

**Manual on restoration strategies
for radioactive-contaminated sites**

Restoration Strategies for radioactively contaminated
Sites and their Close Surroundings
RESTRAT – WP6

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Table of contents

1.	INTRODUCTION.....	1
2.	OVERVIEW.....	2
3.	CLASSIFICATION OF CONTAMINATED SITES	4
3.1	LAYERED TAILINGS AND ROCK PILES	5
3.2	FLOODED URANIUM MINES (CONVENTIONAL UNDERGROUND MINING)	7
3.3	FLOODED URANIUM MINES (UNDERGROUND <i>IN-SITU</i> LEACHING)	9
3.4	CONTAMINATED RIVERS.....	11
3.5	CONTAMINATED ESTUARIES AND TIDAL WATERS	13
3.6	SURFACE DISPOSAL SITES	15
3.7	FLOODED OPEN PITS.....	17
3.8	OPEN BASINS	19
3.9	CONTAMINATED LAKES.....	21
3.10	REFERENCES	23
4.	RESTORATION TECHNIQUES.....	24
4.1	DESCRIPTION	24
4.2	APPLICABILITY	29
4.3	PERFORMANCE	30
4.4	COSTS.....	32
4.5	WORK RATES (LABOUR VOLUME).....	34
4.6	REFERENCES	35
5.	METHODOLOGIES.....	38
5.1	ASSESSMENT OF RADIOLOGICAL AND NON-RADIOLOGICAL HEALTH EFFECTS	38
5.2	PHYSICO-CHEMICAL SOURCE TERM ASSESSMENT	40
5.3	RADIOLOGICAL IMPACT ASSESSMENT	44
5.4	ASSESSMENT OF ECONOMIC COSTS.....	51
5.5	ASSESSMENT OF SOCIAL FACTORS.....	53
5.6	RANKING OF RESTORATION OPTIONS.....	57
5.7	REFERENCES	65
6.	MODELLING APPROACHES AND TOOLS	67
6.1	IMPACT ASSESSMENT MODEL	67
6.2	CHEMICAL SPECIATION	75
6.3	MULTI-ATTRIBUTE UTILITY ANALYSIS	84
6.4	REFERENCES	91
7.	RESULTS FOR EXAMPLE SITES	93
7.1	RANKING OF RESTORATION OPTIONS FOR EXAMPLE SITES	93
7.2	CONCLUSIONS	109
7.3	REFERENCES	113

List of figures

<i>FIGURE 1 : SUB-OPTIMISATION OF OVERALL HEALTH PROTECTION WHERE ONLY RADIOLOGICAL PROTECTION FACTORS ARE INCLUDED IN THE OPTIMISATION PROCESS.</i>	<i>55</i>
<i>FIGURE 2 : OPTIMISATION OF OVERALL HEALTH PROTECTION WHERE RADIOLOGICAL AND NON-RADIOLOGICAL PROTECTION FACTORS ARE INCLUDED SIMULTANEOUSLY IN THE OPTIMISATION PROCESS.</i>	<i>56</i>
<i>FIGURE 3 : BENEFIT COMPONENTS, B, OF CLEAN-UP OPERATIONS.</i>	<i>57</i>
<i>FIGURE 4 : NET BENEFIT OF DIFFERENT REMEDIATION OPTIONS AND THE CORRESPONDING RESIDUAL COLLECTIVE DOSE, S, AFTER CLEAN-UP, FOR THE FIVE JUSTIFIED OPTIONS.</i>	<i>58</i>
<i>FIGURE 5 : ATTRIBUTE HIERARCHY FOR RESTORATION OF A CONTAMINATED SITE.</i>	<i>61</i>
<i>FIGURE 6 : EXAMPLES OF UTILITY FUNCTIONS OF THE DECREASING TYPE; RISK NEUTRAL (LEFT FIGURE), RISK AVERSE (MIDDLE FIGURE) AND RISK PRONE (RIGHT FIGURE) UTILITY FUNCTIONS.</i>	<i>62</i>
<i>FIGURE 7 : GENERIC COMPARTMENTAL SCHEME.</i>	<i>67</i>
<i>FIGURE 8 : CHEMICAL SPECIATION MODELLING FOR ITERATIVE REFINEMENT OF ANALYTICAL DATA SETS</i>	<i>79</i>
<i>FIGURE 9 : URANIUM SPECIATION AS A FUNCTION OF EH, PH AND P(CO₂) FOR THE RANSTAD TAILING SITE (TAILING LAYER).</i>	<i>82</i>
<i>FIGURE 10 : DISTRIBUTION FUNCTIONS FOR WEIGHTING FACTORS FOR MAJOR ATTRIBUTES.</i>	<i>87</i>
<i>FIGURE 11 : DISTRIBUTION FUNCTIONS FOR WEIGHTING FACTORS FOR HEALTH SUB-ATTRIBUTES.</i>	<i>87</i>
<i>FIGURE 12 : DISTRIBUTION FUNCTIONS FOR WEIGHTING FACTORS FOR ECONOMIC SUB-ATTRIBUTES.</i>	<i>87</i>
<i>FIGURE 13 : DISTRIBUTION FUNCTIONS FOR WEIGHTING FACTORS FOR SOCIAL SUB-ATTRIBUTES.</i>	<i>88</i>
<i>FIGURE 14 : THE EXCEL SPREADSHEET WITH THE COMPLETE MULTIATTRIBUTE ANALYSIS MODEL.</i>	<i>89</i>
<i>FIGURE 15 : DISTRIBUTION OF THE SCORES A - E AFTER 12,000 SIMULATIONS.</i>	<i>90</i>
<i>FIGURE 16 : COMPARTMENT SCHEME FOR THE MOLSE NETE RIVER SITE.</i>	<i>94</i>
<i>FIGURE 17 : OVERALL SCORES FOR DIFFERENT REMEDIATION STRATEGIES FOR THE MOLSE NETE RIVER SITE.</i>	<i>96</i>
<i>FIGURE 18 : COMPARTMENT SCHEME FOR THE DRIGG SITE.</i>	<i>97</i>
<i>FIGURE 19 : OVERALL SCORES FOR DIFFERENT REMEDIATION STRATEGIES FOR THE DRIGG SITE.</i>	<i>99</i>
<i>FIGURE 20 : COMPARTMENT SCHEME FOR THE RAVENGLASS ESTUARY.</i>	<i>100</i>
<i>FIGURE 21 : OVERALL SCORES FOR DIFFERENT REMEDIATION STRATEGIES FOR THE RAVENGLASS SITE ("x 10" MEANS THAT THE INDICATED VALUE IS 10 TIMES HIGHER THAN THE ACTUAL ONE).</i>	<i>102</i>
<i>FIGURE 22 : COMPARTMENT SCHEME FOR THE RANSTAD TAILING SITE.</i>	<i>103</i>
<i>FIGURE 23 : OVERALL SCORES FOR DIFFERENT REMEDIATION STRATEGIES FOR THE RANSTAD SITE ("x 100" MEANS THAT THE INDICATED VALUE IS 100 TIMES HIGHER THAN THE ACTUAL ONE)).</i>	<i>105</i>
<i>FIGURE 24 : COMPARTMENT SCHEME FOR LAKE TRANEBÄRSSJÖN.</i>	<i>106</i>
<i>FIGURE 25 : OVERALL SCORES FOR DIFFERENT REMEDIATION STRATEGIES FOR THE LAKE TRANEBÄRSSJÖN SITE ("x 10⁴" AND "x 10³" MEANS THAT THE INDICATED VALUE IS RESPECTIVELY 10⁴ AND 10³ TIMES HIGHER THAN THE ACTUAL ONE).</i>	<i>108</i>

List of tables

TABLE 1 : SUITABILITY OF RESTORATION TECHNIQUES FOR TREATING CONTAMINATED MEDIA.....	29
TABLE 2 : REPORTED EFFECTIVENESS OF RESTORATION TECHNIQUES IN TREATING RADIONUCLIDES	30
TABLE 3 : THE PERFORMANCE OF REMEDIATION TECHNIQUES	31
TABLE 4 : THE COSTS OF REMEDIATION TECHNIQUES	33
TABLE 5 : THE WORK RATES (RESTORATION WORKERS) OF REMEDIATION TECHNIQUES	34
TABLE 6 : URANIUM SPECIATION COMPUTED WITH EQ6 AT THERMODYNAMIC EQUILIBRIUM (TAILING LAYER OF RANSTAD TAILING SITE).....	83
TABLE 7 : ATTRIBUTE VALUES FOR DIFFERENT RESTORATION STRATEGIES AT A HYPOTHETICAL SITE.....	84
TABLE 8 : PARAMETERS USED IN THE NORMAL DISTRIBUTIONS FOR THE SEVEN ATTRIBUTES AND FIVE REMEDIATION OPTIONS.....	85
TABLE 9 : UTILITY FUNCTIONS FOR THE DIFFERENT ATTRIBUTES FOR RESTORATION STRATEGIES AT A HYPOTHETICAL SITE.	85
TABLE 10 : WEIGHTING FACTORS FOR MAJOR ATTRIBUTES AND HEALTH, ECONOMIC AND SOCIAL ATTRIBUTES.....	86
TABLE 11: ECONOMIC AND RADIOLOGICAL DATA (FOR INTEGRATION TIMES OF 100 AND 500 YEARS) FOR DIFFERENT RESTORATION STRATEGIES AT THE MOLSE NETE RIVER SITE. BEST-ESTIMATE VALUES.....	95
TABLE 12 : UTILITY VALUES FOR BEST-ESTIMATE VALUES OF THE ATTRIBUTES FOR EACH OF THE REMEDIATION OPTIONS AT THE MOLSE NETE RIVER SITE.	95
TABLE 13: WEIGHTING FACTORS FOR ATTRIBUTES AND SUB-ATTRIBUTES APPLIED IN THE OPTIMISATION OF REMEDIATION OF THE MOLSE NETE SITE. BEST-ESTIMATE VALUES.	96
TABLE 14 : ECONOMIC AND RADIOLOGICAL DATA (FOR INTEGRATION TIMES OF 100 AND 500 YEARS) FOR DIFFERENT RESTORATION STRATEGIES AT THE DRIGG SITE. BEST-ESTIMATE VALUES.	98
TABLE 15 : UTILITY VALUES FOR BEST-ESTIMATE VALUES OF THE ATTRIBUTES FOR EACH OF THE REMEDIATION OPTIONS AT THE DRIGG SITE.....	98
TABLE 16 : WEIGHTING FACTORS FOR ATTRIBUTES AND SUB-ATTRIBUTES APPLIED IN THE OPTIMISATION OF REMEDIATION OF THE DRIGG SITE. BEST-ESTIMATE VALUES.	99
TABLE 17 : ECONOMIC AND RADIOLOGICAL DATA (FOR INTEGRATION TIMES OF 100 AND 500 YEARS) FOR DIFFERENT RESTORATION STRATEGIES AT THE RAVENGLASS SITE. BEST-ESTIMATE VALUES.....	101
TABLE 18 : UTILITY VALUES FOR BEST-ESTIMATE VALUES OF THE ATTRIBUTES FOR EACH OF THE REMEDIATION OPTIONS AT THE RAVENGLASS SITE.	101
TABLE 19 : WEIGHTING FACTORS FOR ATTRIBUTES AND SUB-ATTRIBUTES APPLIED IN THE OPTIMISATION OF REMEDIATION OF THE RAVENGLASS SITE. BEST-ESTIMATE VALUES.....	101
TABLE 20 : ECONOMIC AND RADIOLOGICAL DATA (FOR INTEGRATION TIMES OF 100 AND 500 YEARS) FOR DIFFERENT RESTORATION STRATEGIES AT THE RANSTAD TAILING SITE. BEST-ESTIMATE VALUES.....	104
TABLE 21 : UTILITY VALUES FOR BEST-ESTIMATE VALUES OF THE ATTRIBUTES FOR EACH OF THE REMEDIATION OPTIONS AT THE RANSTAD TAILING SITE.	104
TABLE 22 : WEIGHTING FACTORS FOR ATTRIBUTES AND SUB-ATTRIBUTES APPLIED IN THE OPTIMISATION OF REMEDIATION OF THE RANSTAD TAILING SITE. BEST-ESTIMATE VALUES.....	104
TABLE 23 : ECONOMIC AND RADIOLOGICAL DATA (FOR INTEGRATION TIMES OF 100 AND 500 YEARS) FOR DIFFERENT RESTORATION STRATEGIES AT THE LAKE TRANEBÄRSSJÖN SITE. BEST-ESTIMATE VALUES.....	107
TABLE 24 : UTILITY VALUES FOR BEST-ESTIMATE VALUES OF THE ATTRIBUTES FOR EACH OF THE REMEDIATION OPTIONS AT THE LAKE TRANEBÄRSSJÖN SITE.....	107
TABLE 25 : WEIGHTING FACTORS FOR ATTRIBUTES AND SUB-ATTRIBUTES APPLIED IN THE OPTIMISATION OF REMEDIATION OF THE LAKE TRANEBÄRSSJÖN SITE. BEST-ESTIMATE VALUES.....	107
TABLE 26 : SUMMARY OF THE RANKING OF REMEDIATION OPTIONS FOR THE EXAMPLE SITES. THE INTEGRATION TIME FOR THE COLLECTIVE DOSE IS 100 YEARS.	110
TABLE 27 : SUMMARY OF THE RANKING OF REMEDIATION OPTIONS FOR THE EXAMPLE SITES. THE INTEGRATION TIME FOR THE COLLECTIVE DOSE IS 500 YEARS	111
TABLE 28 : SUMMARY OF JUSTIFICATION/OPTIMISATION OF REMEDIAL MEASURES AT FIVE EUROPEAN EXAMPLE SITES AND COMPLIANCE WITH IAEA CRITERIA FOR CLEAN-UP.....	112

1. Introduction

This report is submitted as Technical Deliverable No. 14 (TD 14) against the requirements of the RESTRAT (Restoration Strategies for radioactively contaminated Sites and their Close Surroundings) Project.

The RESTRAT project is co-funded by the European Commission under the fourth framework of the Nuclear Fission Safety Programme. Its overall objective concerns the development of a methodology for ranking restoration techniques as a function of contamination and site characteristics.

It is based on an analysis of existing remediation methodologies and contaminated sites and is structured in the following steps:

1. characterisation of relevant contaminated sites;
2. identification and characterisation of relevant restoration techniques;
3. assessment of the radiological impact;
4. development and application of a methodology for the ranking and selection of restoration options;
5. formulation of general conclusions and development of a manual.

This report concerns the manual in which the ranking methodology is explained and applied to example sites. The main attributes that are to be allowed for in the ranking procedure, are extensively discussed and methods for their quantification indicated. Moreover, where appropriate, generic values are given.

When applying the methodology to the example sites, attributes are valued in a site-specific way, based on the site descriptions available in other technical deliverables (TD9 up to 13).

The results for the example sites are shown and conclusions drawn.

The manual is addressed to specialists in the various disciplines, which are involved in the decision-aiding process with respect to the selection of remediation options for radio-active contaminated sites. Those disciplines include:

- radiation protection, radiology;
- environmental sciences (chemistry, physics...);
- economy;
- sociology.

2. Overview

In this manual the methodology for ranking restoration options is explained and applied to example sites that are representative for major categories of contaminated sites.

In chapter 3 classes of contaminated sites that are considered in the RESTRAT study are defined and example sites indicated. Only mid-sized sites that are contaminated as a consequence of local events or practices are taken into account.

Sites contaminated through terrestrial sources (related mainly to mining activities and waste disposal) and through aquatic sources (rivers, lakes and estuaries) are considered.

Potentially relevant techniques for restoration of such contaminated sites are indicated in chapter 4. They may be divided into following classes: physical removal of the contamination, possibly followed by separation of the most contaminated fraction, physical containment of the contaminated medium and immobilization of the contaminants. The characteristics of those techniques playing a role in the ranking procedure are listed and their normalised values (ranges) indicated as they have been determined from a literature review.

For the ranking of the restoration options a multi-attribute utility (MAU) type of analysis has been chosen with a view to the various categories of attributes that are being taken into account:

- radiological health detriment;
- economic costs;
- social factors.

The methods of assessing the attributes are explained in chapter 5.

Collective doses to the population and to the restoration workers constitute the radiological health attributes.

The latter doses could be assessed straightforward, for the assessment of the doses to the population a more complicated impact model had to be built for each site considered.

Among the economic costs, those directly related to the remediation operations and to the disposal of the waste generated, are the most important ones. They have been assessed in a generic way from the extensive literature review, indicated in chapter 4. Other costs such as those related to monitoring and survey have been evaluated in a site-specific way.

Social factors such as reassurance and disturbance of the population concerned were very difficult to be quantified directly. They were replaced by objective measures that were easier to be quantified such as residual doses, activities left on site, volumes of waste to be transported.

Each of the restoration options are then given a score determining utility functions for converting attribute values into utility values and assigning weighting factors to the attributes.

In chapter 6 the models and software applied for assessing the collective dose to the population and for ranking the remediation options (MAU analysis) are explained. Also the chemical speciation model applied for determining the distribution coefficients in the impact model in a more precise and reliable way is included and a method for iterative refinement of the parameter values added. For the assessment of the radiological impact and for the MAU analysis of the remediation options, uncertainty and sensitivity aspects are discussed.

In chapter 7 the results of the application of the methodologies to the example sites are shown and briefly commented. Generic conclusions are also formulated.

Two CD-ROMs (Brendler, 1999; Hedemann Jensen, 1999) are added to this manual.

The former (Brendler *et al.*, 1999) contains the software of the impact assessment models developed for the example sites on the basis of the BIOPATH/PRISM codes, as well as the chemical speciation code MINTEQA2, which may be or may not be included in the impact assessment modelling. The input parameter values for the impact assessment models and the database necessary to use the chemical speciation code are also present on the CD-ROM.

The latter (Hedemann Jensen, 1999) contains the Crystal Ball software used for the multi-attribute utility analyse of the remediation options at the example sites/case. On this CD-ROM also the database is included in which literature values, characterizing the performance of the restoration techniques, have been collected.

References

Brendler V, Stiglund Y and Nordlinder S. (1999) Risk Assessment Model for Use in Site Restoration. Software and User Instructions. (CD-ROM). RESTRAT - TD 7. Forsschungszentrum Rossendorf, Dresden, Germany.

Hedemann Jensen P. (1999) Crystal Ball software and RESTRAT.mdb database. (CD-ROM). Risoe N.L., Denmark.

3. Classification of contaminated sites

The focus of the RESTRAT project is put onto an already limited subset of contaminated land, those stretching over mid-size ranged areas with typical diameters between several tens to several thousands of metres. Larger areas, e.g. the landscapes contaminated by the Chernobyl accident, or very small spots such as a single building are not in the scope of this project. Also, severe contaminations resulting from an accident are outside this scope. From a literature screening of radioactively contaminated sites of the aforementioned type (e.g. to be found in Vandenhove *et al.*, 1999; International Atomic Energy Agency, 1996; European Commission, 1994; Merkel *et al.*, 1995; Merkel and Helling, 1998), and from own visits to several of them, various major classes could be identified. Of course, the features used for this classification can also be defined in a different way. Thus, the categories utilized in this approach are not the only alternative, but at least they proved to be useful for the purpose of establishing a generic methodology for risk assessment.

The classes are characterised by the phenomenological description of the site and the processes most important for the fate of the contaminants, such as the basic chemistry, generation, internal transport, outflow, binding etc. These processes are in turn strongly influenced by the outer shape and inner structure of a given site, and also of its geochemical constituents. Typical exposure pathways, examples and generalized compartment schemes for each class are given in the following sections. All examples that were dealt with in detail as example sites in the RESTRAT project are printed in bold. Generic compartment schemes are indicating the main compartments typical for a given site category, their mutual relationship and the flow of matter between them, including the contaminant. Also water entry points and paths through which the contaminant may leave the system are shown. In all compartment scheme figures, the source term compartments are indicated by a bold frame, optional compartments by a dashed frame.

It is worth mentioning that on several sites that radionuclides not only acted as contaminants but chemical-toxicological contaminants also posed severe problems. Thus a combined risk assessment for both types of hazard would be best suited for such places. The major contaminants can be grouped into four generic classes, which are listed below along with some typical representatives for site restoration.

- I actinides originating either from original ore or from nuclear waste: ^{238}Pu , ^{239}Pu , ^{240}Pu , ^{241}Pu , ^{241}Am , ^{237}Np , ^{234}U , ^{235}U , ^{238}U , ^{230}Th , ^{226}Ra , ^{228}Ra , ^{222}Rn , ^{210}Pb , ^{231}Pa , ^{227}Ac ;
- II radioisotopes of other elements, originating from low- or intermediate level radioactive waste: ^{14}C , ^{36}Cl , ^{63}Ni , ^{90}Sr , ^{93}Zr , ^{94}Nb , ^{99}Tc , ^{107}Pd , ^{126}Sn , ^{129}I , ^{135}Cs , ^{137}Cs ;
- III non-radiating toxic heavy metals and metalloids, originating from ore processing activities (enriched trace metals and processing agents) and to a lesser degree also from municipal or industrial waste: Hg, Cr, Pb, Cu, Cd, As, Bi, Sb;
- IV toxic / carcinogenic organic compounds, originating from municipal or industrial waste: PCBs, dioxins.

In the RESTRAT project only contaminants of classes I and II are taken into account in the risk assessments.

3.1 Layered tailings and rock piles

Phenomenological Description

The sites concerned mainly originate from uranium ore mining and milling activities, but also coal mining leading to the dumping of slags and ashes may be important. The size of a pile can be in the range from $10 \times 10 \text{ m}^2$ till $100 \times 100 \text{ m}^2$ with a shape as a conical heap, a truncated cone heap, a heap on a valley side, or a partial or complete fill of a valley. Usually there is no sealing under these piles. Its material can either consist of original rocks, of processed ore, or of mixtures of both. Additional material like industrial or municipal waste may also be found. Either due to pure technological reasons, or by purpose, most of this sites exhibit a horizontally layered structure. Some of the sites have one or more cover layers made of different material (plastics, bentonite, ...), and there may also be vegetation on top of the site, either artificially engineered or self-developed by nature. Water outlets on the bottom of such sites may be engineered, but only few of them are connected to a water purification facility.

Main Processes

There is horizontal and vertical flow of water, which enables chemical reactions such as leaching processes or the weathering of rocks and minerals. Other chemical reactions may be started due to microbiological processes, mainly oxidation of minerals and other components of the heap, but also the establishment of biofilms on mineral surfaces. The most complex chemistry will be observed in cases where also organic compounds (as part of litter) is incorporated into the heap, the degradation processes will heavily influence the chemical environment for the other contaminants. Biological activities will result in the emanation of gases such as methane, ammonia, hydrogen sulphide or carbon dioxide. Due to the inner structure of a heap parts of it may not be reached by substantial amounts of water and therefore preserve its initial physico-chemical state for long time periods, up to decades. If the heap components are of a coarse form, also atmospheric gases may penetrate it, accelerating oxidation, leaching and weathering processes.

Contaminants and Exposure Pathways

Contaminants may come from various origins; from original ore (radionuclides of class I: uranium, thorium, radium, radon), from processing activities (metals of class III) or from added litter (compounds of class IV)

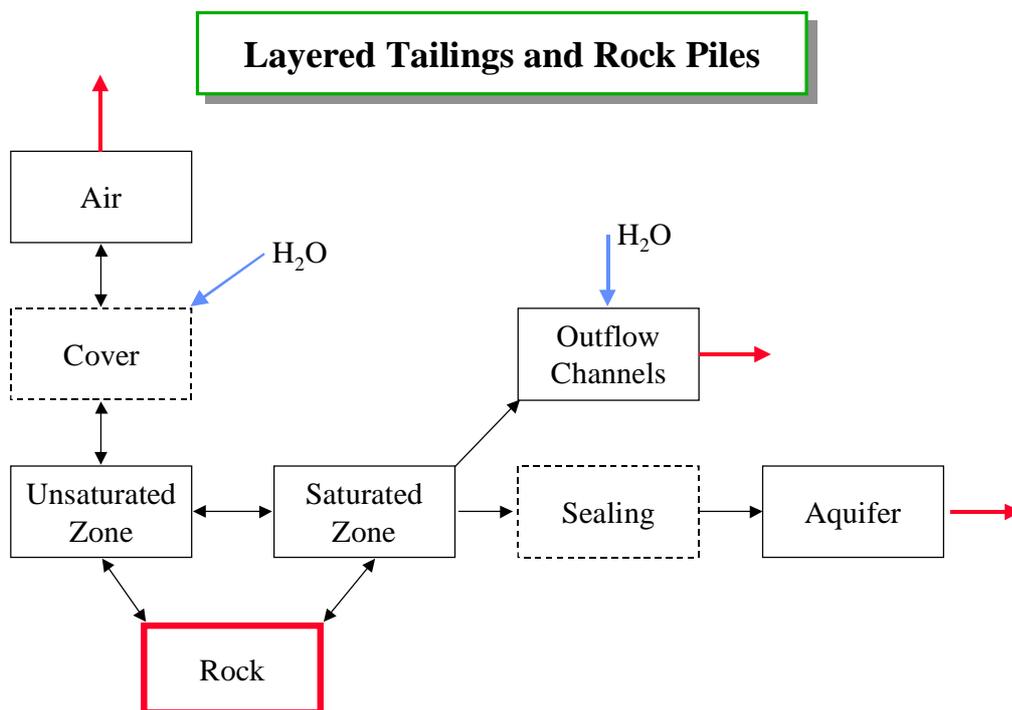
There are several possible exposure pathways. The most important ones are through the use of contaminated aquifers or rivers as sources for drinking water or for irrigation of crops and watering of livestock.

Contamination of the air through resuspension of dust or exhalation of radon may also occur, leading to exposure through inhalation or to contamination of the surroundings, creating secondary sources. These can have either direct (inhalation) or indirect (through crops and livestock) radiological consequences.

Direct radiation from the rock pile or by material from the rock pile used for construction of houses and roads must also be considered.

Examples

A large number of rock piles and mill tailings originating from former uranium mining and milling activities can be found all over Europe. Examples are Schlema / Alberode, Ronneburg or Lengenfeld in Germany, Freital / Gittersee in Germany (here also associated with coal mining), **Ranstad in Sweden**, Sillamäe in Estonia, Pecs in Hungary, Kowary, Kletno and Radoniow in Lower Silesia / Poland, Zirovski in Slovenia, Salamanca, Andujar and Badajoz in Spain.



Generic compartment scheme for layered tailings and rock piles.

3.2 Flooded uranium mines (Conventional underground mining)

Phenomenological Description

The sites concerned consist of a whole underground mine system, with important parts being galleries, shafts, weathering shafts, bore holes - in summary a strongly disturbed rock area. Crushed rock is partially used as backfill. Thus in case of several vertically separated aquifers there are both natural fractures and artificial connections between the layers.

Main Processes

There are strong gradients of temperature, pressure and redox state down the mine (some of them reach depths of more than 1000 metres). When the water rises, it will displace the air (and thus oxygen) and transform the chemical environment to reducing conditions. Due to the higher temperatures observed in mines all chemical reactions will exhibit faster kinetics than usually observed on the surface. Furthermore, biological material may either act as catalyst or even actively process matter. Also, the high hydrostatic pressure that develops during the flooding may influence some chemical processes in an unusual way. Finally, wood degradation products may constitute another important chemical component, namely as complexing and reducing agent.

Water may enter a mine from inner aquifers or from the surface, or from combinations thereof. The state of these infiltrating waters can differ in ion content, pH and redox potential Eh. Gradients in density (due to differing temperature and salt load) may give rise to water circulation in addition to the main water flow from outside into the abandoned mine. In case of several vertically separated aquifers the water exchange between them will occur through flow over fractures, and by diffusion through the separating aquitard (clay horizons etc).

Contaminants and Exposure Pathways

The main radioactive contaminants are of class I (uranium, thorium, radium, radon and their daughter products). They are accompanied by heavy metals of class III, such as lead, cadmium, chromium, or copper, but also arsenic.

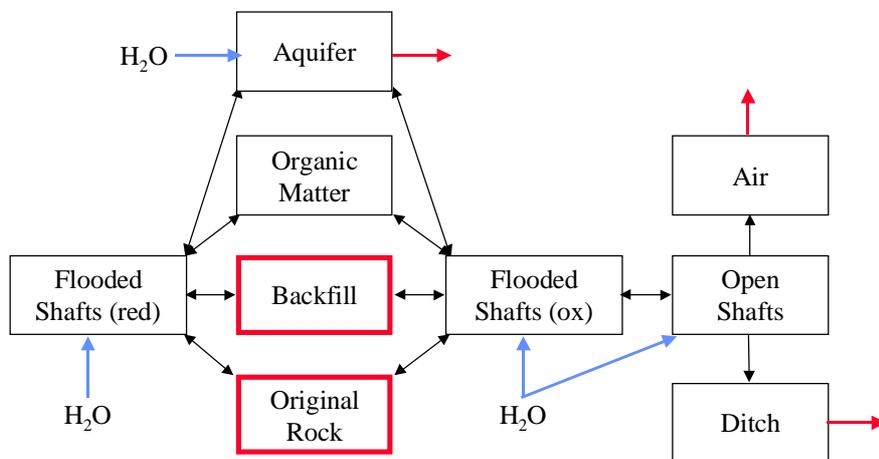
The main exposure pathway is through the use of the contaminated aquifer or river as a source for drinking water. These waters may also be used for irrigation of crops or watering of livestock, creating additional exposure pathways.

An especially critical situation is observed in the transition state between the relinquishment of the mine and the end of the flooding. During that period part of the mine is still open to the atmosphere, but the devices for weathering are already shut down. Hence, there is no longer any forced air circulation, e.g. the exhalation of radon and radioactivity-carrying aerosols through the open ends of shafts becomes uncontrolled.

Examples

Examples can be found in the former uranium mining districts in Saxony and Thuringia (Germany): the Pöhla mine, the Hartenstein mine, or in Bulgaria (mines near Eleshnitsa)

Flooded Conventional Underground Uranium Mines



Generic compartment scheme for flooded conventional underground uranium mines.

3.3 Flooded uranium mines (Underground *in-situ* leaching)

Phenomenological Description

Contrary to the conventional mines as described in the previous section, here even the mechanically undisturbed rock may be influenced because of the special exploitation technique - *in-situ* leaching (ISL) of the rock by diluted sulphuric acid, sometimes after crushing rock blocks to increase permeability. There are cases both with a conventional mine building, and cases where the sulphuric acid is directly injected into the rock and recovered by pumping stations, leaving the situation underground in an even less controlled state. After stopping the mining activities, there will still be a tremendous amount of acid inside the rock.

Main Processes

Usually the solution is pressed through the mine, pumped to the surface, processed to extract most of the uranium, and then injected again into the rock. The dilution by infiltrating water from outside will only take effect in the larger open areas inside the mine (assigned to the compartment "Crushed Rock"), to remove the acid from the micro pores inside the rock (assigned to the compartment "Leached Rock") will take decades. Simply stopping operation will bring the mine, its acid circulation together with the intruding water into a state that can hardly be controlled.

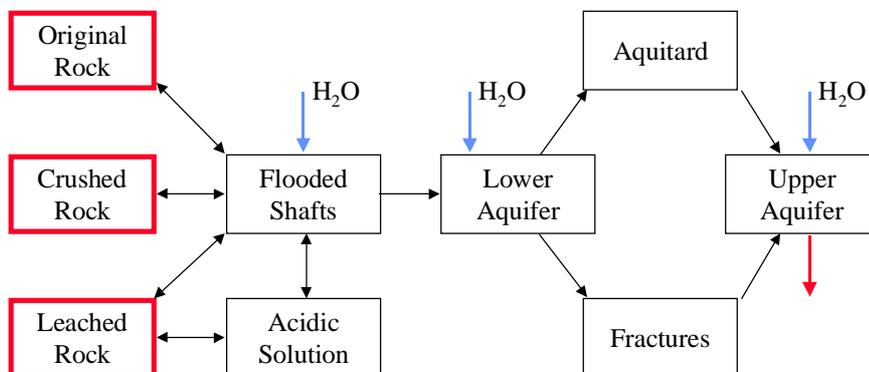
Contaminants and Exposure Pathways

The contaminants and exposure pathways are similar to the ones described in the previous section. Additionally, at least in the first time after abandoning such a mine the water itself may be hazardous because of its acid content.

Examples

This peculiar site category is rather seldom found, major representatives are the mine in Königstein / Saxony in Germany and the Stra deposit in North Bohemia / Czech Republic.

Flooded Uranium Mines (*in-situ* leaching)



Generic compartment scheme for flooded uranium mines (in-situ leaching).

3.4 Contaminated rivers

Phenomenological Description

The sites concerned are rivers or river systems that are contaminated by effluents being discharged continuously from nuclear power plants, civilian and military production facilities of nuclear material, nuclear research centres, deposits or disposals on a routine basis, or by accidental spill-over from nearby reservoirs. A typical phenomenon is the establishing of secondary source terms in the sediments of the river bed.

Main Processes

The horizontal flow rate in the river can span a wide range, also width and depth of the rivers. It starts with small creeks and brooks and may go on to huge streams such as the rivers Ob or Jenissei in Siberia. The lower the flow rate is the more sedimentation processes become important, whereas in turbulent zones resuspension may dominate. Dilution effects occur on each confluence with other rivers. Often the contamination in a tributary is restricted to its length, after joining a bigger stream the contamination level will often decrease below regulatory limits.

The main chemical reactions will be oxidation in the aqueous phase and reduction in the deeper zones of the sediments, creating steep redox potential gradients in vertical directions. This phenomenon can be partially pushed back by bio-turbation activities. Whereas in the oxidation zone surface reactions will dominate (physiosorption, surface complexes, surface precipitation and establishing of coatings on the river bed sediments) in the deeper zones precipitation and co-precipitation may also become important for various radionuclides, because often the lower oxidation states are less soluble.

Contaminants and Exposure Pathways

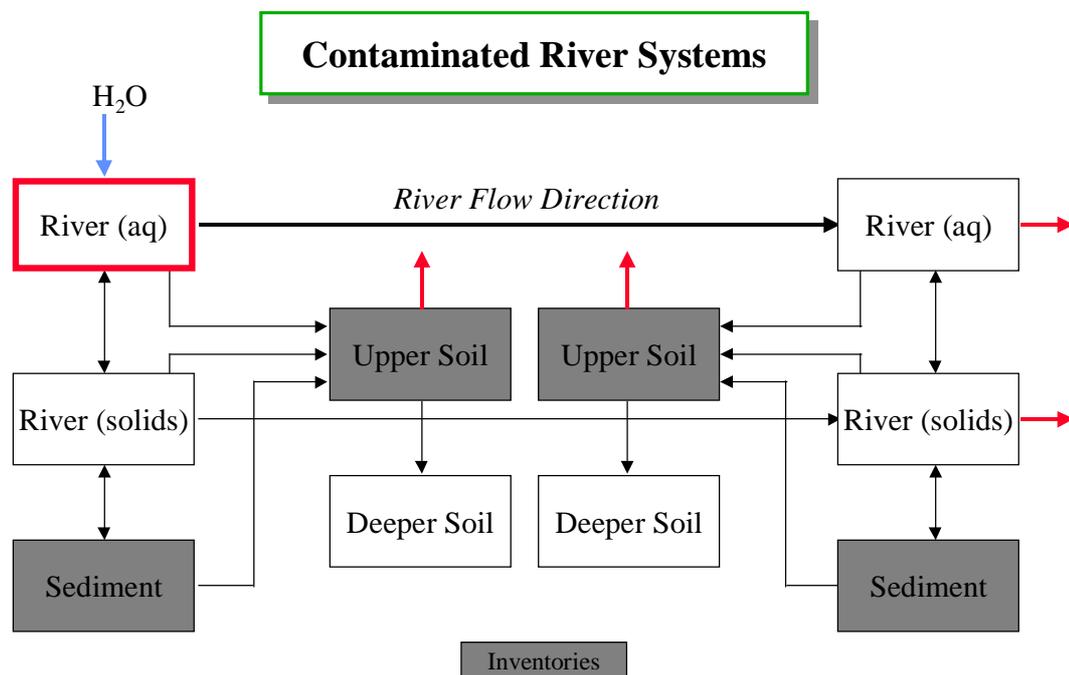
The main contaminants depend on the site and the use of the river.

Their pathway to humans can be directly by using the river as drinking water supply or by eating fish. Indirect ways are the use of water for irrigation of crops or for watering livestock. There are also sites where the river is (regularly or irregularly) dredged, mainly to prevent inundations or enable shipping. Then the dredged sediment may be applied onto nearby agricultural soil which may start other exposure pathways through contaminated crops, meat and milk.

Places with high contamination levels of gamma emitting radionuclides may give rise to important external irradiation of humans. For plutonium and americium, important exposure pathways are also through inhalation.

Examples

Only slightly contaminated sites are part of the rivers **Molse Nete in Belgium** and Bankloop, also in Belgium. Much higher hazard levels are observed along various Russian rivers, such as the Techa.



Generic compartment scheme for contaminated river systems.

3.5 Contaminated estuaries and tidal waters

Phenomenological Description

The sites concerned comprise the whole area of a river's mouth into the sea, starting from the inner estuary with pure river water, including the mixing zone between the river water and tidal waters from the sea (this area can be subdivided into several phases), and stretching into the sea with salty water only slightly effected by the inflow from the river. The actual shape and area can widely differ, depending on geography, river size (width, flow rate, flow volume), and tidal behaviour. The source term can be through discharges in the river or in the open sea, with a tendency to built up secondary source terms on site.

Main Processes

The main phenomena, besides the simple mixing of river water and sea water with the accompanying dilution effects, are the changes in pH, ionic strength and redox potential from the river till the open sea. In the inner zone, the main processes are similar to the ones described for a contaminated river system, like the sedimentation and establishing of a redox gradient. The transition zone adds larger disturbances to the system due to tidal fluctuations. Depending on the flow direction, sedimentation and simultaneously resuspension and even erosion may be observed. But in general the horizontal movement of water bodies should be the dominant flow. In the outer zone, the high salt content may influence the behaviour of some radionuclides.

Contaminants and Exposure Pathways

The main contaminants are of course site-dependent, belonging to the classes I and II.

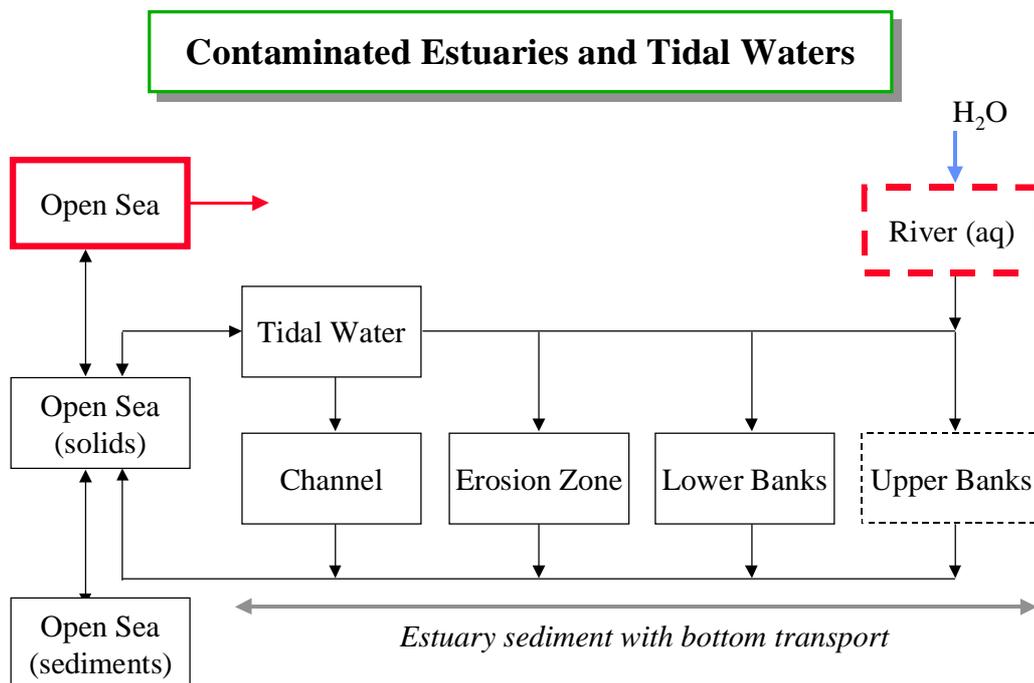
Man can become exposed through the ingestion of fish or shellfish from the estuary waters. For the inner zone one, the utilisation of water for irrigation (closer to the sea water will be too salty for such purposes), gives rise to additional exposure pathways through the ingestion of crops, meat (lamb, beef) and milk.

External exposure and inhalation (in the case of exposure through plutonium, americium or radon) will be important for the sites that are attractive for recreational activities.

Examples

Such sites can be found at the **Ravenglass Estuary / Great Britain** on the Irish Sea south of Sellafield, the phosphate industry in Punta del Sebo / Spain.

The mouths of Ob / Jenissei and the Kara Sea may also be included as example sites.



Generic compartment scheme for contaminated estuaries and tidal waters.

3.6 Surface disposal sites

Phenomenological Description

Sites that may be of concern are those that contain the disposed material in trenches or shallow pits, but also directly on the surface, sheltered by some cover (clay, concrete, plastic membranes). Some of the older parts of such disposals may not have a sealing to the ground and only inadequate covers. The material containing radioactivity can be very heterogeneous, e.g. containing biodegradable materials such as paper and clothes, as well as redundant plant items and steel work, soil, ion exchange resins, concrete debris, and some plastics and rubber. Generally it will be in one or more separate enclosures, like steel containers or concrete ingots. Ditches around the disposal places will collect outgoing water. Additionally, a subsurface drainage system may be installed.

Main Processes

Due to the heterogeneity of the material, the physico-chemical processes can cover many areas. It ranges from leaching processes due to infiltrating water over (bio)degradation of organic material, corrosion of metal containers up to surface reactions and precipitation. The redox state is variable, depending on the oxygen permeability of the covers and on the composition of the disposed material.

Contaminants and Exposure Pathways

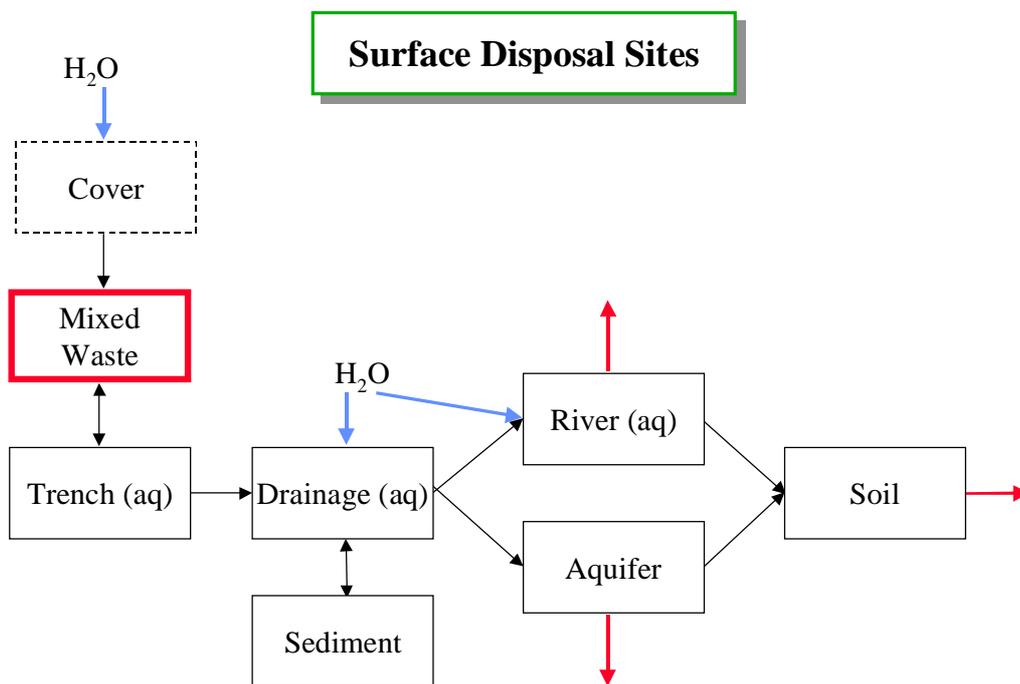
At these sites the inventory shows a broad variety of radionuclides when compared with the other site categories. They may include all radionuclides of classes I and II.

Exposure pathways are transport with the groundwater and river systems to places where the water is used for drinking water preparation. Also contaminated water may be used for irrigation of crops or watering of livestock, so the contaminants can reach humans through the consumption of crops, meat and milk.

Other exposure pathways that may be important are external irradiation and inhalation (this last one only when substantial amounts of plutonium, americium and radium, producing radon, are present in the surface soil).

Example

One example is the **Drigg Site / Great Britain** south of Sellafield for low-level radioactive waste, at least the trenches where originally, waste has been disposed of without an adequate containment or protection of the environment.



Generic compartment scheme for surface disposal sites.

3.7 Flooded open pits

Phenomenological Description

This site category mainly comprises abandoned open-cast uranium mines. So the shape is of a large, irregularly formed basin, filled with water. The size of such artificial lakes can reach several square kilometres, with depths up to 100 metres. Some parts of the bottom and sides of the pit may be covered by backfill material from the mining activities, but usually there is no sealing to the ground. It is also possible that part of the open basin had been used as a landfill / dumping site.

Main Processes

The inflow of water (from cut aquifers or from a surface river) will promote several processes. The primary one is the leaching of the remained ore layers, but also of the backfill. The transition of sulphides and other minerals most often results in an acidification of the whole pit, or of the created lake. This is accompanied by the exclusion of oxygen from the deeper part of the lake. The oxygen supplied by the inflowing water is rapidly consumed by oxidation processes of minerals in reduced form, often catalysed by microbial activity. The transport and diffusion of new oxygen is rather slow compared to these processes. The sheer size of the site will keep the lake and its immediate surrounding rocks in thermodynamic disequilibrium for long time periods. This is especially valid for the formation of new, metastable, mineral phases, which only very slowly may transform into their thermodynamic stable counterparts.

Contaminants and Exposure Pathways

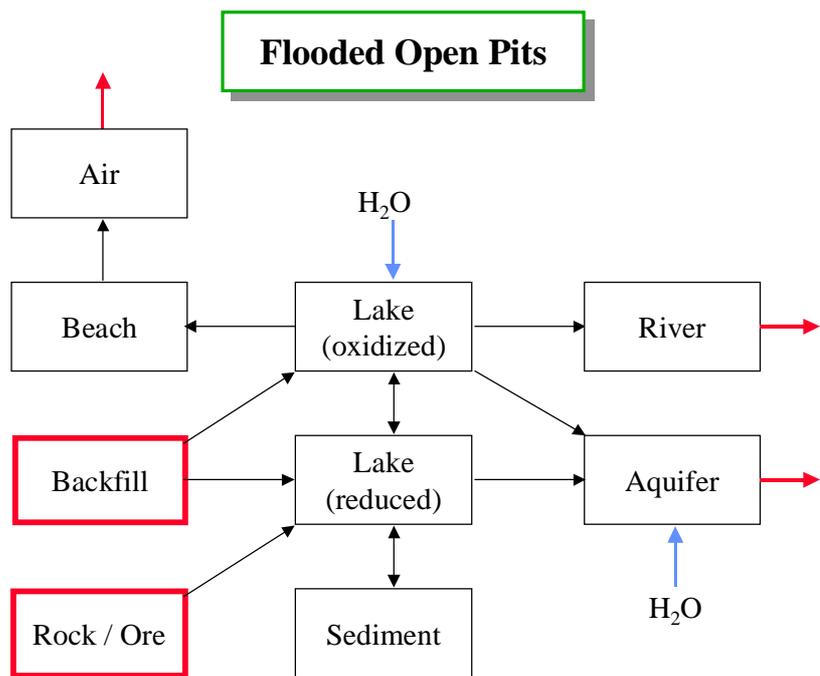
The contaminants are mainly radionuclides of class I (uranium, radium, thorium and their radioactive decay products), but also heavy metals (class III).

Direct exposure pathways are through sub-surface aquifers or a river which is used for drinking water, or through fishes caught in the lake. Indirect pathways are through livestock (meat, milk) drinking contaminated water or through crops grown on land irrigated with contaminated water.

External exposure by occupancy on the shores or during bathing in the lake (here also ingestion of water is possible) must also be envisaged.

Example

A site originating from open-cast uranium mining is the **Lake Tranebärssjön / Sweden**.



Generic compartment scheme for flooded open pits.

3.8 Open basins

Phenomenological Description

Major representatives of this site class are (artificial) tailing ponds and slurry basins from uranium mining and milling, but also from other industries (production of phosphate, aluminium, copper, lead, tin, mercury). Such ponds can be partially dried, with a trend towards falling totally dry. They are usually engineered in a way that there is a sealing layer on their bottom, and a dam with an outlet, often combined with a water cleaning facility. The (purified) water is then released to a river. Sometimes water coming out at the foot of the dam is collected in ditches and pumped back into the pond.

Main Processes

The site is dominated, due to the steadily decreasing moisture content, by precipitation processes, including co-precipitation and surface precipitation. Deeper layers may also exhibit a loss of oxygen leading to a reducing chemical environment. Water flow rates are usually very low, with no directed currents in the basin.

Contaminants and Exposure Pathways

Radionuclides of class I (uranium, thorium, and radium) have to be considered for nearly all of such sites, but most often in combination with other contaminants dependent on the kind of industry the site is based on. Often some mining and milling processes may have enriched heavy, such as lead, copper, mercury, cadmium or chromium, or even arsenic.

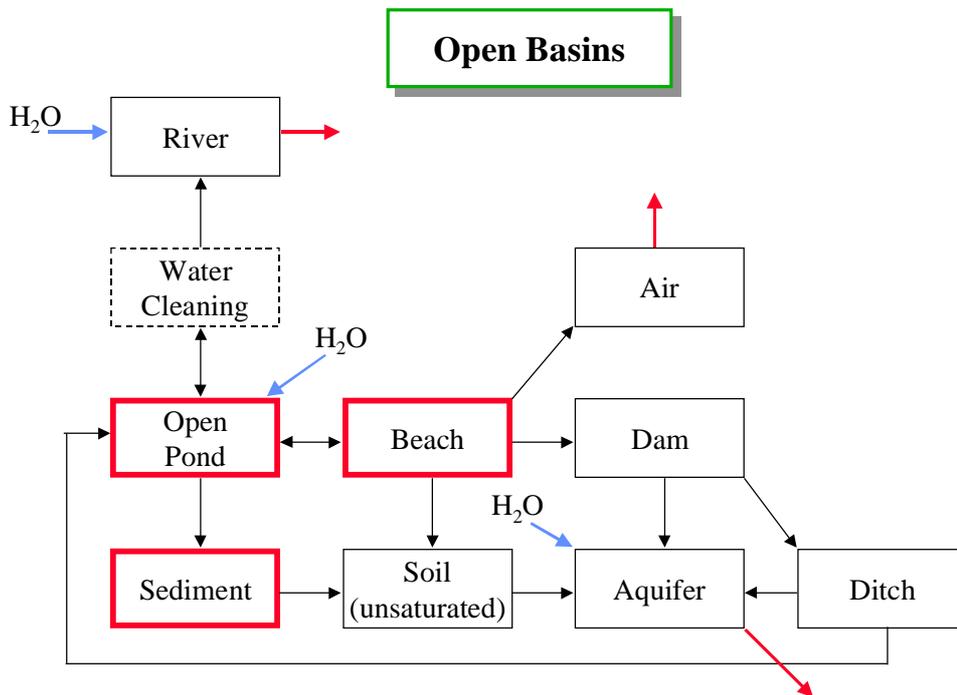
As direct exposure pathways the drinking of water from contaminated river or aquifer must be considered. Such water may also initiate indirect pathways through irrigation of crops and watering of livestock. External irradiation must also be considered.

Another exposure pathway may be through inhalation of air contaminated by dust or aerosols (containing uranium, radium, arsenic) from dried parts of the pond. Also, radon exhalation can contribute to this exposure.

On the other side, there should be no fishing, bathing etc. because the draining of the residual ponds is rather fast in terms of time (certainly below 10 years). During that time the necessary access restriction for the site can easily be maintained.

Examples

- Tailing ponds from uranium mining and milling in Helmsdorf, Dänkritz and Crossen (Saxony / Germany), in Culmützsch und Trüntzig near Seelingstädt (Thuringia / Germany), in Buchovo mill (near Sofia / Bulgaria), in Eleshnitsa (Bulgaria), in Strapod Ralskem (North Bohemia / Czech Republic) or in Feldiora / Romania.
- Sludge basins from the phosphate industry in Tessenderlo / Belgium, Kutina / Croatia, Maardu, near Tallinn / Estonia.



Generic compartment scheme for open basins.

3.9 Contaminated lakes

Phenomenological Description

This site class comprises any water reservoirs (lakes, ponds, artificial storage reservoirs) contaminated either by accidents or by purpose due to injections.

Main Processes

The contaminants can enter the site either as a suspension or truly dissolved. Sedimentation and sorption processes, respectively, will then give rise to secondary source terms on the bed of such lakes. The inflow and throughput of water will lead to dilution effects, but also to a gradual creation of secondary sources away from the original contamination place through resuspension, horizontal transport and desorption / sorption cycles. Contrary to the sites describes in section 1.7 and 1.8 the overall chemistry in these lakes is usually much closer to chemical equilibrium, showing no strong deviations from ambient pH and providing an oxidizing environment. The major phenomena besides surface processes are complexation reactions in the water, here ubiquitous organic matter such as humic acids play an important role. Finally, the transport promoted by eigen- and pseudo-colloids must be taken into account.

Contaminants and Exposure Pathways

The main contaminants can strongly vary, depending on the origin of the contamination.

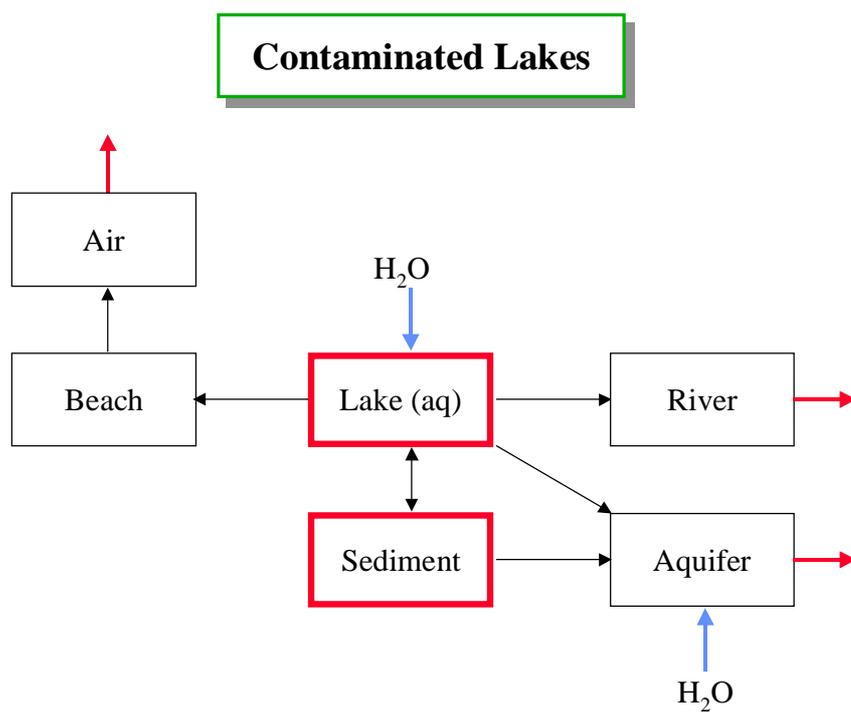
Potential direct exposure pathways include the drinking of water originating from the site (directly or through aquifers) and fish and shell fish consumption. External irradiation may take place during occupancy, which includes bathing.

Another direct exposure pathway may be through inhalation of air contaminated by hydrosols.

The water from the site may also initiate indirect pathways through irrigation of crops and watering of livestock.

Examples

(Lake Karachay – Urals MAYAK)



Generic compartment scheme for contaminated lakes.

3.10 References

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4. Restoration techniques

The selection of restoration techniques which can be appropriately applied to a contaminated site will depend upon a number of factors. These will include:

- the scale of the contamination problem;
- the contaminated medium;
- the radionuclides involved;
- the location of the contaminated site with respect to the local population;
- the location of the contaminated site with respect to a suitable waste repository for any residues.

This chapter describes and quantitatively characterises restoration techniques for treating sites contaminated by radionuclides. Some restoration techniques, which could be potentially useful for restoring these sites, have been omitted on the grounds that, at the time of writing, results have only been obtained from bench-scale investigations and there was inadequate information available to enable their performances to be assessed with respect to radioactive contamination. It is to be expected that techniques which have been omitted by these criteria (e.g. phytoremediation) will continue to develop to a level where they would be included in a review such as this. Also techniques for the restoration of large agricultural or urban areas have not been considered.

After the description, characteristics that will be dealt with, are:

- the applicability of the technique (contaminants and media)
- the performance
- the costs
- the labour volumes

In the accompanying CD-ROM (Hedemann Jensen, 1999) the results of a literature review on those characteristics are stored in the database RESTRAT.mdb. A critical survey of this review may be found in Zeevaert and Bousher (1998).

4.1 Description

Restoration technologies may be divided into four major categories.

- *Removal of source* - normally applied to contaminated soil, although contaminated groundwater or surface water can be removed by pumping. Removal of the contaminated medium may be followed by transportation to a more secure location or a subsequent separation procedure.
- *Separation* - applicable to both contaminated soil and groundwater. Separation technologies, which may be carried out both *in-situ* and *ex-situ* (following excavation or removal of the contaminated medium). Specific techniques include:
 - soil washing;
 - filtration;
 - chemical solubilisation;
 - ion exchange;
 - biosorption.
- *Containment* - barriers may be installed between contaminated and uncontaminated media to prevent the migration of contaminants. Principle techniques include:
 - capping;
 - sub-surface barriers.

- *Immobilisation* - materials may be added to the contaminated medium, in order to bind the contaminants and reduce their mobility and reduce their bioavailability. Immobilisation techniques may be carried out both *in-situ* and *ex-situ*. Principle techniques include:
 - cement-based solidification;
 - chemical immobilisation.

4.1.1. *Removal of Sources*

Conceptually, the most straightforward method to reduce the radiological impact of a site is to remove the medium containing the contaminants to a disposal site. Alternatively, bulk removal is used in conjunction with a separation process where decontaminated material can be returned to the excavation area and a smaller volume of contaminated material is taken to a disposal site.

Bulk removal of contaminated soil may be carried out with excavators (Valentich, 1994; Rice, 1994; Shirley and Schlessler, 1994), bulldozers (Kutlachmedov *et al.*, 1994; Blagoev *et al.*, 1996), vibration cutters or graders (Blagoev *et al.*, 1996). When the contamination is near the surface removal may be carried out by scraping (Blagoev *et al.*, 1996; Roed and Andersson, 1996), or by turf cutting (Bondar *et al.*, 1995; Kutlachmedov *et al.*, 1994; Jouve *et al.*, 1994; Grebenk'ov *et al.*, 1994; Jouve *et al.*, 1993). However, these latter approaches are considered applicable only to the removal of the top 50-75 mm layer of material.

The bulk removal of groundwater, or other contaminated liquids, is likely to be subject to many of the same considerations as bulk removal of solid material, although this option has been less well documented in the available literature. Removal of liquids is generally undertaken in more modest quantities, for a particular purpose (e.g. transfer of wastes for further treatment prior to disposal).

4.1.2. *Separation Techniques*

4.1.2.1 *Soil Washing*

Soil washing separates fine soil particles (silts and clay) from the larger particles by mixing the base soil with water and passing the resultant slurry through a separator. The techniques which may be used to achieve the soil particle fractionation and separation by size include:

- washing and rinsing (Bondar *et al.*, 1995; Bovendeur and Pruijn, 1994; Dworjanyn, 1996; Chilton and Pfuderer, 1989; USEPA, 1996);
- attrition scrubbing (Timpson *et al.*, 1994; Sadler and Krstich, 1994; Peng and Voss, 1994; USEPA, 1996; Groenendijk *et al.*, 1996);
- centrifugation (Goldberg *et al.*, 1994; Mathur *et al.*, 1996; Mista *et al.*, 1995);
- gravity separation (Elless *et al.*, 1994; Bovendeur and Pruijn, 1994; USEPA, 1996);
- hydrocycloning (Peng and Voss, 1994; Bovendeur and Pruijn, 1994; Mathur *et al.*, 1996; Nechaev and Projaev, 1996).

Where the contamination is largely associated with the fine particle fraction this process can be highly effective (USEPA, 1996).

Soil washing is essentially an *ex-situ* process, where the clean granular output stream (soil particles) can be returned to the excavation area. The remaining contaminated soil fines and process wastes are available for further treatment and/or disposal.

4.1.2.2 *Filtration*

Filtration is a process in which contaminants in liquid media (e.g. groundwater) are separated from the liquid media by a porous membrane. The pore size of the membrane can be varied to remove particles

and molecules of various sizes. Depending on what is fed into the filtration system, the process generates two waste streams: a filter cake of solid material, a filtrate of treated (clean) liquid.

The treated (clean) liquid can be reused or returned to its natural environment. The filter cake requires further treatment and/or disposal.

Two membrane filtration systems have been reported (Anderson *et al.*, 1994; USEPA, 1996). This includes an application on surface water at Rocky Flats (Anderson *et al.*, 1994).

4.1.2.3 Chemical Solubilisation

The purpose of chemical extraction is to separate specific radionuclides from the contaminated medium, using a suitable solvent, thus collecting them as a concentrated solution. Separation of the solvent containing the dissolved radionuclide can be an effective method for decontamination but can potentially be a complex and intensive process. The contaminated medium has to be collected and contained in a vessel where the solvent has to be added, ensuring a good contact with the medium. At last the contaminated solvent has to be extracted and collected.

Some factors and agents can influence the solubility of a radionuclide, including:

- the pH;
- complexing agents;
- ion exchange agents to replace the radionuclide in the solid medium;
- oxidation and reduction agents.

This approach may be used as a stand alone technology with the resulting concentrate available for disposal. Alternatively, separation may be used in combination with other technologies where the concentrate is further treated, e.g. through immobilisation or incineration.

An important requirement for this approach is that a solvent can be found which will dissolve the contaminating radionuclide in a particular medium. The choice and effectiveness of the solvent will be dependent upon the chemistry of the radionuclide, its concentration and the nature of the contaminated soil. Therefore, the choice of solvent tends to be site-specific (USEPA, 1996).

4.1.2.4 Ion Exchange

Ion exchange is a process where contaminating ions are removed from a contaminated liquid by replacing them with less harmful ions. The process makes use of materials that have binding sites which have a significantly higher affinity towards the contaminating ions than to ions which are already bound. Contact between the contaminated liquid and the ion exchange material enables the contaminating ion to displace the less harmful ion. The contaminant is removed from the liquid, and is concentrated on the surface of the ion exchange material, while the less harmful ion is released into the liquid.

Treatment of the contaminated groundwater is typically achieved by first extracting the water from a well sunk into the contaminated soil by pumping or collecting as it discharges. The water is filtered and passed, under pressure, through a fixed bed of the ion exchange material where the radionuclide is collected and concentrated. The groundwater, thus depleted of the radionuclide, can be readily returned to the environment (USEPA, 1996).

Two types of ion exchange material are available: reversible and irreversible materials. Reversible materials are usually resins which may be regenerated by stripping off the contaminant (usually with a strong acid). The contaminant can be collected as a highly concentrated, low volume waste, which is available for further treatment. Irreversible ion exchange materials (zeolites) are cheap substances which accumulate the contaminant for later disposal.

4.1.2.5 Biosorption

Biosorption treatments utilise the well-established ability of biomass to accumulate metal ions from solution. The biomass achieves this through accumulation, using a variety of mechanisms (Beveridge, 1989), such as physical adsorption, chemical bonding or production of precipitating agents (e.g. phosphate and sulphide). Both living and non-living biomass can be used to extract metal ions. For living organisms, accumulation can be both extracellular and intracellular (Beveridge, 1989). However, non-living biomass can retain many of the surface properties of the living organisms and, in addition, adsorption may be enhanced by chemically modifying the surface of the material.

Treatment of the contaminated water is accomplished by first filtering and then passing it through a vessel containing the biomass. Here the contaminant is extracted from the solution by biosorption onto the biomass (Volesky, 1990). The water, depleted of the contaminant, may then be returned to the environment. The contaminated biomass is then available for further treatment.

Biosorption can be a fairly rapid process with equilibrium times as low as a few minutes (Singleton and Simmons, 1996). It is accomplished *ex-situ* and may be used to treat contaminated water extracted from a site or the contaminated solutions from soil washing.

4.1.3. Containment

4.1.3.1 Capping

Capping describes a barrier placed over the surface area of the contaminated waste, which may serve several objectives:

- reducing the emission of gases from the waste to atmosphere;
- restricting the infiltration of surface water and, hence, the leaching of contaminants from the site;
- providing shielding from direct irradiation on the site;
- controlling resuspension of contaminated dust to atmosphere.

A cap usually consists of a combination of several layers of different materials. Most of the objectives of capping can be achieved with natural materials, such as layers of soil, gravel, rock or rip-rap. However, the prevention or restriction of infiltration of water is only properly achieved by introducing low-permeability layers. These may be naturally low-permeability soils such as clay (McGregor 1994), or artificial materials such as plastics, geomembranes (e.g. polyvinyl chloride, high density polyethylene, very low density polyethylene, polypropylene, polyester, hypalon), geosynthetic clay liners (usually blankets of bentonite clay with geotextile), asphalt (asphalt-mix, asphalt concrete, asphalt rubber membrane) or cement (concrete).

A typical cap for containing radioactive media may thus consist of a compacted filler, a geomembrane, a layer of compacted clay, another geomembrane and several metres of top soil (USEPA, 1996).

4.1.3.2 Sub-surface Barriers

Sub-surface barriers may be installed around the contaminated zone to confine lateral or vertical migration. They may consist of vertical barriers reaching down to an impermeable natural horizontal barrier, such as a clay zone, to impede ground water flow, or they may completely confine the area, including the bottom, as is the case for land encapsulation. Vertical barriers are mostly *slurry walls* or *grout curtains*.

- *Slurry walls* consist of vertically excavated trenches that are filled with slurry which hydraulically shores the trench to prevent the collapse of the side walls during excavation and produces a barrier to groundwater flow. Slurry walls are generally a mix of bentonite and water, or Portland cement, bentonite and water (USEPA, 1996).
- *Grout curtains* may be constructed in two ways: permeation grouting or jet grouting by mixing. Permeation grouting, takes advantage of the soil's natural permeability by using pressure to inject

the grout, which then flows into the soil. Jet grouting by mixing uses a rotating drill while injecting the grout. This fractures the soil and mixes it with the grout. The spacing of grout curtains (narrow, vertical, grout walls) is selected so that each "pillar" of grout intersects the next, thus forming a continuous wall or curtain. Typical grouts include Portland cement, alkali silicate grouts and organic polymers.

These technologies are primarily concerned with reducing the contamination of groundwater. Consequently, sub-surface barriers are frequently used in conjunction with other technologies. In particular, they are used with capping since this would produce an essentially complete containment structure surrounding the waste mass. Where total land encapsulation is to be considered, the area to contain the waste must first be excavated, after which a liner or other impermeable material can be installed.

4.1.4. Immobilisation

4.1.4.1 Cement-based Solidification

Solidification processes reduce the mobility and solubility of contaminants by stabilising them into a solid matrix and may be accomplished *in-situ* or *ex-situ*.

In-situ solidification is carried out by injecting immobilising agents directly into the contaminated medium using auger/caisson head systems or injector head systems. The estimated throughput rate is 40 to 80 tonne h⁻¹ for shallow soil mixing and 20 to 50 tonne h⁻¹ for deep soil mixing (USEPA 1996).

Ex-situ technologies require excavating the contaminated material and mechanically mixing it with the solidifying agents.

The objective is to limit the spread of contaminants via leaching and to slow the rate of emission of any radon within the solidified mass. The end product of solidification should be a monolithic block of high structural integrity.

Cements generally used for soil stabilisation may be subdivided into three major types (Malone and Lundquist, 1994):

- Portland cements;
- pozzolanic materials such as fly ash (non crystalline silica) or lime (calcium);
- blends of pozzolanic and Portland cement, with additives such as bentonite or other clays (to lower permeability), calcium chloride (as a setting agent and for freeze resistance during curing), lignosulfonates (as setting agents and viscosity modifiers) and gypsum (to increase strength and reduce permeability).

4.1.4.2 Chemical Immobilisation

Chemical immobilisation reduces the solubility of radionuclides by creating a stable or inert solid phase which binds the radionuclide and hence restricts its migration into groundwater. This can be achieved through the addition (either *in-situ* or *ex-situ*) of a chemical binding agent.

In-situ chemical immobilisation involves injecting immobilising reagents directly into the contaminated medium using auger/caisson head systems or injector head systems. The estimated throughput rate is 40 to 80 tonne h⁻¹ for shallow soil mixing and 20 to 50 tonne h⁻¹ for deep soil mixing (USEPA, 1996).

Ex-situ technologies require excavating the contaminated material and mechanically mixing it with the immobilising reagents. A full-scale polyethylene extruder can process material on a scale of 900 kg h⁻¹. The extruded material will cool and set within a few hours (USEPA, 1996) and the solidified material is then stored in containers, for disposal, or re-buried.

Reagents commonly used are thermoplastic polymers (asphalt bitumen, paraffin and polyethylene) and thermosetting polymers (vinyl ester monomers, urea formaldehyde and epoxy polymers) (Chisholm, 1994; Kalb and Adams, 1994; Pollock *et al.*, 1996; USEPA, 1996).

4.2 Applicability

The differences in approaches employed by the restoration techniques means that they will be applicable to different situations. Of particular importance in the initial selection of appropriate techniques for a given site are:

- the media to which a given restoration techniques may be effectively applied;
- the radionuclides which will be affected by the restoration technique;

Any decision on the appropriateness of a particular technique will also involve consideration of a number of site-specific factors such as volume of waste, the locality of the site and accessibility. However, site-specific factors are not discussed any further here.

4.2.1. Media

Each restoration technique will be appropriate to treating different contaminated media. The contaminated media fall into three broad categories:

- ground water, surface water, slurry and waste water;
- soil, sediment, sludge, tailings;
- bulk waste, buried waste.

Table 1 summarises the suitability of different restoration techniques for treating these categories of contaminated media. This may be used for general guidance for eliminating certain restoration approaches. Site-specific factors may further reduce the number of restoration options available.

Table 1 : Suitability of restoration techniques for treating contaminated media

Remediation technique	Media		
	Ground water, surface water, slurry, waste water	Soil, sediment, sludge, tailings	Bulk waste, buried waste
Removal of source	✓	✓	✓
Separation			
Soil washing	✗	✓	✗
Flotation	✗	✓	✗
Filtration	✓	✗	✗
Chemical solubilisation	✗	✓	✗
Ion exchange	✓	✗	✗
Biosorption	✓	✗	✗
Containment			
Capping	✗	✓	✓
Sub-surface barriers	✓	✓	✓
Immobilisation			
Cement-based solidification	✗	✓	✓
Chemical immobilisation	✗	✓	✓

Note: ✓ suitable for a given medium; ✗ unsuitable for a given medium.

Based on data given in EPA (1996).

4.2.2. Contaminants

The suitability of restoration techniques for treating different contaminants depends upon the chemical and physical properties on the contaminant. Table 2 shows the radionuclides for which the different restoration techniques are cited as being effective. It should be noted that this table should be used for guidance only. Site-specific factors (e.g. the chemical and physical state of the radionuclide) must also be taken into account.

In general, the removal of the source, containment and immobilisation are likely to be suited for most radionuclides. However, separation processes are more radionuclide-specific.

Table 2 : Reported effectiveness of restoration techniques in treating radionuclides

Remediation technique	Radionuclide						
	Co	Sr	Cs	Ra	Th	U	Pu
Removal of source	✓	✓	✓	✓	✓	✓	✓
Separation							
Soil washing			✓	✓	✓	✓	✓
Flotation				✓		✓	✓
Filtration				✓		✓	✓
Chemical solubilisation	✓		✓	✓	✓	✓	✓
Ion exchange		✓		✓		✓	
Biosorption	✓	✓	✓			✓	✓
Containment							
Capping	✓	✓	✓	✓	✓	✓	✓
Sub-surface barriers	✓	✓	✓	✓	✓	✓	✓
Immobilisation							
Cement-based solidification	✓	✓	✓	✓	✓	✓	✓
Chemical immobilisation	✓	✓	✓	✓	✓	✓	✓

Note: ✓ reported to be effective for a given radionuclide.
 Primary source of data was EPA (1996).

4.3 Performance

The performance of each restoration technique is determined by its effectiveness against the contaminants (radionuclides) and the time during which they remain effective.

The means by which the effectiveness is expressed will depend upon the technique and the medium to which it is applied. It is expressed from the viewpoint of the impact assessment in which it has to be introduced and is therefore characterised by one of the following terms:

- the decontamination factor (the factor by which the level of contamination is decreased) for source removal and separation techniques;
- the permeability for containment techniques;
- the reduction in radionuclide mobility for immobilisation techniques.

The service life of some restoration technique must also be taken into account; in particular with respect to barriers such as applied in containment techniques. This can be an important consideration as many radioactive wastes remain potential sources of contamination for very long periods of time.

Another important factor to be taken into account for the separation techniques is the reduction in the volume of waste, which has primarily economic consequences (and possibly a slight decrease of the decontamination factor in comparison with the total removal).

The derivation of the performance data, summarised in Table 3, may be found in Zeevaert and Bousher (1998). The specific value for a particular site will fall within the range of values given for the different restoration technique. The uncertainty has been taken into account by assuming triangular (for ranges of less than one order of magnitude) and log-triangular (for ranges of greater than one order of magnitude) distribution functions with the maximum and minimum taken from ranges in Table 3 and the mode taken to be a site-specific value.

Table 3 : The performance of remediation techniques

Remediation Technique	Unit [†]	Value	Service Life
Source Removal			
Soil excavation	DF	1 - 20	NA
Soil scraping	DF	1 - 20	NA
Separation			
Soil washing	DF RF	1 - 10 50 - 98 %	NA
Flotation	DF RF	1 - 10 28 - 97 %	NA
Filtration			NA
<u>NB</u>			
Range for all radionuclides	DF	2 - >100	
U, Pu, Am	DF	>100	
Ra	DF	2	
α-emitters	DF	7	
Chemical solubilisation	DF	1 - 20	NA
Ion exchange (liquids)			NA
Cs	DF	20 - 100	
U	DF	3 - 100	
<u>NB</u> A RF value is not meaningful here, given that a liquid waste is produced.			
Biosorption (liquids)			NA
Range for all radionuclides	DF	2.5 - >100	
Containment			
Capping	k	1×10^{-12} - $1 \times 10^{-9} \text{ m s}^{-1}$	100 - 1000 a
Subsurface barriers	k	1×10^{-12} - $1 \times 10^{-8} \text{ m s}^{-1}$	100 - 1000 a
Immobilisation			
Cement-based solidification	MRF	5 - 25	100 - 1000 a
Chemical immobilisation	MRF	5 - 50	UNK

Note: [†] DF = decontamination factor; RF = waste reduction factor for separation; k = permeability coefficient; MRF = mobility reduction factor; NA = not applicable; UNK = unknown.

From Zeevaert and Bousher (1998).

4.4 Costs

The direct costs associated with the application of a restoration technique to a contaminated site can be divided into three categories:

- the cost of treating the contaminated material;
- the cost of excavating the contaminated material (and transportation);
- the cost of disposing of contaminated waste arising from the treatment (and transportation).

The cost of treating will be dependent upon the choice of remediating technology and will include capital costs, operational costs and maintenance costs.

The cost of extracting or excavating the contaminated material will be a consideration for *ex-situ* techniques or for the technique of source removal. The cost values will also take account of the transport costs and be largely dependent on the quantity of medium being excavated rather than the remediation technique.

The cost of disposing of the waste after treating the contaminated material will also take account of the transport costs. They will be especially important for source removal, followed or not by separation.

The derivation of the costs for each restoration technique are described in detail in Zeevaert and Bousher (1998). These are summarised in Table 4.

Due to the uncertainties associated with these values the same approach was followed as in previous section.

Indirect costs, such as those associated with monitoring and loss/gain of income and taxes, are very site specific and cannot be estimated straightforwardly from the characteristics of the restoration techniques. They are described in Section 5.4.

Table 4 : The costs of remediation techniques

Remediation Technique	Cost
Source Removal	
Soil Excavation	
Excavation costs (including transport)	50 - 150 EUR m ⁻³
Excavation costs (including transport and RCRA disposal)	450 - 800 EUR m ⁻³
Disposal cost for radioactive material (including transport)	2000 - 3000 EUR m ⁻³
Soil Scraping	
Scraping costs (including transport)	1 - 3 EUR m ⁻² (surface area)
Separation	
Soil washing	
Soil washing costs	150 - 500 EUR m ⁻³
Cost of excavation and transport (prior to washing)	50 - 150 EUR m ⁻³
Cost of disposal of radioactive residue (including transport)	2000 - 3000 EUR m ⁻³
Flotation	
Capital costs	15 - 240 EUR m ⁻³
Operational costs	0.8 - 3.8 EUR m ⁻³
Cost of excavation and transport (prior to flotation)	50 - 150 EUR m ⁻³
Cost of disposal of radioactive residue (including transport)	2000 - 3000 EUR m ⁻³
Filtration (liquids)	0.1 - 3.8 EUR m ⁻³ (liquid)
Costs of disposal of radioactive residues (used filters)	2000 - 3000 EUR m ⁻³
Chemical solubilisation	
Separation costs	130 - 670 EUR m ⁻³
Cost of excavation and transport (prior to solubilisation)	50 - 150 EUR m ⁻³
Cost of disposal of radioactive residue (including transport)	2000 - 3000 EUR m ⁻³
Ion exchange (liquids)	1.3 - 2.5 EUR m ⁻³ (liquid)
Costs of disposal of radioactive residues	2000 - 3000 EUR m ⁻³
Biosorption (liquids)	1 - 3 EUR m ⁻³ (liquid)
Costs of disposal of radioactive residues	2000 - 3000 EUR m ⁻³
Containment	
Capping	30 - 45 EUR m ⁻² (surf. area)
Subsurface Barriers	
Slurry walls	510 - 710 EUR m ⁻² (barrier)
Grout curtains	310 - 420 EUR m ⁻² (barrier)
Immobilisation	
Cement-based solidification (<i>ex-situ</i>)	25 - 150 EUR m ⁻³
Cost of excavation and transport (prior to immobilisation)	50 - 150 EUR m ⁻³
Cement-based solidification (<i>in-situ</i>)	50 - 310 EUR m ⁻³
Chemical immobilisation (<i>ex-situ</i>)	60 - 420 EUR m ⁻³
Cost of excavation and transport (prior to immobilisation)	50 - 150 EUR m ⁻³
Chemical immobilisation (<i>in-situ</i>)	60 - 420 EUR m ⁻³

From: Zeevaert and Bousher (1998).

4.5 Work rates (labour volume)

The work rates associated with each restoration technique are especially important for assessing the collective dose to the restoration workers. They can vary over a range of values. This arises from the fact that these techniques have been applied to a variety of contaminated media, on a variety scales and under a variety of conditions. They are derived in Zeevaert and Bousher (1998) and are summarised in Table 5

The specific values for a particular site and the uncertainties are dealt with in the same way as in previous section.

Table 5 : The work rates (restoration workers) of remediation techniques

Remediation Technique	Work Rate (Restoration Workers)
Source Removal	
Soil excavation	0.2 - 1 manh m ⁻³
Soil scraping	0.03 - 0.3 manh m ⁻² (surf. area)
Separation	
Soil washing	0.05 - 0.5 manh m ⁻³
Excavation and transport of the soil (prior to separation)	0.2 - 1 manh m ⁻³
Flotation	0.05 - 0.5 manh m ⁻³
Excavation and transport of the soil (prior to separation)	0.2 - 1 manh m ⁻³
Filtration (liquids)	0.4 - 1.4 manh m ⁻³
Chemical solubilisation	1 - 2.5 manh m ⁻³
Excavation and transport of the soil (prior to separation)	0.2 - 1 manh m ⁻³
Ion exchange (liquids)	0.4 - 1.4 manh m ⁻³
Biosorption (liquids)	0.4 - 1.4 manh m ⁻³
Containment	
Capping	0.03 - 0.3 manh m ⁻² (surf. area)
Subsurface barriers	0.06 - 0.4 manh m ⁻³ (barrier volume)
Immobilisation	
Cement-based solidification (<i>ex-situ</i>)	0.05 - 0.5 manh m ⁻³
Excavation and transport of the soil (prior to immobilisation)	0.2 - 1 manh m ⁻³
Cement-based solidification (<i>in-situ</i>)	0.06 - 0.4 manh m ⁻³
Chemical immobilisation (<i>ex-situ</i>)	0.05 - 0.5 manh m ⁻³
Excavation and transport of the soil (prior to immobilisation)	0.2 - 1 manh m ⁻³
Chemical immobilisation (<i>in-situ</i>)	0.06 - 0.4 manh m ⁻³

From: Zeevaert and Bousher (1998).

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5. Methodologies

5.1 Assessment of radiological and non-radiological health effects

The ranking procedure for restoration options at contaminated sites, as will be explained in section 5.6, is based on the radiation protection principles of justification and optimisation. In such an optimisation study, risk for health effects (radiological and non-radiological) or health detriment is one of the major attributes.

Health effects from exposure to contaminants are generally divided into two categories:

- effects for which the probability of development is proportional to the dose (somatic and genetic effects) and for which it is assumed that exposure to even very low doses presents a non-zero risk (no threshold for effects)
- effects that only occur above a given threshold level of dose (somatic effects)

In the case of exposure to ionising radiation, the two types of effect are also referred to as stochastic and deterministic effects.

Non-threshold substances include genotoxic carcinogens and mutagens; threshold substances include non-genotoxic carcinogens and substances causing toxic effects other than cancer and genetic effects. U.S. Environmental Protection Agency (USEPA) classifies agents as carcinogenic and non-carcinogenic.

5.1.1. Risk from exposure to ionising radiation

During the past decade, new information about the carcinogenic effects of radiation has come from epidemiological studies of Japanese atomic bomb survivors; patients irradiated therapeutically; workers exposed to radiation in various occupations; and populations residing in areas of high natural background radiation. New data have also come from long-term studies of the carcinogenic effects of irradiation in laboratory animals and from experiments on neoplastic transformation in cultured cells. The new data have been summarised in reports from United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR) (1988) and National Academy of Sciences (NAS) / Biological Effects of Ionizing Radiation (BEIR) (1990).

In the case of radioactive contaminations in the environment, radioactivity concentrations will rarely reach levels that are of concern with respect to deterministic effects. As a consequence, in the case of site contaminations such as considered in this study, only stochastic effects are taken into account.

In the stochastic region, a radiation dose will involve a risk commitment, *i.e.* a commitment of an increased cancer death probability rate in the future, after a minimum latent period that may be from a few years in the case of leukaemia to tens of years for other malignant conditions. Any change in the age-specific death probability rate would therefore occur later in life, when the risk of death from other causes is also higher. The risk committed by a radiation dose at a given age can therefore not be added to the background risk at the same age.

The risk for stochastic effects is proportional to the radiation dose incurred. A lifetime dose of 1 Sv starting at age 0 will result in an average lifetime risk, r_{rad} , of fatal cancer of about 0.05 Sv^{-1} . The average loss of life expectancy per unit lifetime dose can be calculated as the product of the average lifetime risk, r_{rad} , and the average loss of life expectancy per cancer, l (15 years):

$$r_{rad} \cdot l = 0.05 \text{ cancer} \cdot \text{Sv}^{-1} \cdot 15 \text{ years} \cdot \text{cancer}^{-1} \approx 1 \text{ year} \cdot \text{Sv}^{-1}$$

The collective loss of life expectancy from a given collective dose, S_{rad} , can then be calculated as:

$$L_{rad} = S_{rad} \cdot r_{rad} \cdot l$$

The collective exposure, S_{rad} , can be expressed over many generations as long as the age distribution of the exposed population does not deviate significantly from the one which has been used to determine the average lifetime risk, r_{rad} .

The methodology for assessing radiation doses is explained in section 5.2.

For potential exposures arising from scenarios that are not certain to occur, the risk and associated loss of life that is to be expected, are reduced with respect to a normal exposure, in proportion to the probability of occurrence of the scenario concerned.

5.1.2. Risk from exposure to toxic chemicals

As far as non-threshold effects are concerned, non-radiological health effects, e.g. from exposure to chemical contaminants can in principle be described in the same way as the exposure to radiation. The attributable lifetime risk from an individual lifetime exposure to a specific chemical contaminant can be calculated by a proper lifetime integration of the exposure, the risk per unit exposure of the contaminant as a function of age and the survival function as a function of age. However, the available information on risk factors for exposure to non-radiological carcinogens is scarcer than for exposure to ionising radiation.

As far as threshold effects are concerned, the risk is assessed by comparing the exposure level with a reference level of exposure, below which a health effect is very unlikely. A toxic hazard quotient may be derived in the following way:

$$\text{Toxic Hazard Quotient} = E / RfD \text{ (or } RfC)$$

Where E is the exposure level, RfD is the reference dose (by ingestion) and RfC is the reference concentration in air (for inhalation)

As a rule, the greater the value of the hazard quotient, the greater the concern for potential threshold effects ought to be. However, the quotient should not be interpreted as a statistical probability.

5.1.3. Risk from combined exposure to toxic chemicals and ionising radiation

Combined exposure to radiation and chemical carcinogens should be expressed in a common risk scale in order to determine the total expected detriment from that exposure.

With respect to non-threshold effects the impact of a combined exposure may be assessed adopting some assumptions two of the more important ones being:

- the lifetime cancer risk, r , is linearly related to the exposure, E , also known as the *linearity hypothesis* which can be expressed as $r(E) = k \cdot E$, and
- no synergetic effects exist between exposures to radiological and non-radiological carcinogens, i.e. the total lifetime risk of a combined exposure of $E_1 + E_2 + E_3 + \dots$ can be described by the sum of risks as $r(E_1 + E_2 + E_3 + \dots) = k_1 \cdot E_1 + k_2 \cdot E_2 + k_3 \cdot E_3 + \dots$

With these assumptions the effect of a combined collective exposure to ionising radiation and toxic heavy metals and chemicals can be described as a total collective loss of life expectancy:

$$L_{total} = L_{rad} + L_{chem,1} + L_{chem,2} + L_{chem,3} + \dots = L_{rad} + \sum_i L_{chem,i}$$

When considering both threshold and non-threshold health effects, difficulties are encountered. Several possible approaches have been suggested, e.g. by USEPA, CRARM (Commission on Risk Assessment and Risk Management) and WHO (World Health Organization). However, a general consensus on a unified approach on the combination of threshold and non-threshold health risks does not yet exist.

In this study risks from non-radioactive contaminants have not been assessed.

5.2 Physico-Chemical Source Term Assessment

5.2.1. Introduction

The source term for radioactive contamination is strongly dependent on the chemical conditions present at the site. Most of the relevant radionuclides can occur in many different chemical forms in an environmental compartment, each having a different mobility, transfer coefficients to and between living matter, and even toxicity. Thus, estimation of the transport behaviour and the resulting risk to the population requires exact knowledge about which forms a contaminant actually predominantly exists and how this is controlled by environmental parameters. The observable variation in existing forms for a given chemical element is termed chemical speciation, the definition is as follows:

Chemical speciation is the distribution of one or more chemical elements between all its possible species (distinct chemical entities such as simple ions or neutral molecules, ion pairs, associates, complexes, hydrolysis products, pure minerals, solid solutions, gases, surface complexes) in a given system, usually consisting of several phases: an aqueous solution in contact with one or more solid phases and the gas phase, and possibly other fluid phases (organics), colloids and aerosols.

Species distributions determine whether a contaminant is mainly a dissolved component - and thus easily transported and taken up - or is immobilized through precipitation or adsorption onto a surface. Therefore, changes in speciation can either accelerate or slow down radionuclide migration. Many of the processes affecting the source term are influenced by the speciation. Their importance may vary, so the following list should not be considered a ranking:

- radioactive decay;
- complexation reactions (with organic and inorganic ligands), through:
 - hydrolysis;
 - dissociation;
 - association / polymerization;
- oxidation state changes / redox reactions;
- precipitation and dissolution of solid phases;
- co-precipitation (inclusion and surface precipitation) of trace components;
- physical and chemical sorption onto mineral surfaces or colloids;
- formation of solid solutions (mixed mineral phases);
- ion exchange;
- extraction (in case of several fluid phases);
- formation of colloids;
- formation of aerosols;
- processes involving biological material, such as biosorption, biologically catalysed redox reactions, enzymatic reactions, metabolisms.

Several of these processes are controlled by parameters from outside, which can be considered as fixed by certain environmental (natural and man-made) conditions. However, for some of these parameters feedback can be observed; the internal physico-chemical processes of an environmental compartment will influence parameters such as pH or redox potential, either amplifying or extenuating already established trends from outside the compartment. Without a thorough description of all parameters and processes, leading to a comprehensive, "full-system" modelling, no sound prognostics are possible.

5.2.2. Unfolding the K_d concept

Up to now, the retardation of contaminants due to interactions with minerals are mainly dealt with using distribution coefficients. A distribution coefficient (or K_d value) is defined as the ratio of the sorbed (fixed, immobilized) and unsorbed (free, truly dissolved) fraction of a component (chemical element) under equilibrium conditions. That means, however, subsuming many physico-chemical processes into one parameter, which is a severe weakness of the K_d principle (Hayes *et al.*, 1991; Puigdomenech and Bergström, 1994). Distribution coefficients are very difficult to measure with a good precision and accuracy. Literally by definition, because of their incorporation of very different basic physico-chemical phenomena, they are dependent on so many parameters that even slight changes in one system parameter (say the Eh or the content of a major cation, or the occurrence of a new mineral phase, etc) can drastically change the distribution coefficient. To measure the effect of all combinations of these parameters is impossible. That means, all K_d values used nowadays in risk assessment or other prognostic studies are just snapshots for specific locations of the site valid only for the time of the measurement. This in turn assigns them very large uncertainties.

A much better strategy is the decomposition of the K_d value into the main basic processes defining it. Such an approach will unfold the single value K_d into a vector of parameters, such as Eh, pH, concentrations of the various components, surface areas, and temperature. Apparently this is a step backwards, but it has the great advantage that all these parameters can be measured with more reliability and precision. Knowing the functional relationships between these processes and how they contribute to the K_d allows a computation of K_d rather than a measurement. Moreover, simulations with variable parameter values may easily yield a K_d surface as a function of the "primary" parameter vector, even for hypothetical conditions. Also, some long-term effects that can render conventional distribution coefficients meaningless (co-precipitation, diffusion of the trace element into crystal lattices) can be accounted for in a better way. Another application is expressing K_d as a function of time, related to better-defined time dependencies of other basic parameters. Furthermore, it becomes possible to identify those parameters affecting the K_d strongest. Consequently, extra measurements can be designed efficaciously to reduce the K_d uncertainty.

Sorption phenomena play an especially important role when looking at the processes responsible for K_d values. Whereas the simplest (and older) sorption models do not distinguish between the various processes contributing to the overall sorption, newer model approaches at least describe separately the effects of the electrostatic attraction between a surface and an ion having charges of opposite sign, and the effects coming from a chemical reaction of an ion with a reactive surface site. The most advanced approach, and the one used in the RESTRAT project, are the surface complexation models (SCM), which can well be dealt with in the usual thermodynamic framework (Dzombak and Morel, 1990; Davies *et al.*, 1978).

A successful application of the aforementioned " K_d -unfolding" requires a good knowledge about the chemical speciation: in the simplest case the determination of the dissolved, precipitated and sorbed fractions of a contaminant. To do so *in-situ* requires enormous efforts: both time and analytical measurement equipment. Even then in certain cases the speciation can not be determined directly. Therefore, in most cases the chemical speciation is computed by special software packages, based on proven algorithms and good sets of thermodynamic and analytical data.

5.2.3. Chemical Equilibria Computation

The computation of chemical speciation basically means solving the set of equations describing the chemical equilibria that must be fulfilled simultaneously in a given system. These equations can be grouped into two main classes; the mass balance equations and the mass action equations. This leads to

a system of non-linear equations with all “free” concentrations being unknown. However, the total concentrations of all elements are known. An additional restriction in ionic systems is the charge balance. Thus, one activity (mostly H^+ / pH, but any other charged species will also do) can not be varied. Finally, the internal norm is that the concentration of the solute H_2O is fixed, so yet another component must not be specified. Therefore, oxygen is usually not explicitly represented by a master species.

The resulting system of non-linear equations is solved iteratively by varying the free concentrations. This involves for example the following steps:

- Solution of all reactions in the homogeneous (aqueous) solution and on surfaces.
- Iteration for activity coefficient corrections.
- Check of saturation indices: does any precipitation / dissolution occur ?
- Test new multi-phase assemblages, if necessary.
- Compute changes in mass for every phase.

The most popular way of computation consists of a two-level process. First, starting values for the variable parameters are estimated and a robust minimisation method is applied. In a second step a hybrid Newton-Raphson procedure with incorporated relaxation is used. During each iteration step the activity coefficients are held constant, they are computed again after all concentrations have been determined. The result is the species distribution for all chemical elements (maybe even split according to their various redox states), the activity and activity coefficients of all species, the redox potentials, and the saturation indices for all minerals.

To help in the selection of an appropriate software package for (geo)chemical speciation modelling, the following questions should be worked through before starting the modelling:

- Does the program handle redox reactions, kinetic rate laws, adsorption, multiple phase equilibria?
- Is the applied activity coefficient model adequate for the system under investigation?
- Which mathematical methods, especially minimisation approaches, are applied?
- What is the performance of computational speed and numerical robustness?
- Can the user access an internal database? If so: is it possible to introduce changes, exclusions, additions via input file options?
- Does the software provide graphical output or other post-processing tools?
- Which operating system and programming language is necessary?
- Are manual and/or source code available, how can support be obtained?

Additionally, features which may be important include: upper concentration limit, charge balance check, initial values, ability to cope with changes in volume, temperature or pressure. There is a variety of speciation programs to choose from, some very popular ones are listed in the RESTRAT TD 2 (Chapter 4).

The most sophisticated chemical speciation software does not give any sensible results without a comprehensive and reliable database. The chemical data foundation can be divided into two main groups, site-specific parameters and reaction-specific parameters.

5.2.4. Site-Specific Analytical Data

Site-specific parameters give the analytical composition of the system (components and their concentrations). They must be determined experimentally in the field and from samples.

The analytical composition can be obtained by measuring the following values:

- Temperature.
- Pressure (total system and partial pressure of all gaseous components).
- Elementary composition, concentrations for all components (total or, better, for each species separately):
 - pH, ionic strength, humidity.
- Composition of solid phases:
 - identification of rocks and their mineral matrix.
- Redox state:
 - Eh, oxygen partial pressure, potentials of important redox pairs.
- Surface properties:
 - Specific surface area, active sites, site densities, crystal size, structural disorders, charge distribution, surface films (biological matter!).

5.2.5. Reaction-Specific Thermodynamic and Kinetic Databases

Reaction-specific parameters describe the underlying basic reactions in a given system, they are universal and, therefore, the same for all sites. They can, hopefully, be extracted from databases or the literature. In some cases it may also become necessary to determine them in specifically designed laboratory experiments. In general, these parameter sets are not unique in the sense that they are dependent on the applied model: activity coefficients, surface complexation model (SCM), species set, mixing model, kinetics, colloids, aerosols.

- Thermodynamic parameters:
 - equilibrium constants;
 - solubilities;
 - enthalpies, entropies, and Gibbs free energies;
 - heat capacities;
 - partial molar properties;
 - activity coefficients.
- Kinetic parameters:
 - rate constants with the corresponding rate laws.
- Radioactive decay rates.
- Degradation rates for biological material.
- Parameters for biosorption.

If adsorption phenomena have to be taken into account, the available analytical and thermodynamical data will determine which model has to be used, starting from a simplistic K_d approach to sophisticated surface complexation models. A comprehensive mineralogical investigation of the soils and rocks present in the system is essential in either case. If one moves to surface complexation, thermodynamic data describing the surface itself, and for the complexes formed on it must be retrieved.

5.3 Radiological impact assessment

As explained in a preceding section (5.1), risks for health effects constitute a major attribute in the ranking procedure of restoration options. In this study only health effects from exposure to radioactive contaminants are considered. Moreover, only normal exposure is taken into account. This means that collective doses are a direct measure for the risks of health effects. The radiological dose to the population due to the radioactive contamination of the site, for the various restoration options, are to be considered along with doses to the workers carrying out the restoration techniques.

The radiological dose to the restoration workers can be straightforwardly assessed from the labour volumes required for the restoration techniques (unit labour volumes multiplied with the volumes or areas of contaminated soil/sediment) and the radioactive contamination levels at the site. The radiological dose to the population on the other hand can only be assessed through the use of a more comprehensive biosphere¹ model; i.e. a model that describes and calculates the releases, transfers and accumulations of the contaminating radionuclides between and in the biosphere media at the site concerned. From the concentrations or inventories of the radionuclides in those media, the radiological dose to the population can then be derived.

5.3.1. Model Categories

Several categories of impact assessment models exist.

- With respect to time dependency, a distinction can be made between equilibrium models and dynamic models. In dynamic models, the concentration or inventories of radionuclides in the biosphere media are calculated in a time-dependent way, from transfers between, and losses from the media, that are mostly described by first-order differential equations. In equilibrium models, a simple approach, assuming continuous equilibrium of radionuclide concentrations between the biosphere media is adopted. The concentrations of the radionuclides in the biosphere media can then be derived easily from each other, applying single algebraic equations (often ratios) or analytical solutions of the linear differential equations, describing the transfers between the media.
- With respect to the type of mathematical ways of computation, distinction may be made between analytical and numerical models. This distinction only applies to dynamic models; equilibrium models are obviously always analytical. Analytical models consist of analytical solutions of the first-order differential equations or algebraic equations expressing the concentration values of the radionuclides in the biosphere media. These values may be a function of time, in the case of a dynamical model. In numerical models the differential equations are solved numerically, applying time steps that are accordingly small with respect to the turn-over rate of the radionuclides in the biosphere media.
- With respect to spatial complexity, simple compartment models and multiple compartment models, up to finite elements or finite differences models may be distinguished. Simple compartment models deal with the biosphere as a small number of large, physically defined entities, areas or volumes (i.e. compartments) between which transfers of radionuclides take place and that are assumed to be instantaneously well mixed. Multiple compartment models are obtained by dividing the biosphere into a larger number of compartments, yielding more and smaller compartments. In a finite elements or finite differences model, a very large number of very small entities are created. The assumption of instantaneously well mixing in the successive elements becomes then physically justified. This type of models is in general only applied to transport of radionuclides by diffusion and advection in aqueous and solid media (geologic formations, aquifers). They require a very good characterisation

¹ According to the Biospheric Model Validation Study (BIOMOVS II, 1993), the biosphere is "that portion of the earth's environment inhabited by living organisms. It comprises of parts of the atmosphere, the hydrosphere and the lithosphere. The biosphere includes the human habitat in the widest sense of the term."

of the site to be modelled and a sufficient computation time. Other models may also calculate radionuclide concentrations at discrete points in the environment.

- With respect to dealing with uncertainties, deterministic and probabilistic models can be distinguished. In a deterministic model, each parameter is given only a single deterministic value leading to a single result or output (calculated endpoint). In a probabilistic model, to certain parameters, a range of possible values is assigned, modelled by corresponding random laws, from which a value will be chosen at random at each run of the model, leading to a range of results (or pdf's) for multiple runs of the model.

In this project the characterisation of the sites and the aim of the study only allow the use of a generic type of model. A dynamic, numerical compartment model is applied, following a probabilistic approach for dealing with uncertainties. It is based on the BIOPATH/PRISM model of Studsvik (Bergström *et al.*, 1982).

5.3.2. Exposure scenarios

An exposure scenario can be defined as a chronological sequence of features, events and processes, commonly known as FEPs², leading to an exposure of the public. For assessing the radiological impact on the population, two types of exposure scenarios can be considered: normal evolution scenarios and probabilistic scenarios (potential exposures).

The normal evolution scenario groups the FEPs, which are certain or nearly certain to happen. For such a scenario, the radiological impact may be expressed in terms of radiological dose or radiological risk. This risk is the product of the radiation dose with the risk conversion factor of the dose.

The probabilistic scenarios also includes FEPs with a limited probability of occurrence, but which may lead to large radiological consequences. For this type of scenarios, the radiological impact is to be expressed in terms of radiological risks. This risk is the product of the radiation dose, should the scenario occur, with the probability of occurrence of the scenario and the risk conversion factor of the dose.

As has already been explained, this study only allows for the normal scenarios which are expected from the actual conditions of the sites.

5.3.3. Calculated Endpoints

In the case of normal evolution scenarios, calculated endpoints of the impact assessment model should include :

- the maximum annual individual effective committed³ dose to an average individual of the critical group of the public (see 'critical group' in section 5.3.7);
- collective effective dose commitments to the public, truncated in time and space (see 'dose calculations' in section 5.3.7).

However because of the uncertainty involved in the assessment of the collective dose, this dose is often not considered.

² Features, events and processing intervening in evolution scenarios and influencing the migration of the radionuclides and the exposure of humans.

³ Individual committed dose = the dose to an individual due to the ingestion or inhalation of radioactive material, integrated over a certain time after the intake increased with the external dose to the individual at the time of exposure. The time over which the internal dose is integrated is representative for the expectation of life of the individual exposed; i.e. 50 years for an adult and 70 years for a child.

In case probabilistic scenarios (potential exposure) are allowed for, calculated endpoints should also include radiological risks.

In this study, the maximum annual individual dose to the critical group before any restoration work has been carried out on the site, will be calculated for the sake of comparison with the IAEA criteria, for clean-up of contaminated land (International Atomic Energy Agency, 1996).

The collective dose to the public will be calculated with and without the restoration measures carried out as a measure for the radiological health detriment, one of the major attributes in the ranking methodology of the restoration options. Also the collective doses to the restoration workers, due to exposures during the restoration works, are to be included in the total health detriment.

5.3.4. Time Frame

From recommendations of the International Commission on Radiological Protection (1997) and of the United Nations Scientific Committee on the Effects of Atomic Radiation (1982) time periods of 100 and 500 years have been selected over which to carry out dose calculations. The former time constitutes a rounded-off value of the average lifetime of an individual, the latter one can be considered as a maximum over which an institutional control over the site may be effective and the radiological background can be remembered (administrative memory). Within this time period (500 years) the climate may also be supposed not to have changed to an important extent.

Moreover the National Radiological Protection Board states that "in decision making, less significance should be attached to collective dose estimates relating to periods beyond 500 years into the future than to those relating to shorter time periods." (Barraclough *et al.*, 1996). Changes in the biosphere and in the behaviour of exposed population groups, such as changes in agricultural practices, dietary or living habits, may occur over relatively short time periods. However these changes are difficult to predict in a quantitative way. Therefore in this generic approach, a constant biosphere is assumed in which the 'hypothetical' critical group(s) will behave in a similar way as today (see also "reference biosphere methodology" in BIOMOVs (1996), Biospheric Model Validation Study).

5.3.5. Transfers in the biosphere

Releases of contaminating radionuclides from a source and subsequent transfers in the biosphere, including losses from the system, are identified and characterized by the processes bringing them about. They can be mathematically expressed by the use of algebraic equations or differential equations, that are derived from the phenomena (FEP's) governing them.

Very important processes, governing the transfers of radionuclides in the biosphere, are sorption processes (adsorption, desorption). They divide the radionuclides between the solid phase and the aqueous phase, according to a distribution coefficient (K_d). In general they have a large influence on the rate constants of the transfers and on the transport of the radionuclides. Several transfer processes or transport phenomena involve only one phase; the solid phase (e.g. sedimentation, resuspension, bioturbation, erosion) or the liquid phase (e.g. diffusion, infiltration, groundwater flow). Some transfers or transport involve both phases (e.g. flow of surface water, irrigation).

Influences from restoration options may be introduced in the following way:

- Removal of sources (or other radionuclide inventories), possibly followed by separation, can be characterised by a decontamination factor (DF) which can be applied directly to the corresponding sources or radionuclide inventories.
- The performance of a containment system is characterised by the permeability value k (in saturated conditions). This can be applied to the water infiltration rate through the source or other radionuclide inventory within the barriers.

- The influence of immobilisation can be characterised by a reduction of the release, or of the mobility of the radionuclides, which can be interpreted as an increase of its retardation with respect to its water transport.

5.3.6. Exposure pathways

Radionuclides present in the various compartments of the biosphere may expose man through various exposure pathways. The exposure can take place internally or externally.

External exposure may be due to the presence of radionuclides in the source or in other biosphere compartments, such as top soil, surface water, bed sediment. They can irradiate man directly or through contamination of his body surface or clothes.

Internal exposure can take place through inhalation or ingestion.

Exposure through inhalation is due to the contamination of air with radionuclides originating from one of the compartments mentioned above. The contamination is brought about by resuspension of soil or sediment particles (aerosols) or by emanation of radioactive gases (radon, carbon-14).

Exposure through ingestion is due to the contamination of the food chain (food crops, animal products, fish) or/and of drinking water. The contamination of the food chain may be brought about through :

- contaminated soil, leading to the contamination of:
 - vegetation (food crops and feeding crops for livestock) by root uptake;
 - livestock by soil uptake when grazing;
- contaminated water, leading to the contamination of:
 - soil and vegetation, by irrigation;
 - bed sediment and fish in contaminated surface water;
 - livestock, by watering;
- contaminated bed sediment, leading to the contamination of:
 - soil, by dredging and application of the sediment onto agricultural land;
 - surface water.

The contaminated drinking water may originate from surface waters or aquifers from which drinking water for the public is prepared.

For the restoration workers, only external exposure and exposure through inhalation have to be considered. Mostly they are directly exposed from the source, which may be constituted by soil or sediment, or from the surrounding soil or sediment.

5.3.7. Dose Calculations and Critical Groups

The doses to the population that are to be assessed are individual doses to an average member of the critical group, and collective doses to all exposed individuals (cf. Calculated Endpoints).

By definition the critical group is a group of members of the public which is reasonably homogeneous with respect to its exposure for a given radiation source and given exposure pathway(s) and is typical of individuals receiving the highest dose by the given exposure pathway from the given source (International Atomic Energy Agency, 1996).

Critical groups are in general supposed to present normal living and dietary habits and obtain their high dose from the contamination of their environment and or of their food. For the general population,

critical groups are often identified as self-sustaining farmers or fishermen living in the contaminated area.

Annual doses for exposure through inhalation of an average member of a critical group are calculated for each radionuclide, by multiplying the concentrations of the radionuclide in the air with the inhalation rate of the individual, the annual time of exposure and the corresponding dose factor for inhalation.

Annual doses for exposure through ingestion of an average member of a critical group are calculated for each radionuclide, by multiplying the concentrations of the radionuclide in the food products with the quantities of food products ingested annually and the corresponding dose factor for ingestion.

Annual doses for external exposure from each radionuclide are calculated by multiplying the concentrations of the radionuclide in the exposing media with the corresponding dose rate factor and the annual exposure times, taking into account possible reductions for shielding or exposure geometries.

Collective doses are the sums of the individual doses of all exposed individuals, over the time periods considered. However making this sums may be a very tedious procedure. An easier, be it less accurate, estimate of the collective dose through ingestion may be derived from the total production of contaminated drinking water, the total yield of food products on the contaminated agricultural areas and the yield of fish caught in the contaminated surface waters over the time period considered. They are then to be multiplied with their average radionuclide concentrations and with the corresponding dose factors for ingestion and summed up in order to obtain the collective dose.

The collective doses through inhalation and external exposure of the public are derived from the corresponding individual doses mentioned above, multiplied with an equivalent number of man.years of exposure over the time period considered and taking into account possible reductions of contamination levels with time.

The collective doses of the restoration workers consist only of inhalation and external irradiation components. They are calculated in the same way as the annual doses to the members of the critical group of the population, but replacing the annual exposure time by the total labour volume of the restoration work.

5.3.8. Uncertainty Analysis

The uncertainty of the calculated endpoints of the impact assessment model should be specified.

Basic types of uncertainty that may be distinguished are:

- scenario uncertainty;
- model uncertainty;
- parameter uncertainty.

The scenario uncertainty deals with the uncertainty of the scenario description: how different components will respond to a certain event and what will be the possible evolution of the system. In the case of risk assessments from scenarios, that are not certain to occur, also the probability of occurrence of the scenario is a source of uncertainty. Since this type of uncertainty is mostly very difficult to quantify, it is often accounted for by adopting pessimistic assumptions with respect to the scenario and system evolution. This type of uncertainty is not considered in this study (only scenarios, that are quasi-certain to occur, are taken into account).

Model uncertainty represents the lack of confidence about the mathematical model being a valid formulation of the assessment problem. Model uncertainties can be assessed through model validation by comparing model predictions with data sets that are independent of the data used to develop the

model. They are also not further considered in this study.

Parameter uncertainty considers the uncertainty about the true value of the parameters used in the model equations. This may be due to the lack of knowledge or to the random character of the parameter value. For assessing the uncertainty of the assessment endpoint due to the propagation of the uncertainty of the model parameters, several types of analysis can be applied.

For a model, consisting of relatively simple equations, uncertainty analysis can be performed using analytical methods, such as variance propagation.

For complex models, numerical methods are preferred to perform an uncertainty analysis. The most commonly applied technique is Monte Carlo simulation. Other approaches could involve differential uncertainty analysis and non-probabilistic methods, such as fuzzy sets, but are not considered here. Monte Carlo analysis is usually performed using one of two random sampling processes : Simple Random Sampling (SRS) or Latin Hypercube Sampling (LHS). In SRS a random value is taken from the distribution specified for each uncertain model parameter for calculating a simple estimate of the desired endpoint. This process is repeated for a specific number of samples or iterations. The result is a set of endpoint values from which discrete percentiles (e.g. 5, 50 and 95th percentile) or a probability distribution may be derived, provided that a sufficient number of iterations has been performed.

SRS is less efficient than LHS, requiring a larger sample size for an empirical approximation of the probability distribution of the assessment endpoint than LHS.

In LHS the range of each uncertain variable is divided into a number of intervals of equal probability, according to the number of iterations (sample size) required. For each run of the model the variables are assigned a value randomly selected from another interval each time. This process has the property of being able to cover the full range of each variable with a relatively small sample size. This method is applied in the PRISM programme (Gardner *et al.*, 1983).

When setting up a quantitative uncertainty analysis it should also be determined whether or not any of the parameters are correlated with each other. For known or suspected dependencies among model parameters that can be quantified through correlation coefficients, simulation techniques are available to address these dependencies. Dependencies which are suspected but hard to quantify can be included using subjective judgement.

5.3.9. *Sensitivity Analysis*

Several methods exist for investigating the sensitivity of model parameters or the influence of the uncertainty in parameter values on the total uncertainty of the endpoint values.

One of the simplest methods is to use the scatter plots of the Monte Carlo samples of the uncertain parameters against the model results to identify possible groupings. Other methods include regression, multiple and stepwise regression between model results and parameter values, correlation coefficients and partial correlation coefficients, correlation ratios and others. More details are to be found in (International Atomic Energy Agency, 1989 and National Council on Radiation Protection and Measurements, 1996).

Very frequently rank correlation coefficients (for instance Spearman's rank correlation coefficient, that is applied in PRISM) are used.

In this method a regression is performed on the rank order of the model results against the rank order of the uncertain model parameters. The index of sensitivity for the uncertain model parameters is the

square of Spearman's rank correlation coefficient.

5.4 Assessment of economic costs.

One of the major attributes in the ranking procedure of restoration options, next to the risk for health effects, is the economic cost. Important economic cost categories that can be distinguished include:

- pure restoration costs;
- waste disposal costs;
- loss/gain of income/taxes for the authorities;
- survey and monitoring costs.

5.4.1. Pure restoration costs

These costs consist of two important components:

- capital costs (initial investment costs) that are generally incurred once. It mostly concerns infrastructure (large tools) and apparatuses. They depend on the size of the contaminated area or quantities of contaminated material to be treated.
- O&M (operation and maintenance) costs or running costs that are usually evaluated on an annual basis. These costs include mainly *labour costs* and *consumable costs*. They can be considered to be directly proportional to the size of the area or the quantities of material that are contaminated.

Mostly reliable data for assessing costs have to come from studies of similar situations in the past. However the estimations of costs are hampered by the lack of past experiences.

Prices of some large tools or apparatuses are available but labour and consumable costs are very site-specific and may vary to a large extent.

In order to be able to apply costs from past experiences, generalised unit costs are derived; these are costs per unit volume of material to be treated or per unit surface area to be restored.

In this study data on unit costs have been collected from a literature survey. They are indicated in chapter 4 ('Restoration techniques').

5.4.2. Waste disposal costs

These costs are important when large quantities of contaminated material are generated which require disposal.

Contaminated waste will be brought about in all restoration options. Yet the quantities and associated costs arising will be important only for the case of source removal with, or without, subsequent separation, that involve the disposal of large volumes of contaminated material (soil, sediment, tailings etc). In fact, for many sites, the waste disposal costs may be the highest cost category, seriously affecting the economic feasibility of source removal and separation.

In cost assessments for waste disposal, unit cost values will also be applied. However, allowance has to be made for the level of radioactivity of the waste. For wastes with a very low radioactivity level, an industrial type of disposal can be applied, which is a much lower cost per unit of waste quantity, than for the disposal of radioactive waste.

The bulk removal of sources such as contaminated soil, without subsequent separation, may bring about large quantities of waste with a very low level of radioactivity. In this case, an industrial type of disposal, such as mentioned above, may suffice. The removal of the highly active residue that remains after a separation, will require a type of disposal for radioactive waste. However the quantities of the

waste to be disposed of are much lower after a subsequent separation, than without separation, which mostly results in a lower waste disposal cost for source removal with separation.

Generic values for unit costs are indicated in chapter 4 ('Restoration techniques').

5.4.3. *Losses or gains of taxes*

A special category of economic costs that can be considered, are losses or gains of taxes (or other forms of economic costs or profits) for the authorities, that are charged with the restoration of the contaminated site.

In general, they are lower than the costs of remediation and waste disposal mentioned above, but they should not be neglected *a priori*.

The losses or gains of taxes, to be included, are direct consequences of the restoration. Such losses mostly originate from activities that are no longer possible on the site such as agricultural activities in the case of capping of contaminated soil, or fishing activities at a river where the contaminated bed sediment has been capped. Gains can originate from activities that are made possible through the restoration such as soil reclamation for agricultural or residential purposes for instance.

The activities mentioned above may also be at the basis of individual profits or losses (of jobs or properties). However, these individual costs may not be put at the same level as the other economic costs considered up to now, since they are not borne by the authorities, that are also charged with the other costs of the restoration. The individual costs will be included in the ranking procedure as social attributes.

Indemnification of people losing their jobs or properties on the other hand should be included in the economic cost (because borne by the authorities).

The assessment of economic costs, related with losses or profits as consequences from site restoration, may be based on objective measures or economic values, such as:

- the price of agricultural products for an agricultural area;
- the price of building grounds (plots) etc...

Or they can be based on subjective measures or values, such as:

- the price people are willing to pay for transport or for moving away, and for making use of the facilities of the site.

In one way or another, costs per unit surface area can be derived for the restored/not restored site, according to its agricultural, recreational, residential, industrial or landscape value, depending on its possible uses. The taxes, associated with these costs or with individual jobs related to the uses mentioned above, can be deduced and considered as economic costs (losses or profits).

They have been included only for the example site of the Molse Nete river. However, no losses in productivity were caused by the implementation of remedial measures the other example sites.

5.4.4. *Monitoring costs*

Costs for monitoring and survey are another special category of economic costs, to be borne by the authorities.

Generally speaking, monitoring costs will be reduced after the restoration of a contaminated site. However, the gain will be much lower than the costs of restoration and waste disposal discussed earlier.

If no restoration of the contaminated site is carried out, the monitoring and survey of the site will have to be continued to its full extent over a certain time, depending on the effective half-lives of the important radionuclides. Normally it consists of a monitoring of the contamination on-site and a survey in the environment for the purpose of the radiological protection to the public.

After the restoration, the extent of the monitoring programme may be reduced, according to the type of restoration carried out. When extensive parts of the contaminating source are removed (after or without separation), the monitoring can be reduced considerably. On the other hand, when the radioactivity is remaining on the site, as is the case when capping or immobilisation has been carried out, then the reduction of the monitoring programme will be lower. In this case the monitoring system will have to verify the condition of the barrier or the fixation of the radionuclides by the immobilising agents.

The major components of the monitoring costs are personnel costs and consumables. Measuring samples in a specialised laboratory may also be an important cost component.

For a well characterised site, the costs of a monitoring system are easy to be quantified.

The monitoring programme for each restoration option can easily be derived from the programme before restoration, by adapting the location of the sampling or measuring points and the frequency of measuring, to the restoration options applied.

This has been applied in the example case of the Molse Nete river.

In a generic approach, a simple objective measure for the extent and the cost of a monitoring system could be the radiological risk to a critical group. If such a risk value is not available, the fraction of the activity left on-site could be a measure, but this should be reduced in relation with a measure for the efficacy of the restoration system (in the case of containment or immobilisation).

In this way the costs for monitoring and survey can be derived directly from the costs of the monitoring system present before remediation. This has been applied at the Drigg and Ravenglass sites.

5.4.5. Special aspects

Values of costs derived from past experiences are only rough approximations and consequently consideration should be given to the inherent uncertainties in the estimates.

When costs are indicated, they should always be related to the time at which they are arising, because of:

- inflation, which reduces the face value of money with time;
- change of unit costs relative to the general price level;
- the higher value of money in the present than in the future (even when allowing for inflation).

Two main conventions have been drafted in order to compare costs arising at different times:

- present worth evaluations, where total costs = investment costs + present worth of future running costs;
- annualised cost estimates = annuity investment cost, spread over lifetime + annual operational cost.

The cost values indicated in this report are mainly referring to the same time period (nineties) and are therefore not converted.

5.5 Assessment of social factors

Next to the radiological protection factors considered so far (economic cost and radiological health detriment), also non-radiological protection factors are to be considered in the optimisation analysis

leading to the ranking of restoration options at contaminated sites. Whereas radiological protection factors are related to the level of radiological protection achieved, including doses averted, costs and other disadvantages incurred in averting doses, non-radiological protection factors are defined as those, which are not. It is very difficult to generalise about these factors, although they can have an important or even overriding influence on the decisions taken.

We can distinguish between social and political factors. Social factors may include the following among many factors:

- perception of the hazard posed by the radiation from radioactive materials at the site,
- psychological impacts,
- reassurance provided by the implementation of remedial measures,
- anxiety caused by implementation of measures,
- individual and social disruption resulting from implementation of measures,
- loss of income.

Although some of these factors to a certain extent are related to the level of protection achieved they are all considered to be non-radiological protection factors or social factors. The political input, however, is always deemed to include only non-radiological protection factors, but is not further considered in this study.

Non-radiological protection factors, such as social factors, are not easily quantifiable, will vary markedly between countries, and may have opposing influences on the choices of intervention levels. Anxiety for instance can be caused by the change induced by the implementation of remedial measures. However, the absence of protective measures can also cause anxiety, which is often exacerbated by a lack of objective information.

Important social factors, that have been considered in this study, are *disturbance* and *reassurance*. Of these two, it is assumed that the dominating is reassurance because of its more or less permanent nature. Reassurance and psychological harm may be related. A decreasing reassurance can be interpreted as an increasing risk of psychological harm. Also reassurance and radiation health factors may be related, a decreasing dose level resulting in an increasing reassurance. Consequently the risk of psychological harm to the affected population, r_{psy} , would be proportional to the level of residual dose and would therefore have the same unit as the radiation risk factor, r_{rad} , (Sv^{-1}) which gives the risk of radiation induced stochastic health effects. If it were possible to determine the risk of psychological effects per unit dose, in terms of loss of life expectancy, the ratio of both risk factors could then enable the determination of the weight of the social factors (cf. section 5.6)

Intuitively, the ratio r_{psy}/r_{rad} is expected to be less than one and probably significantly less than one. However, this value judgement will completely depend on the specific situation. The experience gained after the Chernobyl accident was that socio-psychological factors were given much higher weight than radiation factors. In a non-accidental situation like remediation of contaminated sites with small exposures of the affected population the social factors would probably be given far less weight than in a major accidental situation like Chernobyl. Consequently, the value of r_{psy}/r_{rad} can be assumed to be significantly less than 1.

In this study, reassurance has been linked to both the residual dose and the fraction of activity remaining on the site after the remedial measure has been implemented. However, the residual dose and remaining activity are not necessarily correlated. A remedial measure that has left all the activity on site in a contained form (capping, surface barriers etc.) might give a substantial dose reduction and thus a low value of the residual doses. Detailed information on how social factors like reassurance are linked with individual doses and activity concentration on site is not available. Therefore, the utility value for reassurance has been taken to be maximum (100) for the option with the minimum value of collective dose and minimum value of remaining activity fraction on-site and minimum (0) for the option with the maximum value of collective dose and maximum value of remaining activity fraction on-site.

Disturbance has been linked to the volume of waste to be transported to the waste disposal site. The value of the utility function for disturbance has been taken to be maximum (100) for the minimum amount of waste to be transported from the site and minimum (0) for the maximum amount of waste to be transported from the site.

Optimisation of overall health protection would be the responsibility of the decision-maker with guidance from radiation protection experts as well as experts in the fields of social and psychological sciences.

It has been suggested that non-radiological protection factors should be included within the radiation protection framework. However, there is then a risk of "double-counting" of these factors, which may be accounted for in the numerical guidelines (action levels etc.) and again at the time of the decision. The optimisation of radiological protection is shown in Figure 1.

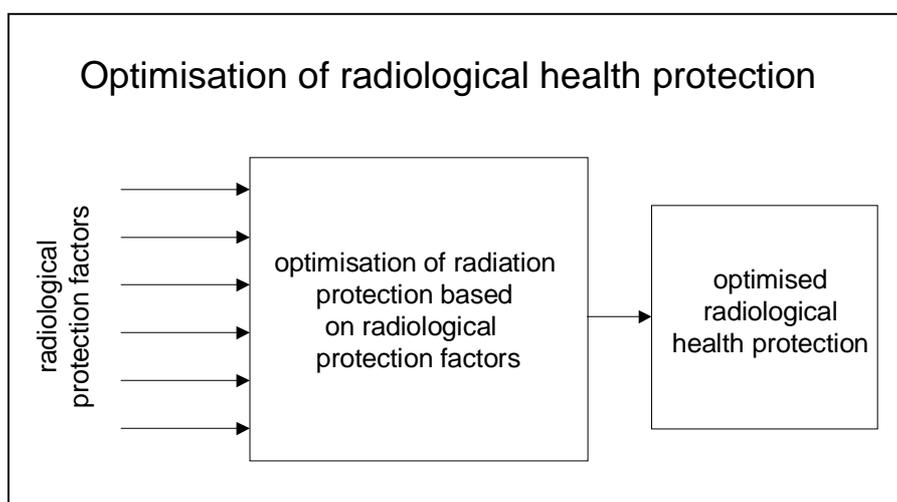


Figure 1 : Sub-optimisation of overall health protection where only radiological protection factors are included in the optimisation process.

The inclusion of socio-psychological factors in the radiation protection framework would also give very arbitrary levels of 'radiation protection'; the level of socio-psychological impact depending not only on the presence of radiation but to a large extent on other non-radiological protection factors, such as the attitude of the mass media, the political climate and the general level of information in the population.

Therefore, to achieve an optimised overall health protection, non-radiological protection factors should enter the optimisation process in parallel with radiological protection factors to form an overall strategy as shown in Figure 2.

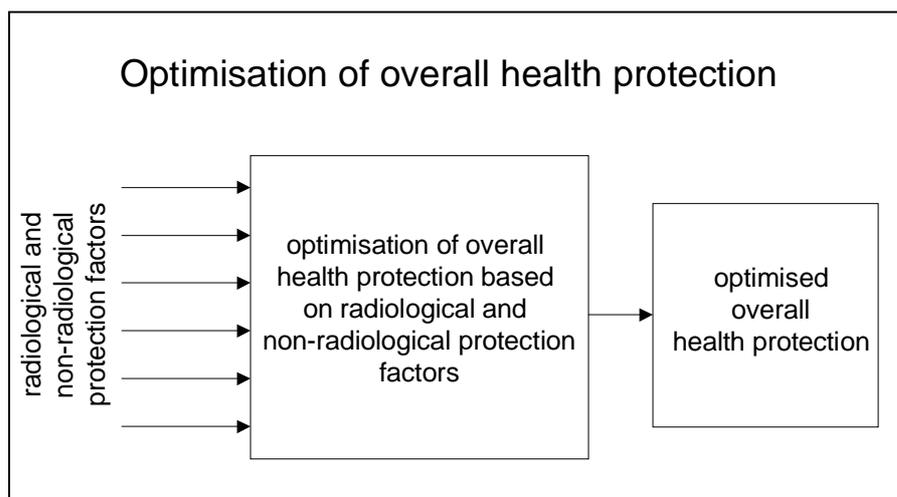


Figure 2 : Optimisation of overall health protection where radiological and non-radiological protection factors are included simultaneously in the optimisation process.

5.6 Ranking of restoration options

5.6.1. Justification and optimisation

The procedure for ranking restoration options for contaminated sites has been based on the radiation protection principles of justification and optimisation. In this study the protection of the public against radiation exposure from contaminated land is addressed and the justification/optimisation procedure is applied to the remedial or protection action for reducing this exposure.

The justification principle requires that the overall effect of the actions involved in the remediation should do more good than harm, taking account of relevant radiological and non-radiological factors. The optimisation principle requires that, among the justified remediation options, the one with the highest net benefit should be selected.

A short review of the justification and optimisation principles is given below.

5.6.1.1 Justification

Clean-up of contaminated land will introduce some benefit to the affected populations. The benefit of undertaking clean-up includes a large number of components or attributes, i , which quantify relative partial benefits, b_i . These partial benefits, depending on the circumstances, can be 'positive' benefits, or advantages, and 'negative' benefits, or disadvantages. Without intervention, the attributes, such as radiation doses - both individual and collective doses - and the anxieties they cause, will represent disadvantages as shown on the left side of Figure 3. After remediation, the disadvantages will have been reduced or even eliminated, and new attributes may have been introduced, as shown on the right side of Figure 3. Some of the new attributes may be advantageous, e.g. the reassurance produced by the remedial measure; others will be disadvantageous, e.g. the cost of the remedial measures and the collateral harm they may cause.

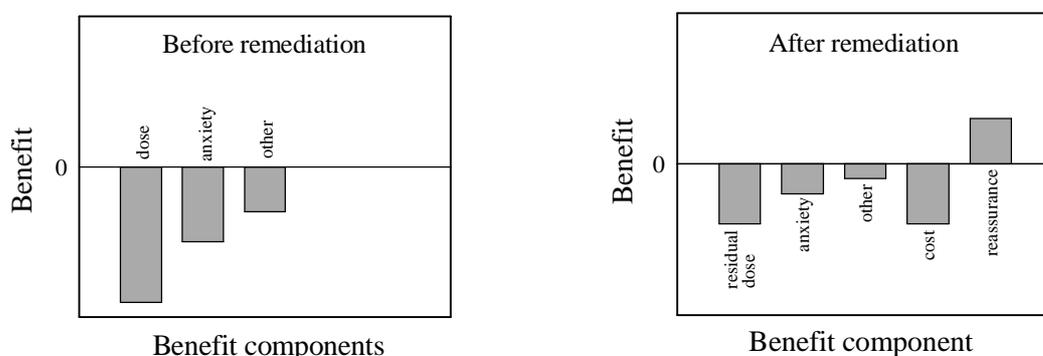


Figure 3 : Benefit components, b , of clean-up operations.

Clean-up is justified when the net benefit, ΔB , is positive:

$$\Delta B = \sum_i b_i(\text{after clean - up}) - b_i(\text{before clean - up}) = \sum_i \Delta b_i > 0$$

The net benefit, ΔB , of a clean-up operation will depend on several factors (attributes), e.g. avertable collective dose, ΔS , monetary costs of a clean-up operation, C , anxiety of the contamination, A , reassurance by the clean-up, R , etc. Thus the net benefit, ΔB , is a function of all the relevant parameters:

$$\Delta B = \Delta B(\Delta S, C, A, R, \dots)$$

The application of the justification principle to clean-up situations requires prior consideration of the benefit that would be achieved by the clean-up and also of the harm, in its broadest sense, that would result from it. It is emphasised that justification must consider non-radiological risks as well as radiological risks, *e.g.* chemical risks, and risks from industrial and transportation operations. Each of the benefit components, b_i , has to be expressed in the same units. These units must be in like quantities or values. For example, since costs are expressed in monetary terms, equivalent monetary values may be assigned to other parameters. Alternatively, other units of value must be used for example equivalent years of lost life.

5.6.1.2 Optimisation

Normally, there would be a range of justified remediation options for which the net benefit is positive. The optimum remediation option would be the one for which the net benefit, ΔB , is maximised, as shown on the left side of Figure 4. Option 1 is the no-remediation option for which the net benefit is zero. The options 4 to 8 are all justified because their net benefits are positive. Option 6 is optimum because the net benefit is maximum. The optimum remediation option does not necessarily mean the option with the lowest residual annual doses, either individual or collective, because there are additional considerations for determining the net benefit. This is illustrated in the right side of Figure 4 where options 7 and 8 entail a lower residual annual dose but give a smaller net benefit than the optimum option 6. If all remediation options have a negative net benefit, the no-remediation option would be the preferable.

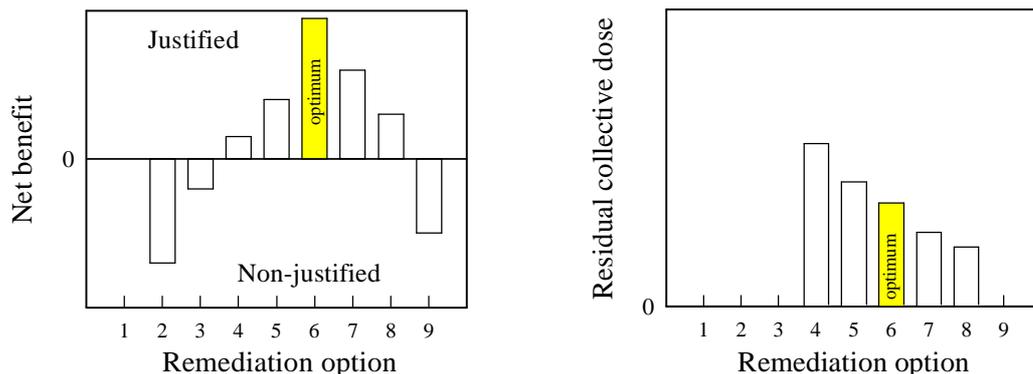


Figure 4 : Net benefit of different remediation options and the corresponding residual collective dose, S , after clean-up, for the five justified options.

The concept of optimisation of protection is practical in nature. Optimisation provides a basic framework of thinking - that it is proper to carry out some kind of balancing of the resources put into protection, and the level of protection obtained. The reduction in dose can only be achieved by the expenditure of some effort and by allocating additional resources. In such cases, it is necessary to decide whether the dose saving that is likely to result is worth the effort of achieving that saving. This is entirely consistent with the optimisation principle.

5.6.2. Decision-aiding techniques

Most decisions require multiple criteria to be taken into account. The field of multiple criteria analysis offers a number of approaches, which take explicit account of multiple criteria in providing structure and support to the decision-making process. In case of restoration of contaminated sites there are several criteria or attributes that need to be considered when choosing an 'optimum' restoration strategy. When the performance and costs of all the protection options have been assessed, a comparison is needed to define the optimum protection option. When the optimum is not self evident, the comparison can be carried using a quantitative decision aiding technique. The result of the

application of the quantitative techniques is known as the analytical solution. If there are non-quantified, radiological protection factors to be taken into account, the analytical solution may not be the optimum solution, which then will have to be determined more intuitively.

Some *decision-aiding* techniques available for use in carrying out decision analysis have been described in detail in ICRP Publication No. 55 (International Commission on Radiological Protection, 1989). The primary objectives of these techniques are to identify the various factors influencing the decision, to quantify them, and systematically to examine the trade-offs between them, so that the process can be made open to the people responsible for the decision and to public scrutiny.

A simple decision-aiding technique, that may suffice in many simple problems is *cost-benefit analysis*. However this technique is limited to quantitative comparisons between readily quantifiable attributes, such as avertable individual and collective risks from exposure to radiation and the monetary cost associated with the clean-up.

A decision-aiding technique that is capable of accepting input data of both a quantitative and a qualitative nature, and which can be used in a wide variety of situations, is *multi-attribute utility analysis*. Less quantifiable factors that are relevant to the decision, such as the reassurance provided by the clean-up but also the anxiety it causes, may then be taken into account

5.6.3. *Cost-benefit analysis*

Cost-benefit analysis involves a balancing of costs in order to establish optimum levels of radiation protection. Optimisation of protection results in the best available combination of costs of radiation protection, X , and detriment, Y , so the sum of the costs ($X + Y$) is minimised. The optimisation process will therefore maximise the net benefit, B . The optimisation condition is fulfilled at a value of collective dose, S_{opt} , where the increase in cost of protection per unit collective dose balances the unit reduction of collective dose:

$$\left(\frac{dX}{dS}\right)_{S_{opt}} = - \left(\frac{dY}{dS}\right)_{S_{opt}}$$

This way of obtaining the optimisation of protection has also been called *differential cost-benefit analysis*. The level of protection defined by the above equation is such that a marginal increase in the cost of radiation protection is balanced by a marginal reduction in the cost of radiation detriment.

The principal characteristic of cost-benefit analysis is that the factors entering the analysis are commonly expressed in monetary terms. In these circumstances the collective dose is transformed into a monetary valuation using a reference value of avoiding a unit collective dose, α . This quantity can be related to the risk per unit dose, R (about 0.05 cancer Sv^{-1}), and the statistical loss of life expectancy per radiation induced cancer, l (about 15 years cancer^{-1}), with some allowance for loss of quality of life for non-fatal cancers and severe hereditary effects. The average loss of life expectancy per unit effective dose, L , can thus be calculated to be:

$$L = R \cdot l \text{ [year} \cdot \text{Sv}^{-1}\text{]}$$

The value of L is approximately one year per sievert.

Within the international radiation protection community it has been argued that a society for protection purposes should spend *at least* what correspond to the GNP (Gross National Product) per capita to save a statistical year of lost life and probably somewhat more. So-called *willingness-to-pay* studies have resulted in values of 200,000 USD \pm 100,000 USD per saved year of statistical life, corresponding to 8 GNP \pm 4 GNP per capita for rich European countries. Therefore, the value of α can roughly be found from the following relation:

$$GNP \cdot R \cdot l < \alpha < 10 \cdot GNP \cdot R \cdot l$$

For rich European countries the value of GNP per capita is of the order of 25,000 USD year⁻¹, which would give a reference value of α between 25,000 USD Sv⁻¹ and 250,000 USD Sv⁻¹. The Nordic radiation protection authorities have recommended a maximum value of α of 100,000 USD Sv⁻¹ (Swedish Radiation Protection Institute, 1991).

The cost-benefit analysis methodology is limited to quantitative comparisons between the protection costs and the detriment costs. In order to include other relevant factors, *e.g.* the distribution of individual doses within the collective dose, it is possible to extend the framework of cost-benefit analysis. This extension allows different values to be assigned to the unit collective dose through an additional component of the detriment cost depending on the individual dose levels involved. The extension can be expressed as:

$$Y = \alpha \cdot S + \sum_i \beta_i \cdot S_i$$

where S_i is the collective dose of individual doses E_i in the i th group and β_i is the additional value assigned to a unit collective dose in the i th group.

5.6.4. Multi-attribute utility analysis

The essence of multi-attribute utility analysis is to use a scoring scheme (or multi-attribute utility function) for the relevant factors with the property that if the score (or utility) is the same for two options there is no preference for one or the other. As basis for comparison between options or alternative strategies, a simple multi-attribute value function approach can be used. There are two major components of such value functions:

- the evaluation of each alternative strategy with respect to the considered attributes, known as *utilities*, u
- scaling factors which reflect the relative importance of each of the attributes, known as the *weights*, w .

The use of utility functions allows the introduction of factors that are not easy to quantify in monetary terms as is required in cost-benefit analysis. The utilities and weighting factors can be expressed in an additive form to give an overall evaluation of each of the alternative strategies, i , or options:

$$U_i = \sum_{j=1}^n w_j u_{ij}$$

U_i is the overall evaluation of option i , w_j is the weight assigned to the attribute j , and u_{ij} is the score or utility of the n factors associated with each of the alternative i on attribute j . The higher the value of U_i , the better the overall ranking of the option. Normally, weighting factors are measured on a ratio scale and normalised to sum to 1 or 100.

There will, however, be uncertainties on the parameters used to calculate the values of the utility functions, u , and there will also be uncertainties on the values assigned to the weighting factors, w . These uncertainties can be included in the calculations of scores, U_i , by using software that is capable of building a model for the scores, $U_i(x, y, \dots)$ in which uncertainty distributions can be assigned to the values of each of the attributes, x, y, \dots , that defines the utility functions, $u_i(x), u_i(y), \dots$, and to the weighting factors, w , for each of the attributes.

5.6.4.1 Attributes for contaminated sites

The analysis should address both radiological and non-radiological issues. Examination of the first of these will, in principle, be straightforward since it involves only the radiation detriment to be averted and the costs associated with the remedial action (including both the direct cost of the action and costs to affected parties). Examination of the second class of issues will involve, in addition to consideration

of other hazards (such as those associated with chemical contaminants), economic and social considerations, some of which are beyond the scope of radiation protection.

The major attributes that are often the most relevant to be considered for remediation of a contaminated site, are *radiation induced health effects*, *monetary costs* and *social costs*. Each of these attributes can be divided into several sub-attributes. The ones considered in this study are:

- *Health attributes*
 - collective doses to population
 - doses to remediation workers
- *Economic attributes*
 - costs of remedial actions (incl. costs of labour and monitoring)
 - costs of monitoring of remedial options
 - costs of disposal of generated waste (in broad categories)
- *Social attributes*
 - reassurance of the public
 - discomfort, disturbance and anxiety from the remedial action

The attribute hierarchy to be used for selection of an optimum restoration strategy can be structured as shown in Figure 5.

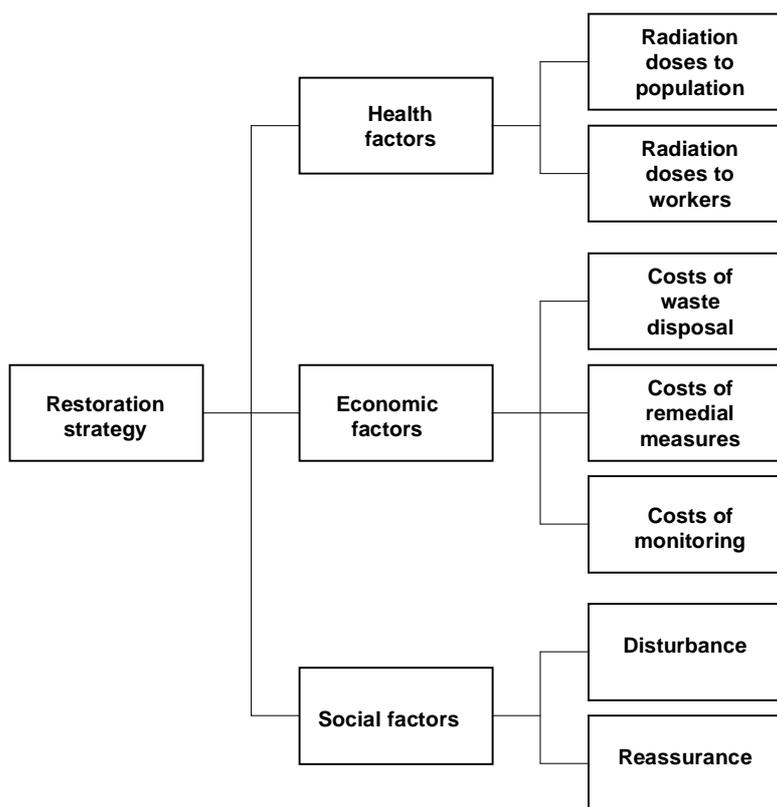


Figure 5 : Attribute hierarchy for restoration of a contaminated site.

5.6.4.2 Utility functions

A utility, u , or utility function, $u(x)$, will express the score or utility of a given attribute with value, x , for a given protective option. A risk neutral utility function can in general terms be defined as:

$$u(x) = 100 \cdot \left(1 + \frac{x_{\min} - x}{x_{\max} - x_{\min}} \right)$$

where $(x_{\min}; x_{\max})$ is the value range of the attribute considered.

The aim of scoring is to assign values to each alternative reflecting the contribution to the overall evaluation from their performance on each end-attribute (sub-attribute). One way of defining the scores (utilities) is to assign the alternative which does best on a particular attribute a score of 100 (or 1) and to assign the alternative which does least well a score of 0. All other alternatives are assigned intermediate scores, which reflect their performance relative to these two end points. A major advantage of this methodology is that the utility functions need not necessarily be linear. They can also have a concave or convex shape, either as increasing or decreasing functions. Functions of the decreasing type are shown in Figure 6 below. Risk averse and risk prone utility functions are shown. The former decrease faster nearer the worst consequences, being more sensitive to variations at the upper end of the range of consequences, whereas the latter decrease slower at the upper end.

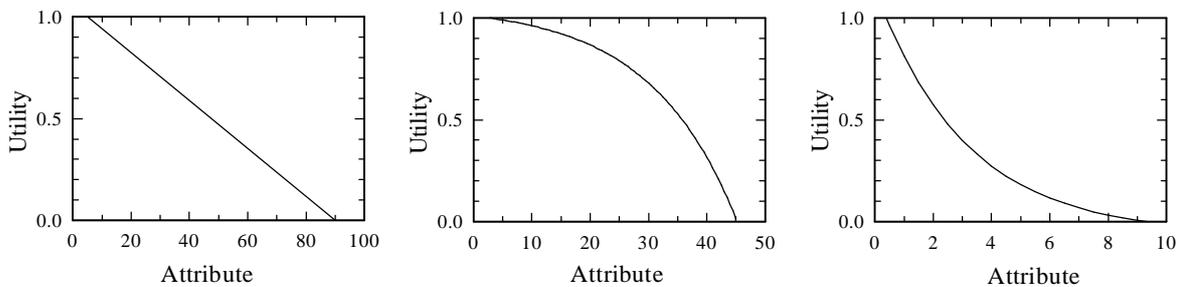


Figure 6 : Examples of utility functions of the decreasing type; risk neutral (left figure), risk averse (middle figure) and risk prone (right figure) utility functions.

5.6.4.3 Weighting factors

The determination of weighting factors is a very difficult task. Different decision-makers might come up with rather different sets of weighting factors for the same attribute. Therefore, there is a need for a systematic assessment of weighting factors and a simple scaling method is proposed in the following.

Weighting factors for major attributes

The primary or major attributes considered in are the *economic*, the *health related* and the *social* attributes, which are difficult to determine as they are ‘measured’ in different units. The methodology used here is to establish conversion/scaling constants between the weighting factors that can be expressed as:

$$\frac{w_{\text{economic}}}{w_{\text{health}}} = C_1 \quad \text{and} \quad \frac{w_{\text{social}}}{w_{\text{health}}} = C_2$$

The sum of the weighting factors for the major attributes should be 1:

$$w_{\text{economic}} + w_{\text{social}} + w_{\text{health}} = 1$$

which would determine the weighting factors as:

$$w_{health} = \frac{1}{1 + C_1 + C_2} \quad \text{and} \quad w_{economic} = \frac{C_1}{1 + C_1 + C_2} \quad \text{and} \quad w_{social} = \frac{C_2}{1 + C_1 + C_2}$$

The value of C_1 can be determined from the following ratio if the population is exposed only to ionising radiation:

$$C_1 = \frac{w_{economic}}{w_{health}} \cong \frac{w_{economic}}{w_{dose, pop}} = \frac{R_{economic}}{\alpha \cdot (R_{dose, pop} + R_{dose, work})}$$

The parameters R_{dose} and $R_{economic}$ denote the range of the collective doses to the affected population and the range of monetary costs, including the equivalent cost of the collective dose to the workers engaged in the remediation, over the remediation options, respectively. If the affected population is exposed also to non-radiological carcinogens, e.g. heavy metals, the total detriment in terms of collective loss of life expectancy from cancers attributable to the combined exposure should be determined.

With respect to the scaling factor, C_2 , it has been indicated in section 5.5 (Assessment of social factors) that, if it were possible to determine the risk of psychological effects per unit residual dose, r_{psy} , in terms of loss of life expectancy, this factor could be determined as:

$$C_2 = \frac{w_{social}}{w_{health}} \approx \frac{r_{psy}}{r_{rad}}$$

and the value of the scaling factor C_2 in this study could be assumed to be less than 1, e.g. 0.2 - 0.3.

Weighting factors for health sub-attributes

Health sub-attributes in relation to site restoration include health effects from exposure of the population and workers to both radiological and non-radiological carcinogens as well as from accidents due to the remedial measures at the site. The health attributes considered here include *radiation induced stochastic health effects* to the affected population and to the workers. The conversion/scaling constants for the health attributes can be expressed as:

$$\frac{w_{dose, pop}}{L_{dose, pop}} = \frac{w_{dose, work}}{L_{dose, work}} = C$$

where $L_{dose, pop}$ and $L_{dose, work}$ is the range of the collective loss of life expectancy from radiation exposure of the population and from radiation exposure of the work force, respectively. The sum of the weighting factors for the health sub-attributes should be 1:

$$w_{dose, pop} + w_{dose, work} = 1$$

which would determine the scaling constant, C , as:

$$C = \frac{1}{L_{dose, pop} + L_{dose, work}}$$

As the range of collective loss of life expectancy, L , is given as the product of the range of collective exposure, R , the risk per unit exposure, r , and the loss of life expectancy per cancer, l , the weighting factors can be determined as:

$$w_{dose, pop} = C \cdot R_{dose, pop} \cdot l \cdot r_{rad} \approx C \cdot R_{dose, pop}$$

$$w_{dose, work} = C \cdot R_{dose, work} \cdot l \cdot r_{rad} \approx C \cdot R_{dose, work}$$

Weighting factors for economic sub-attributes

Economic sub-attributes include the *monetary costs of the remediation operation* including labour costs, the *monetary costs of waste disposal* including the transport of the waste and *monetary costs of monitoring* the radiological conditions at the site. The conversion/scaling constants for the economical sub-attributes can be expressed as:

$$\frac{w_{remedia}}{R_{remedia}} = \frac{w_{waste}}{R_{waste}} = \frac{w_{monitor}}{R_{monitor}} = C$$

where R_i is the cost range of the given sub-attribute, i over all the different remediation options. The sum of the weighting factors for the health sub-attributes should be 1:

$$w_{remedia} + w_{waste} + w_{monitor} = 1$$

which would determine the scaling constant, C , as:

$$C = \frac{1}{R_{remedia} + R_{waste} + R_{monitor}}$$

The weighting factors can then be determined as:

$$w_{remedia} = C \cdot R_{remedia}$$

$$w_{waste} = C \cdot R_{waste}$$

$$w_{monitor} = C \cdot R_{monitor}$$

Weighting factors for social sub-attributes

The basic social sub-attributes considered in this study include *reassurance* and *disturbance*. The conversion/scaling constants for the social sub-attributes can be expressed as:

$$\frac{w_{reas}}{w_{distur}} = C$$

The sum of weighting factors should be 1:

$$w_{distur} + w_{reas} = 1$$

which would determine the weighting factors as:

$$w_{distur} = \frac{1}{1+C} \quad \text{and} \quad w_{reas} = \frac{C}{1+C}$$

It is assumed that reassurance is given a considerably higher weight than the weight given to disturbance and it is proposed here that $C \approx 5 - 7$.

Further research studies are needed before qualified value settings of weighting factors for social sub-attributes can be done. Such research should be performed in close collaboration between experts in the fields of radiation protection and social and psychological sciences.

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6. Modelling approaches and tools

6.1 Impact assessment model

6.1.1. General scheme

The modelling applied to assess the impact on the population in the RESTRAT project is based on compartment theory. This implies that the site concerned is divided into a number of physical defined areas or volumes, i.e. compartments, between which transfers of pollutants can take place. The transfers of contaminants between the compartments are described by rate constants, also called transfer coefficients, which express the fractional output (turn-over) of the contaminants from the compartments per unit of time.

A generic compartment structure is presented in Figure 7. Possibly relevant processes bringing about releases and subsequent transport and transfers of radionuclides in the biosphere are indicated.

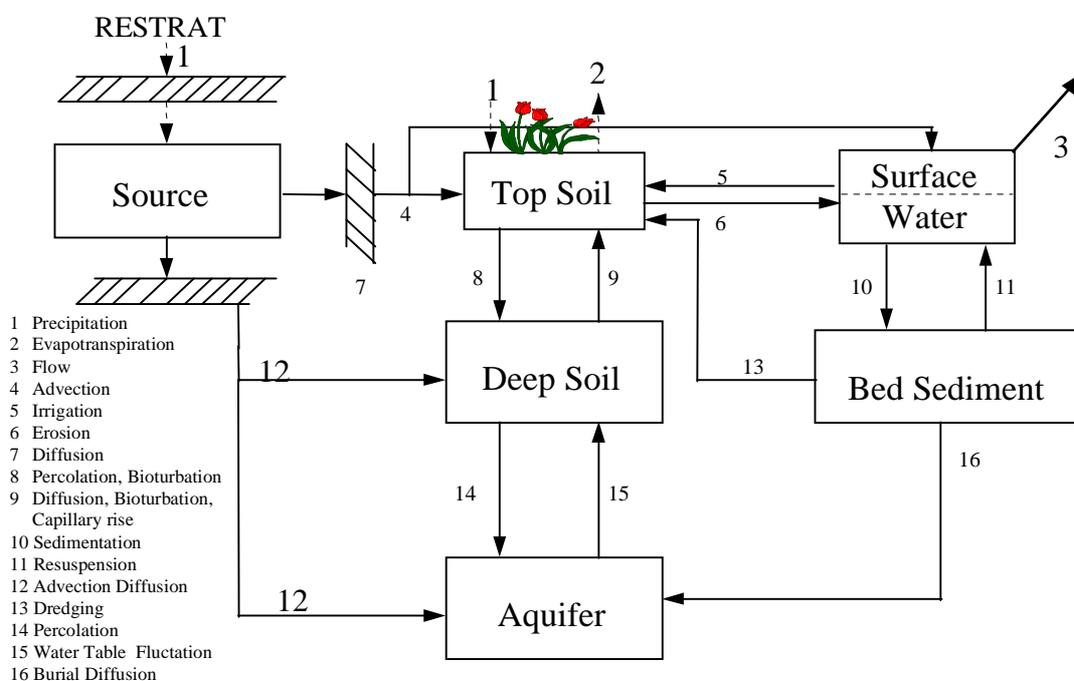


Figure 7 : Generic compartmental scheme

Basic compartments of a simple, generic compartment biosphere model include :

- terrestrial media : top soil, deep soil, vegetation, surface air, (animals);
- aquatic media : surface water, bed sediment, aquifer, (fish).

The source can be situated in one of these compartments or may constitute a separate compartment.

For each example site in the RESTRAT project a compartment system has been developed. They are shown in chapter 7. The compartment models are elaborated with BIOPATH. This is a general tool (Bergström *et al.*, 1982), which can be used for varying types of compartment models, as long as they

are based upon first-order differential equations. Mathematically, this is expressed by a set of first order linear differential equations with constant or time varying transfer coefficients (rate constants).

The general assumptions for compartment models are that :

- the outflow of an element from a compartment is only dependent on the quantity of the element in that particular compartment;
- each compartment is instantaneously well mixed;
- all elements within a compartment have the same probability of leaving the compartment.

Instantaneous and homogeneous mixing of the compartments is a valid assumption in cases where the time studied is long compared with the staying time of the elements within the compartments. Such ideal compartments are of course a simplification of the real situation, but in general, the compartments can be conceived adequately in order to represent reality with satisfactory accuracy. Compartment models have shown to be reliable for modelling the transfer of trace elements in the biosphere, as has been demonstrated in several international model validation studies, such as the Biospheric Model Validation Study (BIOMOVS, 1996), and Validation of Environmental Model Predictions, VAMP (International Atomic Energy Agency, 1993).

The amount of elements (radionuclides or other contaminants) in a given compartment is dependent on:

- the source term for the system, such as the direct release to one or several compartments, or generation within them by decay from a parent substance;
- the outflow to and inflow from other compartments;
- decay, degradation or losses to sinks.

In the BIOPATH code the compartments are considered as reservoirs and the relationship between the amounts of a contaminant in the reservoir system is expressed mathematically in vector form as:

$$\dot{Y}(t) = K \cdot Y(t) + Q(t) - \lambda \cdot Y(t)$$

The vector \dot{Y} refers to changes per unit time of the inventory Y of the contaminant in the different reservoirs of the system at time t . The coefficient matrix K (year^{-1}) describes the transfer rates between the reservoirs and the matrix $Q(t)$ (amount/year) constitutes the source term to the reservoirs.

$$\lambda = \ln(2)/T^{1/2}$$

expresses the decay constant, where $T^{1/2}$ is the physical half-life of the contaminant.

The BIOPATH code includes different methods for solving the equation systems. The method used in the RESTRAT project is LINDIF (Forssén, 1977) which is a semianalytic solution method, in which the solution is derived from the eigen vector of the coefficient matrix. In theory, this method will give an exact solution if the source term is constant. Speaking in terms of radionuclides, this implies in practice a single nuclide. Furthermore, if the coefficients of the matrix vary too much, the differential equation system will become stiff and the rounding errors will bring about large inaccuracies. If the time-step in the calculation is sufficiently short in comparison with the half-life of the nuclides it will give acceptable results even for chains of radionuclides.

6.1.2. Release and transport processes

For the estimation of the source term in the dose assessment, information about the inventory and physical/chemical properties of the contaminants (nuclides) has to be made available. This kind of information is shown in chapter 3 (classification of the sites).

The main process for release and migration of radionuclides from the site is the transport with water. This can occur either via percolation of rainwater through a repository or via contact of the contaminants with a water-body (contaminated sediments or groundwater in contact with a repository). Furthermore, weathering and physical transport of sediment can be important processes for the transport out of the contaminated site.

A very important feature in the transport of the nuclides is the distribution of the nuclides between the aqueous and the solid phase. This distribution is characterised by the K_d parameter, which expresses the fraction of element in solid form relative to the fraction in soluble form. This implies that an immediate steady-state condition is achieved, the circumstances of which can be discussed. Default K_d -values are valid for most nuclides but in order to increase the reliability in the assessment, site specific K_d -values should be used (section 6.2).

Radionuclides released from the contamination source or repository will participate in the ecological cycle within the biosphere. This implies that they will be transported and exchanged between various biosphere components, such as water, sediments, soil and biota.

An adequate compartment scheme has been put together for each example site, based on the physical situation and potential exposure scenario. Simplifications have been made in order to get a useful compartment scheme. A short description follows on the transport processes, which must be considered when developing a compartment system for dose assessments.

The flow of ground- and surface water are the main processes resulting in the transport of pollutants. Other processes are atmospheric transport, biological transport due to animals and finally interaction by man.

6.1.2.1 Transport in groundwater

The groundwater is treated as one compartment where the transport (transfer of the nuclides out of the compartment) is dependent on the water flow and the sorption of the nuclides in the solid phase. The transfer coefficients are then determined by the water turnover rate and the distribution coefficient K_d . This is a rough simplification but can be considered valid for continuous release over long periods and short transport distances. This is often one of the major transport processes from the contamination source.

Transfers of the nuclides out of the groundwater compartment may also be brought about by extraction of groundwater for various uses, such as for irrigation and drinking water purposes.

6.1.2.2 Surface waters

There are three main processes for the transport of radionuclides or other elements released to surface waters. These are outflow, transfer to the sediments and extraction of water for various uses, such as irrigation and drinking water. The first process is in general the most effective in depletion of the contamination levels in the water.

Radionuclides in sediments may be transferred back into the water column through resuspension and in general it is only a small fraction of surface water that is used for irrigation, watering cattle or drinking. Moreover radionuclides deposited onto soils through irrigation migrate to groundwater which is drained

into surface waters. In addition erosion of soil particles to which radionuclides are attached may cause a transfer back to surface waters.

- Water turnover

Water turnover (outflow) causes a transport out of the system of nuclides in soluble as well as in particulate form. The flows have in general seasonal fluctuations with maximum values during spring and autumn. In dose assessment of long periods these calculations can be performed using annual mean values.

- Transfer to sediments

Elements in water will adsorb on particulate matter causing a transfer to the sediments. They may also be transferred to the sediments by diffusion or bioturbation. Earlier studies have, however, pointed out that particle settling is the main process for the transfer of relatively immobile radionuclides to the sediments (Bergström and Nordlinder, 1991; Nordlinder *et al.*, 1996). This is therefore the only process needed to be considered as most radionuclides of interest can be classified as relatively immobile. The effectiveness of this process is among other things dependent on the element's properties and mass sedimentation rates. The adsorption to suspended matter is described by use of the K_d -parameter. This implies that an immediate steady-state condition is achieved, the circumstances of which can be discussed (as was mentioned earlier). The K_d -values depend strongly on the characteristics of the water and the sediment and on the nuclide considered. Values found in the literature are varying over several orders of magnitudes.

- Transfer from sediments

The behaviour of radionuclides in sediments is among other things dependent on the conditions in them. All the chemical processes were simplified so that all radionuclides transferred to the sediments were assumed to be effectively retained. The losses of radionuclides from the sediments were only due to physical processes such as resuspension and growth of sediments.

Resuspension will cause a transfer of the nuclides from the sediments back into the water column. It has been shown to be a major process for maintaining increased levels of radionuclides in water for shallow lakes (Sundblad *et al.*, 1991).

The settling of fresh sediments on top of the existing layer results in a transfer of radionuclides to deeper sediment layers in compartment models, as the mass of the top layer remains constant. However, this will not always be the case, especially when dredging of sediments is carried out. The sediments can then be put on soils leading to an effective transfer of radionuclides to these soils.

6.1.2.3 Agricultural soils

- Transfers to soil

When surface- or ground water containing radionuclides are used for irrigation, the radionuclide fraction which is taken up in the soils leads to the contamination of the soil.

Also dredging of sediments from surface water bodies may bring about contamination of the soil. If the sediment is applied for amending agricultural soil, this may be the most important transfer of nuclides to soil.

- Transfers from soil

Nuclides deposited on soils migrate downwards at various rates dependent on the properties of the soil and on the effectiveness of the element's sorption on solid matter. Diffusion and bioturbation lead to a transport from upper soils to deeper situated layers from which they can be transported back to surface water by ground water runoff. Erosion by wind and water may cause a transport to surface water as well.

In the models set up in this project, advection of dissolved radionuclides with infiltrating water was taken into account as the only process responsible for the loss of radionuclides from the upper soil layer. It is expected that neglecting the other processes may lead to conservative estimates of the concentrations in soil.

6.1.2.4 Biological uptake

Radionuclides may be transferred to various types of biota from the media such as soil and water. The calculation of the resulting concentrations in the biota is based on steady-state conditions between the media and the biota. This is valid for long-lived nuclides within the biosphere and for mean values over a long period or for integration over long times.

Nuclides in soil are partly transferred to plants via root uptake. This uptake is described by the use of a root uptake factor also called soil-to-plant concentration factor, which gives the ratio of the concentration of the nuclides in the plant to the one in the corresponding dry soil. Data for different types of plants are available for most elements, though the values can vary considerably for the same element. The uptake is, among other things, dependent on soil characteristics. There is a spectrum of parameters governing the root uptake such as the vegetation type, type of soil, pH, organic matter, climate and the element-specific properties.

Nuclides in irrigation water are also transferred partly to plants through direct deposition. In this case the concentration of the radionuclides in the plant can be calculated by means of the interception fraction and taking into account a decay through subsequent weathering, characterised by a weathering decay constant. For some types of food crops translocation has to be considered of the radionuclides from the outer surface of the crops to the edible parts. The fraction of the radionuclides reaching the edible part at harvest is given by the translocation factor.

Concentration of radionuclides in sea food are obtained by use of concentration factors from total concentration in water to edible parts of the sea food species. Those factors are based on empirically found data and are valid for steady-state conditions. They implicitly include all transfer pathways from the surrounding environment.

Radionuclides in water, soil or plants may be transferred to livestock through watering, feeding or grazing. From the intakes of the animals, their milk and meat will become contaminated.

The concentrations of the radionuclides in milk and meat can be calculated by using milk and meat distribution factors, giving the fraction of the radionuclides daily ingested, appearing as concentrations in milk and meat respectively.

6.1.2.5 Influences of restoration options

The effects of restoration techniques may be given in different ways, depending on the type of the technique.

If the technique consists of removing contaminated media, its effect is characterised by a so-called decontamination factor (Df). The source term is to be divided by the Df-factor in order to reduce it in proportion to the effect of the restoration technique.

When containment systems (capping and subsurface barriers) are considered, the effect of the technique can be expressed by the permeability of the barriers, characterizing the amount of water infiltrating through the barriers. Allowance has to be made for the partly insaturated conditions of the barriers.

The application of immobilisation techniques will reduce the release and mobility of the radionuclides in the media treated. This can be taken into account by a decrease of the leachable fraction or an increase of the K_d value of the nuclides concerned.

6.1.3. *Exposure pathways*

As explained in section 5.2, the outcome of the impact assessment will be doses. Both individual and collective doses to the restoration workers as well as to the general public are to be assessed. However the doses to the restoration workers can be calculated straightforward from the contamination levels and do not require a complicated biosphere model such as BIOPATH.

The exposure of man can be external or internal.

External exposure can be due to contamination of surfaces of different types e.g. soil/sediment or surface water, by direct contamination of the surface of human bodies or by contaminated particles in the air. The external doses usually plays a minor role compared to the internal doses. For nuclides with high gamma energies, as cobalt-60 and caesium-137, external irradiation is an important pathway.

The internal doses are due to radiation from nuclides within the human body. The nuclides reach the body via intake of food and water, or due to inhalation. The nuclides will be either eliminated or will participate in metabolism in the body, dependent on its chemical and physical properties.

Exposure pathways which have been considered as being potentially relevant for the example sites are:

- consumption of contaminated water;
- consumption of milk and meat contaminated through the watering of the cattle;
- consumption of milk and meat contaminated through the grazing of the cattle of pasture from contaminated soil;
- consumption of fish and shellfish from contaminated surface waters;
- consumption of garden and agricultural products, contaminated through irrigation or amendments to the soil;
- inhalation of contaminated aerosol;
- external irradiation on contaminated fields or banks of surface waters, or in contaminated water or air.

Due to site specific situations additional exposure pathways may exist.

For each of the exposure pathways, the consumption habits and exposure durations of the critical group have been evaluated.

6.1.4. *Uncertainty and sensitivity analysis*

As explained in section 5.3 uncertainties of the endpoints of an impact assessment model may be due to scenario uncertainty, model uncertainty an parameter uncertainty. Also uncertainties due to computational errors could be added to this list.

Firstly, two terms of importance with respect to the confidence in model results have to be defined: verification and validation. The former term applies to the accuracy in the numerical methods, which are used in the mathematical codes applied in the models. The codes need to be verified before entering the step of validation when the model results are compared and evaluated against independent data sets. Verification is necessary to avoid programming mistakes etc.

With respect to uncertainty due to computational errors, verification tests have shown that they are of much lower importance than uncertainties due to conceptual modelling and parameter values. This has been shown in several verification studies, where the solution methods applied in BIOPATH have been tested. Both BIOPATH and PRISM, have been parts of a total model intercomparison where all models used the same expressions and parameter values for all rate constants and where doses to man were calculated from a variety of exposure pathways (Klos *et al.*, 1992).

Uncertainties due to specification of the scenario involve the whole methodology applied for the assessments. One simplification is that the scenarios describe constant conditions over time, i.e. no evolution of the biosphere is considered. There are high uncertainties coupled to any prognoses of future states of the biosphere and also of future behaviour of man.

Elements of model uncertainty include the division of the ecosystem into compartments between which there is an annual transfer of water and other material. Each compartment was assumed to comprise a physical area with uniform chemical and physical properties. Radionuclides were also assumed to be homogeneously distributed within the compartments. This is not always the case in reality. Simplifications and assumptions in process descriptions were also contributing to model uncertainty.

The uncertainties in results mostly considered are those due to parameter uncertainty. These uncertainties are relatively easy to quantify by use of error propagation methods, in contrast to uncertainties due to conceptual modelling.

In respect of their uncertainty, parameter values are to be classified according to their origin or information source, from which they are derived. In order of decreasing preference, (i.e. increasing ranges of uncertainty associated with the estimates of the parameter values) they can originate from measurements on the site, from a literature review of experimental data, or taken by default (generic values).

Data originating from measurements or investigations on the site considered are the most valuable. Parameters for which site-specific data should be available concern for instance the type and inventory of pollutants present, habits of the local population (characteristics of the critical group), turnover in surface water.

Where local data are not available or not representative enough, data from a literature review of field experiments on sites or in conditions, that are similar to the one under study can be applied. Meteorological data, determining infiltration rates, production data of agricultural products and many other transfer parameters in the biosphere are examples of parameters, where such data may be considered to be appropriate.

If no site-specific values or experimental data from a literature review are available, one has to have recourse to default or generic values, which may come for instance from other impact assessment models. Also expert judgement and information extracted from elicitation exercises may be useful. Dose factors are examples of parameter values for which the generic values available are widely used.

Not only the site-dependency, but also the variability of parameter values with time has to be allowed for. In this respect the periods over which the assessments are to be carried out, are important. For assessments over 100 or 500 years, such as in this study, the estimations of the ranges of parameter values should not be based on seasonal variations for instance, but on annual averages.

For characterizing and quantifying the uncertainty ranges of the parameter values, probability density functions (pdf) are used.

If only a very limited amount of data is available, to such an extent that only minimum and maximum values can be derived, uniform distributions are most appropriate. In case still less data are available, for instance one generic or default value, experts in the specific area should be asked to give their advice and a conservative approach should be adopted.

If next to the minimum and maximum values, also a most probable value may be estimated (from the literature or from site-specific data), a triangular distribution can be applied.

For the parameters for which an abundant amount of data are available, normal distributions are adequate.

Linear and logarithmic distributions are being used. Logarithmic distributions (log-uniform, log-triangular, log-normal) are preferred when the ranges between the minimum and maximum values or over four standard deviations would cover substantially more than one order of magnitude.

In this study, the PRISM model is used for the uncertainty and sensitivity analysis. PRISM is a general tool for addressing the uncertainties in any model due to the uncertainty or variability in parameter values (Gardner et al., 1983). The PRISM system consists of three main subprograms, each one described below:

- In PRISM 1, random parameter values are generated by using a systematic sampling method, Latin Hyper Cube. As input to PRISM 1, the mean values or best estimate, type of distributions, standard deviations and the upper and lower limits are given for each parameter. These data are then used to define probability density functions. The Latin Hyper Cube method, used to generate the sets of values from the given distributions, is an efficient Monte Carlo sampling technique which produces random values within the whole desired range. In addition, correlation between model parameters can be taken into account, irrespective of the type of distribution the parameter values are drawn from.
- In PRISM 2 the model is run for each set of input parameter values generated with PRISM 1.
- PRISM 3 statistically evaluates and summarises the joint set of model parameters and predictions. The general statistics for the distribution of each parameter and the response of the model to this distribution contain the following: arithmetic mean, standard deviation, coefficient of variation, geometric mean, percentiles (5, 25, 50, 75, and 95%), and the five highest and five lowest values, respectively.

In order to evaluate the influence of changes of parameter values on the model output, correlations between the model parameters and the responses as well as between the responses themselves are also considered. Two correlation coefficients are calculated: the simple Pearson correlation coefficient, and Spearman Rank, which is the correlation of the ranked values of the parameters and model responses. Associated with each correlation coefficient is their percent covariation (COVAR). This represents the percent variance that one variable accounts for in another variable or response. In the cases of correlated model parameters and responses, percent COVAR indicates the amount of variability in the model response that is explained by the variability of that particular model parameter. The regression procedures are used to obtain the relationship between model parameters and model responses. The parameters to be entered into the regression analysis are selected from those, which give the greatest improvement on the sum of squares of regression. From these analyses the relative contribution to the total uncertainty from each parameter is obtained. Furthermore, parameters and processes contributing significantly to the uncertainty in results can be identified.

The software of the impact assessment models for the example sites can be found in the accompanying CD-ROM (Brendler, 1999). The sets of input parameters necessary for the impact calculations are also included.

6.2 Chemical speciation

6.2.1. Physico-chemical Site Characterization

Measurements of physico-chemical site characteristic are, in many cases, far from being trivial. A good introduction to the matter is given in Chapter 9 - "Geochemistry and the design of sampling programs" in (Deutsch, 1997). Other relevant monographs are also recommended (Broekart *et al.*, 1990; Manahan, 1994; Fränze, 1993).

The most critical step is the very first one, the sampling itself. Many natural environments are very heterogeneous systems making it difficult to collect representative samples. Thus, large sample sets are required to obtain error estimates. Major chemical components may also falsify some analytical results due to matrix effects. But even more important is the fact, that such natural systems are sensitive to small changes in external parameters like temperature, pressure or oxygen content. Collecting a probe from a natural system or installing a sensor or other devices in it to perform measurements on site means a more or less serious disturbance to the system. The next step, taking the sample to an analytical laboratory for further analysis presents an even greater danger of changing the sample irreversibly. The greatest care, therefore, has to be applied to the sampling procedure, with preference given to *in-situ* determinations of sensitive parameters such as pH, gas content (oxygen, carbon dioxide) or redox potential.

In any case it is worthwhile consulting environmental or analytical chemists, geochemists or mineralogists before, during and after sampling campaigns, analytical data generation and processing.

6.2.2. Software Selection

The processes mainly influencing the species distribution can roughly be grouped into three categories:

- reactions in homogeneous solution (redox reactions, hydrolysis, complexation, etc);
- the formation of pure and mixed solid phases;
- and reactions on mineral surfaces.

The modelling of chemical speciation has to take them all into account, which does not pose a problem with regard to the first two categories; nearly all programs available now can handle them, including redox reactions. When it comes to the third category, many programs cannot cope or can just offer the simplistic K_d approach, that has already been critically discussed in section 5.2.2. Finally, two programs were selected for the speciation modelling part in the integrated risk assessment framework. Both are available as source code, which is essential for any adaptations which are necessary to create interfaces between chemical modelling and risk assessment modules. Moreover, they have been in use for many years, have been checked by a number of validation programs and are recommended by international organizations. They are both discussed in more detail below.

6.2.2.1 EQ3/6

Developed at the Lawrence Livermore National Laboratory of the U.S.A. (Wolery, 1992), the geochemical speciation code EQ3/6 rapidly became a wide-spread application. Government institutions especially favour it because the program underwent a rigorous verification procedure in connection with its use for the Yucca Mountain Site Characterization Project (YMSCP), leading to a certificate that approves its use in quality-affecting work (August 17, 1994). The program is available in source code and runs under both UNIX and PC operating systems. It is supplemented by a very exhaustive set of manuals.

The program is actually a suite of modules. EQPT is dealing with the database management. EQ3NR computes any equilibria in homogeneous solution (hydrolysis, complexation, redox reactions), and can also handle redox disequilibria. Components can be defined to be in equilibrium with a mineral or a gas

phase, the charge balance is always maintained. It does compute saturation indices, but oversaturated minerals are not precipitated. EQ6 performs the computation of reaction pathways with mineral precipitation governed by kinetic rate laws. Minerals can be pure phases or solid solutions. The following reaction pathways (each with temperature jumps) are possible: fluid-centred flow-through open systems, closed systems, and titrations (including mixing of two solutions).

EQ3/6 is accompanied by several thermodynamic databases that are well-documented and can be changed or expanded without difficulties. Some of them can be applied also to high ionic strengths, utilizing the Pitzer model for activity coefficients in electrolyte solutions.

Its main limitations are the lack of any sorption model and inability to do parameter scans (to generate 2D-speciations in EQ3NR). Furthermore, contrary to EQ3NR, EQ6 does not allow redox disequilibrium. Finally, the performing speed is rather low compared to other speciation codes, hampering its use as a subroutine in more complex programs.

The software suite was primarily used for the quality assessment of the primary analytical dataset, and to obtain good starting estimates for the speciation modelling inside the integrated PRISM/BIOPATH/MINTEQA2 framework.

6.2.2.2 MINTEQA2

The chemical speciation software MINTEQA2 (Allison *et al.*, 1991) is called a “geochemical assessment model for environmental systems” by its authors. It is distributed by the US Environmental Protection Agency (EPA), Center of Exposure Assessment Modeling. The software is based on the well-known MINEQL code (Westall *et al.*, 1976).

The software has the following features:

- computation of chemical speciation for the homogeneous aqueous phase or for mixed phase systems, including simultaneous consideration of various minerals and a gas atmosphere;
- fixed activities (bulk mineral, indefinite atmosphere etc.) can be defined;
- it can also handle redox reactions;
- sorption phenomena can be dealt with by means of seven different sorption models, incorporating three surface complexation models (SCM): the Diffuse Double Layer model, the Constant Capacitance model and the Triple Layer model;
- it is available in source code format (FORTRAN77) with special adaptations to UNIX systems and the MS-DOS operating system for PCs. These system-dependent parts are rather small, so exporting of the software to other computer systems should be straightforward;
- an external thermodynamic data base (with its own proprietary format) is distributed together with the software, including some sorption data and organic compounds, which is mainly based on the WATEQ3 (Ball *et al.*, 1981) data base as developed by the U.S. Geological Survey;
- there is a separate user interface program for interactive creation of data input files (problem description files) available.

Its main limitations are the lack of a kinetic mode, an inability to handle solid solutions, and shortcomings in scope, quality and documentation of the internal thermodynamic database. The latter can of course be overcome by setting up an one's own database, or, at least, by carefully checking and correcting of the one accompanying MINTEQA2. Also, the convergence of the numerical algorithm for the solution of the mass equilibria equation system is not always guaranteed. This is especially the case where the default master species for an element is actually not present in measurable amounts in the real system under investigation. An example is the modelling of systems under reducing conditions containing uranium. The pre-set master species for uranium is the uranyl cation, which at redox potentials below 50 mV is hardly present. Numerical convergence can be drastically improved by delivering starting estimates for such basic species of at least two orders of magnitude close to their real free concentration. Occasionally another peculiarity can be observed when a mineral is marked as

excluded from consideration that is not the thermodynamical stable one in the defined chemical system. Here it may become necessary to also exclude the more stable minerals containing the same element as the excluded one.

The MINTEQA2 code was initially chosen for incorporation into the risk assessment code because of its handling of surface complexation as the most dominant process directly effecting distribution coefficients. The software for this code as it is applied to the example sites, is present in the accompanying CD-ROM (Brendler, 1999).

6.2.3. Database Selection

During the RESTRAT project work, various databases have been scanned for their respective thermodynamic data concerning hydrolysis (if applicable), complexation and precipitation / dissolution reactions:

- OECD Nuclear Energy Agency - Thermodynamic Database (NEA TDDB) Recommendations for uranium as of 1992 (Grenthe *et al.*, 1992) with the corrections included in the corresponding americium volume (Silva *et al.*, 1995);
- LLNL Gembochs COM Data Base in Version 8 Release 2, as distributed with EQ3/6 Version 7.2b from August 1995, it includes also the NEA TDDB values for uranium (Wolery, 1995) and some extensions based on experimental results from the FZ Rossendorf;
- LLNL Gembochs ALT Data Base, which is in large parts identical to the COM database, but already taking into account the NEA TDDB recommendations for neptunium and plutonium (Wolery, 1995);
- NAGRA Chemical Thermodynamic Database: Redox coupled version for EQ3/6 and PHREEQE, version from August 1994 (Pearson and Berner, 1991; Pearson *et al.*, 1991; Pearson, 1994);
- NIST Critically Selected Stability Constants, Version 2.0 from October 1995 (Smith *et al.*, 1995);
- CHEMVAL 6.S database as described in: CHEMVAL 2: Final Report EUR 16648, 1996 (Falck *et al.*, 1996);
- ZZ-Hatches database, NEA Release 9 from November 1996 (Cross and Ewart, 1991; Bond *et al.*, 1992; Bond *et al.*; 1996).

In the RESTRAT project, the chemical speciation modelling was primarily based on the LLNL Gembochs ALT database. This data base was corrected for several erroneous entries, aqueous complex species such as $\text{Ca}_2\text{UO}_2(\text{CO}_3)_3$ (Bernhard *et al.*, 1997) and $\text{UO}_2(\text{SO}_4)_3^{4-}$ (Geipel *et al.*, 1997) were added. Databases necessary for using the MINTEQA2 speciation codes for the example sites are included in the accompanying CD-ROM (Brendler, 1999).

In certain cases, even a comprehensive literature and database survey may not yield sufficient information for a proper chemical speciation modelling. Then data approximation and estimation can help to proceed with the task. The simplest forms are chemical analogues, i.e. using data from lanthanides for trivalent actinides, or data from cerium(IV) to model plutonium(IV). A next step would be the introduction of correction terms based on trends in ion size, charge density, electronegativity, effective dielectricity, ionization energy, crystal lattice parameters or solubility enthalpies in case of solids. In some cases, *ab-initio* calculation of thermodynamic parameters based only on atomic properties may be possible, or estimations based on some model theories, such as successfully applied to the prediction of free energies of formation for several mineral phases in the U(VI)-SiO₂-H₂O system (Clark *et al.*, 1998).

In special cases, when even data estimation fails, it may even be necessary to perform own experimental studies to fill critical data gaps. More detailed information about requirements in connection with thermodynamic data, focussed on radionuclide, can be found in another EC project report originating from the JETDEM concerted action (Fanghänel *et al.*, 1999).

6.2.4. Quality Assurance

Any data consistency and reliability check of the experimental analytical data demands a solid thermodynamic data foundation. Therefore, the very first step is to establish a sound thermodynamic database for the relevant problem. The emphasis must be on the completeness of basic species, aqueous complexes, and minerals, and on the correctness and accuracy of thermodynamic data.

Then, field and laboratory data can be checked for internal consistency to a certain degree by means of chemical speciation modelling. This in fact should be done, because in almost any real-world application case the initially available experimental data set must be expected to be sparse and partly erroneous. Speciation modelling will help to establish a verification and ranking for all necessary information. Then it can also be decided whether and what additional experiments need to be carried out.

The speciation modelling output must be screened with regard to several items:

- most easily detected are any charge imbalances that indicate either wrong analytical concentrations for a major anion (a suspicious candidate here is the carbonate content) or cation, or that such a major constituent of a solution has not been analysed at all;
- some programs allow the independent computation of a system pH based on its composition, this then has to be compared with the experimental value;
- a measured “global” redox potential of the system should be probed by independent information about various redox couples, and be compared with measured dissolved oxygen contents;
- the next blocks of information from speciation modelling are saturation indices for precipitated minerals. This must be compared with findings about the geochemical and mineralogical composition of the solid matrix in contact with the aqueous phase. But it must be kept in mind, that simultaneous chemical equilibrium is the exception to the rule in natural systems, even on large time-scale. This is especially true in heterogeneous phase reactions, such as precipitation and dissolution. Modelling must take this into account, e.g. by applying kinetic rate laws (when available) or at least by excluding some kinetically-hindered minerals from consideration.
- the last point to be mentioned here is the computed partial pressure of CO₂, that can indicate equilibrium between solution and the atmosphere, but also wrong analytical values for the carbonate content.

After such single-point speciation modelling a screening of the speciation variation should follow. This should be considered as a kind of primitive sensitivity analysis to identify those components with still critical uncertainties. It means scanning the speciation as function of the most influential parameters, usually pH, Eh, carbon dioxide partial pressure, and the concentration of some major complexants for the contaminant under consideration. Additionally, the information yield from chemical speciation can make the whole source term description more realistic, i.e. by specifying some components to be in equilibrium with either a mineral that is in excess or a large open gas reservoir (namely CO₂ in the atmosphere), with the welcome side-effect of reduction of the enormous parameter space.

To summarize, the following steps are to be taken when applying chemical speciation modelling to verify the chemical source term model used in risk assessment (Figure 8 depicts some important ones):

- collect thermodynamic and analytical data sets;

- check of analytical data consistency with chemical speciation modelling;
measure suspicious data again, compute missing data;
- identify system constraints: equilibria with minerals or gas phases;
- screen speciation as a function of important physico-chemical parameters;
- compute K_d values;
measure suspicious mineral composition and properties again;
- perform a full sensitivity analysis and identify most influential parameters;
increase data accuracy for these parameters;
- run the complete risk assessment model.

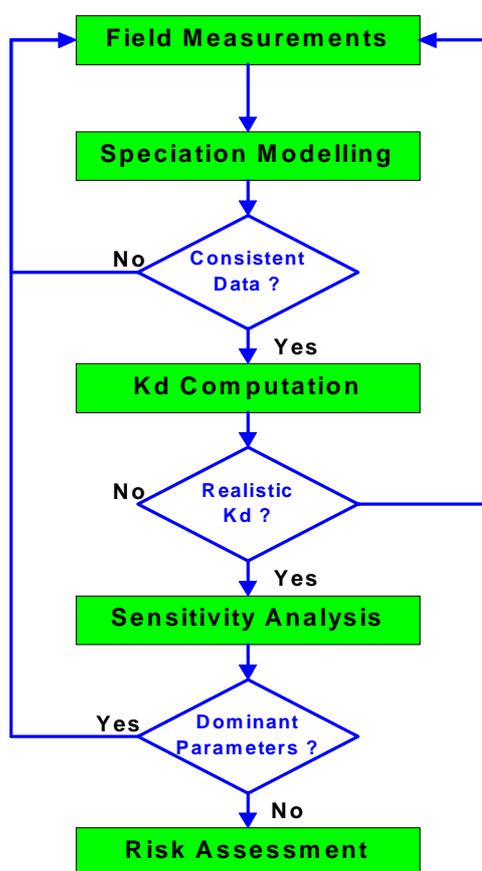


Figure 8 : Chemical speciation modelling for iterative refinement of analytical data sets

6.2.5. Application Examples

In order to illustrate the approach outlined in the previous sections, the development of the data set for the chemical speciation in one of the Ranstad Tailing site compartments is given. The compartment structure (see chapter 7, section 7.1.4) includes several compartments with an aquatic phase, all of them were characterised. Namely this involves five compartments: ground waters from the tailings layer, the moraine layer, the limestone layer, and surface waters from the collecting ditch on the west side of the tailing and from the so-called M-lake (Magasineringsjön) storage reservoir. From here on, only the tailings layer will be considered.

6.2.5.1 Primary Analytical Data Set

There have been extensive analytical campaigns starting as early as in 1965, with enforced activities after 1992 continued until present. Most of their results are reported in (Sundblad *et al.*, 1996). The data were mainly obtained from on-site bore hole samples. Available primary (measured) data sets include:

- annual average values of pH, temperature, conductivity and a number of anion and cation concentrations for surface and lake water;
- metal and radium concentrations of groundwater samples;
- precipitation and temperature curves;
- annual curves of water discharges, pH and contents of sulphate, iron, nickel, cadmium, aluminium, manganese, magnesium, calcium, and uranium at various stations (surface, lake and ground water).

Based on these data sets, other values were computed, such as the amount of transported metals per year. The analytical values relevant for the aforementioned five compartments were taken from this data pool, supplemented by some additional material from Studsvik.

The characterisation of the relevant solid phases is not as satisfactory as that for the aqueous phase. There are only rather general mineralogical investigations for this site published, which are supplemented by some elementary analysis of the tailings layer. Analysis is complicated by a considerable content of organic matter of various origin and in different state of degradation.

From the available data it can be concluded, that the tailing layer is built from leached alum shale, mostly consisting of quartz, illite, and feldspars, with a significant content of pyrite (13%) and organics (22%).

6.2.5.2 Supplementary Measurements

A first check of the data pool revealed several general features:

- distribution over a wide range, even for samples drawn at the same day from probe bore holes just 100 m apart;
- unusually high values for iron and aluminium, to a lesser degree this is also valid for lead, nickel, manganese, cadmium and arsenic;
- no analysis for anions in the groundwater layers;
- there are no details available about the sampling, conditioning, filtering and analytical procedures applied in the analysis.

Therefore, additional measurements were performed as part of the RESTRAT project to close gaps in the previous investigations, and to verify the accuracy of those measurements. During the analytical campaign in August 1996 samples were taken by Studsvik Eco & Safety AB (with the temperature and pH determined *in-situ*) and shipped to FZ Rossendorf, where the analysis was performed. The samples were analysed twice, first without further treatment, and second after filtering, done in three steps for 450, 100, and 15 nm particle size. The following experimental methods were used: ion chromatography, inductively coupled plasma - mass spectrometry, flame - atom adsorption spectrophotometry, γ -spectrometry, pH measurements.

6.2.5.3 Chemical Speciation Modelling for the Tailings Layer

The new results, together with the results from the previous sampling campaigns, were checked for consistency by modelling the chemical speciation, first in a single-point calculation with EQ3NR from the EQ3/6 package. This was followed by parameter scans, again performed with EQ3NR, and finally by reaction path modelling with EQ6.

For all further computations, performed either as scans over a free parameter with EQ3NR, or as reaction path modellings with EQ6, the (kinetically hindered) precipitation of dolomite was suppressed. Also, iron-containing minerals others than $\text{Fe}(\text{OH})_3$ (or HFO - hydrous ferric oxides) were not allowed to precipitate, because their formation kinetics is far too low in relation to the flow rate of the water inside the layers. Also, thermodynamic stable solid phases such as ferrite-Zn, goethite, haematite, magnetite, various nontronites, and trevorite were suppressed in all models because they do not form directly from oversaturated solutions due to kinetic hindering. From the computations the following general conclusions could be drawn:

- The reported dissolved iron and aluminium concentration from the older measurements clearly exceed the saturation limits for many minerals of these elements. Obviously those samples were not filtered properly, so the analytical values incorporated fine-disperse and colloidal material. The values from filtered samples from 1996 show, that both are dissolved iron and aluminium only trace components.
- There are no *in-situ* determinations of the redox state or measurements of the oxygen content in the water available. Therefore, only estimated values for the redox potential in the various waters could be derived using modelled speciation and computing Eh from the concentrations for the redox pairs $\text{NO}_3^-/\text{NH}_4^+$ and $\text{NO}_2^-/\text{NH}_4^+$. This gave a mean redox potential of 346 mV for the tailings layer, equal to slightly reducing conditions, as to be expected from water with contact to reducing minerals like pyrite, and a significant amount of organic matter.
- The water table inside the tailings layer is dominated by sulphates of magnesium and calcium.

In the next modelling step, the uranium speciation for the homogeneous aqueous phase was calculated with EQ3NR: as function of the pH, of the partial pressure of carbon dioxide p_{CO_2} , and of the redox potential Eh. The resulting speciation patterns are shown below in Figure 9.

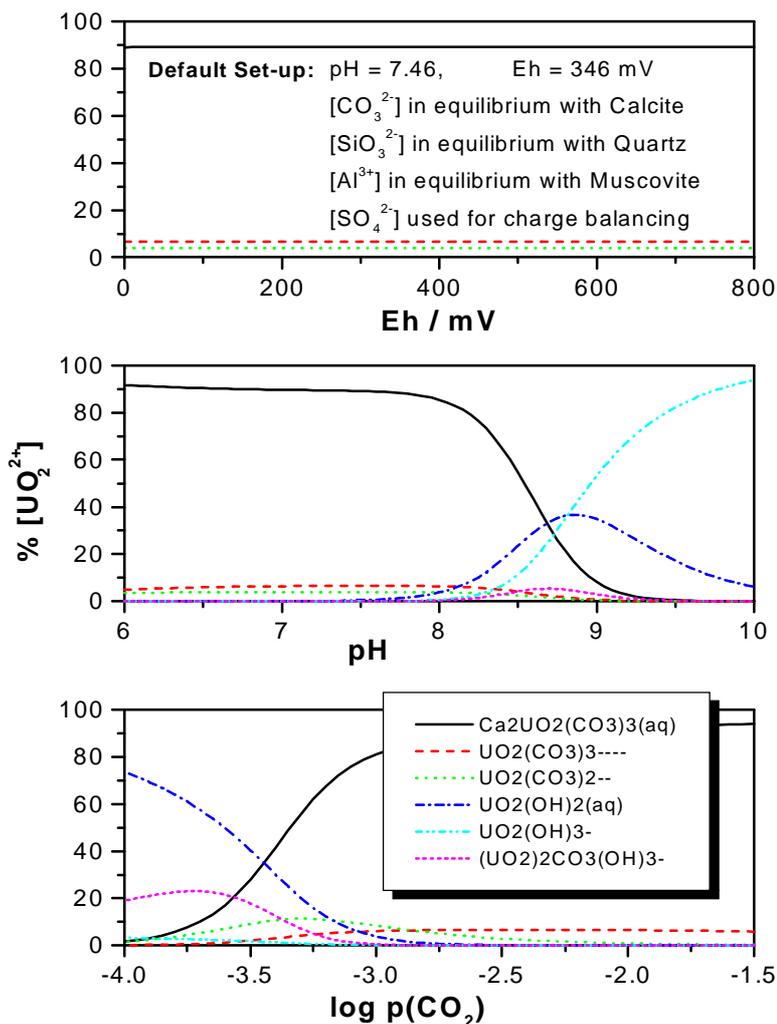


Figure 9 : Uranium speciation as a function of Eh, pH and $p(\text{CO}_2)$ for the Ranstad Tailing site (tailing layer)

First, it became clear that actually the system is redox-indifferent over a wide range of the redox potential, only below an Eh of 0.0 mV the speciation of uranium changes with the appearance of U(IV) species. Thus the approximation of the Eh as discussed above is fully justified. Over large ranges in pH (up to 8.6) and in p_{CO_2} the neutral aqueous complex $\text{Ca}_2\text{UO}_2(\text{CO}_3)_3(\text{aq})$ comprises nearly all of the uranium. At higher pH values a negatively charged hydrolysis species, $\text{UO}_2(\text{OH})_3^-$, becomes dominant, whereas at very low carbon dioxide contents the respective neutral hydrolysis product, $\text{UO}_2(\text{OH})_2(\text{aq})$, is the main species.

An EQ6 reaction path run was then used to model the thermodynamic equilibrium state of the system expected to be reached after the long time scale covered by the risk assessment. The species $\text{N}_2(\text{aq})$, N_3^- , $\text{HN}_3(\text{aq})$, and $\text{UO}_2(\text{N}_3)_x$ had to be excluded in order to avoid that the redox potential shifts to very anoxic values around -200 mV. At equilibrium, the minerals calcite, diaspore, fluorapatite, quartz and thorianite should eventually precipitate. This will remove phosphate and thorium nearly quantitatively from the solution, also 97% of the aluminium and 39% of the silica. However, the assumed presence of calcite instead of dolomite reduced the measured content of calcium only by 0.9%. For iron, the assumed stable phase of $\text{Fe}(\text{OH})_3$ was not saturated. The modelled precipitations decreased the pH to 7.21, and increase the redox potential Eh to 356 mV, which in both cases was still inside the

uncertainty. The corresponding uranium speciation is given in Table 6. It is clearly dominated by the neutral complex species $\text{Ca}_2\text{UO}_2(\text{CO}_3)_3(\text{aq})$.

Table 6 : Uranium speciation computed with EQ6 at thermodynamic equilibrium (Tailing layer of Ranstad Tailing Site)

Species	Molality	Mol % of Total U
$\text{Ca}_2\text{UO}_2(\text{CO}_3)_3(\text{aq})$	8.9825E-07	89.5
$\text{UO}_2(\text{CO}_3)_3^{4-}$	6.5215E-08	6.5
$\text{UO}_2(\text{CO}_3)_2^{2-}$	3.9075E-08	3.9
$\text{UO}_2(\text{OH})_2(\text{aq})$	9.9927E-10	0.1

6.2.5.4 Selection of Final Data Set

For the tailing layer it can be summarized, that modellings with EQ3NR and EQ6 demonstrated that the contents of silicon, aluminium, iron, manganese and thorium measured in the solution are too high with respect to the saturation limits of many minerals. Probably the analysis also included colloidal or microcrystalline material. The saturation indices indicated, that:

- total silicon content may be determined by equilibrium with quartz;
- total carbonate content may be determined by equilibrium with calcite;
- total aluminium content may be determined by equilibrium with an aluminosilicate, e.g. muscovite.

Despite missing direct experimental values for the redox state, an Eh value of +346 mV could be derived from modelling based on the redox couples $\text{NO}_3^- / \text{NO}_2^- / \text{NH}_4^+$.

For the selection of a set of best values, average values (MEAN) and standard deviations (STD) were calculated assigning equal weights to values from the FZR analysis and from bore hole 111L (which most closely resembles the situation inside the tailings layer), with the exception of iron, aluminium, lead, and thorium, where only the FZR analysis was used.

A critical review of the selected data shows, that for most of the major anions and cations the analytical values have reasonable standard deviations (if available). Remarkable exceptions are the content of zinc, manganese and aluminium.

6.3 Multi-attribute utility analysis

The methodology of multi-attribute utility analysis for ranking remediation strategies for a contaminated site, as described in Section 5.6.4, is illustrated here with a hypothetical case. The attributes include monetary costs of the remedial measures, radiological doses to population and clean-up workers, and the social attributes reassurance and disturbance. Linear utility functions, so-called risk-neutral utility functions, have been used and uncertainties included in terms of value distributions of the attributes. The weighting factors assigned to the different attributes have been determined by use of scaling factors in terms of weighting factor ratios, and their values were sampled around a most probable value.

6.3.1. Assessment of attribute values

Attributes for a hypothetical contaminated site have been given the values shown in Table 7. Disturbance and reassurance is assumed to be related to the volume of waste produced by the remedial measures and to the activity left at the site after the implementation of the remedial measures, respectively. Other ways of expressing these attributes can be postulated.

Table 7 : Attribute values for different restoration strategies at a hypothetical site.

Restoration strategy	Collective dose to population [man Sv]	Collective dose to workers [man-Sv]	Monetary costs of restoration [kEUR]			Fraction of activity left on-site	Waste volume (m ³)
			Remediation	Monitoring	Waste disposal		
A	2,000	0	0	50,000	0	1	0
B	1,600	4	380,000	40,000	30,000	0.8	10,000
C	1,200	8	300,000	30,000	100,000	0.6	30,000
D	400	6	600,000	10,000	150,000	0.2	40,000
E	40	10	900,000	50,000	0	1	0

Different software systems can be used for multi-attribute analyses. In the RESTRAT study the Crystal Ball/Excel system has been used. Rather complicated models can be developed in Excel and it can be used as an integral part of the software system Crystal Ball from the company *Decisioneering* (1996). In Crystal Ball uncertainties can be assigned to model parameters and correlations made between them. The software of Crystal Ball, as it has been applied to the examples, is present in the accompanying CD-ROM (Hedemann Jensen, 1999).

To include uncertainties in the analysis, each of the attributes has been assigned a probability distribution (illustrated here by normal distributions). A negative correlation has been assumed between remediation costs and population doses (correlation coefficient of -0.8). A normal distribution functions has been used for the seven attributes for each of the remediation options A - E, and the parameters used are shown in Table 8.

Table 8 : Parameters used in the normal distributions for the seven attributes and five remediation options.

Remediation option	5 - 95%tiles in the normal distributions						
	Population dose [kman·Sv]	Worker dose [manSv]	Remediation costs [MEUR]	Monitoring costs [kEUR]	Waste costs [kEUR]	Activity fraction left	Waste volume [1000 m ³]
A	1.7 - 2.3	0.83 - 1.2·10 ⁻⁵	0.83 - 1.2·10 ⁻⁶	42 - 58	0.83 - 1.2·10 ⁻³	0.79 - 0.94	0.83 - 1.2·10 ⁻⁶
B	1.3 - 1.9	3.3 - 4.7	0.32 - 0.44	33 - 47	25 - 35	0.67 - 0.93	8.4 - 12
C	1.0 - 1.4	6.7 - 9.3	0.25 - 0.35	25 - 35	84 - 120	0.50 - 0.70	25 - 35
D	0.33 - 0.47	3.3 - 4.7	0.50 - 0.70	8.4 - 12	130 - 180	0.17 - 0.23	33 - 47
E	0.033 - 0.047	8.4 - 11.6	0.75 - 1.00	42 - 58	0.83 - 1.2·10 ⁻⁵	0.79 - 0.94	0.83 - 1.2·10 ⁻⁶

6.3.2. Assessment of utility functions

Risk neutral utility functions have been used here to illustrate the utility functions for the different attributes as shown in Table 9.

Table 9 : Utility functions for the different attributes for restoration strategies at a hypothetical site.

Attributes	Utility function
Population dose	$u_{pop}(x) = 100 \cdot \left[1 + \frac{40 - x}{2,000 - 40} \right]$ for $40 \leq x \leq 2,000$ man·Sv
Worker dose	$u_{work}(x) = 100 \cdot \left[1 - \frac{x}{10} \right]$ for $0 \leq x \leq 10$ man·Sv
Remediation costs	$u_{remedia}(x) = 100 \cdot \left[1 - \frac{x}{900,000} \right]$ for $0 \leq x \leq 900,000$ kEUR
Monitoring costs	$u_{monitor}(x) = 100 \cdot \left[1 + \frac{10,000 - x}{50,000 - 10,000} \right]$ for $10,000 \leq x \leq 50,000$ kEUR
Waste costs	$u_{waste}(x) = 100 \cdot \left[1 - \frac{x}{150,000} \right]$ for $0 \leq x \leq 150,000$ kEUR
Reassurance	$u_{reas}(x) = 100 \cdot \left[\frac{1}{2} \cdot \left(\frac{40 - x}{2,000 - 40} \right) + \frac{1}{2} \cdot \left(\frac{0.2 - y}{1 - 0.2} \right) \right]$ for $40 \leq x \leq 2,000$ man·Sv and $0.2 \leq y \leq 1$
Disturbance	$u_{distur}(x) = 100 \cdot \left[1 - \frac{x}{40,000} \right]$ for $0 \leq x \leq 40,000$ m ³

The utility functions, $u(x)$, have been programmed in the Excel spreadsheet and distributions have been assigned to the attributes, x , according to the procedure given in Hedemann Jensen (1999).

6.3.3. Assessment of weighting factors

As discussed in Section 5.6.4, two different methods have been used in this study to determine the weighting factors, w . For attributes at the same hierarchy level given in the same unit, e.g. monetary costs, the weighting between the different attributes have been related to their value ranges, R , by the relation $(w/R)_1 = (w/R)_2 = (w/R)_3 = \dots = C$. The weighting of attributes at the same hierarchy level for which the units are different, as they are for the social attributes, has been determined by assigning a value to the ratio of their weighting factors as $w_2/w_1 = C_1$, $w_3/w_1 = C_2$, $w_n/w_1 = C_{n-1}$.

The values of the weighting factors for the hypothetical site as determined by these two methods are shown in Table 10.

Table 10 : Weighting factors for major attributes and health, economic and social attributes.

Major attributes		
Health weights	Economic weights	Social weights
$C_1 = \frac{w_{economic}}{w_{health}} = \frac{R_{economic}}{\alpha \cdot (R_{dose,pop} + R_{dose,work})} = \frac{(900,000 + 50,000 + 0) - (0 + 50,000 + 0)}{100,000 \cdot ((2,000 - 40) + (10 - 0))} = 4.57$ $C_2 = \frac{w_{social}}{w_{health}} \approx 0.2 - 0.3$		
$w_{health} = \frac{1}{1 + C_1 + C_2}$ $= \frac{1}{1 + 4.57 + 0.25} = 0.172$	$w_{economic} = \frac{C_1}{1 + C_1 + C_2}$ $= \frac{4.57}{1 + 4.57 + 0.25} = 0.785$	$w_{social} = \frac{C_2}{1 + C_1 + C_2}$ $= \frac{0.25}{1 + 4.57 + 0.25} = 0.043$
Sub-attributes		
Collective doses	Monetary costs	Social factors
$C = \frac{1}{R_{dose,pop} + R_{dose,work}}$ $= \frac{1}{(2,000 - 40) + (10 - 0)}$ $= 5 \cdot 10^{-4}$	$C = \frac{1}{R_{remedia} + R_{monitoring} + R_{waste}}$ $= \frac{1}{(900,000 - 0) + (50,000 - 10,000) + (150,000 - 0)}$ $= 9.2 \cdot 10^{-7}$	$C = \frac{w_{reas}}{w_{distur}}$ $\approx 5 - 7$
$w_{dose,pop} = C \cdot R_{dose,pop}$ $= 5 \cdot 10^{-4} \cdot (2,000 - 40)$ $= 0.995$ $w_{dose,work} = C \cdot R_{dose,work}$ $= 5 \cdot 10^{-4} \cdot (10 - 0)$ $= 0.005$	$w_{remedia} = C \cdot R_{remedia}$ $= 9.2 \cdot 10^{-7} \cdot (900,000 - 0)$ $= 0.828$ $w_{monitor} = C \cdot R_{monitor}$ $= 9.2 \cdot 10^{-7} \cdot (50,000 - 10,000)$ $= 0.037$ $w_{waste} = C \cdot R_{waste}$ $= 9.2 \cdot 10^{-7} \cdot (150,000 - 0)$ $= 0.135$	$w_{reas} = \frac{1}{1 + C}$ $= \frac{1}{1 + 6}$ $= 0.14$ $w_{distur} = \frac{C}{1 + C}$ $= \frac{6}{1 + 6}$ $= 0.86$

The weighting factors shown in this table have been built-in the Excel spreadsheet. To include uncertainties in the analysis, each of the weighting factors have been assigned a triangular probability distribution as shown in Figure 10, Figure 11, Figure 12 and Figure 13. The probability is maximum at the central value and 0 at the beginning and end of the interval. The distributions have been assigned according to the procedure given in Hedemann Jensen (1999).

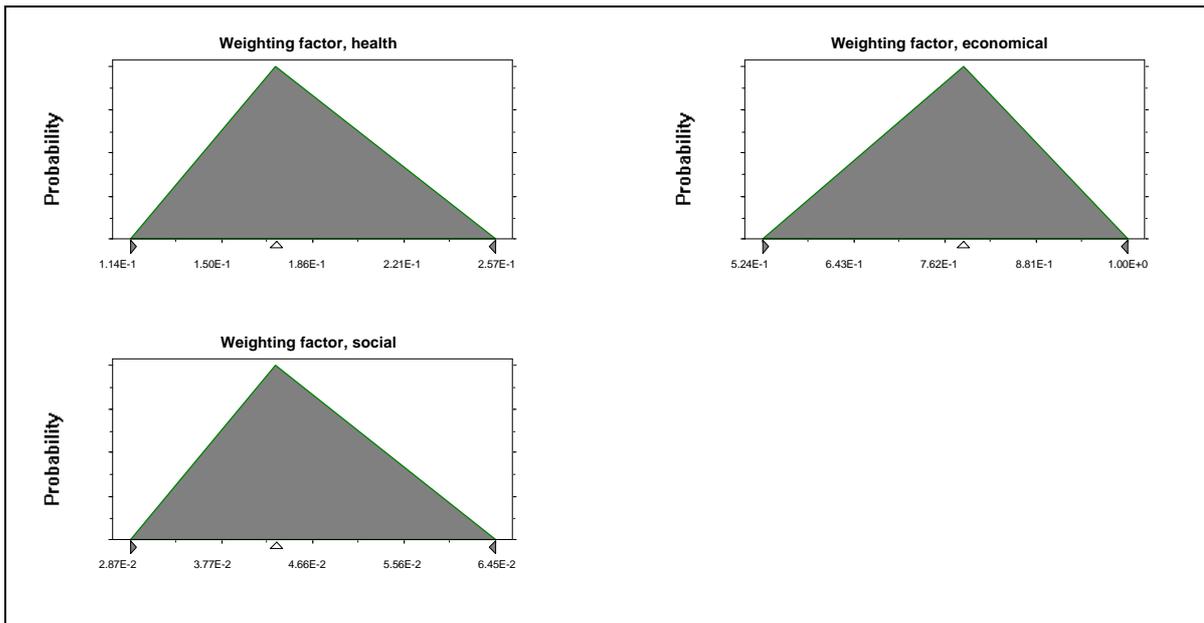


Figure 10 : Distribution functions for weighting factors for major attributes.

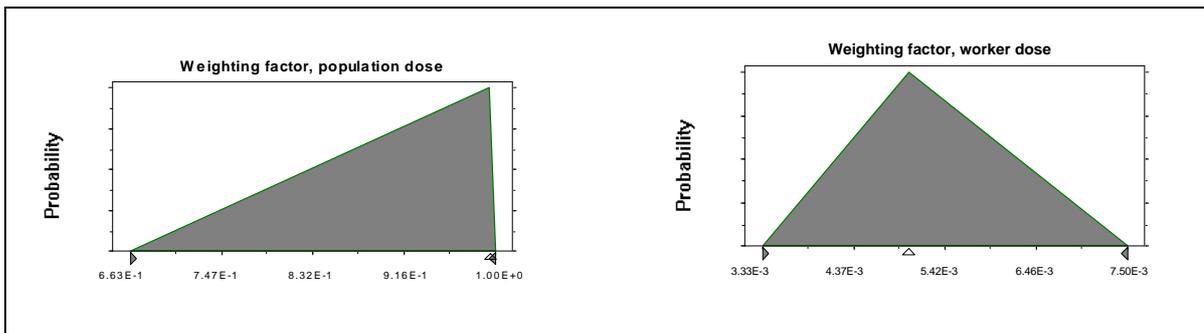


Figure 11 : Distribution functions for weighting factors for health sub-attributes.

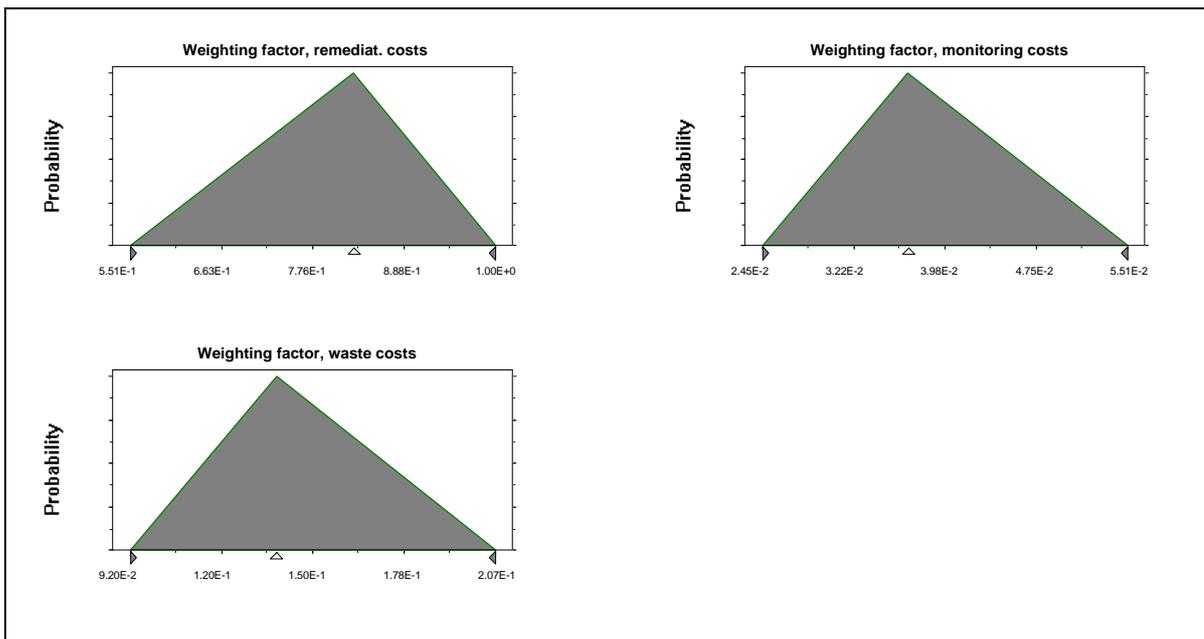


Figure 12 : Distribution functions for weighting factors for economic sub-attributes.

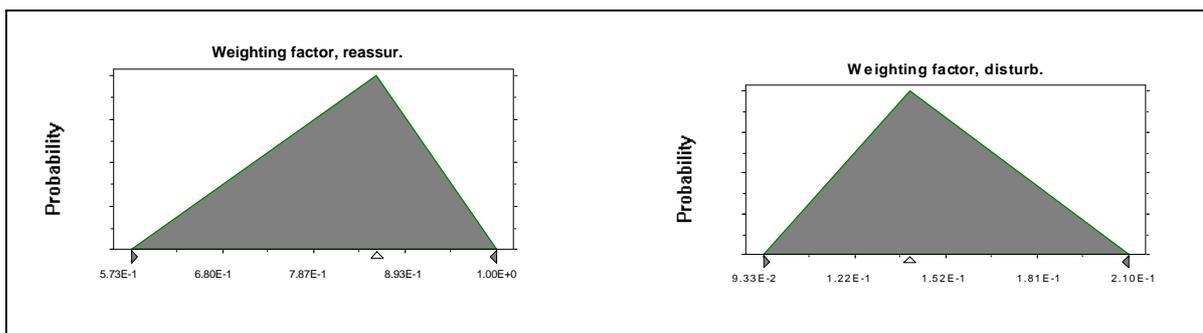


Figure 13 : Distribution functions for weighting factors for social sub-attributes.

6.3.4. Assessment of scores for remedial measures

As discussed in Section 5.6.4 the best (optimised) strategy or option amongst a set of strategies or options, i , can be expressed by the overall score, $U_i(x)$, which has its maximum value at the optimum:

$$U_i(x) = \sum_{j=1}^n w_j \cdot u_{ij}(x) \quad \text{and} \quad U_{opt} = \max \langle U_i(x) \rangle$$

The overall scores, $U_i(x)$, for the remedial options A - E for the hypothetical site have been programmed into the spreadsheet in the cells B30 - B34 as shown in Figure 14 according to the procedure given in Hedemann Jensen (1999).

Latin Hypercube sampling was used to simulate the scores for the options A - E. The values of the attributes, x , were sampled within the probability distributions given in Table 8 resulting in corresponding values of the utility functions, $u(x)$. The weighting factors were sampled within the probability distributions given in Figure 10, Figure 11, Figure 12 and Figure 13. The chart in Figure 14 shows the scores after 12,000 simulations. The probability distributions of the forecasts of the scores $U_A(x) - U_E(x)$ are shown in Figure 15 after 12,000 simulations. The lower right picture in Figure 15 shows the median values of the scores, $U(x)$ based upon the median values of the utilities and weighting factors. This picture also shows the contribution to the median value of total score from the health, economical and social attributes.

	A	B	C	D	E	F	G	H	I	J	K
1	SITE DATA ON DOSES, COSTS AND WASTE VOLUMES										
2	Health attributes		Economic attributes			Social attributes					
3	Collective doses		Monetary costs of restoration			Activity	Waste				
4		Population	Workers	Remediat.	Monitoring	Waste	fraction	Waste			
5	OPTION	[manSv]			[kECU]		left on-site	[m ³]			
6	Option A	2.00E+03	0.00E+00	0.00E+00	5.00E+04	0.00E+00	1.00E+00	0.00E+00			
7	Option B	1.60E+03	4.00E+00	3.80E+05	4.00E+04	3.00E+04	8.00E-01	1.00E+04			
8	Option C	1.20E+03	8.00E+00	3.00E+05	3.00E+04	1.00E+05	6.00E-01	3.00E+04			
9	Option D	4.00E+02	6.00E+00	6.00E+05	1.00E+04	1.50E+05	2.00E-01	4.00E+04			
10	Option E	4.00E+01	1.00E+01	9.00E+05	5.00E+04	0.00E+00	1.00E+00	0.00E+00			
11	A	B	C	D	E	F	G	H	I	J	K
12	UTILITY FUNCTIONS U(x)										
13		Collective doses		Monetary costs of restoration			Social factors				
14	OPTION	Population	Workers	Remediat.	Monitoring	Waste	Reassur.	Disturb.			
15	Option A	0.00E+00	1.00E+02	1.00E+02	0.00E+00	1.00E+02	0.00E+00	1.00E+02			
16	Option B	2.04E+01	6.00E+01	5.78E+01	2.50E+01	8.00E+01	2.02E+01	7.50E+01			
17	Option C	4.08E+01	2.00E+01	6.67E+01	5.00E+01	3.33E+01	4.37E+01	2.50E+01			
18	Option D	8.16E+01	4.00E+01	3.33E+01	1.00E+02	0.00E+00	9.08E+01	0.00E+00			
19	Option E	1.00E+02	0.00E+00	0.00E+00	0.00E+00	1.00E+02	5.00E+01	1.00E+02			
20	A	B	C	D	E	F	G	H	I	J	K
21	WEIGHTING FACTORS										
22						Health	Economic	Social	Sum		
23						1.72E-01	7.85E-01	4.30E-02	1.00E+00		
24					Normalised	1.72E-01	7.85E-01	4.30E-02	1.00E+00		
25		Population	Workers	Sum	Remediat.	Monitoring	Waste	Sum	Reassur.	Disturb.	Sum
26		9.95E-01	5.00E-03	1.00E+00	8.28E-01	3.70E-02	1.35E-01	1.00E+00	8.60E-01	1.40E-01	1.00E+00
27	Normalised	9.95E-01	5.00E-03	1.00E+00	8.28E-01	3.70E-02	1.35E-01	1.00E+00	8.60E-01	1.40E-01	1.00E+00
28	A	B	C	D	E	F	G	H	I	J	K
29	OPTION	Scores									
30	Option A	7.63E+01									
31	Option B	5.15E+01									
32	Option C	5.71E+01									
33	Option D	4.19E+01									
34	Option E	3.02E+01									
35											
36											
37											
38											
39											
40											

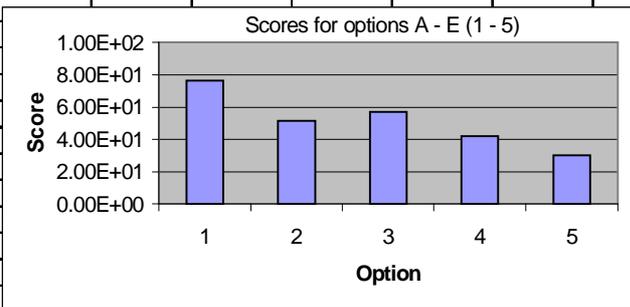


Figure 14 : The Excel spreadsheet with the complete multiattribute analysis model.

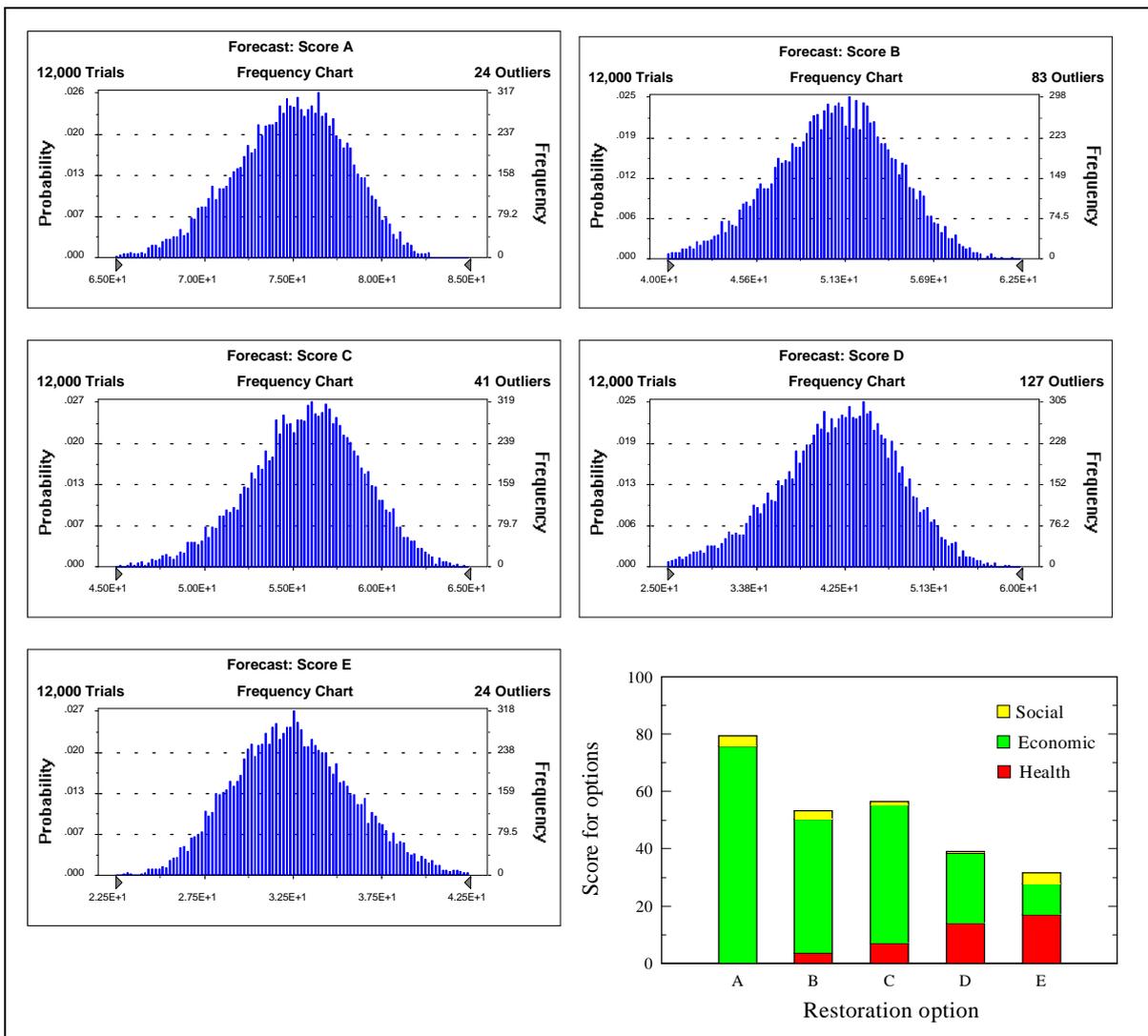


Figure 15 : Distribution of the scores A - E after 12,000 simulations.

It appears from Figure 14 that the remediation option A has the highest score. This option would therefore be the optimum amongst the five options A - E.

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7. Results for example sites

7.1 Ranking of restoration options for example sites

Multi-attribute utility analyses and cost-benefit analyses have been used to arrive at an optimum remediation strategy among a number of different strategies for five contaminated sites in Europe.

The remediation techniques considered include:

A	No remediation		
B	Removal of sources	Soil excavation	
C1	Physical Separation	Soil washing	
C2		Filtration	
D1	Chemical Separation	Chemical solubilisation	
D2		Ion exchange	
D3	Biological Separation	Biosorption	
E1, E2	Containment	Capping	
E3, E4		Subsurface barriers	
F1	Physical Immobilisation	Cement-based solidification	<i>Ex-situ</i>
F2			<i>In-situ</i>
G1	Chemical Immobilisation	Chemical Immobilisation	<i>Ex-situ</i>
G2			<i>In-situ</i>

The characteristics of these techniques are given in chapter 4.

The attributes considered in the multi-attribute utility analysis, include:

- *Health attributes*
 - collective doses to population
 - doses to remediation workers
 - exposure to heavy metals or toxic chemicals
- *Economic attributes*
 - costs of remedial actions (incl. costs of labour and monitoring)
 - costs of monitoring of remedial options
 - costs of disposal of generated waste (in broad categories)
 - loss/gain of taxes due to loss/gain of income
- *Social attributes*
 - reassurance of the public
 - discomfort, disturbance and anxiety from the remedial action

- loss/gain of income

In order to derive utility values for the attributes, linear utility functions, so-called risk-neutral utility functions, have been used and uncertainties included in terms of value distributions of the attributes. The weighting factors assigned to the different attributes have been determined by use of scaling factors in terms of weighting factor ratios, and their values were sampled around a most probable value.

The ranking results are presented below. More details are given in TD 8 (Hedemann Jensen, 1999).

7.1.1. Molse Nete River site

Since 1956, controlled releases of low-level radioactive effluents have been made from nuclear facilities in the region of Mol in the north-eastern part of Belgium. The Molse Nete River has been contaminated with the radionuclides ⁶⁰Co, ¹³⁷Cs, ²³⁹Pu, and ²⁴¹Am as a result of these discharges into the river. The riverbanks have been contaminated through dredging of bed sediment out of the river. Subsequently, also agricultural soils have been contaminated through the application of the dredged sediment onto agricultural land for the purpose of soil amendment.

The compartment scheme for the Molse Nete River site, that has been used for the radiological impact assessment, is shown in Figure 16.

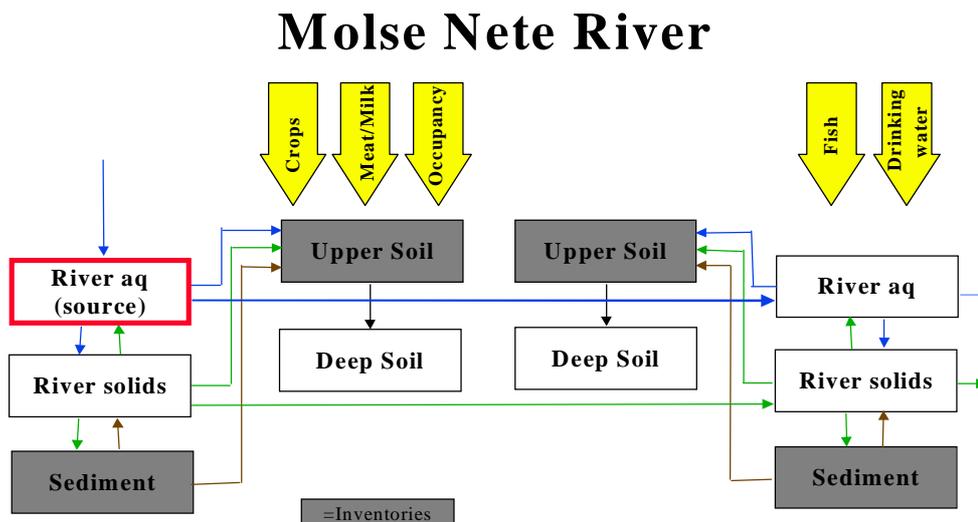


Figure 16 : Compartment scheme for the Molse Nete River site.

The restoration options included for the Molse Nete River site have been identified in TD 11 (Sweeck and Zeevaert, 1999). The economic and radiological data for quantifying the various attributes for each of those options are shown in **Error! Reference source not found.** The corresponding utility values of the attributes are indicated in Table 12. The weighting factors of the attributes calculated by assigning values to their ratios, are given in Table 13.

Table 11: Economic and radiological data (for integration times of 100 and 500 years) for different restoration strategies at the Molse Nete River site. Best-estimate values.

Restoration strategy	Collective dose to population [man Sv]		Collective dose to workers [man·Sv]	Monetary costs of restoration [kECU]				Fraction of activity left on-site	Waste volume (m ³)
	100 y	500 y		Remediation	Monitoring	Waste disposal	Inc. loss Tax loss		
	A	16	51					0	0
B	1.6	5.1	6.1·10 ⁻⁴	3,570	1,000	19,580	0 0	0.1	26,520
C1	4.5	14	1.8·10 ⁻³	12,870	2,000	13,260	0 0	0.3	5,300
D1	1.6	5.1	1.6·10 ⁻³	13,970	2,000	13,260	0 0	0.1	10,600
E1	negli.	negli.	2.6·10 ⁻³	4,250	3,200	0	68 1,360	1	0
F1	negli.	negli.	6.7·10 ⁻³	6,220	3,200	0	68 1,360	1	0
F2	negli.	negli.	1.8·10 ⁻³	5,810	3,200	0	68 1,360	1	0
G1	negli.	negli.	6.7·10 ⁻³	8,340	3,200	0	68 1,360	1	0
G2	negli.	negli.	1.8·10 ⁻³	5,810	3,200	0	68 1,360	1	0

Table 12 : Utility values for best-estimate values of the attributes for each of the remediation options at the Molse Nete River site.

Option	Utilities										
	Health			Economic				Social			
	Population		Worker	Waste	Remedi.	Tax loss	Monito.	Reassurance		Inc. loss	Disturb.
	100 y	500 y						100 y	500 y		
A	0	0	100	100	100	100	0	0	0	100	100
B	90.0	90.0	90.9	0	74.4	100	68.8	95.0	95.0	100	0
C1	71.9	72.5	73.1	32.3	7.87	100	37.5	74.8	75.2	100	80.0
D1	90.0	90.0	76.1	32.3	0	100	37.5	95.0	95.0	100	60.0
E1	100	100	61.2	100	69.6	0	0	50.0	50.0	0	100
F1	100	100	0	100	55.5	0	0	50.0	50.0	0	100
F2	100	100	73.1	100	58.4	0	0	50.0	50.0	0	100
G1	100	100	0	100	40.3	0	0	50.0	50.0	0	100
G2	100	100	73.1	100	58.4	0	0	50.0	50.0	0	100

Table 13: Weighting factors for attributes and sub-attributes applied in the optimisation of remediation of the Molse Nete site. Best-estimate values.

Health factors			Economical factors		Social factors	
100 years	500 years		100 years	500 years	100 years	500 years
0.057	0.157		0.929	0.803	0.014	0.039
	100 years	500 years	Remediation costs	0.37	Reassurance	0.63
Dose population	1	1				
Dose workers	0	0	Waste disposal costs	0.51	Disturbance	0.11
Non-radiation	-	-	Monitoring costs	0.084	Loss/gain of income	0.26
			Loss/gain of taxes	0.036		

The values of the attributes, from which the utility values are determined and the weighting factors indicated above have all been sampled in a triangular distribution between 1.5^{-1} - 1.5 times the most probable value given in **Error! Reference source not found.** and Table 13. Negative correlation between collective doses and remediation costs has been applied with a correlation coefficient of -0.8 .

The overall scores of the remediation options have been determined from the weighted sum of utilities as shown Figure 17. The left-hand picture shows the results for an integration time of 100 years for the collective dose, the right-hand picture shows them for an integration time of 500 years. The error bars represent the 5th and 95th percentiles of the distributions of the scores.

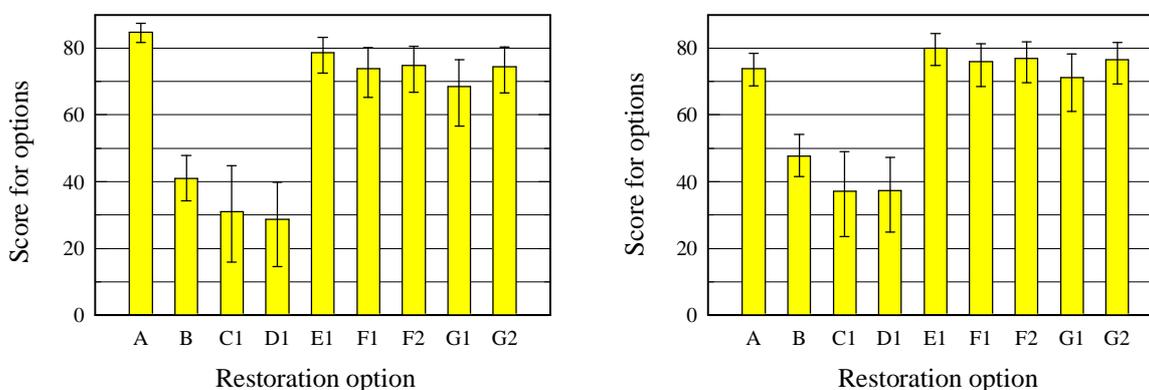


Figure 17 : Overall scores for different remediation strategies for the Molse Nete River site.

In Figure 17 there is practically no difference between the scores for an integration time of 100 and 500 years due to the low weight of the health attributes although the score for option A is somewhat lower for the longer integration time. The options A, E1, F1, F2, G1 and G2 have practically an equal score which makes it rather difficult to distinguish which is the optimum. For a 500-years integration time the option E1, capping, has the highest score, and this option can therefore be considered as the optimum.

7.1.2. Drigg site

The Drigg site is situated in West Cumbria about nine km south of Sellafield in the UK on the coast of the Irish Sea. The site is placed just west of the village of Drigg, 300 m north of the tidal estuary of the River Esk. Since 1959 the site has been used for the disposal of low-level radioactive waste. It is operated by British Nuclear Fuel plc (BNFL) for the shallow burial of solid waste, mostly from the Sellafield site. Several small streams cross the site. The dominating radionuclides giving rise to the low doses to the local population, mainly from milk consumption, are ^{60}Co , ^{137}Cs , ^{239}Pu , and ^{241}Am .

The source is the low-level radioactive waste in solid form deposited at the Drigg site. After interaction between the solid phase and the infiltration water the activity is transported as leakage via drains to Drigg stream. Irrigation was assumed of agricultural soil of water from the stream and dredging of the sediments is occurring on a yearly basis.

The compartment scheme for the Drigg site, that has been used for the assessment of the radiological impact, is shown in Figure 18.

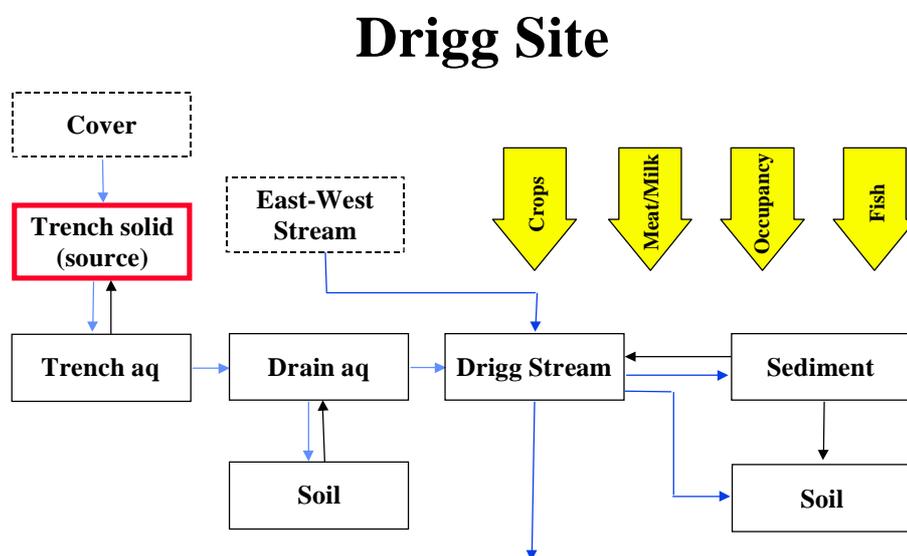


Figure 18 : Compartment scheme for the Drigg site.

The restoration options included for the Drigg site have been identified in TD 9 (Bousher, 1999a). The economic and radiological data for quantifying the various attributes for each of those options are shown in Table 14. The corresponding utility values of the attributes are indicated in Table 15. The weighting factors of the attributes calculated by assigning values to their ratios, are given in Table 16.

Table 14 : Economic and radiological data (for integration times of 100 and 500 years) for different restoration strategies at the Drigg site. Best-estimate values.

Restoration strategy	Collective dose to population [man Sv]		Collective dose to workers [man·Sv]	Monetary costs of restoration [kECU]			Fraction of activity left on-site	Waste volume (m ³)
	100 y	500 y		Remediation	Monitoring	Waste disposal		
A	49	120	0	0	75,000	0	1	0
C2	0.93	3.3	1.5·10 ⁻⁹	380,000	750	31,000	0.01	12,500
D1	9.9	33	1.7·10 ⁻⁹	300,000	7,500	100,000	0.1	41,000
D2	16	51	3.7·10 ⁻¹⁰	1,000,000	15,000	31,000	0.2	12,500
D3	13	42	7.1·10 ⁻¹⁰	1,300,000	7,500	31,000	0.1	12,500
E1	0.43	1.9	5.5·10 ⁻¹²	3,500	75,000	0	1	0
E3	2.9	11	6.9·10 ⁻¹⁰	6,300	75,000	0	1	0
F1	4.2	10	2.8·10 ⁻⁹	55,000	75,000	0	1	0
F2	4.2	10	1.4·10 ⁻⁹	190,000	75,000	0	1	0
G1	2.9	7.2	1.9·10 ⁻⁹	130,000	75,000	0	1	0
G2	2.9	7.2	9.4·10 ⁻¹⁰	55,000	75,000	0	1	0

Table 15 : Utility values for best-estimate values of the attributes for each of the remediation options at the Drigg site.

Option	Utilities								
	Health			Economic			Social		
	Population		Workers	Waste	Remediat	Monitorin g	Reassuran.		Disturban.
	100 y	500 y					100 y	500 y	
A	0	0	100	100	100	0	0	0	100
C2	99.0	98.8	46.4	69.0	70.8	99.0	99.5	99.4	69.5
D1	80.5	73.7	39.3	0	76.9	90.0	85.7	82.3	0
D2	67.9	58.4	86.8	69.0	23.1	80.0	74.4	69.6	69.5
D3	74.1	66.0	74.6	69.0	0	90.0	82.5	78.5	69.5
E1	100	100	99.8	100	99.7	0	50.0	50.0	100
E3	94.9	92.3	75.4	100	99.5	0	47.5	46.1	100
F1	92.2	93.1	0	100	95.8	0	46.1	46.6	100
F2	92.2	93.1	50.0	100	85.4	0	46.1	46.6	100
G1	94.9	95.5	32.1	100	90.0	0	47.5	47.8	100
G2	94.9	95.5	66.4	100	95.8	0	47.5	47.8	100

Table 16 : Weighting factors for attributes and sub-attributes applied in the optimisation of remediation of the Drigg site. Best-estimate values.

Health factors			Economical factors		Social factors	
100 years	500 years		100 years	500 years	100 years	500 years
0.0034	0.0083		0.996	0.990	0.00086	0.0021
	100 years	500 years	Remediation costs	0.882	Reassurance	0.86
Dose population	1	1				
Dose workers	0	0	Waste disposal costs	0.068	Disturbance	0.14
Non-radiation	-	-	Monitoring costs	0.050	Loss/gain of income	-
			Loss/gain of taxes	-		

The values of the attributes, from which the utility values are determined and the weighting factors indicated above have all been sampled in a triangular distribution between 1.5^{-1} - 1.5 times the most probable value given in Table 14 and Table 16. Negative correlation between collective doses and remediation costs has been applied with a correlation coefficient of -0.8 .

The overall scores of the remediation options have been determined from the weighted sum of utilities as shown in Figure 19.

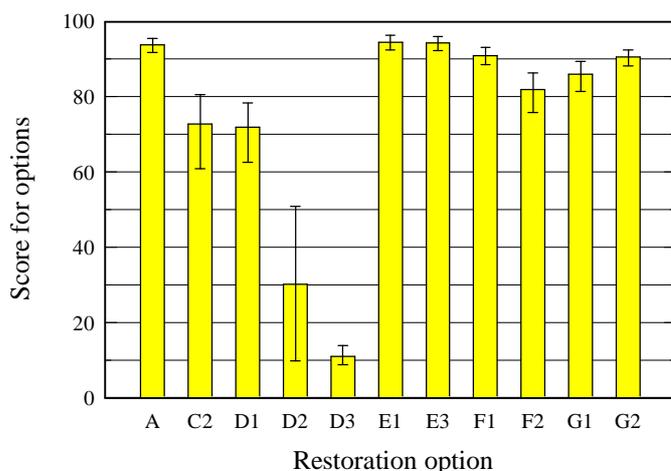


Figure 19 : Overall scores for different remediation strategies for the Drigg site.

Figure 19 shows the results for an integration time of 100 and 500 years for the collective dose (identical values). The error bars represent the 5th and 95th percentiles of the distributions of the scores.

It appears from this figure that option E1 (capping) and E3 (sub-surface barrier) have the highest score, closely followed by the option A (no remediation). Also option F1 (physical immobilisation, *ex-situ*) and G2 (chemical immobilisation, *in-situ*) have a high and practically equal score. Therefore, it might be difficult to pick an optimum solution among the options E1, E3, A, F1 and G2.

7.1.3. Ravenglass site

The Ravenglass estuary is situated in West Cumbria on the coast of the Irish Sea. It encompasses the tidal reaches of the River Esk, Irt and Mite and its northern part directly borders on the Drigg site. The principal source of the estuary is the Irish Sea as the rivers contribute only a smaller part. The sediments are contaminated via the Irish Sea from waste discharges from the Sellafield nuclear fuel reprocessing plant. The main radionuclides of the contamination are ¹³⁷Cs, ²³⁹Pu, and ²⁴¹Am.

This environment presents some problems considering the use of remediation techniques, as it is both tidal, dynamic and can be turbulent. The area is within the Lake District National Park and the public has therefore access to the area. As a consequence of the area characteristics *ex-situ* techniques will provide the best options for remediation of the site.

The compartment scheme for the Ravenglass site, that has been used for the assessment of the radiological impact, is shown in Figure 20.

Ravenglass

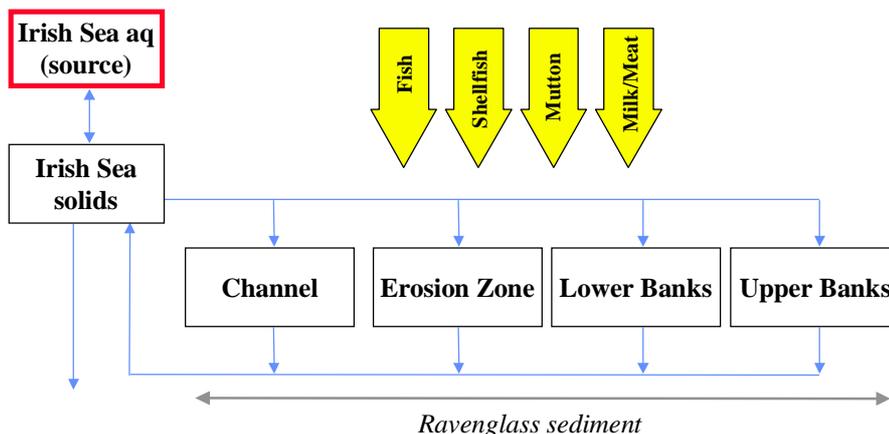


Figure 20 : Compartment scheme for the Ravenglass Estuary.

The restoration options included for the Ravenglass site have been identified in TD 12 (Bousher, 1999b). The economic and radiological data for quantifying the various attributes for each of those options are shown in Table 17. The corresponding utility values of the attributes are indicated in Table 18. The weighting factors of the attributes calculated by assigning values to their ratios, are given in Table 19.

Table 17 : Economic and radiological data (for integration times of 100 and 500 years) for different restoration strategies at the Ravenglass site. Best-estimate values.

Restoration strategy	Collective dose to population [man Sv]		Collective dose to workers [man·Sv]	Monetary costs of restoration [kECU]			Fraction of activity left on-site	Waste volume (m ³)
	100 y	500 y		Remediation	Monitoring	Waste disposal		
A	28	29	0	0	52,500	0	1	0
B	15	15	0.92	130,000	3,000	780,000	0.05	1.3·10 ⁶
C1	23	24	1.01	520,000	10,500	650,000	0.2	2.6·10 ⁵
D1	7.7	8.2	2.29	720,000	10,500	1.1·10 ⁶	0.2	4.3·10 ⁵

Table 18 : Utility values for best-estimate values of the attributes for each of the remediation options at the Ravenglass site.

Option	Utilities						
	Health		Economic			Social	
	Population	Workers	Waste	Remediat.	Monitorin g	Reassuran.	Disturban.
A	0	100	100	100	0	0	100
B	67.3	59.8	29.1	81.9	100	83.7	0
C1	24.0	55.9	40.9	27.8	84.8	54.1	80.0
D1	100	0	0	0	84.8	92.1	66.9

Table 19 : Weighting factors for attributes and sub-attributes applied in the optimisation of remediation of the Ravenglass site. Best-estimate values.

Health factors			Economical factors		Social factors		
100 years	500 years		100 years	500 years	100 years	500 years	
1.08·10 ⁻³	1.11·10 ⁻³		0.999	0.999	2.71·10 ⁻⁴	2.78·10 ⁻⁴	
	100 years	500 years	Remediation costs		0.385	Reassurance	0.86
Dose population	0.90	0.90	Waste disposal costs		0.588	Disturbance	0.14
Dose workers	0.10	0.10	Monitoring costs		0.026	Loss/gain of income	-
Non-radiation	-	-	Loss/gain of taxes		-		

The values of the attributes, from which the utility values are determined and the weighting factors indicated above have all been sampled in a triangular distribution between 1.5⁻¹ - 1.5 times the most probable value given in Table 17 and Table 19. Negative correlation between collective doses and remediation costs has been applied with a correlation coefficient of -0.8.

The overall scores of the remediation options have been determined from the weighted sum of utilities as shown in Figure 21.

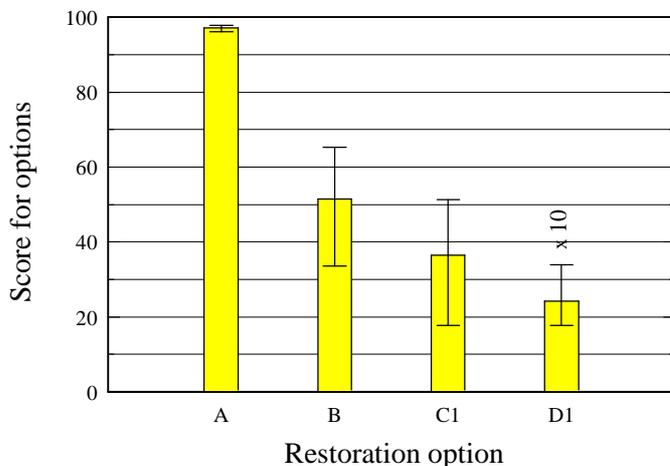


Figure 21 : Overall scores for different remediation strategies for the Ravenglass site ("x 10" means that the indicated value is 10 times higher than the actual one).

Figure 21 shows the results for an integration time of 100 and 500 years for the collective dose (identical values). The error bars represent the 5th and 95th percentiles of the distributions of the scores.

It appears from this figure that option A has the highest score. The scores for options B and C1 are both significantly lower score than for option A. Due to the highest total costs for option D1 this option has the lowest score. The 'no remediation' option A is thus the optimum solution for the Ravenglass site and also the cheapest. There is no difference between the scores for the two different integration times due to the low weight of the health attributes.

7.1.4. Ranstad tailing site

The Ranstad Tailing site is situated in the southern part of Sweden, in the Billingen-Häggum district about 20 km south of the city of Skövde. The tailings have been produced from a former uranium processing plant of the Swedish AB Atomenergi, which operated the uranium from a nearby open pit mine. The mill tailing consists of crushed alum shale from which uranium has been extracted by leaching. The contaminants are mainly ²³⁸U in addition to significant levels of manganese and nickel.

In order to remediate a mill tailing different restoration techniques can be considered. In the case of the Ranstad mill tailing site three different categories of remediation techniques has been looked upon; containment, immobilisation and separation. Containment is a good alternative in order to reduce the amount of infiltrating water and the entrance of oxygen into the tailing. It is the percolating water together with oxygen that governs the weathering processes in the tailing. If the weathering processes stops then the amount of contaminants leaching from the tailing will be strongly reduced.

For the Ranstad tailing site two different types of capping have been considered. The first one consisting of 0.5 m of moraine, as it was on the tailing before the remediation started, and another one consisting of 1.6 m of different soil types, as was actually performed 1991-92.

The compartment scheme for the Ranstad tailing site, that has been used for the assessment of the radiological impact, is shown in Figure 22.

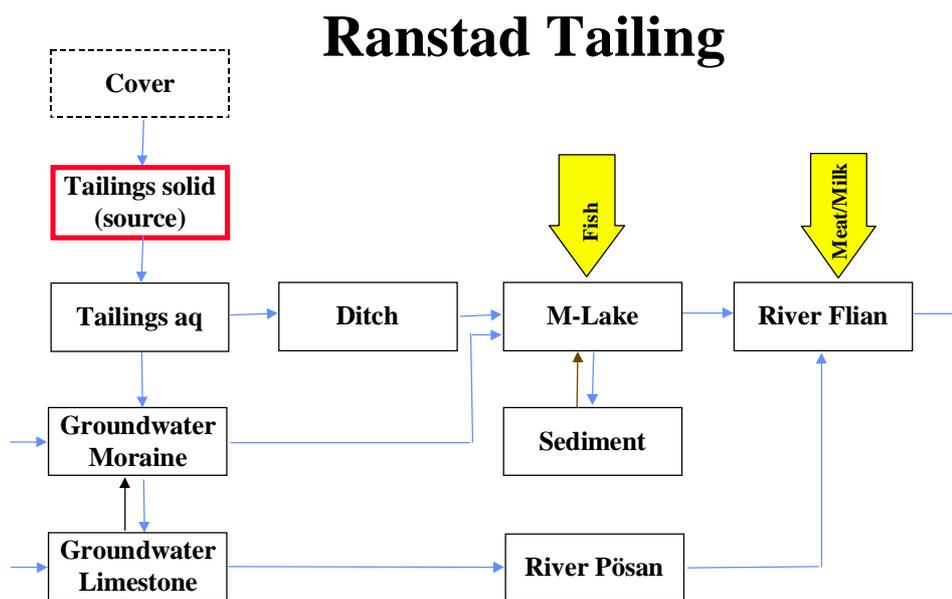


Figure 22 : Compartment scheme for the Ranstad Tailing site.

The restoration options included for the Ranstad tailing site have been identified in TD 10 (Stiglund and Aquilonius, 1999a). The economic and radiological data for quantifying the various attributes for each of those options are shown in Table 20. The corresponding utility values of the attributes are indicated in Table 21. The weighting factors of the attributes calculated by assigning values to their ratios, are given in Table 22.

Table 20 : Economic and radiological data (for integration times of 100 and 500 years) for different restoration strategies at the Ranstad tailing site. Best-estimate values.

Restoration strategy	Collective dose to population [man Sv]		Monetary costs of restoration [kECU]		Fraction of activity left on-site	Waste volume (m ³)
	100 y	500 y	Remediation	Waste disposal		
A	0.59	24	0	0	1	0
C1	0.23	9.4	640,000	38,000	0.4	4.5·10 ⁵
D1	0.13	5.5	730,000	38,000	0.2	1.5·10 ⁵
E1	0.37	15	9,500	0	1	0
E2	0.19	8.1	16,000	0	1	0
F2	0.051	1.8	23,000	0	1	0
G2	0.034	1.1	32,000	0	1	0

Table 21 : Utility values for best-estimate values of the attributes for each of the remediation options at the Ranstad tailing site.

Option	Utilities						
	Health		Economical		Social		
	100 years	500 years	Remedia.	Waste	Reassurance		Disturbance
				100y	500 y		
A	0	0	100	100	0	0	100
C1	64.7	63.8	12.3	0	69.6	69.4	0
D1	82.7	80.8	0	0	91.4	90.4	66.7
E1	39.6	39.3	98.7	100	19.8	19.7	100
E2	71.9	69.4	97.8	100	36.0	34.7	100
F2	96.9	96.9	96.8	100	48.5	48.5	100
G2	100	100	95.6	100	50.0	50.0	100

Table 22 : Weighting factors for attributes and sub-attributes applied in the optimisation of remediation of the Ranstad tailing site. Best-estimate values.

Health factors		Economic factors		Social factors		
100 years	500 years	100 years	500 years	100 years	500 years	
7.64·10 ⁻⁵	2.97·10 ⁻³	1.0	0.996	1.91·10 ⁻⁵	7.43·10 ⁻⁴	
	100 years	500 years	Remediation costs	0.95	Reassurance	0.86
Dose population	1	1				
Dose workers	-	-	Waste disposal costs	0.05	Disturbance	0.14
Non-radiation	-	-	Monitoring costs	-	Loss/gain of income	-
			Loss/gain of taxes	-		

The values of the attributes, from which the utility values are determined and the weighting factors indicated above have all been sampled in a triangular distribution between 1.5^{-1} - 1.5 times the most probable value given in Table 20 and Table 22. Negative correlation between collective doses and remediation costs has been applied with a correlation coefficient of -0.8 .

The overall scores of the remediation options have been determined from the weighted sum of utilities as shown in Figure 23.

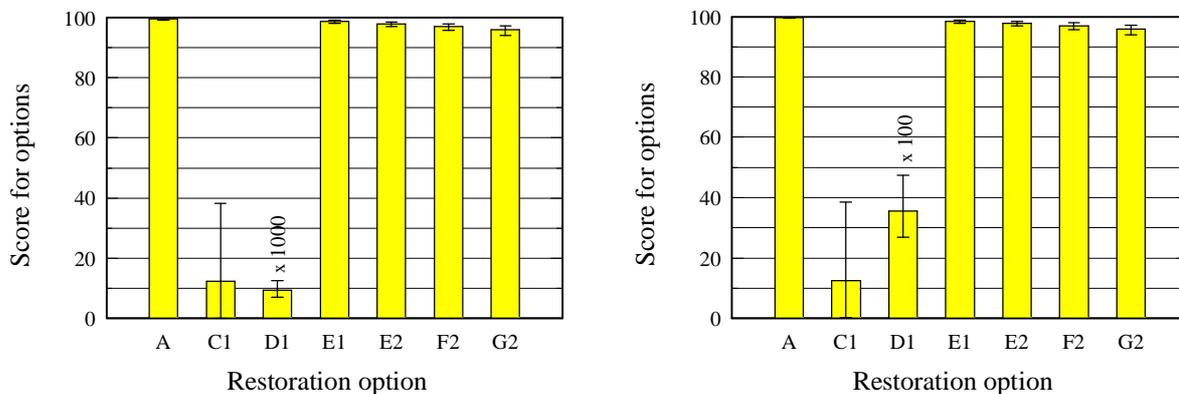


Figure 23 : Overall scores for different remediation strategies for the Ranstad site ("x 100" means that the indicated value is 100 times higher than the actual one)).

The left picture shows the results for an integration time of 100 years for the collective dose and the right picture shows them for an integration time of 500 years. The error bars represent the 5th and 95th percentiles of the distributions of the scores.

As can be seen from Figure 23, option A has the highest score. The options E1, E2, F2 and G2 have all a more or less equal score, not significantly lower than that of option A. The options C1 and D1 both have a low score due to high remediation and waste disposal costs. The 'no remediation' option A can thus be considered as the optimum solution for the Ranstad site and also the cheapest. There is no significant difference between the scores for the two different integration times except for option D1.

7.1.5. Lake Tranebärssjön

The location of the Lake Tranebärssjön site is approximately 5 km east of the Ranstad tailing site. It is a former uranium mine (open pit mining) which was in operation between 1965 and 1969. The lake has been existing only since 1990, when the mine was flooded by water. Its dimensions are 2000 m length, 100-200 m width, and 15 m depth, giving an open area of 250 000 m².

The lake Tranebärssjön is not considered to be a radiological problem even though the Swedish Radiation Protection Agency have decided that ²²⁶Ra should be measured four times a year at the outlet of the lake. During the last three years the radioactivity has not exceeded 10 mBq L⁻¹. Since there is a lack of information in order to restore a lake this study has focused on the restoration of the outgoing water from the lake. Both physical separation (filtration) and biological separation (bio-sorption) have been considered as restoration options for the lake.

The compartment scheme for the Lake Tranebärssjön site, that has been used for the assessment of the radiological impact, is shown Figure 24.

Lake Tranebärssjön

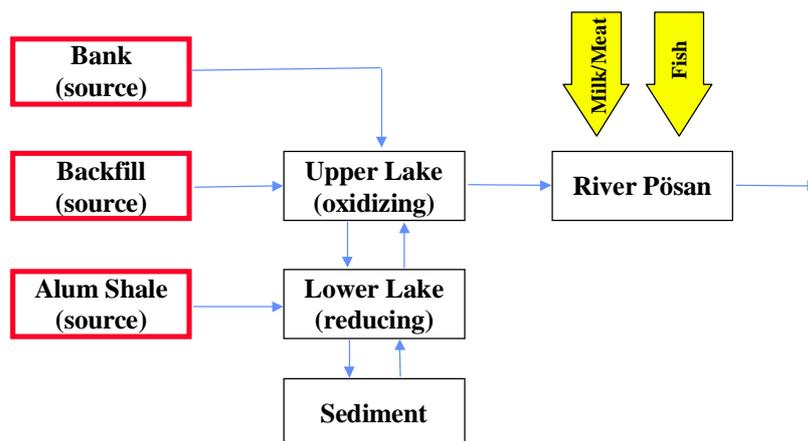


Figure 24 : Compartment scheme for Lake Tranebärssjön.

The restoration options included for the Lake Tranebärssjön site have been identified in TD 13 (Stiglund and Aquilonius, 1999b). The economic and radiological data for quantifying the various attributes for each of those options are shown in Table 23. The corresponding utility values of the attributes are indicated in Table 24. The weighting factors of the attributes calculated by assigning values to their ratios, are given in Table 25.

Table 23 : Economic and radiological data (for integration times of 100 and 500 years) for different restoration strategies at the Lake Tranebärssjön site. Best-estimate values.

Option	Collective doses to population [man Sv]		Monetary costs [kECU]	Fraction of activity left on-site
	100 y	500 y		
A	0.069	0.27	0	1
C2	0.0081	0.033	400,000	0.1
D3	0.002	0.0089	700,000	0.03

Table 24 : Utility values for best-estimate values of the attributes for each of the remediation options at the Lake Tranebärssjön site.

Option	Utilities					
	Health		Economic	Social		
	100 y	500 y		Reassurance		Disturbance
				100 y	500 y	
A	0	0	100	0	0	100
C2	90.9	90.8	42.9	91.8	91.8	42.9
D3	100	100	0	100	100	0

Table 25 : Weighting factors for attributes and sub-attributes applied in the optimisation of remediation of the Lake Tranebärssjön site. Best-estimate values.

Health factors			Economic factors		Social factors	
100 years	500 years		100 years	500 years	100 years	500 years
$7.64 \cdot 10^{-5}$	$2.97 \cdot 10^{-3}$		1.0	1.0	$1.91 \cdot 10^{-5}$	$7.43 \cdot 10^{-4}$
	100 years	500 years	Remediation costs	1	Reassurance	0.86
Dose population	1	1				
Dose workers	-	-	Waste disposal costs	-	Disturbance	0.14
Non-radiation	-	-	Monitoring costs	-	Loss/gain of income	-
			Loss/gain of taxes	-		

The values of the attributes, from which the utility values are determined and the weighting factors indicated above have all been sampled in a triangular distribution between 1.5^{-1} - 1.5 times the most probable value given in Table 23 and Table 25. Negative correlation between collective doses and remediation costs has been applied with a correlation coefficient of -0.8 .

The overall scores of the remediation options have been determined from the weighted sum of utilities as shown in Figure 25.

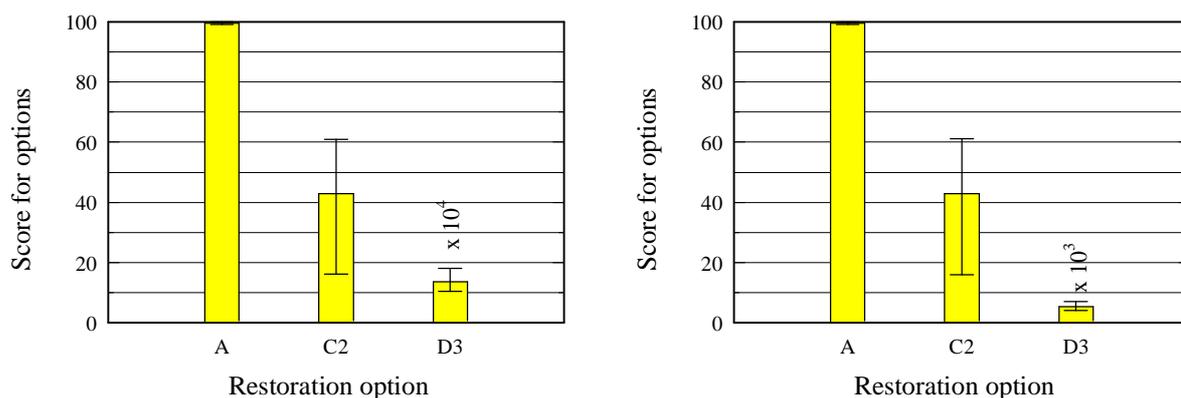


Figure 25 : Overall scores for different remediation strategies for the Lake Tranebärssjön site ("x 10^4 " and "x 10^3 " means that the indicated value is respectively 10^4 and 10^3 times higher than the actual one).

The left picture shows the results for an integration time of 100 years for the collective dose and the right picture shows them for an integration time of 500 years. The error bars represent the 5th and 95th percentiles of the distributions of the scores.

As can be seen from Figure 25, option A has the highest score. The options C2 and D3 both have a significantly lower score than that of option A due to high remediation costs. The 'no remediation' option A can thus be considered as the optimum solution for the Lake Tranebärssjön site and also the cheapest.

7.2 Conclusions

The ranking of various remediation options at the five European example sites, as carried out with multi-attribute utility analysis, is summarised in Table 26 and Table 27. The ranking of the remediation options at each example site nearly all give the result that 'no remediation' is the best option, *i.e.* having the highest score. The reason is the dominating weight of the economical attributes compared to the health and social attributes. The rather low collective doses and the potential for only low collective dose savings by remediation together with relatively high economic costs of the remedial measures are the cause of the low weights given to health and social factors. In addition, the low health and social weights are responsible for an only marginal difference between the scores for the situations where collective doses have been determined for a time period of 100 and 500 years.

Cost-benefit analyses based on best-estimates of collective dose and monetary costs would lead to a similar conclusion; none of the remedial measures considered for each site being justified. If more extreme values of collective doses or of their monetary equivalence are included in the cost-benefit analyses some of the remedial measures considered for the sites of Molse Nete River and Drigg would be justified. For the sites of Ravenglass, Ranstad and Lake Tranebärssjön no remedial measures are justified, not even if more extreme values of the collective doses are included.

The individual doses to critical groups without remedial measures being introduced at each of the example sites have also been compared with the IAEA criteria for clean-up of contaminated land (International Atomic Energy Agency, 1997). If a dose constraint for controlled practices would be applied to the outcome of the remediation process at the sites, some remediation might be needed at all sites.

The results of the justification and optimisation procedures applied above are summarized in Table 28.

Table 26 : Summary of the ranking of remediation options for the example sites. The integration time for the collective dose is 100 years.

Ranking No.	Molse Nete river	Drigg	Ravenglass estuary	Ranstad	Lake Tranebærssjön
1	No remediation (discharge stop ^a) (A)	Capping (E1)	No remediation (A)	No remediation (A)	No remediation (A)
2	Capping soil/sediment (E1)	Sub-surface barrier (E3)	Source removal (B)	Capping 0.5 m (E1)	Filtration (C2)
3	Physical immobilisation <i>in-situ</i> (F2)	No remediation (A)	Soil washing (C1)	Capping, 1.6 m (E2)	Biosorption (D3)
4	Chemical immobilisation <i>in-situ</i> (G2)	Physical immobilisation <i>ex-situ</i> (F1)	Chemical solubilisation (D1)	Physical immobilisation (F2)	
5	Physical immobilisation <i>ex-situ</i> (F1)	Chemical immobilisation <i>in-situ</i> (G2)		Chemical immobilisation (G2)	
6	Chemical immobilisation <i>ex-situ</i> (G1)	Chemical immobilisation <i>ex-situ</i> (G1)		Chemical separation (D1)	
7	Soil/sediment removal (B)	Physical immobilisation <i>in-situ</i> (F2)		Soil washing (C1)	
8	Physical separation (C1)	Filtration (C2)			
9	Chemical separation (D1)	Chemical Solubilisation (D1)			
10		Ion exchange ^b (D2)			
11		Biosorption ^b (D3)			

^(a)All options are with discharge stop

^(b)Very costly techniques (liquid treatment over many years)

Table 27 : Summary of the ranking of remediation options for the example sites. The integration time for the collective dose is 500 years

Ranking No.	Molse Nete river	Drigg	Ravenglass estuary	Ranstad	Lake Tranabärssjön
1	Capping soil/sediment (E1)	Capping (E1)	No remediation (A)	No remediation (A)	No remediation (A)
2	Physical immobilisation in-situ (F2)	Sub-surface barrier (E3)	Source removal (B)	Capping 0.5 m (E1)	Filtration (C2)
3	Chemical immobilisation in-situ (G2)	No remediation (A)	Soil washing (C1)	Capping, 1.6 m (E2)	Biosorption (D3)
4	Physical immobilisation ex-situ (F1)	Physical immobilisation <i>ex-situ</i> (F1)	Chemical solubilisation (D1)	Physical immobilisation (F2)	
5	No remediation (discharge stop ^a) (A)	Chemical immobilisation <i>in-situ</i> (G2)		Chemical immobilisation (G2)	
6	Chemical immobilisation ex-situ (G1)	Chemical immobilisation <i>ex-situ</i> (G1)		Chemical separation (D1)	
7	Soil/sediment removal (B)	Physical immobilisation <i>in-situ</i> (F2)		Soil washing (C1)	
8	Physical separation (C1)	Filtration (C2)			
9	Chemical separation (D1)	Chemical Solubilisation (D1)			
10		Ion exchange ^b (D2)			
11		Biosorption ^b (D3)			

^(a)All options are with discharge stop

^(b)Very costly techniques (liquid treatment over many years)

Table 28 : Summary of justification/optimisation of remedial measures at five European example sites and compliance with IAEA criteria for clean-up.

Site	Justification by cost-benefit	Compliance with IAEA criteria	Optimised strategy
Molse Nete river	'No remediation' has the highest net benefit (0) on central estimates; some options are justified on extreme values of doses	Remediation usually needed (constraint) or sometimes needed (no constraint) on grounds of annual individual doses	'No remediation' (100 years); Capping soil and sediment (500 years);
Drigg	'No remediation' has the highest net benefit (0) on central estimates; some options are justified on extreme values of doses	Remediation almost always needed (constraint) or usually needed (no constraint) on grounds of annual individual doses	Capping
Ravenglass estuary	'No remediation' has the highest net benefit (0) on central estimates and also on extreme values of doses	Remediation almost always needed (constraint) or usually needed (no constraint) on grounds of annual individual doses	'No remediation'
Ranstad	'No remediation' has the highest net benefit (0) on central estimates and also on extreme values of doses	Remediation sometimes needed (constraint) or rarely needed (no constraint) on grounds of annual individual doses	'No remediation'
Lake Tranebärssjön	'No remediation' has the highest net benefit (0) on central estimates and also on extreme values of doses	Remediation sometimes needed (constraint) or rarely needed (no constraint) on grounds of annual individual doses	'No remediation'

7.3 References

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