without causing a major impact on the hydrogeology of the aquifer. This technology appears to be on the verge of commercial application and exploitation with a number of commercial materials appearing on the market. A funnel and gate system using clinoptilolite to remove strontium-90 is currently being designed for application on the AECL Chalk River Laboratory site in Canada [32]. Examples of reactive barrier technologies which are currently under investigation are listed in Table II; this is not an

exhaustive list and is intended mainly to demonstrate the variety of systems under investigation, as well as the versatility of this technology.

#### *7.1.2.2. In situ immobilization*

In situ immobilization refers to technologies which facilitate the immobilization of radioactive contaminants within the subsurface, but which do not rely on the degree of engineering inherent in reactive barrier technologies. Such systems could employ the injection of chemical into an aquifer to change the chemistry, pH, or redox potential, resulting in the precipitation of the target contaminant. Potential examples include the injection of alkaline liquids to raise the pH, or the injection of oxidizing agents such as hydrogen peroxide to raise the redox potential. One example of in situ precipitation which has been in use for some time is the injection of lime suspension into flooded uranium mine workings at the Ronneburg site in Germany (see Vogel in Annex V). The concept of in situ precipitation of heavy metals by injection of hydrogen sulphide gas has been proposed [36], as has the precipitation of lead as insoluble phosphate salts [37]. Such an approach requires a good understanding of the local hydrogeology, as well as an adequate control of the system to contain the treatment agents and prevent their migration beyond the treatment zone. One disadvantage of this technology is that, once implemented, the process control is limited.

One variation of this approach is the stimulation of indigenous or introduced micro organisms which can, in turn, change the chemical environment through their growth. One example of this approach is where bacteria are stimulated to immobilize uranium through the generation of reducing conditions in contaminated plumes originating from uranium milling and mining operations [13]. Although this approach has undergone extensive research, it has not yet been applied at full scale. Among the potential problems with this approach is the re-oxidation (and re-mobilization) of the uranium once the treatment process is complete.

#### *7.1.2.3. Electrokinetic barriers*

An electrokinetic barrier functions by the employment of a series of electrodes placed within a contaminant plume to screen contaminants from the plume as it passes through the barrier. The process generally involves the passage of a low direct current through the contaminated region between positive and negative electrodes. Cationic contaminants tend to accumulate near the negative electrodes where they can be either removed through pumping or sorbed in situ. Electrokinetics is a tried and tested technology for processing soils; it has been used for years for the removal of salts from agricultural soils. Its potential application to contaminated soils and groundwaters has received a significant amount of attention in recent years. It has the advantages of being effective in low permeability strata for which other in situ techniques are less effective. A major disadvantage of this approach is that as the groundwater flow rates increase, the cost of the electricity required to maintain the barrier may rise prohibitively. Another disadvantage is that the presence of metallic objects, such as buried pipes, can significantly inhibit the process efficiency and increase costs. This approach has not been used for the large scale remediation of radioactive sites, but it has been applied to the remediation of heavy metal contaminated land. In the case of radioactive groundwater remediation, it is best considered to be at the pilot scale and not yet a fully proven technology.

### 7.1.3. Contaminant flushing

The concept of contaminant flushing involves the use groundwater flow, either natural or enhanced, to purge the contaminated groundwater from the region of concern. Along with the processes of retardation, dispersion, dilution, and radioactive decay, contaminant flushing can be effectively applied in given situations, resulting in the attenuation of groundwater contamination down to acceptable levels.

#### 7.1.3.1. Natural flushing

In natural flushing, the existing groundwater flow conditions are employed to flush the contaminant out of the region of concern. Natural flushing can be an appropriate remedial solution for aquifers with high groundwater velocities and dispersivities because these properties will promote the attainment of the required level(s) of dilution. Natural flushing may also be the most cost effective and complete remediation technology for aquifers with low groundwater velocities and dispersivities when coupled with institutional controls (e.g. access limitations) that provide enough time to achieve contaminant flux.

Although this is not strictly a remediation technology (as defined in the introduction to this section), natural flushing is an approach which should always be considered when choosing a remediation technology. Natural flushing provides the baseline case against which all other approaches can be compared. In some cases, the advantages gained, or the costs incurred, by adopting an alternative remediation technology may be such that flushing is the most appropriate option available. Alternative remediation technologies should only be employed if they provide real, quantifiable benefits over natural flushing (i.e. if they are worth the additional investment).

The selection of natural flushing is not a “do nothing” remedial option; rather, its application will require a significant degree of monitoring throughout the lifetime of the process and implementation of institutional controls to ensure effective protection of human health and the environment. If this approach is to be adopted, it is essential that the hydrogeology and contaminant transport characteristics of the system are well characterized. In addition, the mathematical modelling of the geo-hydrological and mass transport situation, both now and in the future, is advisable and probably essential to its successful implementation. Natural flushing is recognised as a legitimate method for achieving compliance with groundwater protection standards by the US Environmental Protection Agency (EPA). For natural flushing to be approved by the EPA, it is required that the following features can be demonstrated:

- compliance to cleanup criteria is to be achieved within 100 years or less;
- the groundwater is not currently being used as a source of community water (or other beneficial use), nor is it projected to be so used within the natural flushing period;
- established concentration limits are not projected to be exceeded at the end of the natural flushing period; and
- adequate monitoring will be established and maintained throughout the flushing period.

Natural flushing has been identified as a potential approach for the remediation of groundwater contaminated by leachates arising from uranium mill tailing sites [38] and is near to being approved for that application.

### 7.1.3.2. Active flushing

Active flushing or gradient manipulation [38] is an enhanced version of natural flushing described above and is a candidate for those sites where natural flushing is proceeding too slowly, such as sites with very low hydraulic conductivity or low gradients. It is achieved by adding water to a contaminated aquifer to increase the groundwater velocities in a specific direction. The water addition can be achieved either via injection wells or infiltration trenches. The successful application of this approach requires a source of uncontaminated water to supply the injection process. As with natural flushing, an efficient monitoring programme is essential, and mathematical modelling of the consequences of water addition is advisedly done prior to implementation and throughout the full duration of the remediation process.

## 7.2. EX SITU REMEDIATION TECHNOLOGIES

Ex situ remediation technologies involve the extraction and subsequent treatment of the contaminated groundwater. One of the main advantages of ex situ remediation is that the above ground treatment process can be engineered and controlled to a much greater degree than is possible with an in situ process. In many cases, the treatment processes employed are essentially the same as those used in situ systems, it is simply the manner in which the process is brought to bear on the contaminant that is different. A conceptual flow diagram for an ex situ remedial action system is shown in Figure 5.

In almost every case, ex situ remediation generates two products for disposal, the remediated groundwater and the contaminant containing stream. The contaminant containing stream can be a solid, liquid, or sludge/slurry and it will require final disposition in an appropriate disposal facility. For a technology to be considered viable, it should generate far lower volumes of the contaminant containing stream than the volume of groundwater treated. In general terms, provided that the efficiency of the remediation is acceptable, the greater the volume reduction which has been achieved with the process, the more successful the remediation technology. However, it should be borne in mind that when treating radioactively contaminated groundwater, the concentrated form of the contaminant may have negative, as well as positive, consequences. Although criticality problems are highly unlikely (assuming that fissile radionuclides are present), there may be a potential problem with regard to the radiological classification of the waste. Concentration of the radioactivity could result in a higher classification (e.g. “intermediate” rather than “low level” radioactive waste) with an associated substantial increase in the final disposal costs. In some countries, although low level waste disposal sites may be available, it could be that intermediate level disposal sites are not. In other countries, the final disposal option for radioactive waste may depend on the half-life of the contaminants (radionuclides), too. Thus, the available disposal options (and costs) can vary, depending on the nature of the contamination. When considering what ex situ process to employ, the final disposal route (and related costs) should always be high on the list of criteria used to evaluate and select a technology. It is quite possible, of course, that the most effective or efficient removal technology is not the one which is the most cost effective or environmentally acceptable.

Key to the success of any ex situ remediation is the efficiency of the extraction process. The most common extraction system would involve the installation of a series of extraction wells from which the contaminated groundwater is pumped. If the contaminated groundwater is at shallow depths, then a so called “well point network” would be appropriate. Well points are

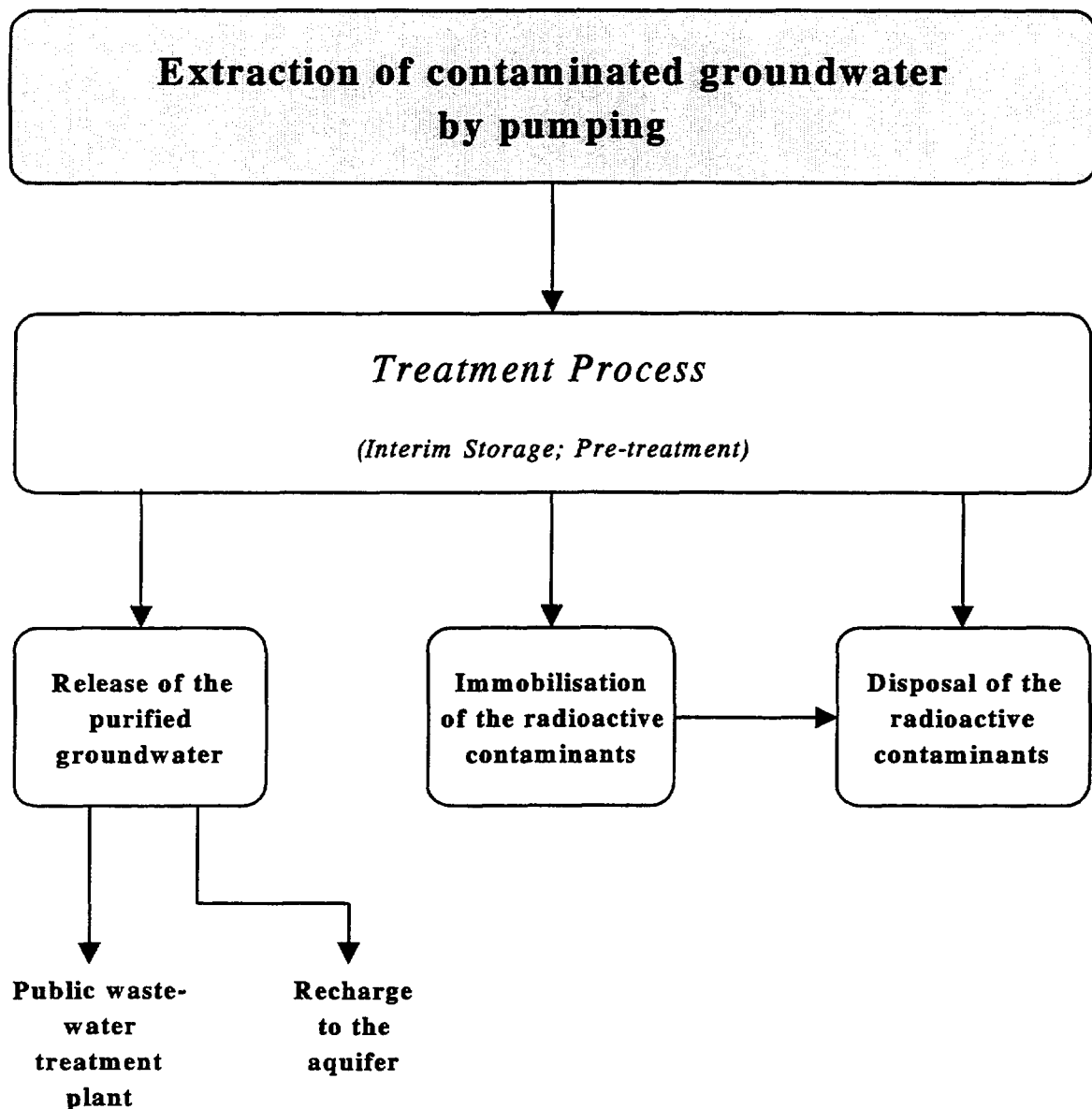


FIG. 5. Conceptual flow diagram for an ex-situy remedial action system.

generally of a smaller diameter than extraction wells and are driven into the ground rather than being drilled. For the extraction of fluid, they are also generally connected to a central vacuum pump rather than having individual down hole pumps. An alternative to pumping is the use of trenches to collect seeping or intercepted groundwater. As with well points, this is only practical for the collection of shallow groundwaters.

There are a variety of pumping strategies which could be applied to the extraction of contaminated groundwater. For example, it might be necessary to combine periods of pumping with periods of no pumping (sometimes referred to as "pulsed pumping"). The periods of no pumping would allow contaminants to diffuse out of less permeable zones or to desorb from the aquifer matrix. The length of time that the pumping may be suspended will depend on how long the aquifer takes to re-establish its equilibrium contaminant concentration. If this approach is not adopted and extraction pumps are run continuously, false end points may result. This is possible when the contaminant concentration in the groundwater falls below the targeted level, but if the

remediation process is stopped the contaminant concentration in the aquifer starts to increase with time. This result can occur due to a re-establishment of the equilibrium processes described earlier. Even with optimum pumping strategies, many pore volumes of groundwater may need to be extracted to achieve a desirable level of cleanup.

It should be noted that for many sites, continuous pumping may provide a faster and more complete remediation than pulsed pumping. There are two reasons for this. By continuously pumping, the pores between the soil particles are continuously swept with groundwater, effectively maximizing the concentration gradient of contaminant molecules between the soil particles and the groundwater. This maintains a maximum diffusion rate, which is the rate limiting step in groundwater remediation. Conversely, allowing the contaminant equilibrium concentration to be reached essentially stops the diffusion process, effectively stopping the remediation. Additionally, continuous pumping can result in a more complete remediation because the groundwater is continually captured, rather than intermittently as with pulsed pumping. Thus, contaminants are not allowed to be swept away with the groundwater that continues to flow. However, continuous pumping does result in very low contaminant concentrations in the extracted groundwater and this may be costly if the ex situ treatment costs of the groundwater are based on the volume of groundwater treated. A compromise of the two pumping strategies is to lower the extraction flow rate when the mass transfer of the contaminant is diffusion rate limited. However, a lower flow rate will result in a smaller groundwater capture zone and a lower flow rate may not be compatible with an ex situ groundwater treatment system sized for a higher flow rate.

The extraction bore holes used for groundwater recovery need to be installed with care. It is particularly important that the drilling of boreholes does not lead to an increased spread of contamination due to the introduction of preferential flow paths. Historically, preferential flow paths have been observed in the annulus of wells that have not been properly sealed with low permeability material, such as bentonite or grout.

The remediated groundwater, which is the other product of an ex situ remediation process, also needs to be disposed of. The disposal options which are available include re-injection into the aquifer, passive infiltration into the aquifer, disposal to land/marine/surface water bodies, evaporation, and even disposal to foul sewer systems, and so on. Re-injection of treated groundwater up gradient of a contaminated region effectively combines the technology of hydraulic flushing with the ex situ treatment. This approach has been proposed for the Wismut uranium mining sites in Germany for the treatment of contamination resulting from in situ leaching operations (see Annex V). Re-injection can also be performed down gradient of the contaminated region and, depending on location, volume, and site conditions, the down gradient re-injection can serve as a hydraulic barrier to flow from the contaminated region, effectively containing the contaminant plume.

Whichever disposal option is used, it is important that the environmental impacts of that particular disposal is considered. This should, of course, include consideration of other potential contaminants in the water (e.g. nitrate, sulphate) which may or may not have been influenced by the remediation technology employed, since this would probably have been specifically targeted at the radioactive constituents. In some cases, it is possible that a secondary treatment system may be required to remove nonradioactive contaminants, thereby allowing for the disposal of the remediated groundwater.

### **7.2.1. Collection and immobilization**

A potential approach to the remediation of radioactively contaminated groundwater is to solidify (i.e. immobilize) the recovered contaminated water in a stable matrix with a cement. This is unlikely to be an efficient or practical process due to the large amount of groundwater which would require solidification. Another approach is the sorption of contaminants onto clay (or other sorbents) and the subsequent calcination of the clay (or sorbent) to form a compact solid for final disposal.

Solidification may become a more practical process, however, if it is combined with an evaporation stage where the water loss is allowed to occur in an open system, thus leaving the contaminant behind in a more concentrated state. The immobilization/solidification of liquid wastes following an evaporation stage is a very common treatment process for liquid radioactive wastes wherein matrices such as bitumen, epoxy resins, and cement have been used. Similar approaches have also been applied to some radioactively contaminated sludges.

The only examples of this evaporative approach being applied on a large scale to radioactively contaminated groundwater is in the treatment of groundwaters contaminated by uranium mill tailings. In these cases, contaminated waters have routinely been extracted and pumped as a fine mist over the ground surface to facilitate evaporation. An alternative (though less commonly used approach) is to pump the contaminated water into a holding pond where evaporation takes place. In these cases, no engineered immobilization occurs and the contaminants are distributed either within the soil or in the bottom sediments of the pond.

When evaporation is considered as part of the treatment strategy for contaminated groundwater, several precautions are necessary. A primary concern is the control of airborne radionuclide contamination which can occur as a result of spraying a fine mist or when all the moisture has evaporated. If the evaporation relies on weather conditions and solar energy, the rates of evaporation can vary substantially at different times of the year. If the evaporation does not rely on the weather conditions and solar energy, but rather by energy input by engineered means, the costs of such energy can be very high. Also, evaporation of radioactively contaminated groundwater will concentrate the radionuclides and precautions should be considered because of elevated dose rates and (under certain conditions) criticality concerns.

### **7.2.2. Collection and treatment technologies**

In collection and treatment technologies, the recovered groundwater is treated in one or a number of process steps to separate the groundwater from its radioactive contaminants. The radioactive contaminants may be collected as a solid, a liquid or a sludge/slurry. The advantage of this approach is that process conditions can be carefully controlled and a number of treatment process can be combined in series, thereby allowing the sequentially targeting of specific contaminants in the groundwater. There are a large number of ex situ treatment processes available, some of which have been in use for many years in the chemical and wastewater treatment industries. The tried and tested nature of these systems makes them highly attractive as treatment options because they are typically off the shelf technologies and are, in many cases, relatively easy to install and operate. In recent years, a number of new and innovative technologies have also been developed which complement those already available. Regardless of the pedigree of these system, most can be grouped into two broad classes, *viz.*, they are based on either precipitation or sorption (ion exchange). A notable exception to this, however, is phytoremediation which is also discussed below.

### 7.2.2.1. *Precipitation processes*

Precipitation is a physicochemical process wherein dissolved (generally inorganic) species are transformed into less soluble or insoluble compounds under changed chemical conditions. The alteration of species in the dissolved state to insoluble compounds facilitates their physical removal from the system by processes such as sedimentation or filtration. Precipitation is a well established technology which can be easily fitted into an integrated treatment process; it has been successfully applied to the removal of heavy metals from liquid waste streams for many years. Precipitation is generally facilitated in three different ways (or combinations of the three ways) as follows:

- the pH is modified, e.g. by chemical addition;
- the redox potential is changed, e.g. by chemical addition; and
- the chemical composition is changed, e.g. by chemical addition.

The essence of any precipitation process is the continuous addition of chemically active materials. This addition can be problematic since the efficient treatment depends on adding the reactants in the proper proportions, on the appropriate mixing conditions, and maintenance of the influent water chemistry, all of which may vary and be difficult to control. This can cause problems since, for example, unless proper doses of reactant (“dosing”) and the water chemistry are closely coupled, over and under dosing can occur. Under dosing can obviously cause a problem since it reduces the removal efficiency, however, over dosing can also be a problem. This is because over dosing can result in undesirable/unacceptable levels of process chemicals in the effluent water stream. Precipitation, like other conventional treatment processes, is nonselective in that ions that are not of specific interest will also precipitate along with the target species. This may result in increased levels of waste material to be disposed of (i.e. increasing the costs of disposal).

On the other hand, the nonselective nature of precipitation may also have some advantages. For example, the precipitation of calcium brought about by an increase in pH will also result in the co-precipitation of strontium. Co-precipitation is a process by which ionic species will precipitate at lower concentrations than would normally be expected, due to the precipitation of another ionic species which has similar chemical properties. This approach has been successfully applied at full scale for the removal of strontium from contaminated groundwater at the Chalk River Laboratories in Canada [32].

Examples of precipitation processes employed to remove radioactive contaminants include the application of ferric chloride to remove radium and uranium from liquid effluents and groundwaters resulting from uranium mining operations. The addition of ferric chloride results in the precipitation, under alkaline conditions, of iron as ferric hydroxide, which in turn results in the co-precipitation and sorption of certain radioactive contaminants. This process has been successfully employed to remove radium and uranium from contaminated waters generated by uranium mill tailings [39]. Alternatively, in neutral and weakly alkaline mine water, radium can be removed by co-precipitation with barium sulphate in association with aluminum and ferrous sulphate. In weakly acidic solutions, barium chloride can be added to stimulate the precipitation of radium.

Recent research and development activities in the field of precipitation technologies for remediation have included bio precipitation technologies. The bio precipitation approach employs

biological systems to bring about the chemical changes needed to effect precipitation. Examples of bio precipitation include enzymatic uranyl phosphate precipitation [40], uranium precipitation through microbial generation of reducing conditions, and the bio precipitation produced in sulphate reducing bioreactors [41]. Although the application of these technologies to removal of radionuclides is at the bench and field scale, there has nonetheless been a full scale application of a sulphate reducing bioreactor to remove heavy metals from groundwater [6, 42].

#### *7.2.2.2. Adsorption and ion exchange systems*

The use of adsorption and ion exchange processes for the decontamination of wastewater streams is a mature technology which is routinely applied in the chemical process industry. Ion exchange involves exchanging anions or cations between an ion exchange material and the wastewater. Adsorption is a similar process, which does not include the sorbing material losing a counter ion (i.e. the sorbed species become attached to surfaces of solid sorbent materials without there being any mass loss). There are many sorption and ion exchange materials available, including clays; by-products such as fly ash; organic materials such as peat; biological materials such as fungal and algal biomass; and commercial products such as zeolites. Ion exchange resins can be recharged (i.e. the process can be reversed) once they have become saturated. However, where this approach is used, the resulting radioactively contaminated recharge (wash) waters will require a controlled disposal.

Although these are mature technologies, there are limitations to the remedial application of adsorption and ion exchange processes including such effects as clogging by oils, greases and suspended solids; damage resulting from changes in chemistry; and damage by chemical reactants such as strong oxidants. A common application of these processes is as a polishing stage after a precipitation process has been applied to remove the bulk of the contaminants [47].

#### *7.2.2.3. Phytoremediation*

Phytoremediation involves the use of plants to remove contaminants from the subsurface and produce a harvestable biomass [44, 45]. The growing and harvesting of plants is a relatively inexpensive operation, and it can be accomplished with minimal effort. A notable negative aspect of phytoremediation is that there is a risk of inadvertent transfer of contaminants down the food chain through the ingestion of plant material (the biomass) by animals. There is also the problem that the biomass which is harvested has to be disposed of; one possible route to disposing of the biomass is incineration, though, of course, this too may not be environmentally benign.

Phytoremediation is an in situ technology when applied to the remediation of contaminated soil. However, when applied to the treatment of groundwaters, it is probably better done so as an ex situ process. In the ex situ configuration, the extracted groundwater could be diverted through specially constructed beds of plants to allow the maximum contact between plants and contaminants. Reed bed technology has been applied to the treatment of acid mine drainage and landfill leachate at conditions ranging from pilot to field scale. One interesting variation on conventional phytoremediation is the use of constructed wet lands. These systems operate on a larger scale than reed beds and the operative metal removal processes include both chemical and microbiological processes along with the accumulation of plant biomass material.

#### *7.2.2.4. Complementary technologies*

There are a large number of complementary technologies which can be applied as ex situ processes. The most common of these are technologies which improve the performance of precipitation processes. These include sedimentation, filtration, coagulation, flotation, and, potentially, magnetic separation. It is not within the scope of this report to document or discuss these processes in detail, however.

#### **7.2.3. Process integration**

Process integration refers to the effective combination of complementary process stages to maximize contaminant removal. Process integration is commonly applied in ex situ (as opposed to in situ) processes where it is easier to combine and mix multiple technologies. Its application to in situ conditions is more difficult to envisage, however, in principle, in situ and ex situ combinations could be potentially feasible. It is recommended that opportunities for process integration should always be investigated, since it is through such integration that the maximum contaminant removal is often achieved.

As an example of process integration, a typical flow sheet for the removal of uranium from groundwater contaminated by uranium milling operations may include the following (for an example, see Annex III):

- (1) clarification of the solution by addition of coagulants such as polyacrylamide, or ferrous or ammonium sulphate, followed by separation via thickening and filtration;
- (2) precipitation of uranium and other radionuclides by the addition of barium salts or ferric chloride; and
- (3) a final cleaning stage employing ion exchange or adsorption technologies.

A similar flow sheet is followed at the Helmsdorf water treatment plant in Germany which treats up to 200 m<sup>3</sup>/h of uranium and arsenic containing groundwater. The major process stages are as follows:

- (1) filtration to remove suspended solids;
- (2) ion exchange to remove uranium;
- (3) precipitation and flocculation to remove arsenic and radium; and
- (4) a final liquid solid separation using flotation and filtration technologies.

There are many other examples of successful application of remediation technologies on a large scale for contaminated groundwaters. In general, the chosen approach for a given contamination problem will depend on many local and site specific factors, economic considerations, and so on, as have been discussed above.

## **8. SUMMARY AND CONCLUSIONS**

Although the environmental restoration industry is still relatively young, there is a fair amount of experience around the world in planning remedial options for addressing potential

health risks and threats to the environment posed by radioactively contaminated surface and groundwaters. As a consequence of building this experience, there have been a number of lessons learned. To date, most of these pertain to studying the problem and planning reasonable approaches, rather than the successful completion of cleanup actions. One of the objectives of this technical report has been to capture and describe the methodologies, management processes, and national experiences that could be usefully applicable to Member States of the IAEA which are facing problems associated with radioactively contaminated groundwater sites. A number of off the shelf and innovative technologies have been identified which have real potential to cleanup/remediate radioactively contaminated groundwater. From the in situ technologies, these are the active and passive containment barriers, in situ chemical/bacterial injection, and electrokinetic barriers. Innovative ex situ technologies are phytoremediation with reed beds and constructed wetlands, and magnetic separation techniques. However, many of the remediation technologies identified in this report have not truly progressed beyond the conceptual or bench scale stage. In most cases, existing technologies currently being implemented involve classic waste water treatment techniques utilizing proven chemical and physical treatment technology.

Relatively few large scale, extensive, and successful cleanup actions have been implemented for radioactively contaminated groundwater. Remediation of sites containing radioactively contaminated groundwater is complex and numerous factors contribute to the difficulty, as shown below:

- lack of historical experience;
- implementation difficulties;
- magnitude of the task, varying from near routine to exceedingly challenging, if even capable of being accomplished with existing technologies and resources;
- lack of mature understanding of risk consequences and environmental effects;
- unfavourable cost/benefit ratios;
- other more demanding or opposing national priorities;
- technical impracticalities based on existing technologies
- existence of unclear or conflicting goals;
- indecisiveness on the part of decision makers;
- conflicting, nonexistent or poorly constructed sets of regulations and laws;
- lack of leadership, imagination, objectivity, knowledge, connectivity of the affected parties or planners/decision makers, or freedom of action on the part of the technical community and other groups whose efforts are needed to achieve successful cleanup/remediation; and
- unworkable or poorly constituted institutional control/management options.

At most (perhaps all) of the known radioactively contaminated groundwater sites around the world, people are not currently dependent upon the contaminated groundwater as a source of drinking water. Therefore, the potential health risks due to groundwater exposure pathways lie in the future. In most regions of the world, groundwater is a precious and valuable resource and has significant socio-- political ramifications. As a consequence, political leaders and decision makers often find themselves faced with difficult alternative selection issues. And, because of the often emotional views of the public, the affected populations and others, the cost/benefit ratios for remedial action are sometimes construed as being unimportant or of secondary consideration.

A wide range of management and technical options have been identified in this technical report for consideration by interested parties. The remedial options range from the natural flushing alternative (allowing nature to take its course while continuing to monitor the situation) to aggressive extraction and treatment in an effort to reduce the radioactive contamination to "background water quality." Each site should be treated as a unique case, with its own special requirements, conditions and socio- political factors. In some cases, the natural flushing alternative may be quite appropriate and in other cases, the need to implement some amount of engineered remediation system is necessary. In all cases, it is hoped that a rational approach can prevail to achieve optimal results based on logical, cost effective and risk reduction based applications of remediation technologies.

Many of the programmatic and site specific factors and considerations that should be considered by decision makers for addressing the issues involved with the remediation of radiologically contaminated groundwater have been identified in this technical report.

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## **Annexes**

### **CASE HISTORIES OF NATIONAL EXPERIENCE**

#### **Introduction**

National experiences vary among countries that are planning or implementing remedial action to address radioactively contaminated groundwater. Many Member States of the IAEA have developed varying degrees of knowledge on site histories, source terms, the behaviour of radionuclides in groundwater systems, hydrogeologic conditions, risk assessments, technology evaluations, and public awareness relations. Although no single country has accumulated vast experience or records of success in the remediation of radioactively contaminated groundwater, some countries such as Canada, Germany, and the United States of America have successfully implemented remedial actions, including such conventional approaches as pump and treat. Some countries are experimenting with innovative technologies for remediating contaminated groundwater, such as reactive barriers, bioremediation, and phytoremediation. These annexes present narratives of the experience of selected Member States.



## **Annex I**

### **UNITED STATES OF AMERICA — THE UMTRA PROJECT**

#### **I-1. INTRODUCTION**

In the aftermath of the Cold War, the United States of America has begun addressing the environmental consequences of five decades of nuclear weapons production and testing. The buildup and stockpiling of nuclear materials and weapons required an extensive manufacturing and testing effort that generated large volumes of waste and resulted in considerable environmental contamination. In many cases, remediation will be supplemented with the implementation of monitoring, land and water use restrictions, and other institutional controls to protect human health and the environment over the long term.

##### **I-1.1. Contamination from nuclear weapons production, research, development, and testing activities**

Radioactive and hazardous substances arising from nuclear weapons production, research, development and testing activities, as well as from other Department of Energy (DOE) nuclear and non-nuclear programmes, have contaminated groundwater and soils on and in the vicinity of DOE sites. Some waste streams have been discharged to the environment with and without prior treatment. These include relatively small, localized releases that may have resulted from accidents; larger planned releases of process effluents; and releases on a much larger scale, such as atmospheric dispersion and fallout from nuclear weapons testing. In other cases, containment systems such as tanks, drums, lined cribs or landfills have lost their integrity and wastes have leaked into adjacent soil and water. Contaminated media (including groundwater) have also resulted from spills and other inadvertent releases during process operations or maintenance.

The processes which have generated radionuclides and radioactive waste during this period, leading to the contamination of soils and groundwater, include the following:

- Uranium mining, milling, and processing;
- Isotope separation (enrichment);
- Fuel and target fabrication;
- Reactor and particle accelerator operations;
- Chemical separations;
- Weapons component fabrication and testing;
- Weapons supporting operations;
- Research, development and demonstration activities.

The contaminated environmental media and primarily water, soils, and biota. Nuclear weapons production activities have resulted in a legacy of 1500 million cubic metres of contaminated water and over 70 million cubic metres of contaminated soils. Non weapons activities have contaminated an additional 350 million cubic metres of water and around 6 million cubic metres of soils. Radioactively contaminated groundwater is known to exist at 39 sites in the United States of America. At several DOE sites, contaminated groundwater has also migrated outside of the controlled site boundary. An example of this is the Paducah Diffusion Plant in the State of Kentucky. At this site, the groundwater has become contaminated by Tc-99, a long lived radionuclide, and trichloroethylene, a hazardous cleaning solvent that was once commonly used at the site. The contamination resulted from leaks, poor waste disposal practices, and discharges

which occurred on the site many years ago. Over time, the contaminants reached the groundwater; the contamination gradually dispersed until several large plumes of contaminated groundwater had formed. The DOE has been investigating this contamination for several years, has tried to identify the sources, and has begun an interim removal of the contaminants, along with the implementation of controls on the groundwater plumes. Until a final decision is made concerning the remediation of this contamination, and an action plan is implemented, DOE will be providing an alternate water supply to the public where the groundwater contamination has reached hazardous levels.

Other sites known to have experienced offsite groundwater contamination include Fernald, Hanford, Kansas City Plant, Los Alamos National Laboratory, Brookhaven National Laboratory, Lawrence Livermore National Laboratory, Oak Ridge National Laboratory, Pantex Plant, Rocky Flats Environmental Technology Site, and the Savannah River Site\*

### **I-1.2. Contamination from uranium mill tailings and former processing activities**

The Uranium Mill Tailings Remedial Action (UMTRA) Project in the United States of America was established by the US Congress in 1978 to assess and clean up contamination at 24 designated former uranium processing sites. To date, UMTRA has completed groundwater remedial closure actions at two sites, Maybell, Colorado and Spook, Wyoming. After the initial characterization of the contaminant plumes and hydrological conditions, with the use of existing data, it was determined that the sites qualified for a “supplemental standards” designation due to their limited use groundwater. The application of supplemental standards was determined to be sufficiently protective of human health and the environment based on the conclusion that there was no direct exposure pathway to receptors (i.e. members of the affected population).

Further details of the conceptual model, risk assessment, and remedy selection can be found in the final Maybell, Colorado Uranium Mill Tailings Remedial Action Plan (1997) ; the Spook, Wyoming UMTRA Site Observational Work Plan (1995) ; the Spook, Wyoming UMTRA Environmental Assessment of Contaminated Groundwater (1997) ; and the Spook, Wyoming UMTRA final Groundwater Compliance Action Plan (1997).

Of the remaining former processing sites, seven are targeted for the “no further action” remedy selection, nine are targeted for the “passive” remedy selection, two are targeted for “active engineered” remedy selection, and one will most likely be targeted for a combination of remedy selections.

For most of the UMTRA sites, there is a need to collect additional data in varying amounts. One site, Riverton, Wyoming, has completed detailed data collection, and its draft analytical and numerical contaminant transport modelling has been completed. Riverton will be the first UMTRA site proposed to the regulatory agency (US Nuclear Regulatory Commission). Formal submittal to the regulatory agency is planned to occur next year.

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\* Linking Legacies, US DOE, Rep. DOE/EM-0319 (1997).

The two sites targeted for active engineered remedy selection are Tuba City, Arizona and Monument Valley, Arizona. Both of these sites are located on Native American Reservations. Indian representatives feel strongly that the groundwater must be cleaned up regardless of cost. The contaminants of concern are sulfates, nitrates, and uranium. To date, workshops held on evaluating Innovative Technologies for cleanup/remediation of the Tuba City site have not identified an innovative technology with practicable application. A technical team continues to evaluate the concept of enhanced pump and treat. One possible enhancement could involve the application of blast fracturing and/or excavation of horizontal wells. Particular challenges to be addressed during the selection process for a cleanup remedy include the limited recharge fluxes, low hydraulic conductivities within the saturated zone, very low extraction well yields, treatment methods for removing contaminants from the water, and solid waste disposal.

The estimated remediation completion costs for an active engineered UMTRA site are on the order of \$50 to \$100 million. The cost estimates include escalated dollars over the lifetime of the project, as well as a contingency fund. Furthermore, there may be real uncertainty as to whether the pump and treat approach can meet the cleanup goals. The cost estimates are based on the assumption that the contaminant concentrations can be reduced to their cleanup goal levels after 3 pore volume extractions. The conceptual well field design has a hydraulic requirement that there be hundreds of vertical extraction wells operating over the course of the remediation project.

Two examples of US Programmes to address radioactively contaminated groundwater are presented, one for the UMTRA Project (abandoned uranium facilities, etc. ) and one to address contamination problems at US weapons and laboratory sites (e.g. the Hanford Site in the State of Washington, where weapons material, plutonium, was produced in nuclear reactors and processed for manufacture of nuclear weapons). The examples described here typify the problems faced by the US Department of Energy in the remediation of contaminated sites.

## **I-2. THE UMTRA PROJECT**

### **I-2.1. Background and setting**

From 1943 to 1970, much of the uranium ore mined in the United States of America was processed by private companies under procurement contracts with the US Atomic Energy Commission. This ore was used in national defence research, weapons development, and the developing nuclear industry. After fulfilling their contracts, many of the uranium mills closed and left large quantities of waste, such as uranium mill tailings and abandoned mill buildings at the sites.

Beginning in the late 1960s and 1970s, serious concerns arose about direct gamma radiation, radon gas, and uranium decay products at the abandoned sites, as these were determined to be potentially hazardous to human health from long term exposures. Public concern about health hazards associated with mill tailings and radiation lead to engineering and radiological studies to identify mill sites around the country. As a result of these studies, the US Congress passed the *Uranium Mill Tailings Reclamation Control Act* (UMTRCA) on November 8, 1978. This essentially established the US Department of Energy's Uranium Mill Tailings Remedial Action (UMTRA) Project.

The UMTRCA directed the US Department of Energy (DOE) to stabilize, dispose of, and control, in a safe and environmentally sound manner, uranium mill tailings at designated inactive uranium mill sites. Further, Congress directed private companies to implement similar actions at uranium and thorium sites that were still active in 1978. Under the UMTRA Project, DOE has been performing remedial action of the surface contamination since 1983. Surface remedial action has been completed at 22 former processing sites.

Encapsulating the tailings and other contaminated materials into an engineered disposal cell eliminates the continued source of pollutants into the groundwater, but does not address the residual groundwater contamination that resulted from milling activities and from years of uncontrolled tailings the effects of seepage. The US DOE has, in the past, developed and continues to implement measures to protect human health and the environment by complying with prescriptive/risk based federal groundwater final, standards (60 FR 2854-2871 *Standards for Remedial Actions at Inactive Uranium Processing Sites*; final rule US Environmental Protection Agency, January 11, 1995) in a cost effective and publicly acceptable manner.

### **I-2.2. Surface remedial actions**

Land contaminated with uranium mill tailings ranged from 8 ha at the Spook, Wyoming site to 248 ha at the Ambrosia Lake, New Mexico site. Most sites resided either in urban settings or in river flood plains. The stabilization of the surface contamination at the sites was almost evenly divided between on site and off site disposal. The amount of contaminated materials ranged from approximately 65 000 cubic metres to 4 400 000 cubic metres.

### **I-2.3. Climate**

All UMTRA sites except Canonsburg, Pennsylvania site are in the western United States of America, generally in arid and semiarid environments. Fifteen sites are in dry climates and receive 30 to 50 cm annually; and three sites receive more than 50 cm annually.

### **I-2.4. Surface water**

Twenty-two sites are near surface water bodies, including major river such as the Colorado River. Perennial streams and ponds occur near many of the sites. Ephemeral and intermittent washes and arroyos also occur near many of the sites.

### **I-2.5. Groundwater**

Groundwater contamination in varying degrees has been observed at all but one of the sites. Milling activities at some of the sites created subsurface zones of saturation with contaminated groundwater in geological formations that previously did not contain groundwater. Seepage of contaminated water has affected the naturally occurring underlying aquifers at most of the sites. Some of the common constituents of concern include uranium, nitrate, sulfate, molybdenum, and selenium.

The estimated total volume of contaminated groundwater at the UMTRA sites is 40 000 000 cubic metres. At sites with contaminated groundwater, the per cent of off site contamination ranges from none to 98 per cent.

### **I-2.6. Ecological resources and wetlands**

Most sites are in areas dominated by desert shrub or desert grasslands or desert grassland plants. Riparian plant communities along rivers, streams, washes, and arroyos occur at or near most sites. Wetlands occur at or near many of the sites.

### **I-2.7. Land use**

Land use in and around UMTRA sites in urban areas ranges from industrial and commercial to residential and public. In rural settings, land use includes farming and ranching. Groundwater is often used for domestic uses, watering livestock, and irrigation. No one is drinking contaminated groundwater at any of the sites.

### **I-2.8. Remedial action selection process**

The DOE is in progress of continued site characterization, monitoring, and making remedial action decisions. The UMTRA Project has implemented a decision making framework for selecting site specific groundwater remedial action remedies that are protective of human health and the environment. The determination of site specific groundwater remedial action strategies takes into account site specific groundwater conditions; human and environmental risks, public participation, and cost. The approach is designed to be sufficiently flexible to allow for interim actions, such as alternate water supply systems, and institutional controls.

The decision making framework utilizes a logic diagram to identify the appropriate groundwater remedial action strategy or strategies for a site. Each termination step in the decision process is protective of public health and the environment. The final Programmatic Environmental Impact Statement for the Uranium Mill Tailings Remedial Action Groundwater Project (October 1996) details the logic supporting the programmatic decision making framework.

To date, UMTRA experience with remediation options for contaminated groundwater has identified critical areas of importance that include, but are not limited to:

- Public involvement;
- Up-front planning;
- Consistent and programmatic approach to decision making;
- Uncertainty analysis; and
- Cost benefit analysis.

Philosophical approaches to establishing clean up goals can be based on site specific risk coupled to applicable or relevant and appropriate requirements such as CERCLA guidance or prescriptive and generic standards such as UMTRA requirements. Both can be effective. The United States of America currently is using both approaches.

## **Annex II**

# **REMEDIATION OF RADIONUCLIDE CONTAMINATED GROUNDWATER IN CANADA**

## **II-1. BACKGROUND**

Canada has had a relatively long history in the mining, milling, and refining of radium and uranium ores, with significant activities dating back to the 1930s when radium was a valuable isotope. During World War II, the focus of ore extraction and processing shifted to uranium, and in 1946 Canada's first research reactor went critical. The prototype of the CANDU reactor went critical in 1962, and currently there are 22 CANDU power reactors (20 in Ontario, 1 each in Quebec and New Brunswick) supplying electricity to the grid. Research reactors are currently operated at 7 universities and institutes across the country, and the Chalk River Laboratories (CRL) operates both research and isotope production reactors. Canada has never had a nuclear weapons programme, although experimental reprocessing of spent fuel was undertaken at CRL in the 1950s. Since that time, the power reactor programme has employed natural uranium in a once through cycle, and the enriched fuel used in some research reactors is not recycled. Isotope enrichment operations in the country are limited to the production of heavy water.

Uranium mine and mill tailings represent the largest quantity of radioactive wastes in Canada, with a total mass exceeding 100 000 000 tonnes. Most of this volume is located in the Elliot Lake area, where the uranium ores and, consequently, the tailings, contain large quantities of pyrite. Sulphide oxidation has been the key influence on contaminant mobilization from tailings areas, and three strategies have been applied to manage this contamination. Lime and barium chloride additions are being used to precipitate sulphates, metals, and radionuclides from tailings dam seepages and groundwater plumes that have discharged to the surface. The flooding of tailings, and, on a more experimental basis, the addition of large quantities of degradable organics to the surface of tailings, have been used to reduce oxygen ingress and the consequent oxidation of sulphides. Experimental tests using reactive permeable barriers are also in progress. Permeable walls of organic materials (inoculated with anaerobic bacteria), calcium carbonate, and sand and gravel have been placed in shallow aquifers containing low pH, high sulphate, and high dissolved metal contaminant plumes. Sulphate reduction to sulphide in the reactive wall, coupled with neutralization, has significantly reduced groundwater contaminant concentrations. Studies to determine the longevity of the reactive permeable walls are continuing.

Uranium and radium refinery wastes have been another (historic) source of shallow aquifer contamination. Uranium, arsenic, and (rarely) radium contamination of near surface groundwaters has resulted from the storage of these wastes in shallow trenches. At all of the sites affected by such contamination, the aquifer's radioactive contamination has been controlled or restricted to local flow systems that discharge into nearby wetlands, and lime and ferric chloride additions have been used to co-precipitate the contaminants. All of the refinery wastes at these sites are scheduled to be removed to a permanent disposal facility. The current treatment of discharging contaminated groundwaters will continue until the source materials have been removed. Following the removal, groundwater contamination will be re-assessed to determine whether any other remedial actions will be required.

A number of surficial aquifers at Chalk River have been contaminated with radionuclides released from infiltration pits and solid waste storage sites. Two groundwater collection and treatment operations are currently running, with <sup>90</sup>Sr being the target radionuclide. In one case,

groundwater is collected following discharge to a small surface watercourse, while, in the other, a series of wells are used for collection. Treatment consists of the addition of calcium hydroxide, sodium carbonate, and powdered zeolite to remove the radionuclides through a combination of precipitation and sorption, followed by filtration and solidification of the dewatered solids in cement. In one case, the process has been very effective in reducing  $^{90}\text{Sr}$  concentrations to drinking water levels while achieving a volume reduction of more than 700. In the second operation, the treatment has, to date, been only moderately successful, and process refinement is continuing. Design work for a passive remediation system, consisting of a funnel and gate groundwater collector followed by gravity flow through clinoptilolite, is in progress for a third  $^{90}\text{Sr}$  plume at Chalk River.

All of the activities listed above, except heavy water production, have generated radioactive wastes, and these wastes have been managed with varying degrees of effectiveness. To date, the contamination of groundwater with radionuclides has been a limited, but real, problem in Canada. Such problems fall readily into sources arising from the following:

- (1) uranium and radium mining and milling,
- (2) uranium and radium refining, and
- (3) post reactor wastes.

One other source of radioactive wastes has been the refining of niobium ores, and this issue will be addressed in the discussion of contamination of refining residues, because most of the known stock of niobium refinery waste is co-located with uranium radium refinery wastes.

## II-2. PURPOSE AND SCOPE

This contribution provides a short perspective on activities in Canada that address the remediation of groundwater that has become contaminated with radionuclides. A summary is given of activities in the cleanup and control of groundwater contamination arising from uranium mining and milling, from radium and uranium refining, and from research reactors and the commercial and medical uses of radioisotopes.

## II-3. URANIUM MINING AND MILLING

Although uranium mining has been conducted in a number of regions of Canada, by far the largest mining and milling operations were located in the vicinity of Elliot Lake, in central Ontario. Mines here operated from 1955 to 1996, when production ceased as the Elliot Lake ore bodies could no longer compete with the much higher grade deposits being developed in the Athabaska sandstone of northern Saskatchewan. Table II-1 summarizes the quantities and disposition of tailings generated at mines operated by Rio Algom — data from the Denison, Stanrock, and Canmet properties have yet to be compiled. The Elliot Lake area features a rolling, glaciated bedrock surface with intermittent, generally thin, deposits of glacial till and outwash sands and gravels. Most of the mill tailings (and mine waste rock) are stored in surface impoundments, frequently created by building permeable dams across low points in a bedrock controlled valley.

TABLE II-1. TAILINGS IN THE ELLIOT LAKE MINING DISTRICT

Mine	Operating Period		Tailings' Quantity		Effluent Treatment		Surface stabilisation
	From	To	Tonnes	Placement	From	To	
Pronto	1955	1960	4 000 000	Surface	1960s	present	1970s
Nordic	1957	1968	12 000 000	Surface	1980s	present	1970s
Lacnor	1957	1960	3 000 000	Surface	1980s	present	1970s
Buckles	1957	1958	0	Surface			
Milliken	1958	1964	0	Surface			
Quirke	1956	1990	46 000 000	Surface, flooded	1970s		1990s
Panel	1958	1990	16 000 000	Surface, flooded	1970s		1990s
Spanish American	1958	1959	500 000	Deep lake			

The mill tailings consist predominantly of sand and silt sized particles, and possess moderate permeabilities in many of the impoundment areas. The hydraulic conductivity of bedrock in the Elliot Lake area is generally very low, and local glacial till, where present, is also a relatively low permeability material. Water discharged to the tailings and precipitation in excess of evapotranspiration requirements infiltrate through the tailings, but when the tailings basin is located over bedrock or till, little additional downward movement is possible. Where permeable dams surround all or portions of the tailings basin, much of the tailings infiltration drains through the dams to surface watercourses. Such flow systems are widespread among the Elliot Lake tailings impoundments, and barium chloride and lime treatment systems in settling basins are the norm for treating contaminated surface waters and dam seepage. When permeable materials underlie the tailings, however, water that percolates through the tailings may recharge the aquifer and migrate away from the tailings area in the subsurface, and there are some cases where this has indeed happened.

The Elliot Lake tailings contain up to 7 weight per cent of pyrite, and the dominant reaction in tailings pore water is the oxidation of the iron sulphide, producing very high concentrations of dissolved iron (>5000 mg/L) and sulphate (11 000 mg/L), and pH below 4.8. Many other elements are also dissolved in these waters, however, including uranium, radium, and decay chain thorium and actinium. Morin et al. reported uranium concentrations of as much as 8500 µg/L, and <sup>223</sup>Ra, <sup>226</sup>Ra, <sup>227</sup>Ac, and <sup>227</sup>Th concentrations of 3.1, 5.9, 11.8 and 22.2 Bq/L, respectively, in groundwater in a sand aquifer which is adjacent to the Nordic Main tailings area near Elliot Lake [II-2, II-3].

In those studies [II-2, II-3], the authors found that the plume of tailings contaminated groundwater was advancing through the sand aquifer at velocities of up to 10 m/a, whereas

groundwater velocities were on the order of 400 m/a. Although the calcite represents less than 1 weight per cent of sands in the Nordic Main aquifer that are unaffected by tailings leachate, calcite dissolution appeared to be the key agent in controlling the advance of radionuclides and other contaminants in the tailings plume. Increases in pH, calcium, and carbonate concentrations resulting from calcite dissolution result in the precipitation of gypsum, siderite, iron, aluminum, and silicon hydroxides, and co-precipitation or sorption of most of the radionuclides. Fifteen years after the Nordic Main tailings area went into service, groundwater uranium and  $^{226}\text{Ra}$  concentrations 20 metres down gradient of the tailings boundary were 10  $\mu\text{g/L}$  and 0.1 Bq/L, respectively.

Indirect groundwater remediation has been undertaken at the Elliot Lake uranium mine tailings sites (and at other mine tailings areas where sulphide oxidation is the dominant cause of pore water contamination) by reducing oxygen ingress to the tailings. One method to accomplish this has been to keep the tailings flooded, and this is being implemented at the Quirk and Panel tailings areas through the construction of impermeable dams [II-4]. A second, and currently experimental, approach has been to place substantial quantities of organic matter (e.g. peat) on the surface of tailings areas.

Permeable reactive walls are currently being investigated for the direct treatment of groundwater contaminated with leachate created by the oxidation of sulphide rich tailings. In the course of reducing sulphate in the leachate to low solubility sulphide, many metals are precipitated or co-precipitated, and the solution pH increases. In 1993, a test cell was installed in a sand aquifer adjacent to a Sudbury area tailings impoundment. The approach is conceptually simple (see Figure A-II-1). A key attraction is the passive nature of the treatment; no pumping system is required to deliver the contaminated groundwater to the treatment agents. Waybrandt et al. investigated mixtures capable of reducing sulphate to sulphide, using a variety of organic materials (composted sewage sludge, composted sheep manure, leaf mulch, wood chips, and sawdust) amended with agricultural limestone and inoculated with sediments from an anaerobic creek bed. Using simulated tailings leachate, the study concluded that mixtures of rapidly degradable and more refractory organic substrates (e. g. composted sewage sludge, wood chips, and sawdust) were most effective, providing a rapid onset to effective sulphate reduction (from 3300 to <35 mg/L in 20 days) and greater anticipated longevity [II-5].

In the field test of the method, Blowes et al. [II-4] used a mixture of leaf mulch, pine mulch, and pine bark as the source of organic carbon, agricultural limestone for pH control, anaerobic creek sediment as a source of sulphate reducing bacteria, and coarse sand and gravel (to increase permeability). Within 7 months of installation, sulphate concentrations decreased from >3500 mg/L up gradient of the reactive wall to <250 mg/L at the down gradient boundary of the reactive wall, with  $\text{SO}_4^{2-}$  concentrations of <10 mg/L within the wall. Ferrous iron concentrations decreased from >600 mg/L to <5 mg/L. The pH increased rapidly from up gradient values of <5 to above 6.3. Similar concentrations were observed one year after installation of the test cell, and the experiment is continuing. Additional reactive walls containing different organic carbon sources have since been installed and are being tested. These tests are being conducted in a plume from nickel mine tailings, so there is no information on the effects on radionuclides, but it is probable that the precipitation of sulphides (and potentially aluminum and silicon hydroxides) would co-precipitate radioisotopes.

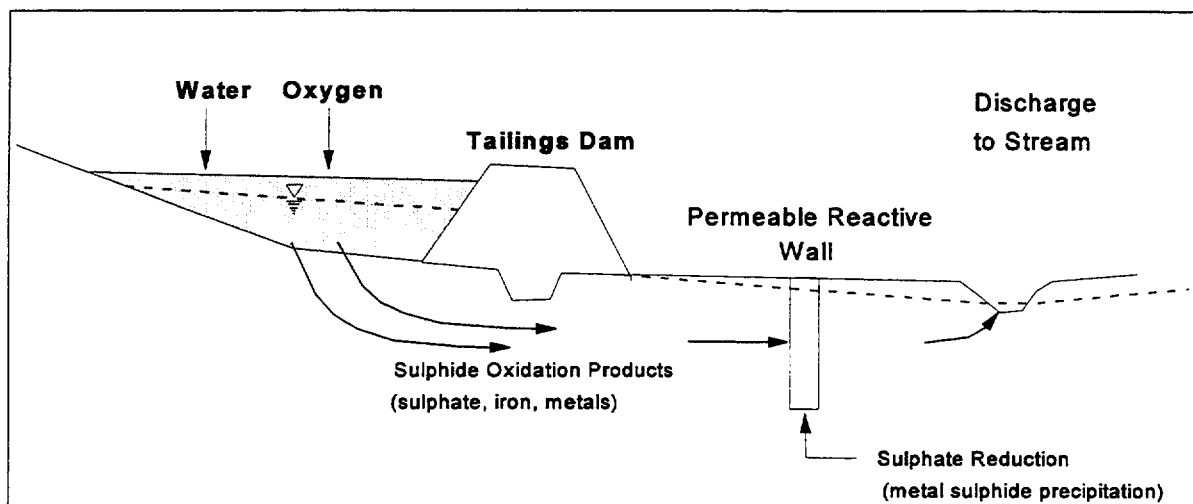


FIG. II-1. Schematic diagram of the treatment of tailings-contaminated groundwater using a permeable reactive wall.

Information on the operational life of such reactive walls is currently being collected. They will eventually require replacement of materials, as the organic substrates are consumed and as mineral precipitates reduce the permeability of the wall and direct groundwater flow around, rather than through, the cell.

#### II-4. URANIUM REFINERY WASTES

In 1932, Eldorado Gold Mines Ltd began construction of a radium refinery in the town of Port Hope, located on the north shore of Lake Ontario approximately 100 km east of Toronto. Routine radium production (and minor uranium refining) was in effect in 1935; in 1942 the refinery began large scale production of uranium, and this has continued to the present. In the 1940s the Canadian government purchased Eldorado and operated it as a crown corporation; in the 1980s the refinery operations were sold to Cameco Corp., the current operators.

Over the course of the refinery's operation, wastes have ended up in a number of locations within Port Hope, and in two rural sites just west of the town, known as Welcome and Port Granby. Table II-2 lists the periods of use for each of the sites or regions where refinery related wastes are located, and provides a summary of the volumes and estimated average concentrations of the key contaminants in the source materials. Since 1988, refinery wastes have been either recycled or placed in retrievable storage on the refinery property

Wastes in Port Hope were distributed in dumps in ravines, in former sand and gravel pits, in the local municipal landfill site, and in some cases found their way into backfill around buildings in the town. Apart from the municipal landfill site, refinery wastes and contaminated soils have largely been removed some industrial and refinery wastes were transferred to the Welcome site in the early 1950s, and the contaminated soils stored at Chalk River were removed from a variety of locations in Port Hope in a programme that focused in particular on commercial and residential properties. Uranium and arsenic are present in groundwaters down gradient of the municipal landfill site, but this is augmented with more conventional landfill leachate species.

There are no local groundwater users, and to date groundwater remediation has been limited to the placement of a low permeability cover on the landfill site.

TABLE II-2. VOLUMES AND AVERAGE CONTAMINANT CONCENTRATIONS IN URANIUM REFINERY WASTES

		Port Hope	Welcome	Port Granby	Chalk River
Dates of Use	From	1932	1948	1955	1976
	To	1948	1955	1988	1977
Material Volumes					
Radium Waste	m <sup>3</sup>		7650	6800	
Industrial Refuse	m <sup>3</sup>		3750	55000	
Refinery Wastes	m <sup>3</sup>			89700	
Contaminated Soil	m <sup>3</sup>	119 000	154 000	149000	60000
Contaminated Harbour Sed.	m <sup>3</sup>	54 000			
Average Concentrations					
<sup>226</sup> Ra	Bq/g	8	40	50	4
Uranium	mg/kg	150	500	1000	700
Arsenic	mg/kg	150	4000	4000	100

Wastes at the Welcome site are located in shallow trenches excavated in sands and gravels, at the top of a 5% slope towards the northwest and a watercourse about 500 m from the trench area. The sand and gravel is up to 15 m thick beneath the trenches, but thins down slope towards the watercourse. This thinning is marked by springs and seepages where flow in the shallow aquifer discharges to surface. Between 25 and 60 m of silty sand till underlies the sand and gravel, limiting connection between the shallow aquifer and the region's limestone bedrock, which is at least locally an aquifer. Trench depths in this unsaturated zone operation were constrained by the shallow water table, but infiltration carries contaminants into the aquifer, where they migrate to the northwest and discharge in the hillslope seepages.

Groundwater concentrations of up to 0.29 Bq/L of <sup>226</sup>Ra, 3600 µg/L of U, and 8000 µg/L of As have been measured in observation well samples at the Welcome site. Groundwater discharge from the hillslope springs, and surface runoff from the site, are routed to treatment ponds where ferric chloride is added, and then to a pair of sedimentation ponds, before draining to the local creek. On average, the treatment reduces arsenic concentrations to less than 25 µg/L, radium to less than 0.06 Bq/L, and uranium to less than 330 µg/L. To date, there has been no evidence of groundwater contaminant transport beyond the local springs and surface water treatment system.

The Canadian federal government accepted responsibility for wastes generated by the Port Hope refining operations up to the time of sale to Cameco. The government has been seeking a permanent disposal facility for the Port Hope area wastes, and plans to remove wastes and contaminated soils from the Welcome site when a disposal facility is available. The current strategy for the Welcome site, therefore, is to maintain the current surface and groundwater discharge collection and treatment operation until all of the solid wastes and contaminated soils have been removed from the site, and assess the need and options for additional groundwater remediation at that time.

The Port Granby waste management facility is located adjacent to a steep, 35 m high bluff along the Lake Ontario shoreline. The site consists of a nearly level area of about 1.5 hectares, bounded on the south by the Lake Ontario bluff, and to the east and west by short, steep, ravines that have eroded back from the lake shore. Between 1960 and site closure in 1988, most of the waste and contaminated soil were placed in trenches excavated approximately 5 m into the local surficial sands, with trench depth limited by the water table. Between 1955 and 1960, however, wastes were also dumped into the northern ends of the two ravines, locally known as the East and West Gorges.

The sands in which the waste trenches were excavated are underlain by up to 18 m of dense silts, 3 to 5 m of sandy silt till, a second sand unit up to 20 m thick, a basal clayey silt till, and Paleozoic carbonate bedrock. The silt and sandy silt till isolate groundwater flow systems in the deeper sand and the bedrock from groundwaters affected by leaching from the waste trenches. Shallow groundwater contaminated by infiltration through the trenches and through the wastes that were dumped into the head of the site's two ravines discharge in springs and seepages in East and West Gorges. Collection ponds have been constructed near the mouths of the two ravines, and the water is then pumped back up to treatment ponds located at the northern edge of the site.

The maximum concentrations of  $^{226}\text{Ra}$ , U, and As in samples from observation wells on the Port Granby site reported in [II-13] are 2.88 Bq/L, 145 000  $\mu\text{g/L}$ , and 1660  $\mu\text{g/L}$ , respectively [II-6]. Hydroxides in some of the refinery wastes have generated groundwater with a pH up to 11.8, and leachates are very elevated in a wide variety of major ions and fluoride. The latter element arises from waste produced in the course of manufacturing  $\text{UF}_6$ , where much of the dissolved arsenic in the contaminated groundwater is present as a fluoride complex. Contaminated groundwaters from the two ravine discharge areas, as well as surface runoff, are treated with ferric chloride, following acid addition to reduce the pH to 7, and the water is then routed through two settling basins before release from the site. Treated waters contain, on average, 0.14 Bq/L of radium, and 2100  $\mu\text{g/L}$  of As. No data for uranium concentrations in the treated water were reported. The ineffectiveness of the ferric chloride treatment at Port Granby (in contrast to operations at the Welcome Waste Management Facility) has been linked to the complexation of arsenic with fluoride in the Port Granby site's contaminated ground and surface waters.

As is the case at Welcome, operations at Port Granby are expected to continue in their current mode until the solid wastes have been removed and transported to a permanent disposal facility. Part of the subsequent remediation at the Port Granby site will be a re-evaluation of groundwater contamination and an assessment of the need for further groundwater remediation.

In 1976, a programme was launched to remove soils contaminated with uranium refinery waste from several locations in Port Hope. Clean ups of residues from a niobium smelting operation in Ottawa, and radium contaminated soils from a site near Toronto were also under way

at the time, and AECL agreed to establish a storage facility at the Chalk River Laboratories (CRL), locally known as Area F. A total of 119 000 tonnes (74 000 m<sup>3</sup>) of contaminated soils were placed in a surface storage site, with a final cover consisting of 0.3 m of clayey silt and 1 m of sand and topsoil. The wastes were placed on 2 to 5 m of unsaturated sand, overlying a shallow groundwater flow system that discharges to a nearby wetland.

The clayey silt cover installed on Area F was intended to reduce the amount of infiltration through the contaminated soil, but this barrier has been largely ineffective. Surface waters draining the Area F groundwater discharge area are routinely monitored, and to date there has been no evidence of contaminants from the site. The site performance has also been monitored at 5 year intervals by drilling and sampling soils and pore waters through the wastes and the underlying sands. The most recent such study was conducted in 1993; it found that radium has continued to exhibit almost no mobility, but that both arsenic and uranium are being leached from the contaminated soils. To date, however, neither of these elements have entered the local groundwater flow system both are removed from infiltrating water by sorption to iron oxide coatings on the sands that lie beneath the waste [II-7] concluded that there was sufficient capacity in the iron oxyhydroxide coatings of the sands to retain arsenic inputs for several decades and that the same was likely to be the case for uranium.

## II-5. POST REACTOR WASTES

Canada has five sites where radioactive wastes arising from reactor operations (and to a very limited extent from accelerator operations) are stored. The three Canadian provincial utilities that supply nuclear generated electricity (Ontario Hydro, Hydro Quebec, and New Brunswick Power) store spent fuel in a combination of pools and above ground dry storage canisters located at the generating stations. Each also manages low and intermediate level wastes in a combination of above ground storage and in shallow subsurface engineered structures. Ontario Hydro has centralized all of its low and intermediate level wastes at the Bruce Nuclear Power Development site in south central Ontario, while Hydro Quebec and New Brunswick Power manage all wastes at the station. AECL operates waste management facilities at its research facilities the Whiteshell Laboratories (WL) in Manitoba and the Chalk River Laboratories (CRL) in Ontario. Facilities at WL provide storage only for wastes generated on site; CRL manages its own wastes and those arising from medical and commercial isotope production and use in Canada.

At all locations other than CRL, waste management operations date from, at the oldest, the mid 1960s. By that time, there was sufficient experience in radioactive waste handling and storage that the facilities have been quite successful in preventing the release of radionuclides, and no significant groundwater contamination problems have been identified. Such has not been the case at Chalk River, where operations began in the mid 1940s, and a number of plumes of groundwater containing radionuclides have developed. The remainder of this section will discuss conditions and remediation operations at CRL.

The CRL site consists of a 37 km<sup>2</sup> property bordering the Ottawa River, 190 km northwest of Ottawa. Bedrock at the site consists of crystalline high grade metamorphics of Precambrian age, predominantly with a granitic composition. The area has been repeatedly glaciated, leaving a rock surface with a hummocky topography and moderate relief; about 15% of the CRL site is bedrock outcrop or areas with less than 1.5 m of unconsolidated sediment. Unconsolidated

sediments of late glacial or post glacial age cover the remainder of the site these consist of bouldery sand till, fluvial sands (and minor silts) aeolian sands in sheet and small dune deposits, and local recent accumulations of organic sediments in wetlands. Throughout Chalk River's operating history, dune sands have been used for all subsurface waste management operations. Waste placement above the water table has been a requirement for all facilities, and the only places on the CRL site where depths to the water table are greater than 3 to 4 metres are upland regions in high permeability soils.

Figure II-2 is a map of the CRL site that shows the locations of waste management facilities. Table II-3 provides a brief commentary on features of the facilities that involve the storage of wastes in the subsurface. All of the sites overlie unconfined groundwater flow systems in sand aquifers, with flow velocities of tens of centimetres per day, and subsurface flowpath lengths of between 100 and 1000 metres, and as the table indicates, all except Area F have resulted in the contamination of these aquifers with radionuclides. Almost all of this contamination has resulted from wastes placed in soil trenches (most of which are covered only with sandy soils) and the aqueous wastes pumped to the infiltration pits of the Liquid Dispersal Area (LDA).

Despite the wide variety of radioisotopes present in the CRL waste management areas, the nuclides that have resulted in groundwater contamination that extends appreciably beyond the source areas are limited to a short list. By far the most widespread and abundant radionuclide in groundwater at CRL is tritium, almost all of which is present in the waste as tritiated water. Once released from the source, tritium is transported at groundwater velocity, with no interaction with aquifer solid phases. Second in terms of both frequency of occurrence and quantity in site aquifers is  $^{90}\text{Sr}$ , but this isotope has been the key target for groundwater remediation programmes that have been instituted to date. Strontium interacts with the granitic sands of the CRL aquifers, and though the reactions extend beyond ion exchange sorption is limited and mostly reversible. Radiostrontium generally migrates at a few per cent of the velocity of the transporting groundwater.

Radiocarbon, most of which is known or expected to be present as carbonate species, does not appear to interact appreciably with local aquifer solids. Observed concentrations in site aquifers are much lower than either tritium or  $^{90}\text{Sr}$ ;  $^{14}\text{C}$  is notable in the Area C aquifer plume primarily because it is the only isotope with an appreciable groundwater distribution other than tritium. The remaining radionuclides noted in Table A-II-3 to be present in CRL groundwater ( $^{60}\text{Co}$ ,  $^{106}\text{Ru}$ , and  $^{125}\text{Sb}$ ) are encountered in aquifer plumes emanating from the (LDA). The mobile components of these isotopes are all known to be present as anionic or neutrally charged species. Almost all other radionuclides have undergone only very limited migration since their placement in CRL waste management areas, being retained in the wastes and by sorption to natural sediments in the unsaturated zone. Small quantities of some of these radioisotopes have been transported into the underlying aquifer, but their distribution, even after as much as 50 years, is limited to their source areas.

There are two groundwater remediation projects currently operating at CRL, and a third is under development. In all three cases, reduction of  $^{90}\text{Sr}$  to concentrations equal to or less than the Canadian federal maximum concentration for drinking water (currently 5 Bq/L, 10 Bq/L before 1996) is the key treatment target, although in the case of the Chemical Pit plume, other isotopes are present and at least their partial removal is desirable. Both of the current remediation systems are pump and treat operations; the system under development will operate by passive treatment.

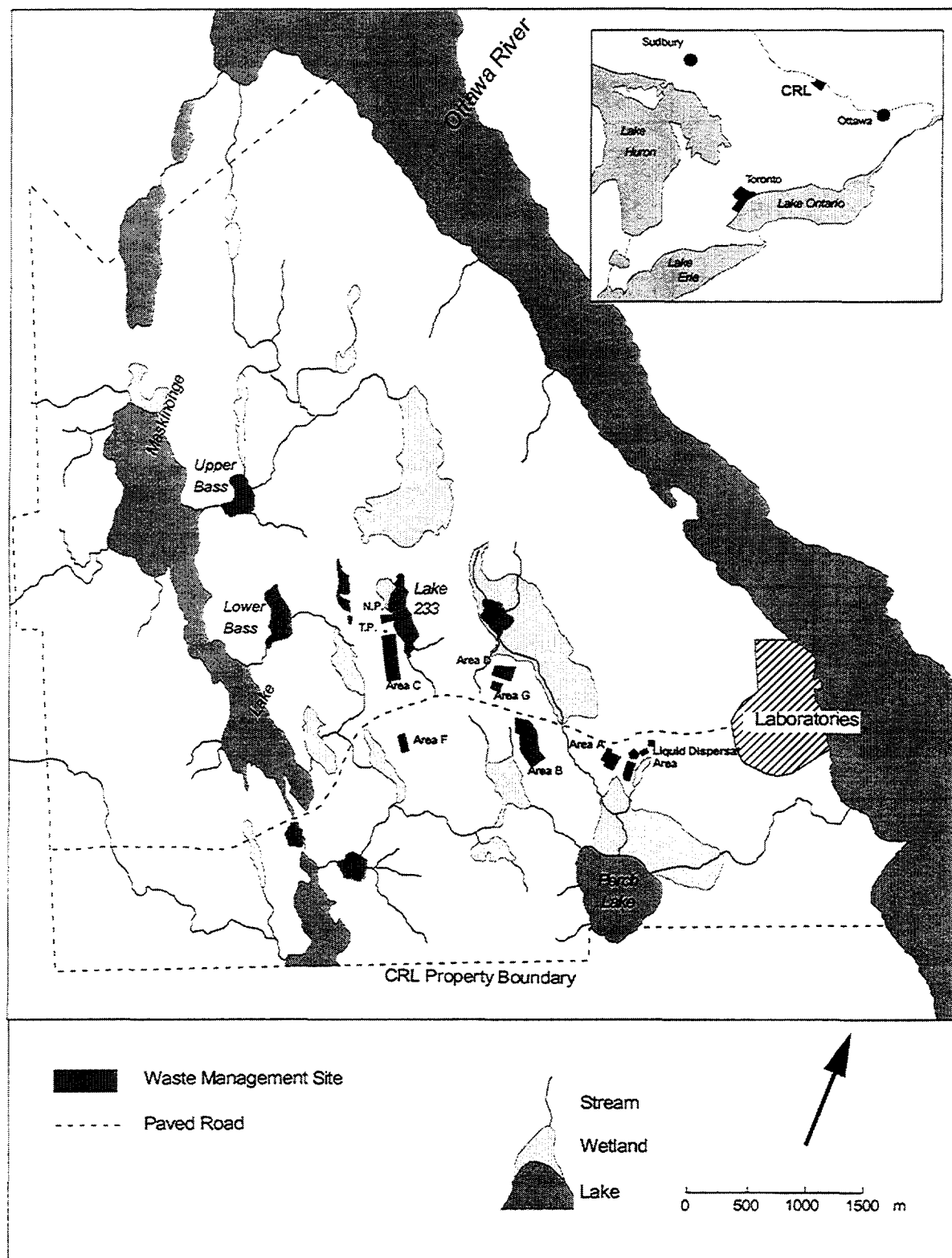


FIG. II- 2. Map of the Chalk River Laboratories.

TABLE II-3. OPERATING PERIODS AND GENERAL CHARACTERISTICS OF SUBSURFACE WASTE MANAGEMENT AREAS AT CRL

Waste Mgmt. Area	Period of Operation	Description
<i>Solid Wastes</i>		
Area A	1946–1953	1.2 ha compound used for solid wastes in engineered structures and soil trenches, and aqueous liquid wastes in pits and trenches. Contains a wide variety of radionuclides; <sup>90</sup> Sr is the dominant groundwater isotope.
Area B	1953–	14 ha compound used for both low level (soil trench) and intermediate level solid waste (engineered structures) to 1963. Since then used for intermediate level waste only. The site contains actinides, fission and activation products, and medical/commercial isotopes. <sup>90</sup> Sr emanating from part of the soil trench area is the dominant groundwater isotope.
Area C	1963–	4.5 ha compound containing 90 000 m <sup>3</sup> low level solid waste in soil trenches. The site contains tritium, fission and activation products, and medical/commercial isotopes. Tritium and <sup>14</sup> C are the dominant groundwater isotopes.
Area F	1976–1977	74 000 m <sup>3</sup> of soil from Port Hope, Ont. Contaminated with Ra, U, As. There is no groundwater contamination from the site.
<i>Liquid Wastes</i>		
LDA	1953–	Four compounds containing infiltration pits for aqueous wastes, with one compound still in use. The wastewaters contain tritiated water, mixed fission and activation products, and small quantities of actinides. Tritium, <sup>90</sup> Sr, <sup>60</sup> Co, <sup>106</sup> Ru, and <sup>125</sup> Sb are the dominant groundwater radionuclides.
Nitrate Plant	1953–1954	Accidental release of mixed fission products (now predominantly <sup>90</sup> Sr and <sup>137</sup> Cs) to an infiltration pit. <sup>90</sup> Sr is the dominant groundwater radionuclide

The first groundwater pump and treat operation at CRL was established to remove <sup>90</sup>Sr from groundwater contaminated by wastes in soil trenches in Area B. The contaminant plume terminates in the upper reach of a watercourse located approximately 40 m from the waste management area's perimeter, and the watercourse receives very little uncontaminated groundwater. As a result, a collection sump installed in the stream just downstream of the termination of the contaminant plume has served as the collection point.

Collection and treatment at the "Spring B" began in 1992, initially as a batch mode operation, with upgrading to semi continuous treatment in 1993 and to a computer automated system in 1995. The treatment process involves combined chemical precipitation and ion exchange onto powdered zeolite added directly to the wastewater, followed by solid liquid separation using cross-flow micro filtration and gravity settling. The filtrate is neutralized and passed through cation exchange zeolite columns in a final polishing step prior to discharge. The process operates at room temperature, and the generic treatment technology has been patented. During 1996, the plant treated  $1.8 \times 10^6$  L contaminated groundwater, and produced 12 standard drums (205 L) of cemented solid waste that meets US NRC guidelines for leachability and mechanical strength. The 730-fold volume reduction is slightly short of the design objective of a factor of 1000. The concentration of <sup>90</sup>Sr in the treated effluent water averaged 7.8 Bq/L,

meeting the Canadian drinking water objective in place at the time that treatment operations commenced.

The chemical pit, one of the infiltration pits in the liquid dispersal area, is less than 100 m from the discharge zone for the local unconfined sand aquifer. The primary remedial action at the chemical pit has been to remove it from service, and this was done in 1992 when waste treatment systems at CRL were upgraded. A variety of radionuclides (dominated by  $^{90}\text{Sr}$ ) continue to be leached from contaminated sediments, however, and in 1994 construction of a modular treatment plant started. At the same time, a groundwater interception system consisting of a line of four pumping wells and four observation wells used for draw down control was installed. The automated treatment plant began processing contaminated groundwaters in late 1995 using lime sodium carbonate additions to co-precipitate dissolved radionuclides, with powdered sorbent addition to improve removal efficiency, and rotary drum vacuum filtration to separate solids and liquids. The solids are mixed with cement and drummed.

To date, removal efficiency has not been as good as that achieved in the Area B operation — effluent  $^{90}\text{Sr}$  concentrations are currently about 50 Bq/L from a feed containing an average of 650 Bq/L. The removal of  $^{60}\text{Co}$ , which is present in the feedwater as an anionic complex with dissolved organics at concentrations of up to 400 Bq/L, has been limited to date to about 30%. Given the short half-life of  $^{60}\text{Co}$ , and the assumed institutional control period of 100 years, this low removal efficiency is not seen as a major problem, although higher removals are certainly desirable. Of greater concern has been the fact that in 1996 the volume reduction from the feedwater to the drummed solidified waste has been limited to a factor of 285. Efforts to improve the treatment plant performance are continuing.

A third groundwater remediation project at CRL is in the planning stage, again with  $^{90}\text{Sr}$  as the target isotope. In 1953–54, a pilot plant for the decomposition of ammonium nitrate liquid wastes arising from fuel reprocessing experiments was constructed and operated; processing was terminated following a major upset that released mixed fission products to an infiltration pit overlying a shallow sand aquifer. With decay, the only two isotopes that are still present in significant quantity are  $^{137}\text{Cs}$  and  $^{90}\text{Sr}$ . Caesium has remained within 20 m of the infiltration pit, but a 350 m long plume of  $^{90}\text{Sr}$  in the local sand aquifer has formed and is now at the edge of the groundwater discharge area. A passive system intended to remove the  $^{90}\text{Sr}$  from groundwater prior to surface discharge is being designed, using a “funnel and gate” wall to direct the contaminated water into a collection sump, and a gravity flow treatment cell — a bed of clinoptilolite, to remove  $^{90}\text{Sr}$ .

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## **Annex III**

### **CLEANING OF RADIONUCLIDES FROM GROUNDWATER AT SITES CONTAMINATED BY URANIUM MINING AND MILLING ACTIVITIES IN THE RUSSIAN FEDERATION**

#### **III-1 INTRODUCTION**

Despite the sharp decrease in production output and a number of government decrees on the environmental protection, informed experts agree that the environmental conditions in Russian industrial regions are still deteriorating. The communal waste products and industrial wastes are seen to be growing and the atmospheric pollution is increasing (especially since rapid growth of automobile transport with extremely low performance of exhaust control systems). Many water sources have become contaminated due to insufficient purification of effluents and their recycling for domestic and industrial purposes.

In a previous report (Development in Cleaning Effluents of Branch Facilities, by V.V. Shatalov et al., 1995), it was noted that water recycling in industrial uranium production facilities was one of the most economical and resource-saving means of ensuring environmental protection. The major advantages for this approach include the following:

- a lesser degree of purification is required for the recycling water as compared with the quality of effluents dumped into reservoirs;
- it is only necessary to treat certain flows from selected regions, and not the entire volume of the combined liquid wastes;
- stage-wise introduction of local installations is feasible; and
- there can be concomitant utilization of valuable components in the liquid wastes.

The economy of this methodology lies with the following:

- reduction in total use of water and the amount of water to be dumped (e.g. the main task will result in a 40–50% reuse of cleaned effluents for industrial and domestic needs);
- reduction in amounts or elimination of the fines charged for the dumping of untreated liquid waste; and
- lower capital and operational costs when introducing local purification for separated flows (i.e. in comparison with cleaning up all liquid wastes to permissible concentration limits).

Calculations show that new facilities equipped with water recycling systems are, on the average, ten times less expensive than construction of effluent cleaning systems for sanitary treatment of the same capacity. A facility that is changed over for water recycling requires 8–10 times less fresh water (e.g. this was demonstrated at the Malyshevskaja deposit's dressing plant, whose design includes integration of effluents after ore preparation, comminution, beneficiation and cleaning of tails). The integrated discharges are directed to ore preparation and comminution operations and, after demineralization by adsorption, to flotation and treatment of the concentrate.

### III-2. GROUNDWATER TREATMENT AT URANIUM MINES

The main direction for decreasing the environmental impacts of uranium mine waters contaminated by radionuclides is purification and the controlled use of the cleaned water for technical and household purposes.

It should be emphasized that, in general, not all of the cleaned mine water could be used just for the mine itself, and so it is important to provide other possibilities of water utilization.

Depending on hydro technology and the radiochemical conditions, the cleaning of mine waters can be conducted by means of the following:

- co-precipitation,
- adsorption, or
- combination of absorption/adsorption (sorpitive cleaning).

The profitability of the process is determined by the rate of uranium recovery.

The uranium mine waters typically contain bicarbonate and carbonate ions in plentiful supply. Therefore, neutral and weakly alkaline mine waters contain uranium as di- and tri-carbonate complexes. The effective recovery of uranium can be achieved only after destruction of the complexes.

Radium is co-precipitated by barium sulphate after the addition of barium salts. Sulphates of aluminium and iron (in the ferrous state) were used as coagulants and co-precipitating reagents.

In the case of sulphates at concentrations in mine waters of over 150 mg/L, the radium recovery of more than 90% was obtained after adding stoichiometric quantities of barium chloride at 40 mg/L. The influence of barium chloride consumption on the rate of radium precipitation is shown in Table III-1.

TABLE III-1. THE INFLUENCE OF BARIUM CHLORIDE CONSUMPTION ON THE PRECIPITATION OF RADIUM

BaCl <sub>2</sub> consumption, mg/L	Content of Ra in mine water, Bq/L		The rate of water purification, %
	Initial	Final	
20	2.6	0.30	88.0
40	2.6	0.24	90.6
60	2.6	0.24	90.7
100	2.6	0.02	99.3
160	2.6	0.06	97.7

For mine waters with a low sulphate content, it is advisable to use sulphate salts as coagulants for the effective recovery of radium, thorium and, in the case of uranium, a partial recovery. However, it is impossible to decrease the uranium concentration to less than about 0.3–0.4 mg/L.

The investigations for recovery of uranium and other radionuclides from sulphate-carbonate solutions by co-precipitation were carried out by mixing the solutions for 0.5–1 hour at a temperature 20C. The solutions were analysed after separation from the residue (Table III-2).

TABLE III-2. THE INFLUENCE OF SOME COAGULANT CONSUMPTION ON THE REMEDIATION OF MINE WATERS CONTAINING URANIUM AND OTHER RADIONUCLIDES

Reagents consumption, mg/L			Concentration in mine waters after purification		
BaCl <sub>2</sub>	FeSO <sub>4</sub> ·3-	CaO	U, mg/L	Ra, Bq/L	Th, Bq/L
-	-	-	1.5	2.6	3.7
35	20	60	0.5	0.10	0.37
35	40	120	0.5	0.11	0.15
35	60	70	0.5	0.09	0.20
35	80	75	0.48	0.06	0.06
35	120	125	0.46	0.10	0.06

The All-Russian Research Institute of Chemical Technology (ARRICT) has developed integrated circuits for cleaning mine waters of uranium deposits with complex saline composition. They provide a 25% removal in the case of Ra and remove 90% of total radionuclides. The developed technological schemes take into account the mine water type, as well as the radionuclide occurrence form. The major operations, in their sequential order, consist of the following:

- removal of suspended particles, radionuclides and bacterial impurities in a mixer and thickener, using barium chloride, polyacrylamide, aluminium sulphide and tri-sodium phosphate, followed by recleaning of the suspended particles on filters;
- treatment of mine water with barium chloride and polyacrylamide, followed by recovery of the uranium from the suspension through filtration on an ion exchange resin of the AMP type;
- treatment of mine water with barium chloride or pyrolusite, polyacryl amide with iron, or aluminum sulphate, followed by the recovery of uranium using an ion exchange resin of the BPI-1AP type.

The mine water cleaning scheme at the one uranium processing plant with a capacity of 2000 m<sup>3</sup>/h was introduced in 1993.

### III-3. DEVELOPMENTS IN THE PURIFICATION AND CONDITIONING OF DRINKING WATER

In 1992–1996, the ARRICT was conducting research on modern water purification and conditioning with technologies applicable to different regions of Russia. Special attention was given to environmentally threatened areas (i.e. the territories affected by the Chernobyl accident and the Chemical Enterprise “Mayak”, Chelyabinsk). To provide a universal level of operational

characteristics for the systems under development, it would be necessary to simultaneously remove the major contaminants from the water to be purified. Examples of such contaminants are as follows:

- organic and inorganic impurities, including insecticides and pesticides;
- colloidal impurities and suspensions containing up to 50% of such hazardous elements as lead, excessive quantities of iron, etc.;
- dissolved forms of toxic metals such as mercury, cadmium, nickel, chromium, manganese, etc., including certain forms of caesium and strontium; and
- harmful bacteria, microbes and other micro organisms.

As the basic purification method, adsorption was selected with application of locally developed materials (e.g. selective inorganic and organic ion exchangers) and additional micro filtration.

Integrated adsorption and micro filtration loadings provide a complex purification for all types of contaminants, as well as for caesium and strontium, as required by the World Health Organization.

On the basis of these developments, the Institute has designed individual and collective filters and has, in the pilot plant scale, manufactured about 5000 units. These filters have passed intensive testing in the Gomel region, Belarus, and the Kurgan region in Russia. Some new generation filter prototypes have been developed on the basis of the devised filtration and adsorption materials. These filters comply with all the current requirements.

The Research Institute Mosvodokanal Proekt has conducted tests on various filter types, including those of foreign make. These tests have proven that the filters, which combine selective adsorbency with micro filtration, exceed all other types in their performance characteristics.

In 1994, on the basis of these developments, the first Russian commercial certified operation was set up and utilized for the production of highly conditioned bottled water. The plant, with a capacity of about 9 m<sup>3</sup> per hour, was commissioned in early 1995 in the city of Zelenograd. The water purified at the plant has excellent taste and contains absolutely no bacterial micro flora, which makes it perfectly suitable for long term storage.

#### III-4. EXTRACTION OF RADIONUCLIDE Cs-137 FROM MILK, WHEY, AND WATER

This section discusses removal of radionuclides from milk, whey, and water and the findings and methodology discussed are broadly applicable to the remediation of radioactively contaminated groundwater.

In 1954, a number of accidents took place in several countries resulting in radioactive contamination of the environment. Since 1957, there have been approximately 10 such cases, including accidents in the Russian Federation. Through its accumulation with cattle fodder (i.e. contamination of plants), the radioactivity passes into the food chain, especially in dairy products.

Therefore, in May 1986, the caesium permissible concentration limit was set up for the food stuffs delivered into the European Community (EC) States from the third world and other countries. For dairy products and children's food, the permissible concentration limits for caesium-137 and caesium-134 are 379 Bq/kg, and, for all other products, 600 Bq/kg.

The Russian Federation Public Health Ministry has established the following temporary radionuclide limits permissible in main dairy products:

- Milk, drinking water = 370 Bq/L
- Condensed milk = 18500 Bq/L
- Dry milk, curds, sour cream = 3700 Bq/L
- Cheese, dairy butter = 7400 Bq/L.

After making the cheese and curds, the radionuclides will remain in the whey. Upon drying, 5000 tonnes or 250 freight cars of radionuclide-contaminated whey have been produced.

Throughout the world, there are only a few commercial plants that can decontaminate milk and milk whey. A very efficient and compact adsorption-decontamination treatment method has been developed in Russia for water and dairy products using mineral, organic/mineral or polymer adsorbents sized +0.63 mm. The method provides an effective decontamination of liquid natural or condensed dairy products with an activity level of 3700–370 000 Bq/L. Water or milk are treated in special apparatus for 1–2 hours with stirring, followed by the separation of saturated adsorbent on a fixed arch sieve or a vibrating screen. Depending on the thoroughness of decontamination, the radioactivity of the cleaned water is characterized by the data shown in Table III-3.

TABLE II-3. RESULTS OF WATER CLEANING WITH USE OF DIFFERENT ADSORBENTS

Adsorbent	Activity, $10^3$ Bq/L		Nuclide removal %
	Initial	Final	
Clinoptilolite	592	148	77
Silica gel	592	11.8	98
Silica gel	151.7	7.4	95
Silica gel	44.4	2.59	94

In case a more thorough decontamination of water should be required, the operation can be repeated with a fresh portion of adsorbent (the second stage). The separated adsorbent is then returned to the first stage. The adsorbent from the first treatment is subsequently disposed of by burial.

### III-5. DEVELOPMENT OF A LIQUID LOW LEVEL RADIOACTIVE WASTE TREATMENT

In 1986–1988, the Institute was carrying out research on synthesis and the properties of selective inorganic adsorbents. In 1989, the first Russian commercial plant for processing liquid low-level radioactive waste (LLRW) from the navy was set up at the Repair and Transport Enterprise (RTE) ATOMFLOT, Murmansk. Up to the present time, the RTE has processed over 5000 cubic metres LLRW of various composition, including saline decontamination solutions.

In 1994, the facility started processing effluents coming from the Russian Navy. The recently developed reconstruction programme will raise the plant capacity to 5000 m<sup>3</sup> annually. The plant will be capable of treating all LLRW types coming from the Severodvinsk region, solidifying the radioactive waste into cement blocks. The adopted approach fully meets the recommendations of the IAEA and the requirements of the new Radioactive Material Treatment Law.

## **Annex IV**

### **UKRAINE (CHERNOBYL EXCLUSION ZONE)**

#### **IV-1. OBJECTIVES AND SCOPE**

This contribution discusses experience in the area of remediation of radioactively contaminated groundwater, acquired in Ukraine<sup>1</sup> during decade of efforts aimed at mitigating the environmental consequences of the Chernobyl accident.

The main subject of this contribution is the remediation measures conducted at the Chernobyl Nuclear Power Plant (CNPP) site in the aftermath of Chernobyl accident to protect groundwater resources from radioactive pollution and to minimize transport of radioactivity between surface and groundwater pathways (see Figure IV-1).

The discussion contained in this Annex is divided in following topics:

- emergency phase groundwater protection measures (Section IV-4);
- problem of groundwater contamination at “Red Forest” radioactive waste dump site (Section IV-5);
- groundwater remediation of the CNPP cooling pond (Section IV-6);
- current management strategy for groundwater contamination problems at the CNPP site (Section IV-7).

Sections IV-4 to IV-6 are devoted to retrospective analysis of the efficacy of groundwater remedial measures conducted in the Chernobyl zone in 1986–1994, and Section IV-7 discusses results of recent groundwater risk assessment studies and implications from these analyses to groundwater remediation strategy. Lastly, we summarize in Section IV-8 the lessons from the groundwater remediation experience at Chernobyl, which may be applicable to management of other radioactively contaminated sites in other parts of the world.

#### **IV-2. CNPP SITE HYDROGEOLOGY**

The CNPP is situated in the northern part of the Kiev Polessje (Woodlands) Region of Ukraine. This region is characterized by a moderately continental humid climate net positive precipitation over evaporation. Precipitation averages 600 mm/a. The landscape is mostly flat. The soil mantle is composed of light mineral and organic soils having low clay content, low cation exchange capacity, and is slightly acidic. The first aquifer below the surface is the Quaternary unconfined aquifer composed mostly of highly permeable (hydraulic conductivity 1–10 m/d) alluvial and fluvio glacial sandy deposits (Figures IV-2 and IV-3). The depth to groundwater occurs 1–2 m in river flood plain areas to 10 m and more in watershed areas. The

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<sup>1</sup>After the Chernobyl accident, combined physical and chemical techniques have been used, some of which may also be useful for large scale groundwater remediation [IV-1–IV-7].

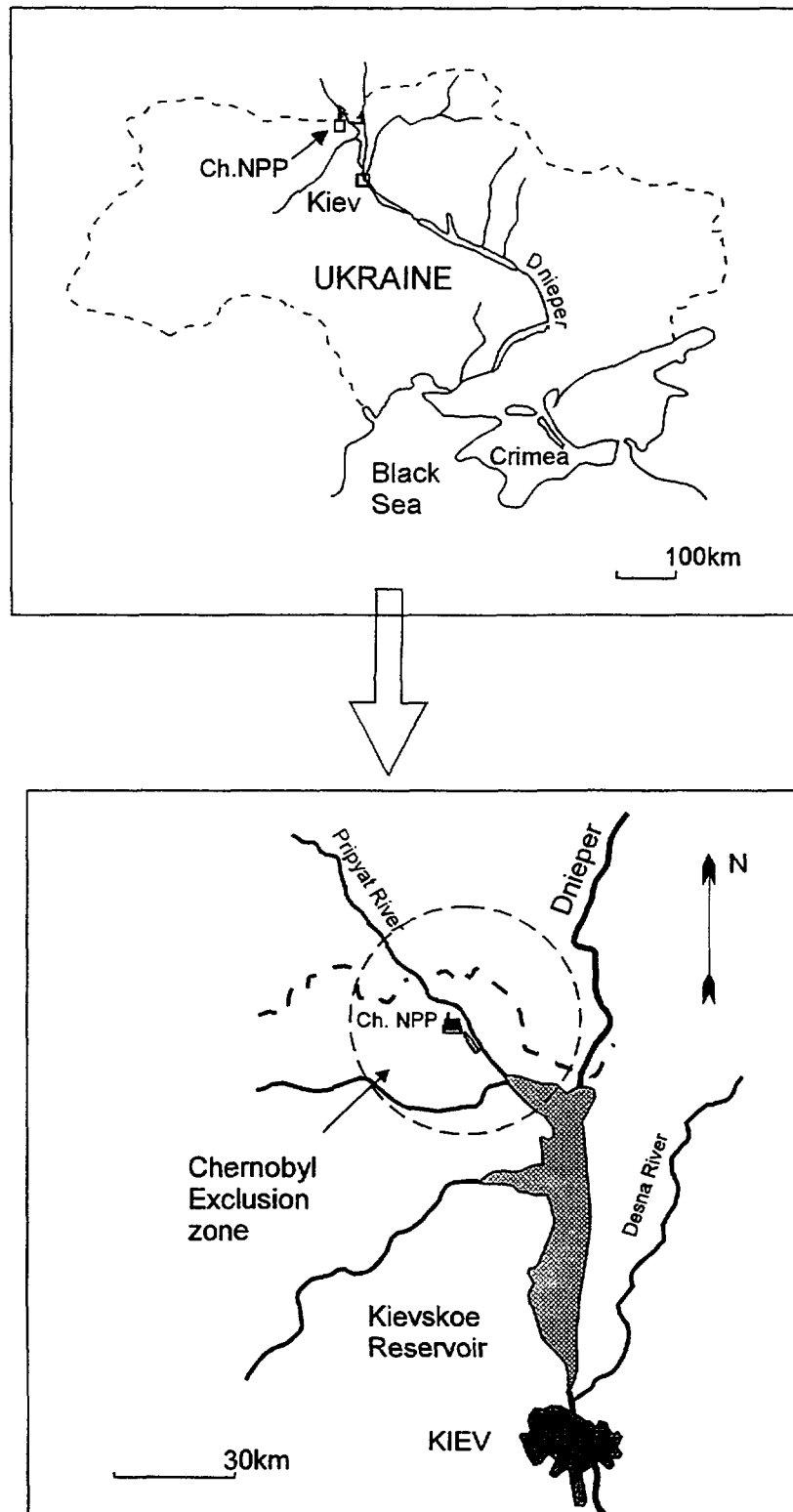


FIG. IV-1. Location of the Chernobyl nuclear power plant.

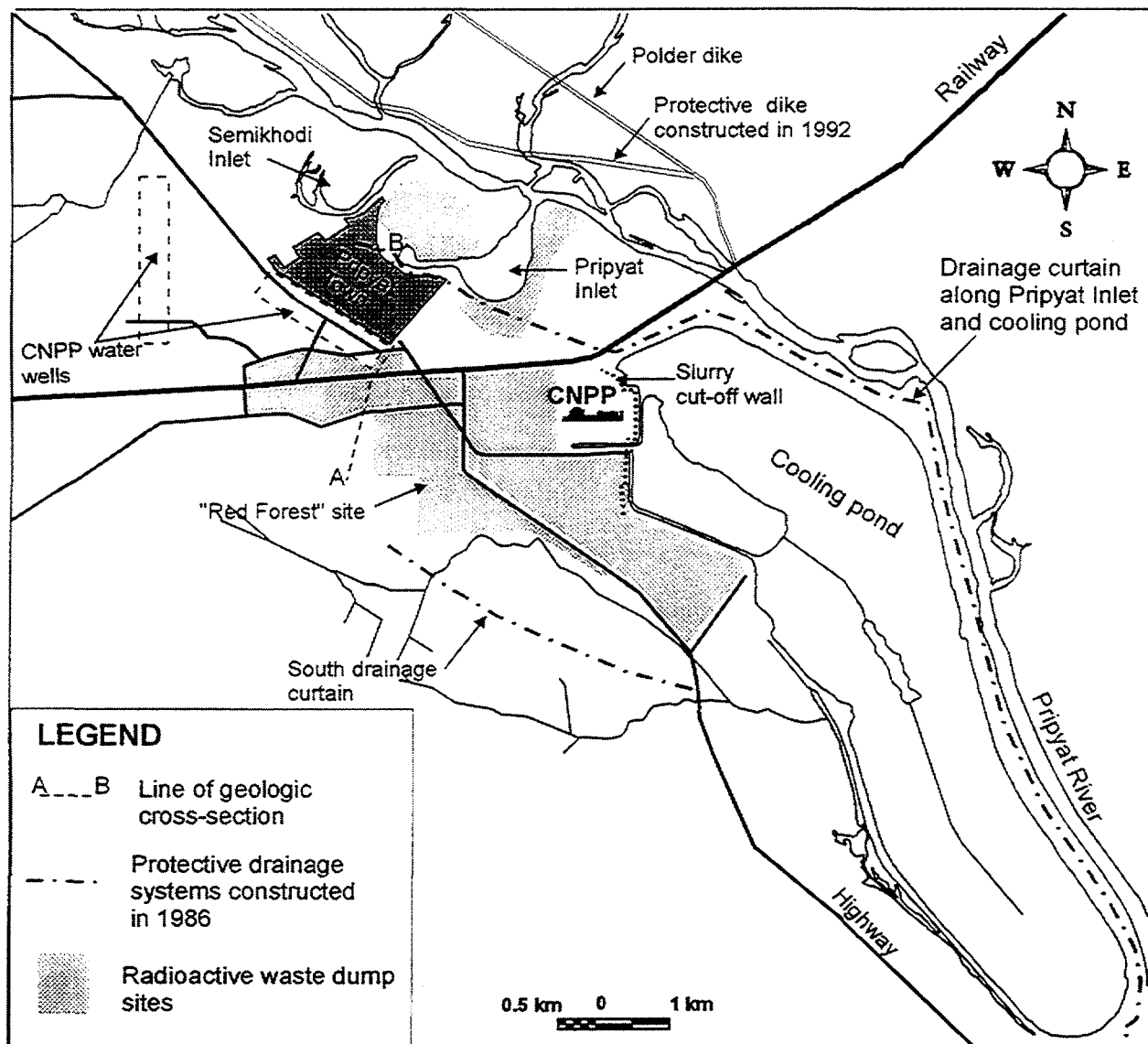


FIG. IV-2. Map of the Chernobyl nuclear power plant site.

aquitard layer composed of clay marls of the Eocene separates the upper unconfined aquifer and the confined aquifer in the sandy deposits of the Eocene. The hydraulic conductivity of aquitard layer is estimated at  $10^{-3}$ – $10^{-2}$  m/d.

The Eocene aquifer is exploited by the Pripjat Town water well field, the water being used for the supply of the CNPP. The study area is covered by a dense network of rivers, streams and agricultural drainage channels (with an average surface water channel density of 0.3 km/km<sup>2</sup>) which form tributaries to the Pripjat River. The runoff coefficient (i.e. percentage of annual river basin precipitation to runoff) is 10–30%. Groundwater discharge to rivers accounts for about 30% of the river basin runoff. In general, the environmental conditions of the study area (i.e. humid climate, acidic soils, high permeability and low sorption capacity of sediments, developed drainage network) favour migration of radioactive fallout contaminants to the groundwater system.

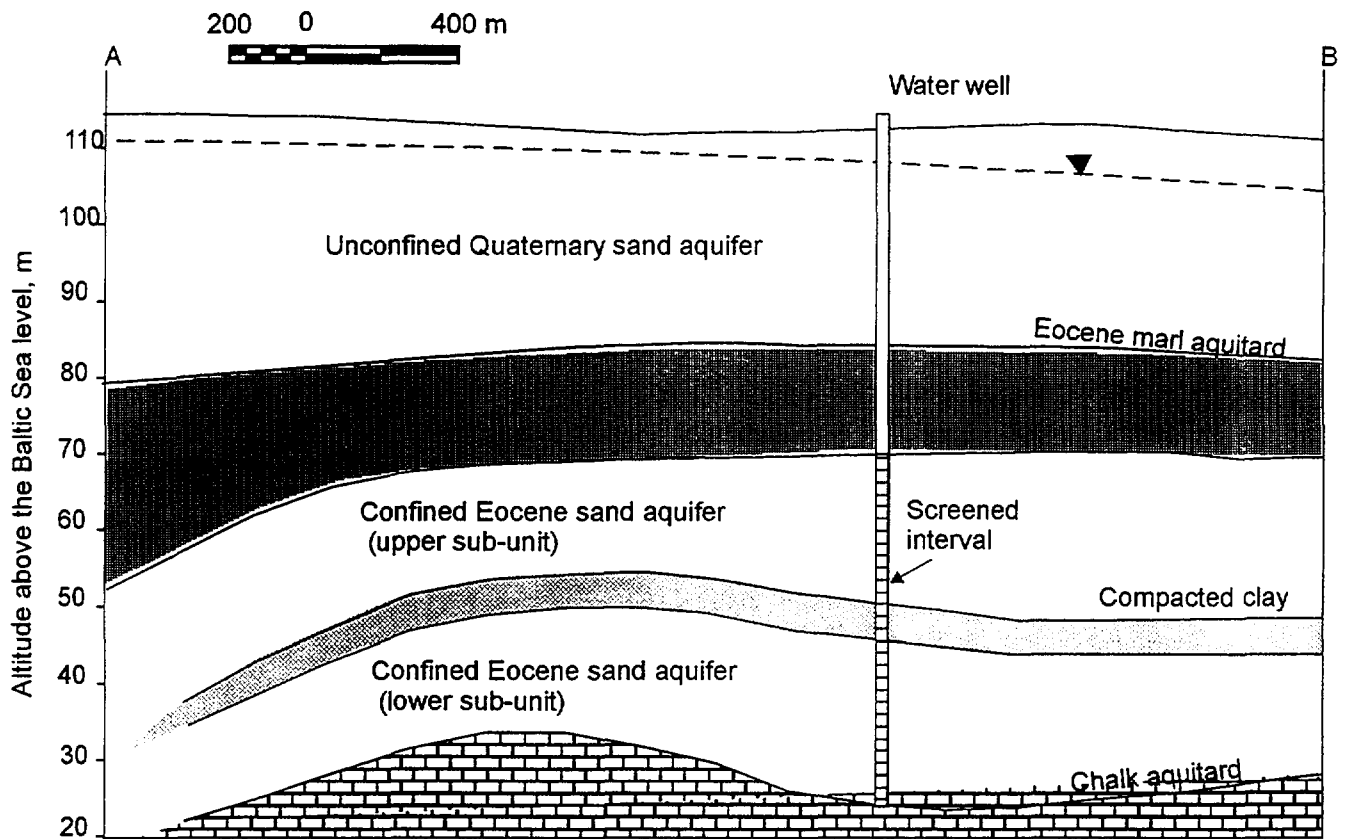


FIG. IV-3. Geologic cross section of the study site.

#### IV-3. SOURCES OF RADIONUCLIDE MIGRATION TO THE GROUNDWATER SYSTEM

Most radioactive fallout released during the Chernobyl accident was deposited in the upper portion of the Dnieper River basin. The majority of relatively "coarse" (up to 100  $\mu\text{m}$ ) reactor fuel "hot particles" from the destroyed Unit 4 of the CNPP containing relatively refractory  $^{90}\text{Sr}$  were deposited in the 30-km radius "exclusion" zone surrounding the CNPP. Surface contamination densities by  $^{90}\text{Sr}$  and  $^{137}\text{Cs}$  reach in the most contaminated locations of the 30-km zone thousands of curies per square kilometre. Over  $3.7 \times 10^{14}$  Bq of  $^{90}\text{Sr}$  is deposited in the Pripyat River flood plains, which are liable to be inundated during the spring flood, and in the polder areas (wetlands), which are liable to be flooded by surface- and groundwater during snow melts and in rainy seasons.

A number of surface water reservoirs adjacent to the CNPP were severely contaminated by radioactivity. Of particular concern was contamination of the CNPP cooling pond, situated on the flood plain of the Pripyat River near the destroyed Unit 4. Radioactive fallout and releases of liquid radioactive wastes from cleanup of Unit 4 resulted in accumulation of  $>10^{13}$  Bq of long-lived radionuclides in the cooling pond system [IV-8].

A major redistribution of radioactive contaminants in the environment surrounding the CNPP occurred due to cleanup activities carried out at the CNPP site in the first two years following the accident. Clean up wastes (radioactive debris, upper soil layers, and vegetation) with a total volume of about  $10^6$  m<sup>3</sup> were bulldozed directly into the local sandy soil in 2–3 m

deep unlined trenches. These radioactive waste dumps eventually became sources of serious groundwater radiological contamination.

Thus, radioactive substances in the CNPP 30-km zone are present in a variety of locations. Some of these sources of radioactivity pose a significant hazard to groundwater resources. In particular, the quantity of radioactivity concentrated in unorganized radioactive waste dumps is comparable to the radioactive contamination dispersed over the whole 30-km exclusion zone.

Radionuclides in the fuel fallout were initially contained in an almost insoluble matrix, which is the peculiarity of Chernobyl contaminants compared to global nuclear tests fallout. In the years that followed the Chernobyl accident, dissolution and release of radionuclides from the fuel matrix occurred. The release kinetics has been rather slow. The typical first order radionuclide release rate constants are estimated at about  $10^{-8}$ – $10^{-9}$  s<sup>-1</sup> [IV-11]. The radionuclides <sup>137</sup>Cs and <sup>90</sup>Sr show different geochemical behaviour after being leached from fallout. The <sup>137</sup>Cs is rapidly fixed by clay minerals and organic matter present in soil and, as a result, only about 1–10% remains in a mobile (i.e. water soluble and ion exchangeable) form. In contrast, up to 60–80% of the <sup>90</sup>Sr in soils of CNPP zone remained in highly mobile chemical forms in 1995. Little was known prior to the accident about the environmental fate of this specific type of radioactive contamination. The lack of understanding of geochemical behaviour of Chernobyl contaminants has lead to miscalculations and a number of incorrect remedial decisions in the early period of mitigation of the environmental consequences of Chernobyl accident.

#### IV-4 EMERGENCY PHASE GROUNDWATER PROTECTION

Because of the geographical location of the CNPP site in the upper drainage area of the Pripyat–Dnieper River system, the potential threat from the hydrological river transport of fallout radionuclides to the Ukrainian capital Kiev and other downstream populations was one of the major concerns from the first days after the accident. The population of the Dnieper basin region is approximately 32.5 million. About 9 million people are using Dnieper River as their drinking water source, while other exposure pathways include fishing and consumption of irrigated agricultural products. This issue is still of concern to the population and controversy over the water protection measures in the Chernobyl zone is continuing.

Facing the potential radiological hazard, soon after the accident the government of the USSR enacted a large scale programme to protect the Pripyat–Dnieper system from the secondary radionuclide contamination. As early as 30 May 1986 (i.e. one month after the accident) a special governmental decree was adopted entitled “Decision on systems for protection of groundwater from radioactive contaminants at the Chernobyl NPP site”. The groundwater protection measures were developed by the “Gidroproekt” institute (Moscow). These measures were intended to exercise control on the 120 km<sup>2</sup> area adjoining the CNPP, and pursued the following remediation objectives [IV-12]:

- prevent radionuclide transport by groundwater via the shallow unconfined Quaternary aquifer to the Pripyat River;
- prevent radioactive seepage from the CNPP cooling pond into the Pripyat River;
- protect the confined aquifer in the Eocene deposits, which is used as a drinking water source for CNPP from radioactive contamination.

To achieve these objectives the following protective systems were conceived and installed (some of them partially) at the CNPP site during summer of 1986 (Figure IV-2) :

- South “drainage curtain” about 5 km long representing a row of 54 vertical drainage wells (each 30 m deep).
- Drainage curtain along the bank of the Pripyat Inlet (5 km long; composed of 96 wells).
- About 30 metre deep slurry cut-off wall barrier around CNPP (to Eocene marl aquitard). Drainage wells were planned inside the cut-off wall to lower groundwater levels below the damaged Unit 4.
- Drainage curtain between the Pripyat River and cooling pond (13.5 km long, composed of 177 wells).

The aim of some of the drainage systems was to increase the thickness of the unsaturated zone at the site, and to decrease vertical hydraulic head gradients and seepage fluxes between the Quaternary unconfined aquifer and confined Eocene aquifer. In addition, some of the drainage systems were intended to intercept seepage of contaminated groundwater from the Quaternary aquifer to the Pripyat River.

The drainage wells were interconnected by a header pipe more than 25 km long. Treatment of contaminated drainage water represented a serious problem, because no technology existed at that time in the USSR to treat such large volumes of contaminated water. The research programme to develop water treatment technology was initiated in parallel with construction of protective systems. As an interim solution, it was decided to release the drainage water to the CNPP cooling pond.

The groundwater remediation measures discussed above were combined with a number of other surface water protective measures, including construction of river sediment traps, installation of zeolite containing dykes on streams and small rivers, etc. [IV-13].

In the first month after the accident, information on radioactive contamination of the area adjacent to the CNPP was incomplete and controversial. Accordingly, groundwater remedial measures were based on the “worst case” analysis. For example, early Chernobyl environmental analysis assumed that chemical radionuclide species in the fallout were in a highly water soluble form [IV-12]. In general, most counter measures in that period were implemented without detailed scientific assessment of their radiological benefit and cost efficiency, due to the lack of time for research and the lack of necessary experience.

By mid summer 1986, the level of the radioactive contamination of the territory immediately adjacent to the CNPP was found to be 10 to 100 times lower than the worst estimates of May 1986. In addition, the initial assumption about high solubility of the Chernobyl fallout was not confirmed. Laboratory studies on radionuclide leaching of the fuel particles performed at that time indicated very low mobility of Chernobyl contaminants. Field observations during the first 6 months after the accident showed that no significant breakthrough of radionuclides had occurred to the groundwater system. Based on these data, and on updated radionuclide transport assessments by the end of 1986, the groundwater remediation projects were stopped. Only a 2.2 km segment of the planned 8.5 km slurry wall around the CNPP was

completed. The protective drainage systems were put on reserve and some pumps and drainage collectors being later dismantled and removed.

The costs of incorrect remedial assessments were high and practically useless. For example, the costs of installing the drainage curtain along the cooling pond alone were 22 million roubles<sup>2</sup>. Another negative consequence was the unnecessary exposure to mitigation workers. The typical exposure rates at the CNPP site in the summer period of 1986 were about 0.1–1 R/h.

The mistakes of the emergency phase groundwater remediation measures were made because of the lack of adequate radiological information, as well as the absence of reliable forecasts of radionuclide transport in the hydrogeological environment. The decision makers at Chernobyl did not have at hand nor developed emergency response action plans, nor site specific hydrogeological databases and radiological assessment models. As a result, protective features of the local hydrogeological environment have been significantly underestimated.

In addition, during the first several years following the event, the Chernobyl accident was surrounded by an atmosphere of secrecy, which was inherent to the Soviet political and administrative system of that time. The normal peer review of remediation projects by scientists outside the nuclear sector institutes was not carried out. Previous nuclear accidents (e.g. Kyshtim accident in Southern Urals in 1957) were covered up by authorities, and scientists and society were not able to study and learn from these events [IV-8].

#### IV-5. PROBLEM OF GROUNDWATER CONTAMINATION AT THE RED FOREST RADIOACTIVE WASTE DUMP SITE

An early mitigative measure implemented in 1987 was the *in situ* burial of contaminated soil, vegetation, and other radioactively contaminated debris, including the "Red Forest" pine trees (killed by high levels of radiation), within about an 8 km<sup>2</sup> area adjacent to CNPP Unit 4. A volume of about 1 million m<sup>3</sup> of waste was bulldozed by engineering army troops in shallow 2–3 m deep unlined trenches and soil mounds. The goals of this measure were to reduce the external exposure to CNPP workers, and to mitigate the potential risk of atmospheric transport of radioactivity associated with a forest fire of the dead pine "Red Forest" (Figure IV-2). The decision to dispose waste in sandy shallow trenches near the groundwater table was supported by geochemical data from laboratory leach testing which indicated very low solubility of CNPP near field fallout fuel particles.

The optimistic predictions of 1987 about high stability of the fuel "hot" particles, which were based on short term laboratory studies, were not confirmed in the subsequent years by field data. Beginning in 1988, increases in the mobility of radioactive contaminants in the hydrological and hydrogeological environment at the Chernobyl site have been observed due to weathering and dissolution of "hot" particles [IV-11]. From 1987 to 1989, the radiological situation at the "Red Forest" site has worsened because of a 1 to 2 m rise of the groundwater table, which inundated some burials. The rise in the groundwater table was probably provoked by increased infiltration recharge because of the changed evapotranspiration conditions caused by demolition of vegetation and removal of the top soil layer. Inspection of several trenches at the "Red Forest"

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<sup>2</sup> Unless otherwise noted, cost estimates are given in USSR 1984 roubles; 1 USSR rouble (1984) = US\$1.6.

site in the autumn of 1989 has revealed groundwater contamination by  $^{90}\text{Sr}$  at concentrations two orders of magnitude above drinking water standards<sup>3</sup>. Groundwater contamination at the “Red Forest” site and other adjacent radioactive waste dump sites are viewed as a serious environmental problem [IV-9].

Data from the “Red Forest” site characterization studies conducted in 1990–91 indicated that intense radionuclide migration to groundwater occurred from waste burials.

The critical radionuclide was  $^{90}\text{Sr}$ . It was estimated that  $^{90}\text{Sr}$  migration from waste burials can produce a radionuclide plume in the groundwater with a concentration exceeding the drinking water standard at a distance up to several hundred metres downstream from the trenches containing wastes. However, there was no risk of discharge of highly contaminated groundwater to Pripyat River, which is located at several kilometre distance from the “Red Forest” site. At the same time, some conservative analyses indicated the possibility of contamination of the Eocene aquifer. Also, there was a possibility that some unknown burials with higher activity waste may exist in the immediate vicinity of Pripyat River [IV-14].

Several groundwater remedial alternatives were worked out and evaluated for the area occupied by radioactive waste dumps by the Institute of Industrial Technologies (NIPIPT, Moscow) [IV-14, IV-15]. Excavation and disposal of waste was deemed a non-viable option because of the large volume of waste, high exposures to remediation personnel, and high costs of constructing a repository for final disposal of long-lived transuranic waste. Remediation cost estimates were rather high even for some less labourious remedial alternatives.

Several remedial proposals were based on the application of innovative technologies and required conducting additional extensive research and development works and field testing prior to a large scale application [IV-14] and are discussed as follows:

- isolation of trenches by narrow (3–8 cm thick) vertical or slanted slurry screens, combined with in situ stabilization of soil in the trench by silicatisation (i.e. injection of liquid glass), and capping by impermeable material from the top;
- creation of a geochemical barrier to radionuclide migration via well injection of reactants into the aquifer, which would cause co-precipitation of dissolved radionuclides;
- creation of permeable sorption barriers to emitting radionuclides using an adsorbent-filled trench excavated in an aquifer across the groundwater flow and intensification of radionuclide migration using an induced electric field.

In 1991, the Soviet Union collapsed. During the subsequent years most remedial research activities at the CNPP site by Russian institutes, including NIPIPT, have been curtailed due to the crisis and economic difficulties in the Ukraine. It became evident that there is no potential for implementation of costly groundwater remediation proposals given the changed political and economic situation.

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<sup>3</sup>The USSR drinking water standard for  $^{90}\text{Sr}$  constitutes 14.8 Bq/L (400 pCi/L).

The cooling pond has a surface area of 22.9 km<sup>2</sup> and a volume of 150 × 10<sup>6</sup> m<sup>3</sup> (Figure IV-2). The water level in the pond is 7 to 8 m above the low level of the Pripyat River. The sides of the pond for most of the perimeter are formed by a levee, built from highly permeable (hydraulic conductivity 5–25 m/d) local alluvial sands. This design results in extensive seepage through the levee. Seepage and evaporative losses are compensated for by pumping water from the river. Drainage ditches were constructed at the base of the levee to intercept the seeping water and prevent the formation of wetlands. The drainage water from ditches discharges to the Pripyat River. Seepage loss from the cooling pond are estimated at 70–100 × 10<sup>6</sup> m<sup>3</sup>/a. Seepage loss is composed of the discharge of the levee drainage ditches (55–80 × 10<sup>6</sup> m<sup>3</sup>/a, groundwater travel time to the Pripyat River 1–2 months) and subsurface seepage discharge through the bottom of the cooling pond that is not intercepted by the drainage ditches (15–20 × 10<sup>6</sup> m<sup>3</sup>/a, groundwater travel time 1–2 years) (see Figure IV-4).



FIG. IV-4. Alternative designs of protective drainage systems aimed to capture radioactive seepage discharges from the cooling pond.

As a part of the emergency response groundwater remediation programme, a 13.5 km long drainage curtain consisting of 177 drainage wells (each 240 mm diameter and 20 m deep) were constructed to protect the Pripyat River from radioactive seepage discharge from the cooling pond (Figures IV-2 and IV-4). The project debit rate of the drainage system was  $103 \times 10^6 \text{ m}^3/\text{a}$ . The drainage curtain was intended to capture the contaminated groundwater seeping from the pond and treat it using decontamination facilities. However, because water treatment technology has not been yet developed, as an interim solution was planned to return drainage water back to the pond. Because monitoring data did not reveal progressive groundwater contamination by the end of 1986, the drainage curtain was put on reserve after completion.

Another remedial alternative, evaluated in May 1986, was the construction of the cut-off wall through the levee to the marl aquitard. This alternative was rejected because of its estimated high cost of 12.5 million roubles for the 14 km long, 20 m deep, 0.15 m thick slurry wall. By comparison, the drainage curtain project cost was 3.5 million roubles; actual construction cost was 22 million roubles [IV-16].

By the end of 1988, an increase in  $^{90}\text{Sr}$  concentrations had been observed in the water of the cooling pond, in the drainage discharges, and in some groundwater monitoring wells between the cooling pond and the Pripyat River (Figure IV-5). An increase in  $^{90}\text{Sr}$  concentration in the pond water column from 1987 to 1989 was probably due to radionuclide leaching from the fuel particles contained in the bottom sediments of the pond, while the subsequent decreases were due mainly to dilution.

Several research programmes were initiated in 1989–1990 to address the problem of increased  $^{90}\text{Sr}$  mobility in the cooling pond system and to evaluate remedial alternatives. The new careful assessments revealed conceptual shortcomings of the drainage curtain design. Groundwater modelling has shown that about 30% of the drainage curtain discharge would be the water coming from the Pripyat River (Figure IV-6). The additional volume of seepage river water pumped by the drainage curtain and returned back to the pond may exceed evaporation losses from the pond [IV-17]. Other concerns were that returning all seepage water back to the pond may cause (over the long term) an increase of the TDS levels in the water above the 1 g/L operating level of the CNPP [IV-17] and that operating the drainage curtain would increase migration of radionuclides to groundwater from contaminated sediments [IV-16]. Therefore, the drainage curtain has not been brought into operation.

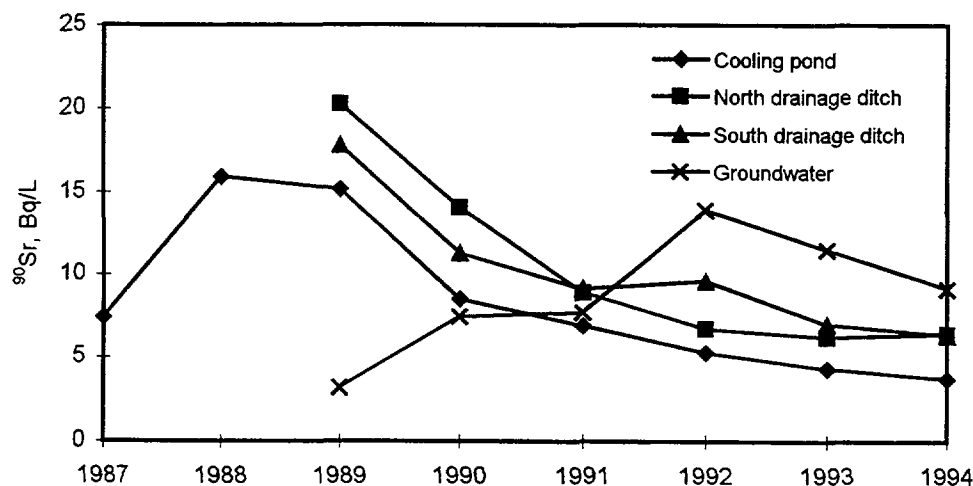


FIG. IV-5. Radioactivity of the surface and groundwater at the cooling pond of CNPP.

As an alternative remedial solution, construction of an open drainage system between the cooling pond and Pripjat River was started in 1991. The new remedial system was intended to collect the drainage water discharging from the ditch running along the North East flank of the pond levee, as well as to intercept subsurface seepage to the Pripjat River (Figure IV-4, b). The intercepted seepage water had to be returned to the pond using 6 pump stations, installed along the drainage perimeter. In view of the difficult economic situation, a relatively cheap drainage system design has been adopted, with no measures to consolidate the sandy bottom and the sides of the constructed drainage channel. Excavation of the open drainage ditch was completed in 1994. Since that time, several spring floods of the Pripjat River have caused erosion of the constructed channel, and the system is in non-working condition [IV-16].

Costs necessary to repair and consolidate the open drainage channel with gravel are estimated at 1 million roubles. The operational costs are also rather high (e.g. the pump energy consumption alone is estimated at 1.5 million kW/a). In addition, an estimated 4.5 million roubles are required to construct a separate system which would collect and pump back to the pond the contaminated discharges from the second drainage ditch running along the South West flank of the levee of the cooling pond [IV-16].

As discussed in recent retrospective cooling pond countermeasure analyses [IV-8], several strategic mistakes have been performed in the course of the cooling pond remedial efforts.

No apparent technical reason existed to initiate the cooling pond remedial construction works in conditions of high radiation exposures just one month after the accident. Even for the most mobile constituent, the solute travel time from the cooling pond to the Pripjat River through the subsurface pathway is at least one year. In reality, it took up to six years before significant subsurface breakthrough of  $^{90}\text{Sr}$  to the Pripjat River occurred.

Both the drainage curtain built 1986 and the open drainage system completed in 1994 were focused on protecting the Pripjat River from radionuclide migration from the cooling pond through the subsurface pathway (Figure IV-6). However, based on recent analyses [IV-18], the most important  $^{90}\text{Sr}$  migration pathway was through the cooling pond levee to drainage ditches; the subsurface pathway to Pripjat River was of only minor importance (Figure IV-6). Unfortunately, the pathway through the levee did not receive appropriate attention in the remedial

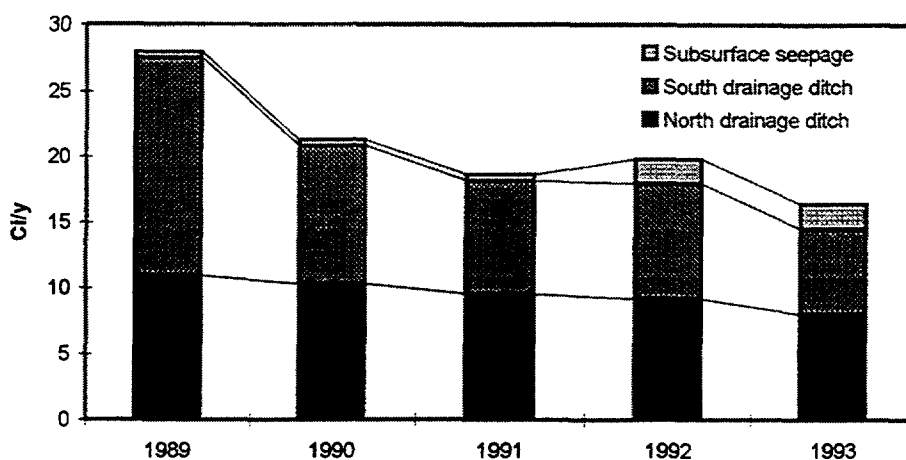


FIG. IV-6. Releases of  $^{90}\text{Sr}$  from the cooling pond to the Pripjat River via various pathways.

designs. On the other hand, the early assessments drastically overestimated the radionuclide transport via the subsurface migration pathway, which was primarily the result of a poor monitoring system and a lack of understanding of radionuclide transport and fate in the hydrogeologic system of the cooling pond [IV-8].

#### IV-7. CURRENT STRATEGY FOR MANAGEMENT OF GROUNDWATER CONTAMINATION PROBLEMS

Very low effectiveness of ground and surface water remedial measures conducted in the early aftermath of the Chernobyl accident demonstrated that future remedial efforts require careful scientific grounding and optimization. A purposeful research programme "Radioecology of the water systems in the zone of the influence of Chernobyl accident" was conducted in 1992–95 by the Ministry on Chernobyl Affairs of Ukraine. As a result of this programme and other studies, the current concept for water protection and remediation for the areas contaminated by the Chernobyl accident was developed.

#### IV-8. HEALTH RISK BASED REMEDIATION CRITERIA

It is now widely recognized that remediation criteria and standards should be based on health risks. The early post accident groundwater remedial analysis at Chernobyl did not have clearly defined health risk based criteria. Instead, the radionuclide drinking water standards were often used as relevant decision criteria. The use of drinking water standards as criteria for groundwater remediation is questionable, because there is no residential population in the Chernobyl 30-km zone, which serves an institutional control to prevent public access to highly contaminated land [IV-8].

As discussed earlier in this paper, the major health concern was the off-site radionuclide transport from the Chernobyl zone to the Pripyat–Dnieper river system. Results from the dose assessments for the Dnieper River exposure pathway allowed consideration of the hydrological remediation measures at Chernobyl from the health risk perspective. Based on the assessments, a value of 2 Bq/L has been specified for  $^{90}\text{Sr}$  concentration in the lower reaches of the Pripyat River on the border of the Chernobyl 30-km zone as a main criterion for decision making regarding ground- and surface-water remedial measures. Keeping with this criterion would permit water to be used safely at any point in the downstream Dnieper River system. (The above concentration value corresponds to a dose limit value of 1 mSv/a, established by Ukrainian law on protection of the population from the consequences of Chernobyl accident.)

Projected average lifetime fatal cancer risks from Dnieper exposure pathway for the population of Dnieper regions were estimated to be on the order of  $10^{-5}$ . For some critical subgroups (e.g. professional fishermen), evaluated risks were 4–5 and even more times higher. It was also established that compared with other sources of radiation exposure (e.g. external exposure from surface contamination, internal exposure from ingestion of contaminated agricultural products), the Dnieper pathway contributes a rather small portion of radiation dose (e.g. 6% of total dose in 1993 for the person in Kiev Region). For people living in the Southern Ukraine this value is 20–30% and more. Because the Pripyat–Dnieper pathway poses rather low radiation risk, only relatively inexpensive remedial measures in accordance with the ALARA principle may be justified in the Chernobyl zone.

On the other hand, the sociological studies have shown that in the public opinion, the potential danger to health from water consumption was greater than from other sources. Therefore, water remediation measures are important to minimize the psychological stress from radiation hazards for the population consuming radioactively contaminated water.

#### IV-9. CURRENT AND PROJECTED ESTIMATES OF RADIONUCLIDE RELEASES VIA THE GROUNDWATER PATHWAY TO THE RIVER NETWORK

Radiological monitoring data were used to identify the sources of radioactive contamination of the Pripyat River. It was established that direct surface water interactions with contaminated river flood plain soils during flooding events is the major  $^{90}\text{Sr}$  mobilization mechanism [IV-19].

The contributions of groundwater sources to contamination of the river network in the Chernobyl zone currently are relatively small. Based on monitoring data,  $^{90}\text{Sr}$  transport by groundwater from non-point sources in the Chernobyl zone to the Pripyat River was estimated to be less than  $5.9 \times 10^{10}$  Bq/a in 1992, which is 1.5% of the total  $^{90}\text{Sr}$  transport by Pripyat River. This is not surprising because in the contaminated catchment areas, usually 95 to 98% of fallout activity was contained in 1992 in the upper 5 to 10 cm of soil. The process of vertical radioactive contaminant migration through the vadose zone to groundwater has been delayed due to the slow release of radionuclides from the matrix of fuel particles, and due to retardation in the unsaturated zone caused by sorption on soil. The typical  $^{90}\text{Sr}$  vertical migration rates in the soil profile are estimated to be from 0.1 to 1.5 cm/a, while maximum rates do not exceed 2–6 cm/a [IV-20].

According to modelling studies, groundwater migration is not expected to cause significant increase of radionuclide transport to rivers in the future. The maximum annual projected  $^{90}\text{Sr}$  transport by groundwater to the Pripyat River from the non-point sources in Chernobyl 30-km zone was conservatively estimated at  $7.4 \times 10^9$  Bq/a about 50 years from now. The maximum  $^{90}\text{Sr}$  transport with groundwater from point sources of contamination at the CNPP site (i.e. radioactive waste dumps, radioactive “hot spots” on the surface) was conservatively estimated at  $1.3 \times 10^{11}$  Bq/a in about 100 years [IV-18]. The estimated maximum release rates equal approximately 0.02% per year from initial radionuclide inventory in the catchment area. Comparison of the above estimates of the long term  $^{90}\text{Sr}$  groundwater transport to the Pripyat River with current hydrologic contaminant releases due to the surface water wash out process (e.g.  $3.7 \times 10^{12}$ – $1.48 \times 10^{13}$  Bq/a in 1989–1995) shows that the groundwater pathway has a potential for contributing only marginally to the overall  $^{90}\text{Sr}$  transport to the Dnieper system.

#### IV-10. ON SITE RISKS FROM THE GROUNDWATER EXPOSURE PATHWAY

Bugai et al. compared the health risks from different radiation sources in the Chernobyl 30-km zone: 1) from contaminated soil (due to external irradiation and ingestion of contaminated agricultural products; the major dose forming radionuclide is  $^{137}\text{Cs}$ ), and, 2) from using  $^{90}\text{Sr}$  contaminated groundwater from the unconfined aquifer. The risk from surface contamination was estimated to be about one order of magnitude higher compared to the groundwater risk (Figure IV- 7). Thus, in the case of diffused surface contamination by radioactivity, as in the Chernobyl zone, the groundwater contamination is not a priority health concern.

The risk cost benefit analysis of contamination of the CNPP water wells exploiting the Eocene aquifer was conducted as shown in the reference [IV-21]. The basic premise of the utilized risk cost benefit assessment framework is that the value of avoiding a groundwater contamination incident (i.e. cost of groundwater remediation measures) is not higher than expected (probabilistic) costs that would be incurred if the incident were allowed to occur [IV-22]. The risk was defined as the expected costs of well field failure due to exceedence of the  $^{90}\text{Sr}$  regulatory standard for drinking water in extracted groundwater. The probability of well field failure was estimated using the Monte Carlo technique. Based on estimated low economical risk of water well failure and was concluded that "no remedial action alternative" is the preferred management strategy for the CNPP well field.

#### IV-11. IMPLICATIONS TO SURFACE AND GROUNDWATER REMEDIATION STRATEGY

Identification of sources of Pripyat River contamination combined with risk assessment studies was used to optimize water protection strategy within the 30-km Chernobyl zone. Various hydrological off-site radionuclide transport scenarios have been simulated using computer models of the Dnieper Reservoir system. Construction of dykes around the contaminated flood plain areas on the left bank of the Pripyat River was chosen as the most effective remedial option at a reasonable cost. The protective dyke was completed by the end of 1992. Subsequent monitoring data confirmed modelling predictions of the effectiveness of this measure. More than  $3.7 \times 10^{12}$  Bq of  $^{90}\text{Sr}$  were prevented from being washed out from the flood plain of the Pripyat during the flooding of summer 1994. A similar dyke is being constructed currently to isolate the contaminated area at the right bank of the Pripyat River which is adjacent to the CNPP.

The groundwater migration sources, except for the cooling pond, provide a relatively insignificant contribution to radioactive contamination of the river network in the Chernobyl zone. Therefore, remediation of these sources (including the "Red Forest" site) cannot substantially influence off-site risk caused by hydrologic transport. From the perspective of on-site radiological risks, there is no reason to remediate groundwater alone without comprehensive remediation of other environmental media. Based on the above considerations, the "passive" management strategy is currently adopted for groundwater contamination problems, consisting of developing a reliable monitoring system for the contaminated groundwater sites. (Implicit in this strategy is the assumption that the exclusion zone will remain a long term institutional control over the entire contaminated area.) The monitoring issue is of particular importance because the existing groundwater monitoring system in the Chernobyl zone suffers from a number of shortcomings, including an insufficient number of wells (which are unevenly distributed and inappropriately sited) and the well designs are often not suited to monitoring purposes [IV-23].

Monitoring alone entails a potential risk for a much greater amount of remediation in the future because contamination spreads and point sources of radioactivity transform to diffused sources. Therefore, the attitude towards groundwater remediation may change in future, if the general management strategy of the authorities towards the Chernobyl zone evolves from institutional control (current practice) to comprehensive remediation and returning the contaminated site to productive use.

The cooling pond represents a special case, because it is an important source of  $^{90}\text{Sr}$  migration to Pripyat River, especially during low water seasons. Because remediation of subsurface radioactive releases from the pond is a complex and expensive task, and because the CNPP is expected to be closed in the year 2000, a "no action" strategy is currently considered as

the most reasonable approach to the immediate future management of the pond. The proposed long term solution after the reactors are stopped is that the cooling pond would be partially drained to reduce seepage discharges and radionuclide migration to Pripjat River to an insignificant level. One consequence of this remediation alternative would be that large areas of the contaminated bottom sediments of the cooling pond would be exposed to the atmosphere. To prevent atmospheric transport of radioactivity, contaminated sediments may be covered by a soil screen or retrieved and disposed in the special repository [IV-8].

#### IV-12. LESSONS FROM CHERNOBYL GROUNDWATER REMEDIATION EXPERIENCE

Several common lessons emerged from the review of groundwater remedial actions carried out to mitigate the environmental consequences of the Chernobyl accident, which may be useful for decision makers working with radioactively contaminated sites in other countries.

##### **Planning of remedial measures**

Remediation criteria and objectives must be clearly defined and spelled out early in the assessment phase of groundwater remedial activities. In contrast, the common practice at Chernobyl in the early post accident period referenced drinking water standards instead of establishing risk based criteria. For example, in the case of the “Red Forest” radioactive waste dump site, considerable resources were spent in 1991–92 on analyses of remedial alternatives and development of remediation technologies. Later, in view of the fact that groundwater did not pose significant off-site health risk, remediation projects were abandoned.

Another lesson is that, groundwater contamination problems should be considered within the “big picture” of various radioactive contaminant exposure pathways/risks. As the Chernobyl case study shows, for regional pollution by radioactive fallout (even in generally unfavourable hydrogeological conditions), groundwater is a less important agent of radioactive contaminant mobilization to the river network than surface water interactions. Therefore, remedial efforts were re-directed to isolation of contaminated flood plain areas from inundation by surface water.

A peer review of proposed actions is an important component of effective management of remediation projects. The economical losses and the negative impacts to remedial workers’ health from the unnecessary groundwater remediation projects at Chernobyl in 1986 might have been avoided, if there had been peer review of the proposed countermeasures by independent scientists.

##### **Social and economic aspects**

The Chernobyl experience demonstrates that decision makers and the ordinary public often suppose that groundwater contamination entails much higher risk than it really does. This reality needs to be accounted for in management of groundwater contamination problems.

Groundwater remediation often requires big financial investments, while potential benefits are often limited and/or highly uncertain. The promising formalized approach to decision making in groundwater remediation analyses accounting for uncertainties is economic risk cost benefit analysis [IV-22], which has been applied recently to a number of real problems of groundwater contamination by radioactivity, including those related to the Chernobyl accident [IV-21, IV-24].

## Remedial technologies

In many cases, engineers at Chernobyl did not have at hand needed remedial technologies. In particular, subsurface permeable geochemical/sorption barriers have been often identified as a conceptually preferable remedial solution (e.g. for the cooling pond problem and the “Red Forest” problem [IV-14, IV-17]). Another unsolved technological problem was treatment of large volumes of water contaminated with  $^{90}\text{Sr}$ . This highlights the need for research and development activities on these and other innovative groundwater remedial technologies.

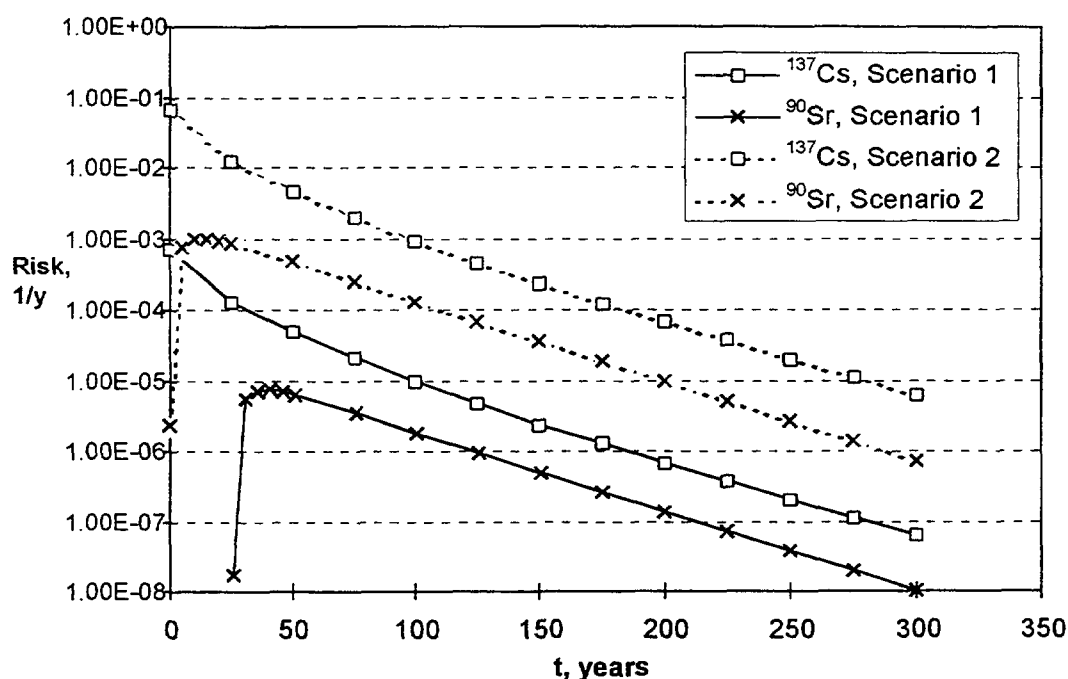


FIG. IV-7. Health risk to hypothetical residents of the 30 km zone from  $^{90}\text{Sr}$  and  $^{137}\text{Cs}$ .  
Scenario 1 — average conditions, Scenario 2 — Red Forest site.

## Environmental transport and fate of radioactive contaminants

Many incorrect remedial actions at Chernobyl were caused by the lack of scientific knowledge on the environmental transport and fate of radioactive contaminants (e.g. environmental behaviour of “hot” particles). Therefore, continued research in the field of contaminant hydrogeology and radioecology is warranted. Unique monitoring data from the Chernobyl case study deserve to be summarized and made available to the international scientific community<sup>4</sup>.

Decisions at Chernobyl were often based on extremely conservative contaminant transport assessments. While conservative models that provide a margin of safety may be appropriate to use in assessment of planned facilities (e.g. radioactive waste repositories), assessment for remedial activities must use realistic analyses and incorporate best science [IV-8]. This would help prevent costly, unnecessary remediation.

<sup>4</sup>Up to now there have been very little international efforts to study and summarize the hydro geological consequences of the Chernobyl accident.

Remedial assessments must carefully account for possible adverse secondary effects from remedial measures (e.g. the situation with the “Red Forest” site, where removal of the soil layer and vegetation during cleanup activities resulted in a changed evapotranspiration regime and subsequent rise of groundwater table, which inundated some trenches with cleanup wastes). Analysis of such long term adverse environmental effects may be a challenging research task for scientists and engineers involved in remediation.

#### IV-13. CONCLUSIONS

The Chernobyl experience shows that groundwater remedial actions have to be planned and implemented with an integral scientific and engineering approach considering the following:

- risks to human health, estimated using state of the art models and realistic radioactive contaminant transport and exposure scenarios, integrated into a “big picture” of various exposure pathways and risks;
- country specific social and economic factors;
- available remedial technologies;
- possible secondary adverse environmental effects of remedial measures.

It seems that in many cases, especially in countries with developing economies where remedial funds are limited, the passive approach, consisting of establishing institutional control on groundwater usage and monitoring further spreading of contamination, may represent an acceptable approach from health risk perspective and cost effective solution of groundwater contamination problems.

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## **PROGNOSIS OF GROUNDWATER CONTAMINATION CAUSED BY FLOODING OF URANIUM MINES IN EASTERN GERMANY**

### **V-1. BACKGROUND**

Large scale uranium mining operations were conducted in the States of Saxony and Thuringia from 1946 through the end of 1990. The German federal government is funding a very large environmental restoration project to rehabilitate the resultant waste piles, mines and tailing ponds in eastern Germany. This project, which is called WISMUT, includes the investigation, assessment, protection, and treatment of groundwater. The radioactive contamination of surface and groundwater is associated with sources such as underground mines, leaching sites, open pits, tailings ponds, and waste rock piles.

The WISMUT sites include the following:

- underground mining sites at Ronneburg and Schlema-Alberoda;
- the in situ leaching site at Koenigstein; and
- the tailings ponds at Seelingstädt (Culmitzsch, Truenzig) and Crossen (Helmsdorf).

New water treatment plants have been installed at Helmsdorf and Poehla. Another water treatment plant will be commissioned early 1998 at the Schlema mine. While at the Poehla and Schlema sites contaminated mine waters are treated to improve the groundwater quality, the plant at Helmsdorf is treating contaminated water of a tailings impoundment to allow subsequent consolidation and coverage of the tailings.

To properly plan the groundwater protection measures and water treatment activities, site specific investigations were carried out on the mining and milling conditions, hydrogeology, infiltration processes, water rock interactions, and the hydrodynamic behaviour expected in the future. In each case, it is important that the principles of justification and optimization are appropriately linked to the decision making process. In deciding the preferred approach to groundwater protection, whether by water treatment or in situ remedial actions, the following requirements apply:

- Prognosis of the environmental impacts of remediation option by modelling;
- Deterministic or probabilistic risk analysis;
- Use of multi criteria (including uncertainty considerations) decision making;
- Maintenance of close contacts and communication with the permitting authorities.

A preventative strategy of groundwater protection includes the controlled flooding of large underground mines Ronneburg, Koenigstein and Schlema-Alberoda to reduce the contaminant load of the discharges. A further action has included long term covering of tailings ponds and waste rock dumps with natural materials, thus reducing the seepage of contaminants to the groundwater.

If an active water treatment is deemed necessary, the following factors should be carefully considered:

- Prevention of immediate radiological risk to people;

- Prevention of long term contamination of aquatic systems;
- Prevention/reduction of risks to industrial water consumers;
- Maintenance of safe operations;
- Safe disposal of residues and wastes;
- Insurance of low capital and operating costs.

Currently, WISMUT employs chemicals and polymers for the precipitation and adsorption of contaminants of concern. One of the sites is also utilizing ion exchange. Depending on the ultimate remedial action cleanup goals, the implementation of these ex situ treatment methods has the potential of being technically feasible, but also (most likely) at a very high cost. Remedial action cleanup goals for the contaminated groundwater are still being developed in consultation with the regulatory agencies. Complicated issues, which are sometimes associated with the containment and extraction of contaminated groundwater, appear to be minimal at most WISMUT sites, however.

## V-2. INTRODUCTION

Uranium mining in Eastern Germany was terminated in 1990 after more than 40 years of large scale exploitation resulting in a cumulative production of about 200 000 t of uranium. These operations had significant impact also on the hydrosphere in the mining regions of Saxony and Thuringia.

In the Gera-Ronneburg region (Thuringia), the legacy left behind by uranium mining comprises a vast system of interconnected underground mines and a large open pit mine in Lichtenberg. Underground mining voids total  $28 \times 10^6 \text{ m}^3$ , the floodable pore volume of the partially backfilled open pit amounts to some  $15 \times 10^6 \text{ m}^3$ . With a mine and groundwater pumping rate of between 700 and 1000  $\text{m}^3/\text{h}$ , the depression cone from the mines currently extends over an area of more than 70  $\text{km}^2$  and affects a rock mass of some 15 billion  $\text{m}^3$ .

In the Aue mining district of Saxony, uranium production amounted to approximately 80000 tonnes of uranium from mines developed down to a depth of some 1800 m. Underground mining voids that were not backfilled total some  $40 \times 10^6 \text{ m}^3$ . These voids are hydraulically connected, both horizontally and vertically, by 62 sloping levels. Mine development and production drifts amounted to a total length of 4200 km, 80 shafts, and numerous mining chambers.

To date, about two thirds of the Schlema-Alberoda mine has been flooded. In the Ronneburg mining district, flooding was initiated in November 1997 following extensive preparations.

## V-3. OBJECTIVES OF AND CONDITIONS FOR MINE FLOODING IN THE RONNEBURG REGION

Remediation of the legacy left behind by uranium mining aims at reducing radiological, chemical and other exposure to the public and the environment to an acceptable level. Remedial action is carried out in full regulatory compliance. Flooding of the Ronneburg deposit meets long term environmental requirements, e.g. reduction of acid generation from pyrite and subsequent

mobilization and discharge of radionuclides, metals, and salts into ground and surface waters and the elimination of radioactive emissions.

Mining workings to be flooded comprise the workings of the former Reust, Schmirchau, Paitzdorf, Drosen, and Beerswalde mines. The vertical extension of the flooding zone is about 900 m. The volume to be flooded is in excess of  $50 \times 10^6 \text{ m}^3$ .

Hydrogeological conditions and substantial structural differences between individual mining fields give rise to the following requirements of a general nature that must be met in order to control pre- and post-flooding water qualities:

- avoidance of large scale circulation of water and active intervention with regard to water stratification;
- protection of receiving waters against unacceptable contaminant loads;
- restoration, to the extent feasible, of pre-mining conditions in catchment areas of receiving streams; and
- permanent suppression of oxidation processes in draw down areas and improvement of water quality.

The option preferred by WISMUT consists of continuous flooding up to an optimum elevated groundwater level in order to reduce natural leaching processes in the unsaturated zone in the long term.

Realistic forecasting of the flooding process and of the flows involved as well as of the anticipated final groundwater levels is subject to the availability and reliability of a multitude of parameters describing flooding zones and water ingress.

The floodable mine volume (Table V-1) is not limited to stops, shafts and development drifts but also includes open, collapsed, or partly backfilled voids from the exploration, development and mining activities. The later type of voids are estimated for the Ronneburg mine field at about  $28 \times 10^6 \text{ m}^3$ . They are of specific hydraulic relevance to the dynamics of flooding and subsequent flows.

Due to loosening and deformation of the rocks directly influenced by mining operations in particular in the roof, a floodable pore space of  $7 \times 10^6 \text{ m}^3$  is estimated and offers possibilities for direct infiltration of meteoric waters.

In addition to man-made mine cavities in the rock mass and to the enlarged pore space immediately adjacent to mine workings, geological voids in the rock mass not directly influenced by mine workings within the depression cone are anticipated in the order of  $4 \text{ to } 10 \times 10^6 \text{ m}^3$ . The wide range of this estimate caused by the fact that due to geological bedding conditions the rock mass within the depression cone is only partially dewatered. In addition, the void's structure involving depositional porosity and narrowness of joints as well as relatively isolated karst holes reduces the cavity share in limestone which was dewatered by gravity and will be subject to replenishment from flooding. There is nevertheless potential for a volume of floodable geological voids to be reckoned with and which might affect both the duration of the flooding process as well as potential gradients and isostatic compensation flows as the flooding proceeds.

TABLE V-1. ESTIMATED MINE VOLUME (WORKINGS AND VOIDS) FOR FLOODING OF THE RONNEBURG MINE

	10 <sup>6</sup> m <sup>3</sup>
Open mine workings	19.3
Backfilled mine workings, block cavings, wood etc.	9.3
Dewatered pore/joint volume adjacent to mine workings	7.0
Pore volume of back filled open pit and "innenkippe"	15.4
Dewatered pore/joint volume within depression cone	4–10
<b>Total</b>	54.7–60.7

Waste rock material deposited in the open pit's internal dump and waste rock placed in the pit for backfilling purposes offer a floodable pore volume of  $1.6 \times 10^7$  m<sup>3</sup> which will have considerable impact on the duration of flooding but only minor impact on the dynamics of the flow.

Towards the end of the flooding process, the anticipated ratio of mine cavities to geological voids impacted by mining activities to virgin geological voids to pore volume of open pit fill will be 50% to 11% to 11% to 28%.

The total volume to be flooded as well as the volume of anticipated recharge is in the first place dependent on the shape and extension of the depression cone. Therefore, determining the exact volume of the draw down by monitoring groundwater prior to and during the flooding process is an essential condition for predicting the flooding process.

Recharge of the flooding volumes is primarily by direct infiltration of meteoric water. In order to monitor varying infiltration rates it will make sense to select a number of infiltratopes, i.e. zones overlain by geomechanically disturbed rock and outcropping aquifers. Total infiltration into the flooding voids will average some 725 m<sup>3</sup>/h in normal year, in wet years it will be approximately 1000 m<sup>3</sup>/h.

#### V-4. RESULTS OF BOX-MODELLING TO PREDICT THE FLOODING PROCESS AT THE RONNEBURG MINE

Discretisation of the flooding voids into boxes (20 mining fields and specific areas of high infiltration) and cells (up to 22 levels) was required to allow three dimensional and transient modelling of the flooding process in a complicated rock mass characterized by a folded Silurian joint aquifer and numerous interconnections between mining fields and blocks via drift systems, rise drifts, and boreholes. In preparation for flooding an abundance of hydraulic dams and "brakes" have been installed underground. In sensitive areas mine workings were totally backfilled.

This gave rise to a total of more than 300 spatial unit boxes to which volumes of mining cavities and geological voids, amounts of water ingress and discharge as well as alimentation volumes were assigned.

Box specific volumes of infiltration, calculated for dry, normal, and wet years and the effects on infiltration rates by future surface remedial activities can be used to predict the flooding process and subsequent dynamics of groundwater as well as to predict local groundwater levels in specific mining fields.

Conventional simulators fail to model the flooding process within this system and the flow fields in the post flooding stage. Therefore, a new programme code was developed that allows a free structure of flow balance boxes within the flooding voids thus operating with a minimum of problem related balance boxes. For the first time, dynamics of flooding were calculated using that code.

For the calculation of the post flooding stage the box model was linked to a conventional pore model.

Water balancing does not merely consider flood waters in the saturated zone but also includes all operational artificial water transfers between mining fields.

Major results of the modelling include:

- Uneven distribution of cavities and groundwater restoration rates generate flows between mining fields during the flooding process.
- Predictions of changes in the groundwater regime during the flooding process is also possible in cases where the water balance is controlled by technical intervention;
- In the post flooding stage, hydraulic shortcuts via the drift systems cause dominate flows between the mining fields to occur rather in the drifts than in the aquifer;
- This would cause, among others, the groundwater to flow through the mine areas surrounding the backfilled open pit;
- Once the final flooding level volume will be reached, flood waters are expected to flow out almost entirely in a single valley which is characterized by its hydrogeological connection to the pore aquifer, its valley situation and location close to mine workings (Fig. V-1) ;
- The box model produced to allow balancing and forecasting of flooding processes also permits predictions of water table levels in individual mining fields and of volumes and directions of flow between boxes and cells. It is anticipated that the flooding process will take a minimum of 11 to 13 years for the flooding waters to flow out into the Gessen valley;
- During the flooding process, groundwater restoration will be slow in high cavity mining fields such as the Lichtenberg open pit. Compensation flows may occur between sources and sinks. The latter may be influenced by artificial tapping or alimentation;

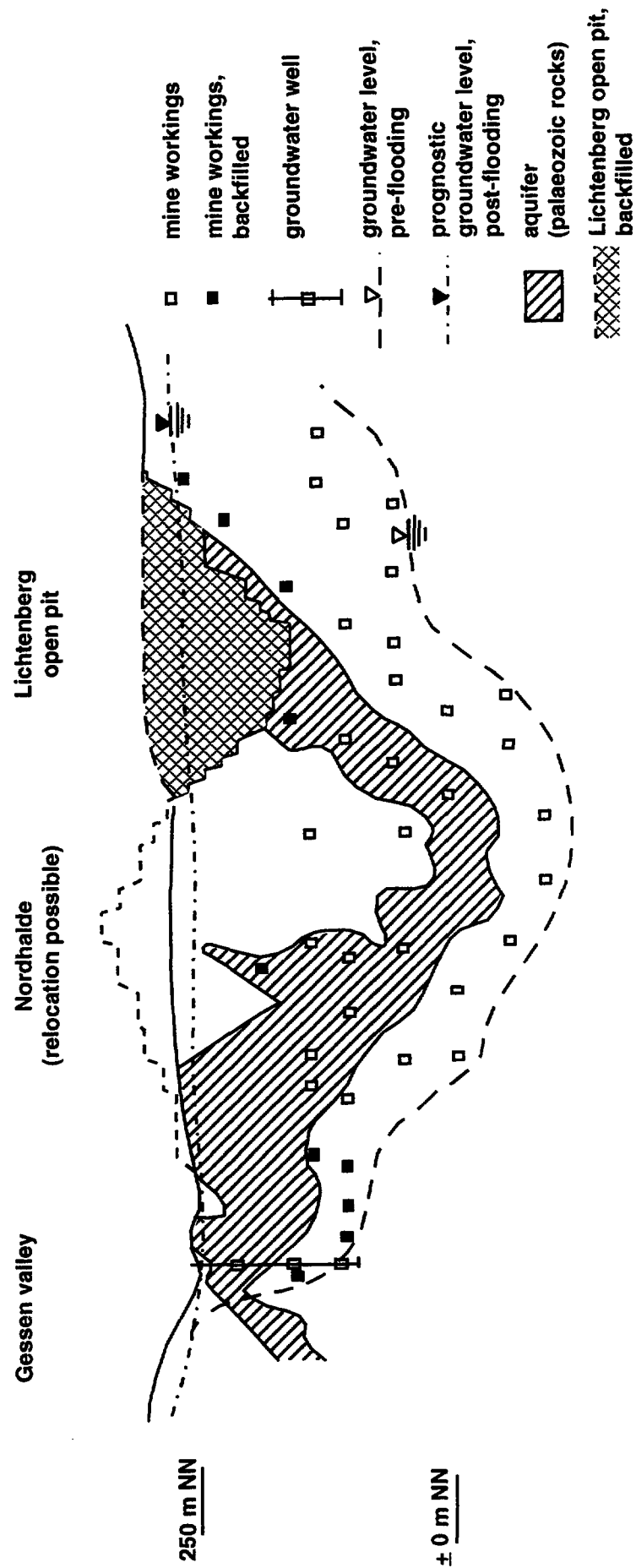


FIG. V-1. Hydrogeological connection to the pore aquifer, its valley situation and location close to mine workings.

SW

NE

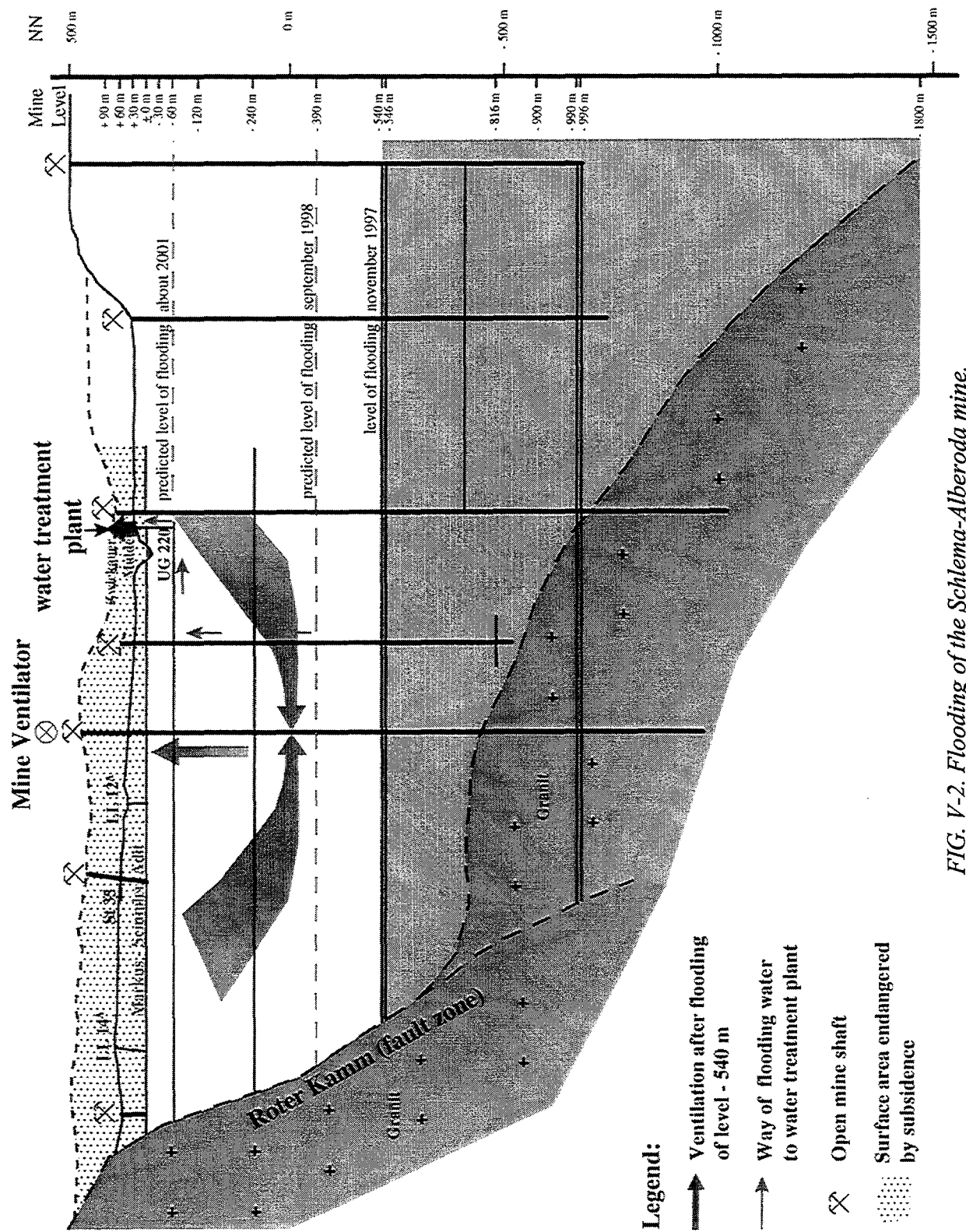


FIG. V-2. Flooding of the Schlema-Alberoda mine.

Using monitoring results of flooded parts of the mine and laboratory investigations as well as geochemical modelling it is anticipated that flooding waters flowing out at the surface, into the Gessen and Wipse valleys, will show initial concentrations as listed in the following table:

TABLE V-2. EXPECTED INITIAL CONCENTRATIONS OF OUTFLOWING MINE WATERS AT GESSEN AND WIPSE VALLEYS

Parameter	Unit	Gessental valley forecast	Wipsetal valley forecast
Water volume	m <sup>3</sup> /h	approx. 250	<50
pH		3-4	3-4
U <sub>nat</sub>	mg/L	<6	<4
<sup>226</sup> Ra	mBq/L	<500	<500
Fe <sub>tot</sub>	mg/L	<400	<400
SO <sub>4</sub>	mg/L	< 10000	<4000
hardness	dH	<300	<200

#### V-5. PREDICTED FLOODING PROCESS IN THE SCHLEMA-ALBERODA MINE

The rehabilitation strategy for the Schlema-Alberoda mine provides for the hydraulically controlled flooding of underground mine workings along with the filling of shafts and near surface mining voids and continued ventilation of some upper mine workings (Fig. V-2).

Flooding of the mine workings will reduce contaminant discharge of environmental relevance via the atmospheric and aquatic pathways in the medium and long term. Exclusion of air in the wake of flooding the workings is predicted to reduce physical and chemical mobilization processes in particular of arsenic, uranium, and radium.

When flooding waters will reach the level of 390 m asl, i.e. about 60 m below the level of the receiving stream, control of the flooding becomes a necessity to limit potential damage at the surface. This necessitates construction and operation of a water treatment plant designed to cope with up to 700 m<sup>3</sup>/h of pumped mine waters. Operating life of the plant may vary between 20 and 30 years.

Three dimensional modelling was used to predict the hydrodynamics of the flooding process and of thermodynamic convection processes in the flooding zone.

First, the mine workings were discretised using AutoCAD, assigning parameters such as geometry, voids, and conductivity to the respective levels and mining fields.

Filling of the flooding zone is the result of approx.  $4 \times 10^6$  m<sup>3</sup>/a of direct infiltration through near surface mining areas and of up to  $3 \times 10^6$  m<sup>3</sup>/a of groundwater intruding via the flanks.

Major input data included the geothermal gradient, conductivity rates of shafts, drifts, mining zones and virgin rock, thermal conductivity, specific thermal capacity as well as volumes

TABLE V-3. PREDICTED CONCENTRATIONS FOR OUTFLOWING FLOODING WATER

Parameter	Unit	Quality
pH		7.0
Dry matter	g/L	4
Filterable matter	g/L	0.02
Ca	g/L	0.3
Mg	g/L	0.3
Na	g/L	0.5
K	g/L	0.05
SO <sub>4</sub>	g/L	1.8
HCO <sub>3</sub>	g/L	1.1
Cl	g/L	0.1
U	mg/L	5.4
Ra	Bq/L	3.5
As	mg/L	3.5
Fe	mg/L	7.9
Mn	mg/L	4.6
Cu	mg/L	0.02
Pb	mg/L	0.002
Zn	mg/L	0.1
Ni	mg/L	0.02

and location of inflow. Cooling down of the rock mass as a result of mine ventilation was a thermal boundary condition to be considered.

The transmission constant or k-value is a parameter of specific sensitivity when it comes to predict realistic convection processes. This concerns in particular the permeability of shafts and drifts as well as of mining zones. Evaluations have indicated that shafts and those development drifts which constitute hydraulic connections between shafts determine the permeability of mining zones. Given the presence of numerous shafts, drifts and mining blocks, the mine as a whole is a perfectly communicating system in hydraulic terms.

Density differences of the water due to geothermal water temperatures are the driving force for the generation of convection boxes.

The first step in the modelling process was to calculate various design models to be used for phenomenological considerations to explain modes of action. It became apparent that permeability coefficients, mine geometry as well as hydraulic and thermal boundary conditions are relevant parameters that influence the generation and intensity, respectively, of convection cells.

Modelling of thermo hydraulic processes was by 3D models designed for that purpose. Extremely time consuming calculations showed that convection processes will inevitably occur and even persist at average water temperatures of more than 30C once the flooding has come to an end.

In the case of the Schlema-Alberoda mine, modelling results show that warm waters will rise in the shafts to cool down in the upper levels of the mine and subsequently sink down via the mining zones and the disturbed rock surrounding the shafts. Therefore, it can be anticipated that cold infiltration waters and warm waters will mix in greater depths. Stratification and limitation of convection to defined mine areas would presuppose large scale hydraulic sealing of vertical connections at all levels. This is not feasible due to time and economic constraints. Investigations are under way to determine the extent to which convection might promote contaminant mobilization in drifts, sumps, mining blocks, and the disturbed surrounding rock or whether rapid exchanges will rather reduce concentrations in the flooding water following discharge of mineralized pore waters and contaminants from the mining zones. For the time being it is anticipated that concentrations in the flooding water which is currently at the 600 m level below the surface will remain stable for some years after the end of the flooding process and will then gradually decrease to natural background levels.

The concentrations predicted for the flooding water flowing out at the surface are shown in Table V-3.

## V-6. CONCLUSIONS

With regard to potential emissions and to preventive and active water conservation by the timely construction and operation of water treatment plants and to in situ measures to be taken in the mines, reliable forecasts concerning the duration of the flooding process and the evolution of the flooding media are indispensable.

Numerical models are suited for this job to the extent that the sufficiently represent the complexity of mine workings and disturbed rock. For simulation calculations to provide reliable results, input data from monitoring must be collected at an early stage.

During the flooding process, models must be calibrated on the basis of monitoring results so that results might be upgraded with regard to the end of flooding situation which is of environmental relevance.

This might also give rise to technical measures to be taken at an early stage to modify dynamics of flooding by dam construction or injections or to influence water quality by in situ treatment with a view of limiting the subsequent contaminant discharge.

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## **Annex VI**

# **RADIOACTIVELY CONTAMINATED LAND AREAS AND WATERS IN BULGARIA**

## **VI-1. INTRODUCTION**

The major source for radioactive contamination of large land areas and groundwater in Bulgaria has been the uranium industry (mining and milling). The total contaminated area is approximately 20 km<sup>2</sup>, including 4 km<sup>2</sup> of forest. Parts of the contaminated areas are mountainous, in low productive regions, but the major part is agricultural land along the rivers Maritsa, Tundzha and Struma which had been used for in-situ leaching.

## **VI-2. HISTORY OF THE URANIUM INDUSTRY IN BULGARIA**

The first uranium explorations were in the Buhovo region near Sofia. The explorations had been performed by German geologists before the Second World War but no mining resulted from those activities. The uranium industry in Bulgaria actually began immediately after the end of the war. The first mines for uranium ore were started under the Goten peak in Stara planina (the Balkan mountains), next to Buhovo village, in 1945, and the ore was sent directly through the railway station of Yana to the former USSR. On a hill at the edge of Buhovo the first uranium ore processing facility was built in 1947. The wastes from this plant were dumped in the Manastirsko dere (Monastery gully) above the Yana village. A tailings pond was constructed in 1958.

New mining sites were discovered, studied and put into operation in other regions but the ores were transported to Buhovo for processing. In 1966, a second milling plant was built in Eleshnitsa which processed ore from South Bulgaria. That plant was more modern but it did not produce the final product (yellow cake) which was dried, ground and packed in Buhovo. The total number of workers for both milling plants was approximately 1000 and the total annual production of the final product was approximately 600–700 tonnes/annually or 35 000 tonnes for the production period of 1947–1992.

Experiments with the method of in-situ leaching for low grade ores started in 1968–69. Approximately 15 sites for in-situ leaching were started up over a period of about 20 years. These sites treated rather poor grade ore (between 0.006 and 0.030%, or from 60 to 300 g/t). The leaching mines reduced the number of underground miners from 5000 in the period 1965–1970 to approximately 500 in 1988. The in-situ leaching was experimentally applied also for leaching uranium from unused low-grade ores in classical underground mines. The method of the heap extraction was applied in both Eleshnitsa and Smolyan. Initially, the combined extraction was with sulphuric acid solutions and, later, the "soda" scheme was applied with Na<sub>2</sub>CO<sub>3</sub>, NaHCO<sub>3</sub> and NH<sub>4</sub>CO<sub>3</sub>. Near the end of operations in the uranium industry, approximately 30% of the uranium concentrate was obtained by in-situ leaching with all of its modifications. During the entire operating period of the uranium industry, only four open pit mines existed — "Dospat", "Eleshnitsa", "Deveti septemvri", and "Senokos". Only "Senokos" was functioning when the uranium industry began its close down.

All of the uranium mining sites are located in the southern part of Bulgaria (south from the Balkan mountains) and only one uranium site is in north Bulgaria (Smolyanovtsi).

### VI-3. ENVIRONMENTAL CONDITIONS NEAR THE PROCESSING (MILLING PLANT) SITES

The environmental conditions near the processing (milling plant) sites are discussed below. In Bulgaria, the uranium industry has established two uranium processing plants — at Buhovo and Eleshnitsa.

#### a) The Buhovo milling plant.

At Buhovo, the tailing ponds have an area of approximately 1.3 km<sup>2</sup> with dumped waste of  $1 \times 10^7$  m<sup>3</sup> and mass of approximately  $1.6 \times 10^7$  tonnes. The average specific activity is less than 5 kBq/kg U and more than 100 kBq/kg Ra. During normal operation of the plant, the process is closed and there should be no release of water. Nevertheless, water has occasionally been released but the content of the radionuclides from the uranium–radium chain is low: uranium 0.5–0.7 mg/L (compared with the limit of 0.6 mg/L) and reactivity is below the limit of 0.15 Bq/L. The corresponding values for the water of rivers Yanestitsa, Lesnovska and Mesta are much below the current limits. The drainage water, however, contains sulphates usually 5–6 g/L (the upper limit is 300 mg/L) and also some metals — Cu, Zn, Fe, Mo, Cd, Mn. Only the concentration of Mn is above the limit.

A special concern is for the large area contaminated with Ra. At some points, the specific Ra activity is 10 kBq/kg and the exposure rate reaches 10 µSv/h (1000 µR/h). The uranium processing plant in Buhovo is located approximately 15 km northeast of the city limits of Sofia. Because of the plant's operation for more than a decade without a tailings pond, approximately  $1.2 \times 10^7$  m<sup>3</sup> had been contaminated with radium. Most of the land use is agricultural but some of the contaminated land is within the limits of the neighbouring villages. In the beginning, immediately after the World War II, the ore was shipped by train through the Yana railroad station to the former USSR. In 1947, a small installation for chemical concentration was built which, after having its capacity increased, processed all the ore mined in Bulgaria up until 1968, when a second plant was built. In this plant, the ore is crushed and ground, oxidants are added to the slurry and, after leaching with acid and the selective extraction of uranium, the slime is neutralized and then dumped by hydro transport. Up until 1958, when a tailings pond was built, the wastes were dumped in Manastirsko dere (Monastery gully) above Yana village. The insoluble components precipitated in the gully and the rest of the slime drifted freely through the Yana village and then through the old river beds of rivers Buhovchitsa and Yanestitsa into the Lesnovska river. This has resulted in contaminated village yards in the Yana village and contaminated land of approximately  $1.2 \times 10^5$  m<sup>2</sup> in area in vicinity of the villages Yana, Gorni Bogrov and Dolni Bogrov (Upper Bogrov and Lower Bogrov), and on both sides of the road to the small town of Elin Pelin. Of the most contaminated land areas, 1.2 km<sup>2</sup> was capped and approximately 2.8 km<sup>2</sup> was forested. Measurements in the mid 1950s identified some hot spots with an exposure rate of 5 µSv/h, and reaching as much as 10 µSv/h in some locations. The long term radiation monitoring has shown that the exposure rates have decreased roughly by 30%.

The principal concern is that of contaminated land lying within the limits of the Yana village and for areas used for growing vegetables and other food stuffs for both people and the cattle. A certain portion of the contaminated land adjoins the tailings pond and the waste area of the nearby steel plant Kremikovtsi. The uranium content in soil is an average of 10 times and, for the most extreme case, 40 times above the mean content of uranium in soil. For the private

gardens, the uranium content is comparable with the natural content in soil. The uranium content of these soil is in average 20 times less than the content of uranium in contaminated soil, but the radium activity is approximately six times less for sediments and three times less for soil.

The measurements for uranium and radium in groundwater show that, due to sorption and filtration, the content and activities are very low and are close to the accepted limits for drinking water, even close to the tailings pond wall. Deep sampling (to depths of 100 m) show that uranium varies within 1–5 micrograms per litre (limit 600 µg/L) while radium in 6 out of 8 deep sampling wells is above the limit of 150 mBq/L, but in three of the closest wells the value is approximately five times that limit. For groundwater, the conclusion is that radium and uranium uptake is not a dominant radiation hazard.

Another radiation hazard is of course the inhalation of radon and radon daughters. The inhalation of dust which is partially radioactive is much less important since the specific activities of the soil and ores are low. A potential source term for dust is the dried edge of the tailings pond when the wind blows towards Yana; because of the distance, however, this hazard is fairly negligible. The measured maximal potential alpha energy for the Yana village is 18–440 MeV/L, the upper value of which corresponds to approximately 30% of the limit for the population. For the villages Gorni Bogrov and Dolni Bogrov, the radon contamination of the air does not differ from other places in Bulgaria.

#### **b) The Eleshnitsa milling plant.**

The environmental impact of the underground mines near Eleshnitsa is determined by:

- *The water from the mines.* The water contains natural radionuclides slurry, chemical agents, oil, and, in the case where combined methods (in situ leaching) had been applied, heavy metals. The main radiation hazard is due to radium and, to a much lesser extent, to uranium, thorium and polonium. The average uranium content of the surface water is above the upper limit of 0.6 mg/L, and the radium content is between 50 and 800 mBq/L (the accepted limit is 150 mBq/L). The radium content quickly decreases below the limit of 150 mBq/L in the tailings ponds.
- *The mined waste rock deposited on the surface.* The rock had been mined during earlier investigations and also during excavations around the ore layer. The mined rocks contain all of the daughter products of uranium in equilibrium. Some rocks are close to the accepted ore concentration limit of 0.025–0.030 %. The presence of radium causes the contamination of the air close to the rock heaps, but this does not extend beyond about 0.5–1 km. The erosion lead to a spreading of natural radionuclides in the hydro system, thus affected an area around the mines. At the present time, there are nearly 300 waste heaps spread over an area of  $8.3 \times 10^5 \text{ m}^2$ . The total volume of the waste is approximately  $1.3 \times 10^6 \text{ m}^3$ , of which the uranium containing waste is  $8 \times 10^6 \text{ m}^3$  with an average uranium content of 0.025% and a total activity of  $8 \times 10^{13} \text{ Bq}$ . The specific activity is estimated to be about 6 kBq/kg.
- *The ventilation systems.* The radon content in the ventilated air is 30–800 Bq/m<sup>3</sup> with potential alpha energies of  $(10\text{--}50) \times 10^5 \text{ MeV/L}$  for the different mines. The maximal recorded values for the closest villages of Seslavtsi and Eleshnitsa are 920 and 6000

MeV/L. The daily average value in Eleshnitsa is below the upper limit of 1330 MeV/L, but in certain periods when temperature inversions occur, the value of the potential alpha energy is close to the limit. The ventilation system also releases inert and radioactive dust and toxic (explosive) gases which are diluted very quickly.

The risk estimates for Eleshnitsa are based only on radon exposure outdoors and do not account for radon indoors. The annual individual exposure was estimated to be 0.82 of the working level (WLM). Hence, the cumulative exposure for a 50 year old person is about 40 WLM — a value which approaches some occupational exposures of miners.

#### VI-4. ENVIRONMENTAL CONDITIONS NEAR SITES OF IN SITU LEACHING

The ore body is about 120–200 m thick and extends over several hundred millions of m<sup>2</sup>. The land dedicated for the in-situ leaching is  $1.6 \times 10^7$  m<sup>2</sup> in area, of which  $2 \times 10^6$  m<sup>2</sup> is non-agricultural land and  $3 \times 10^6$  m<sup>2</sup> is forest. The land which has been actually used for in situ leaching is  $6 \times 10^2$  m<sup>2</sup>, which includes the mining land and the sorption installations. The incoming and the outgoing flow channels are spread in a network with separation distances of about 10 to 30 m between them; hence, all of the tubes and the pumping facilities cover an area of about  $0.5\text{--}0.6 \times 10^6$  m<sup>2</sup> which could be eventually contaminated. The main problem is that the greater part of the land is agricultural and is expected to be returned to the owners, so that the need for remediation is very urgent.

For each site, the quantity of solution which is processed daily ranges from 4 to  $30 \times 10^3$  m<sup>3</sup> with an average concentration of uranium between 5 and 20 mg/L. The concentration of salts varies from 15–20 g/L of which 10–12 g/L are sulphates and the rest are other salts and micro quantities of heavy metals and rare earths. There is a potential hazard for contamination of the surface of the land and also of the surface and undergroundwater. There is no air-contamination. The ion exchange resins are enriched to 40–50 g/kg or 1000–1300 kBq/kg; these could represent a radiation hazard if they were dumped or discarded.

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

The most serious problem is the radioactive contamination of the deep groundwater which reaches the region of the ore. The uranium concentrations there may reach 20–30 mg/L and that of radium 1–2 Bq/L. The same problem exists for the shallow groundwater in the case of accidental release and tube defects. For the shallow water, the recorded maximal concentrations are 3–4 mg/L uranium and 0.5 Bq/L Ra in spite of the dilution. In general during in-situ leaching, due to evaporation and hydro transport, the level of the groundwater is diminished which leads to a certain restriction of the contamination. After the leaching has been stopped, the contaminated water migrates. The water is slightly acidic and, if it reacts with the carbonates, a certain hydrochemical barrier could be formed. That is not a clear problem, however, and certain investigations are necessary to assess this possibility. Some of the sites (Bolyarovo, Tenevo, Okop) are close to places where water is used for drinking; this proximity could lead to a decrease

in the quality of the drinking water. After the closing of the uranium mining occurred, a special order was issued to prescribe the continuation of the solution recycling without adding acid. For most of the sites, the flow decreased and the salts began to deposit in the tubes and the filters of the sorption columns. The deposited salts have an increased radioactivity so that a possible contamination hazard can be expected when the tubes will have to be disassembled. When this problem was identified, it was ordered to stop the recirculation. For only one site (Selishte) was there not a problem.

The remediation of the in-situ leaching sites is not finished. The radiation hazard is less important than that of the chemical and acid contamination, sulphates and some heavy metals. It can be concluded that the main hazard is due to the sulphuric acid, the quantity of which is estimated to approximately 2.5 million tonnes. This problem is extremely important especially upon closure of the uranium industry. A case of special interest are the areas in the Gorna Trakiiska nizina (Upper Tracian plain) near Plovdiv where the soil is very good for agriculture and the deep water is considered to be an important reserve.

## VI-5. CONCLUSIONS

The main contamination problems for the uranium industry presently are due to its very sudden closure and the lack of needed funding for investigations and remediation procedures. Both land areas and surface/subsurface waters have been affected. These problems should be solved as quickly as possible although the contaminated areas are not large and the size of the population living close to them is relatively small. The radionuclides can migrate through underground and surface water and therefore purification facilities should be built wherever possible. The tailings ponds and waste heaps should be stabilized and rehabilitated to reduce or prevent water migration and radon exhalation. The intake of radionuclides from the in-situ leaching sites can be reduced by neutralization and site remediation. All of the rehabilitation measures can be realized with the labour of the workers of the former uranium industry, thus reducing the social tensions, as well as providing a cleaner, more acceptable environment for the local population. The most urgent problem to be addressed is the tailing pond located near the first milling plant at Buhovo.

## ANNEX VII

### NATIONAL EXPERIENCE ON GROUNDWATER CONTAMINATION AND REMEDIATION ASSOCIATED WITH URANIUM MINING AND MILLING IN THE CZECH REPUBLIC WITH CASE EXAMPLE FROM THE NORTH BOHEMIAN CRETACEOUS AREA

#### VII-1. BACKGROUND

In the Czech Republic, the pertinent groundwater contamination problems are associated with uranium mining and milling sites. The environmental problems of radioactive contamination are being addressed on two levels, operational and conceptual, as discussed below.

(1) *Operational level* — This involves monitoring, evaluation, and, following the limits of possible pollution of individual constituents of the environment which are given by regulations, preventive measures to avoid pollution and damage of the environment. Monitoring plans are being approved by the hygienic service. This activity is ensured by staff located at the individual areas.

(2) - *Conceptual level* — This can be characterized by conceptualizing and planning the most suitable remediation methods for the contaminated remains, residues and groundwater after closeout of mining and milling activities. This level is fundamental while the uranium production programme is being slowed down and closed out. This activity is carried out by the Czech mining company, DIAMO, headquarters staff and the staff in the individual areas.

The main environmental effects caused by the activities of exploration, mining and processing of uranium are described below.

- (a) There has been a change of the regime and chemical composition of groundwaters in the upper Cretaceous sedimentary complex in Northern Bohemia caused by the co-existence of classical deep mining and ISL technologies.

The main contaminants are ammonium, sulphate, and nitrate ions. The volume of contaminated water is 186 mil. m<sup>3</sup> in the cenomanian horizon and 72 mil. m<sup>3</sup> in the turonian horizon.

- (b) There are huge volumes of tailing impoundments covering large areas. The remediation of these tailing impoundments has two phases, preparations (planning) and realization.

In the preparations phase, the following actions occur:

- groundwater quality and surrounding biosphere is monitored and evaluated;
- environmental and population risk assessments are carried out; and
- proper remediation materials are obtained and tested.

TABLE VII-1. TAILING IMPOUNDMENTS IN THE CZECH REPUBLIC

<i>Tailing impoundment</i>	<i>Area</i> ha	<i>Volume</i> m <sup>3</sup> × 10 <sup>-3</sup>
Straz	187	19 200
GEAM	90	9800
MAPE	292	24 000
Pribram	44	240
Western Bohemia	20	2800

In the realization phase, the following factors are achieved:

- obtain and apply the proper technologies for the impoundment water treatment;
- install a cover over the tailing impoundment body to prevent water seepage into it and, also, radon emanations out of it; and
- incorporate (i.e. integrate) the site features into the surrounding landscape as part of the environmental restoration.

The insulation (isolation) of the tailings from the biosphere is the main objective of the remedial actions. In this regard, the following requirements have to be fulfilled:

- all measures have to be done in accordance with long term safety requirements;
  - the groundwater has to be protected and of acceptable quality;
  - radon emissions have to be within the given radiological limits; and
  - escape of radioactive elements into the biosphere has to be reduced to acceptable limits, if not eliminated altogether.
- (c) There are huge volumes of waste rock dumps to be dealt with. From an environmental point of view, the waste rock dumps are important due to their (negative) effect on appearance of the landscape, as well as their potential for spreading contamination to surface and groundwaters and to the atmosphere in the form of dust particles.

There are three main approaches to solving the problem of waste dumps, as follows:

- leave the dump in place, reshape and cover it with inert materials, and continue the long term monitoring and evaluation of groundwater quality;
- use the waste rocks for the tailings impoundments remediation and for other purposes after the area has been remediated; or

TABLE VII-2. SIZE OF WASTE ROCK DUMPS AT VARIOUS SITES IN THE CZECH REPUBLIC

<i>Waste rock dumps</i>	<i>Volume</i>
	m <sup>3</sup> x 10 <sup>3</sup>
Straz	1100
GEAM	3300
Pribram	30 000
Western Bohemia	2100
Jachymov a Horni Slavkov	14 000
Others	7400

- use the waste rocks as a building material (but, of course, this depends on its suitability to be used in this way).
- (d) Contamination of littoral belts of streams and stream sediments has been caused by mine water effluent discharges. There is only one example of a systematic effort to remediate such an environmental problem in the Czech Republic and it is the Ploucnice river in Northern Bohemia. The task was begun as a result of the government decision Nr. 366/92 and efforts on it lasted for two years; the results were examined, and the decision was made to continue the programme up until 1997. The principal results have been presented at the IAEA Symposium on "Radioactive Releases into Environment," which was held in Vienna, Austria in May (1995).
- (e) There has been a change in the stream regimes associated with the mine water effluent discharges and/or dewatering of areas associated with mining activities. This topic is partly associated with the above topic D. However, the main problem is associated with the volume of water effluents, which is sometimes equal to, or even higher than, the volume of the stream water, and then seriously changes the quality of the stream water.

The problem with dewatering of the surface streams occurs mainly in the Pribram area, where the mine area was between two streams basins, the Pribram Brook and the Kocaby River. The deep mine dewatering has influenced the flow of both streams, but the main problem is that, after the mine was closed and flooded, the Pribram brook does not have the same natural inflow of groundwater which it had before, because the this water now goes into the Kocaby river basin.

- (f) A change has occurred in the regime and chemical composition of groundwater in contact with old mine workings. After the exploration and mining activities were carried out and completed, many old and abandoned mine workings were left. Where these are in contact with groundwater, they can rapidly change its regime and chemical composition. It is then necessary to monitor such changes and evaluate the results. If the contamination levels are

not in accordance with the given regulations, some improvement measures have to be planned and implemented.

### **Summary of remediation concepts in the Czech Republic**

The Czech company, DIAMO, had worked out the ecological concepts already in 1994. The conceptual framework summarises the state of production and remediation works, the aims of ecological policy, assumptions concerning their achievement, a course of action for achieving the environmental goals.

The principal problems identified are the following:

- concentrations of contaminants in discharged mine waters;
- subsequent concentrations of contaminants in the surface and groundwaters; and
- the existence of secondary problems associated with "harmful" mine water contamination of sediments.

According to the remediation concepts that were established, the following aims can be defined in way of needed remedial actions:

- the quality of mine water discharges has to be such that, according to the legal limits, it will not endanger human health or the environment, nor should it increase the other risk of any other exposures.
- In the end, the technical activities in the area should be completed, leaving it to its natural destiny (i.e. in its natural state).
- The degree and extent of "secondary" problems should be determined and they should be eliminated as being of influence on the environment or of concern to the public.

The proposed course of action to reach the desired final state is as follows:

- (1) Creation of a suitable monitoring system with respect to specific conditions of all remediation stages — from the planning and preparation of technical solutions to the final phase involving only observation and documentation of the desired goal.
- (2) Selection and setting of acceptable risks (real and possible) and evaluation of their influence on the public and the environment and the public.
- (3) Proposal of technical solutions that will allow the smooth change-over from the remediation stage (with water purification) to the observational stage and free discharge of water.
- (4) Proposal of variant technological measures and selection of optimum technologies from the point of view of results to be achieved with spent means and materials.
- (5) Realization of proposed technical measures and remedial operations.

- (6) Feedback obtained through monitoring. Ongoing evaluations of the environmental state and assessment of the remedial actions efficiency.
- (7) Solution correction (re-calibration of methods) or end of remediation.

From the successful conclusion of remediation, it is necessary that, in the early phase (e.g. first stage after risk evaluation), the aims are clearly stated and the means established to allow accumulation of needed documentation (evidence) in the future.

To reach the successful conclusion, it is necessary to work public relations into the remediation programme. As experienced by DIAMO, It turns out that the neglect for on-time and complete public knowledge can bring serious complications at a high technical level in enforcing and solving contamination problems. As a matter of fact, timely, well conducted public relations and the providing of complete information can bring large savings in the overall budget available for remediation.

## VII-2. HISTORICAL REVIEW OF URANIUM PRODUCTION ACTIVITIES

The major area of the Czech Republic is formed by the Czech massif, geological structure, which belongs to one of the most important uranium ferocious provinces of Europe. Its mining history reaches far into medieval times. The term "Pitchblende" — pitchy looking ore from the silver mines of Jachymov — first appeared in the books by Georgius Agricola (1494–1555). Pierre and Marie Curie succeeded in preparing radium and later also polonium from the same pitchblende in 1898.

The relatively unimportant production of uranium ores used in the glass and ceramic industries and for the production of radioactive preparations was replaced after 1945 by an intense development of uranium ore exploration, mining and processing activities. Many important deposits, namely Horni Slavkov (1946), Pribram (1947), Zadni Chodov (1952), Rozna and Olsi (1956), Vitkov II (1961), Okrouhla Radoun (1962), Dylene (1964), Hamr and other deposits of the north Bohemian Cretaceous (1963-1968) were discovered during this period.

The last few years have been under a considerable contraction programme in the uranium industry in the Czech Republic. This was caused by the restriction of the uranium export to the former Soviet Union and the free market conditions in the Czech economy. This represents a decrease in uranium production from 2500 t in 1989 to 1539 t in 1992 and a further decrease to approximately 400–500 t in 1997. All the exploration, mining and processing activities were operated by the state for more than 50 years.

## VII-3. SOME PROBLEMS OF REMEDIATION WORKS IN THE CZECH REPUBLIC

The state financed the development of uranium production and the state enterprise transferred the main part of its profit to the state in the past. Under those economic conditions, the enterprise did not create the sufficient economic reserves for remediation. This is a reason, why the state funds the on-going uranium production contraction programme.

The national budget covers the following:

- liquidation of uranium production capacities;

- social welfare of laid off employees, according to legal obligations;
- remediation of the Straz area after in situ leaching (ISL);
- environmental investment and the so called Old Remainders Programme, i.e. liquidation of negative influence on the environment caused by uranium.

Based on the contraction programme, DIAMO is responsible for remediation activities. All activities must have a project. The project has to address the technical issues, the social programme for employees, environmental impact statement and financial budget required for the project realization. The project is then approved by the Ministry of Industry and Trade and is the basis for the project realization and its funds from state budget.

The classical decommissioning has three steps. The first step is the decommissioning of the underground, the second step consists of decommissioning of the surface facilities or their conversion to other purposes. The final step is the reclamation of the whole area influenced by uranium production activity.

There is a quite different situation in the area of the North Bohemian Cretaceous deposits. These are shallow and subhorizontal deposits with very complicated hydrogeological conditions. The deep mining of these deposits was different in comparison to vein type deposits. The mined out space has to be fully backfilled by special concrete to prevent any subsidence of the overlying formations. ISL uranium production using sulfuric acid on the Straz deposit also requires very different conditions for its remediation, decommissioning and close-down. All the operations in the area are closely connected with each other, mainly because of groundwater management.

DIAMO started and at the beginning co-financed "The Project of Old Remainders Inventory and Liquidation". The whole inventory of all remainders from uranium production, especially from the 50s and 60s, is the main aim of this project. The main problems are a treatment of contaminated water from old mines, decreasing of radon emanation, waste rock dumps reclamation and collapsing of old mine workings. Review of the main remainders left after uranium exploration, mining and processing activities in the Czech Republic:

- 500 exploration and production sites ,
- 360 exploration shafts,
- 300 exploration and production edits,
- 52 waste dumps at the shaft sites with 90 million metric tons of waste rocks,
- 14 tailings impoundments containing approximately 44 million metric tons of tailings,
- 1 area of ISL of 6.5 square kilometres.

#### VII-4. ENVIRONMENTAL PROBLEMS SOLUTION IN THE CZECH REPUBLIC

The DIAMO company is realising its activities in many places in the Czech Republic.

The following six have been the main mining and processing areas.

- Straz-Hamr area, the Ceska Lipa district;
- Dolni Rozinka, the Zdar nad Sazavou district;
- Mydlovary, the Ceske Budejovice district;

- Pribram, the Pribram district;
- Western Bohemia, the Tachov, Karlovy Vary and Sokolov districts;
- Okrouhla Radoun, the Jindrichuv Hradec district.

In addition to these areas, there were many exploration activities across the whole Czech Republic.

#### **VII-4.1. The main influences on the environment caused by uranium ores exploration, production and processing**

- (a) Change of the flow and chemical composition of groundwater in the upper Cretaceous sedimentary complex in Northern Bohemia caused by coexistence of classical deep mining and ISL technologies;
- (b) Large areas and huge volumes of tailing impoundments;
- (c) Huge volumes of waste rock dumps;
- (d) Contamination of littoral belts of streams and stream sediments caused by mine water effluent discharge
- (e) Change of stream regimes associated with the mine water effluent discharge and/or dewatering of areas associated with mining activities. This topic is partly associated with the topic D.
- (f) Change of regime and chemical composition of groundwater in contact with old mine workings

### **VII-5. CASE EXAMPLE OF AN IMPACT OF URANIUM PRODUCTION ON THE WATER QUALITY IN THE NORTH BOHEMIAN CRETACEOUS**

#### **VII-5.1. Introduction**

The classical uranium deep mining in the area of Straz-Hamr in the so called Straz block of the North Bohemian Cretaceous started at the end of the 60s and in a comparatively short time developed to a great scale. Two deep mines (Hamr and Krizany) were operating in this region. Parallel to this deep mining operation uranium production using ISL was developed not far from the Hamr deep mine. Both contrasting production methods influence each other, mainly in groundwater management. The third operation in the area, the Straz mill tailings impoundment built in 1980, also has a big influence on the groundwater. Its remediation will have to take account of all the operations in the area.

The geological situation can be concisely described as follows: the Cretaceous sediments of Cenomanian and Turonian age are unconformably underlain by metamorphosed Proterozoic rocks. The workable uranium concentrations occur mainly at the base of the Cenomanian.

From the hydrogeological point of view there are two important aquifers in the region. The upper Turonian unconfined aquifer containing high-quality potable water and the lower Cenomanian confined aquifer. The quality of the Cenomanian water is to a great extent affected by the occurrence of uranium ores (also in regions not containing workable uranium concentrations) and its water is not suitable for the potable water use.

#### **VII-5.2. Mine water from the deep mining in the region**

The quality of mine water reflects the main problem of the mining region as a whole, i.e. the coexistence of two different production operations close to each other, i.e. ISL on the Straz deposit and the deep mine at the Hamr deposit, approximately 1500 metres apart. From the point of view of the origin of the mine water and its chemical composition neutral pH and acid mine water are distinguishable. Neutral pH water is not influenced by ISL technology, acid mine water is affected by the leaching solutions of ISL.

The acid mine water is pumped predominantly from the region of the drainage cross-cut and from the forefield of the deep mine towards the ISL fields. From the areas not adjacent to the ISL fields only neutral pH water is pumped.

##### *VII-5.2.1 Technology of mine water purification*

The mine water purification process is very complex. There are some different types of mine water purification procedures.

Neutral mine water is cleaned in the central decontamination station (CDS) in two production lines:

- turbid mine water (pumped from mud sumps with uranium content) ;
- clear mine water (pumped from drainage wells).

The purification is accomplished by the following technological processes:

- sedimentation;
- flocculation;
- clarification;
- filtration;
- anion exchange (for uranium extraction) ;
- discharge into the retention basins and into the stream.

Acid mine water is subjected to a preliminary purification in the neutralization station (NDS-6) using the following technological processes:

- neutralization;
- sedimentation;
- filtration;
- chlorination.

The water purified by these processes is then fed into the CDS where it undergoes the final purification and is then released into the stream.

The filtration cake from the NDS is collected and deposited in the tailings impoundment.

The technological schemes described above have been in operation in the CDS since 1988 and in the NDS since 1986. At present these technologies treat about 15 Million m<sup>3</sup> a year without any important problems regarding quality and the released water meets all the water quality requirements.

#### *VII-5.2.2. Environmental impacts of released mine water in the region*

Mining of uranium, particularly in the years before the start of the CDS caused a contamination of the sediments of the PlouPnice river and its floodplain. The extent of this contamination is apparent from the results of an airborne gammaspectrometric survey of the Ploucnice river region from its source down to the town of Ceska Lipa. In 1992, s.p. DIAMO launched an extensive detailed programme for the exploration of the most important contaminated localities. The results showed that the contamination is not continuous but concentrated in small "hot spots" having the area of about tens to thousands of square metres. Any hazard affecting environment and public health is very low. It is, therefore, assumed that remediation measures will not be required.

The discharge of mine water was a problematical issue in the past, i.e. before the purification capacities of the CDS and NDS were put into operation. The concentrations of contaminants then exceeded the background levels several fold. This resulted in the creation of local contaminated areas in the floodplain of the PlouPnice river. Nevertheless the evaluation of hazards connected with the sites affected by this contamination so far now show that the contamination does not represent a significant hazard for the population and the environment.

The release of mine water will continue until approximately 2001. Nevertheless the surface watercourses will still be affected by the current remediation activities in the ISL area and from decommissioning of the surface installations and equipment of the mine area.

#### **VII-5.3. The Straz mill tailings impoundment**

The Straz mill tailings impoundment served as the final place for tailings deposition. These tailings originated from processing of the Hamr and Krizany deep mines, low-grade uranium ores.

The Straz mill tailings impoundment is situated in the so called Tlustec block, which, together with the Straz block, forms the northern part of the Bohemian Cretaceous basin. The Tlustec block is sunken for about 600m in comparison with the Straz block. The zone of the Straz fault, which is the border between these blocks, occurs near the tailings impoundment. Many smaller faults are connected with this fault. Three aquifers are present in this area. Confined Cenomanian and Turonian aquifers have a piezometric level between 0 and 25 m under the surface. This depends on the topography. The upper Coniacian aquifer is unconfined. These aquifers are separated by impermeable clayey layers.

Only the upper Coniacian aquifer has direct contact with tailings impoundment water. This aquifer is formed by fine-grained sandstones and has a thickness between 3 and 90 m. Sandstones have a very low content of rock cement and therefore they are very porous. The underlying rocks

of this aquifer are formed by a marly sandstones strata up to 100 m thick. This strata forms a border between Coniacian and Turonian aquifers.

Filtration coefficient in permeable sandstones has a value of  $10^{-5}$  m/s and in impermeable sandstones of  $10^{-8}$  m/s.

The Straz tailings impoundment has an area of about 180 ha (1.8 km<sup>2</sup>). It is divided into two parts: the 1st and the 2nd stage.

The western part (the 1st stage) of the tailings impoundment has been in use since 1980 and about 1.42 million m<sup>3</sup> of tailings have been deposited here. The tailings impoundment dams are formed by sandy-loamy materials. In the lower part, the impoundment is sealed by plastic foil. The present height of the dams is 28m. On top of this part, there is a lake, where mud were sluiced.

The building of the eastern part (the 2nd stage) of the tailings impoundment started in 1988, because large production development was planned at that time. This part was used for tailings deposition for only one year. The project for this stage was developed using old requirements. It did not require thorough bottom sealing. It was supposed that the bottom would be sealed by fine-grained particles from tailings. Because the deposit of tailings was stopped very soon, the bottom has a practically original character. At present, this part of the tailings impoundment is a lake with an area of about 90 ha (0.9 km<sup>2</sup>). This lake contains water contaminated by tailings from the Straz uranium mill. The water surface in this lake is approximately on the same level as the groundwater. The main contamination is 3 to 5 g/L of SO<sub>4</sub> ions. Underlying rocks of the impoundment are formed by permeable Coniacian sandstones with a filtration coefficient  $k_f = 2.5 \times 10^{-5}$  m/s.

Bedrock is created partly by old fish pond sediments and partly by fine-grained particles of sluiced tailings. Water seepage through the tailings impoundment dams is caught by the system of drain ditches and pumped back. Rainfall water from the tailings impoundment surroundings is caught by a second system of drain ditches and discharged into the PlouPnice river. Because the tailings impoundment complex was built in the lowest part of a shallow valley, it dams the natural draining age of groundwater.

Requirements for the environmental safety of the impoundment operation increased after 1989 and an extensive monitoring system was created. Observations were compared with the results of mathematical modelling. Using mathematical modelling, the following questions were investigated:

- how to minimize the influence of the impoundment on its surroundings under present conditions;
- how to restore the impoundment whilst ensuring protection of groundwater.

The system of monitoring wells was developed to observe the tailings impoundment influence on groundwater. Every week, the groundwater level is measured and static samples of groundwater are taken for chemical analysis. According to the needs, this sampling is completed by dynamic sampling, which helps to determine hydrogeological characteristics in the underground. These direct methods of taking chemical samples are completed by auxiliary geophysical methods, which determine underground parameters along the defined lines.

Regular measurements of the tailings impoundment influence on the groundwater offered were very important. Although the tailings impoundment is situated on permeable bedrock, spread of pollution was very slow and in some areas it stopped. These results led to doubts about the quality of this monitoring system. It was decided to work up a detailed study of the groundwater flow in the tailings impoundment area.

#### **VII-5.4. In situ leaching area**

ISL has attracted considerable public interest since 1990. There were many ideas about its future, ranging between the immediate closure and continuation of a relatively very high production levels. Both are not acceptable, because both of them are extreme solutions.

Therefore a huge programme of research and verifying works started. The results were summarised in a series of reports called Analysis of ISL. The first report published in 1990 had 60 pages only. The second one was a result of about one year works and had about 400 pages and many appendices and was completed in 1991. It was also translated into English. The third one consists of ten volumes and thousands of appendices. Its name is "Analysis of ISL on the Straz Deposit with Special Respect to Remediation Preparation and Remediation of the Environment". It also set up the initial conditions for remediation proposals.

##### *VII-5.4.1. Initial Conditions for Remediation Actions*

#### **Cenomanian aquifer**

Amount of contaminated water:	186 million m <sup>3</sup>
Area influenced by contamination:	24 km <sup>2</sup>
Main contaminants: SO <sub>4</sub>	3792.0 thousand t
NH <sub>4</sub>	91.4 thousand t
Al	413.0 thousand t
U	approx. 1.0 thousand t
Total amount of TDS <sup>5</sup> :	4.8 million t

#### **Turonian aquifer**

Amount of contaminated water:	80 million m <sup>3</sup>
Area influenced by contamination:	7.5 km <sup>2</sup>
Main contaminants: SO <sub>4</sub>	22.0 thousand t
NH <sub>4</sub>	1.3 thousand t
Total amount of TDS:	25–30 thousand t

Water in the Cenomanian aquifer can be divided into two types according to its quality as follows (because it is highly contaminated water the word solution is used from the technological point of view):

- concentrated Cenomanian solution;
- diluted Cenomanian solution;
- acidic Turonian solutions.

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<sup>5</sup> Total dissolved solids.

The following table shows the main qualitative characteristics of solutions and not influenced water before mining.

TABLE VII-3. MAIN QUALITATIVE CHARACTERISTICS OF SOLUTIONS AND NOT INFLUENCED WATER BEFORE MINING

	Unit	Cenomanian Solutions	Affected Cenomanian water	Affected Turonian water	Cenomanian water before	Turonian water before
pH	-	0.5	1.8–2.8	2.5–7.0	6.7	6.7
TDS	mg/L	50 000–100 000	5000–20 000	500–5000	140	100
SO <sub>4</sub>	mg/L	33 000–80 000	3300–13 000	50–3300	33	35
NO <sub>3</sub>	mg/L	600–1400	5–100	5–1000	<1	5.21
F	mg/L	150–250	5–50	0.5–25	<1	<1
U	mg/L	1–30	0–15	<1	0.02	0.01
Ra	Bq/L	50–90	30–70	0.1–1.0	8.739	0.074
H <sub>2</sub> SO <sub>4</sub>	mg/L	15 000–30 000	500–5000	<500	NA	NA

There is a risk of contamination spreading over a bigger area and a bigger volume of groundwater because of instability of the groundwater regime after the returning to natural flow conditions. There is also a risk of contamination overflow from lower Cenomanian aquifer to upper Turonian aquifer because of the structural-tectonic conditions. Mathematical modelling shows that leaving of 3 g/L in Cenomanian aquifer can cause 2x to 5x higher amount of ammonia ion in comparison to the drinking water standard in Turonian aquifer. Therefore the basic limits for Cenomanian groundwater decontamination should be lower than 3 g/L of TDS.

There is a big influence on the surface ecosystems in the area covering over 650 ha. The landscape and soil conditions were changed through a combination of forest activities, soil chemical composition was changed because of contamination and the water regime in the area was changed due to cutting off the forests and extent stripping operations.

#### *VII-5.4.2. The state of ISL before the end of March 1996*

The DIAMO activity at that time was based on three decrees of the Czech government as follows:

- No. 366/92 to results of a complex evaluation of ISL in the Ceska Lipa district and following work procedure to determine production and remediation of the Straz deposit;
- No. 429/93 about a change of concepts of uranium production and processing contraction programme and mothballing of the Hamr mine I;
- No. 244/95 to realization of uranium production and processing contraction programme in the Czech Republic.

Mainly the government decree No. 366/92 was important for the future remediation course in the area of Hamr-Straz. It determined the so called transition period with a special regime of ISL. During that period the basis for decision making process about the future of ISL were prepared. The uranium production was kept with acidification of well-fields on the lowest technological level to allow the continuous production to finish and remediation works beginning. That regime continued until the end of March 1996. The report from that time period was completed at the end of 1995. Also the environmental impact statement was worked out and is the part of the Basic Remediation Concepts of ISL.

#### *VII-5.4.3. The state of ISL after the beginning of April 1996*

On March 6, 1996 the new decree (No. 170/96) of the Czech government was issued. The main elements of this decree are as follows:

- to proclaim the liquidation of ISL in Straz from April 1, 1996;
- to set up obligatory limits of total dissolved solids (TDS) concentrations in water of Turonian aquifer dependent on its future assumed use in the influenced area;
- to submit a progress report annually year on May 31.

Based on this decision the frame course of liquidation and remediation of ISL and the programme of evaluation works in the preparation period of remediation of the area and rock environment influenced by ISL in the Ceska Lipa district was worked out.

#### *VII-5.4.4. Remediation aims*

There are three main aims of ISL remediation:

- (i) gradual decreasing of TDS in groundwater of the Cenomanian aquifer to reach limits which will reduce the risk for drinking water of the Turonian aquifer. Based on the present results it is known that the Turonian aquifer will be influenced even if the average of TDS in the Cenomanian aquifer is 3 g/L. Therefore the reached limit should be under 3 g/L of TDS and it will be achieved by gradual decreasing salinity in the Cenomanian aquifer. At the same time the gradual verification of a remediation limit and optimization of technologies has to be performed;
- (ii) gradual decreasing of TDS in groundwater of the Turonian aquifer to reach limits for drinking water;
- (iii) gradual incorporation of leaching fields surface into ecosystems with respect to the territorial systems of ecological stability;

Remediation of the Straz deposit includes remediation of the Cenomanian aquifer, the Turonian aquifer and incorporation of leaching fields surface into ecosystems. To reach the decreasing of TDS in solutions in the deposit there will be surface technologies — Station for liquidation of acidic solutions — Stage I and Stage II. (SLKR I and II) using evaporation technologies.

#### **(a) Remediation of the Cenomanian Aquifer**

Remediation of the Cenomanian aquifer will be ensured by recovery of solutions to the surface and their desalination. A part of the cleaned water will be discharged into a stream and a part will be injected into the Cenomanian aquifer for maintaining the needed hydrogeological conditions. Injection will be located out of the contaminated area not to dilute contaminated solutions. There are two main periods.

- (i) Preparation period. This period will ensure the hydraulic under balance of groundwater to reach the control over all solutions in the underground and increase the concentrations of solution in the central part of the deposit. Therefore the thickened solutions will be injected back into the Cenomanian aquifer;
- (ii) Remediation period. This period will ensure decreasing of TDS until the given limit concentrations are achieved. During this period the technology for processing of products of the evaporation station will be built. Possible commercially used products such as sulphuric acid, aluminium oxide will be the output in this period;

#### **(b) Remediation of the Turonian Aquifer**

Remediation of contaminated part of the Turonian aquifer will be performed in combination of three following procedures:

- (i) Injection of contaminated solutions into the hydraulic barrier (the Cenomanian aquifer) ;
- (ii) Selective recovery of the most contaminated solutions and their processing at membrane technologies;
- (iii) Discharge of less contaminated solutions into a stream (the PlouPnice river).

#### *VII-5.4.5. Water management between 1996 and 2004*

This period can be characterized by many changes in groundwater flow associated with remediation actions. The whole preparation period and the beginning of remediation period will continue in co-existence of classical deep mining and in-situ leaching. All actions are targeted to decrease the negative influence of this co-existence.

#### *VII-5.4.6. Conclusions*

This example shows the importance of evaluation of all aspects associated with mining mainly the final phase of it before the operation starts.

During the last five years a lot of exploration, research and evaluation works has been performed. For example it was geological exploration for evaluation of geological aspects of groundwater communication between both aquifers and their mutual influence. Also a lot of works has been done in the area of contaminated water processing technologies.

All the present results show that the total amount spent for remediation action could be smaller if all works were done during production from its beginning or even before it.

The total expected amount of money which will be spent for contaminated groundwater processing is expected at more than 1.5 billion USD in following 30 years.

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## GLOSSARY

*The definitions given below may not necessarily conform to definitions adopted elsewhere for international use.*

<b>ALARA</b>	An acronym for “As Low As Reasonably Achievable”, a concept meaning that the design and use of nuclear facilities, and the practices associated with them, should be such as to ensure that exposures are kept as low as reasonably practicable, with technical, economic and social factors being taken into account.
<b>Active flushing</b>	An engineered (artificially enhanced) version of natural flushing, often used to increase the groundwater magnitude and flow velocity.
<b>Adsorption</b>	See sorption.
<b>Alluvium</b>	A surface accumulation or near surface deposit of unconsolidated or poorly consolidated gravel, sand, clays or peats that are loosely arranged, unstratified or not cemented together.
<b>Bio-barrier</b>	A low permeability barrier which employs the growth of bacteria to block the pores in a geological formation, thereby retarding fluid flow.
<b>CERCLA</b>	Comprehensive Environmental Response Compensation and Liability Act (United States of America).
<b>Clastic dyke</b>	Geologic formation which can facilitate vertical transport of contaminants (preferential pathway).
<b>Cut-off wall</b>	A vertical barrier installed to prevent the horizontal migration of groundwater.
<b>dH</b>	Deutsche Härte = one degree dH = one gram CaO/100 Liter H <sub>2</sub> O.
<b>Displacement barrier</b>	A barrier constructed by forcing the barrier material into the ground without any associated excavation.
<b>Electrokinetics</b>	The use of an electrical field to remove contaminants from the groundwater or from soil.
<b>Excavated barrier</b>	A barrier constructed by removing soil material and replacing it with the a desired barrier material.
<b>Ex-situ technology</b>	A process applied external to the contaminated region, above ground.
<b>Extraction</b>	Removal (extraction) of groundwater via pumping.

<b>Funnel and gate</b>	A variation of a reactive barrier wherein low permeability barriers are employed to channel contaminated groundwater through a reactive barrier of treatment zone.
<b>Gradient, hydrological</b>	The rate of change in total hydraulic head per unit distance of flow in a given direction.
<b>Gradient manipulation</b>	See active flushing.
<b>Hydraulic containment</b>	Containment achieved through the manipulation by hydraulic means of the groundwater flow around a particular region of contamination in order to prevent further migration or movement of the contaminants.
<b>In situ technology</b>	A process applied in place (within the ground or contaminated region).
<b>Ion exchange</b>	A usually reversible exchange of one ion with another, either on a solid surface , or within a lattice. A commonly used method for treatment of liquid waste.
<b>Marl</b>	Friable earthy deposit consisting of clay and calcium carbonate.
<b>Mixed wastes</b>	Radioactive waste that contains non-radioactive toxic or hazardous materials that could cause undesirable effects in the environment. Such waste has to be handled, processed and disposed of in such a manner that takes into account the chemical as well as its radioactive components.
<b>Natural flushing</b>	The application of the existing groundwater flow and geochemical attenuating conditions to flush (remove) the contaminant from the region of concern.
<b>pH</b>	Negative $^{10}\log$ of $H^+$ - concentration; a unit of measure for acidity/alkalinity.
<b>Phytoremediation</b>	The use of plants to remove contaminants from the subsurface into a harvestable biomass.
<b>Plume</b>	The spatial distribution of a release of airborne or waterborne material as it disperses in the environment.
<b>Precipitation</b>	A standard chemical method that can be used in the treatment of liquid wastes where radionuclides are removed from the liquid by either forming or being carried by the insoluble product of a chemical reaction made to occur within the liquid.
<b>Reactive barrier</b>	Groundwater permeable geochemical barriers installed across the flow path of the contaminant plume allowing it to flow through while at the same time removing the contaminant (e.g. radioactive species) through interactions with the reactive component of the barrier.

**Sorption**

A broad term referring to the interaction of an atom, molecule or particle within pores or on the surfaces of a solid, the 'substrate'. Absorption is generally used to refer to interactions taking place largely within the pores of solids, in which case the absorption capacity of the solid is proportional to its volume. Adsorption refers to interactions taking place on solid surfaces, so that the capacity of a substrate is proportional to the effective specific surface area. Chemisorption refers to actual chemical bonding with the substrate. Physisorption refers to physical attraction, e.g. by weak electrostatic forces.

**Vadose zone**

Subsurface zone extending from the surface to the top of the capillary fringe overlying the groundwater.

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