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Remediation of Sites with Dispersed Radioactive Contamination

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REMEDIATION OF SITES WITH DISPERSED RADIOACTIVE CONTAMINATION

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FOREWORD

The IAEA attaches great importance to the dissemination of information that can assist Member States with the development, implementation, maintenance and continuous improvement of systems, programmes and activities that support a sustainable nuclear fuel cycle and nuclear applications, including the legacy of past practices and accidents. The IAEA has therefore initiated a comprehensive programme of work covering all aspects of environmental remediation:

(a) Technical and non-technical factors influencing a decision on environmental remediation;
(b) Site characterization techniques and strategies;
(c) The assessment of remediation techniques;
(d) The assessment of technical options for the cleanup of contaminated media;
(e) Post-restoration compliance monitoring;
(f) The assessment of the costs of remediation measures.

Dispersed low level contamination poses a particular challenge to those charged with its remediation. Many techniques are not efficient below certain concentration thresholds or entail more severe impacts on certain environmental compartments than the contamination itself. In such cases justification for remediation may not be given on radiation protection grounds, but remediation may still be demanded by the public.

This report examines a variety of technological options for dealing with dispersed low level contamination. The approaches are broadly grouped into the three categories of:

(1) Non-intervention;
(2) Containment;
(3) Removal.

The techniques related to the above three categories may need to be chosen in conjunction or consecutively for any given site in order to arrive at an efficient and long term solution. These techniques are briefly outlined and their advantages and limitations are discussed. The need for a holistic design for the remedial action is stressed.

The extensive body of references provided will help the reader to find more detailed information.

The IAEA officer responsible for the preparation of this report was W.E. Falck of the Division of Nuclear Fuel Cycle and Waste Management.

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1. INTRODUCTION

1.1. BACKGROUND

To respond to the needs of Member States, the IAEA launched an environmental remediation project to deal with the problems of radioactive contamination worldwide.

The term ‘remediation’ is used in this report to encompass all activities leading to reduced exposure to radiation and to an improved environmental and/or economic value of a site; it does not necessarily imply restoration to a pristine environmental state. The terms ‘rehabilitation’ and ‘restoration’ are often used interchangeably. In the context of this report remediation is taken to mean management of contamination (i.e. removal, containment and monitored non-intervention).

The IAEA environmental remediation project includes an IAEA Co-ordinated Research Project [1], as well as the participation of IAEA experts in concrete remediation projects when requested by individual Member States.

The IAEA has prepared several documents dedicated to particular technical or conceptual areas (see Table 1), including documents on the characterization of contaminated sites [8, 14], technical and non-technical factors relevant to the selection of a preferred remediation strategy and technique [3, 17], overview of applicable techniques for environmental remediation [5], options for the cleanup of contaminated groundwater [10] and planning and management issues [19, 20]. In addition, a number of other IAEA publications dealing with related aspects have been compiled under different IAEA projects; these include TECDOCs on the remediation of uranium mill tailings, the decontamination of buildings and roads and the characterization of decommissioned sites.

Detailed procedures for the planning and implementation of remedial measures have been developed over the past decade or so. A critical element is the characterization of the contamination and of the various environmental compartments in which it is found, in order to be able to evaluate the applicability of remediation techniques [8]. The chemical or mineralogical form of the contaminant will critically influence the efficiency of the remediation technique chosen. Careful delineation of the contamination will ensure that only those areas or volumes of material that are actually contaminated are treated [8]. This, in turn, reduces the amount of any secondary waste generated. Understanding the processes of migration and dispersal is particularly important when natural attenuation is to be relied upon as a mechanism for remediation and dose prevention (see Section 4.1).

TABLE 1. RELATIONSHIP BETWEEN RELEVANT TOPICAL DOCUMENTS PRODUCED OR UNDER DEVELOPMENT

Safety	Management	Databases	Technology	Special topics
Management of Radioactive Waste from the Mining and Milling of Ores [2]	Factors for Formulating a Strategy for Environmental Restoration [3]	A Directory of Information Resources on Radioactive Waste Management, Decontamination and Decommissioning, and Environmental Restoration [4]	Technologies for the Remediation of Radioactively Contaminated Sites [5]	Extent of Environmental Contamination by Naturally Occurring Radioactive Material (NORM) and Technological Options for Mitigation [6]
Monitoring and Surveillance of Residues from the Mining and Milling of Uranium and Thorium [7]	Characterization of Radioactively Contaminated Sites for Remediation Purposes [8]	Design Criteria for a Worldwide Directory of Radioactively Contaminated Sites (DRCS) [9]	Technical Options for the Remediation of Contaminated Groundwater [10]	The Long-term Stabilization of Uranium Mill Tailings: Final Report on the Co-ordinated Research Project 2000–2004 [1]
Remediation of Areas Contaminated by Past Activities and Accidents [11]	Compliance Monitoring for Remediated Sites [12]	A Worldwide Directory of Radioactively Contaminated Sites (DRCS), http://www-drcs.iaea.org [13]	Site Characterization Techniques Used in Environmental Restoration Activities [14]	Remediation of Sites Contaminated by Hazardous and Radioactive Substances [15]

TABLE 1. RELATIONSHIP BETWEEN RELEVANT TOPICAL DOCUMENTS PRODUCED OR UNDER DEVELOPMENT (cont.)

Safety	Management	Databases	Technology	Special topics
Derivation of Remediation Values for Areas with Radioactive Residues from Past Activities and Accidents and their Implementation [16]	Non-technical Factors Impacting on the Decision Making Processes in Environmental Remediation [17]		This report	
Removal of Sites and Buildings from Regulatory Control Upon the Termination of Practices [18]				

Although contamination may originally be dispersed, secondary processes may concentrate, fractionate or otherwise redistribute the radionuclides; for example, it was observed that Chernobyl fallout was concentrated in geomorphological depressions by near surface migration processes (Fig. 1) [21]. Conversely, erosion may lead to the dispersion of radionuclides, particularly in mining environments.

The application of a remediation technique requires holistic studies examining the technical feasibility of the proposed measures, including analyses of their impact. Consequently, input from various scientific and engineering disciplines, including the health sciences, chemistry, physics, geology, microbiology and environmental engineering, is necessary in order to develop technical solutions. It is also necessary to include information on the political, social and economic context. Factors such as the overall project cost versus availability of resources over time, public perception and the availability of skilled workers need to be considered [17].

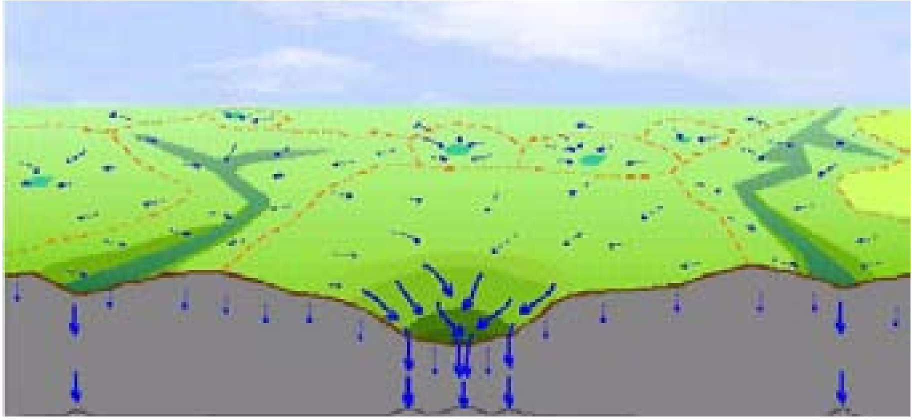

FIG. 1. Schematic representation of radionuclide accumulation in topographical depressions due to near surface migration, soil erosion, etc. [21].

It is essential that the process of decision making be transparent, as this makes the decision taken more defensible to possible public scrutiny and is also an essential element of quality control. Formal methods to aid decision making have been developed over the years. A recent IAEA document describes these in more detail [17]. A generic scheme of decision making and technology assessment is given in Fig. 2.

Techniques for remediating sites with well defined contaminated areas containing relatively high concentrations of contaminants are well established. The remediation of dispersed contamination, however, still constitutes a challenge when considering factors such as expected dose, cost, public perception and anxiety, and minimal disturbance of the environment. Any remedial action in an area containing dispersed contamination has to be justified, in that it must remove or reduce the source term or prevent the likelihood of exposure to a harmful dose. In order to optimize the intervention, a reasonable balance has to be found between the risk of exposure due to the potential radiation dose and the cost of the remedial action [22]. Similar considerations apply to further lowering the residual contamination over larger areas following prior remedial measures.

As large scale remediation operations can be disruptive to agriculture, natural ecosystems or (engineered) waste management sites, suitable implementation strategies that take into account the socioeconomic boundary conditions need to be developed [20]. An earlier IAEA publication, which was developed following the Chernobyl accident, focused on the planning of emergency response and remediation measures [20]. Since its publication in 1991, extensive experience in related fields has been gained [23]. One of the main conclusions from this experience is that the answer to the problem should

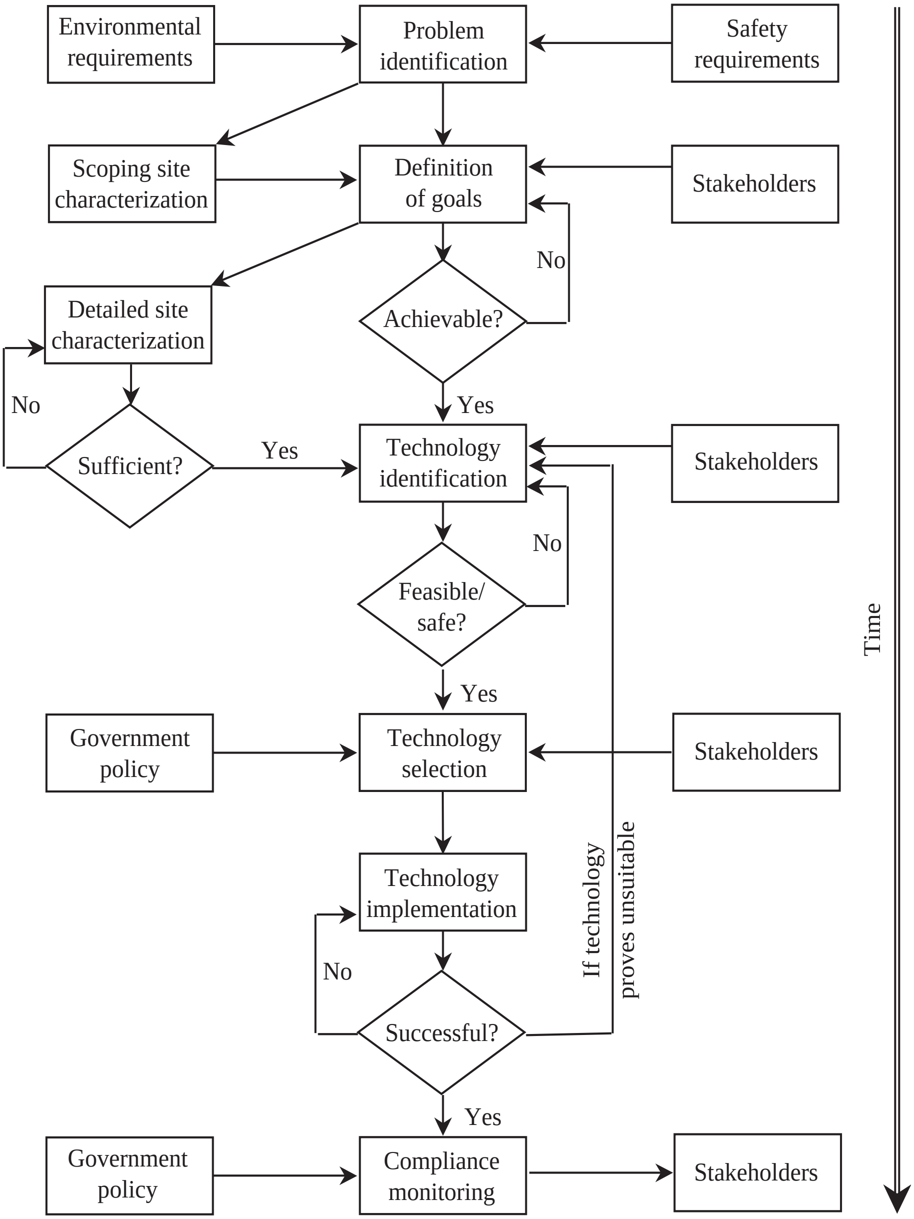

FIG. 2. Phases of decision making in remediation strategies and technologies [17].

be sought in low cost, low intensity, low maintenance (‘passive’) and often low technology solutions. Treating the contamination typically gave rise to the problem of having to dispose of large quantities of slightly contaminated material, mostly topsoil [24]. In the case of short lived nuclides, in situ methods to prevent direct exposure or uptake via the food pathway have been successfully developed [25].

1.2. SCOPE

This report describes remediation techniques that are applicable to dispersed radioactive contamination at a variety of sites, including surface soil, the vadose zone, surface water, sediments and groundwater. This report does not consider protective actions or emergency countermeasures to reduce uptake in humans and domestic animals through the food pathway, for example by feeding Prussian blue to cattle [26]. Strictly speaking, these are not remediation measures, since they only disrupt certain exposure pathways; they may, however, be needed in intervention situations to supplement remedial measures. Guidelines on their application have been published elsewhere [27].

This report is intended for individuals interested in the design, selection, review or approval of projects to remediate sites containing dispersed radioactive contamination and provides a basic overview of the state of knowledge for decision makers in government and at the community level and for consultants.

1.3. STRUCTURE

This report is divided into seven sections. Section 2 outlines the nature of dispersed radioactive contamination. Section 3 explains the fundamental technical choices in making decisions about remediation and outlines basic evaluation criteria influencing technology selection. Section 4 discusses the monitored natural attenuation and other non-intervention options. Section 5 describes the technical options for containing dispersed contaminants, while Section 6 is devoted to removal techniques. A summary evaluation of the technological choices is provided in Section 7.

2. DISPERSED RADIOACTIVE CONTAMINATION

A variety of activities and accidents may result in dispersed (non-point) sources of radioactive contamination. For the purpose of this report, dispersed contamination refers to the occurrence of concentrations of radioactive isotopes distributed over a wide area, where complete removal and disposal of the source is not practicable. Such contamination may present a hazard to humans and the environment. Examples of sources include inadequate practices for the management and disposal of radioactive waste, accidental radioactive releases to the environment, major nuclear accidents, nuclear weapon tests and incidents involving radionuclides at nuclear installations or other user establishments (such as hospitals and industrial and research facilities) [28]. A practice is any human activity that potentially introduces additional sources of exposure or exposure pathways or extends exposure to additional people or modifies the network of exposure pathways from existing sources so as to increase the exposure or the likelihood of exposure of people or the number of people exposed. Past practices that were not adequately controlled have led to significant radioactive contamination of areas in Member States. Some old facilities that processed radioactive material when radiation protection criteria were not as stringent as they are today are sources of radioactive contamination. Radioactive contamination can also result inadvertently from human activities involving processes in which naturally occurring radioactive material (NORM) can be concentrated, in areas not normally controlled by regulatory bodies, to levels beyond the limits set for practices [6]. Such activities include conventional ore mining and processing (such as copper ore mining, phosphogypsum production or mineral sands processing).

Contamination may take various forms and have different impacts on human health and environmental media; for example, a deliberate or accidental discharge of material produced during mining tends to produce dispersion plumes [28–31]. Conversely, accidents involving nuclear power plants have, in the past, led to isolated but widely dispersed hot spots of activity distributed across otherwise uncontaminated areas (e.g. Dounreay and Chernobyl). The two cases are distinct and pose very different problems for workers engaged in remediation.

International Commission on Radiological Protection Publication 60 stipulates that “remediation measures shall be justified by means of a decision aiding process requiring a positive balance of all relevant attributes relating to the contamination. In addition to the avertable annual doses, both individual and collective, other relevant attributes shall be assessed” [32]. Thus “remediation shall (a) reduce the doses to individuals or groups of individuals being exposed; (b) avert doses to individuals or groups of individuals that are likely to arise in the future; (c) prevent and reduce environmental impacts from the radionuclides present in the contaminated area” [12]. A generic reference level for aiding decisions on remediation is an individual existing annual effective dose of 10 mSv from all sources, including natural background radiation. At existing annual effective doses of less than 10 mSv, remediation would not normally be justified on radiation protection grounds. An existing annual equivalent dose of 100 mSv (inclusive of all existing contributions, including doses due to the natural background radiation) to any organ will justify intervention under almost any circumstances [22]. Decisions on remediation will usually have to take into account other factors besides radiation protection and, in particular, the views of those groups of the public that may be affected by the situation and by any remediation action. In this context, the selection of non-intrusive and less environmentally disruptive techniques will be preferable [17].

Finally, whether the contamination remains dispersed over time will depend on the chemical and mineralogical characteristics of the contaminant, environmental transport processes and receptors. Physical, chemical and biological processes leading to the concentration and fractionation of radioactive species are discussed further in Section 4.

3. FACTORS INFLUENCING TECHNOLOGY SELECTION

3.1. FUNDAMENTAL TECHNICAL CHOICES

The objective of any technique used in a remediation project is either to remove or reduce the source term or to block the exposure pathways. This can be achieved in a variety of ways and needs to be tailored to the contaminants and pathways of interest. It may be necessary to use a suite of techniques to achieve the remediation objectives, especially for source term isolation or removal.

In the case of dispersed contamination, a rigorous assessment of the actual and potential pathways is required to determine the optimal action. This assessment begins with the identification and consultation of records, if available. The historical assessment needs to be confirmed by a physical site characterization, for example by walk-over gamma ray measurements. Detailed sampling and analysis may be needed to more clearly identify hot spots and to delineate materials that do not require further attention. In recent years, a variety of strategies and techniques for efficient site characterization have been developed [8, 33, 34].

Discrimination between zones containing different levels of activities is a major challenge in reducing the areas and volumes of material to be considered [8, 35]. Once a site has been characterized, the decision makers are faced with their first fundamental choice for the intended remedial action. They must decide whether they will:

(a) Leave the site undisturbed, while establishing a monitoring scheme for determining the evolution of the site. This option relies on natural processes to prevent significant exposure. The entire process needs to be carefully monitored so that alternative action can be initiated if required.
(b) Contain or restrict the mobility of the radioactive contaminants: this involves immobilizing the contaminants inside the area in which they already exist, reducing the potential for further migration or entry into active pathways of exposure.
(c) Remove the radioactive contaminants from the site, using an appropriate treatment scheme: this involves extracting, concentrating and then safely disposing of the contaminants at another location.

The three generic options that represent the fundamental technical choices for remediation can be summarized as monitored non-intervention, containment and removal. Each of these fundamental technical choices will direct decision makers to follow substantially different paths with regard to their subsequent choices, actions and potential results, making available significantly different technological options for application (Fig. 3). In addition, since a variety of remediation techniques exist for containing, reducing or removing contamination, the technologies illustrated in Fig. 3 are grouped by the primary emphasis of the technology into containment and separation or extraction. The groupings are not necessarily mutually exclusive; for example, a barrier system may be used to contain and extract a contaminant, and, in some cases, the use of a particular technique may occur on or off the site.

This choice, as discussed in Refs [3, 17], cannot be made solely on the basis of scientific or engineering considerations. In addition to technical constraints, there may be a wide range of regulatory and socioeconomic constraints on the selection of an appropriate remediation or disposal strategy [17, 36]. Regulations in Member States may favour certain techniques and prohibit or discourage others. International agreements may also preclude or restrict some strategies. As has been discussed in detail elsewhere [29], the local population may want to participate in the remediation decision making process; public acceptability can be a major factor in selecting a particular remediation technique. Active inclusion of the public will increase their knowledge and awareness of the problem, increase acceptance of the

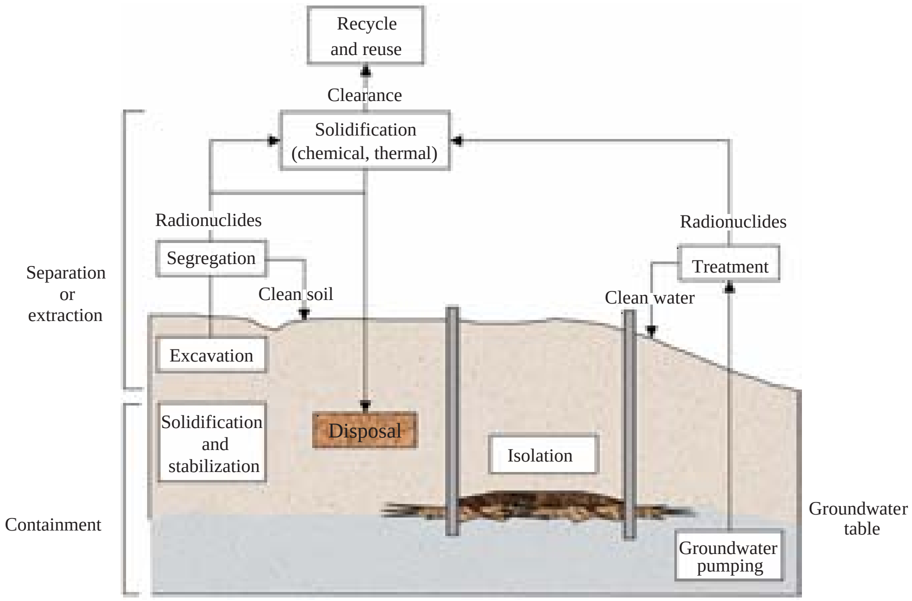

FIG. 3. Classification of remediation technologies by function (adapted from Ref. [34]).

remediation technique selected for deployment and increase acceptance of restrictions on land use that may result. Participation may also enhance the public's willingness to support the long term maintenance of remediation measures and related installations.

Once these points have been clarified, measures may be chosen taking the considerations described in Sections 3.2–3.8 into account.

3.2. TECHNOLOGY SOURCES

A wide variety of remediation techniques are now commercially available or at the demonstration stage. Although most of these techniques are generic in nature, others use proprietary formulations of reactants and other agents or applications that are protected by patents or similar means. Since the field is continuously developing, formal methods to assess the applicability and effectiveness of techniques have been developed [37]. Decision aiding tools make clearer the evaluation criteria used to support the selection of a technique to meet the remediation objectives [3, 5, 12, 38].

Approaches to selecting techniques vary from country to country. Some countries regularly undertake technology assessments to help ensure that proposed projects are effective and efficient [39–49]. The findings typically are made accessible in technology directories [50–54] or bibliographies [55, 56]. There are also international, semi-governmental, industry or research community sponsored initiatives [57–65]. Technology and technology suppliers directories are available [66, 67]. Other Member States and organizations rely on informal approaches, for example based on personal judgement by experts and managers, to select techniques.

Once measurable remediation objectives have been established, several factors affect the decision making process [3]. These basic evaluation criteria include engineering and non-engineering considerations:

(a) Effectiveness in remediating the contamination;
(b) The costs associated with the remediation programme;
(c) Occupational safety and health risks associated with the technique;
(d) Prior experience with application of the technique;
(e) Sustainability of any institutional control required;
(f) Socioeconomic considerations.

3.3. EFFECTIVENESS

The use of the term ‘effectiveness’ in this section refers to the ability of the technique to contain, reduce or remove the contaminant in order to prevent exposure of humans or undue harm to other properties of the site. Typically, an overarching objective is that the remediation improves the radiological situation by removing or reducing the source term or eliminating exposure pathways that entail a health risk and that it is not detrimental to the environment [68]; for example, the annual dose is decreased, or the functionality of soils is retained or future land uses are not unnecessarily restricted.

Site specific considerations exert a significant influence on the effectiveness and efficiency of the chosen remediation method. Since the mineralogical and geochemical characteristics of the contaminant vary among contaminated sites, remediation methods are not universally effective and efficient. Methods to model and predict the effectiveness of techniques under consideration have been developed [69, 70]. The anticipated performance of a given technique can be simulated and compared with similar results from other techniques to facilitate the selection [71]. The remediation action will be complemented by a post-remediation assessment and monitoring programme to ensure its efficacy [72] and that also may be part of any institutional control required for residual contamination [13, 33, 73].

Steps have already been undertaken to incorporate remediation activities into the ISO 9000/14000 quality management systems [71, 74]. Record keeping is an integral part of quality assurance and/or quality control. It is essential that records be kept of remedial actions undertaken, so that at any later point in time their performance can be evaluated against the original design. Having comprehensive documentation available also facilitates intervention in the event of unsatisfactory performance.

3.4. COST

The term ‘cost’ in this section covers the direct expenditure of funds associated with the remediation technique. These include funds for licensing fees for the technique, for equipment, labour and materials to deploy the technique, for design, construction management and treatability studies, for operation and maintenance, and for monitoring and the disposal of residual waste. Standard engineering cost principles can be applied to develop cost estimates for remediation techniques. Major cost elements can be long term monitoring, surveillance and maintenance. Depending on the time for which institutional control is required, provisions need to be made for funding these activities over periods of decades or even centuries.

Cost data for a wide variety of remediation techniques are available from various sources; for example, the appendix of a recent IAEA document [17] provides an overview of remediation costs, drawing on national directories such as the Historical Cost Assessment System (HCAS) [75] in the United States of America, which provides useful material for a relative cost assessment of the techniques listed.

3.5. OCCUPATIONAL SAFETY AND HEALTH

The term ‘occupational safety and health’ in this section covers the potential hazards and risks to workers involved in implementing the remediation technique [3]. Safety risks may result from accidents during deployment. Health risks may result from workers being exposed to radionuclides. Occupational risks vary substantially among techniques and may be an important consideration in selecting a given technique.

Information on safety and health risks can be obtained from standard reference sources, regulatory standards, medical surveillance, safety studies, toxicological data and epidemiological studies [76, 77].

3.6. SECONDARY ENVIRONMENTAL IMPACTS

The implementation of a remediation project may result in a variety of environmental impacts in addition to those resulting from the contamination itself. Environmental risk involves adverse impacts on ecological receptors located on or off the site due to a significant disturbance to the site ecosystem and its surroundings as a result of remediation; for example, certain techniques, such as the removal of topsoil or soil washing, may remove surface contamination at the cost of destroying the soil ecosystem.

Depending on the size of the site, an area larger than the actual contamination may be required for installations, the intermediate storage of waste and so on. The removal, transport and disposal of residual waste may result in environmental impacts and risks at locations other than those of the original contamination. There is, for example, little benefit in removing a contaminant that is well fixed on a low volume of soil, only to produce a high volume of an aqueous waste with the contaminant in a soluble or mobile form. In addition, the remediation techniques chosen may generate large quantities of secondary waste and may pose risks of exposure of the public or operators that exceed the risks of quiescent contamination [78].

Environmental risk resulting from the implementation of remedial actions may also extend to possible impacts on natural resources such as surface water, groundwater, air, geological resources or biological resources. The potential for environmental risk may be an important factor in decision making, since remediation technologies are more likely than others to produce adverse impacts on ecological receptors, including habitat disruption, or to generate damage to natural resources.

3.7. PRIOR EXPERIENCE

The term ‘prior experience’ in this section covers experience of implementing the remediation technique at other sites. It can be very useful to know whether the technique has been used successfully in the past. Information on previous deployments is available from a number of sources, including vendors, regulatory authorities, professional organizations, Internet databases, trade associations and publications (see Section 3.2).

3.8. SOCIOECONOMIC CONSIDERATIONS

The term ‘socioeconomic considerations’ in this section covers political, social and economic factors that may influence the selection of a remediation technique and its application at a site with dispersed radioactive contamination. The legal and institutional framework, prevailing socioeconomic boundary conditions and public perceptions can influence the choice and deployment of techniques to remediate sites with dispersed radioactive contamination [17]. The level of public reassurance generally increases with the degree of intervention and hence cost of the operation [29].

4. NON-INTERVENTION

4.1. BASIC CONSIDERATIONS

A decision not to intervene in site cleanup implies reliance on the capacity of natural media (rocks, soils, sediments and groundwater) to retard contaminant migration (i.e. natural attenuation) or on physical, chemical and biological processes to reduce activity levels to below those of concern (i.e. dilute and disperse). In either case, environmental monitoring is required to verify that such an approach is effective for the system under investigation. It should be noted that, ultimately, all remediation options that do not entail complete removal of the contaminant source would de facto revert to this solution if the half-lives of the radioactive species exceed a few hundred years.

It is also important to draw a distinction between those radioelements that occur naturally and those that do not, such as technetium and the transuranics. In the case of the former, reference may be made to the known geochemistry of the element in a given environmental medium. This provides a degree of confidence in predicting future migration behaviour. For artificial radioelements, experience is generally limited to laboratory data or small scale field trials. The use of ‘chemical analogies’ in this context may be necessary but is far from ideal [79].

4.2. DILUTION AND DISPERSION

Radioactive materials are discharged routinely into the air and surface waters, both fresh and marine, from nuclear facilities worldwide. Prior to 1985, dilute and disperse was also used for solid nuclear waste, although this practice has since ceased following the London Convention [80] and OSPAR (Convention for the Protection of the Marine Environment of the North-East Atlantic) Convention agreements [81]. Fewer restrictions apply to non-nuclear radioactive waste, including effluents from mining and industrial activities producing NORM. In the North Sea, for example, radioactive scale from pipe work is removed in offshore facilities, milled and released with no further treatment [82].

The effectiveness of dilution in aqueous media is critically dependent on the speciation of the radioelement under the prevailing environmental conditions [83, 84]. This will control factors such as solubility, adsorption to surfaces, bioavailability and toxicity. Many radiologically important elements may be concentrated by geochemical and/or biological processes, leading to secondary sources of potential contamination [85–87]. Similarly, physical dispersion of solids may not always be effective if the size and density of the particles differ significantly from the ambient environment.

There is no doubt that, even where not proscribed by legislation, the dilute and disperse option is opposed by regulators, environmental groups and the public at large.

4.3. NATURAL ATTENUATION

4.3.1. Principles

The concept of natural attenuation has received a great deal of attention in recent years [88–91]: it constitutes the least invasive approach to environmental restoration. The concept is not new; for example, it forms an integral component of the design criteria for geological repositories that depend on geochemical processes to retard radionuclide migration to the biosphere. It is not entirely without financial cost. Reliance on natural attenuation requires adequate monitoring, owing to the evolution of natural systems with time and our incomplete understanding of the processes operating. The effects of any change in land use or in water abstraction would also need to be assessed, hence the increased use of the term ‘monitored natural attenuation’ in the literature [92–94].

A large number of processes can contribute to natural attenuation, as discussed below. Figure 4 illustrates the effect of some of these processes on the migration and concentration distribution of radionuclides. In order to be effective, they must prevent or delay the arrival of a radionuclide at a receptor until such time that it will have decayed to an insignificant level.

4.3.2. Physical processes

4.3.2.1. Radioactive decay

The half-lives of radionuclides now present in the environment range from microseconds to many millions of years. For higher members of the natural series (^{234}U, ^{235}U, ^{238}U, ^{232}Th), together with some transuranics (e.g. ^{239}Pu) and fission products (e.g. ^{99}Tc, ^{129}I), no substantial decay will have occurred even on the longest assessment timescale. However, many other isotopes produced by nuclear fission (e.g. ^{60}Co, ^{90}Sr, ^{137}Cs) or contained in industrial NORM will not persist beyond a few hundred years. Clearly, it is therefore essential that a detailed radionuclide inventory be compiled before deciding to adopt natural attenuation as a management policy at any given site. The extreme fractionation between members of a decay series caused by chemical processing precludes the assumption of secular equilibrium in the majority of cases [95].

4.3.2.2. Filtration

In most situations the dominant exposure pathway is via flowing water. Resistate minerals (e.g. monazite, zircon and barite), other insoluble materials, for example cement, or particulate matter on to which radionuclides have become bound may be retarded by filtration. This will depend on the relative size of the particles and the pore distribution of the host medium, although even small colloids may be removed by fine grained clay matrices or fibrous peat. In the case of aquifer transport, adequate characterization of the hydrogeological flow regime (permeability, hydraulic conductivity, heterogeneity, fracture distribution) is a prerequisite for a quantitative assessment. Variably saturated conditions and geotechnical issues also have to be taken into account for surface deposits.

4.3.2.3. Volatilization

Radon produced by decay of parent radium isotopes will escape from well ventilated soils or heaps, and hence the progeny will be subject to

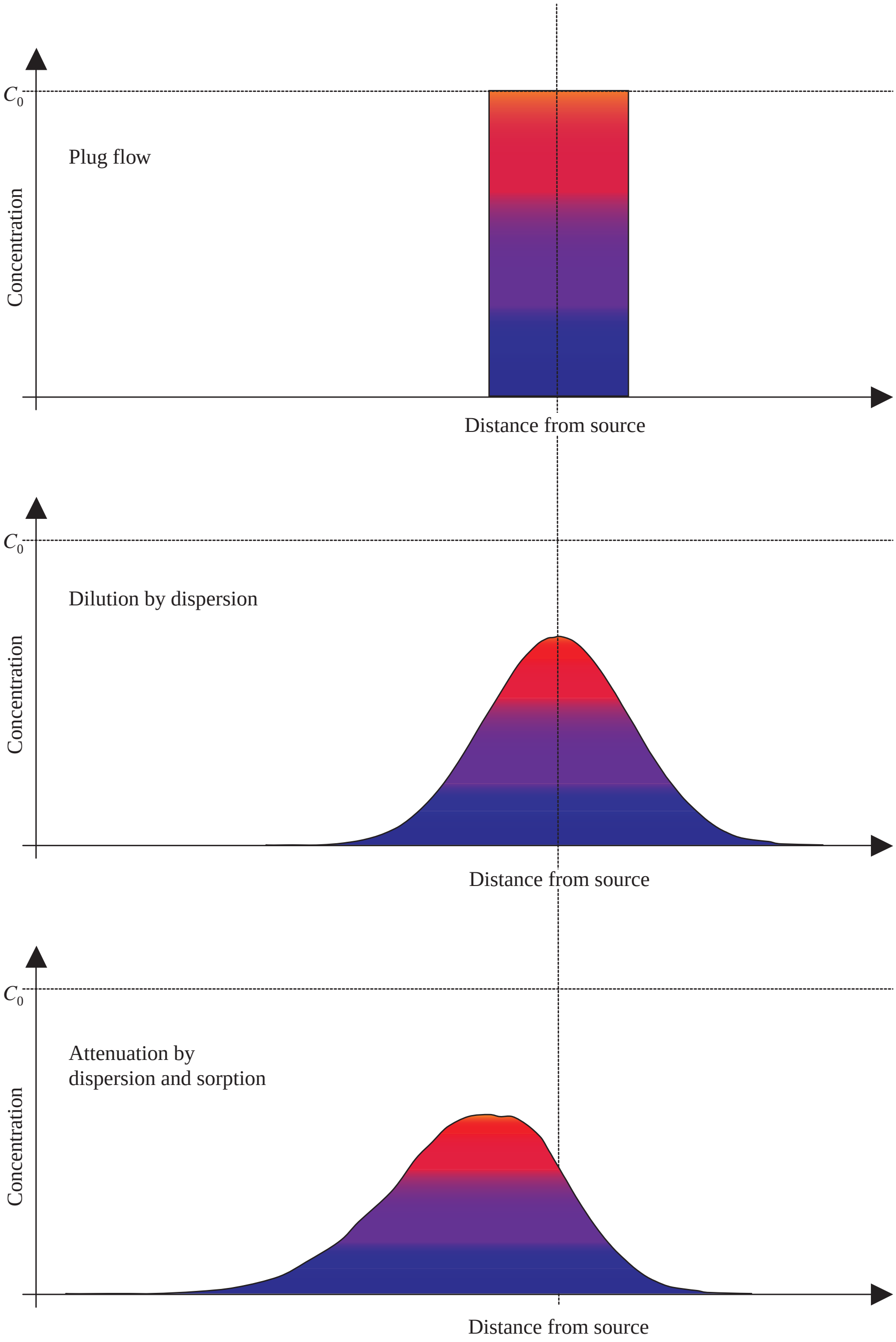

FIG. 4. Transport mechanisms effecting dilution and attenuation.

atmospheric dispersion. Methylated and permethylated forms of bismuth, lead, polonium and selenium (e.g. Ref. [96]) microbially generated in the subsurface can also be volatilized.

4.3.3. Chemical processes

4.3.3.1. Precipitation

Relatively few natural series radioelements and no artificial isotopes will exist in sufficient mass concentrations to precipitate as a pure phase from surface, pore or groundwaters: the exceptions are uranium, lead and thorium.

Uranium is a relatively mobile element in the near surface zone, owing to the stability of U(VI) aqueous complexes. However, it may be precipitated by reduction to U(IV) or in the form of uranyl minerals, principally phosphates, silicates, arsenates, vanadates and oxyhydroxides, several of which may occur simultaneously at the same locality [97]. Around 150 such phases have been identified and their formation represents the dominant fixation mechanism for uranium in weathering environments [97]. It follows that the amount of uranium released to groundwaters or surface waters from these secondary sources will depend on the solubility and dissolution rate of the phases as a function of pH and water composition. Unfortunately, too few quantitative data exist at present to allow predictive modelling [98, 99], an issue that needs to be addressed.

Lead, for example, may precipitate as the insoluble sulphide galena (PbS) that will incorporate ^{210}Pb by isotopic substitution. Thorium occurs only in the tetravalent state and is substantially insoluble except at very low pH. Where mobilized, for example in acid mine drainage (AMD), fixation occurs rapidly, often within a few micrometres, via the formation of silicates or, in the absence of silica, oxyhydroxides [100].

4.3.3.2. Co-precipitation

Radionuclides present at very low mass concentrations can nevertheless form solid phases by co-precipitation in mineral lattices. An important example from both a nuclear and a NORM perspective is the high selectivity shown by radium for barite, a mineral that has been very well characterized [101] and is also exploited in a remediation context (e.g. Refs [102, 103]). It is likely that transuranic isotopes would be similarly incorporated in uranium and lanthanide bearing minerals. There is evidence that this would be the case from extensive laboratory studies [104], although it remains to be confirmed in the field. Establishing the geochemical controls on migration of artificial radioelements is the major challenge to workers involved in the remediation of legacy nuclear sites.

Co-precipitation on ferric oxyhydroxide flocs is an extremely efficient removal mechanism for a large number of radioelements in solution. As the contaminants tend to be released upon crystallization to goethite, the process is often classified under the more general heading of sorption.

4.3.3.3. Sorption

In its strictest sense, sorption refers to the non-specific and reversible uptake of ionic species at charged surface sites. Used loosely, it has come to encompass aspects of co-precipitation, ion exchange and a number of ion specific interactions that are more appropriately termed complexation. The distinction is not made here other than in the case of co-precipitation, described above, as the latter clearly extends beyond the surface, resulting in the formation of a defined mineral phase [105].

Clay minerals typically show a strong affinity for radionuclides in the cationic form. Geological media with a high clay mineral content are therefore more likely to effect attenuation. Adsorption and ion exchange would be expected to play an important role in retarding the migration of soluble monovalent and divalent ions. Examples include the pronounced retention of caesium on zeolites (e.g. clinoptilolite) and the substitution of strontium for interlayer cations in smectites. Surface sorption is an important transient for multivalent ions in the formation of new mineral phases.

4.3.3.4. Complexation by organics

A number of radionuclides exhibit significant migration potential in the presence of aqueous, low molecular weight organic compounds [106]. Equally, however, immobile organic matter in the form of peat [107–109] or organic rich horizons in soils and sediments may provide an excellent substrate [107, 110] for radionuclide retention. These phenomena have been studied extensively in the context of ‘natural analogue’ studies for the performance assessment of radioactive waste repositories [86, 111–113]. Uranium approaching percentage levels has been reported in peat from Canada and northern Europe, whereas iodine, often considered to be a conservative tracer in such assessments, has been shown to be fixed in organic rich lacustrine deposits [114].

4.3.4. Biological processes

Biomineralization, 'biosorption' and microbially mediated phase transfer also can effect attenuation. These processes are discussed in more detail in Sections 5.5–5.7, 6.8 and 6.11. The complex biogeochemical processes effecting the fixation or mobilization of metals, including radionuclides, in various types of soil ecosystems have been studied with increased intensity in the aftermath of the Chernobyl accident [115] and in other remediation contexts [116].

4.3.5. Definitions and applicability

Whether to intervene or to rely on natural attenuation can only be determined on a site by site basis [117]. Factors militating against intervention include:

(a) The areal extent of the contamination;
(b) The accessibility of the site;
(c) The proximity to sensitive receptors;
(d) The radionuclide inventory;
(e) The time frame;
(f) The presence or absence of co-contaminants;
(g) The chemical and mineralogical characteristics of the material;
(h) In the case of surface deposits, the geotechnical stability;
(i) The transmissivity of the host medium.

A comprehensive site investigation programme is essential to determine these factors.

The degree of confidence that can be ascribed to natural attenuation in preventing harmful exposure or environmental damage is proportional to the level of characterization of that site. Developing an understanding of the physical, chemical and biological processes operating is more crucial in the case of natural attenuation than if the contamination were to be removed physically from the site.

A decision to apply monitored natural attenuation (Fig. 5) as the preferred management strategy will invariably be made by considering a combination of scientific, economic and political criteria. Ideally it should be based on a prior risk analysis of the specific site and follow an established technical protocol [118]. Models exist for evaluating the likely performance of such systems, for designing the monitoring programme and for the interpretation of monitoring data [112, 113]. Given a backdrop of scarce resources, various initiatives are under way to promote the acceptance of natural

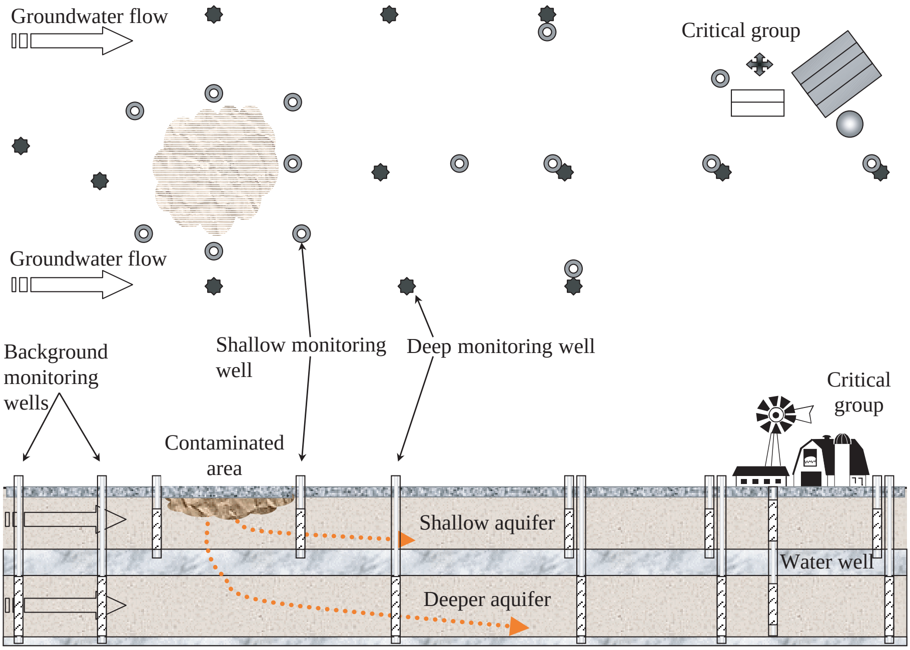

FIG. 5. The principle of monitored natural attenuation.

attenuation as a part of a cost effective and environmentally sound solution for radioactively contaminated sites worldwide (e.g. Ref. [119]).

It may be possible to modify some property of the media hosting the contamination in such a way that the retention capacity of the site is substantially enhanced. Examples include the addition of certain minerals, revegetation (see Section 5.6) and the creation of wetlands [120] (see Section 5.7). Strictly, any attempt to modify the characteristics of a contaminated site represents a form of intervention; for this reason, such measures are discussed further in the next section.

4.4. ALTERNATIVE LAND USES

When extensive areas have been contaminated, many of the discussed remediation methods may be too expensive to carry out or too intrusive. In particular, when the land was used for agricultural purposes, alternative uses may need to be considered. Such alternative uses may range from switching to different crops to turning to completely different uses, such as parkland.

When selecting an appropriate countermeasure for a given area and contamination problem, the optimum solution will depend, apart from radiological criteria, as much on economic, social and political factors as on sound scientific considerations [121].

Many studies have been targeting possible agricultural countermeasures in response to concentration levels in food and agricultural crops exceeding the applicable standards. Most studies have been conducted to test the effect of different physical and chemical countermeasures. However, information on the long term effect of countermeasures, and especially of a change to non-food crops, is still scarce.

When investigating alternative crops, the principal questions to be addressed are:

(a) Can an alternative crop be found that is suited to the climate and soil conditions prevailing in the contaminated area?
(b) What is the fate of the radionuclide in the cultivation system and along conversion routes?
(c) How does the radionuclide in question behave during biomass processing and what is the expected radionuclide concentration in the end products?
(d) What is the exposure during biomass cultivation and processing?
(e) Would production and utilization of the alternative crop be economically feasible?
(f) What are the overall prospects for the chosen alternative crop as an alternative land use for large contaminated areas?

In order to understand the fate of the various radionuclides and their distribution in products, residues and waste, one needs to know the various radionuclide fluxes. These depend on the initial deposition levels, crop accumulation factors, which in turn depend on plant and soil characteristics, and the radionuclide accumulation in the produce (e.g. wood, rape or beetroot). Whether residues and waste need to be treated as radioactive waste depends on the radionuclide concentration and the applicable exemption limits.

Crops used for liquid biofuel (oils, alcohol) production, such as rape, wheat, sugar beet, barley, potatoes and winter rye, may be suitable alternative crops.

The data in Table 2 indicate that crops with a low transfer factor (TF) to the useable product can be found and that the resulting liquid biofuels are almost free from activity, and that ^{137}Cs levels in the waste and residues are generally of no concern.

Examples from Belarus, however, show that caesium levels in oil cake from rapeseed oil (~2000 t/ha) and the pulp and vines from sugar beet (~4000 t/ha)

TABLE 2. CAESIUM TRANSFER FACTORS TO DIFFERENT PLANT PARTS OF SOME POTENTIAL BIOFUEL CROPS [122]

Crop	Plant component	Caesium TF (10^{-3} m^2/kg)
Spring wheat	Straw	0.23–0.36
	Grain	0.13–0.16
Winter wheat	Straw	0.27–0.44
	Grain	0.08–0.18
Rye	Straw	0.43–0.60
	Grain	0.17–0.29
Spring rape	Green mass	0.33–0.81
	Straw	0.38–0.92
	Seeds	0.27–0.66
Brassicaceae	Seeds	0.037–3.4[a]
Peas	Seeds	0.69–1.25
	Straw	0.82–1.45
Leguminosae	Seeds	94 (12–750)[a]
Sugar beet	Root	0.43[b]
Root crops	Root	0.025–11 (1.1–110)[a]
Green vegetables	Leaves	0.07–4.86[a]
	Leaves (peaty)	260 (25–2700)[a]
Sunflowers	Straw	1.48–2.88
	Seeds	0.43–0.82

[a] From Ref. [123] (average and 95% confidence limits).
[b] From Ref. [124].

may be too high for use as animal fodder and for incineration and that they may have to be disposed of as radioactive waste [125]. This, however, applies primarily to cases of high contamination levels, since only about 3% of the total ^{137}Cs and 6% of the total ^{90}Sr taken up will be potentially involved in the soil–plant–fodder–animal–human chain [126]. Moreover, oil cake and sugar beet pulp usually constitute only 10% of fodder, and hence generally animal fodder can be used without restriction [126]. The actual cost of liquid biofuel typically is significantly higher than that of fossil fuels, and hence a price subsidy or other measures may be needed [125, 126].

On the other hand, the production of rapeseed and processing to edible rapeseed oil are profitable technologies [122, 126], and the levels of caesium and strontium in the rapeseed oil after three filtrations and bleaching are below the detection limit [126].

The feasibility of valorization of contaminated land by willow short rotation coppice (SRC) for energy production has been addressed in various studies [125, 127–129]. Coppicing is a method of vegetative forest regeneration by cutting trees at the base of their trunk at regular time intervals. Fast growing species of the *Salix* genus (willows) are frequently used in a coppice system because of the ease of their vegetative reproduction and the large biomass produced. The harvested biomass is converted into heat or power (with an appropriate off-gas treatment). As such, this non-food industrial crop is a potential candidate for the valorization of contaminated land that has use restrictions. SRC may be preferred over traditional forestry since revenues come sooner after establishment and more regularly (every 3–5 years). SRC yields are also high on good agricultural soils, and its use is not a drastic change in land use; SRC is easy to introduce and it is easy to return the land to the production of food crops. SRC may also be considered as complementary to forestry, given the different culture requirements of both vegetation systems. Forests perform well on sandy soils, whereas SRC requires soils with a sufficient water retention capacity. SRC has additional potential advantages in a contamination scenario: since it is a perennial crop, dispersion of radionuclides will be limited. Harvest can be in winter, when the soil may be covered by snow, resulting in radiation protection of the workers. Finally, SRC cultivation is not too labour intensive, which is also an advantage with respect to exposure.

Willow SRC may be a suitable rehabilitation tool for highly contaminated land, but only if the radionuclide levels in the wood are below the exemption limits for fuelwood, if the average yearly dose received during coppice cultivation and coppice wood conversion is acceptable, if SRC can be grown successfully in the contaminated territories (soils, climate), if the cultivation of SRC is technically feasible and if SRC production and conversion are economically profitable.

It has been shown [125, 127–129] that for soils with a medium to high fixation (finer textured soils) and sufficient potassium availability, the TF ratio of concentration in plant biomass to concentration in soil is $<10^{-5}$ m^2/kg, and wood can be safely burnt and the ashes can be disposed of without concern. For light textured soils, however, with a low radiocaesium fixation and low soil potassium, the TF to wood is around 10^{-3} m^2/kg, and concentrations in wood may be elevated enough that the prevailing exemption limits are reached. Given that TFs for common forestry and for straw of winter wheat and rape are comparable, the same applies for burning wood or straw for energy.

SRC has generally a high annual yield of about 12 t/ha, but sandy soils are only suitable for SRC production if well fertilized and irrigated. Only during the conversion phase and when burning highly contaminated wood (3000 Bq/kg) do doses in the vicinity of ash collectors exceed the level of 1 mSv/a for a member of the general public [125]. Contributions from other possible exposure pathways are negligible (external exposure during cultivation and transport, inhalation dose in the combustion plant and doses to the public following wood burning).

Crop yield and the capital cost of the conversion units are among the most important parameters affecting system profitability. At the production site, a minimum yield of 6 t/ha/a is required for Belarus production conditions and of 12 t/ha/a for western European conditions, if all other parameters are optimal [125, 128]. Heating schemes may be a viable option for wood conversion in Belarus, whereas electricity generation schemes are not. Subsidies would be required in Europe to make wood conversion economically feasible. It has also been concluded that the existence of a contamination scenario does not necessarily hamper the economic viability of the energy production schemes studied. The cost associated with the disposal of contaminated ashes was estimated as less than 1% of the biofuel cost and will not affect economic feasibility.

Forestry can also be considered to be an adequate alternative land use [130, 131]. Soil to wood TFs to coniferous and deciduous wood are around 10^{-3} m^2/kg [132] and are hence comparable with the TFs to willow wood observed for low fertile soils with limited caesium fixation. They are high compared with the TFs observed for willow in finer textured soils and soils with an adequate potassium status. Moreover, the annual biomass increase is only 6 t/ha for forests and may attain 12 t/ha for SRC grown on soils with an adequate water reserve and fertility status. SRC may hence be a more promising land use option on these types of soil than traditional forestry. On soils with a low water reserve (e.g. sandy soil), however, willow yield without irrigation is too low to be economically feasible, and forestry may hence be the preferred option [129].

Fibre crops are also potential alternative crops for agricultural land with restricted use. Potentially suitable crops are the annual fibre crops hemp (*Cannabis sativa* L.) and flax (*Linum usitatissimum* L.). Hemp and flax are well known arable crops that have been cultivated for centuries. Ukraine has a legacy of flax and fibre hemp cultivation, but in Belarus there is only some flax production. Since the early 1990s the acreage for production of flax and hemp has declined dramatically in Ukraine. Establishment of fibre crops on contaminated arable land is generally of no radiological concern [133]. The TFs observed to hemp fibres are a factor of 4 to 50 higher than the TFs observed to flax. Cultivation is hence generally restricted to not too contaminated areas

(<1000 kBq/m^2). For both crops it holds that contamination levels in the waste products (oil seed cake, chaff, ash after burning of straw) may, however, be high enough that they should be considered as radioactive waste. The economics of this land use has not, however, been investigated [133].

The introduction of alternative crops in a contamination scenario may be a feasible and adequate remedial option. Although there are some scenarios in which energy production from SRC and potentially other alternative crops on contaminated arable land is radiologically safe and economically feasible, installing this cultivation system on a large scale requires extensive logistics, infrastructure and initial investment. Implementation is likely only to be successful with adequate political support.

There are a number of additional types of alternative land use, such as the creation of parkland. Such measures, however, would largely be administrative and would amount to 'institutional control' and therefore are not considered further in this report.

4.5. AGRICULTURAL COUNTERMEASURES

Many studies have been concerned with possible agricultural countermeasures in response to concentration levels in foodstuffs and agricultural crops exceeding the permissible levels in the wake of the Chernobyl accident; for example, the efficiency of countermeasures for radioactively contaminated natural and agricultural ecosystems was evaluated recently in the context of Chernobyl in Ref. [134]. To this end, a database of 5261 experiments carried out during 1987–1999 and their respective results was compiled by participants from Belarus, the Russian Federation and Ukraine [134]. The main evaluation criterion was the efficiency of experimental treatments in reducing radionuclide concentrations in final products as compared with untreated controls. It is important to note, however, that the majority of countermeasures do not intend to influence soil or groundwater concentrations, but aim to break exposure pathways.

Countermeasures can be based on a selection of crops that exhibit smaller radionuclide uptake than crops used previously, on food processing to reduce radionuclide contents or on choosing non-food crops, resulting in either case in a produce from the contaminated land that is radiologically acceptable [25, 121, 135]. Impacts on the dose received by people and on the ecology and economy of the affected area may vary enormously: a change in crop variety will have a much smaller impact than more radical changes such as the substitution of vegetables by cereals or changing from arable land use to animal husbandry.

Assessments have shown that substituting crops and fertilization are the most effective countermeasures in plant production. The efficiency of countermeasures, expressed by the reduction factor of radionuclide concentration in final products, was found to be of the order of 3 to 9, depending on the soil and individual crops [134]. Substituting crops may not be expensive, but its viability depends on a variety of economic conditions; for example, on contaminated sandy soils in Belarus it would be possible to grow rye that has an acceptable concentration of ^{137}Cs, but market demand for rye is poor and recently has been tending to zero. Another example are the impediments for the utilization of sugar beet: there is no sugar processing industry in the contaminated area in Belarus and no agricultural infrastructure, such as suitable farm machinery, for cultivating the plants.

Fertilizer application will suppress the uptake of certain radionuclides, mainly due to competitive effects. Thus potassium dosages will generally decrease the soil to plant transfer of ^{137}Cs, certainly when the soil is low in potassium. Reported reduction factors have varied between studies, but overall reduction factors ranging between 1.1 and 5.0 have been obtained [25, 134, 136–138]. The efficiency of potassium additions strongly depends on the exchangeable potassium content in the soil. For soils with a low to optimal potassium content, high dosages of potassium fertilizer are very effective and profitable. For soils with a high potassium content, only moderate dosages of potassium fertilizer are recommended to replace the potassium removed with crop yields [139, 140].

The behaviour of ^{90}Sr and its uptake by plants are controlled by its similarity to calcium. Many investigators have found a significant correlation between strontium TFs and the reciprocal of the exchangeable calcium content [141]. Consequently, much of the research and actions to reduce strontium uptake by plants has centred on the use of lime as a soil based countermeasure. The use of lime has reduced strontium uptake by up to 40%, the use of limed compost by up to 60% [141]. Generally, the reduction factor of radionuclide uptake by agricultural crops varies widely, from 1.1 to 3, depending strongly on the initial soil pH. The liming effect is most pronounced for acid soils [27].

Countermeasures that aim to provide the optimum (from the plant production point of view) rates of fertilizer application are the most viable, since the investment on fertilizer is paid back in the form of additional crop yields, and frequently profits are made. It has to be noted that the addition of nitrogen fertilizer should be moderate, as high dosages appear to stimulate the accumulation of ^{137}Cs and ^{90}Sr in plants [142–145]. Phosphorus dosages should be in accordance with crop responses and the phosphorus content of the treated soil, and should be crop specific.

The humus content of a soil is important because of its tendency to form co-ordinate bonds with calcium and strontium, which are stronger than the binding by ion exchange sites on soil minerals. Organic matter addition has resulted in strontium TF reductions by a factor of 1.2 to 7. The latter value was obtained after the addition of 15% organic matter to a sandy soil [141]. Field experiments on a podzoluvisol (loamy sand) soil in Belarus that increased humus content from 1.5% to 3.5% resulted in a reduction of ^{137}Cs and ^{90}Sr activity in perennial grass by a factor of 2 [142, 144].

Chemical amendments, such as zeolites, ammonium hexacyanoferrate (AFCF) or clay minerals, also reduce radionuclide uptake by plants, since the radionuclides are trapped and so rendered less available for plants [146–149]. A reduction factor of 4.6 in ^{90}Sr transfer has been obtained for a sandy soil after the addition of 1% zeolites [149], and a factor of 25 by applying 10 g AFCF per square metre [146]. However, the investigation of zeolites and clay amendments in field trials on a loamy sand soil in Belarus resulted in only a rather low reduction of activity in cereals [150]. Taking into account the high cost of transport and application of amendments at reasonable rates, for example 15 t/ha, this type of countermeasure is generally not affordable in conventional agriculture.

A more radical improvement of private hay land and meadows in all Chernobyl affected rural settlements of Belarus, the Russian Federation and Ukraine (where the total annual dose is above 1 mSv or the activity per unit area exceeds 555 kBq/m^2) is recommended. This countermeasure combines the liming of acid soils, fertilization (including the basic application of organic fertilizers), destruction of old turf, sowing of new grass stand and regulation of soil water (drainage), if needed; for example, radical meadow improvement has resulted in a reduction of grass activity by a factor of 1.7 to 3.5 [151], but other applications have achieved reduction factors for ^{137}Cs of up to 16–20 [134]. The reduction factor of surface meadow improvement is lower, and is 3.5 on average.

Although strictly speaking not remediation techniques, certain livestock management measures are effective in reducing public exposures. Such measures include feeding complexants, such as Prussian blue, to dairy animals to prevent ^{137}Cs transfer into the milk, or changes in pasture or fodder at critical times to reduce uptake. Achieved reduction factors vary widely between 2 and 15 [134].

Similarly, use and access restrictions for forests are more emergency responses than remediation measures.

Food processing can significantly reduce radionuclide concentrations in products. The efficiency depends on the type of processing and varies widely,

TABLE 3. EFFECTS OF VARIOUS COUNTERMEASURES ON RAPE RADIOACTIVITY [126]

	Activity reduction in rape products	
	^{137}Cs	^{90}Sr
Liming to 6 t/ha	14%	42%
Application of $N_{90}P_{90}K_{180}$ fertilizer	42%	27%
Liming to 6 t/ha + $N_{90}P_{90}K_{180}$ fertilizer	45%	59%
Variety selection	2.5 times	3.0 times
Rapeseed oil processing (crude oil)	250 times	600 times

removing 50–98% of the ^{137}Cs or ^{90}Sr during the production of butter or casein from milk [134].

The relative efficiency of different agricultural countermeasures can be seen from the experiments in which rape was grown on radioactively contaminated land in Belarus (Table 3). The effect of liming is mainly due to a rise of the soil pH and hence the increased availability of exchangeable calcium. Choosing a rape variety with less uptake offers activity reductions of up to three times. The most efficient removal of activity is offered by oil processing, resulting in a reduction of up to 600 times. Concentrations of radionuclides after a three stage filtration and bleaching are below the limits of detection. The combination of oil seed processing with several agricultural countermeasures therefore allows the production of food grade oil practically free from radionuclides and produces a valuable protein by-product (cake as animal fodder) with permissible concentrations of radionuclides [126].

5. CONTAINMENT

5.1. ENHANCED ATTENUATION

Although contaminated media sometimes provide sufficient attenuation capacity, normally these attenuation mechanisms must be enhanced through technical measures. Enhancement of attenuation may be targeted at particular exposure pathways; for example, plant uptake may be minimized or blocked to prevent contaminants from entering the food chain [27, 152].

Simple ploughing or deep soil mixing is not an efficient means of reducing direct surface gamma exposure [153], as such an approach will result in a dispersal of radionuclides over a larger area, thereby increasing the volume of contaminated soil.

Studies subsequent to the Chernobyl accident found that deep ploughing with digging, combined with liming and potassium fertilizer application, can decrease caesium and strontium transfer from soil to plants by a factor of 3 to 4 [23, 150]. The objective of deep ploughing is to skim off the upper 0–5 cm contaminated soil layer and burrow it beneath the turned over arable layer (30–50 cm), thereby preserving most of the soil fertility. Subsequent cultivation practices have to be limited to shallower depths to prevent the contaminated soil layer from being dug up or roots from reaching this layer. This quite cost effective countermeasure had only limited application after the Chernobyl accident because of the thin humus horizon of the predominantly light textured soils in the region [143, 150].

Changing the pH and redox conditions in contaminated zones can enhance attenuation [119, 120, 154–157], particularly in situations in which treatment is otherwise difficult, such as the presence of fractured rock [152]. Oxidation or reduction can be achieved by injecting aqueous solutions of appropriate agents, or by bubbling gases through the contaminated zone [156, 158]. Long term sustainability is uncertain, and competing geochemical processes need to be evaluated carefully [152, 159]. Environments with relatively low redox potential and high organic matter content (e.g. wetlands) tend to trap metals naturally [120], a property that can be utilized (Section 5.7).

The number of sorption sites may be increased by adding clay or zeolites [160] to soils. The addition of reactive minerals, such as lime, apatite [160–162] and its derivatives, such as bone meal [163, 164], may lead to immobilization through the formation of sparingly soluble mineral phases incorporating the contaminating radionuclides.

5.2. LOW PERMEABILITY BARRIERS

Low permeability barriers, which include slurry walls, sealable-joint sheet piles and curtains created by injection grouting [165–167], interrupt water flowpaths and may delay the migration of radionuclides. The construction of soil–bentonite slurry walls and cement–bentonite slurry walls to serve as low permeability barriers is a well established technique, has been demonstrated in civil construction and has been transferred to provide containment at contaminated sites [168]. The efficacy of low permeability barriers, however, depends critically on strict quality control during construction. Such low permeability

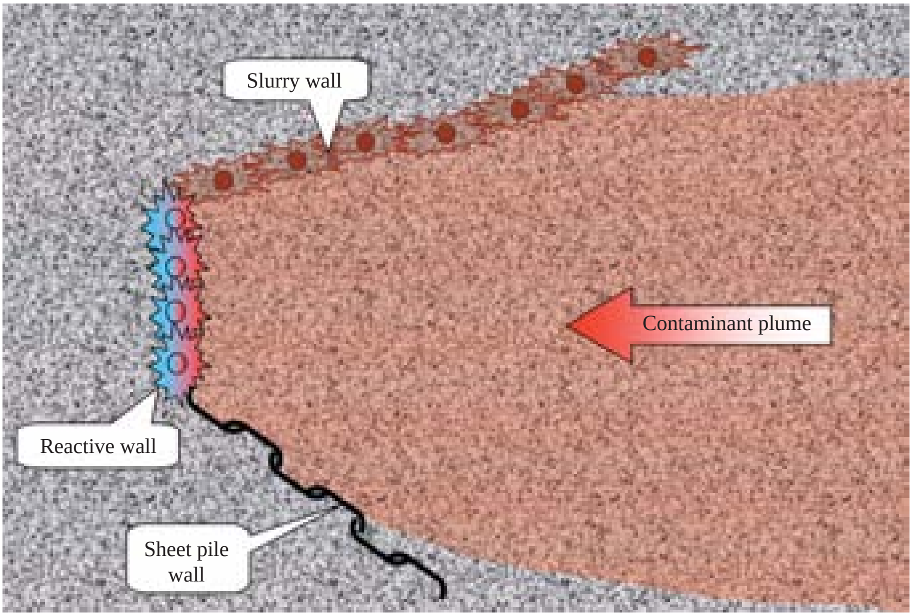

FIG. 6. Sketch of a permeable reactive wall in combination with a Funnel-and-Gate system.

barriers may be used in isolation or as a component of a suite of techniques (Fig. 6).

5.3. PERMEABLE REACTIVE BARRIERS

The use of permeable reactive barriers or walls is distinguished from outright containment by the fact that the contaminant carrier as such (i.e. the groundwater) is not prevented from movement [169–172]. The objective is rather to remove the contaminants from the mobile phase. Permeable reactive barriers are installed by excavating a portion of the aquifer, disposing of the excavated material and replacing it with a permeable material designed to react with the contaminant and remove it from the flowing water (Fig. 6). The advantages over pump and treat systems are that no active pumping or process operation and maintenance is required, thus reducing energy and operation and maintenance costs, no treatment sludges are produced, thus reducing waste disposal costs, and no surface facility is required, which allows the land to be returned to productive use. The systems typically rely on the natural gradient of the groundwater table as the driving force. The barrier material must be designed to remain reactive for periods of many years to decades. Furthermore, the barrier permeability must be sustained throughout the duration of the groundwater treatment. The performance of permeable reactive barrier systems must therefore be monitored so that corrective action can be taken when required.

Permeable reactive barriers have been designed and implemented for the treatment of dissolved metals [169, 173, 174], acid mine drainage [175–177], radionuclides [178–181] and dissolved nutrients [182–184]. Contaminant removal can be effected in a variety of ways [185–187]. Treatment processes include adsorption [188, 189], simple precipitation [190], adsorptive precipitation [183], reductive precipitation [169, 176, 179] and biologically mediated transformations [175, 176, 182].

Changing the redox state can be a very effective method of immobilizing certain radionuclides (e.g. uranium and technetium). These radionuclides have two or more oxidation states, and the more reduced oxidation states are less mobile; for example, reduction of the hexavalent uranyl ion UO_2^{2+} to the tetravalent U(IV) state results in the precipitation of sparingly soluble precipitates, including $UO_{2(s)}$ or mixed U(VI)–U(IV). Zero valent iron is an abundant and inexpensive reducing agent that has been observed to reduce and precipitate uranium and technetium in laboratory studies [178, 179, 191–193]. The oxidation products generated (e.g. ferric hydroxides) can provide a high capacity sorption substrate also for non-redox sensitive species [194], but their long term stability in relation to changes in redox conditions has to be carefully evaluated [195].

Permeable reactive barrier systems containing zero valent iron have been installed for the treatment of uranium, technetium and other metals [179, 180, 196, 197]; these barriers demonstrate excellent removal of uranium and technetium. Examination of the reaction products has been conducted at a series of sites of permeable reactive barriers [179, 198, 199]. Although the results of these characterization studies are inconsistent, all the reports indicate that a portion of the uranium entering the barrier system is reduced to U(IV), whereas some portion may remain in the U(VI) oxidation state. Other metals commonly associated with uranium mine waste, including arsenic, molybdenum, selenium, vanadium and zinc, are also removed from the groundwater, possibly as reduced phases (e.g. V_2O_3) or as sulphide minerals (As_2S_3, ZnS) [180, 200].

Organic reductants, such as sawdust, have also been used to promote the reduction and precipitation of uranium. Passive treatment systems containing organic carbon have been used to remove both uranium and nitrate from groundwater at sites where these two constituents coexist as a result of releases from nuclear weapon production facilities [201].

Sorption can remove contaminants from groundwater and can maintain low concentrations of radionuclides. Sorptive materials that have been evaluated or deployed in permeable reactive barrier systems for treating radionuclides include zeolites (e.g. clinoptilolite [202]), phosphate based adsorbents (e.g. bone char apatite [196] and Apatite II [203]) and hydrous ferric oxides (e.g. amorphous ferric oxyhydroxide (AFO)) [188, 196].

The majority of the reactive barriers installed to date have been continuous barriers installed across the entire width of the plume. Contaminant fluxes also can be focused on the reactive barrier by an array of non-reactive barriers, such as slit or slurry walls [166], to form a Funnel-and-Gate system [204, 205]. Funnel-and-Gate systems reduce the physical length of the treatment portion of the barrier and prevent contaminants from circumflowing the treatment zone. The volume of reactive material required to treat contaminated groundwater is determined by the contaminant concentrations, groundwater geochemistry and flow rate. For many contaminant plumes, the volume of reactive material will be similar, whether a continuous barrier or Funnel-and-Gate configuration is employed. Since the installation of continuous barriers is typically less expensive than that of Funnel-and-Gate systems, this installation technique has been preferred. Furthermore, because Funnel-and-Gate installations focus the flow to across a small cross-sectional area, there is greater potential for clogging by the formation of secondary precipitates.

Depending on the reactive material to be used, deployment techniques may include injection wells (for grouts, gels and soluble reactants) or trenches (see Fig. 7) cut by a suitable excavator (for grouts and particulate material such as granular iron, sawdust, etc.). Development work on efficient methods to emplace reactive barriers with minimal disturbance, even in awkward places, is ongoing. Adaptation of more novel civil engineering techniques, such as directional or horizontal drilling, the use of guar gum slurries for barrier installation [177], hydraulic fracturing [206] and jet grouting techniques [207], can be used for the emplacement of barriers at depths beyond the capabilities of conventional excavation techniques (Fig. 8).

In December 1998 a reactive barrier incorporating a novel wall and curtain design was installed at the Atomic Energy of Canada Limited (AECL) research facility in Chalk River, Ontario, Canada, about 200 km west of Ottawa [202]. The wall and curtain design directs contaminated groundwater to the treatment media, and excludes the overlying uncontaminated groundwater from the treatment zone (Fig. 9). The treatment zone in the wall and curtain barrier contains clinoptilolite, a naturally occurring zeolite mineral that removes ^{90}Sr from the plume via a sorption reaction. Preliminary monitoring of

FIG. 7. (a) Continuous trenching machine used to install the 46 m long, 7.3 m deep and 0.6 m wide granular iron permeable reactive barrier; (b) simultaneous excavation and replacement of aquifer material with granular iron as the horizontal trencher advances [193].

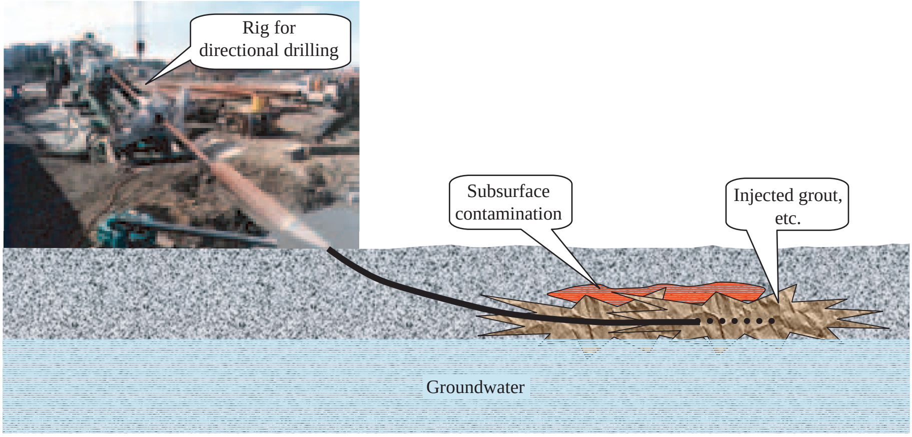

FIG. 8. The principle of directional drilling and grouting.

the barrier performance has suggested excellent removal of ^{90}Sr, from initial concentrations of 0.1–100 Bq/L to below the Canadian drinking water limits [189]. The ongoing monitoring programme has shown that the barrier has treated more than 1.85×10^7 Bq of ^{90}Sr.

A three component Funnel-and-Gate reactive barrier system was installed at a former uranium upgrading site at Fry Canyon, Utah, USA, in

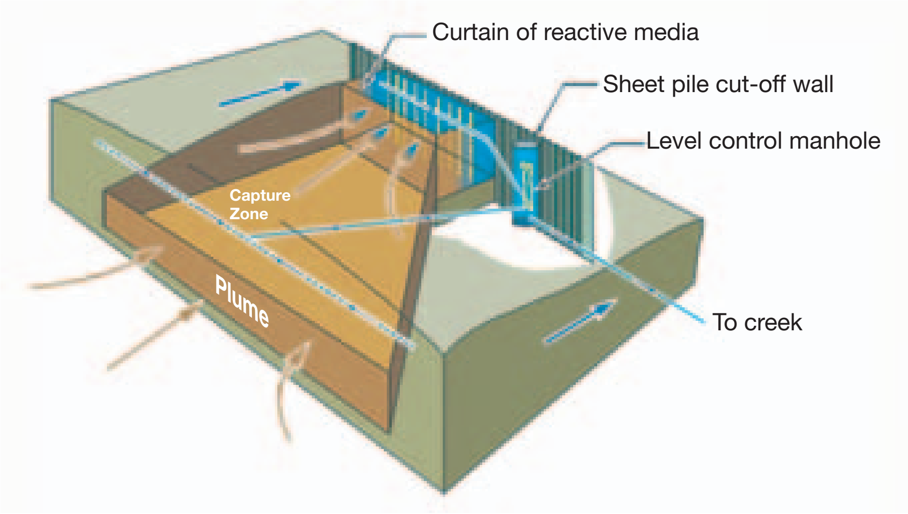

FIG. 9. Trench and gate system [202].

1997 [196]. One of the permeable reactive barrier components contains AFO, and a second contains bone char apatite. Both of these components are intended to assess the potential for uranium treatment by sorptive processes. The third component contains zero valent iron, intended to reduce and precipitate uranium as a reduced uranium oxide. The performance of the barriers has been monitored over three years and 1650 pore volumes. The zero valent iron component demonstrated the greatest removal efficiency (C/C_0 = 0.007), followed by the AFO system (C/C_0 = 0.271) and the bone char system (C/C_0 = 0.412).

Computer simulations conducted using reactive solute transport models can be used to determine design parameters for barrier installation, to predict the potential for barrier clogging and to assess the potential benefits of barrier performance. The performance of a reactive barrier installed at the Elizabeth City US Coast Guard Support Centre [208] was simulated using the reactive solute transport model MIN3P [209]. Comparison of the simulation results with subsequent measurements showed good agreement (Fig. 10). The performance of the permeable reactive barrier installed at Monticello Canyon, Utah, USA, was simulated using the PHREEQC model [180].

The limitations on permeable reactive barrier performance and lifespan include constraints on the reactive material longevity and the barrier permeability. Of these concerns, the potential for barrier clogging and the permeable reactive barrier evolving into an impermeable reactive barrier is the most significant. Since the total mass of contaminant that accumulates in the barrier is modest, the principal precipitates resulting in clogging are the products of reactions between the barrier material and the major ions present in the water, or between the barrier material and the water itself. The use of zero valent iron (Fe^0), the most commonly used reactive material, results in the reduction of water and an increase in the pH to between pH10 and pH11. This increase in pH favours the precipitation of carbonate minerals, principally calcite ($CaCO_3$) and siderite ($FeCO_3$). Over periods of several years to decades, the accumulation of these precipitates potentially may be sufficient to reduce the pore space of the reactive material and limit barrier permeability. Reactive barrier technology has evolved recently, and the oldest barriers are now approaching ten years of operation. Clogging to a degree that is sufficient to impair barrier performance has yet to be observed, although long term monitoring programmes are required to assess this concern.

The long term fate of the reactive barrier after remediation is complete or after the barrier becomes ineffective depends on the nature of the contaminant and on the characteristics of the barrier. Concerns include the potential for remobilization of contaminants retained in the barrier and the potential for clogging in the barrier to alter natural groundwater flow conditions. In many

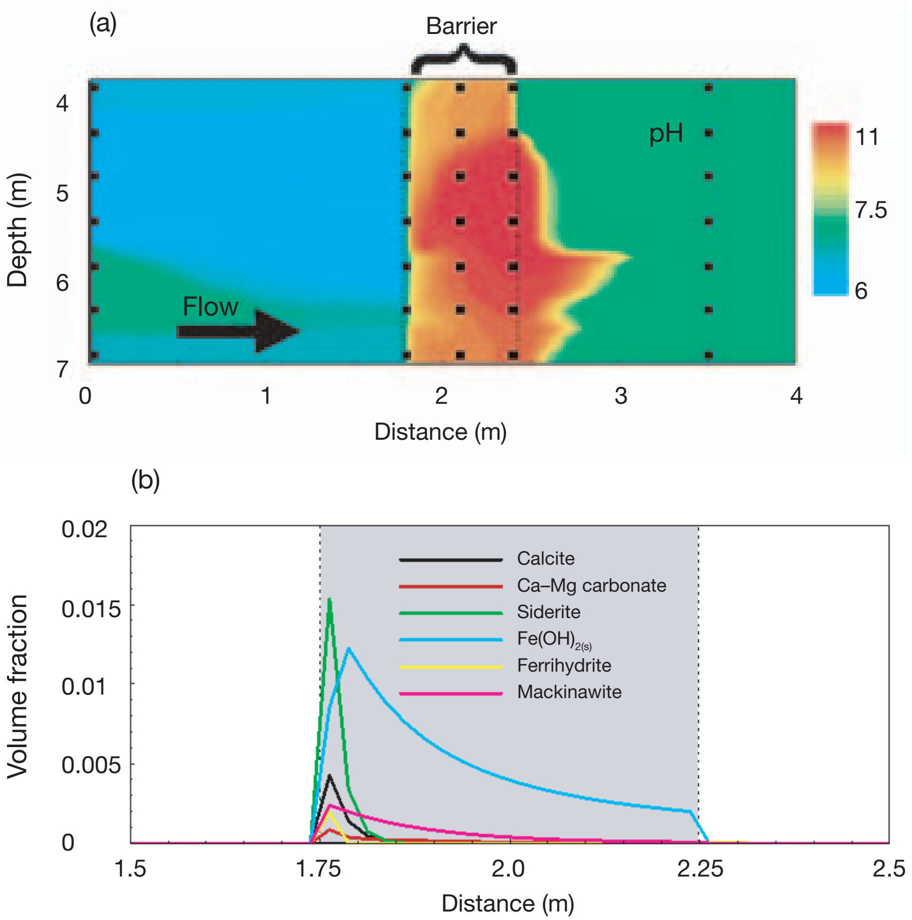

FIG. 10. Simulated heterogeneous reactions at a permeable reactive barrier at the Elizabeth City US Coast Guard Support Centre [209].

barrier systems, the contaminant is converted to a form that is stable in the geochemical environment that prevails in the aquifer. Furthermore, because the mass of contaminant is small relative to the mass of the barrier material, the residual barrier material may be classified as non-hazardous. In these systems, it may be acceptable for the barrier to remain in place. In other cases, the mass of contaminant may exceed soil guidelines, the contaminant may have the potential for remobilization or the contaminant may be sufficiently hazardous to warrant excavation of the reactive material and placement in a secure waste disposal facility. In these cases, excavation of the barrier, or a portion of the barrier, may be required.

Although considerable research on the performance of reactive walls is continuing worldwide, some techniques have reached commercial maturity [210, 211].

5.4. IMMOBILIZATION

Immobilization, unlike physical containment, is intended to affect the contaminated material itself. The objective of immobilization is to change the contaminant form into one that is less susceptible to migration. Two basic options can be distinguished: in situ and ex situ treatment [212, 213].

In situ treatment effects immobilization without the contaminated material being removed. Three major methods to effect in situ immobilization can be distinguished, based on chemical, biochemical or thermal treatments.

Chemical immobilization is based on the injection of a variety of grouts [214] or on changing pH and/or redox conditions in the groundwater, for example [120, 154, 155, 215]. These grouts can be based inter alia on ordinary Portland cement (OPC), water glass (sodium silicate), gypsum or organic polymers, for example acrylic or epoxy resins. The suitability of immobilizing agents via injection depends largely on the hydraulic properties of the contaminated material. OPC and epoxy resins typically have a high viscosity, while water glass and gypsum solutions, or acrylic acid suspensions, can be made up with viscosities equal to that of water. The long term stability of the polymer stabilized material has to be carefully assessed. Breakdown products containing functional groups, such as carboxylic or phenolic groups, may actually act as a vehicle to facilitate transport of radionuclides.

Injection of chemical reductants, including calcium polysulphide, has been used to promote contaminant reduction and precipitation within aquifers. Contaminants that are well suited to remediation using this approach include metals with a lower solubility under reduced conditions. Injection techniques have been used to treat Cr(VI), through reduction to Cr(III) and precipitation of Cr(III) hydroxides. In situ redox manipulation (ISRM) [155, 215] is a variation on a chemical injection system (Fig. 11). When using ISRM, a strong reductant is pumped into the aquifer, converting oxidized Fe(III) bearing minerals to Fe(II) bearing minerals. These reduced phases remain stationary, and react with oxidized dissolved contaminants that migrate through the treated zone in the aquifer [215]. This approach has been demonstrated on a pilot scale to treat groundwater contaminated by Cr(VI) at the Hanford Site in southeastern Washington State, USA [215].

Biochemical or biological methods are based on the introduction or stimulation of microorganisms that change the chemical environment [216].

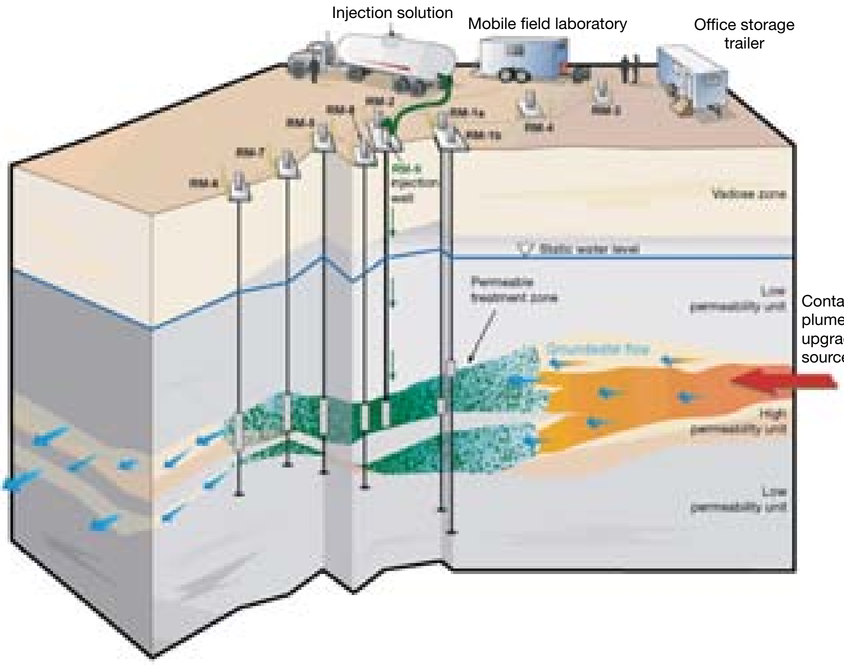

FIG. 11. *In situ redox manipulation (courtesy of Batelle).*

Depending on the circumstances and intentions, a (enzymatic) reductive or oxidative precipitation of radionuclides can be effected. The application would be similar to creating a biowall, as discussed in Section 5.5.

Thermal treatments use heat processes to immobilize the contaminant. Thermal treatment, however, generally is not economically efficient for dispersed radioactive contamination [5, 15].

Ex situ treatments are carried out in some sort of plant, either on or off the site. After treatment, the material is either returned or disposed of in an engineered repository. A number of treatment techniques can be used for both in situ and ex situ treatments, the method of application varying in each case. Ex situ methods are discussed in Section 4.3.

Organic polymers and water glass are also used to immobilize surface contamination. The main effect is to enhance the cohesive properties of topsoils, thus preventing wind and water erosion [217] (Fig. 12). Depending on the formulation, infiltration of rainwater may also be impeded and thus the downward migration of radionuclides retarded.

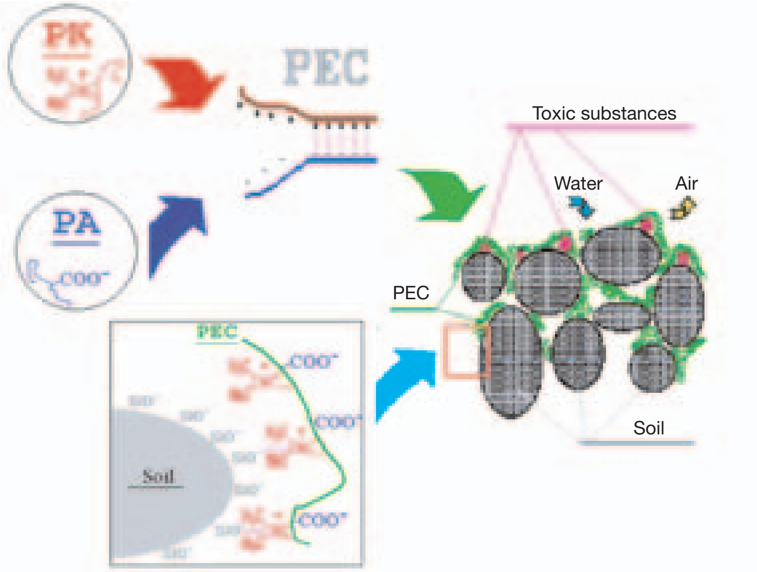

FIG. 12. Binding of soil particles and entrapment of contaminants using organic polymers (S. Mikheykin, VNIICHT, Moscow, personal communication).

Over the years consultants and contractors have developed a wide range of proprietary engineering applications based on the fundamental processes outlined above (e.g. Refs [218, 219]).

5.5. BIOLOGICAL BARRIER WALLS (BIOWALLS)

A biowall is an in situ barrier that relies on biological processes to restrict the migration of radionuclides (Fig. 13). The application of the technology is most appropriate to geological formations with significant permeability (e.g. sands, sandstone and permeable limestones) and no preferential flowpaths such as open cracks and fissures. A biowall can be emplaced downstream from the contaminated site or constructed in situ via the formation of biofilms and biocolloids [220–226]. The development of a biowall requires the introduction of suitable microorganisms and the provision of nutrients and essential elements to further their propagation. Adjustments to the pH or redox potential may also be required to initiate bacterial growth.

The effectiveness of biowalls results from:

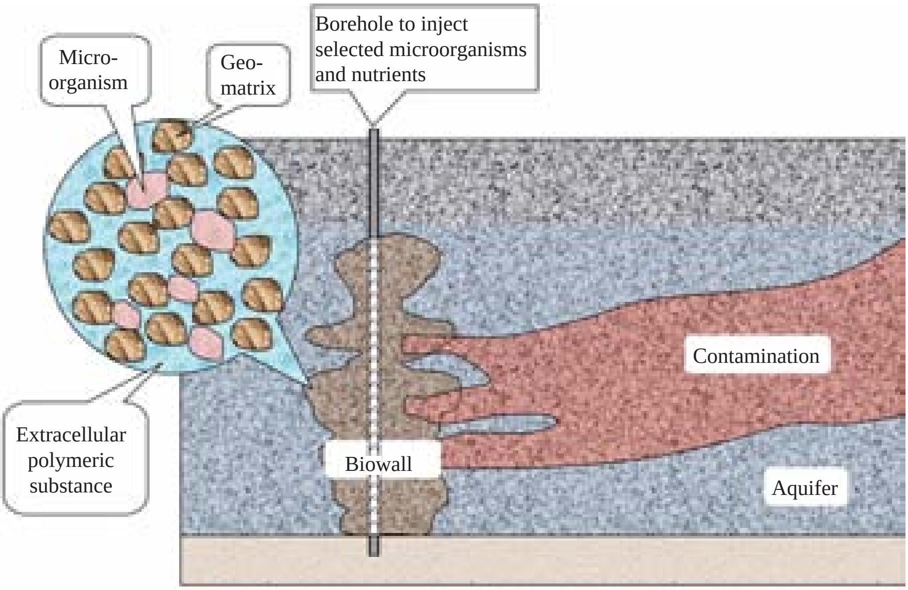

FIG. 13. The principle of a biowall.

(a) The physical reduction of permeability and hence groundwater flow by the microbial population. This effect can be enhanced by the use of ultramicro cells (less than 100 nm). In the course of growth by metabolism they increase in size and may completely fill the pore space.

(b) The generation of metabolites capable of restricting the migration of radionuclides through the barrier wall. Such metabolites are mainly extracellular polymeric substances (EPSs), commonly termed ‘slimes’, which the microbial cells use for attaching themselves to the substrate. These EPSs also fill pore spaces and thus reduce permeability.

(c) The sequestering of radionuclides from groundwaters by complexation [227–229], although it should be noted that subsequent mobilization of these colloidal species could constitute an additional transport mechanism [230]. Microbial action can also be utilized to modify groundwater chemistry (e.g. sulphate reducing bacteria) to immobilize redox sensitive species [231] such as uranium [232, 233] or to prevent the formation of acid drainage [234]. The latter two methods would act in a similar way to an inorganic reactive wall. Several microbial strains are commercially available.

5.6. PHYTOSTABILIZATION

Phytostabilization, the development of a stable and permanent vegetation cover, reduces the risk of erosion of contaminated soil from sparsely or non-vegetated land [235], thus reducing waterborne and dustborne exposure pathways. In addition to preventing erosion, this technique may change the mobility of potentially toxic elements by reducing concentrations in the soil water and on freely exchangeable sites within the soil matrix. Both processes alter the speciation of soil metals, reducing the potential environmental impact. The technologies draw upon fundamental plant and soil chemical processes as well as established agricultural practices. The development of a stable and self-perpetuating ecosystem as a result of this type of treatment may have additional benefits, as in some circumstances plant root activity may also influence metal speciation by changes in redox potential or the secretion of protons and chelating agents. The microflora associated with root systems may also be involved in these processes. The rainwater infiltration rate is reduced by plant induced evapotranspiration, thus reducing the potential for leaching and acid drainage generation [236–238]. Some examples of phytostabilization approaches are presented below.

Soil stabilization is very important on certain types of arable land to prevent horizontal radionuclide migration due to water and wind erosion (see Fig. 14). The ^{137}Cs activity in topsoil in valleys may be increased by 30–80% as

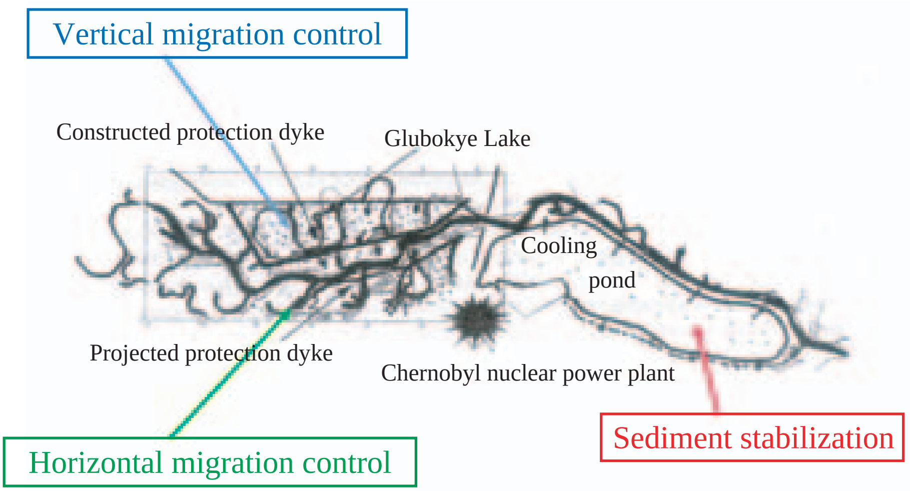

FIG. 14. Phytostabilization approaches at the contaminated area of the Dnieper close to the Chernobyl nuclear power plant.

a result of run-off of fine soil particles, as shown in field experiments in Belarus [142, 150]. Crop rotation with perennial grasses covering up to 50–80% of the cultivated area and avoiding row crops reduces contaminated topsoil loss from 10–20 t/ha to 2–3 t/ha. On slopes, deep soil tillage without overturning the arable layer is needed. Conventional ploughing with overturning of the arable horizon should only be carried out to destroy and plough in old turf [239]. Good cultivation practices, such as ploughing parallel to the slope, rather than up and down, will also reduce erosion.

Wind erosion may occur on sandy soils and on drained peaty soils. It is recommended to eliminate root crops on soils for which soil loss amounts to 8–15 t/ha or more [143, 240]. The major area (50–80%) of crop rotation should be under perennial grasses. A smaller area can be allocated to winter and spring cereals and to annual grasses. In any case, soils should be under vegetation cover throughout the year, preferentially under perennial grasses. In such a manner soil loss due to wind erosion can be reduced to 2 t/ha [239].

Remedial actions are still being investigated to control the radionuclide efflux from the Chernobyl exclusion zone within the Dnieper catchment system. This and adjacent drainage basins form a wide area from which contaminated waters flow and sediments are transported downstream through the Pripyat and Dnieper Rivers across Ukraine and to the Black Sea. Phytostabilization techniques could in this context also be considered as remedial options. Three phytorehabilitation approaches involving willow plantations have been studied [127] (Fig. 14): (1) the effect of willow plantations on vertical migration of radionuclides; (2) the effect on the stabilization of the Chernobyl cooling pond sediments; and (3) for lateral erosion control.

The area of interest for studying the vertical migration control by willows was an extremely contaminated zone of 16 km^2 on the left bank of the Pripyat River (between 3.7 and 18.5 TBq/km^2 ^{90}Sr and ^{137}Cs and 0.37 TBq/km^2 Pu), which is partly protected from spring floods by a dam. Through modelling exercises it was shown that, due to their high evapotranspiration rate, willow SRC stands are expected to lower the groundwater table level by 100–200 cm in fertilized stands. Without fertilization, a lowering of the groundwater table level of less than 50 cm was predicted. Since the immobilization potential of ^{137}Cs and ^{90}Sr in the willow wood is limited, the influence of plant uptake on migration remains low.

Following the closure of the Chernobyl nuclear power plant, the water level of the cooling pond (22.5 km^2; depths between 1.5 and 15 m; with about 111 TBq ^{137}Cs and 37 TBq ^{90}Sr) will drop by 4–7 m, and 15 km^2 of the sediments will become exposed and may be in need of stabilization. To this end the SALIMAT option was investigated [241]. SALIMATs consist of a roll of willow rods (stems) rolled around central disposable tubes that are unwound by dragging them across the lagoon. Small scale tests have demonstrated that SALIMATs establish well on contaminated pond sediments and produce a full vegetation cover during the second year. The approximated cost of the phytostabilization option ranges between €0.8 million and €1.9 million for the reclamation of 15 km^2 of sediments, which is low compared with the prospected cost of removal of the sediments ($6 million, transport and disposal costs not included) or maintenance of the present water level ($200 000 per year).

The project area for horizontal erosion control was on the right bank of the Pripyat River, which was significantly less contaminated than the left bank and is not protected by a dam. After inundation, part of the activity is eroded and transported to the Pripyat River with the withdrawing water. It was calculated that even in the event of extremely high flooding, a dense willow plantation will effectively decrease horizontal soil erosion and the concomitant transport of radionuclides into the Dnieper River system.

Vegetation or revegetation is a commonly employed measure for the capping of engineered waste disposal facilities and mining residues such as spoil heaps [242] or tailings ponds. The final step in closing out an impoundment for uranium mill tailings is the design and placement of a cover that will give long term stability and control to acceptable levels radon emanation, gamma radiation, erosion of the cover and tailings, and infiltration and precipitation into the tailings and heaps. Surface vegetation can be effective in protecting tailings or a tailings cover from water and wind erosion. Factors affecting the effectiveness of surface revegetation on impoundments can be broadly classed into climatological and agrobiological factors. Plants should be chosen to match the local climatic conditions. From an agrobiological perspective, the nature of the ore and the milling process will largely determine whether uncovered tailings are capable of sustaining growth. Considerable efforts to improve unfavourable properties such as low or high pH values and low plant nutrient content will usually be required before tailings can sustain growth. Depending on the substrate, revegetation requires preparation and amelioration of the topsoil, including removal, for example, of acid generating minerals [243, 244]. Techniques and strategies to overcome such difficulties have been developed [245], for example hydroseeding or the use of compost from organic household refuse [246]. The method may be limited to low contaminant concentrations, owing to the (root) toxicity of higher concentrations. An adequate soil cover may need to be established.

Water and wind erosion are the primary concern at unvegetated tailings or waste rock piles. While a vegetation cover will decrease erosion, it may raise concerns that it promotes radon emanation by drying out the tailings and tailings covers. Tree roots may penetrate the contaminated material and disturb the integrity of engineered covers. Conversely, the increased evapotranspiration following vegetation establishment alters the water balance of a tailings or waste rock facility and will decrease seepage. The effect of vegetation due to the effect of plant roots on the physicochemical characteristics of tailing material has not been studied very intensively so far.

At a 35 year old reclaimed site on a uranium mining dump near Schlema, Germany, it was concluded that vegetation cover could reduce infiltration by 40–60% due to interception by the canopy (25–40%) and increased transpiration [247]. It was further found that of the 165 000 g/ha of uranium in the soil (30 cm depth), only 4 g/ha was in the above ground plant parts and 510 g/ha in the below ground plant parts. Since most (90%) of the uranium taken up during the growing season is recycled (returned to the soil) with pine needles, uranium dispersion by uptake through vegetation is minimal. It may be concluded from these preliminary results that forest vegetation may reduce the infiltration rate and will disfavour radionuclide dispersion.

The proper design of tailings covers is crucial to ensure their long term stability with respect to plant intrusion [248]. Since plant roots can penetrate compacted sealing layers (trees can have roots reaching down 3–4 m) and since trees need to have a certain degree of mechanical support in order to minimize the probability of uprooting, a vegetation substrate depth of at least 1.5 m is required. The vegetation substrate layer must be such that the critical suction is not exceeded at the top of the clay seal. It must be thick enough for plants to find sufficient water and nutrients so as to prevent the generation of a high suction at the seal. Cracks resulting from such suctions become accessible to roots and can be widened as further water is extracted.

In addition to the mechanical effects of soil stabilization and water management, revegetation has aesthetic benefits and sometimes cultural connotations, in particular on native or aboriginal lands [249]. The choice of vegetation cover may also effect some sort of institutional control; for example, converting contaminated agricultural land into forestry reserves interrupts a potential exposure pathway via the food chain. It has to be ascertained, however, that no other exposure pathway is opened up, for instance via burning contaminated firewood.

5.7. CONSTRUCTED WETLANDS

Constructed wetlands are engineered, human-made ecosystems specifically designed to treat wastewater, mine drainage and other waters by optimizing the biological, physical and chemical processes that occur in natural wetland systems (Fig. 15). Constructed wetlands can provide an effective, economical and environmentally sound treatment of wastewater, and serve as

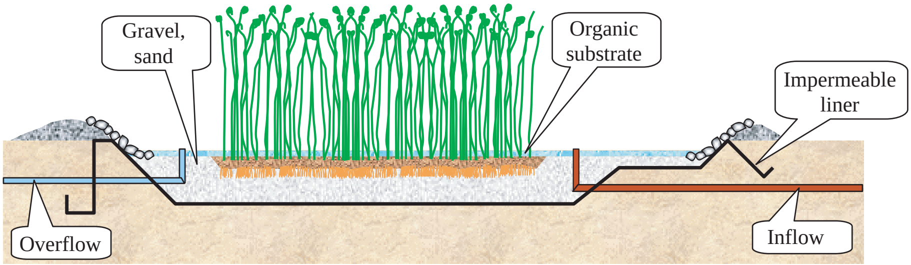

FIG. 15. Schematic cross-section of a constructed wetland.

wildlife habitats. The conceptual design leads to either the (permanent) fixation of the contaminants in situ or to plant uptake with the view to harvesting shoots later for further treatment and disposal. The latter option is discussed in more detail in Section 6.10, while the following largely focuses on in situ fixation.

The concept for such constructed wetlands was originally developed to treat domestic effluents for residual organic material, for example following mechanical and biological (activated sludge) treatment steps [250], and has found widespread application in particular for the treatment of (acid) mine effluents [251–273]. The Chernobyl accident also prompted research in this direction [274]. A critical appraisal of this application is given in Ref. [275], and the mechanisms acting are reviewed, for example, in Refs [276, 277]. The concept was further developed to recover metals from low grade ores or mine water by a process known as phytomining (see also Section 6.10).

Constructed wetland systems are grouped into three main types [278]: (1) free water surface systems; (2) subsurface flow systems; and (3) aquatic plant systems. Free water surface systems, or soil substrate systems, consist of aquatic plants rooted in a soil substrate within a constructed earthen basin that may or may not be lined, depending on the soil permeability and groundwater protection requirements [279] (Fig. 16). Free water surface systems are designed to accept preliminarily treated, low velocity wastewater, in plug flow, over the top of the soil media or at a depth of between 2 and 45 cm. Subsurface flow systems are typically gravel substrate systems that are similar to free water surface systems; however, aquatic vegetation is planted in gravel or crushed stone and wastewater flows approximately 15 cm below the surface of the media. The aggregate typically has a depth of between 30 and 60 cm. No visible surface flow is evident in subsurface flow systems [279]. Aquatic plant systems are also similar to free water surface systems, but the water is located in deeper ponds and floating aquatic plants or submerged plants are used [281]. Where available, natural ponds may be used. Where they exist, natural wetlands and

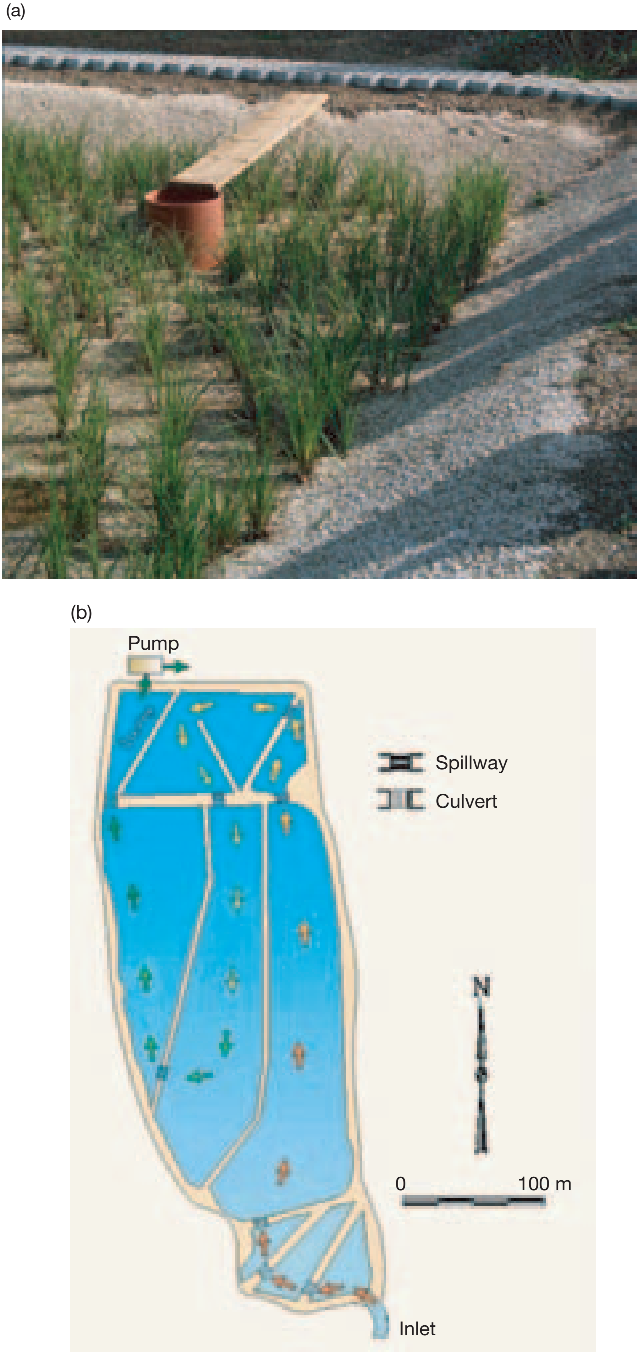

FIG. 16. (a) Image of a constructed wetland; (b) plan view of a constructed wetland [280].

bogs can be used as traps for radionuclides and other metals [282, 283], although this might be better classed as biosorption or natural attenuation, since it is mostly the decaying organic matter that effects retention. Studies on natural analogues for radionuclide migration have demonstrated this mechanism to be effective for thousands of years [114].

Early research revealed that phytoextraction via constructed wetlands (used to purify water) was ineffective because it was difficult to remove inorganic elements that precipitated from the water into the sediments. In addition, floating plant systems, with subsequent biomass harvesting, were determined to be inefficient and uneconomic [284].

After three years of operating a pilot constructed wetland to treat the mine water from the flooded Pöhla Tellerhäuser mine at Wismut, Germany, it was shown that the system removed iron, arsenic, manganese and radium. Removal processes were based on the geochemical characteristics of the contaminants. For manganese and ^{226}Ra, removal was also partially through biofilm formation. Uranium was not removed, given the high pH and the presence of high bicarbonate concentrations [285]. It is hence clear that process effectiveness in constructed wetlands depends on the speciation of the radionuclides concerned [286] and hence on the control of the governing parameters in the surface and pore waters, such as pH [287], and that waters may need to be subject to enhancement by additives [288] or pretreatment [289].

Wetlands may be constructed with the main objective of excluding atmospheric oxygen from material that would generate acid from the oxidation of pyrite and other sulphides [290]. This method, however, is likely to be effective only in regions where precipitation is higher than evapotranspiration (i.e. in temperate and humid tropical climates). Climatic conditions limit the general applicability of wetlands. Extended periods of deep frost as well as arid conditions are unfavourable. If, however, effluents only arise during frost free periods, it may be possible to operate wetlands in fairly high latitudes or altitudes.

Passive water treatment technologies such as constructed wetlands at abandoned mining sites may be appropriate for small contaminant loads. However, long term stability and resilience with respect to external disturbances and recovery are of major concern for both wetland operators and regulators [291]. Technical guidance for designing and operating constructed wetlands may be limited, owing to a lack of long term operational data. Potential seasonal variability and impact on wildlife may negatively affect system operation and securing permits, respectively [292]. Relatively large parcels of land are required and water consumption is high, owing to large evapotranspiration rates in some areas.

6. REMOVAL

6.1. PRECONDITIONS AND CONSTRAINTS

It must be noted that in general any method relying on the removal of contaminated soil is likely to require substitution of the removed material with clean (top)soil. Therefore, in addition to considerations with respect to technical feasibility, an economic source of clean soil will be required to make this option viable. Conversely, a precondition for any removal option is the availability of a suitable disposal site for the excavated materials, whether they are left untreated or whether they are conditioned before emplacement.

6.2. IMMOBILIZATION AND SOLIDIFICATION (EX SITU)

A wide variety of agents have been used, or proposed, for the solidification of excavated materials [212, 213, 293]. Often the objective is not only to immobilize the contaminants but to add value to the waste material by converting it into a useful product, for example for construction purposes. Use in general construction as a substitute for valuable raw materials requires special testing and licensing procedures to ensure environmental compatibility and compliance with quality criteria such as compressive strength, freeze–thaw cycle stability, leachability, etc. Solidified wastes may also be used in the construction of cappings, etc., for (hazardous waste) landfills. In cases where no further use is envisaged, minimization of the volume increase by the solidification agents is desirable to save valuable raw materials and repository space [213]. If only small volumes arise, the material may be combined with material from other waste streams requiring a similar immobilization treatment. Combining waste streams can make the process more economically viable, as products in marketable quantities are produced.

The treatment may be undertaken on or off the site at dedicated facilities. In the case of off-site treatment, the material has to comply with the applicable transport regulations and must meet the appropriate safety criteria while being handled. The additional risk from transporting material must be worked into the respective safety and cost–benefit analyses.

As for in situ immobilization, possible (cold bonding) solidification agents include OPC and other hydraulic binders such as certain fly ashes, gypsum, silicate gels (water glass) [294] and organic polymers, particularly acrylic and epoxy resins. The overall cost of treatment obviously depends on the amounts or volumes requiring treatment, and some binders are more economic in larger quantities than others. Organic polymers, in most cases, appear to be applicable only to smaller volumes of material. The conversion of gypsum ($CaSO_4 \cdot 2H_2O$) into a semi-hydrate (plaster of Paris, $CaSO_4 \cdot \frac{1}{2}H_2O$) requires heat treatment in a kiln. It is only the plaster, not the gypsum, that exhibits the curing properties required of binders.

The main conclusion of a recent report [295] on the European perspective of NORM waste treatment was that immobilization is not widely used or accepted as a treatment. Many companies regard this type of technology as less feasible for NORM waste material and hence have not pursued the development of immobilization techniques as a waste treatment process. However, for treating the radioactive remainder of a separation step, immobilization is widely seen as a treatment with a high potential.

Into this latter classification would also fall ground freezing as a temporary measure to prevent the dispersal of contaminants [296, 297]. Either an impermeable screen around a contamination can be established or the contaminated material itself can be frozen in order to facilitate its handling. In either case, it is unlikely that in the present context of low level dispersed contamination this method would find a field of application.

The removal of a contaminated topsoil layer is, of course, the most effective measure, but generates large quantities of waste and is only applicable to small areas of land [138]. Moreover, the most fertile layer of the soil is removed in the process. The overall efficiency of such a measure depends very much on the operating conditions and on the distribution of the contamination in the vicinity of a critical group [151]. In Belarus, the Russian Federation and Ukraine the removal of contaminated topsoil was recommended for all settlements where the ^{137}Cs activity exceeded 555 kBq/m^2 and for 25–33% of settlements where the ^{137}Cs activity was in the range of 370 to 555 kBq/m^2. It was estimated that it would incur costs of about €325 per inhabitant [151].

6.3. SURFACE AND GROUNDWATERS: PUMP AND TREAT

Pump and treat systems (Fig. 17) were the baseline for remediating groundwaters throughout the 1990s [10, 298–301]. The technology is based on the assumption that contaminant concentrations can be reduced or removed by employing ion exchange or sorption [302] and precipitation processes [303]. Some attempts have been made to use electrolysis [304] or (reverse) osmosis in pump and treat systems. Chemicals have also been added underground in an attempt to enhance recovery rates [305].

A National Academy of Sciences report [306] provides a comprehensive assessment of the effectiveness of pump and treat systems for the remediation

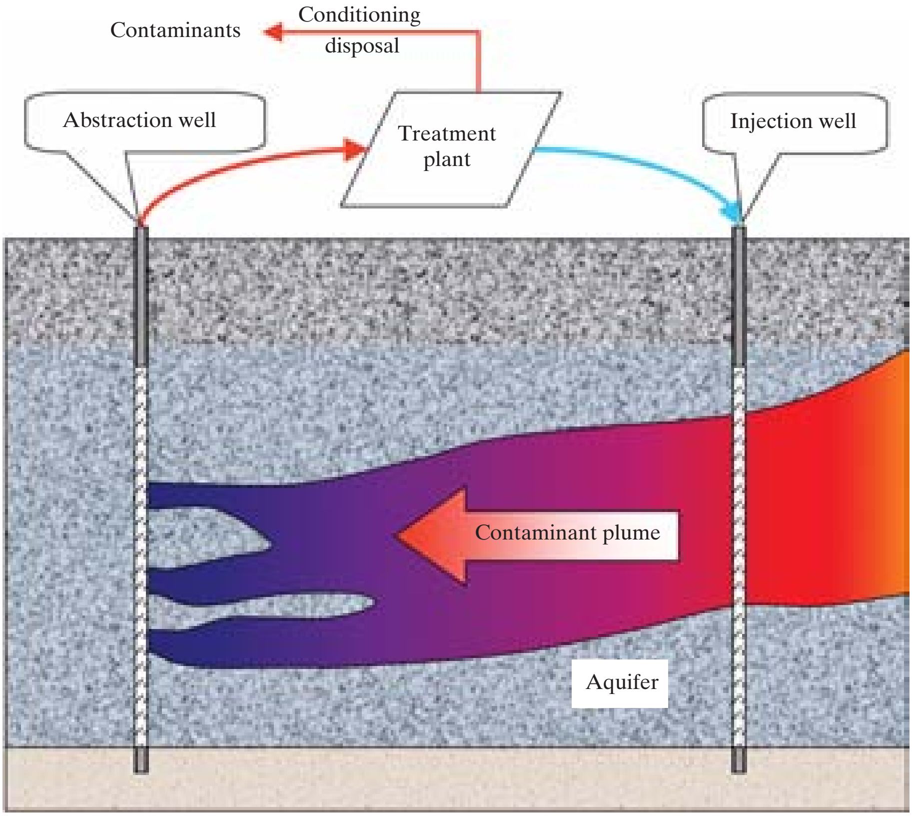

FIG. 17. A pump and treat system.

of subsurface contamination. The report found that pump and treat is inefficient as a source removal technology, although it can reduce source term volumes. In line with other methods based on changing the distribution between two different phases of a contaminant, this method becomes increasingly inefficient as the concentration gradient between, for example, species sorbed on the solid matrix and aqueous species diminishes. Large quantities of groundwater may have to be pumped and treated to remove only small amounts of contaminant. Removal in situ is inefficient, owing to tailing or mass transfer limitations. A further complication arises from the fact that not all pore water is mobile. Contaminants may be trapped in dead end pores and released into the mobile pore water only by diffusive processes, which is one of the mechanisms responsible for the tailing. Although various configurations of abstraction wells, etc., have been investigated with a view to increasing the degree of hydraulic connectedness and hence efficiency [307, 308], these configurations have been unable to overcome the fundamental constraints on diffusion.

Undesirable water properties, for example low pH values, as is often the case with mine effluents or disposal facility leachates, may pose special problems during processing [309]; a neutralization step might be required [310].

For these reasons it is unlikely that simple pump and treat methods will have much scope for application in situations with relatively low levels of contamination.

6.4. ENHANCED RECOVERY

It may be desirable to chemically treat aquifers in order to facilitate or accelerate the recovery of radionuclides or to lower residual concentrations in pump and treat scenarios [305]. Such methods are often termed 'soil flushing' [311]. After removal of the contaminant and before being reinjected, the pumped water is dosed with lixiviants, for example acid, surfactants [312], complexing agents such as EDTA [313, 314] or inert electrolytes, to replace sorbed radionuclides. However, unwanted side effects, such as dissolution of the rock matrix, may be difficult to predict. Some of the available extraction methods are used in hydrometallurgy to enhance metal value recovery [315, 316]. Figure 18 shows the principal layout for the treatment of an aquifer, while Fig. 19 shows the arrangement for treating the unsaturated zone above an aquifer.

Biological in situ leaching may be appropriate as a technique for enhanced recovery. More details on this method are given in Section 6.9.

Electrochemical methods for enhancing recovery of radionuclides in aqueous solutions have been proposed [304, 317–319]. If an electric field is applied to a solution, inorganic and organic ions migrate according to their charges to the respective electrodes (Fig. 20). Two primary mechanisms transport contaminants through the soil towards one or the other electrode: electromigration and electro-osmosis. In electromigration, charged particles are transported through the substrate. Electrolysis arrangements concentrate metal ions on the cathode and can aid the oxidation of organic contaminants. In contrast, electro-osmosis is the movement of a liquid containing ions relative to a stationary charged surface. The direction and rate of movement of an ionic species will depend on its charge, both in magnitude and polarity, as well as on the magnitude of the electro-osmosis induced flow velocity. Non-ionic species, both inorganic and organic, will also be transported along with the electro-osmosis induced water flow.

Different types of electrode material have been tested to improve performance, including porous ceramics and the rather novel carbon aerogels

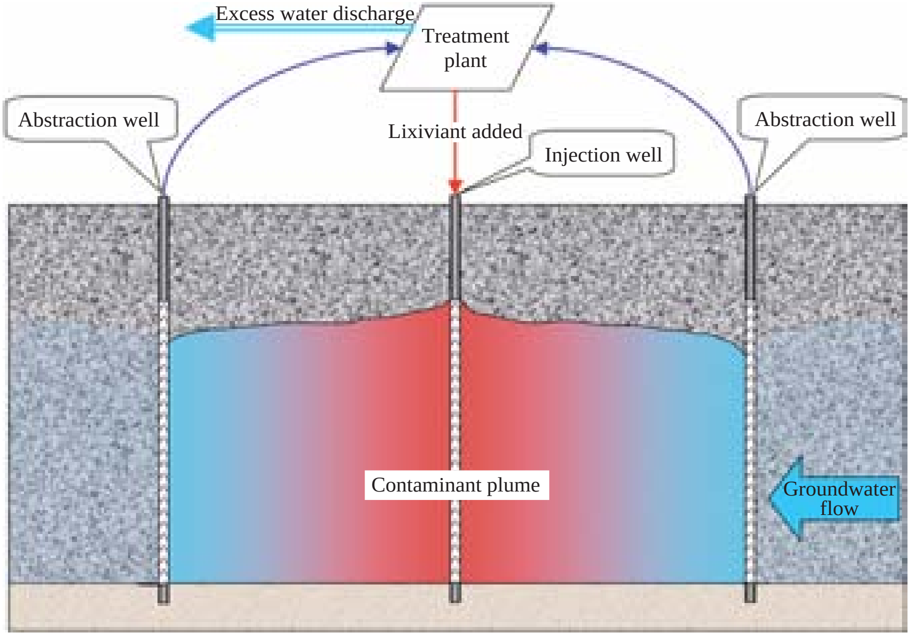

FIG. 18. Sketch of an in situ leaching or enhanced recovery arrangement.

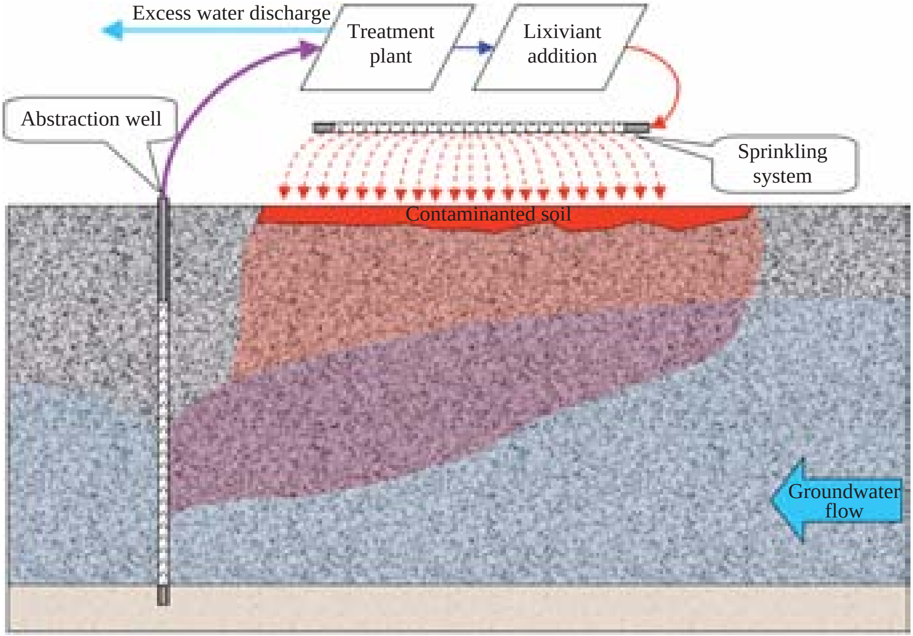

FIG. 19. Sprinkling of contaminated soil in the vadose zone to remove contamination.

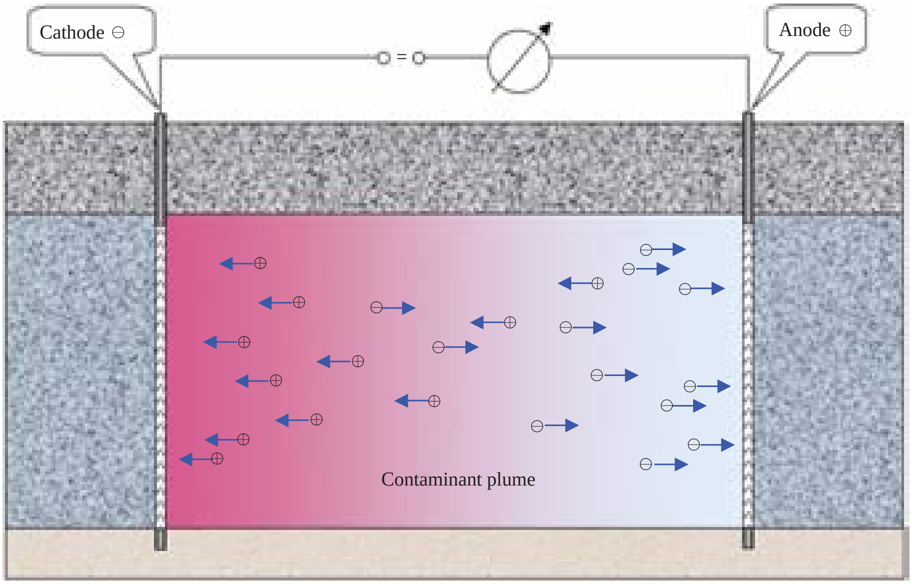

FIG. 20. Generic layout for remediation by electrolysis and electro-osmosis.

that increase the effective surface area [302]. Electro-osmosis may be combined with other techniques to remove contaminants from low permeability geomatrices such as clays. LASAGNA is a technology demonstration project designed to evaluate a combination of techniques [168].

6.5. CHEMICAL EXTRACTION (EX SITU)

Disposal of contaminated soil is likely to be an inefficient option in most cases, owing to the large volumes involved [24], even though compaction may help to reduce the volume [320]. Separation or extraction of the contaminants is preferable where no suitable in situ technique is available, and a variety of proprietary processes have been developed over the past decade or so (e.g. Ref. [321]). Excavated soils can be treated with inorganic and organic solvents to dissolve and remove metals, including radionuclides [322]. The effectiveness and efficiency of a given solvent will depend on the type of binding between the radionuclide and the soil substrate and on the chemical species of the radionuclide. The choice of chemicals that can be applied is much more varied than for in situ treatment, given the better control of the processes and the fact that the operation can be carried out in closed reaction vessels. Considerations of environmental impact from the remediation operation, for example unwanted effects on the groundwater and aquifers, are restricted to considerations that apply to similar industrial operations.

The solvents used can be either salt solutions that aim to replace radionuclides from sorption sites, as in backwashing ion exchangers, or acids that destroy substrates, such as carbonates, chelating agents [323–326] or surfactants [312] that bring sorbed ions into solution, or organic solvents that solubilize neutral species.

The recovery of low concentrations of uranium and other radionuclides from the process solutions can be a problem in itself. It is typically carried out by ion exchange. Suitable exchangers are synthetic organic polymers [327] or certain mineral species [328], such as zeolites.

Physicochemical methods, such as dialysis or membrane separation [329], sometimes in conjunction with electrochemical methods, have also been applied to remove radionuclides from soils.

Treatment residues themselves, such as sewage sludges, may need further treatment to remove radionuclides before they can be utilized or disposed of [330].

While being offered by a wide range of commercial contractors, it must be noted that, typically, extraction or soil washing processes destroy the soil's functionality and result in a sterile product. When reuse as topsoil is planned, addition of humus formers, for example compost, is necessary.

6.6. HYDROMETALLURGICAL METHODS

Ex situ methods to remove metals from soils and rocks, such as heap leaching, have undergone considerable development in the field of hydrometallurgy and solution mining [316, 331–333] and have been adapted to remediation problems [315, 334, 335]. Although the (bio)chemical processes are similar to those discussed in Sections 6.5 and 6.9, these methods are presented here separately because of their industrial application background. Large quantities of material are excavated and piled on specially prepared pads or polders. The leaching solution is sprayed or trickled over these 'heaps' at intervals and the leachate is collected for further processing (Fig. 21). The processing follows similar lines as that for treating pumped groundwaters.

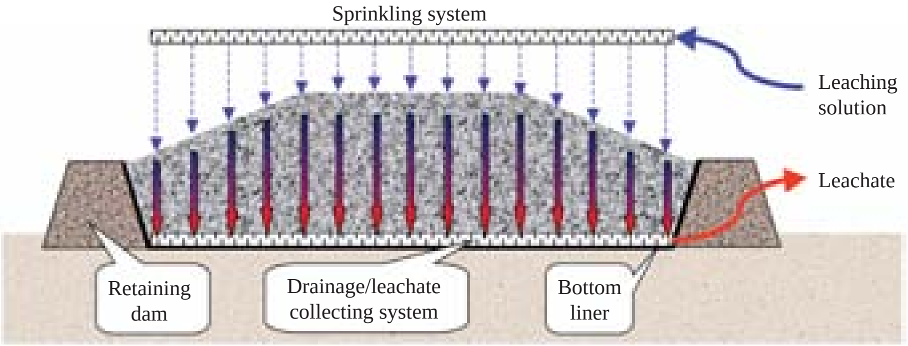

FIG. 21. A heap leaching system.

6.7. SEGREGATION

Contamination is often associated with particular size fractions or mineral phases of a soil. Separation and segregation of the contaminated fraction will greatly reduce the amount of material requiring further treatment and disposal, while freeing the reminder for reuse. Extraction processes can therefore usually be made more efficient by a prior size fractionation of the geological materials [336–338], since contaminants tend to accumulate in the clay fraction because of the larger number of sorption sites on clay minerals and their often higher specific binding energy. Radionuclides and other heavy metals may also substitute for common ions of similar ionic radius and charge as counter ions in interlayers. In certain cases neoformation of clay minerals can incorporate radionuclides in the mineral lattice.

A variety of separation techniques have been borrowed from mineral processing, including mechanical sieving and screening, hydraulic size fractionation in, for example, settling tanks or hydrocyclones, specific gravity separators such as shaking tables or sluices, surface chemistry related processes such as froth floatation, and processes based on the different magnetic susceptibilities of different minerals. A combination of these techniques may be required to isolate the relevant fractions. Segregation is often the first step before one of the above chemical extraction methods is applied. The latter are also referred to as soil washing, if they form part of an extraction procedure.

6.8. BIOSORPTION

Scientific and technological developments over the past few decades have made possible the application of biological processes to the solution of complex environmental problems (Refs [339–341] and the proceedings of the international biohydrometallurgy symposia). Microorganisms, that is to say single cell organisms such as bacteria or fungi, have been used as minute biological reactors that can efficiently and economically carry out specialized operations. Microbial biomass, whether living or not, has been shown to selectively sequester and retain elements from dilute aqueous solutions via a process named biosorption.

Through the process of biosorption the biosorbed species are selectively removed from the solution and are retained inside the microbial cells (biomass) in concentrations that are several orders of magnitude higher than those in the original solution. Heavy metals and radionuclides are taken up into cellular components such as the cell walls of certain microorganisms, which then can be harvested, carrying along the sequestered radionuclides. Biosorption is being explored in hydrometallurgy to concentrate metal bearing solutions, for example from heap leaching, and in the treatment of contaminated mining effluents [291, 316, 342, 343].

Engineering developments in the area of biosorption have led to the design of engineered biosorbents, microbial biomass cells or cellular components immobilized on or within various matrices, thus acquiring the form of small particles such as those of conventional adsorbents (e.g. activated carbon) or ion exchange resins. Organic cellular material derived from higher plants or algae have also been proposed as the basic material for manufacturing biosorbents that can be used for the extraction of metals, including radionuclides [344–354].

Biosorption methods are largely ex situ methods applicable for diluting contaminated solutions such as groundwaters or seepage. The contaminated solution is pumped into engineered reactors, in which it contacts the immobilized microbial biomass under optimized conditions (solution pH, flow rate, etc.). The contaminants are retained in an insoluble form by the biomass and the treated solution is let out of the reactor. The process of biosorption is reversible under certain conditions, which means that after the biosorbent is exhausted it could potentially be used for regeneration, releasing the previously held radionuclides in a small volume of the regenerating solution. Alternatively the biosorbent could be used once through and then disposed of appropriately.

Biosorption is an equilibrium process, with solution pH playing the role of the master variable, since it defines the speciation of the elements in the solution. This also means that the key driving force that dictates the biosorptive uptake capacity of the biomass in terms of mass of biosorbed species per unit mass of biosorbent (also referred to as the loading capacity) is the residual concentration of the contaminants after treatment and not the initial contaminant concentration [355].

The optimal biosorption pH depends on the biomass used and on the elements being removed; for example, the biosorption of uranium by the fungus *Rhizopus arrhizus* appears to be optimal at pH4, with significant reduction of the metal uptake capacity as the pH drops to pH2. The increased concentration of hydrogen ions at the acidic pH along with the chemical effects on the cell walls of the microorganisms are responsible for this reduction in capacity [356, 357]. However, increasing the pH towards neutral values may again create operational problems, depending on the composition of the contact solution. The hydrolysis and subsequent precipitation of ferric ions which adsorb on to (coat) the biosorbent adversely affect the biosorption process [227].

The biosorption of metals by algal biomass is another example in which the sequestering of metals such as lead, zinc or copper by microorganisms such as *Chlorella vulgaris*, *Chlorella regularis* or *Chlamydomonas* sp. is optimal in the range of pH6 to pH8. The biosorption of oxyanions such as chromates or selenates by the same type of algae has an optimal biosorption pH in the acidic range of pH2 to pH3 [358, 359].

Biosorption of ^{226}Ra by several types of microorganisms, such as *Rhizopus arrhizus*, *Aspergillus niger* and *Streptomyces niveus*, exhibited an optimal contact pH in the neutral to alkaline range, with corresponding radium equilibrium uptake capacities in the range of tens of MBq/g. It is therefore obvious that optimization of biosorption processes should be made on a case by case basis and requires increased care so that the process will perform satisfactorily.

Considerable efforts have been made to understand the underlying mechanisms of biosorption and to improve the process efficiency. The available information has shown that cell walls are the major biosorption functional sites for heavy metals, uranium and thorium. It has also been shown that EPSs play a significant role in biosorption [360]. The molecular level understanding of the biosorptive processes is still limited to selected pairs of metals and microorganisms. The microbial biomass provides ligand groups on to which the metal species bind. In addition, sorptive and hydrolysis processes play a role. Three major classes of microbial biopolymers (proteins, nucleic acids and polysaccharides) provide biosorption sites. Different ionic species of a given element might exhibit preference for a different binding site. Should the preference of one metal ion for a ligand be similar to that of another ion, a biosorption competition effect might be observed if both elements are simultaneously present in the contact solution.

A model of bisorption competition effects that is based on Pearson's classification of metals [361] has been reported as a basic tool for understanding such effects. On the basis of this model, significant ionic competition effects can be observed for metals belonging to the same Pearson classification class. Elements belonging to different classes demonstrate limited competition, while elements belonging to the Pearson's classification borderline class are affected by the presence of competing co-ions [362]. Additional systematic work for the mechanistic understanding of biosorptive processes and the associated ionic competition effects is required.

Numerical simulation techniques play an important role in designing and assessing remediation processes, including those using biotechnological methods [363–365]. Although biosorption using inactive microbial biomass has been demonstrated to be effective in substantially removing (and in some cases recovering) targeted radionuclides such as uranium, radium and thorium from contaminated solutions, a full scale commercial application is not yet available. The use of living microorganisms in innovative reactor configurations has recently been under investigation for the same purposes as conventional biosorption. This approach to the biological sequestering of metals has substantially different requirements and operating conditions than conventional inactive biomass biosorption. This alternative biotechnological approach is often referred to as bioaccumulation or bioprecipitation and is showing excellent potential.

6.9. BIOLEACHING

Bioleaching occurs naturally when microorganisms assist in the slow weathering of out-cropping sulphide ore bodies. Bioleaching is an established biotechnological process for the dissolution and hence mobilization of valuable metals from ores by microorganisms [366–371]. Metals for which this technique is mainly employed are copper, cobalt, nickel, zinc, gold, silver and uranium. It is estimated that about 20–30% of the world's copper production originates from bioleaching; in the case of uranium it is judged to be about 5–10%. Bioleaching has also been promoted as a cost efficient method for metal value recovery in developing countries, and its applicability in this context has recently been reviewed [372].

Bioleaching also has scope for application in reworking waste material from mining for enhanced recovery of metals, including radionuclides, which has the potential to reduce the environmental burden. The method has been explicitly applied to the remediation of uranium and other mining legacies [373, 374]. The pathways of the resulting contaminated waters have to be carefully controlled, for example by arrangements similar to those for heap leaching (Fig. 21). Microbially mediated leaching processes frequently have the unwanted side effect of AMD generation, for example by pyrite oxidation [375].

The types of ore that are amenable to bioleaching comprise sulphides, carbonates and oxides. The groups of microorganisms involved are mainly bacteria and fungi. In some cases algae and lichens may also play a role. Various mechanisms are involved, depending on the type of ore in question.

In the case of sulphidic minerals the predominant dissolution causing microorganisms are acidophilic (meaning organisms living between pH0 and pH5) bacteria of the sulphur and iron cycles, namely *Acidithiobacillus* (abbreviation *A.*, former name *Thiobacillus*) *ferrooxidans*, *A. thiooxidans*, *Leptospirillum* (abbreviation *L.*) *ferrooxidans*, *A. caldus*, *Metallogenium* sp., *Sulfobacillus thermosulfidooxidans*, *Sulfolobus* sp., *Acidianus brierleyi* and several others. The species of *Acidithiobacillus* live in the moderate temperature range (0–45°C), *Metallogenium* and *Sulfobacillus* thrive at elevated temperatures (40–65°C) and *Sulfolobus* and *Acidianus* are thermophiles growing from 65 to 90°C.

The dissolution is generally effected by two mechanisms, depending on the type of mineral to be dissolved:

(a) Pyrite and molybdenite and a few other minerals of the same structure can only be dissolved by an oxidizing attack on their crystal lattice, owing to their electronic configuration (non-bonding outer orbitals) [376]. The bacteria able to do this are the Fe(II) oxidizing *A. ferrooxidans*, *L. ferrooxidans* and *Acidianus* sp. This mechanism is known as the thiosulphate mechanism.

(b) All other sulphidic minerals possess bonding outer orbitals and thus are more or less dissolvable by a hydrolytic attack involving protons. In addition, Fe(III) ions further the dissolution by an oxidizing attack. These minerals may consequently be dissolved by all the above mentioned bacteria of the sulphur and iron cycles. This dissolution process is known as the polysulphide mechanism.

In both cases, the dissolution of the mineral is mainly effected by bacteria attached to the surface of the respective mineral. The compounds mediating such attachment are EPSs (‘slimes’). The EPSs consist, from a chemical point of view, mainly of lipids, carbohydrates, sugar acids (uronic acids) and complexed, inorganic ions such as Fe(III) ions. The distance between the bacterial cell and the mineral substrate surface is of the order of 20 to 50 nm. This space is filled with the EPS, creating a reaction space with unknown conditions of pH, redox and ion concentrations; the reaction space is an extension of the radius of action of the cell, thus allowing it to augment its food supply. As a consequence, biological leaching becomes considerably accelerated (sometimes more than 100-fold) compared with the purely chemical process utilizing Fe(III) ions and/or protons only. In the latter process the freely suspended planktonic cells also have to be considered, since their effect is mainly the reoxidation of the iron ions in solution. Bioleaching is thus an interface process and belongs to the area of nanobiotechnology.

Final products of dissolution are metal cations, Fe(III) ions, sulphate and/or sulphuric acid. The energy of the oxidation is partially conserved by the bacteria for metabolic purposes and growth. The bacteria possess specialized cell components allowing them to conserve some of the energy in a utilizable form (ATP, NADH). Furthermore, they need only carbon dioxide from the air to build up their cell mass and inorganic trace elements. These are therefore very specialized organisms; this type of metabolism is called lithoautotrophy.

The above mentioned bacteria are generally not important for carbonate and/or oxide ores. Bacteria and fungi are used for dissolving such minerals, which, due to an unbalanced metabolism, excrete organic acids. This requires an ample supply of exogenous carbon sources, which they metabolize, and as a consequence of either too much substrate, or a lack of essential nutrients or trace elements such as nitrogenous compounds or minerals, excrete partly in an intermediate oxidation state. Excreted acids are, for example, citric, oxalic, succinic, malic, acetic and/or formic acid, and sugar (uronic acids) or amino acids. These acids dissolve and/or complex metal cations and thus solubilize them.

The bioleaching technique is employed in several forms.

(a) In the case of low grade ores that for economic reasons cannot be processed by conventional roasting or other similar processes, a heap leaching process is applied (Fig. 21). In the majority of cases in which this technique has been applied to date, the ore contained copper, zinc and trace elements. A limited number of experiments of this type have been performed for extracting uranium from low grade ores. One experiment was carried out near Ronneburg, Germany, by Wismut in the 1980s, another one at Elliott Lake in Canada. For this purpose large amounts of low grade ores are placed on leach pads (plastic liners) or dumped in valleys with a known and impermeable geological strata and sprinkled regularly with acidified bacteria-containing solution (which originates from similar operations or from acid mine waters). The dissolved metals and sulphate plus sulphuric acid are left to accumulate to a concentration at which extraction processes such as solvent extraction, ion exchange and/or electrowinning become viable. Residence times for such operations range from several months to a few years. If these heaps are constructed without consideration of the underlying geology and/or abandoned without care, acid mine–rock drainage (ARD or AMD) may result. Abandoned mines, mine shafts, open pits, etc., might also produce and release ARD. Owing to the acidity combined with dissolved heavy metals, this might result in serious environmental damage and/or even create new ecosystems (as in Rio Tinto, Spain).

(b) Bioleaching has been employed experimentally at the field scale in Germany to treat heavy metal contaminated sediments [377]. The generic scheme from the raw sediment to a viable soil substrate is illustrated in Fig. 22.

(c) In the case of sulphidic concentrates, bioleaching is increasingly used for extracting precious metals. In recent years several plants have gone into operation that use acidophilic leaching bacteria for extracting gold, nickel and cobalt. The operation usually consists of stirred tanks (bioreactors) with volumes of up to 1000 m^3 in continuous operation. Residence times are in the range of 3 to 7 days.

Another form of application is in situ bioleaching (see Fig. 18) [378]. This technique is applied: (1) to treat ore bodies that are located too deep underground for conventional mining; (2) to treat leftover ore from other mining activities, as was the case with the uranium bioleaching at Elliott Lake, Canada; and (3) to avoid damage to the surface environment. Consequently,

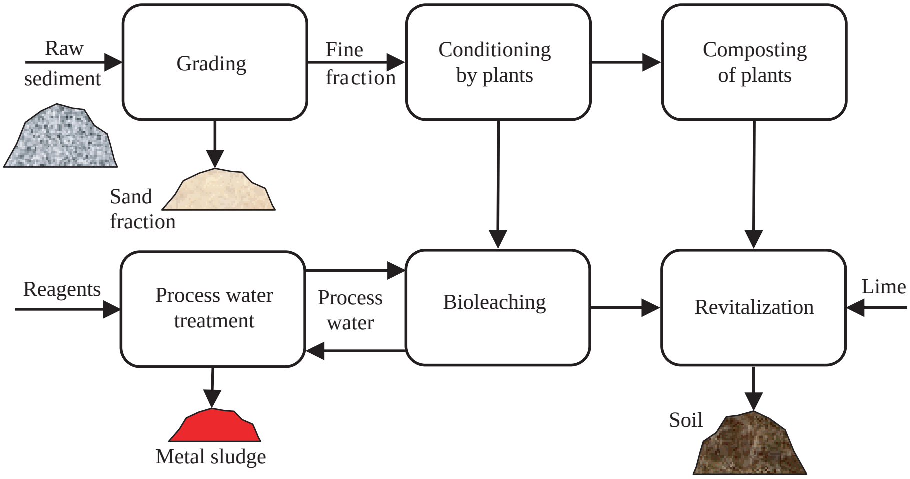

FIG. 22. The experimental process from raw contaminated sediment to reconditioned soil, after Ref. [377].

for bioleaching the host rocks are typically prepared by blasting to perforate them and by the installation of pumping in and pumping out pipe systems for the application, recirculation and extraction of leach solutions. The pregnant solution is then processed in an extraction plant to extract the valuable metals. Depending on the geological setting, such in situ leaching may pose different hazards to the environment. Sometimes, losses of the leach solution that enter unnoticed underlying aquifers via faults or fissures in the separating geological strata are encountered.

In the case of radioactive minerals, there may also be another, unwanted effect: an enhanced emission of radon. Comparison of the radon emission rates and bioleaching activity at the above mentioned leaching waste heaps near Ronneburg, Germany, has shown that high cell numbers of leaching bacteria were found at sites with high radon emissions, whereas at sites with low emissions only low cell numbers occurred. An explanation for this effect comes from the mineralogy of the ore. At Ronneburg the uranium is embedded in pyrite. Once this pyrite has been attacked by bioleaching, radon is liberated and may escape into the atmosphere. This causes an additional exposure for the local population and requires measures to reduce or even inhibit the biological process.

6.10. PHYTOEXTRACTION

6.10.1. Overview

The use of plants to remove contaminants from the environment and concentrate them in above ground plant tissue is known as phytoextraction [379–394]. Phytoextraction requires that the target metal (radionuclide) be available to the plant root, absorbed by the root and translocated from the root to the shoot; biomass production should be substantial. The metal (radionuclide) is removed from the site by harvesting the biomass, after which it is processed either to recover the metal or further concentrate the metal (by a thermal, microbial or chemical treatment) to facilitate disposal.

Research and development efforts have focused on two areas: (1) remediation of contaminants such as lead [395–397], arsenic, chromium, mercury and radionuclides [129, 386, 398–406]; and (2) mining, or recovery, of inorganic compounds, mainly nickel and copper, having intrinsic economic value.

Successful implementation of phytoextraction depends on [284]:

(a) The bioavailability of the contaminant in the environmental matrix;

(b) Root uptake;
(c) Internal translocation of the plant;
(d) Plant tolerance.

Plant productivity (i.e. the amount of dry matter that is harvestable each season) and the accumulation factor (the ratio of metal in plant tissue to that in the soil) are important design parameters [406–408]. This is clearly exemplified by the following set of equations and tables [406]. The percentage yearly reduction in soil activity can be calculated as:

$$\text{Annual removal (\%)} = \frac{\text{TF} \times \text{yield}}{W_{\text{soil}}} \times 100 \tag{1}$$

where TF is the transfer factor or bioaccumulation factor (TF = $C_{\text{plant}}/C_{\text{soil}}$, with C_{plant} the concentration of the radiocontaminant in the plant (Bq/g) and C_{soil} the concentration of the contaminant in the soil) and W_{soil} the weight of the contaminated soil layer (kg/ha). As is evident from Eq. (1), the annual removal percentage increases with yield and TF. However, TF and yield values are not independent: a high yield is often associated with lower TFs because of growth dilution effects.

Phytoextraction typically requires several years of operation, and the future trend in radionuclide concentration in the soil can be calculated from:

$$C_{\text{soil},t} = C_{\text{soil},t=0} \exp\left[-\left(\frac{\text{TF} \times \text{yield}}{W_{\text{soil}}} + \frac{0.69}{t_{1/2}}\right) \times t\right] \tag{2}$$

The second term in the exponent of Eq. (2) accounts for radioactive decay ($t_{1/2}$ is the half-life of the radionuclide). For some radionuclides with long half-lives (e.g. $t_{1/2}$ for ^{238}U is 4.5 × 10^9 a), this component will not affect the phytoextraction potential. For others, for example ^{137}Cs and ^{90}Sr, with half-lives of 30 years, the phytoextraction potential will be affected; that is, a yearly loss of 2.33% in activity occurs merely through radioactive decay. Equation (2) assumes a constant bioavailability of the contaminant (i.e. a constant TF).

Table 4 shows, for a calculated example, the percentage annual removal per hectare for different crop yields and TFs, based on a 10 cm deep soil layer that has a mass of 1500 t for a soil density of 1.5 kg/dm^3. It should be borne in mind that if the contamination is spread to a depth of 20–50 cm in the soil, annual removal with the biomass is reduced by a factor of 2 to 5, respectively, compared with the figures presented in Table 6.

TABLE 4. PERCENTAGE YEARLY REDUCTION OF SOIL CONTAMINATION DUE TO PHYTOEXTRACTION AND RADIOACTIVE DECAY [406]

TF (g/g)	Annual reduction due to phytoextraction (%)					Annual reduction due to phytoextraction and decay (%)				
	Yield (t/ha)					Yield (t/ha)				
	5	10	15	20	30	5	10	15	20	30
0.01	0.003	0.007	0.01	0.013	0.02	2.33	2.34	2.34	2.34	2.35
0.1	0.033	0.067	0.1	0.133	0.2	2.36	2.40	2.43	2.46	2.53
1	0.33	0.67	1.00	1.33	2.00	2.66	3	3.33	3.66	4.33
2	0.67	1.33	2.00	2.67	4.00	3	3.66	4.33	5	6.33
5	1.67	3.33	5.00	6.67	10.00	4	5.66	7.33	9	12.33
10	3.33	6.67	10.00	13.33	20.00	5.66	9	12.33	15.7	22.33

Note: $t_{1/2}$: 30 a; soil depth: 10 cm; soil density: 1.5 kg/dm^3.

Yields of more than 20 t/ha and TFs higher than 0.1 (Table 4) may be regarded as upper limits, except for strontium. This would result in an annual reduction percentage of 0.1% (decay excluded). When TF equals 1, the annual reduction is about 1%. Table 5 gives some ranges for TFs for the natural radionuclides uranium and radium, predominant contaminants in the NORM industries, and the long lived fission products ^{137}Cs and ^{90}Sr.

By rearranging Eq. (2) the number of years needed to attain the required reduction factor as a function of annual removal percentage can be calculated. Table 6 presents the number of years required to attain a reduction of the contaminant concentration up to a factor of 100, given an annual extraction

TABLE 5. RANGES FOR TFs (RATIO) BASED ON DATA FROM REFS [122, 123, 141, 146, 402, 403, 409–416]

	Total range (Bq/g plant to Bq/g soil)	Comment on conditions for upper limit
Cs	0.00025 – 7.5	*Brassica*, organic soil
Sr	0.0051 – 22	Green vegetables, sandy soil
U	0.000006 – 21.13	Tubers, sandy soil
Ra	0.00029 – 0.21	Grass, sandy soil

TABLE 6. CALCULATED NUMBER OF YEARS REQUIRED TO DECONTAMINATE A SOIL FOR A REQUIRED (DESIRED) REDUCTION FACTOR AND A GIVEN ANNUAL REMOVAL PERCENTAGE

Desired reduction factor	Activity remaining, $C_{\mathrm{soil},t}/C_{\mathrm{soil},t=0}$ (%)	Annual removal (%/a)							
		20	15	10	5	3	2	1	0.1
5	20	7	10	15	31	53	80	160	1650
10	10	10	14	22	45	76	114	229	2301
20	5	13	18	28	58	98	148	298	2994
50	2	18	27	37	76	128	194	389	3910
100	1	21	28	44	90	151	228	458	4603

Note: Soil depth: 10 cm; soil density: 1.5 kg/dm^3.

percentage or percentage reduction in radionuclide activity varying between 0.1% and 20%. With an annual removal of 0.1% it would take more than 2000 years to decontaminate a soil to 10% of its initial activity; with an annual removal of 1%, more than 200 years are required. It is hence clear that measures would need to be taken to increase the annual removal efficiency through crop selection, or to increase the bioavailability by applying soil additives and through technical measures (e.g. decreasing the tilled soil depth).

In most cases one has limited control over the depth of the contamination, although it may be feasible and advantageous to excavate and pile the soil to the desired soil depth for phytoremediation purposes. One possibility is to excavate the soil and spread it on geomembranes, which impedes roots from penetrating to deeper layers. These membranes will also limit contaminant dispersal to the underlying clean soil, but a substantial area may be needed for treatment. Decreasing the tilled soil depth increases the removal percentage according to Eq. (1) and may intensify root–soil contact, and may result in an increased TF.

The other factors influencing radionuclide bioavailability, such as crop selection and measures to increase the bioavailability of the radionuclide of concern, are generally radionuclide specific. To maximize the metal content in the biomass, it is necessary to use a combination of improved soil management measures, for example optimizing the soil pH and mineral nutrient contents, or the addition of agents that increase the availability of metals.

Apart from the application of soil additives to increase export with the plant biomass, plant selection may also be important for improving the phytoextraction potential [417]. As already mentioned, there is a significant interspecies variability in TFs (Table 9). Since the values are seldom obtained for similar soil and growth conditions, the effect of plant species on the TFs cannot be unambiguously derived. Observed differences between plant varieties or cultivars have been up to a factor of 2 [403].

Improved genotypes with optimized metal uptake, translocation and tolerance, and improved biomass yield, may also be an approach to improved phytoextraction. Plant breeding [418] and genetic engineering may open further alleys to develop hyperaccumulating plants [419–421], but actual research and technology development is mostly limited to heavy metals.

Although positive effects have been obtained following applications of soil amendments that increase element bioavailability, the effect of continuous treatment on soil quality, plant growth and bioaccumulation is not clear. There also remains the question of long term effectiveness: will TFs remain constant or will they decrease as radionuclide concentrations decrease.

Effective extraction of radionuclides and heavy metals by hyperaccumulators is limited to shallow soil depths of up to 30 cm. If a contamination is found at substantially greater depths (e.g. 2–3 m), deep rooting perennial crops could in principle be employed, but the fraction of their roots exploring the contaminated zone would be small and hence also the phytoextraction potential.

There are concerns that contaminated leaf litter and associated toxic residues [422] may result in uncontrolled dispersion of the contaminants. Finding a safe use or disposal route for contaminated biomass will be a major element in developing a phytoextraction scheme.

Little is known about the economics of phytoextraction, which not only depends on the extraction efficiency but also on the costs associated with crop management (i.e. soil management, sowing or planting (yearly returns for annual crops), harvesting, post-harvest biomass transport, biomass treatment, potential disposal costs and site monitoring). The treatment of 1 m^3 of contaminated soil (10 m^2 for a 1 dm soil layer) will result in about 10–20 kg of biomass (~2–4 kg of ash) annually.

6.10.2. Uranium removal

Free UO_2^{2+} is the uranium species most readily taken up and translocated by plants. Since this uranium species is only present at a pH of pH5.5 or less, acidification of uranium contaminated soils may be necessary for phytoextraction [401]. The uranyl cation also binds to the soil solids and organic matter, reducing the extent of plant uptake [423, 424]. Therefore, in addition to acidification, soil amendments that increase the availability of uranium by complexation may also be required. In testing the role of acidification and chelating agents on the solubilization of uranium it was found [402, 403] that, of the organic acids and chelating agents tested, citric acid was the most effective for increasing uranium in the soil solution. Following citric acid treatment (20 mmol/kg) the uranium accumulation in Indian mustard (*Brassica juncea*) was increased 1000-fold [403] and in beet (*Beta vulgaris*) tenfold [402, 403].

Similar results were obtained when testing the potential for phytoextraction of uranium from a low level contaminated sandy soil using rye grass (*Lolium perenne* cv. Melvina), Indian mustard (*Brassica juncea* cv. Vitasso) and redroot pigweed (*Amarathus retroflexus*) [129]. The annual removal of the soil activity with the biomass was less than 0.1%. Addition of citric acid increased uranium uptake up to 500-fold, and extraction percentages of 2–5% appear achievable. Citric acid addition, however, resulted in a decreased dry weight production (all plants tested) and even plant death and crop regrowth (in the case of rye grass). Depending on the desired contamination reduction factor (e.g. 5–50), it would still take between 30 and 200 years for the target to be met (Table 6).

6.10.3. Strontium removal

Table 7 shows the annual crop removal of ^{137}Cs and ^{90}Sr. It is clear from this table that in normal agricultural systems the annual caesium flux is small compared with the reservoir present in the soil. The ^{137}Cs removal rates are all less than 1%, and the highest removal is found for grassland. The high sorption of ^{137}Cs in soil and the typical potassium levels in soil required for optimal plant growth all limit removal rates.

The removal of ^{90}Sr with biomass is higher than that of ^{137}Cs because the ^{90}Sr availability is typically tenfold above that of caesium. The TFs of ^{90}Sr in green vegetables and *Brassica* plants are typically around unity and the upper levels are around 10 [409]. Phytoextraction of ^{90}Sr has not yet been investigated at the field scale. The high removal rates in agricultural crops (Table 7) suggest that phytoextraction may be worth while to explore.

The highest transfers of ^{90}Sr are typical for leguminous perennial grasses (*Trifolium* family) and *Brassica* plants [25]. Field experiments in Belarus were carried out at the Belarussian Research Institute for Soil Science and Agrochemistry (BRISSA) [425] on light-textured soil contaminated with ^{90}Sr (Table 8). It was found that cow clover (*Trifolium pratense*) has annual green mass yields of up to 65–75 t/ha (6–7 t/ha dry mass). The ^{90}Sr removal values were in the range 2.5–3.6% of the total radionuclide reservoir in the soil. A change of soil pH from neutral (pH6.8) to moderately acid (pH4.9) enhanced the ^{90}Sr transfer by a factor of almost 2, but the yield of clover was reduced, so the total

TABLE 7. ANNUAL REMOVAL BY CROP BIOMASS OF ^{137}Cs AND ^{90}Sr FOR SOME AGRICULTURAL CROPS, EXPRESSED AS A FRACTION OF TOTAL CONTENT IN THE TILLED LAYER (ARABLE CROPS) OR IN THE 0–12.5 cm LAYER (GRASSLAND)

	Yield (dry matter) (t/ha)	Caesium TF (g/g)	Caesium crop removal (% of total in soil)	Strontium TF (g/g)	Strontium crop removal (% of total in soil)
Cereals (grain)	5–7	0.0004–0.25	0.0005–0.06	0.02–0.94	0.0037–0.22
Potato tuber	6–10	0.003–0.89	0.0006–0.3	0.03–1.4	0.006–0.5
Leafy vegetables	5–10	0.008–1.7	0.001–0.6	0.45–9.1	0.07–3.0
Grassland	10–15	0.01–1.0	0.007–1.0	—	—

Note: The TF ranges were derived from Ref. [409].

accumulation of radionuclide per unit area was increased only by a factor of 1.5. It should be noted that when the clover is used as animal fodder, the greater part of ^{90}Sr activity will end up in dung and in normal agricultural practice would be returned back to the fields. An alternative, non-dispersive use of the biomass has not yet been developed for this example in Belarus.

It may hence be concluded that, except for ^{90}Sr, annual removal of contaminants with plant biomass is generally too low to allow phytoextraction to be efficient without soil additives that increase bioavailability. The high removal rates in agricultural crops for strontium suggest that phytoextraction could be explored with benefit for this element.

6.10.4. Caesium removal

Given its similarity to potassium, the soil potassium status will affect ^{137}Cs availability [141, 426–430]. Generally, the higher the soil potassium, the lower the TF. Extremely low soil fertility with regard to potassium may increase ^{137}Cs TFs tenfold to 100-fold [427, 431, 432], but will also decrease plant growth. A decrease in pH [136, 426] and decreased ammonium levels [141, 398, 433–436] generally increase caesium soil to plant transfer, but the effects are generally limited (a factor of 2).

TABLE 8. STRONTIUM-90 ACCUMULATION BY CLOVER ON PODZOLUVISOL LOAMY SAND SOIL IN BELARUS (DEPOSITION: 37 kBq/m^2) [425]

Fertilizer treatment	Crop yield (green mass) (t/ha)	^{90}Sr activity (Bq/kg)	^{90}Sr accumulation in yield (kBq/ha)	^{90}Sr accumulation (% of total soil concentration)
		For soil pH (KCl) = 4.9		
P_{60}	36	243	8 809	2.4
$P_{60}K_{60}$	46	238	10 948	3.0
$P_{60}K_{120}$	54	223	12 098	3.3
$P_{60}K_{180}$	65	207	13 455	3.6
		For soil pH (KCl) = 5.9		
P_{60}	40	198	7 871	2.1
$P_{60}K_{60}$	49	188	9 165	2.5
$P_{60}K_{120}$	57	178	10 191	2.8
$P_{60}K_{180}$	72	160	11 440	3.1
		For soil pH (KCl) = 6.8		
P_{60}	46	169	7 732	2.1
$P_{60}K_{60}$	55	153	8 339	2.3
$P_{60}K_{120}$	64	151	9 589	2.6
$P_{60}K_{180}$	75	123	9 194	2.5

The effect of ammonium addition on the phytoextraction potential of ryegrass and *Brassica* grown on caesium contaminated soil has been tested [398]. Ammonium addition increased the dry weight yield by 20% and the TF by 80%, resulting in a TF of 0.8 g/g. With a realistic yield of 20 t/ha under field conditions, this would result in an annual reduction of 3.3% (decay included). This would imply in turn that 50 years of continued phytoextraction would be needed to reach a reduction of the soil contamination level by a factor of 5 (cf. Table 6).

Amarantus species have TFs as high as 3.2 g/g [437, 438]. With a yield potential estimated at around 30 t/ha/a (based on two harvests per year) and a target fourfold reduction in soil activity, the phytoextraction process would require 16 years to complete.

In a normal agricultural land use system the annual ^{137}Cs removal with plant yield is rather small compared with the total amount of contamination derived ^{137}Cs present in the soil [439]. It is known that the highest caesium uptake typically occurs in perennial grasses. As found recently in several field experiments in Belarus, ^{137}Cs removal rates for perennial grasses with an annual dry matter yield of 2–5 t/ha are less than 0.1% [425]. Phytoextraction of caesium in normal agricultural practice therefore appears not to be a very efficient process.

6.10.5. Phytoextraction project in Belarus

The phytoextraction effect of rape (*Brassica* sp.) is significant. Rape has a high ability to accumulate ^{90}Sr [126, 239]. In BRISSA field experiments the annual accumulation of ^{90}Sr in pods and straw reached approximately 3% of the ^{90}Sr content in the soil (Table 9). Radionuclides incorporated in straw ploughed in just after harvesting will be unavailable for one to two subsequent growing seasons, until the final mineralization of the straw. The degree of ^{90}Sr immobilization by straw is comparable in size to the reduction of soil contamination due to radioactive decay. The phytoremediation effect of growing rape may be increased by removing straw from the field and disposing of it safely. However, the disposal option is likely to be rather expensive and will deprive the soil of the necessary raw material for humus formation. Thus while phytoremediation with rape appears feasible in principle, it might be more sustainable to operate the scheme as a means for enhanced attenuation.

Available data indicate a significant interspecies variability in the transfer of radionuclides from soil to plants. However, hard experimental data for the evaluation of phytoextraction potential and for the development of an appropriate crop rotation scheme are rather scarce. Experimental data from Belarus show differences in the accumulation of ^{137}Cs for 32 varieties of spring rape between years of up to 1.8–2.7 times, and for ^{90}Sr of up to 1.8–4.0 times. It should be noted that these differences are radionuclide specific, meaning that one variety that accumulates less ^{137}Cs does not necessarily accumulate less ^{90}Sr [126]. The experimental results from Belarus allow the identification of varieties that have the desired uptake properties: more uptake for phytoextraction purposes or less uptake for minimizing the radionuclide content in the food pathway [134, 239].

TABLE 9. STRONTIUM-90 ACCUMULATION BY VARIETIES OF SPRING RAPE RELATED TO PODZOLUVISOL LOAMY SAND SOIL WITH A DEPOSITION OF 37 kBq/m^2 (1997–1998) [126]

Variety	Seed yield (t/ha)	^{90}Sr activity (Bq/kg)		^{90}Sr uptake (kBq/ha)		^{90}Sr uptake (% of total soil content)		
		Seeds	Straw	Seeds	Straw	Seeds	Straw	Total
Hanna (standard)	1.9	265	663	514	5 141	0.14	1.39	1.53
Yavor	1.9	240	648	451	4 873	0.12	1.32	1.44
Likosmos	2.0	264	686	541	5 628	0.15	1.52	1.67
Lirovel	1.8	275	688	501	5 005	0.14	1.35	1.49
Licoll	2.2	310	868	682	7 638	0.18	2.06	2.25
PF 7118/93	2.2	314	879	675	7 561	0.18	2.04	2.23
PF 7045/91	2.1	319	479	670	4 019	0.18	1.09	1.27
PF 7056/92	1.9	322	902	570	6 383	0.15	1.73	1.88
Iris	1.9	338	744	629	5 532	0.17	1.50	1.67
Orakel	2.1	344	826	726	6 968	0.20	1.88	2.08
PF 5045/88	2.2	345	759	742	6 527	0.20	1.76	1.96
PF 7369/94	2.3	358	967	816	8 815	0.22	2.38	2.60
Lizonne	1.7	395	1 027	675	7 025	0.18	1.90	2.08
PF 7410/94	1.9	407	1 140	765	8 570	0.21	2.32	2.52
Liazon	2.0	436	1 221	855	9 571	0.23	2.59	2.82
PF 7041/91	1.8	477	1 336	844	9 456	0.23	2.56	2.78
PF 7008/91	2.4	478	1 338	1 166	13 063	0.32	3.53	3.85

6.11. RHIZOFILTRATION

Rhizofiltration is the use of plants to sequester compounds from aqueous solutions through adsorption on the roots or assimilation through the roots and eventual translocation to the aerial biomass (phytoextraction). Rhizofiltration is being investigated for the removal of radionuclides from aqueous waste streams, including groundwater and wastewater [440].

Rhizofiltration is particularly effective in applications with low concentrations and large volumes of water. Plants that are efficient at translocating metals to the shoots should not be used for rhizofiltration, since additional contaminated plant residue is produced [441].

The removal of a radionuclide from an aqueous waste stream is governed by the plant dry weight production and the concentration factor, CF (ratio of Bq/g plant to Bq/mL water or soil solution). Since adsorption in (waste)water per volume is lower than in soil, the CF is higher than the TF. This becomes clear when considering the relationship between the TF and the CF, which is TF = CF/K_D, in which K_D is the solid–liquid distribution coefficient of a radionuclide (e.g. dm^3/kg) (i.e. the ratio of radionuclide activity concentration in the solid phase to that in the soil solution). Since the value of K_D for most radionuclides is generally substantially higher than 1 [27, 442, 443], it is clear that the CF exceeds the TF by the same factor and that rhizofiltration is generally more effective than soil phytoextraction.

A plant suitable for rhizofiltration applications can remove toxic metals from solution over an extended period of time with its rapid growth root system. Various plant species have been found to effectively remove toxic elements such as arsenic, copper, cadmium, chromium, nickel, lead and zinc from aqueous solutions [444, 445].

Pilot scale research on rhizofiltration has found that the roots of sunflowers (*Helianthus annuus* L.) reduced levels of lead, copper, zinc, nickel, strontium, cadmium, U(VI), manganese and Cr(VI) to concentrations near to or below regulated discharge limits within 24 h [441, 446]. Beans and mustard were less effective than sunflowers in uranium removal [447]. Virtually all uranium was concentrated in the roots, and almost none in the shoots. Removal was higher (by a factor of 2) at pH5 than at pH7.

Uranium is clearly removed much faster from contaminated pond water than caesium and strontium (Fig. 23) [447]. Sunflowers showed higher caesium and strontium removal rates than timothy, meadow foxtail, Indian mustard and peas.

However, rhizofiltration has its limits [447]. In an experiment with rather highly contaminated wastewater (1 mg/L U) and high flow rates (1.05 L/min), 95% of the uranium was removed by 6 week old sunflowers grown for 2 weeks in the wastewater, resulting in effluent concentrations of 40–70 µg/L, which are above the 20 µg/L drinking water limit.

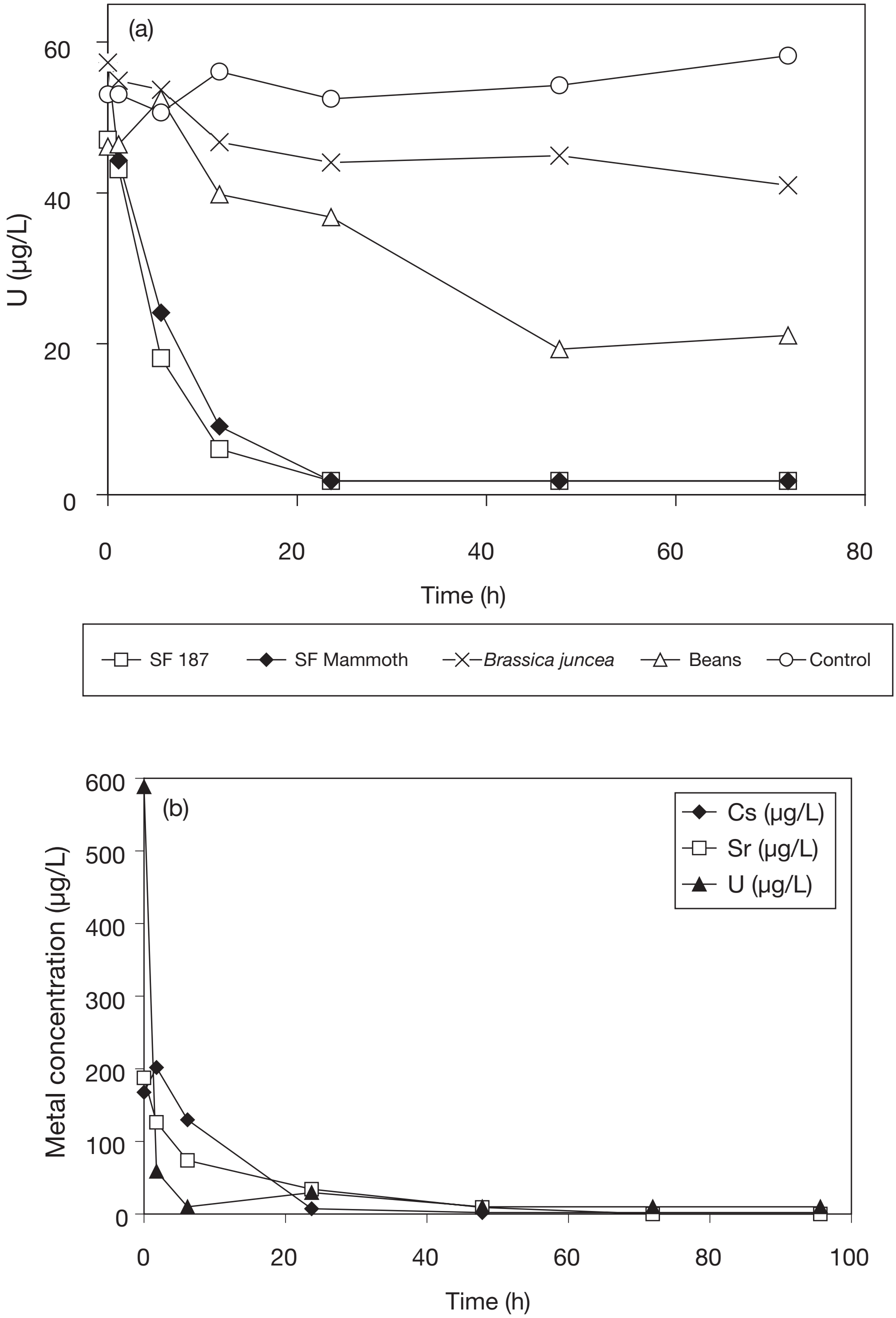

FIG. 23. Removal of uranium by different sunflower cultivars (a) and removal of caesium, strontium and uranium by sunflowers (b) [447].

7. SUMMARY AND CONCLUSIONS

Dispersed low level contamination poses a particular challenge to those charged with its remediation. Many techniques are not efficient below certain concentration thresholds or entail more severe impacts on certain environmental compartments than the contamination itself. In such cases justification for remediation may not be given on radiation protection grounds, but remediation may still be demanded by the public.

This report examines a variety of technological options for dealing with dispersed low level contamination. The objective of any technology used in a remediation project is either to remove or reduce the source term or to block exposure pathways. This can be achieved in a variety of ways and needs to be tailored to the contaminants and pathways of interest. The approaches are broadly grouped into the three categories of:

(a) Non-intervention;
(b) Containment;
(c) Removal.

While this conceptual grouping is useful for understanding the objectives behind certain technologies, it may be noted that various technologies could be grouped into more than one category. The functionality of a certain technology may change over time or may combine, for example, containment with removal.

The decision of whether to remediate actively would be based on predictive modelling and a risk assessment. If there are no acute exposure pathways, the decision not to intervene may be justified. However, the effects of natural attenuation would be monitored to verify that the contamination behaves as predicted.

Reliance on natural processes may need to be complemented by a change in the use of the contaminated land in order to minimize exposures and the uptake of radionuclides. Such change in land use may extend to (temporarily) dedicating agricultural land to forestry or may simply involve a change of crops. In coping with the widespread contamination following the Chernobyl accident, such agricultural countermeasures have found wide application for retaining some use of the land.

There are various ways by which enhanced attenuation can be attempted. Typically such measures intend to change the controlling geochemical parameters, such as pH or redox potential, to values that would disfavour migration. Enhancement of sorption binding capacities and intensities by soil additives has also been attempted.

Such measures gradually lead to more invasive measures to contain a contamination. Outright containment by impermeable or low permeability barriers and liners may encounter logistic problems for dispersed contamination. Apart from the fact that this would involve major civil engineering work, the effectiveness may be difficult to maintain.

Permeable reactive barriers seem to offer a viable alternative for a wide range of contaminants. Such barriers are intended to fix in situ a contaminant passing by with the groundwater flow. The fixation mechanisms typically are sorption or precipitation, or a combination of both. Frequently, the reactants in the barrier are also meant to induce a change in the redox of the radionuclide (or heavy metal) concerned, leading to a less mobile species. Various construction methods are employed, including trenching and injection curtains. A rather more novel application are biowalls that utilize microbial processes to effect fixation.

It may also be possible to immobilize a contaminant in situ, rather than waiting for it to migrate into a reactive barrier. Technologies involve, for example, either the injection of grouts to bind contaminants and to reduce soil permeability, or the injection of reactants to precipitate the radionuclides. In the latter case, the reactant may be compressed air or another oxidant to oxidize the iron in the groundwater to ferric oxyhydroxides, which act as a soprtion substrate. Compared with reactive barriers, however, the amount of civil engineering work is likely to be higher.

In certain environments the dispersal of surface contamination in the form of contaminated dust may be of concern. Organic polymers and other binders have been successfully used to suppress dust generation. Reducing erodibility may also be an important first step in establishing a stable vegetation cover. In addition to mechanically stabilizing the topsoil, phytostabilization can establish a well determined cycling of radionuclides in a limited ecosystem, thus preventing their further dispersal.

Plant systems and more specifically aqueous plant systems are actively used to sequester radionuclides from surface waters, including mine drainage and drainage from waste disposal facilities. While such (constructed) wetlands have a certain attraction in requiring relatively little attention once established, the longevity of the solution very much depends on the stability of the ecosystem created. When, for example, the wetland dries out, there is a risk of the radionuclides trapped in the root system or the shoots becoming remobilized. In climates with severe winters, such constructed wetlands may only function during the summer vegetation period.

The baseline techniques for contamination removal from soils and groundwater are excavation and pump and treat, respectively. While excavation removes the contamination, if carried out judiciously, it creates equivalent volumes of material that must be treated and disposed of. In the case of dispersed contamination this may potentially lead to large volumes of waste and hence may not be feasible. The excavated material also may have to be replaced with other, uncontaminated material, for which no resource may be available.

Pump and treat methods are based on the assumption that the source can be completely removed by physical displacement. Experience and predictive modelling of the controlling processes have, however, shown that almost always a residual contamination will remain. This residual contamination will manifest itself as a long concentration 'tail' under a steady pumping regime and may result in rising concentrations when pumping is stopped or interrupted. For this reason, nowadays pump and treat is considered more a (dynamic) containment technique than a real removal technique. This probably applies even if enhanced recovery techniques are applied, such as injection of lixiviant into an aquifer. Likely recovery rates can be estimated taking into account the experience of the oil industry and the mining industry employing in situ leaching [378]. More than half of the original concentrations of the target material (oil, metal value) may remain underground. Leaching techniques have also been applied to the vadose zone, but little information exists on their efficiency for the removal of non-biodegradable materials.

If applied, ex situ extraction techniques will be able to overcome hydrodynamic limitations, by agitating the excavated material as a slurry, etc. Arrangements such as heap leaching have the advantage that the soil or rock structure can be broken down and thus the limitations due to inhomogeneities can be overcome. This may not hold, however, at the microscopic level. Fine grained material may need to be disintegrated considerably to achieve reasonable recovery rates.

Heap leaching, like various other leaching methods, may involve (micro-) biological components. Bioleaching can be actively promoted by stimulating the growth of specialized microorganisms. These microorganisms influence their geochemical environment and may involve the radionuclides in their metabolic activity, which leads to a mobilization of the radionuclides. The main advantage of bioleaching is seen in obviating the use of large quantities of lixiviants, such as strong acids, and the ensuing need to dispose of these lixiviants once the remediation is completed.

The efficiency of any ex situ removal process can be significantly enhanced by appropriate characterization and preceding segregation of contaminated materials. Inorganic and organic contaminations tend to be preferentially associated with the fine fraction of soils, and hence a grain size separation will greatly reduce the amount of material that requires treatment.

It is important to note that any extractive technique applied to a topsoil is likely to impair seriously its functionality and fertility, resulting in a sterile material.

Over the past few decades many processes for removing radionuclides and other metals from solution have been developed. In addition to the traditional precipitation, sorption and ion exchange methods, various biotechnology methods, such as biosorption, have been developed. Although data on large scale application in a remediation context are still lacking, the experience gained from metallurgical applications is promising.

Higher plants can also be used to extract metals, including radionuclides, from soil. The uptake is very plant and radionuclide specific. After the Chernobyl accident, plant uptake by various agricultural species was extensively studied, with a view both to identifying minimal uptake conditions and to maximizing uptake in a phytoextraction context. The data showed that the rate of sequestration would be 1–2% of the total soil reservoir at best per crop cycle, resulting in proportionately long treatment times. Addition of complexing agents to the soil can help mobilize radionuclides such as uranium and facilitate plant uptake, resulting in sequestration rates of up to 5%. The potentially negative effect of such additives on plant growth and biomass yield, however, has to be carefully evaluated. The effective depth of phytoextraction is limited by the penetration depth of the roots, which is typically the tilling depth in agricultural soils. Adequate uses or disposal routes for the harvested contaminated biomass need to be found.

Another technique involving higher plants to sequester radionuclides from solution is rhizofiltration, in which the geochemical environment created by plant roots is utilized to remove radionuclides from solution. A variety of physicochemical processes may actually be at work. Radionuclides that are less mobile under reducing conditions, such as uranium, and that sorb more readily, are more accessible to fixation by rhizofiltration than radionuclides that belong to the alkaline or earth alkaline groups (caesium and strontium).

This report shows that a wide variety of remediation techniques have been developed over the years. Dealing with dispersed radionuclides in the topsoil, however, remains difficult. There are serious trade-offs to be made between the degree of invasiveness and the efficiency of a technique. The sheer volume of material to be treated often precludes the use of certain techniques known to be efficient. Where direct exposure is not a concern, techniques and strategies that prevent further dispersion, and hence limit or interrupt pathways by which the radionuclides would enter the food pathway, seem to be the most appropriate.

Similarly, removal of radionuclides from groundwater tends to be rather inefficient. Containment to prevent further dispersal, for example by reactive barriers, appears to be a better choice.

In all cases where direct exposure is not a concern, monitored natural attenuation should be the baseline against which active remediation measures should be evaluated.

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GLOSSARY

AMD. Acid mine drainage.

ARD. Acid rock drainage.

ATP. Adenosine triphosphate.

CF. Concentration factor for a radionuclide between different environmental compartments, for example soil solution and plants (ratio of Bq/g plant to Bq/mL water or soil solution).

electrowinning. Concentration of metals from a pregnant solution using electrolysis techniques.

NADH. Nicotinamide adenine dinucleotide.

pregnant. Said of metal bearing leach solutions after contact with the ore.

TF. Transfer factor for a radionuclide between different environmental compartments, for example soil and plants. Unit depends on the original (activity) concentration units for the respective compartments.

SRC. Short rotation coppice. Woodland management scheme whereby rapidly growing tree species are cut every few years to harvest the biomass.

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