RESTRAT Final Report

Restoration Strategies for Radioactively Contaminated Sites and their Close Surroundings.

RESTRAT

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1. INTRODUCTION

A number of nuclear facilities in Europe are reaching the end of their as designed life expectancy. While the main technical part of the installations will be subject to a controlled decommissioning, in many cases contamination has been dispersed over the site or is contained by methods which may be unreliable for long-term storage. Restoration of such sites seems to be indicated both for the sake of the protection of the population and to relieve otherwise costly control of the site. However, the clean-up by conventional techniques is very expensive and may be very questionable on the basis of cost-benefit evaluations. Those problems are well-known, e.g. by the large clean-up programmes of former military sites in the USA and will become acute when decisions have to be taken about the future of sites, as well in Eastern as in Western Europe. The US experience shows that the application of alternative techniques, e.g. *in-situ* remediation is hampered by the lack of experiences on the site and of transparent risk assessments, considering the exposure of the present and future populations and of the restoration workers.

In order to tackle the most urgent problems related to site restoration, a robust and transparent decision-aiding methodology and relevant data are needed.

2. OBJECTIVES

The main objective of this project is to develop a methodology for ranking restoration techniques as a function of site- and contamination characteristics. A manual is to be elaborated in which this methodology is explained and applied to representative example sites of major classes of restoration cases.

The project is structured in following steps:

- identification and characterization of relevant cases (example sites);
- characterization of relevant restoration techniques;
- development of a risk assessment methodology (model);
- development of a ranking methodology of restoration options;
- evaluation of the restoration options at the example sites and formulation of conclusions
- elaboration of the manual.

Five representative example sites are studied:

- the BNFL Drigg site : a low-level waste disposal site (by Westlakes S.C.)
- the Ranstad tailing site : a restored uranium mining and milling site (by Studsvik EcoSafe)
- the Molse Nete : a contaminated freshwater river (by SCK•CEN)
- the Ravenglass estuary : a contaminated estuary (by Westlakes S.C.)
- the Tranebärssjön lake : a contaminated freshwater lake (by Studsvik EcoSafe)

They are representative of five major classes. The former two are related to solid sources of contamination, the latter three to liquid sources.

The characterisation of the example sites has to be carried out with a view to the assessment of the radiological impact and the ranking of the restoration options. Characteristics to be investigated encompass general ones (with respect to geography, geology, hydrology, meteorology and anthropology), physico-chemical properties (of surface soils, surface waters and groundwaters) and radiological features (sources, contaminations, exposure pathways). Particular attention shall be paid to physico-chemical processes influencing the source term evolution and subsequent behaviour of radioactive substances in the environment.

Possibly relevant restoration techniques are to be identified and their characteristics for use in the risk assessment model and ranking procedure to be determined. They encompass essentially: the applicability, costs, performance, side effects (principally waste arisings). An extensive literature review will be carried out and data on those techniques collected in a database

The ranking methodology for restoration options will be based on the radiation protection principles of justification and optimisation. The criteria and attributes that are to be taken into account, will be defined and quantified for the combinations of cases/restoration techniques considered. Radiological criteria (individual/collective doses or risks/to workers and population), economical ones (e.g. costs of restoration operations and waste disposal) and social ones (e.g. reassurance of the public, disturbance) will be taken into account. A review of international guidance from IAEA and ICRP in this respect will be drafted.

With respect to the radiological impact assessment a compartmental type of biosphere model (based on the BIOPATH/PRISM software of Studsvik) will be used. It has to be coupled with a chemical speciation model(s) in order to be able to take into account important physico-chemical characteristics influencing the behaviour of the radionuclides. The radiological impact on the population before and after the implementation of the restoration measures will be assessed. Collective doses will be determined as a measure of the total radiological health detriment; individual doses for the sake of the application of the IAEA criteria on clean-up of contaminated land. Also collective doses to the restoration workers will be taken into account. With respect to the collective doses to the population, uncertainty analysis will be carried out and the parameters contributing most to the uncertainty, identified.

In the manual the general methodology for ranking restoration options is described and the results for the example cases discussed.

3. PROGRESS

3.1 WP1 : Case Studies

In addition to the four specific contaminated sites to be studied according to the technical annex of the project proposal,

- the BNFL Drigg site (WP11) by Westlakes Scientific Consulting
- the Ranstad tailing site (WP12) by Studsvik
- the Molse Nete river (WP13) by SCK•CEN
- the Ravenglass estuary (WP14) by Westlakes Scientific Consulting,

a fifth site has been introduced,

- the Tranebärssjön lake (WP15) by Studsvik.

They are representative of five major classes. The former two are related to solid sources of contamination (wastes), namely discharges of solid waste, and relics of mining and milling. The latter three are related to liquid sources of contamination (wastes, sediment, ore body), namely contaminated freshwater rivers, estuaries and freshwater lakes.

The example cases have been characterised for the evaluations of the impact, to be carried out in WP4 and for the ranking of the restoration options, to be performed in WP5. Characteristics investigated encompass general ones (with respect to geography, geology, hydrology, meteorology and anthropology), physico-chemical properties (of surface soils, surface waters and groundwaters) and radiological features (sources, contaminations, exposure pathways). Also, potentially relevant restoration techniques have been listed for each site. The quantification of the parameters associated with each of the restoration options was carried out in WP3.

3.1.1 Drigg Site / Great Britain

General Characteristics

The Drigg Site is located on the coast of the Irish Sea about 9 km south of Sellafield (Great Britain). The site lies just west of the village of Drigg and about 300 m north of the tidal estuary of the River Esk at Ravenglass (see Figure 1). It has been used for the disposal of low-level radioactive waste since 1959. It is now operated by British Nuclear Fuel plc (BNFL) for the shallow burial of solid waste, mostly arising from the Sellafield site. Disposals of radioactive waste to the trenches and vaults at Drigg have all taken place in the north-western part of the 120 ha site and the present planning consent relates only to the 36 ha northern section.

The geology of the Drigg area consists of a complex heterogeneous sequence of glacial sediments overlying an irregular bedrock surface of Triassic sandstone. The glacial sediments range from compact boulder clays, through silts to coarse, highly permeable, sands and gravels. The whole site is underlain by at least one clay horizon. The clays restrict the downward percolation of surface infiltration but create the potential for lateral groundwater flow.

Several small streams cross the site, of which the most important are the Drigg stream and the East-West-stream which flows into the Drigg stream. The main Drigg stream flows off-site (c.$1\times10^6 m^3a^{-1}$) through cattle grazed pasture, before joining the River Irt which subsequently flows into the estuary at Ravenglass.

The site has an mean rainfall of 950 mm a^{-1} and a potential evapotranspiration rate of 560 mm a^{-1}. The mean temperature at the site lies between 5·8 and 11·5°C. The mean wind speed, which is a predominantly southerly wind, is 3·8 m s^{-1}.

The area next to the site is only sparsely populated (71,000), there is no local occupancy assumed for the site itself. Local agriculture is dominated by dairy cattle and sheep husbandry. There is little commercial fishing in the near vicinity of the site. The Drigg site is not a source of potable water.

Physico-Chemical Characteristics

Analyses of the streams at different locations about the site was also undertaken as part of the project. Samples of water and sediment (aqueous and solid phases) were taken and analysed in parallel by FZ Rossendorf and Westlakes S.C.The results are summarised in Table 1. This shows that the site is characterised by waters dominated by the sodium and calcium cations and the chloride and bicarbonate anions, with also a comparatively high silicate content. Iron and aluminium are mostly present in colloidal form.

The waters exhibit a low redox potential and a high ammonia contents. This will have a major influence on the chemical speciation of the redox sensitive contaminants such as plutonium and, to a much lesser degree, americium.

The sediment of the Drigg stream predominantly consists of illite.

This data was subsequently used in the calculation of site-specific distribution coefficients (see WP2).

Figure 1 : *Aerial View of Drigg Site.*

Table 1 *: Physico-chemical characterisation of stream water at the Drigg site.*

Component	Drain mol L^{-1}	Drigg Stream mol L^{-1}	East-West Stream mol L^{-1}
F^-	nd	4·5E-07	nd
PO_4^{3-}	2·0E-06	1·3E-06	3·5E-06
NO_3^-	7·1E-05	4·7E-04	8·1E-04
NO_2^-	3·7E-07	3·E-06	7·4E-06
NH_3	2·9E-05	1·7E-05	4·5E-05
SO_4^{2-}	5·7E-04	3·8E-04	3·2E-04
CO_3^{2-}	2·5E-03	1·6E-03	1·2E-03
Cl^-	2·4E-03	1·6E-03	1·5E-03
SiO_2	1·7E-04	1·2E-04	1·2E-04
K^+	1·2E-04	2·0E-04	2·5E-04
Na^+	2·4E-03	1·4E-03	1·1E-03
Ca^{2+}	1·9E-03	1·3E-03	1·2E-03
Mg^{2+}	5·5E-04	3·1E-04	2·9E-04
Fe	4·1E-05	2·6E-05	5·1E-06
Al^{3+}	1·7E-04	2·3E-06	2·9E-06
Zn^{2+}	nd	5·1E-07	6·7E-07
U	nd	8·1E-08	4·2E-10
Th	nd	7·3E-09	7·3E-09
Pb^{2+}	nd	4·6E-07	bdl
Ni^{2+}	nd	2·8E-07	1·2E-07
Mn^{2+}	nd	bdl	3·6E-08
Cd^{2+}	nd	1·6E-07	bdl
As	nd	4·3E-08	2·0E-08
pH:	6·45	6·92	6·57
Eh / mV:	59·7	96·0	157·1

nd = not determined

bdl = below detection limit

Radiological Characteristics

Initial disposal of waste were in seven shallow trenches. These were of most interest to this project. However, this practice has discontinued and the trenches, as they were filled, were covered with clay layer to reduce infiltration. This represented a partial remediation of this part of the site and for the purposes of this project calculations were made to estimate the behaviour of radioisotopes when the clay layer was absent. Therefore, the unremediated site was, in effect, a hypothetical situation.

The radioisotopes causing most concern were identified as uranium-238, caesium-137, plutonium-239 and americium-241. Estimates of their total inventories in the seven trenches were calculated and are given in Table 2.

The radionuclide concentration and total annual discharge rate from the trenches were estimated assuming unimpaired infiltration of the trenches. The results of these calculations are summarised in Table 3.

Table 2 : *Estimated inventory of radionuclides in trenches.*

Radionuclide	Inventory TBq
Caesium-137	2·4
Uranium-234,235,238	41·3
Plutonium-239,240	1·6
Americium-241	0·7

Table 3 : *Estimated radionuclide content and annual discharge in leachate from trenches.*

Radionuclide	Concentration kBq m^{-3}	Annual discharge MBq a^{-1}
Caesium-137	48·6	267
Uranium-234,235,238	28·1	93
Plutonium-239,240	8·6	47
Americium-241	6·5	36

Restoration Options.

Restoration techniques which are considered by the RESTRAT project are described in WP3. The selection of techniques which were suitable for the Drigg site took account of the nature and quantity of the waste, its location, its accessibility for treatment, the distance from a suitable point of disposal and the likely impact on the workforce.

The waste in the trenches at the Drigg site mostly contains radioactive objects rather than contaminated soil. However, the leachate into the streams will represent a secondary source of contamination. Public access to the site is restricted. However, the contaminated waste is readily accessible for remediation. Therefore, treatment on the site would be the most appropriate option.

A number of remediation techniques were rejected as being inappropriate to this site. In particular, since the waste is already located in a repository the removal of the source to another site is not considered a sensible option. The physical state of the waste also meant that separation approaches such as soil washing and flotation are rejected.

The remediation measures for this site fall into two categories: those which treat the leachate and those which treat the solid waste. The following options were considered most appropriate for the Drigg site:

A. No remediation

C2. Filtration

D1. Chemical Solubilisation

D2. Ion Exchange

D3. Biosorption

E1. Capping

E3. Sub-surface Barrier

F1. Physical Immobilisation, *ex-situ*

F2. Physical Immobilisation, *in-situ*

G1. Chemical Immobilisation, *ex-situ*

G2. Chemical Immobilisation, *in-situ*

3.1.2 Ranstad Tailing Site / Sweden

General Characteristics

The Ranstad Tailing Site is situated in the southern part of Sweden, in the country of Västra Götaland, at the borderline between the communities of Skövde and Falköping (Figure 2). The location of the area is about 20 km south of the city of Skövde, in the Billingen-Häggum district next to the Billingen mountain.

At the industrial facilities for the uranium mining and milling in Ranstad, 1.5 million tonnes of alum shale were mined between 1969 and 1984. About 215 tonnes of uranium oxide were extracted from the ore.

In 1984 the mining permit ceased and, as a consequence, the planning for the remediation began. This consisted of covering the mill tailings area with a multi-layer system (the dry depository) as well as filling the open pit mine with weathered shale and water (the wet depository).

In the mill tailing area (Figure 3), about 10^6 m^3 of tailings have been deposited, covering an area of 250 $m^{2.}$ The tailings consists of crushed alum shale, leached slag, from the uranium mining, and dams containing slimes from the uranium processing and from purification of the water.

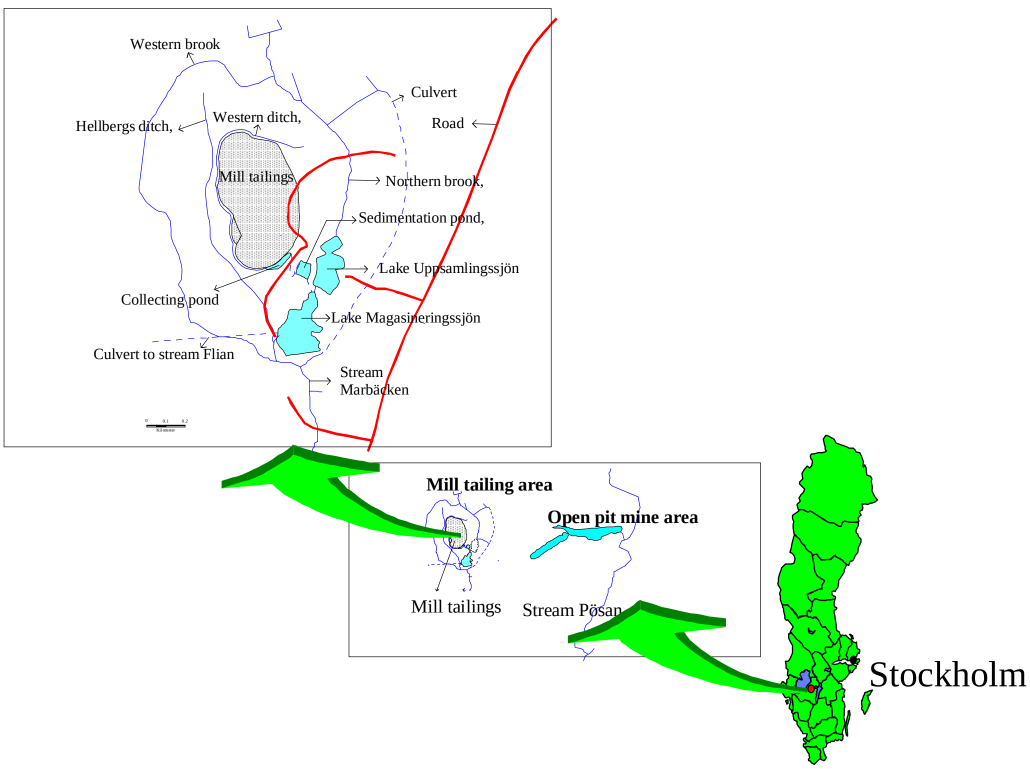


Figure 2 : *Geographical location of the Ranstad tailing site and details of the mill tailing area.*

The mill tailings are situated in an old discharge area with peat occurring in spots. Originally the Marbäcken stream flowed across the area.

When the residues from the extracted alum shale were placed in the area, two lakes were formed, Lake Uppsamlingssjön and Lake Magasineringssjön. The leachate-water is drained to a collecting pond, from which the water is pumped to the purification plant and sedimentation pond. Further on the leachate-water flows over a triangular weir to Lake Magasineringssjön. From the Lake Magasineringssjön the water is pumped via a culvert across the Billingen mountain to the Flian and Slafsan watercourses which discharge into the River Hornborgaån, which eventually discharges into Lake Hornborgasjön.

The area is sparsely populated, with some detached farms in the near vicinity. Two small villages, Stenstorp, south-east of Ranstad, and Skultorp, north-east of the tailing site, are situated within five kilometres of Ranstad. Stenstorp is inhabited by about 2000 people and Skultorp by about 3500. Forest and pastureland mainly cover the area around the Ranstad site.

Figure 3 : *View of the Ranstad tailing area*

Physico-Chemical Characteristics

The leachate from the tailing area, as well as the subsurface water, is being collected in a ditch surrounding the tailings, the Western ditch (Västra diket). Since the covering of the mill tailings during the years 1991-1992, the content of weathered minerals in the leachate has decreased. This is due to the almost oxygen-free environment, which has been established in the tailings. The infiltration and the oxygen content are measured continuously in the tailings. These measurements have shown that the sealing layer is almost impenetrable for oxygen.

Available primary (measured) data sets include:

- annual average values of pH, temperature, precipitation, conductivity and water discharges
- annual concentration values of sulphate, iron, nickel, cadmium, aluminium, manganese, magnesium, calcium, and uranium at various stations (surface- and ground water).

The results are summarised in Table 4

Table 4 : *Physico-chemical characteristics for Ranstad Tailing Compartments (mol/litre).*

Component	Compartment				
	Tailing	Moraine	Limestone	M-Lake	Ditch
F-	1.1E-4		7.1E-6		
PO_4^{3-}	2.0E-6	< 2.1E-5	5.3E-7	3.2E-8	1.5E-7
NO_3^-	1.2E-4	1.1E-5	7.1E-5	1.8E-5	9.0E-6
NO_2^-	3.2E-6	< 1.1E-5	2.9E-7	2.8E-7	3.0E-7
NH_4^+	4.5E-4		7.1E-6	1.7E-5	1.5E-4
SO_4^{2-}	2.3E-2	2.6E-4	5.4E-4	5.8E-3	1.0E-2
CO_3^{2-}	3.6E-3	3.7E-3	3.0E-3	1.6E-3	4.8E-4
Cl^-	5.6E-4	3.8E-4	3.2E-4	2.2E-4	1.9E-4
SiO_2	6.8E-5	4.6E-4	2.5E-4	3.3E-5	2.2E-4
K^+	1.2E-3	3.4E-5	3.8E-5	4.0E-4	1.0E-3
Na^+	8.6E-4	2.7E-4	2.9E-4	1.1E-3	5.9E-4
Ca^{2+}	1.1E-2	1.6E-3	1.7E-3	5.3E-3	9.3E-3
Mg^{2+}	1.0E-2	1.4E-4	2.1E-4	7.6E-4	2.6E-3
Fe	3.6E-7	3.6E-7	3.6E-7		
Al^{3+}	1.2E-7	2.0E-7	7.0E-8	1.1E-7	
Zn^{2+}	4.6E-7	3.2E-6	7.2E-8	1.7E-7	3.0E-7
U	1.0E-6	5.1E-9	4.9E-9	3.0E-8	1.0E-7
Th	1.3E-9	3.2E-9	3.1E-7	1.3E-9	
Pb^{2+}	2.0E-8	5.8E-8	8.5E-9	2.8E-9	3.2E-9
Ni^{2+}	6.3E-7	5.2E-7	7.5E-8	1.3E-7	1.8E-6
Mn^{2+}	8.7E-5	5.6E-6	7.1E-6	8.5E-6	6.9E-5
Cd^{2+}	4.2E-9	1.3E-9	3.0E-10	3.1E-9	3.6E-9
As	2.5E-8	1.6E-8	1.0E-8	3.4E-8	5.1E-8
pH	7.46	6.93	7.85	7.8	6.88
T (in °C)	8.8	9	9	7.9	8.5
Eh (in mV)	346	800	800	318	374

Radiological Characteristics.

The contaminants in the water are not only radionuclides, such as ^{238}U, but also manganese and nickel.

In the Ranstad area the critical group concerned consists of farmers living in the neighbourhood. They consume locally produced meat and drink milk from cows locally farmed. The fish are taken from Lake Magasineringssjön. The drinking water is taken from the limestone aquifer underneath the tailings.

Fish are cultivated in Lake Magasineringssjön, by an active fishermen’s club located in the area. The arable land around the Ranstad site is mainly used for grazing of cattle or for growing crops used as over-wintering fodder for animals.

Restoration Options

In order to remediate a mill tailings, different restoration techniques can be considered. In the case of Ranstad mill-tailing site, three different categories of remediation techniques have been envisaged, containment, immobilisation and separation.

Containment reduces the amount of infiltrating water and the entrance of oxygen into the tailings. It is the percolating water together with oxygen that governs the weathering processes in the tailings. If the weathering processes stop, the amount of contaminants leaching from the tailings will be strongly reduced.

For the Ranstad tailing site two different types of capping have been considered. The first one consists of 0.5 m moraine, which covered the tailings before the remediation started, the other consisting of 1.6 m of different layers, which was applied during 1991-92.

Immobilisation is a technique where the aim is to reduce the mobility and solubility of contaminants. This can be done either by injecting solidifying material into the tailings, physical immobilisation, or by injecting immobilising reagents, chemical immobilisation. Since these methods would reduce the leakage from the tailing considerably they have been included in this study.

Separation techniques are useful in order to separate the contaminants from the tailings into a concentrated solution. Both physical and chemical separation can be used for this purpose. Even though such methods are not likely to be used when large amounts are to be separated, due to high costs, these techniques have been considered for the Ranstad tailing site.

The following options have been considered for the Ranstad tailing site:

- A. No remediation
- C1. Soil washing
- D1. Chemical Solubilisation
- E1. Capping (0.5 m)
- E2. Capping (1.6 m)
- F2. Physical Immobilisation, *in-situ*
- G2. Chemical Immobilisation, *in-situ*

3.1.3 Molse Nete River / Belgium

General Characteristics

Since 1956, controlled releases of low-level radioactive effluents have been made by the nuclear activities in the region of Mol into the River Molse Nete. The Molse Nete is a small river in the north-eastern part of Belgium (Figure 4).

At the Molse Nete site, the surface is a fine sand layer of 2 to 10 m thick. The underlying soil is constituted by the Kasterlee formation, which is a homogeneous fine sand layer (mode: 150 µm) of about 15 m thick At the base of this formation, small flint pebbles and lenses of clay can be found. The Kasterlee formation is lying upon the Diest formation, which is about 120 m thick at the Molse Nete site and is formed by clayey and medium to very coarse-grained sands.

The water table of the aquifer lies between one to two meters beneath ground level. The lower part of the Kasterlee formation contains a significant clay content which limits the water migration into the underlying Diestiaan aquifer.

The Molse Nete drains approximately 62 km^2. The relief of the region is flat and the slope of the Molse Nete is rather small, about 0.4%. The river has an average water velocity of 0.41 m/s (0.24 - 0.47 m/s) and a flow rate of 2.9 m^3/s in the wet season and 0.28 m^3/s in the dry season.

The site climate is temperate with a slight maritime influence. The annual average temperature is about 10°C. In the summer, a maximum of 36.2 °C can be reached, in the winter temperature can drop to -19 °C. The annual average precipitation, measured over the period 1968 to 1983, is 757 mm, with a minimum of 527 mm and a maximum of 996. The evapotranspiration amounts to 450 mm / year on an average. During more than 4 months per year, the relative humidity is greater than 90%.

The area surrounding the site consist of 75% meadows (mainly for milk production) and arable land, with large parts not used any longer, 10% woodland and 15% urban area. A special feature of the Molse Nete is its regularly dredging, where the sediment deposited on the banks is sometimes used to ameliorate the grassland or vegetable gardens. The river is occasionally used for fishing and swimming. Only a few houses are built along the river banks. The population density within a circle of 10 km is about 300 people/km^2. The nearest larger community downstream the discharge point is the center of Geel, with a few ten thousand inhabitants, at 1 km (shortest distance) from the river.

Physico-chemical Characteristics

Samples of the water column of the river were taken by SCK and shipped to FZ Rossendorf for analysis. For most of the ions and cations the analytical values are in good agreement with the results of measuring campaigns in the past. Analysis of the water samples from the two points of investigation did not reveal larger changes, so the water can be regarded as homogeneous throughout the course of the river, at least inside the defined site area. The results are summarized in Table 5.

With respects to the river sediments not much is known about the mineralogy. They mostly consist of quartz sands and clays based on mica, glauconite and flint. The soil is based on glauconite and iron-enriched sandstone.

Main components are chlorides and (hydrogen)carbonates of sodium and calcium, under oxidizing redox conditions at neutral pH. The silicate content is rather high, whereas iron and aluminium are only trace components, thereby not heavily influencing the contaminant speciation. Moreover the observed ammonia and nitrate concentrations indicate that the water is slightly polluted.

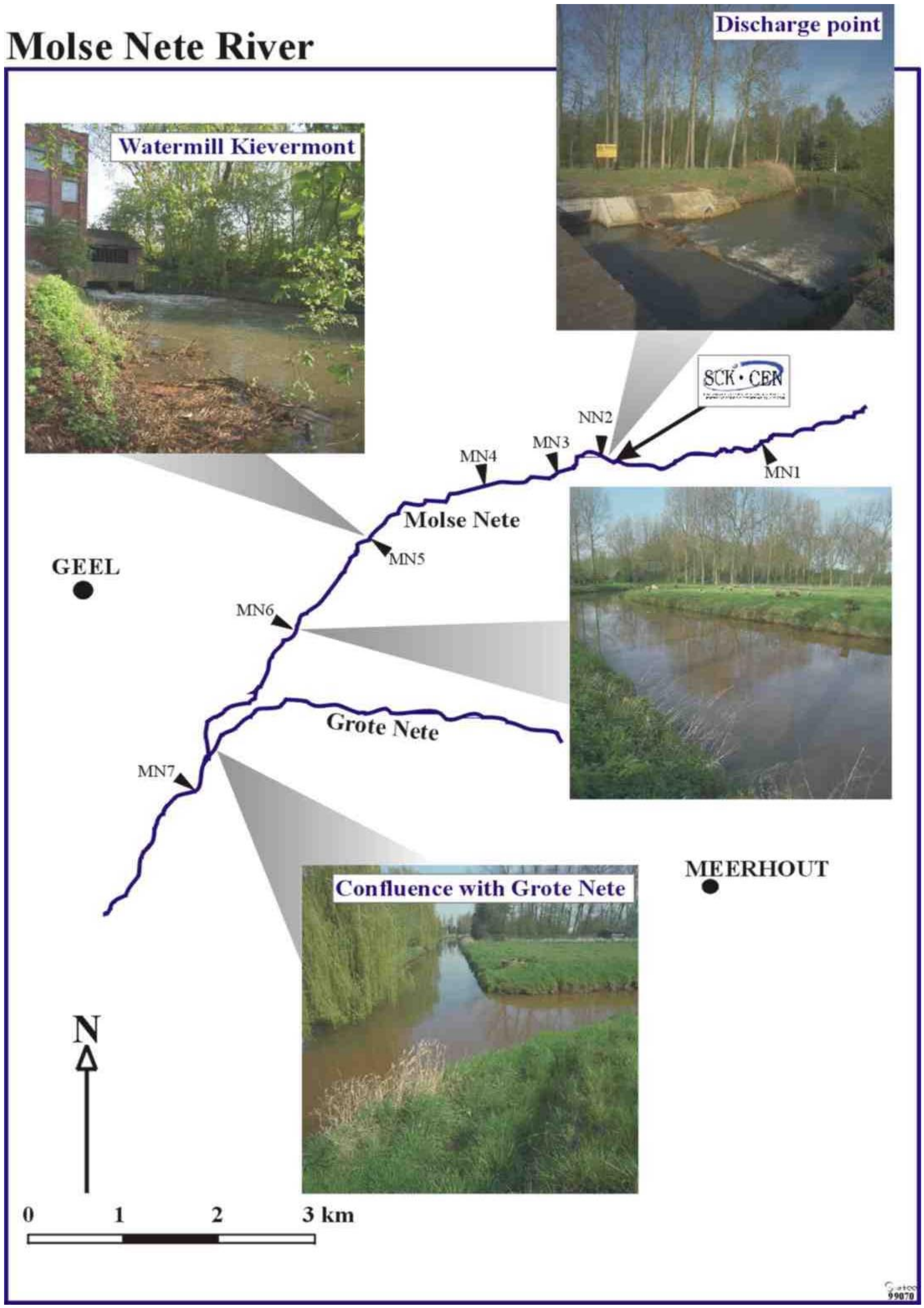


Figure 4 : *Views on the river of the Molse Nete*

Table 5 *Physico-chemical characteristics of the Molse Nete*

Distance:	0.7 km mol / L	3.7 km mol / L
PO_4^{3-}	< 2.105E-05	< 2.105E-05
NO_3^-	3,290E-04	3,790E-04
NO_2^-	< 2.174E-05	< 2.174E-05
SO_4^{2-}	7,777E-04	9,900E-04
CO_3^{2-}	1,560E-03	1,613E-03
Cl^-	1,461E-03	1,492E-03
SiO_2	1,844E-04	1,873E-04
K^+	2,535E-04	3,018E-04
Na^+	1,770E-03	1,823E-03
Ca^{2+}	1,173E-03	1,267E-03
Mg^{2+}	2,802E-04	3,863E-04
Fe	8,953E-07	8,953E-07
Al^{3+}	8,747E-08	8,005E-08
Zn^{2+}	8,321E-07	3,304E-07
U	2,479E-09	6,260E-09
Pb^{2+}	2,413E-10	2,413E-10
Ni^{2+}	1,457E-07	2,095E-07
Mn^{2+}	2,257E-07	2,390E-06
As	4,5514E-08	4,271E-08
pH:	7,20	7,20

Radiological Characteristics

The most important radionuclides, discharged into the Molse Nete are ^{60}Co, ^{137}Cs, ^{239}Pu and ^{241}Am. Over the period 1961 to 1990, about 130 GBq ^{60}Co/y, 37 GBq ^{137}Cs/y, 1.3 GBq ^{239}Pu/y and 0.68 GBq ^{241}Am/y were released. The discharges of radionuclides are nowadays lower.

The radionuclides ^{137}Cs, ^{60}Co, ^{241}Am and Pu, characterised by a high adsorption capacity, tend to accumulate in the soil and sediment and subsequently might lead on the long term to non-negligible doses.

Since releases have been going on for over forty years a lot of activity can be found in the sediments and in the soil next to the river. The river is dredged every five years and the sediment is then put on the banks. Some of the dredged sediment is subsequently applied onto agricultural soil. Irrigation of fields and pastures with water from the river is occurring a few times every year.

The radionuclide concentrations as measured in soil samples of the river banks during the period 1989 – 1990 are shown in Table 6.

Table 6 : *Radioactivity concentrations of soil samples (Bq kg-1) from the banks of the Molse Nete*

Distance to the discharge point (km)	^{60}Co	^{137}Cs	^{241}Am	*Pu*
+ 0.05	210	610	157	
+ 0.65	560	1120	450	250
+ 0.80	49	152	20	
+ 1.05	610	1290	104	
+ 1.65	69	96	18	
+ 2.30	380	890	710	550
+ 2.40	17.1	58	2	
+ 3.35	0.56	230		
+ 3.70	685	1200	190	140

The most important exposure pathways are :

- for ^{60}Co, the external irradiation through occupancy
- for ^{137}Cs, the ingestion of tubers, green vegetables and meat
- for ^{239}Pu and ^{241}Am, the ingestion of root vegetables and inhalation of dust.

Restoration Options

A first category of restoration techniques to be considered is the removal of radioactive sources (soil and sediment). This is obviously an effective measure, but implies the disposal of large volumes of soil and sediment as radioactive contaminated waste, which is very costly. The costs are reduced to reasonable values by only removing soils from fields and pastures that were assumed to have been contaminated with river sediment, and not those that were contaminated through irrigation with river water only.

The replacement of the removed soil by clean, fertile soil makes it again suited for agricultural activities and increases the cost only by a negligible amount.

The volumes of contaminated soil and sediment to be disposed of can be reduced further by the application of separation techniques on the excavated material. This is at the cost of a higher concentration of radionuclides in the waste remaining for disposal and consequently of a higher unit price of waste disposal. However, in this case the considerable reduction of volume of waste to be disposed of, more than counterbalances the higher unit cost of the waste disposal. The separation techniques taken into consideration are a physical one, soil washing and a chemical one, chemical solubilization.

A third category of restoration techniques that can be applied is containment. The capping of the contaminated soil and river sediment with asphalt concrete, or a hot asphalt mix yields a very effective barrier against water infiltration. Additionally, the major advantage of this technique is its low over-all cost. The dose to the population will be reduced to a negligible value but at the cost of a surface soil, that is no longer suited for agricultural practices. As a consequence the drawback of this technique is the loss of income (social attribute) and taxes (economical attribute) from agricultural activities. The application of only a layer of fertile soil would not suffice to make the soil again suited for agricultural activities, as it would in the case of soil removal.

The application of subsurface barriers, another containment technique is obviously not suited for this site.

A last category of restoration techniques to be considered is immobilisation; cement-based (physical) and polymer-based (chemical) immobilisation can be applied. Although immobilisation has not such a drastic influence on the soil water balance as capping has, the soil will not be suited for agricultural practices after treatment. Whether the application of a fertile soil layer will suffice for making the soil again usable for agricultural purposes, is questionable. As a consequence, the advantage of the reduction of the collective doses to negligible values is at the cost of the drawbacks of a loss of income (social attribute) and a loss of taxes (economical attribute) as for capping.

To conclude with, following restoration options were considered most appropriate for the Molse Nete river site:

A. No remediation

B. Soil and sediment removal

C1. Soil and sediment washing

D1. Chemical Solubilisation

D2. Ion Exchange

E1. Capping

F1. Physical Immobilisation, *ex-situ*

F2. Physical Immobilisation, *in-situ*

G1. Chemical Immobilisation, *ex-situ*

G2. Chemical Immobilisation, *in-situ*

3.1.4 Ravenglass Estuary / Great Britain

General Characteristics

The Ravenglass estuary is situated in West Cumbria (Great Britain) on the coast of the Irish Sea.

The site encompasses the tidal reaches of the Rivers Esk, Irt and Mite and occupies a total area of about 7·3 km^2 (see Figure 5). Its northern end of the estuary directly borders the Drigg example site.

The principal source of the estuarine sediments is the Irish Sea; the rivers contribute a much smaller fraction. The sediments can be divided into three categories (facies) reflecting their source of origin. The main classes are estuary deposits (channel facies, erosional facies, and bank facies), relic deposits and dune material. The channel facies mainly comprise of coarse grained sand. Bank facies deposits consist mainly of fine grained sediments, of sandy silt and silt grade. Erosional facies have a mixed grain size of silty sand grade. Measured erosion rates vary from 0 to 6 mm a^{-1}.

Channel facies have a mean sedimentation rate of 11 mm a^{-1}, but deposition is reported to be considered highly sporadic, partly in association with mobile beds. Lower bank facies have relatively high sedimentation rates of 28 mm a^{-1}, whereas the upper bank facies (salt marshes and intertidal pastures) have sedimententation rates of only 4 mm a^{-1}.

The hydrology is strongly influenced by seasonal temperature changes and the circulation patterns of the Irish Sea. An estimated 3 10^6 m^3 of water and 100 10^3 kg of sediment enters and leaves the estuary during each tidal cycle.

The site has a mean rainfall of 980 mm a^{-1} and a potential evapotranspiration rate of 510 mm a^{-1}. The mean temperature at the site lies between 6·6 and 12·7°C. The mean wind speed, which is predominantly from the south-west, is 4·5 m s^{-1}.

The area next to the site is sparsely populated (71,000). The small village of Ravenglass (population 200) is situated next to the estuary. The area is also used for recreational activities, both by the local population and by tourists. These activities include fishing, bathing, rowing and living on house boats. There is some pasture for cattle and over-wintering sheep.

Physico-Chemical Characteristics

Analyses of the estuarine waters at different point on the estuary were carried out as part the project. This information was used to summarise the chemical composition of the main aqueous phases involved in the modelling of the estuary. This is summarised in Table 7. It is clearly shown that the composition of the estuarine water is almost identical to that of the Irish Sea, demonstrating the strong tidal effects. This means a high ionic strength due to the presence of high concentrations of sodium and magnesium chlorides and sulphates.

The pH becomes slightly more basic from the river through the estuary to the open sea. The redox potential is in the reducing range, thus influencing especially the speciation of plutonium.

The principal mineral in the sediment was illite.

The data was subsequently used in the calculation of site-specific distribution calculations (see WP2).

Figure 5 : *Aerial View of Ravenglass Estuary.*

Table 7 : *Physico-chemical characteristics for the Ravenglass Estuary compartments.*

Component	**Compartment / Abbreviation**				
	Upper Banks **UB**	**Lower Banks** **LB**	**Channel** **CH**	**Sediment** **SD**	**Irish Sea** **IS**
	Mean **mol L^{-1}**	**Mean** **mol L^{-1}**	**Mean** **mol L^{-1}**	**Mean** **mol L^{-1}**	**Mean** **mol L^{-1}**
PO_4^{3-}	7·69E-05	-	-	-	-
NO_3^-	5·65E-05	-	-	-	-
SO_4^{2-}	2·64E-03	1·34E-02	2·45E-02	1·17E-02	2·67E-02
CO_3^{2-}	1·09E-04	1·33E-03	2·07E-03	2·00E-02	1·20E-03
Cl^-	4·82E-02	2·67E-01	4·89E-01	3·82E-01	5·07E-01
SiO_2	4·93E-05	3·06E-05	3·37E-05	3·89E-04	3·59E-05
K^+	9·85E-04	4·89E-03	9·42E-03	6·42E-03	9·43E-03
Na^+	4·23E-02	2·31E-01	4·14E-01	3·01E-01	4·19E-01
Ca^{2+}	1·15E-03	5·14E-03	9·75E-03	6·30E-03	9·56E-03
Mg^{2+}	4·57E-03	2·58E-02	4·68E-02	3·47E-02	4·80E-02
Fe	-	-	2·39E-06	5·30E-05	2·51E-06
Al^{3+}	-	-	3·78E-06	4·43E-07	2·08E-06
Zn^{2+}	5·75E-07	5·72E-07	1·52E-07	-	8·41E-08
U	1·81E-09	1·29E-08	1·67E-08	2·71E-08	1·70E-08
Pb^{2+}	-	-	3·62E-09	1·45E-09	2·90E-09
Ni^{2+}	1·57E-07	2·73E-07	-	-	-
Mn^{2+}	8·71E-07	3·60E-07	1·26E-07	3·01E-04	1·01E-07
Cd^{2+}	-	-	8·90E-09	-	-
As	6·01E-08	3·22E-07	-	-	-
pH:	7·02	8·04	7·93	7·33	8·07
Eh / mV:	-	-	122·6	218 0	129·1

Radiological Characteristics

Sediments are contaminated mainly via the Irish Sea from waste discharges from the Sellafield nuclear fuel reprocessing plant and, to a considerably lesser extent, from run-off from the Drigg site into the River Irt and sunsequently into the estuary.

The radionuclides causing most concern were identified as caesium-137, plutonium-239 and americium-241. Estimates of the total inventories for the estuary are 4·5 TBq caesium-137, 4 TBq plutonium-239 and 4 TBq americium-241.

Restoration Options

Restoration techniques which are considered by the RESTRAT project are described in WP3. The selection of techniques which were suitable for the Ravenglass site took account of the nature and quantity of the waste, its location, its accessibility for treatment, the distance from a suitable point of disposal and the likely impact on the workforce.

The estuarine environment presents some problems when considering the use of remediation technologies. The environment is tidal, it is a dynamic environment and, at times, can be turbulent. In addition, the public have access to the area which is within the Lake District National Park. These factors have to be reflected in selecting remediation technologies.

Restoration of the Ravenglass Estuary will primarily be concerned with remediation of the muddy banks of the mud flats and salt marshes which contain the highest levels of radionuclide activity. The radionuclides tend to be associated with the finer particles in the sediment.

Another consideration is that the highest radionuclide activities are not always present at the surface of the sediment.

It is concluded that *ex-situ* remediation technologies will provide the best options for the remediation of this example site. Such approaches also have the advantage that the technologies will be less susceptible to the tides and to the weather.

The above constraints meant that the choice of restoration options are somewhat limited. Those considered to be most practical are as follows:

- A. No remediation
- B. Source Removal
- C1 Sediment Washing
- D1. Chemical Solubilisation

Only the sediments of the upper banks are considered for restoration.

Quantification of parameters associated with each of restoration options, and needed for the RESTRAT project, was subsequently carried out in WP3.

3.1.5 Lake Tranebärssjön / Sweden

General Characteristics

Lake Tranebärssjön is situated close to the Ranstad tailing area (Figure 6). It originates from a former open pit mine that has been subsequently flooded.

In the industrial facilities for the uranium mining and milling in Ranstad, 1.5 million tonnes of alum shale were mined between 1969 and 1984. About 215 tonnes of uranium oxide were extracted from the ore.

In 1984 the mining permit ceased and, as a consequence, the planning for the remediation began. It consisted of covering the mill tailings area with a multi-layer system (the dry depository) as well as filling the open pit mine with weathered shale and water (the wet depository).

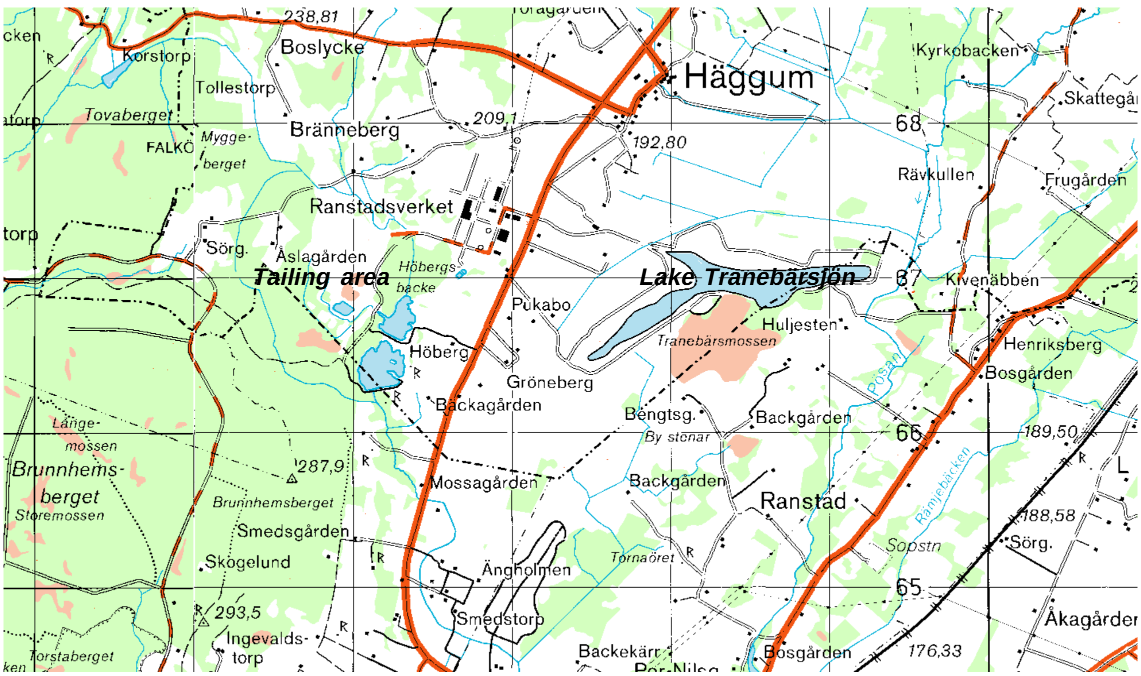


Figure 6 : *Map of the Ranstad area with Lake Tranebärssjön.*

Lake Tranebärssjön originates from a trench, 2000 m long, 100-200 m wide and 15 m deep, left by the alum shale mining operations (Figure 7). The lake has existed since 1990, when groundwater extraction from the open pit mine stopped. The general aim of the remedation was to re-shape the open pit into a lake. An overflow into a nearby Pösan stream keeps the water level constant.

The surroundings of Lake Tranebärssjön vary. One lakeside mainly consists of the undisturbed geological formation, with moraine on the top, then the limestone layer, the alum shale layer, and as the lowest layer of interest sandstone. The other side of the lake, including most of the lake bottom, consists of backfilled limestone and alum shale, covered by a thinner layer of backfilled moraine on the top.

The inflow of water into the lake is dominated by groundwater supplied from the moraine and limestone aquifers. The total volume of the water mass today is about 1 million cubic metres. In the western part of the lake the water depth is about 4 metres while it is about 15 metres (the maximum depth) in the eastern part. The western part consists to a large extent of wetland with plane slopes, while the eastern part, where the outlet is situated, has more abruptly sloping borders. The average flow at the outlet was

28 litre/second during 1997 and 48 litre/second during 1998 and relatively constant during the year except for the snow melting period. Lake Tranebärssjön discharges into the Pösan stream.

Figure 7 : *View of Lake Tranebärssjön*

The area is sparsely populated, with some detached farms in the near vicinity. Two small villages, Stenstorp, south-east of Ranstad, and Skultorp, north-east of the tailings site, are situated within five kilometres of Ranstad. Stenstorp is inhabited by about 2000 people and Skultorp by about 3500. Forest and pastureland mainly cover the area around the Ranstad site.

Physico-Chemical Characteristics

The water body in the lake is stratified by a thermocline at the depth of 8-10 metres. In the upper layer, oxidising conditions prevail and sulphates and carbonates of calcium and sodium, with a slightly basic pH dominate. The lower layer, showing reducing conditions and almost oxygen free conditions at the bottom, has a similar chemical composition, with an increased amount of Ca^{2+}, SO_4^{2-} and CO_3^{2-} (and also manganese and nickel) and the pH value is almost neutral. The water masses are mixed fast and drastically twice a year (on an average), during spring and autumn, which gives rise to an increased colloid content in the surface water. The colloids probably consist of iron oxides hydroxides. This process stains the lake red. The situation is stable only for some days, then the original state of the lake system recovers.

Available primary (measured) data sets include:

- time series and annual averages for pH, temperature, precipitation, water discharges, conductivity and a number of anion and cation concentrations for the two lake water sampling points and the groundwater pipes;
- metal, sulphur and silica concentrations for pore water from the sediment of the lake;

- annual concentration values of sulphate, iron, nickel, cadmium, aluminium, manganese, magnesium, calcium, and uranium at various stations.

The results are summarised in Table 8.

Radiological Characteristics

Lake Tranebärssjön is not considered to be a radiological problem even though the Swedish Radiation Protection Agency has decided that radium-226 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. The contaminants are the same as for the Ranstad tailing site, namely uranium-238, manganese and nickel.

The contaminants in Lake Tranebärssjön arise from three different sources.

- Bank material surrounding the lake, which is the material that had to be taken away before the mining in the open pit could start. The material consisted of alum shale and moraine.
- Backfill material in the lake, which is the same material as above.
- Alum shale. Weathering processes have occurred in the shale during the 30 years when the open pit was being pumped dry.

The transport of uranium from the outlet of the lake, as well as downstream of the Pösan stream, has increased during 1998, compared to the transport during 1996 and 1997 when it was decreasing. One explanation may be that the bank material, which later became backfill material when the mining ceased, is weathered and contributes to the high concentrations of uranium and other heavy metals in the lake. The weathering products are now delivered into the lake by the groundwater.

Since one part of the lake partly is a wetland, it habits different species of birds and ornithologists frequently visit the area. Canoeing as well as other recreational activities on the shores of the lake may also occur. Swimming is not very common, but a beach is located on the south shore of the lake. Fish species (salomonoid species) have been introduced in the lake, so fishing is possible, although not in large amounts since the fish population is quite small.

In the Ranstad area the critical group concerned consists of farmers living in the neighbourhood. They consume locally produced meat and drink milk from cows locally farmed and taking the water from the Pösan stream, fish is also captured in the stream.

The arable land around the Ranstad site is mainly used for grazing of cattle or for growing crops used as fodder for animals.

Restoration Options

Since there is lack of information in order to restore a lake, this study has focussed on reducing the discharge of pollutants via the outflow from the lake. Two different alternatives have been considered as being realistic for the particular lake, firstly the filtration of the outflowing water from the lake through a sandfilter and secondly the passing of the water through a wetland before its discharge into the Pösan river.

Table 8 : *Physico-chemical characteristics for Lake Tranebärssjön (mol/litre)*

Component	Compartment			
	Lake, top layer	Lake, bottom layer	Backfill	Alume shale
F^-	2.9E-5	3.2E-5		
PO_4^{3-}	3.8E-7	4.4E-7		
NO_3^-	8.8E-5	5.8E-5		
NO_2^-	2.2E-7	1.3E-7		
NH_4^+		7.4E-6		
SO_4^{2-}	7.8E-3	1.2E-2	8.7E-3	1.5E-2
HCO_3^-	3.0E-3	5.2E-3	4.6E-3	7.4E-3
Cl^-	3.6E-4	3.6E-4		
Si	8.8E-5	1.7E-4	1.7E-4	2.7E-4
K^+	1.4E-4	1.5E-4	1.6E-4	2.7E-4
Na^+	3.3E-4	3.7E-4	3.5E-4	5.5E-4
Ca^{2+}	8.7E-3	1.4E-2	9.6E-3	1.5E-2
Mg^{2+}	8.1E-4	1.2E-3	7.4E-4	1.5E-3
Fe	1.9E-5	3.6E-4	4.3E-7	1.4E-3
Al^{3+}	4.7E-7	4.9E-7	2.4E-7	3.2E-7
Zn^{2+}	4.4E-7	1.8E-7	2.8E-7	5.0E-7
U	5.6E-7	1.0E-6	7.0E-7	1.2E-6
Pb^{2+}	4.3E-9	6.8E-9		
Sr^{2+}			8.4E-6	1.5E-5
Ni^{2+}	7.1E-7	2.8E-6	2.4E-7	9.5E-7
Mn^{2+}	9.3E-6	1.1E-4	2.8E-7	1.6E-4
Cd^{2+}	3.6E-9	8.9E-9		
As	4.5E-8	8.8E-8		
Th	2.2E-9	2.4E-9		
pH	7.87	6.75	7.04	6.83
O_2(aq)	6.3E-4	3.8E-6		
T / °C	8	7.6	7	7

3.2 WP2 : Physico-Chemical Phenomena

3.2.1 Identification of the main physico-chemical processes and parameters

Chemical speciation is the distribution of one or more elements between all its possible species (distinct chemical entities) in a given system. The species distribution determines whether a contaminant is mainly a solute component - and thus easily transported and taken up - or is immobilized because of precipitation or adsorption onto a surface. Therefore changes in speciation can accelerate or slow down radionuclide migration. The source term is influenced mainly by processes like:

- Radioactive decay
- Complexation reactions (with organic and inorganic ligands), through hydrolysis, dissociation and association / polymerization
- Oxidation state changes / redox reactions
- Physical and chemical sorption onto mineral surfaces, ion exchange
- Precipitation and dissolution of solid phases, co-precipitation (inclusion & surface precipitation) of trace components, formation of solid solutions
- Extraction (in case of several fluid phases)
- Formation of colloids and aerosols
- Processes involving biological material, like biosorption, biologically catalyzed redox reactions, enzymatic reactions

The processes listed above can be described quantitatively by their respective functional terms, each requiring a unique set of parameters. Usually best known are the parameters defining the stationary state of a system. They are specific for each site and must therefore be measured:

- Temperature
- Pressure (total system and partial pressure of all gaseous components)
- Elementary composition and concentrations, this includes pH , ionic strength , humidity
- Composition of solid phases, requires identification of rocks and their mineral matrix
- Redox state, may be described by Eh, oxygen partial pressure, and potentials of important redox pairs
- Surface properties, including specific surface area, active sites, site densities, crystal size, structural disorders, charge distribution, surface films (biological matter !)
- Total water content

A second group of parameters describes the temporal and spatial evolution of a system. It arises from a rather heterogeneous set of disciplines, e.g., chemistry, biology, hydrodynamics, meteorology, and geophysics. Such parameters are not in the scope of this working package.

Finally, there are reaction specific, site-independent parameters:

- Thermodynamic parameters, i.e. equilibrium constants, solubilities, enthalpies, entropies, Gibbs free energies, heat capacities, partial molar properties, activity model coefficients
- Kinetic parameters (rate constants)
- Radioactive decay rates
- Degradation rates for biological material
- Parameters for biosorption

3.2.2 State-of-the-art chemical speciation and transport software

First, an overview of speciation and migration modelling software available was elaborated. Chemical speciation modelling software can be divided into programs to compute speciations with given thermodynamic parameters and programs combining speciation modules with a transport code (often called reactive transport codes or coupled transport codes). Unfortunately, none of the present models include a specific treatment for colloids, they are simply ignored in all cases.

Every program has a number of options to cover different areas of physico-chemical phenomena. Some of them require high learning efforts. Therefore it is hard for the potential user to judge, which is the proper approach for a particular application. To ease such decisions, it was necessary to determine the major selection criteria, and to apply them to some representative software packages, working out their strengths and weaknesses and giving recommendations as to their application. Features to be considered when evaluating speciation modelling software are:

- Can the program handle redox reactions, kinetic rate laws, adsorption, multiple phase equilibria?
- Which activity coefficient models are included?
- Which mathematical methods, especially minimization approaches, are applied?
- How is the performance of computational speed & 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 postprocessing tools?
- Which operating system and programming language is necessary?
- Are manual and/or source code available, how can support be obtained?

Additional features are less important, e.g. upper concentration limit, charge balance check, initialization of values, ability to cope with changes in volume, temperature or pressure etc.

As shown above, there are many speciation programs to chose from when it comes to an integration with the PRISM / BIOPATH software. Applying the selection criteria defined above, two software packages have been selected that are both available in source code. This is essential for the adaptions necessary to create interfaces between chemical modelling and risk assessment modules. Other advantages are that they are in use for many years now, have been checked by a number of validation programs and are recommended by international organizations. The programs are EQ3/6 and MINTEQA2. Both programs cover the whole range of chemical reactions in homogeneous aqueous solutions, including redox reactions, precipitation and dissolution equilibria. The main differences between them are as follows:

- only EQ3/6 is capable of handling with kinetic rate laws
- only MINTEQA2 has sorption models incorporated

3.2.3 Unfolding the K_d – a better approach to risk assessment

At present, all risk assessment codes (in the RESTRAT project the PRISM / BIOPATH program suite was chosen) rely on the wide-spread but simplistic K_d framework. It is built on the concept of distribution (or retardation) coefficients, which are defined as the ratio of the sorbed (fixed, immobilized) and unsorbed (free) fraction of a given component under equilibrium conditions. This widely used concept is, however, too simplistic because many very different basic physico-chemical phenomena (hydrolysis and complexations, redox reactions, mineral precipitation and dissolution, adsorption and ion exchange, colloid formation) are subsumed into just one empirical parameter. Any K_d used in prognostic studies is just a snapshot for a specific combination of Eh, pH, concentrations, and mineral composition, and its value is sensitive to even slight changes in some parameters. This in turn assigns very large uncertainties to them.

A better strategy is to unfold the single K_d value into a parameter vector, i.e. the decomposition of the K_d into its underlying basic processes, knowing the functional relationships between them and how they contribute to the K_d. Then a K_d can be calculated, even for simulated hypothetical scenarios with long-term drifts in the chemical environment. Moreover, those parameters affecting the K_d strongest can be identified, and consequently, extra measurements can be designed specifically to reduce their uncertainty. A realization of this concept requires the integration of a geochemical speciation program, such as MINTEQA2, into the risk assessment code PRISM / BIOPATH.

Besides chemical speciation in homogeneous aqueous solutions, combined with precipitation and dissolution of minerals, the interactions between dissolved species and surfaces are very important. In the literature there are many attempts to describe these interactions, that can be grouped into various phenomena, such as physisorption, chemisorption, co-precipitation, inclusion, diffusion, surface-precipitation, or even formation of solid solutions. Surface complexation in a strict sense only describes the chemisorption and has therefore to be combined with models for the other effects to ensure a proper thermodynamically based speciation model for the elements of interest. On shorter timescales it is nevertheless often the dominating process, having a fast kinetics. Processes like diffusion of sorbed ions into the host mineral and the subsequent formation of mixed crystals or solid solutions may then follow, but require much more time. Based on various textbooks and publications, it was decided to incorporate both non-electrostatic adsorption models (Distribution coefficient (K_d) model, Langmuir adsorption model, Freundlich adsorption model, Ion exchange model) and electrostatic adsorption models (Constant Capacitance model, Diffuse Double Layer model, Triple Layer model).

3.2.4 Thermodynamic databases

To use these sorption models in a geochemical speciation computation, an appropriate thermodynamic data base is required. Some older compilations of sorption data are available, but they exclusively focus on the K_d concept and are therefore of less value to this project. There is in general a lack of sorption parameters in the literature, the data coverage is by far more sparse than in case of the aqueous complex stability constants. Thus, an own general SCM data base was established under the MS Excel 5.0 software, after an extensive original literature review (that will be continued throughout the whole project). It comprises both the Constant Capacitance model, the Diffuse Double Layer model, and the Triple Layer model. At present, the data base covers the minerals ferrihydrite (Fe_2O_3*H_2O), goethite (α-FeOOH), haematite (α-Fe_2O_3), quartz (SiO_2), amorphous silica, pyrolusite (β-MnO_2), kaolinite, TiO_2 (Anatase), Al_2O_3, calcite ($CaCO_3$), muscovite, biotite, feldspars, chlorite, and fluor apatite. The following anions and cations are included: Cl^-, SO_4^{2-}, NO_3^-, CO_3^{2-}, K^+, Ca^{2+}, Mg^{2+}, Na^+, Ni^{2+}, Cu^{2+}, Ag^+, UO_2^{2+}, Al^{3+}, Pu^{4+}, NpO_2^+, Pb^{2+},Th^{4+}, Fe^{3+}, and Cd^{2+}. The data base contains all the intrinsic surface properties for the minerals such as specific surface area S_A, the number of distinct site types, the surface site density Γ or the electrostatic properties (capacities C_n of the various surface layers), and the thermodynamic complex stability constants for all investigated surface complexes.

From the general data base, specific ones for all RESTRAT example sites were derived, following the internal data format required by the MINTEQA2 program. The main radioactive contaminants are uranium, plutonium, americium, cesium and cobalt. Samples from the sites contained considerable amounts of freshly precipitated iron hydroxides such as ferrihydrite $Fe_2O_3 \cdot H_2O$. Their transformation into thermodynamically more stable minerals such as goethite or haematite exhibits very slow kinetics. Ferrihydrite has a very large specific surface and strong absorbing capacities, thus it was chosen as the major adsorbing surface. The Diffuse Double Layer model was selected to describe surface complexation. The respective intrinsic surface parameters, the uranium sorption parameters and the reaction constants for the ions competing with uranium for sorption sites were taken from textbooks and publications. To overcome a still observable lack for certain parameters, a Linear Free Energy Relationship (LFER) was utilized to derive stability constants for the species of interest from experimental results for other components. The finally derived thermodynamic database for sorption onto hydrous ferric oxides, described with the diffuse double layer surface complexation model, is shown in Table 9 below.

Anions are considered to sorb on strong and weak sites with the same stability constants. The simple cations Na^+, K^+, and Cs^+ and the anions CO_3^{2-}, Cl^- and NO_3^- are considered to be non-sorbing. This means that the contaminant ^{137}Cs, present in three of the examples cases, cannot be dealt with in the framework of the SCM, here other approaches such as ion exchange equilibria must be used. There were no data available for Al^{3+}, but an omission of Al^{3+} is not so critical, because the sorption of the contaminants is mainly competing with the major cationic components Ca^{2+} and Mg^{2+}.

Table 9 : *Thermodynamic database*

Ion	Reaction	log K_{intr}	Remark
UO_2^{2+}	$FeOH + UO_2^{2+} + H_2O = FeO\text{-}UO_2(OH) + 2\ H^+$	-3.12	Strong
	$FeOH + UO_2^{2+} + H_2O = FeO\text{-}UO_2(OH) + 2\ H^+$	-5.0	Weak
PuO_2^{2+}	$FeOH + PuO_2^{2+} = FeO\text{-}PuO_2^+ + H^+$	5.4	Strong, LFER
	$FeOH + PuO_2^{2+} = FeO\text{-}PuO_2^+ + H^+$	3.0	Weak, LFER
Pu^{4+}	$FeOH + Pu^{4+} + 3\ CO_3^{2-} + H^+ = FeOH_2\text{-}Pu(CO_3)_3^-$	58.0	Strong
Ca^{2+}	$FeOH + Ca^{2+} = FeOH\text{-}Ca^{2+}$	4.97	Strong
	$FeOH + Ca^{2+} = FeO\text{-}Ca^+ + H^+$	-5.85	Weak
Mg^{2+}	$FeOH + Mg^{2+} = FeO\text{-}Mg^+ + H^+$	-4.6	Weak, LFER
Co^{2+}	$FeOH + Co^{2+} = FeO\text{-}Co^+ + H^+$	-0.46	Strong
	$FeOH + Co^{2+} = FeO\text{-}Co^+ + H^+$	-3.01	Weak
Mn^{2+}	$FeOH + Mn^{2+} = FeO\text{-}Mn^+ + H^+$	-0.4	Strong, LFER
	$FeOH + Mn^{2+} = FeO\text{-}Mn^+ + H^+$	-3.5	Weak, LFER
Am^{3+}	$FeOH + Am^{3+} + 2\ CO_3^{2-} + H^+ = FeO\text{-}Am(HCO_3)_2$	29.0	Strong
SO_4^{2-}	$FeOH + SO_4^{2-} + H^+ = Fe\text{-}SO_4^- + H_2O$	7.78	LFER
	$FeOH + SO_4^{2-} = FeOH\text{-}SO_4^{2-}$	0.79	LFER
SiO_3^{2-}	$FeOH + SiO_3^{2-} + H^+ = Fe\text{-}SiO_3^- + H_2O$	15.9	LFER
	$FeOH + SiO_3^{2-} = FeOH\text{-}SiO_3^{2-}$	8.3	

In addition to the above discussed sorption database, also thermodynamic databases for the complexation, hydrolysis and redox processes had to be selected. Based on own database evaluations and reports in the literature, the ALT database accompanying the EQ3/6 package was mainly used as thermodynamic basis, with some minor modifications and additions.

3.2.5 Integration of chemical speciation modelling into the risk assessment software

At the moment no program is available that could act as a kind of superset of the two selected software packages EQ3/6 and MINTEQA2, including both kinetics and surface complexation. But the actual combination strategy to incorporate speciation codes into the PRISM / BIOPATH software is held so flexible that it needs comparatively small efforts to substitute the present speciation modules by another, better program. Here, the integration of MINTEQA2 may serve as an example for the integration in the risk assessment codes. Moreover, Figure 8 illustrates the data flow and the relationships between the various modules of the combined PRISM / BIOPATH / MINTEQA2 code.

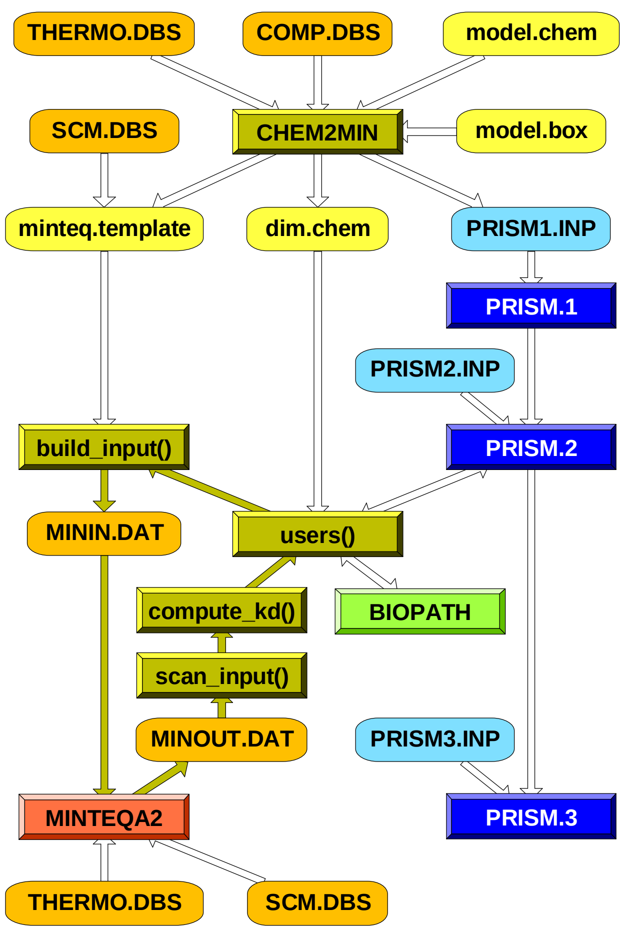


Figure 8 : *Dataflow*

The chemical model is defined in an extra file which is used to create both the input file for PRISM and the template for MINTEQA2. Thus it is assured that the model description is consistent for both parts of the modelling. This file is called MODEL.CHEM and has to be written by the user. It has a well defined, line-oriented structure. After some lines of general information for documentation purposes, detailed chemical data for each compartment of the model set-up are specified. Comment lines can be inserted everywhere to guide the user when checking the input file.

3.2.6 Computation of chemical speciation and distribution coefficients

The results for all RESTRAT example site (**I**: Drigg site, **II**: Ranstad tailing, **III**: Molse Nete river, **IV**: Ravenglass estuary, **V**: lake Tranebärssjön) compartments are presented in Table 10 below. They are each based on runs with 1000 varied parameter sets. Additionally, those parameters contributing most to the uncertainty of the various computed K_d values were identified by means of a ranked regression analysis. The table lists the two most important input factors for each contaminant / compartment couple, together with the amount of improvement in the K_d description attributed to that parameter.

Most of the calculated distribution coefficients (K_d values in m^3/kg) for uranium, americium, plutonium and cobalt fall well into the range used so far for modelling of these sites, but exhibit much smaller uncertainties. Where there were significant deviations, they could always be attributed to site-specific conditions. It should be noted, that all K_d values exhibit a log-normal distribution.

Table 10 : *Distribution coefficients for RESTRAT example sites*

Site and Compartment	Contaminant	log K_d ± σ	1^{st} Factor	2^{nd} Factor
I: Drain	Uranium	-0.42 ± 0.38	C_{HCO3-}: 71.7 %	pH: 8.9 %
	Americium	1.81 ± 0.66	pH: 66.9 %	C_{HCO3-}: 11.9 %
	Plutonium	1.94 ± 0.12	C_{HCO3-}: 63.6 %	C_{solid}: 10.9 %
I: Drigg Stream	Uranium	0.29 ± 0.32	pH: 36.1 %	C_{solid}: 27.4 %
	Americium Plutonium	1.59 ± 0.48 2.38 ± 0.19	C_{solid}: 21.5 % C_{solid}: 67.0 %	C_{HCO3-}: 14.1 % C_{HCO3-}: 8.8 %
II: Tailing Layer	Uranium	-2.16 ± 0.30	pH: 82.2 %	C_{solid}: 11.0 %
II: Moraine Layer	Uranium	-1.50 ± 0.28	pH: 79.3 %	C_{solid}: 17.0 %
II: Limestone Layer	Uranium	-4.69 ± 0.30	pH: 88.3 %	C_{solid}: 9.8 %
II: Storage Pond	Uranium	1.50 ± 0.21	pH: 69.5 %	C_{solid}: 18.6 %
III: River Water	Cobalt	-0.38 ± 0.18	pH: 83.8 %	C_{solid}: 11.9 %
	Americium	1.23 ± 0.12	C_{HCO3-}: 36.4 %	C_{solid}: 30.1 %
	Plutonium	2.49 ± 0.09	C_{solid}: 47.7 %	C_{HCO3-}: 19.9 %
IV: Upper Banks	Americium	-0.73 ± 0.41	C_{solid}: 63.0 %	C_{HCO3-}: 18.5 %
	Plutonium	2.27 ± 0.36	C_{solid}: 69.0 %	pH: 23.5 %
IV: Estuary Channel	Americium	-1.33 ± 0.84	C_{solid}: 94.7 %	pH: 4.2 %

	Plutonium	0.93 ± 0.56	C_{solid}: 96.2 %	pH: 2.8 %
IV: Sediment	Americium	2.03 ± 0.13	C_{solid}: 59.9 %	C_{Mg+2}: 21.7 %
	Plutonium	1.22 ± 0.16	C_{solid}: 38.6 %	C_{Mg+2}: 28.1 %
V: Upper Lake	Uranium	0.48 ± 0.15	C_{solid}: 36.8 %	pH: 34.6 %
V: Lower Lake	Uranium	0.38 ± 0.27	C_{HCO3-}: 84.9 %	C_{solid}: 11.1 %
V: Backfill	Uranium	-0.02 ± 0.29	C_{HCO3-}: 78.3 %	C_{solid}: 9.3 %
V: Alum Shale	Uranium	-0.27 ± 0.30	C_{HCO3-}: 79.7 %	pH: 8.8 %

The following contaminant-specific conclusions can be drawn:

- K_d values for uranium are especially sensitive towards uncertainties in pH and carbonate content, controlling the amount of hydrolysis species and carbonate complexes that reduce the sorbed portion of uranium;
- K_d values for americium are very small for highly mineralized waters;
- Plutonium sorption is best described assuming hexavalent plutonyl ions.

3.3 WP3 : Restoration Techniques.

3.3.1 Identification of relevant restoration techniques

A comprehensive literature review (187 literature sources) on restoration techniques has been collated in a computer database. Restoration techniques are taken to be techniques (or measures) which prevent (or reduce) the radiological impact (or risks) to the population from the residual contamination of contaminated sites. The techniques which were considered by this project were those which had been reported as having been used to remediate radionuclide contamination and there was adequate data to enable the technique to be comprehensively characterised. Techniques which had only been developed through to bench-scale experiments, had not been used to remediate radionuclide contamination or failed to provide sufficient data to characterise the technique were rejected.

Restoration technologies identified as being potentially relevant to this project fell 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. The disposal of the material can present a drawback to this approach.
- ***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). The disposal of the material can present a drawback to this type of approach. 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. Principal 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. Immobilisation techniques may be carried out both *in-situ* and *ex-situ*. Principal techniques include:
    - cement-based solidification;
    - chemical immobilisation.

3.3.2 Characterization of relevant restoration techniques

The restoration techniques which met the above requirements were then characterised in terms of the following criteria:

- ***Applicability***: The contaminants and the media for which they are suited; the length of time for which they would be applicable; and the manpower required to apply them.

- ***Performance***: The effectiveness against the contaminants (radionuclides). This is expressed in terms of:

- efficiency, which is may be measured as:
  - a decontamination factor or percentage removal of contaminant (appropriate to source removal or separation techniques),
  - change in hydraulic permeability (appropriate to containment techniques),
  - reduction in the mobility of contaminants (appropriate to immobilisation techniques).
- reduction in volume of waste (appropriate to removal of source or separation techniques).
- service life of the restoration technique.

- ***Cost***: Capital, operational and maintenance costs and the cost of disposing of the waste.

- ***Side effects***: In particular, the exposure of the workforce carrying out the remediation to the waste.

The ranges of performance, cost and exposure times (to restoration workers), based on data extracted from the literature, are summarised in Table 12. Where values could not be obtained from the literature then these were estimated on the basis of their similarity to other techniques for which values are available. This, along with the fact that data was extracted from literature which describes experiments carried out under a variety of conditions, for different types of waste and for different groups of radionuclides gives rise to a considerable degree of uncertainty associated with many of the values listed in this table. Consequently, criteria associated with each technique are defined in terms of a range of values rather than as a single value.

The application of this data to specific sites requires that individual values are selected from these ranges of values. This choice will be a matter of judgement depending upon factors such as accessibility, the nature of the contaminated waste and local conditions. However, it is assumed that the full range of reported costs and performances represent the uncertainties associated with each site. Triangular (for ranges of less than one order of magnitude) and log-triangular (for ranges of greater than one order of magnitude) distributions are assumed. Therefore, whilst the uncertainties will be independent of the site, the choice of mode for each distribution will be site-specific.

3.3.3 Application to example sites

The restoration options that were considered to be appropriate for each example site have been identified in WP1. They are brought together in

Table 11.

In Table 12 data is shown, which was used to assign values to the performance, costs and manpower exposure parameters for the application of the restoration options to each site. Also calculated were the volume of waste generated by the restoration option and the residual activity fractions left on site for each restoration option. These values were used as part of the ranking of the restoration options in WP5.

As an example, the choice of performance, cost and exposure parameters for the Drigg site (Table 13), the Ranstad tailing site (Table 15) and the Molse Nete river (Table 17) are given hereafter.

The calculated restoration, disposal and monitoring costs for each option along with the volume of waste generated and the activity left on the site are summarised in Table 14 (for Drigg), Table 16 (for Ranstad), Table 18 (for Molse Nete).

Table 11 : *Restoration options for the example sites*

	Drigg site	Ranstad tailing site	Molse Nete river	Ravenglass estuary	Trane-bärssjön lake
A Basecase (No remediation)	X	X	X	X	X
B Removal of Sources					
(Soil/Sediment excavation)			X	X	
C Physical Separation :					
Soil washing		X	X	X	
Filtration	X				X
D Chemical/Biological Separation:					
Solubilisation	X	X	X	X	
Ion Exchange	X				
Biosorption	X				X
E Containment : Capping	X	X[1]	X		
Subsurface Barriers	X				
F Physical Immobilisation : *ex-situ*	X		X		
In-situ	X	X	X		
G Chemical Immobilisation : *ex-situ*	X		X		
in-situ	X	X	X		

X : Restoration options considered

[1] Two thicknesses considered : 0.5 m and 1.6 m moraine

Table 12 : *Performance, cost and workforce exposure values of various remediation technologies.*

Technology	Case †	Medium	Performance Indicator		Cost (EUR)		Workforce exposure (manh)	Service life
Removal of Source	B		*Decontamination factor*		*Extraction (per m^3)*	*Disposal & transport (per m^3)*	*(per m^3)*	
Soil excavation		Solid	1 - 20		50 - 150	450 - 800	0·2 - 1	not applicable
					(per m²)		*(per m^2)*	
Soil scraping		Solid	1 - 20		1 - 3	450 - 800	0·03 - 0·3	not applicable
Physical separation			*Decontamination factor*	*Waste reduction*	*Excavation & separation (per m^3)*	*Disposal & transport of residue (per m^3)*	*(per m^3)*	
Soil washing	C1	Solid	1 - 10	50 - 98%	200 - 650	2000 - 3000	0·25 - 1·5	not applicable
Flotation		Solid	1 - 10	28 - 97%	65 - 390	2000 -3000	0·25 - 1·5	not applicable
			Decontamination factor		*Separation from liquid (per m^3)*			
Filtration	C2	Liquid	2 - >100		0·1 - 3·8	2000 - 3000	0·4 - 1·4	not applicable
Chemical separation					*Excavation & separation (per m^3)*			
Chemical solubilisation	D1	Solid	1 - 20		180 - 820	2000 - 3000	1·2 - 3·5	not applicable
					Separation from liquid (per m^3)			
Ion exchange	D2	Liquid	3 - 100(U), 20 - 100(Cs)		1·3 - 2·5	2000 - 3000	0·4 - 1·4	not applicable
Biological Separation								
Biosorption	D3	Liquid	2·5 - >100		1 - 3	2000 - 3000	0·4 - 1·4	not applicable
Containment			*Resultant permeability (m s^{-1})*		*Total (per m^2 surface area)*		*(per m^2 surface area)*	
Capping	E1 + E2	Solid	1×10^{-12} - 1×10^{-9}		30 - 45		0·03 - 0·3	1,000 y
Subsurface barrier	E3	Solid					(per m³ barrier volume)	
a) slurry walls			1×10^{-12} - 1×10^{-8}		510 - 710		0·06 - 0·4	100 - 1,000 y
b) grout curtains			1×10^{-12} - 1×10^{-8}		310 - 420		0·06 - 0·4	100 - 1,000 y
Immobilisation			*Mobility reduction factor*		*Total (per m^3)*		*(per m^3)*	
Cement-based solidification		Solid						
a) *ex-situ*	F1		5 - 25		75 - 300		0·25 - 1·5	not known
b) *in-situ*	F2		5 - 25		50 - 310		0·06 - 0·4	not known
Chemical immobilisation		Solid						
a) *ex-situ*	G1		5 - 50		110 - 570		0·25 - 1·5	not known
b) *in-situ*	G2		5 - 50		60 - 420		0·06 - 0·4	not known

† *RESTRAT designation code.*

Table 13 : *Site-specific values for the performances, costs and exposure times of remediation techniques appropriate to the Drigg site.*

Remediation option / Operation	Case †	Performance		Cost	Exposure Time
		Indicator	Value		(Restoration Workers)
Filtration (liquid)	C2	DF	100	0·75 EUR m^{-3}	0·7 manh m^{-3}
Chemical solubilisation	D1	DF	10		
Separation				550 EUR m^{-3}	2·3 manh m^{-3}
Excavation and transport (prior to solubilisation)				65 EUR m^{-3}	0·7 manh m^{-3}
Ion exchange (liquids)	D2	DF	5	2 EUR m^{-3}	0·7 manh m^{-3}
Biosorption (liquids)	D3	DF	10	2·5 EUR m^{-3}	0·7 manh m^{-3}
Capping	E1	k	1×10^{-10} m s^{-1}	35 EUR m^{-2} (barrier)	0·05 manh m^{-2} (barrier)
Subsurface barriers	E3	k	1×10^{-9} m s^{-1}	350 EUR m^{-2} (barrier)	0·1 manh m^{-3} (barrier volume)
Cement-based solidification (*ex-situ*)	F1	MRF	10	35 EUR m^{-3}	0·6 manh m^{-3}
Excavation and transport (prior to immobilisation)				65 EUR m^{-3}	
Cement-based solidification (*in-situ*)	F2	MRF	10	350 EUR m^{-3}	0·2 manh m^{-3}
Chemical immobilisation (*ex-situ*)	G1	MRF	10	180 EUR m^{-3}	0·6 manh m^{-3}
Excavation and transport (prior to immobilisation)				65 EUR m^{-3}	
Chemical immobilisation (*in-situ*)	G2	MRF	10	100 EUR m^{-3}	0·2 manh m^{-3}

DF = decontamination factor; k = permeability coefficient; MRF = mobility reduction factor.

† *RESTRAT designation code.*

Table 14 : *Site-specific monetary costs, residual activities and waste volumes for the Drigg site.*

Remediation strategy	**Volume of waste removed from site**	**Fraction of activity remaining after remediation**	**Monetary costs of remediation [EUR]**		
	m^3		**Remediation (incl. labour)**	**Waste disposal (incl. transport)**	**Monitoring costs**
Filtration	0	1	$3{\cdot}8 \times 10^{8}$	$3{\cdot}1 \times 10^{7}$	$7{\cdot}5 \times 10^{5}$
Chemical solubilisation	12500	0·01	$3{\cdot}0 \times 10^{8}$	$1{\cdot}0 \times 10^{8}$	$7{\cdot}5 \times 10^{6}$
Ion exchange	41000	0·1	$1{\cdot}0 \times 10^{8}$	$3{\cdot}1 \times 10^{7}$	$1{\cdot}5 \times 10^{7}$
Biosorption	12500	0·2	$1{\cdot}9 \times 10^{9}$	$3{\cdot}1 \times 10^{7}$	$7{\cdot}5 \times 10^{6}$
Capping	12500	0·1	$3{\cdot}5 \times 10^{6}$	0	$7{\cdot}5 \times 10^{7}$
Subsurface barrier	0	1	$6{\cdot}3 \times 10^{6}$	0	$7{\cdot}5 \times 10^{7}$
Physical immobilisation (*ex-situ*)	0	1	$5{\cdot}5 \times 10^{7}$	0	$7{\cdot}5 \times 10^{7}$
Physical immobilisation (*in-situ*)	0	1	$1{\cdot}9 \times 10^{8}$	0	$7{\cdot}5 \times 10^{7}$
Chemical immobilisation (*ex-situ*)	0	1	$1{\cdot}3 \times 10^{8}$	0	$7{\cdot}5 \times 10^{7}$
Chemical immobilisation (*in-situ*)	0	1	$5{\cdot}5 \times 10^{7}$	0	$7{\cdot}5 \times 10^{7}$

Table 15 : *Site-specific values for the performances, costs and exposure times of remediation techniques appropriate to the Ranstad Site.*

Remediation option / Operation	Case †	Performance		Cost	Exposure Time
		Indicator	Value		(Restoration Workers)
Soil washing	C1				
Chemical solubilisation	D1				
Capping, 0·5 m moraine	E1				
Capping, 1·6 m	E2				
Physical immobilisation (*in-situ*)	F2				
Chemical immobilisation (*in-situ*)	G2				

† *RESTRAT designation code.*

Table 16 : *Site-specific monetary costs, residual activities and waste volumes for the Ranstad site.*

Remediation strategy	Volume of waste removed from site	Fraction of activity remaining after remediation	Monetary costs of remediation [EUR]	
	m^3		Remediation (incl. labour)	Waste disposal (incl. transport)
Soil washing	450000	0·4	640000	38000
Chemical solubilisation	150000	0·2	730000	38000
Capping, 0·5 m moraine	0	1	9500	0
Capping, 1·6 m	0	1	16000	0
Physical immobilisation (*in-situ*)	0	1	23000	0
Chemical immobilisation (*in-situ*)	0	1	32000	0

Table 17 : *Site-specific values for the performances, costs and exposure times of remediation techniques appropriate to the Molse Nete Site.*

Remediation option / Operation	Case †	Performance		Cost	Exposure Time (Restoration Workers)
		Indicator	Value		
Source removal	B				
Soil		DF	7 (Co)	125 EUR m^{-3}	0·8 manh m^{-3}
waste disposal				700 EUR m^{-3}	
Sediment		DF	10 (Cs, Pu, Am)	150 EUR m^{-3}	0·9 manh m^{-3}
waste disposal				800 EUR m^{-3}	
Soil washing	C1				
Soil		DF	4 (Co)	350 EUR m^{-3}	0·95 manh m^{-3}
Excavation				125 EUR m^{-3}	
waste disposal:				2500 EUR m^{-3}	
Sediment		DF	6 (Cs, Pu, Am)	350 EUR m^{-3}	1·05 manh m^{-3}
Excavation				150 EUR m^{-3}	
waste disposal:				2500 EUR m^{-3}	
Solubilisation	D1				
Soil		DF	10	400 EUR m^{-3}	2·3 manh m^{-3}
Excavation				125 EUR m^{-3}	
waste disposal:				2500 EUR m^{-3}	
Sediment		DF	10	400 EUR m^{-3}	2·4
Excavation				150 EUR m^{-3}	
waste disposal:				2500 EUR m^{-3}	
Capping	E1				
Soil		k	10^{-10} m/s	40 EUR m^{-2}	0·25 manh m^{-2}
Sediment		k	10^{-10} m/s	45 EUR m^{-2}	0·28 manh m^{-2}
Physical immobilisation (*ex-situ*)	F1				
Soil		MRF	15	100 EUR m^{-3}	0·95 manh m^{-3}
Excavation				125 EUR m^{-3}	
Sediment		MRF	15	100 EUR m^{-3}	1·05 manh m^{-3}
Excavation				150 EUR m^{-3}	
Physical immobilisation (*in-situ*)	F2				
Soil		MRF	15	200 EUR m^{-3}	0·30 manh m^{-3}
Sediment		MRF	15	250 EUR m^{-3}	0·35 manh m^{-3}
Chemical immobilisation (*ex-situ*)	G1				
Soil		MRF	10	180 EUR m^{-3}	0·95 manh m^{-3}
Excavation				125 EUR m^{-3}	
Sediment		MRF	10	180 EUR m^{-3}	1·05 manh m^{-3}
Excavation				150 EUR m^{-3}	
Chemical immobilisation (*in-situ*)	G2				
Soil		MRF	10	200 EUR m^{-3}	0·30 manh m^{-3}
Sediment		MRF	10	250 EUR m^{-3}	0·35 manh m^{-3}

DF = decontamination factor; k = permeability coefficient; MRF = mobility reduction factor. † *RESTRAT designation code.*

Table 18 : *Site-specific monetary costs, residual activities and waste volumes for the Molse Nete site.*

Remediation strategy	Volume of waste removed from site	Fraction of activity remaining after remediation	Monetary costs of remediation [EUR]		
	m^3		Remediation (incl. labour)	Waste disposal (incl. transport)	Monitoring costs
Source removal	26520	0·1	3570	1000	19580
Soil washing	5300	0·3	12870	2000	13260
Solubilisation	10600	0·1	13970	2000	13260
Capping	0	1	4250	3200	0
Physical immobilisation (*ex-situ*)	0	1	6220	3200	0
Physical immobilisation (*in-situ*)	0	1	5810	3200	0
Chemical immobilisation (*ex-situ*)	0	1	8340	3200	0
Chemical immobilisation (*in-situ*)	0	1	5810	3200	0

3.4 WP4 : Risk Assessment

3.4.1 General approach

In this working package a methodology is developed for assessing the radiological impact on man as a major attribute in the ranking and selection procedure of the restoration options for radioactive contaminated sites. As well impact on the public as the impact on the restoration workers is to be taken into account. When the radiological impact is brought about by an event or a scenario that is -quasi- certain to happen, it is expressed in terms of doses; when the event or scenario shows only a limited probability of occurrence, the impact is expressed in terms of risk. In this study the scenarios considered all show a high probability of occurrence and therefore only doses are considered.

A dose assessment model has been developed for every site. Individual doses to average members of the critical group and collective doses as a measure of the total health detriment are taken into account.

Impact scenarios were developed separately for workers and public. For the workers the dose impact is brought about during the restoration works through inhalation of resuspended dust and through external irradiation. It can be calculated very straightforward from their exposure times and from the contamination levels.

For the public the radiological consequence of a restoration is a dose reduction (aversion); i.e. the dose impact without restoration minus the dose impact with restoration. Basic exposure scenarios for the public include for instance exposures due to agricultural or fishing activities, residence on contaminated sites. In order to calculate these dose impacts a more comprehensive system, the biosphere, is to be considered and the exposure also takes place through ingestion, next to inhalation and external irradiation.

The model used for the dose assessments can be divided into two parts:
- the transport model;
- the exposure model.

The transport model

Transport modelling in this study is based on compartment theory. The system is divided into a number of physically defined areas or volumes, i.e. compartments. A compartment is a component of the biosphere with similar properties but which may vary substantially between different compartments such as surface water, ploughed soil. Exchange between these compartments is described by rate constants expressed in number of turnovers per unit of time. 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 from a compartment is solely dependent upon the quantity of the element in that particular compartment
- the compartment is instantaneously well mixed
- all elements have the same probability of leaving the compartment

The first condition is always fulfilled when the physical amount of the outflow is relatively small and has no influence on the remaining fraction.

In general, compartments can be designed to fulfill the condition of instantaneous and homogeneous mixing with satisfactory precision. This is especially valid in cases where the time studied is long compared with the turnover rate of nuclides within the compartment. Soils for agricultural purposes are continuously ploughed and the current practices of crop rotation reduce the in-homogeneity for pasture land. Water bodies can be considered well-mixed using annual mean concentration for the dose assessment. Sediments may be areas with strong gradients. However, dividing the sediment into layers at different depths can reduce the inhomogeneity within compartments.

The amount of radioactivity in a given compartment is dependent on:

- the source term for the compartment system, such as the direct release to one or several compartments, or generation within them by decay from the parent nuclide
- the outflow to and inflow from other compartments
- radioactive decay

The transfers of radionuclides between the compartments are being brought about by various processes. These processes may be driven by man, such as irrigation or dredging, or may be of natural origin, such as water flow, advection, diffusion, infiltration, resuspension, sedimentation, bioturbation.

Also within the compartments transfers can take place, for instance between phases. Important transfers can take place between aqueous and solid phases in soil or surface waters by the processes of sorption and desorption, characterized in equilibrium by the Kd value. Several transfer processes between compartments may be only applicable to the solid (sorbed) or the aqueous (dissolved) fraction of the radionuclides.

The exposure model

The radionuclides present in contaminated media of the biosphere may expose man through different exposure pathways. These pathways can be of an external or internal nature.

External exposure can be due to contamination of various media (e.g. soil, sediment, surface water or air) which may irradiate man directly or to direct contamination of the surface of human bodies. The external dose to man is calculated from the radionuclide concentrations (or intensities of gamma radiation) in the irradiating media, the duration of the exposure and the corresponding dose rate factors. In literature, dose rate factors can be found for some standard conditions such as immersion in uniformly contaminated water or air, exposure at 1 metre above an infinite, uniformly contaminated slab source etc. Such dose rate factors have been used in this study together with the appropriate reduction factors in order to make allowabce for restricted source sizes and shielding effects.

The internal doses is due to radiation from nuclides within the human body. The nuclides, in general, reach the body via intake of food and water, or via inhalation. The radioactive element will be either eliminated or retained in the body, where it can participate in the metabolism, dependent on its chemical and physical properties. The internal dose to man is calculated from the radionuclide concentrations in foodstuffs, water or air, the consumption or inhalation rates and the dose factors for ingestion or inhahalation. The dose factors used in this study are those that have been published by the International Commission on Radiological Protection. They are based on models of the metabolism of radionuclides inhaled or ingested by man.

The major exposure pathways normally considered (for the general public), 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 on contaminated pasture
- consumption of fish from contaminated surface waters;
- consumption of cereals, potatoes and vegetables, contaminated through irrigation or amendments to the soil;
- inhalation of contaminated aerosol (also for the restoration workers);
- external irradiation on contaminated fields or banks of surface waters, or in contaminated water or air (also for the restoration workers).

Concentrations of radionuclides in vegetative foodstuffs are derived from the concentrations in soil through bioaccumulation factors to the edible part of the plant. Interception and translocation factors

are applied for the direct transfer of radionuclides from irrigation water to the plants, taking into account also the effect of weathering.
Concentrations of radionuclides in animal food products are derived from the intake of radionuclides by the animals. The intake occurs through contaminated feed or pasture (contaminated in the same way as vegetative foodstuffs), contaminated water and through contaminated soil, taken in by the animals while grazing.
Fish may become contaminated through their foodchain or by the uptake of elements by respiration through the gills. In the assessments, bioaccumulation factors from water to fish are applied, which are based on observations and thus implicitly take into account all pathways.

The doses to be calculated consist of individual and collective, effective, committed doses.
Individual doses are to be assessed for an average member of the critical group. Such a group should, by definition, consist of a rather homogeneous, real or fictive group of individuals, likely to obtain the highest exposure due to their location or living or working habits. Critical groups are supposed to present normal habits regarding food and other factors, but to obtain their high exposure from the contamination levels in their environment or food items. A - hypothetical - critical group may consist of a fictive group of individuals in the cases of exposure in the future, assuming that the living or dietary habits of the people have not changed from now and that the critical group is situated at a "bad" location.
In this study critical groups are assumed to consist of 'self-sustaining' farmers or fishermen living in the contaminated area. Both are supposed to be exposed through ingestion of contaminated agricultural products, but the former are also supposed to be exposed through external irradiation and inhalation on the contaminated fields, while the latter are supposed to be exposed through ingestion of contaminated fish and external irradiation while fishing.

The collective doses are the sum of the individual doses normally truncated in time and space. They are a measure of the total radiological health detriment to which they are supposed to be linearly related. A common way to perform calculations of the collective dose to a large, inhomogeneous group is to estimate the mean dose to the actual population. This can be done when statistical data for consumption are available. Another way is to use production data of food crops and fish capture and other similar data.
From international recommendations, time periods of 100 and 500 years have been selected, over which to carry out the dose calculations. Within this time period the climate is supposed not to have changed to an important extent and the - hypothetical - critical group may be assumed to behave in a similar way as today; this means with the same living and dietary habits.

3.4.2 Assessments of the example sites

In this project compartmental models have been developed using the BIOPATH-code, or actually the subprogram ACTIVI contained in the BIOPATH-package, for solving the differential equations and the PRISM-code for addressing uncertainties

BIOPATH is a general tool which can be used for varying types of compartment models, as long as they are based upon first-order differential equations. PRISM is another general tool for addressing the uncertainties in any model due to the uncertainty or variability in parameter values.
In PRISM, sets of random parameter values are generated from given distributions by using a systematic sampling method, Latin Hyper Cube.The Latin Hyper Cube method is an efficient Monte Carlo sampling technique which samples values at random, one from each interval with equal probability of occurrence within the whole value range.
Each set of parameters is used in its turn in the model to yield an output (response). The joint set of model parameters and model responses are statistically evaluated and correlations calculated (the Pearson, and Spearman Rank correlation coefficients). From this type of analysis (sensitivity analysis) the relative contribution to the total uncertainty from each parameter can be obtained and processes contributing to the uncertainty in results identified.

The BIOPATH/PRISM-codes were selected in the RESTRAT project because they could be adapted readily to various compartment systems and because they have been verified and validated in a lot of international studies (e.g. BIOMVS[1] I, BIOMVS II, VAMP[2], BIOMASS[3]). In these studies they proved to be very reliable and valuable. In this project it was also shown to be possible to couple BIOPATH/PRISM with chemical speciation codes, such as EQ3NR and MINTEQA2, another reason for using these codes.

The compartment system and collective doses to the public and to the restoration workers are given in Figure 9 to Figure 18 for each site and restoration option, respectively (see Technical Deliverable 6). Option A is without any restoration, the different options are described in chapter 3.

[1] Biosphere Model Validation Study

[2] IAEA Co-ordinated Research Programme: Validation of Environmental Model Predictions

[3] IAEA Co-ordinated Research Programme: Biosphere Modelling and Assessment Methods

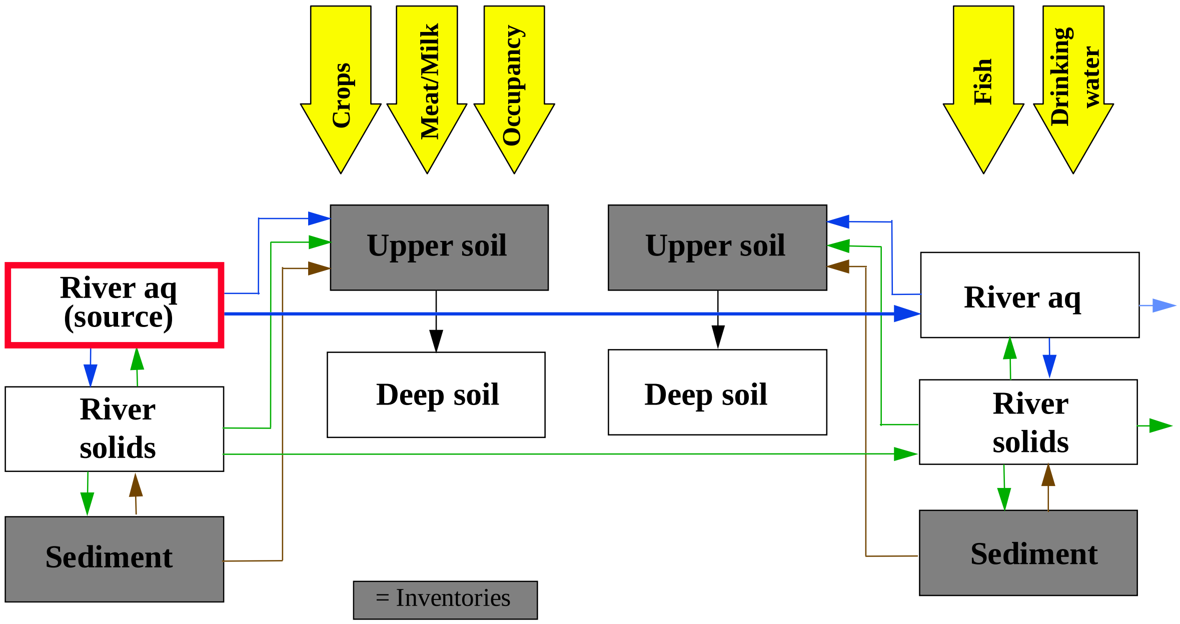


Figure 9 : *Compartment structure for the Molse Nete River and considered exposure pathways.*

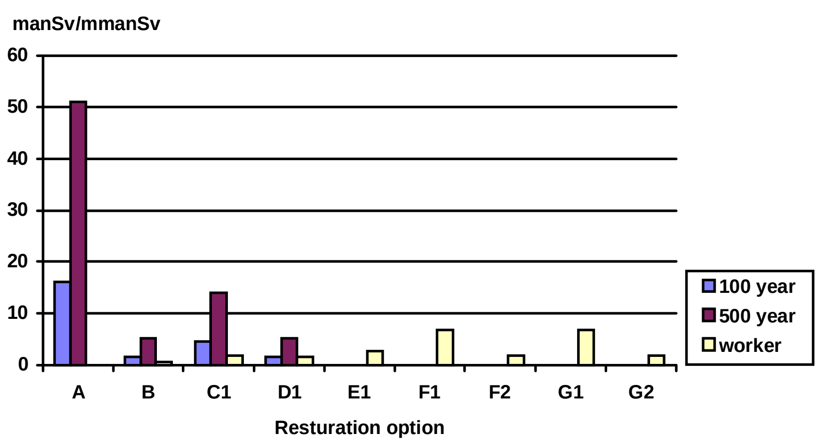


Figure 10 : *Collective doses to public at Moles site truncated at 100 and 500 year (manSv) and workers for restorations (mmanSv)*

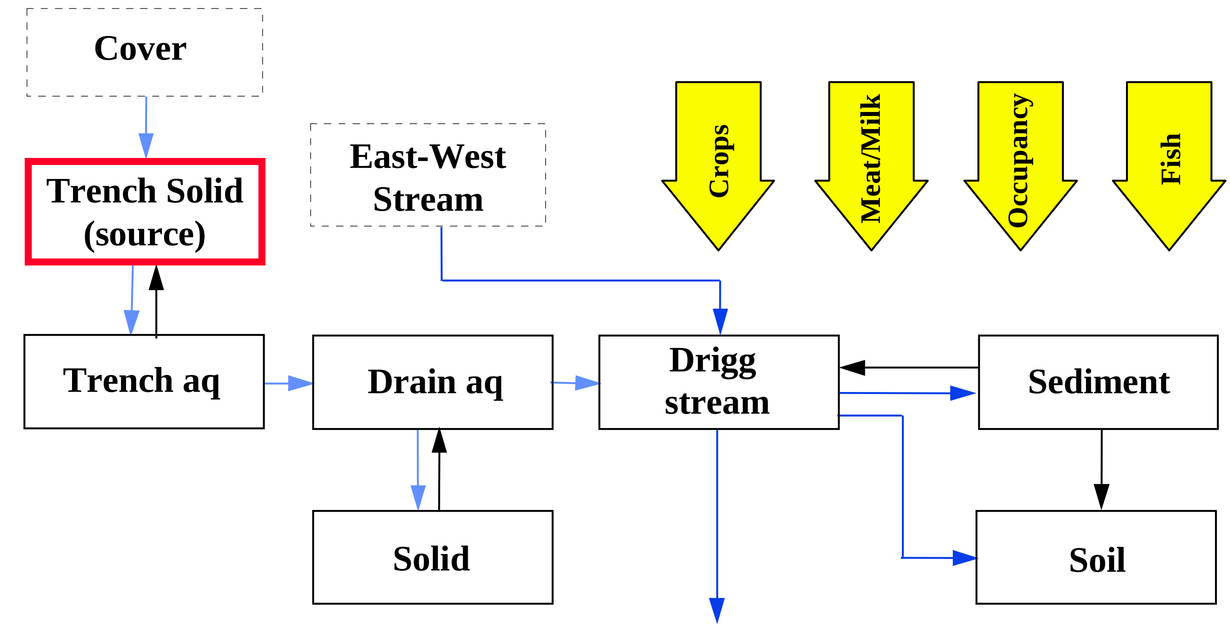


Figure 11 : *Compartment scheme for the Drigg site.*

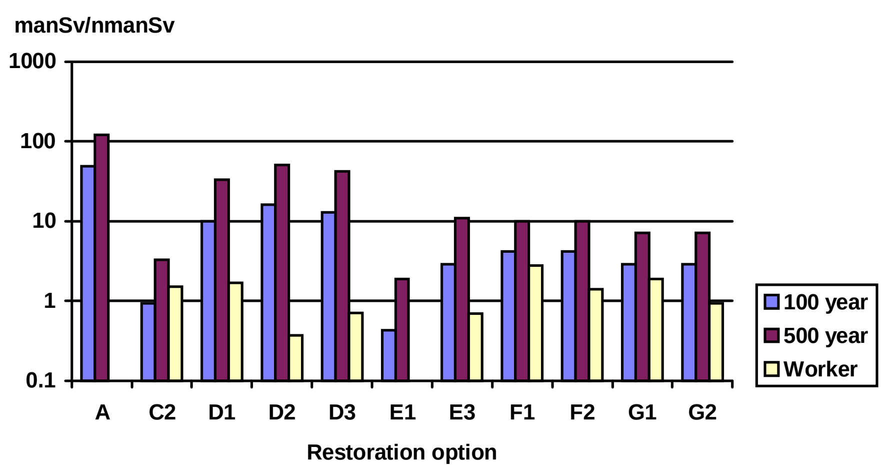


Figure 12 : *Collective doses to public at Drigg site truncated at 100 and 500 year (manSv) and workers for restorations (nmanSv)*

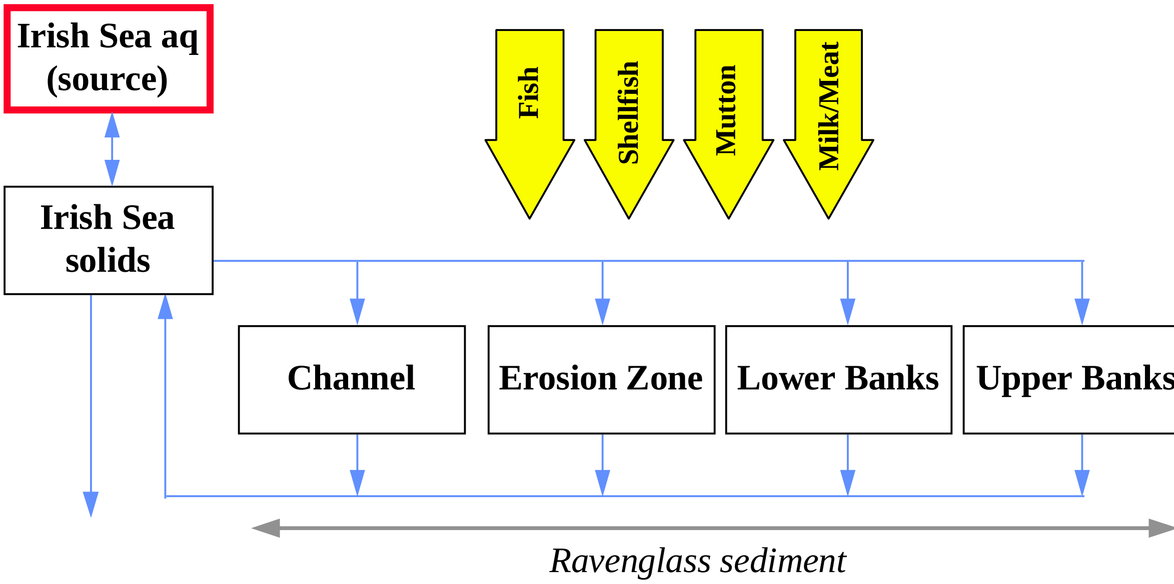


Figure 13 : *Compartment scheme for the Ravenglass estuary.*

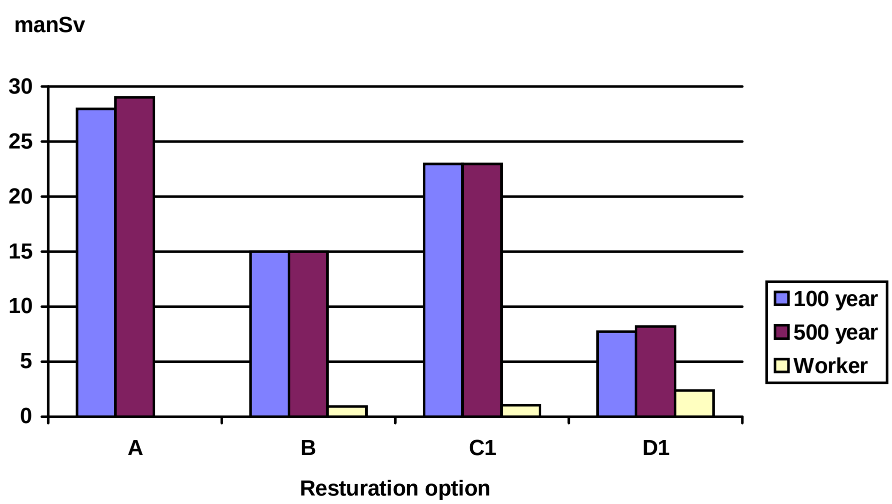


Figure 14 : *Collective doses to public at Ravenglass site truncated at 100 and 500 year (manSv) and workers for restorations (manSv)*

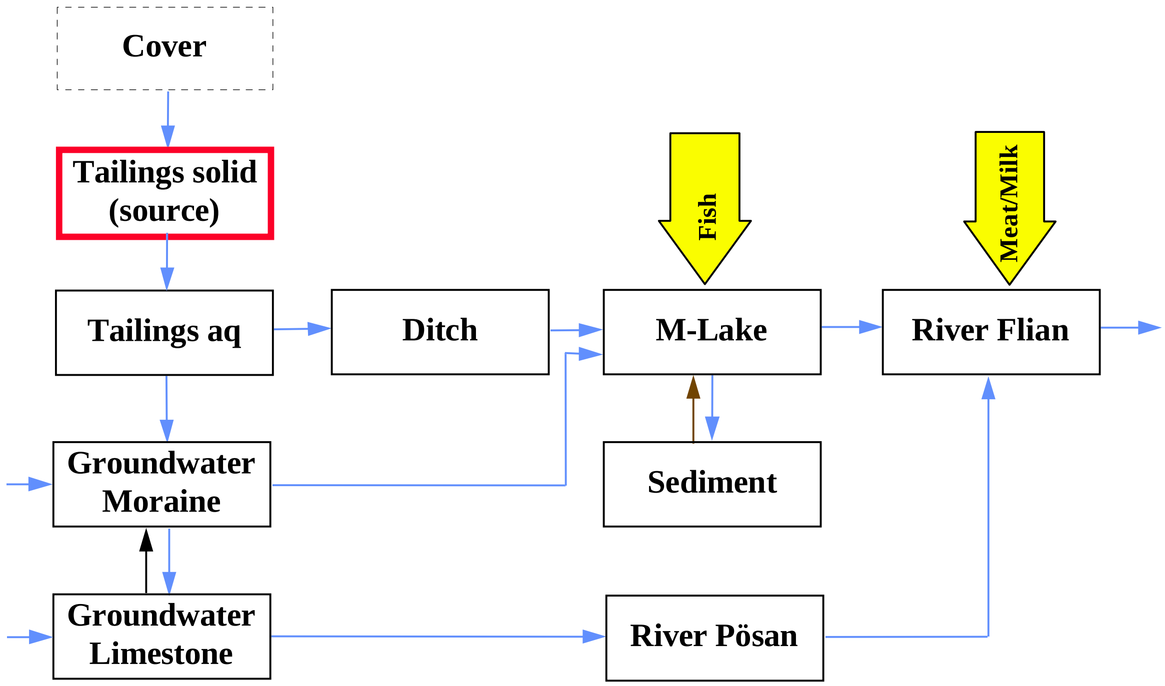


Figure 15 : *Compartment scheme for the Ranstad tailing site.*

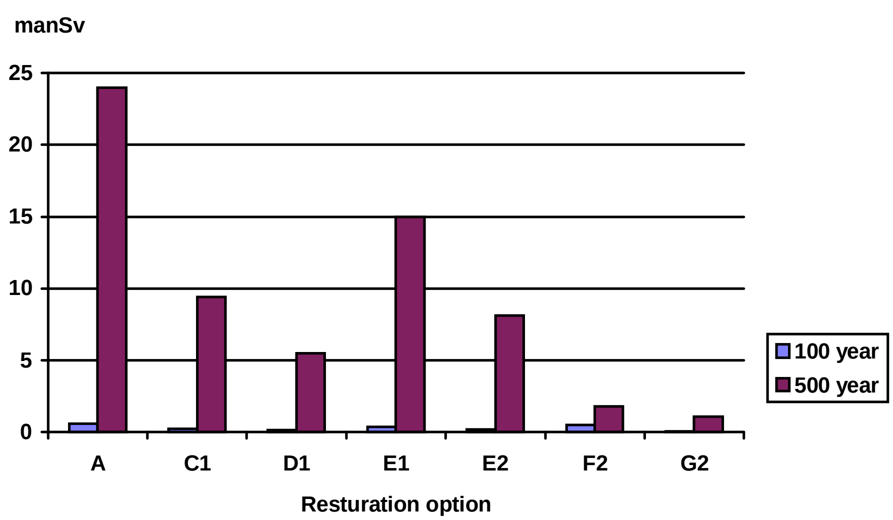


Figure 16 : *Collective doses to public at Ranstad tailing site truncated at 100 and 500 year (manSv), (Workers are assumed to not receive any doses of significance.)*

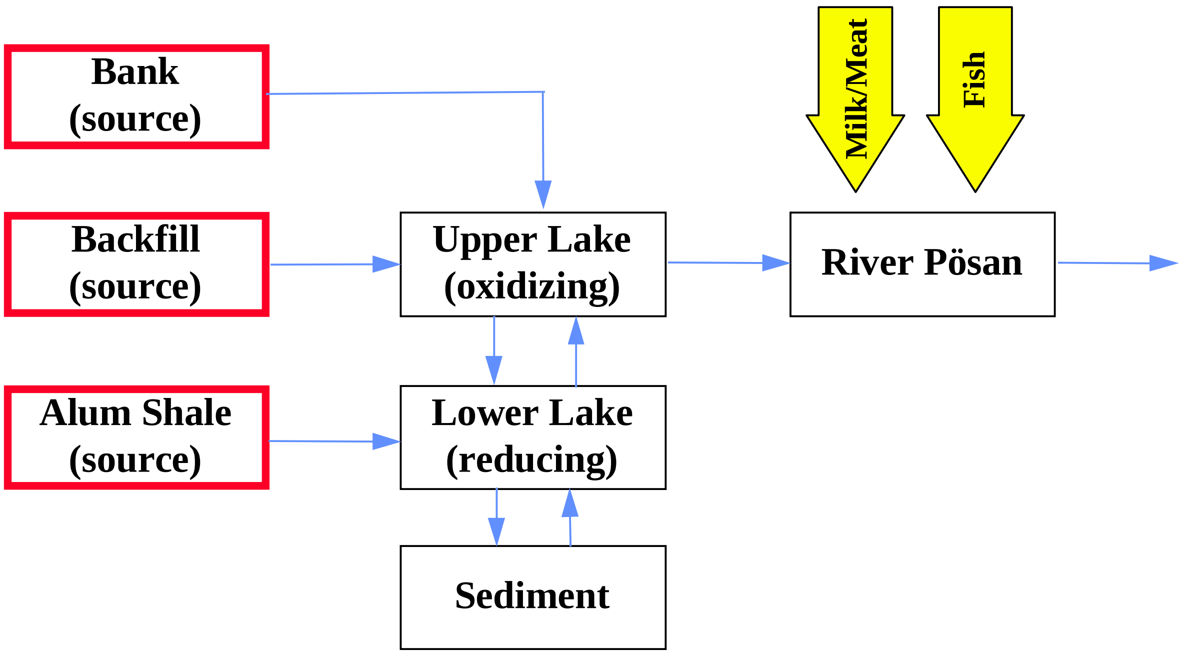


Figure 17 : *Compartment scheme for the lake Tranebärssjön.*

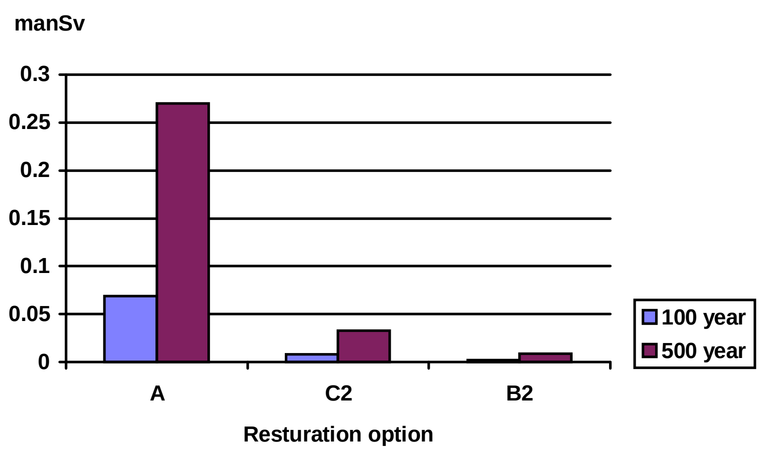


Figure 18 : *Collective doses to public at Lake Tranebärssjön truncated at 100 and 500 year (manSv), (Workers are assumed to not receive any doses of significance.)*

3.4.3 Dose assessments with site-specific K_d values

Additional dose assessments were performed with the same model, but with K_d values that were calculated with the chemical speciation model developed in WP2. As example sites, the Molse Nete River, Drigg and Ranstad tailing site were considered. The collective doses to the public, truncated at 100 year and without any restoration measure implemented, were calculated with the new site-specific K_d values (Table 19) and compared with the results obtained above with generic K_d values originating from the literature (Figure 19). In this figure the ranges of the dose values are given for Co-60, U-238, Pu-239 and Am-241, between the 5^{th} and the 95^{th} percentile.

Table 19 : *Distribution coefficients (Kd) (m3/kg) from literature and from site-specific calculations.*

Site and nuclide	Literature data			Site-specific data		
	Mean	Low	High	Mean	Log Mean	Std of log
River Molse						
Co-60	20	5	100	0.41	-0.38	0.18
Pu-239	250	100	1000	17	1.23	0.12
Am-241	1000	100	2000	310	2.49	0.09
Drigg						
U-238, drain	0.1	0.01	1	3.63	0.56	0.33
U-238, stream	10	1	100	17	1.23	0.48
Pu-239, drain	2	0.01	100	87	1.94	0.12
Pu-239, stream	100	1	600	240	2.38	0.19
Am-241, drain	6	0.001	50	26	1.42	0.30
Am-241, stream	100	1	600	32	1.49	0.26
Ranstad tailing, U-238						
Tailing layer	0.015*	0.002	0.1	0.034	-1.47	0.35
Moraine layer	0.015*	0.002	0.1	0.29	-0.54	0.26
Limestone layer	0.015*	0.002	0.1	0.0023	-2.63	0.31
Storage pond	2	0.2	20	59	1.77	0.19

* one single parameter was used

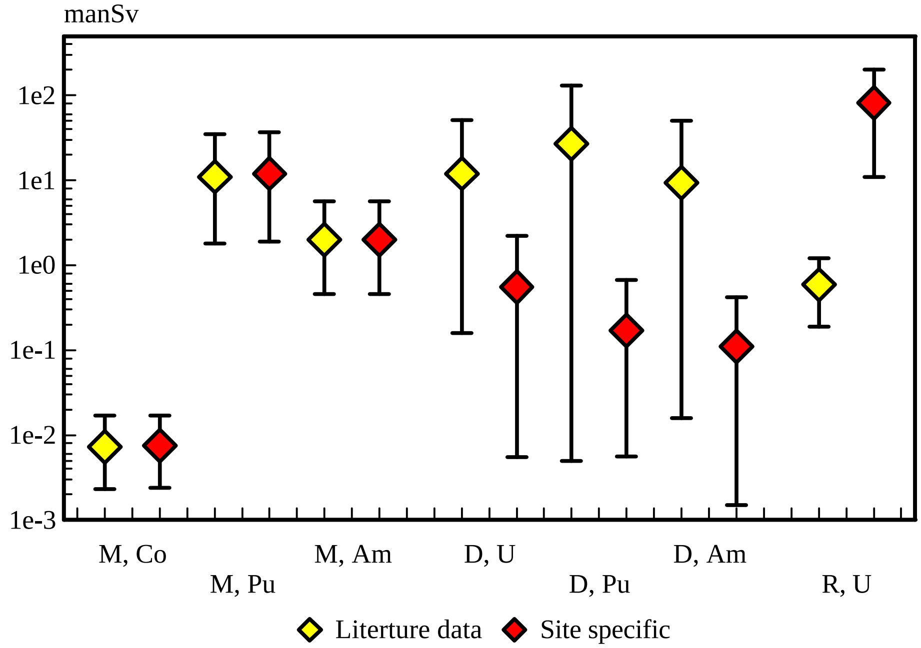


Figure 19 : *Collective dose (manSv) to the public at Molse Nete river (M), Drigg (D) and Ranstad tailing (R)*

For the Molse Nete river, no significant difference has been observed due to the fact that only site-specific K_d values were available for the river water and not for the soil, which is the main source for human exposure (through root uptake of various crops) at that site.

For the Drigg site, the dose values calculated with the site-specific K_d values are considerably lower than the dose values calculated with the literature K_d values. This was due to the fact that the site-specific K_d values are higher than the ones derived from the literature and that the last ones also show a wide range, down to very low values (associated with high dose values).

For the Ranstad tailing site the dose values calculated with the site-specific K_d values are considerably higher than the dose values calculated with the literature K_d values. This is due to the low site-specific K_d values in the limestone aquifer with respect to the ones derived from the literature, leading to higher radionuclide concentrations in this aquifer. Since one of the major exposure pathways is considered to be consumption of water taken from the limestone aquifer, the resulting dose to man is higher for the site-specific conditions.

3.5 WP5 : Selection of Restoration Options.

A review has been made of international guidance and criteria for clean-up and restoration of contaminated areas. Individual doses to the populations being exposed from selected European example sites have been compared to this guidance to examine if clean-up of the sites may be needed. In addition, a methodology based on multi-attribute analysis has been elaborated, including determination of utility functions and weighting factors, to rank identified restoration options. Sensitivity and uncertainty analyses have been used to identify the most important parameters in the ranking process and the uncertainty on the individual scores. The review and the methodology are briefly addressed below.

3.5.1 Overview of international guidance.

The International Commission on Radiological Protection (ICRP) has published a new set of general recommendations in 1990 (ICRP 1990). These recommendations provide a system of radiological protection that distinguishes between two broad categories of situations: *practices* and *interventions*. According to ICRP, `...*the primary aim of radiological protection is to provide an appropriate standard of protection for man without unduly limiting the beneficial practices giving rise to radiation exposure*´ (ICRP 1990, paragraph 100). More specifically, ICRP states that:

> *A system of radiological protection should aim to do more good than harm, should call for protection arrangements to maximise the net benefit, and should aim to limit the inequity that may arise from a conflict of interest between individuals and society as a whole* (paragraph S14)

Clean-up situations can be fitted within the framework of practices and intervention, although this is not always entirely straightforward. A slightly more general approach based on the broader conceptual definitions of practices and intervention provided by ICRP can also be used to simplify the advice. The International Atomic Energy Agency (IAEA) has proposed an approach for developing radiological criteria for clean-up in which the recommendations of the ICRP and of the Basic Safety Standards from six international organisations are taken into account. The proposed IAEA guidance on clean-up is shown in Figure 20.

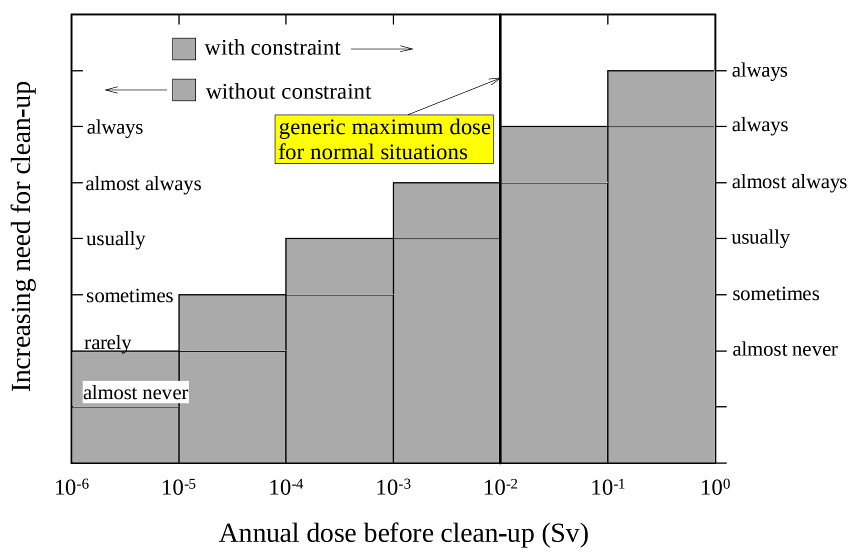


Figure 20 : *Proposed clean-up criteria from IAEA in situations with and without constraints on the clean-up process.*

Draft recommendations from ICRP are similar but with less details for annual doses below the generic maximum dose for normal situations (see Technical Deliverable 1).

3.5.2 Model for optimization of remedial measures

Decisions on restoration of contaminated sites may include other considerations than purely radiological protection considerations. Satisfying the justification principle requires that the overall effect of the actions involved should do more good than harm, taking account of relevant radiological and non-radiological factors. Most decisions require multiple criteria to be taken into account. The field of multiple criteria analysis offers a number of approaches. 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.

One 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*. 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***.

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 linear 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 representing the contribution to the overall evaluation from their performance on each end-attribute (sub-attribute). One way of defining the scores (utilities) is to identify the alternative which does best on a particular attribute and to assign this alternative a score of 100 (or 1) for that attribute. The alternative which does least well should then be assigned a score of 0 for that attribute. 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. For all non-linear utility functions, the knowledge of at least another point or characteristic (in addition to the points 0 and 100 (or 1)) is required to characterise the single utility function, $u(x)$. 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 of the alternative i on attribute j or the utility value of attribute j for the alternative i. 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 are, however, 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, $U_i(x, y, \ldots)$ in which uncertainty distributions can be assigned to the values of each of the attributes, x, y, …. , that defines the utility functions, $u_i(x)$, $u_i(y)$, …. , and to the weighting factors, w, for each of the attributes.

The determination of weighting factors is a 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 has been proposed. The methodology used here is to establish conversion/scaling constants between the weighting factors that can be expressed as:

$$\frac{w_{major,1}}{w_{major,n}} = C_1 \,,\ \frac{w_{major,2}}{w_{major,n}} = C_2 \,, \ldots\ldots\ldots, \frac{w_{major,n-1}}{w_{major,n}} = C_{n-1} \,,\ C_n = 1 \quad \Rightarrow \quad w_{major,i} = \frac{C_i}{1+\sum_{i=1}^{n} C_i}$$

The weighting factors for the major attributes considered in this study, the *economic*, the *health related* and the *social* attributes, are difficult to determine as they are 'valuated' in different units. The ratio between the weighting factors of the economic and health related attributes may be derived easily through the monetary value of the man Sievert. The ratio between the weighting factors for the social and health attributes for instance is much more difficult to derive. If it can be assumed that reassurance is the dominating social factor (because of its more or less permanent nature) and that it could be represented by a risk of psychological harm, with a unit of Sv^{-1}, then this ratio is intuitively expected to be less than one. It has been considered to be significantly less than one for non-accidental situations like remediation of contaminated sites with small exposures.

For sub-attributes, that can be valuated in the same units, such as economic and health related ones, the scaling constants between the weighting factors can be expressed in a simpler way, through the ranges of the values of the attributes, R:

$$\frac{w_{sub,1}}{R_{sub,1}} = \frac{w_{sub,2}}{R_{sub,2}} = \ldots\ldots\ldots\ldots\ldots\ldots = \frac{w_{sub,n}}{R_{sub.n}} = C$$

The scaling constants for the social sub-attributes have been determined in a more general way as described above. Reassurance is given the highest weight because of its permanent nature, and disturbance is given the lowest weight due to its transitional nature.

Model calculations would form the basis for determining whether to carry out remedial actions and to optimise such actions, subject to any constraints, for protection of individuals that otherwise would be exposed. The attribute hierarchy to be used for selection of an optimum restoration strategy can be structured as shown in Figure 21.

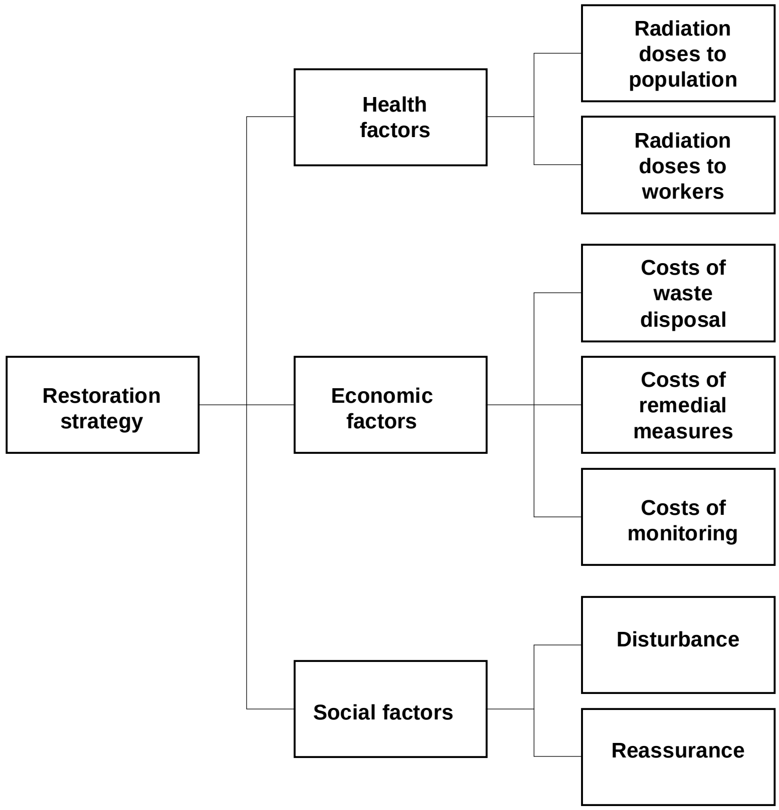


Figure 21 : *Attribute hierarchy for restoration of a contaminated site.*

3.5.3 Ranking of restoration options at the example sites

The multi-attribute utility analysis has been used to evaluate and rank potentially relevant remediation strategies for the five example sites considered. It should be emphasized that some attributes were not evaluated in detail at all the example sites.

Especially some of the economic attributes have been difficult to determine, other economic attributes were determined through generic values of unit costs in the literature.

Social factors posited another kind of difficulties. In order to overcome the problem of the valuation of these attributes, reassurance has in this study been linked to both the residual dose and the fraction of activity remaining on the site after the remedial measure has been implemented and disturbance has been linked to the volume of waste to be transported to the waste disposal site. However, 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 100 for the option with the minimum value of collective dose and minimum value of remaining activity fraction on-site and 0 for the option with the maximum value of collective dose and maximum value of remaining activity fraction on-site. The utility value for disturbance has been taken to be 100 for the minimum amount of waste to be transported from the site and 0 for the maximum amount of waste to be transported from the site.

It has to be noticed that the dose estimates are on the conservative side. The overall picture is expected to remain robust with more realistics economic attributes since the potential dose savings by the suggested remedial measures are rather moderate.

However, with respect to the health factors, not only health risks from radionuclides should be taken into account but also health risks from non-radioactive toxic chemicals (such as present in the Ranstad tailing area).

For radionuclides a collective loss of life expectancy (stochastic health effects) can be calculated from a given collective dose, S_{rad}, an average lifetime risk, r_{rad}, and the average loss of life expectancy per cancer, l (15 years):

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

For exposure to chemical contaminants, non-radiological health effects, can be described in the same way as the exposure to radiation as far as stochastic (non-threshold) effects are concerned. The collective loss of life expectancy from a given collective exposure (man·mg·d^{-1}) of a single non-radiological carcinogen, $S_{chem,i}$, can therefore be calculated as:

$$L_{chem,i} = S_{chem,i} \cdot R_{chem,i} \cdot l$$

where $R_{chem,i}$ is the average lifetime risk per unit exposure rate (cancer·mg^{-1}·d).

Some assumptions are needed in order to assess the impact of a combined exposure of ionising radiation and othertoxic agents like heavy metals. Two of the more important assumptions are:

- 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 $E_1 + E_2 + E_3 + \ldots\ldots$ can be described by the sum of risks as $r(E_1 + E_2 + E_3 + \ldots\ldots) = r(E_1) + r(E_2) + r(E_3) + \ldots\ldots = k_1{\cdot}E_1 + k_2{\cdot}E_2 + k_3{\cdot}E_3 + \ldots\ldots$

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

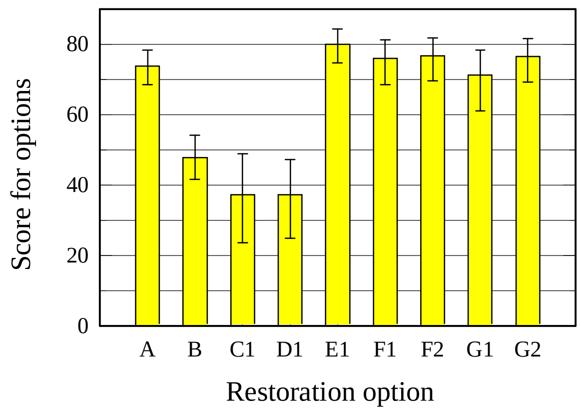
$$L_{total} = L_{rad} + L_{chem,1} + L_{chem,2} + L_{chem,3} + \ldots\ldots = L_{rad} + \sum_i L_{chem,i}$$

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

The detailed results of the ranking of the restoration options, performed at each example site, are given in Technical Deliverable 8. Results for Molse Nete River are presented here in Figure 3. The left-hand picture shows the scores with uncertainty bands corresponding to the 5% and 95% percentiles of the calculated distributions. The right hand picture shows the scores based on central estimates of utilities and weighting factors. The weight of the social, health and economic attributes are also indicated in the right-hand picture.

The top-five most important parameters are the weighting factors for the health attribute, the economic costs, the monitoring cost, the waste disposal cost and the remediation cost.

It appears from Figure 22 that the option E1 (capping), capping, has the highest score, and this option can therefore be considered as the optimum. It also appears that the economic attribute is dominating the overall scores compared to the health and social attributes.



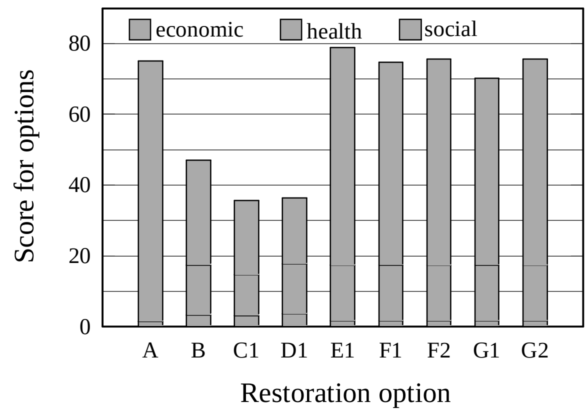


Figure 22 : *Overall evaluation of scores for different remediation strategies for the Molse Nete River site for an integration time of 500 years for the collective dose.*

The ranking of various remediation options at the five example sites, as carried out with multi-attribute utility analysis, is summarised in Table 20 and Table 21. 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 economic 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.

However, it has to be borne in mind that the outcome is dependent on the weighting factors and utility functions applied by the users of the model.

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. 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.

Table 20 : *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	Ranstad	Lake Tranebärssjön
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 *in-situ* (F2)	No remediation (A)	Soil washing (C1)	Capping, 1.6 m (E2)	Biosorption (D3)
4	Chemical immobilisation *in-situ* (G2)	Physical immobilisation *ex-situ* (F1)	Chemical solubilisation (D1)	Physical immobilisation (F2)	
5	Physical immobilisation *ex-situ* (F1)	Chemical immobilisation *in-situ* (G2)		Chemical immobilisation (G2)	
6	Chemical immobilisation *ex-situ* (G1)	Chemical immobilisation *ex-situ* (G1)		Chemical separation (D1)	
7	Soil/sediment removal (B)	Physical immobilisation *in-situ* (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 21 : *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	Ranstad	Lake Tranebärssjön
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 *ex-situ* (F1)	Chemical solubilisation (D1)	Physical immobilisation (F2)	
5	No remediation (discharge stop[a]) (A)	Chemical immobilisation *in-situ* (G2)		Chemical immobilisation (G2)	
6	Chemical immobilisation ex-situ (G1)	Chemical immobilisation *ex-situ* (G1)		Chemical separation (D1)	
7	Soil/sediment removal (B)	Physical immobilisation *in-situ* (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)

3.6 WP6 : Manual

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

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. 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. The attributes that have been taken into account include:

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

The methodologies for assessing the attributes are then explained. They have been elaborated in the other working packages.

The results of the application of the methodologies to the example sites are shown and briefly commented.

Two CD-Roms are added to this manual.

The former 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 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.

4. MAIN ACHIEVEMENTS AND DISCUSSION.

4.1 WP1 : Case Studies

Achievements

Example sites/cases have been selected as representatives for major classes of contaminated sites in Europe. They include :

- the BNFL Drigg disposal site for solid waste;
- the Ranstad tailing site with relies of mining and milling;
- the Molse Nete river, a contaminated freshwater river;
- the Ravenglass estuary;
- the Tranebärssjön lake (at Ranstad), a contaminated freshwater lake.

The information gathered at these sites covered characteristics concerning :

- the geography and the lay-out;
- the geology and the hydrology;
- the meteorology;
- the anthropological activities;
- physico-chemical properties;
- radiological impact.

Discussion

The characteristics of the sites examined were needed for the quantification of the site-dependent attributes (radiological doses, economic costs, social factors) in the ranking procedure of the restoration options. They were in general well-documented and easy to extract from internal documents and open literature except for the physico-chemical parameters.

The physico-chemical parameters that had to be determined/measured, were identified in WP2 and included pH, temperature, ionic composition of the aqueous phases, redox states, organics, mineralogical composition of the solid phases. Data collected from the open literature and from internal documents were not always consistent and showed important information to be lacking. This was especially true for the ionic composition of the aqueous phases and the mineralogical composition of the solid phases. As a consequence our own comprehensive analyses were performed (mainly at FZ Rossendorf and partly at Westlakes S.C.) on water and soil and sediment samples taken from each example site.
A proper sampling approach was established and distributed to the partners in order to ensure representative data sets with a high quality. The water samples were analyzed for all the major anions and cations, pH, Eh and chemical-toxical (and sometimes radioactive) contaminants. Soil and sediment samples were analyzed in particular with respect to the mineralogical composition. Information about this composition has also been derived from site descriptions and pedological an geological maps. Geochemical speciation models were used in WP2 to check the analytical data for consistency and accuracy.

Based on the characteristics of the sites, potentially relevant restoration techniques have been selected at each site.

Technical Deliverables

The site descriptions have been the subject of the following technical deliverables. In these reports also the results of the impact assessments and of the optimisation analyses of the restoration options are given.

Bousher A. Drigg Site: Basic characteristics and evaluation of restoration options. RESTRAT - TD9. 980132/02; Westlakes Scientific Consulting, Cumbria, UK; 1999.

Stiglund Y. and Aquilonius K. Ranstad tailing site: Basic Characteristics and Evaluation of Restoration Options. RESTRAT - TD 10. STUDSVIK/ES-99/21; Studsvik Eco & Safety AB, Nyköping, Sweden; 1999.

Sweeck L. and Zeevaert Th. Molse Nete River site: Basic Characteristics and Evaluation of Restoration Options. RESTRAT - TD 11. BLG-811; SCK.CEN, Mol, Belgium; 1999.

Bousher A. Ravenglass Estuary: Basic characteristics and evaluation of restoration options. RESTRAT - TD 12. 980132/03; Westlakes Scientific Consulting, Cumbria, UK; 1999.

Stiglund Y. and Aquilonius K. Lake Tranebärssjön: Basic Characteristics and Evaluation of Restoration Options. RESTRAT - TD 13. STUDSVIK/ES-99/22; Studsvik Eco & Safety AB, Nyköping, Sweden; 1999.

4.2 WP2 : Physico-chemical Phenomena.

Achievements

Important physico-chemical phenomena influencing the source term evolution and migration of radionuclides in the environment, have been identified from a literature review. They include among others radioactive decay, complexation reactions, oxidation state changes surfaces, physical and chemical sorption onto mineral surfaces, precipitation and dissolution of solid phases. The physico-chemical parameters describing quantitatively these processes have been determined. System-specific parameters were determined from field and laboratory data (see also WP1). The parameters defining its spatial and temporal evolution are not in the scope of this WP. The most important reaction-specific parameters are the thermodynamic ones. They are independent on the site and can therefore be derived from thermodynamic data bases.

The use of single K_d values in impact assessment models gives rise to large uncertainties in the results. In order to solve this problem, the K_d concept is decomposed into the main processes defining it, unfolding a single K_d value into a vector. This introduces many new parameters, but all of them can be determined more easily and more precisely.

An overview of geochemical speciation modelling software was elaborated in order to find an appropriate model that can allow for an unfolding of the K_d and that can be integrated into the risk assessment code. Two software packages have been selected : the EQ3/6 program and MINTEQA2, that cover the whole range of chemical reactions in homogeneous aqueous solutions. The main differences between them are that only EQ3/6 is capable of dealing with kinetic rate laws, and only MINTEQA2 has surface complexion models incorporated.

Thermodynamic databases have been identified from the literature and own databases have been set up for using the sorption models in a geochemical speciation computation.

For both codes ECO3/6 and MINTEQA2, software has been created to organize the data input and transfer to the PRISM/BIOPATH package and to incorporate the speciation models into the risk assessment software. The way of incorporation has been held flexible in such a way that substituting the present speciation modules by another program would need only comparatively small efforts.

For all five example sites, distribution coefficients have been calculated for a number of compartments and for the major radionuclides, using the MINTEQA2 software. Uncertainty and sensitivity analyses have been included.

Discussion

The first results for the example sites prove the applicability of the concept of substitution of empirical K_d values by a theory-based sorption and speciation model. The software implementation is now tested and documented. From the currently available results, several general conclusions can be drawn:

- in many cases the solid concentration C_{solid} is a major impact factor for the distribution coefficient modelling, in case or porous aquifers mainly reflecting the uncertainty of the rock porosity and lacking knowledge about that portion of rock which is not accessible to sorption processes inside the various layers;
- the thermodynamic database situation is far from being satisfactory, especially for redox reactions;
- the sorption data for actinides at lower oxidation states is very sparse and of questionable quality;

- the collection of analytical data for the solid phases, i.e. mineralogical characterization of rocks, sediments, and suspended solids, deserves more attention.

Several recommendations can be made.

In order to better account for the mineralogical inhomogeneity of natural solids (rocks and minerals) the sorption model should be expanded to several instead of just one mineral surface per compartment. To further speed up computation times it seems worthwhile to change the present external call of the speciation code to a fully incorporated subroutine inside the risk assessment model.

As it turned out from at least two of the five examples sites, and as it can be demonstrated in many other cases, many real-world contaminations are in fact a mixture of radionuclides with chemical-toxic contaminants, such as heavy metals, arsenic or organic compounds. Thus such mixed contaminations need to be modelled, too.

The approach towards a better incorporation of physico-chemical phenomena governing the source term at contaminated sites must be further elaborated by inclusion of other aspects that may be important at certain sites. Here, the addition of interactions between contaminants, solids and humics to the integrated model is the most obvious case. Another topic to be included in future versions of the integrated risk assessment would be the co-precipitation of contaminants with “bulk” mineral phase precipitation.

Theoretical considerations, models and appropriate software are only one side of the metal, the other side is a high-quality database. In case of site-specific parameters this mainly aims at improvements in sampling and treatment methods before and during analytical procedures. Recommendations with regard to such methods are given in the RESTRAT TD2, but this must be handled by the investigators of a certain site specifically. However, when it comes to reaction-specific data, the urgently required improvements in quantity and quality of appropriate thermodynamic and kinetic databases, a coordinated European action would give greatest benefits.

Technical Deliverables

Following technical deliverables have been produced within this work package.

Brendler V. Physico-Chemical Phenomena governing the Behaviour of Radioactive Substances. State-of-the-Art Description. RESTRAT - TD 2. Forsschungszentrum Rossendorf, Dresden, Germany; 1999.

Brendler V. Physico-Chemical Phenomena governing the Behaviour of Radioactive Substances. Site-specific Characteristics. RESTRAT - TD 5. Forsschungszentrum Rossendorf, Dresden, Germany; 1999.

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

4.3 WP3 : Restoration Techniques

Achievements

A literature review has been carried out in order to identify and characterise restoration techniques which are applicable to sites contaminated by radionuclides. These were compiled as a MS-ACCESS database.

Techniques which have reached a sufficient level of development for treating radionuclide contamination were classified into four broad categories:

- Removal of sources : bulk removal of contaminated medium
- Separation of contaminated fractions
- Containment : providing barriers between contaminated and uncontaminated media
- Immobilisation : adding material to the contaminated medium in order to bind the contaminants

Each technique was then evaluated in terms of characteristics relevant to the other Working Packages of the RESTRAT project, dealing with risk assessment and ranking of restoration options. Relevant characteristics include the applicability of the technique, economical costs, performance (efficiency, reduction in waste volume, service life), side effects (in particular waste arising and remobilization of other pollutants).

Each characteristic was quantified in terms of a range of values reflecting the uncertainties associated with these values.

Specific values for the characteristics of each technique, which was identified as potentially appropriate to an example site, were selected from the ranges of values identified above. The criteria for selecting these values took account of site-specific factors, such as nature of the waste, nature of the site, distribution of radionuclides throughout the site, ease of access for the remediation technique and the location of a suitable disposal site.

The site-specific cost and performance data were used to calculate both the volume of waste generated and the monetary costs for remediating each example site. This data was subsequently used as attributes in the ranking of restoration techniques for each example site.

Discussion

The aim of this work package has been to identify and characterise restoration techniques, for radionuclide contamination, in terms of a database which can subsequently be used by other working packages within the RESTRAT project. This has been successfully achieved.

However, an examination of the relatively large ranges of values obtained for each parameter within the database reflects the fact that the data has been obtained from a variety of sources. These sources describe the application of restoration techniques to a variety of wastes containing a number of radionuclides, in different environments. In many cases the circumstances in which they have been applied are very poorly defined.

The selection of values for site-specific parameters depend upon factors such as the type of waste, the type of contamination, its distribution and the accessibility of the site for restoration. To date, the choice of values is largely a matter of personal judgement; whilst the uncertainty is taken to be the full range of values assigned to each parameter. This approach needs to be further refined to reduce these ranges of uncertainty and also to provide a framework through which values of site-specific parameters can be determined. This could be achieved through identifying those factors which contribute to the choice of value and a re-examination of the literature to quantify the impact of each factor.

Many sites which are contaminated by radionuclides will also contain non-nuclear contaminants e.g. toxic metals, organics. The application of a restoration technique to such sites is also likely to have an impact upon the non-nuclear contaminants associated with the site. This could be a beneficial (detrimental) effect and contribute to the eventual choice (rejection) of restoration option. Therefore, the impact of restoration techniques, which are relevant to treating sites contaminated by radionuclides, on non-nuclear contaminants requires investigation. This could be achieved through an additional investigation of the literature, and extension of the database, to determine the effectiveness of each restoration option towards other types of contaminants.

Another aspect of applying restoration options to nuclear waste sites which requires further investigation is that of monitoring costs after restoration. To date, monitoring costs have been taken as proportional to the fraction of activity left on site. However, this need not be the case, also the risk presneted by the contaminants left on site should be considered as well as other conditions of the site that may influence the monitoring needs. Therefore, quantification of monitoring costs need further refinement.

Finally, restoration technologies continue to develop and results continue to be published. A number of potentially useful restoration options were discarded as there is currently insufficient data to justify their inclusion in the database. Therefore, it is recommended that the literature should continue to be monitored for data for future applications.

Technical Deliverables

Detailed information is to be found in the technical deliverable TD 3+4.

Zeevaert T. and Bousher A. Restoration techniques: characteristics and performances. RESTRAT - TD 3+4. BLG-816; SCK.CEN, Mol, Belgium or 980132/01; Westlakes S.C., Moor Row, Cumbria, U.K; 1999.

4.4 WP4 : Risk Assessment

Achievements

In this working package a methodology is elaborated for assessing the radiological impact on man from contaminated sites without and with restoration actions carried out.

Following dose values have been assessed:

- maximum annual individual doses to the average member of the critical group (public);
- collective doses (truncated in time and space) to the public;
- collective doses to the restoration workers.

For the restoration workers the doses were calculated very straightforwardly, from working volumes and contamination levels. For the public a more complex system (biosphere model) was developed based on a compartmental approach.

All dose calculations have been performed with uncertainty and sensitivity analyses with respect to variation and uncertainty in parameter values. Uncertainty ranges have been presented for the collective doses for each exposure pathway and nuclide and for each restoration option at the example sites. The most sensitive parameters have also been identified. Especially distribution coefficients (K_d) have been shown to be very important in most cases.

The compartmental models have been elaborated based on the BIOPATH/PRISM codes. The BIOPATH code has been used for solving the differential equations representing the transfers between compartments and the PRISM-code for addressing uncertainty and sensitivity analyses. A chemical speciation codes (MINTEQA2) has also been coupled to BIOPATH/PRISM for using site-specific Kd values.

Following issues have been addressed successfully:

- ➢ determination of the basic compartments in terrestrial and hydrological (aqueous) biospheres, soils, aquifer, water column, sediments;
- ➢ identification of possible transfer processes between compartments or within compartments (between phases);
- ➢ description of potential critical groups in the defined exposure scenarios for the public : essentially self-sustaining farmers and fishermen living on the site;
- ➢ determination of exposure pathways for the public based on agricultural and fishing activities of the critical groups, and for the restoration workers (inhalation and external irradiation);
- ➢ introduction of the effect of the restoration options in the assessment models;
- ➢ determination of the time periods for the collective dose calculations at 100 and 500 y, based on international recommendations.

The methodology used for dose assessments for radionuclides, was also used for risk assessments for heavy metals (Ranstad and Tranebärssjön). However for heavy metals the assessment has been limited to the total intake by humans.

Discussion

The dose assessment is based on models and methods that are generally accepted. Within this study compartmental biosphere models have been elaborated based on the BIOPATH/PRISM software. This has been tested and verified through several international studies, such as BIOMOVS and VAMP. Experiences from these studies have been applied in this study.

Site-specific characteristics may be very important in dose assessments, in a sense that the doses assessed may be heavily influenced by specific features or characteristics of the sites, such as exposure pathways, K_d values etc.. Within the RESTRAT project, as the aim has not been to assess very accurately the radiological impact at the example sites, but only to use the sites for illustration purposes of the ranking methodology for restoration options, a detailed and precise site charaterisation has not been carried out. As a consequence, a rather generic and conservative approach was adopted and the doses assessed at the example sites was not very accurate. For such an approach, compartmental modelling has proved its usefulness.

Moreover, the degree of precision of the impact assessment has also to be in agreement with the level of the dose impact. At the example sites considered, this level was mostly rather low in relation to the high economic costs for restoration (and waste disposal).

Nevertheless the precision of the biosphere modelling at the example sites has been enhanced and the uncertainty reduced by introducing site-specific K_d values, based on chemical speciation of the radionuclides, as indicated in WP2. The importance of using site-specific values of the distribution coefficients has been demonstrated through the decrease of the uncertainty ranges of the doses assessed at the example sites.

A further enhancement of the accuracy, and reduction of the uncertainty, in the impact assessments could be obtained through a more precise characterisation of the sites. This is especially the case with respect to the mineralogical composition of the solid phases where the lack of information about compostion hampers a more precise determination of K_d values. A more precise site characterisation may also enable a further subdivision of the biospheric compartments to be made and more precise dose results to be obtained.

Another area where considerable improvement of the biosphere modelling could be made is the modelling of the migration of the contaminants in soil, both in the unsaturated and saturated zone. The performance of the compartmental model applied is very limited in this respect. It can consider neither different mobilitiy values for different fractions of the contaminants nor inhomogeneities in the transport media. Consequently, it can only give rough estimates for the transport of the contaminants. Substituting the soil compartment by a numerical transport modelling approach based on the physical processes involved and allowing for spatial inhomogeneities, could enable more robuste and defensible results to be obtained. Then also the influence of some of the restoration techniques could be introduced in a better, more site-specific way, based on the processes involved.

With respect to the presence of non-radioactive contaminants, the health risks of these substances need also to be assessed on a common bases with the health risks of the radionuclides. The impact assessment methodology should be enlarged in ordre to be able to combine the risks from both types of contaminants.

Technical Deliverables

The dose assessment model and results for the example sites have been described in the technical deliverable TD 6.

Stiglund Y. and Nordlinder S. Dose Assessment Model for use in Site Restoration. General Methodology and Site-specific Aspects. RESTRAT – TD 6. STUDSVIK/ES-99/18; Studsvik Eco & Safety AB, Nyköping, Sweden; 1999.

4.5 WP5 : Selection of Restoration Options

Achievements

Three methods or approaches for evaluating restoration options for contaminated sites have been applied to the example cases:

- the IAEA criteria for clean-up of contaminated land;
- a simple cost-benefit analysis based on central estimates of collective doses and economic costs;
- a multi-attribute utility analysis, taking into account health related, economic and social attributes.

The model for the ranking of the restoration options using a multi-attribute utility type of analysis has been built in a spread sheet format. A hierarchy of attributes has been set up and utility functions and weighting factors determined. Uncertainties of the scores of the restoration options and sensitivity to the parameters have been evaluated, using a Latin Hypercube sampling method to generate random numbers within the assigned parameter distributions for the utility values and weighting factors.

Important issues in the optimisation of remedial measures are social factors, risk assessment of combined exposures of ionising radiation and toxic metals and assessment of weighting factors.

Important social factors have been identified to be constituted by disturbance and reassurance. In order to overcome difficulties with the quantification of these factors, reassurance has been linked to the residual dose and the fraction of activity remaining on the site after the remedial measure has been implemented, disturbance has been linked to the volume of waste to be transported to the waste disposal site.

It has also been stated that all social factors and other non-radiological protection factors should enter the optimisation process in parallel with radiological protection factors in order to form an optimised strategy of overall health protection (not only radiological protection). To include socio-psychological factors in the radiation protection framework would give very arbitrary levels of 'radiation protection'.

With respect to the health-related attributes, it has been recognised that health risks from non-radioactive toxic chemicals (if present) should be taken into aacount as well as those from radioactive substances.

Non-threshold (stochastic) health effects of a combined collective exposure to ionising radiation and to toxic chemicals can be described as the sum of the collective losses of life expectancy from all contaminants.

When both non-threshold (stochastic) and threshold (deterministic) health effects involved, however difficulties are encountered. Several possible approaches have been suggested, but a general consensus on a unified approach on the combination of both types of health risks does not yet exist.

The determination of weighting factors of the attributes is a difficult and delicate task. A simple scaling method based on conversion constants between the weighting factors have been proposed.

For attributes that can be expressed in the same units, the conversion constants could be determined in a simple way from the ranges of the attribute values.

For attributes that cannot be expressed in the same units, other considerations were taken into account, for instance permanent or transient nature of some of the social effects.

The results obtained for the five example sites are summarized in the table below.

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	'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'

When considering only central estimates of collective dose and monetary costs, a simple cost-benefit analysis would lead to the conclusion that none of the remedial measures considered for each site are justified.

With respect to the IAEA criteria, some remediation might be needed at all sites, if it is assumed that a dose constraint for controlled practices would be applied to the outcome of the remediation process.

Multi-attribute analyses on ranking different 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 economic attributes compared to the health and social attributes.

Discussion

Different remediation measures have been evaluated for the five example sites. The evaluation has been based upon (a) justification of the measures by trade-off between avertable collective dose and monetary costs, (b) compliance with the recommended clean-up criteria from the IAEA, and (c) ranking of scores for the different remediation measures by use of multi-attribute utility analyses. The applied attributes include monetary costs of the remedial measures, the collective dose to the clean-up workers, the collective dose to population, and the social factors reassurance and disturbance (and loss of income for the Molse Nete River site). 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, and their values were sampled around a most probable value.

Although the multi-attribute method has the advantage of allowing the inclusion of factors that are not easy to quantify in monetary terms, there are difficulties with the determination of weighting factors for the different attributes. Without any terms of reference for the weighting between attributes, value settings by a decision-maker could lead to ‘optimised’ results that might be useless because of a subjective bias of the decision-maker in the selection of weighting factors. Therefore, the outcome of any multi-attribute analysis, including the present study, should be judged very carefully in the light of the values assigned to the weighting factors before any firm conclusions could be drawn.

Except for a few countries, criteria for restoration of contaminated sites are far from being fully developed within European countries and elsewhere. The rationale for deriving remediation criteria in the different countries is not very clear and conceptual differences between existing national criteria do exist. Therefore, it seems that national levels for remediation are not always based on an optimisation of protection of the affected population and further development and guidance is therefore needed to aid decisions on implementation of remedial measures at contaminated sites. In the context of remediation of contaminated sites, it is likely that social costs of disruption for those affected by the remedial measures and continuing long-term anxiety about residual levels of contamination for those continuing to live in the area will be important factors, even the dominating. Important research issues in the context of restoration of contaminated sites are the quantification of social factors, optimisation models and model parameters and risk factors for combined exposures of radiation and toxic chemicals.

Social factors

In analysing the inputs to any decision on remedial measures at a contaminated site, it is necessary to decide on the relative importance or weight of each attribute. The relative weighting to be assigned to the attributes may be very different depending on the type of situation, and it may be difficult to achieve a broad consensus on them, thus making it hard to generalise. Nevertheless, it is important to take these subjective attributes into account; they must be considered in a decision-making process that is wider than the justification process solely based on radiation protection considerations. Notwithstanding this advantage of the multi-attribute method it suffers from the weakness of assigning a value to the weighting factors for the different attributes. Without any terms of reference for the weighting between attributes, value settings by the decision-maker could lead to ‘optimised’ results that might turn out as being highly subjective because of a subjective bias in the selection of weighting factors. An important research theme is to further develop the methodology for assigning values to weighting factors for the different advantageous and disadvantageous attributes.

Optimisation models and model parameters

Several software systems for uncertainty analysis and decision-making between competing options are on the market. Decisions are modelled using hierarchical weighted value functions and the system has an extensive facility for visual interactive sensitivity analysis, which enables the decision-maker to explore the implications of changing or priorities and values. Uncertainties can be assigned to model parameters and correlations made between them. Also the sensitivity of the forecast to the different parameters can be analysed including non-linear utility functions. More general and flexible models for multi-attribute optimisation, in which fast changes of parameter distributions and correlations can be accomplished, are desirable. An interactive presentation of the scores of the different remedial options and the uncertainty band for each option is also needed. Research is needed with the aim of developing more flexible and user friendly models for multi-attribute optimisation. Research is also needed to develop a more structured approach for the selection of probability distributions for attributes and to develop a methodology to describe correlations between the attributes.

Combined exposure to radiation and toxic chemicals and metals

Combined exposure to radiation and chemical carcinogens can be expressed on a common risk scale in order to determine the total expected detriment from the combined exposure. Some assumptions are needed and two of the more important assumptions are (1) the lifetime cancer risk is linearly related to the exposure, also known as the *linearity hypothesis*, and (2) no synergetic effects exist between exposures to radiological and non-radiological carcinogens. The European software ASQRAD is suitable for describing the radiation risk from acute or prolonged exposures based upon demographic data and different risk projection models (BEIR, UNSCEAR etc.). The code was designed to be a flexible, easy-to-use tool with the facility to quantify somatic and hereditary effects, based on a wide selection of health effect models for both individuals and populations. If it were possible to express exposure to heavy metal in terms of an equivalent radiation exposure, the ASQRAD model can be used to predict the risk of combined exposures. When both deterministic and stochastic health effects are involved difficulties are encountered. Several possible approaches have been discussed, but a general consensus on a unified approach on the combination of stochastic and deterministic health risks as well as radiological and non-radiological risks does not yet exist. Research is needed to develop a methodology to describe combined exposures of ionising radiation and heavy metals, both for stochastic and deterministic effects.

Technical Deliverables

Two technical deliverables (TD1 and TD2) have been produced, covering the subject of this work package.

Hedemann Jensen, P. Remediation of Contaminated Areas. An Overview of International Guidance. Risø-R-1122(EN), Risø National Laboratory, May 1999 (RESTRAT - TD 1).

Hedemann Jensen, P. Methodology for ranking restoration options. Risø-R-1121(EN), Risø National Laboratory, April 1999 (RESTRAT – TD 8).

4.6 WP6 : Manual

Achievements and Discussion

In the manual the ranking methodology for restoration options is explained, based on the work performed in the other working packages. Moreover, an extensive overview has been made of classes of radioactively contaminated sites, that are within the scope of this project, categorized according to the origin (practices or events) of their contamination. The results for five example sites, which have been considered to be representative for major categories are also shown.

The manual firstly contains a documentation part on the categorization of contaminated sites and on the characteristics of possibly relevant and well documented restoration techniques.

The actual ranking methodology has been developed in two stages. In a first stage a general description is given of the methodology, based on radiological optimization principles. The methodologies for determining the various relevant attributes have also been explained.

In a second stage, the way how, and specific tools (codes) through which, in this project, the attributes have been quantified, is indicated. The determination of the weighting factors and the conversion of the attribute values into utility values for introduction into the multi-attribute utility analysis have been elucidated.

In the last part of the manual, the results of the application of the ranking methodology to the example sites are shown.

It has to be stressed that the results shown are only of illustrative value. First of all the sites have not been characterised in enough detail to make more accurate assessments of the radiological impact than the rough and conservative evaluations that have been provided for. Secondly, the high economic costs for the restoration operations and waste disposal have been estimated from generic values of unit costs, taken directly from the literature. At last the social factors have been substituted by characteristics of the site, which were considered to govern the attitude and the perception of the public to a large extent.

Consequently the utility values derived in this way are somewhat arbitrary and also weighting factors (for social attributes for instance) may be criticised.

However the ranking methodology presented is considered to be widely applicable. It may be used by authorities, stakeholders and other interested groups of the public, that are involved in the decision-shaping process with respect to restoration. Those groups may then use utility functions and weighting factors for the attributes that correspond to their viewpoints. For instance risk-averse utility functions can be used instead of risk-neutral ones and a higher weighting factor can be adopted for the health related attributes than the one derived from the monetary value of the man.Sievert. Even attributes may be omitted or others may be added. As a consequence every group making evaluations in this sense may come up with a different ranking, according to their insight. The analysis of the differences between the outcomes also constitutes a very important step in the decision-shaping process and is made easy by the common form of the multi-attribute framework.

To conclude with the outcome of the ranking of the restoration options for the example sites, as shown here, is to be considered as only one (obtained by a group of a neutral, technical nature) among a whole set of other ones (obtained by other interested groups).

Technical Deliverables

The manual is issued as a technical deliverable for this project (TD 14):

Zeevaert Th, Hedemann Jensen P, Brendler V, Nordlinder S and Bousher A (1999) Manual on restoration strategies for radioactively contaminated sites. RESTRAT - TD14. BLG-819; SCK.CEN, Mol, Belgium.

In this manual, also two CD-ROMs have been included, containing all the software used in the RESTRAT project.

One CD-ROM comprises the software of the risk assessment model.
The other CD-ROM includes:

- the Crystal Ball software for uncertainty and sensitivity analysis for Excel spreadsheet, with applications to a generic example and to the example sites in RESTRAT;
- the database on restoration techniques, RESTRAT.mdb in Access.

5. PUBLICATIONS.

Apart from the technical deliverables mentioned before, following publications or presentations in relation with the RESTRAT project have been made.

Brendler V. Einbindung geochemischer Speziationsmodule in Risk-Assessment Software.Workshop: Geochemische Modellierung, FZK / INE, Karlsruhe, 22.-24. April 1997

Brendler V, Bernhard G and Nitsche H. Coupling geochemical speciation to risk assessment codes. CIC Meeting of the Society of German Chemists, Zürich, November 1997

Brendler V, Bernhard G, Nitsche H, Stiglund Y and Nordlinder S. Coupling geochemical speciation to risk assessment codes. International Conference and Workshop "Uranium Mining and Hydrogeology II", Freiberg, September 1998

Jackson D, Wragg S, Bousher A, Zeevaert Th, Stiglund Y, Brendler V, Hedemann Jensen P and Nordlinder S. Establishing a method for assessing and ranking restoration strategies for radioactively contaminated sites and their close surroundings. Nuclear Energy, 38(4), 223-231, 1999.

Nitsche H and Brendler V. Radionuclide Migration and Transport in the Vadose Zone: R&D Needs in Measurement and Modelling. Fourth Intern. Symposium and Exhibition on Environmental Contamination in Central and Eastern Europe, Warsaw, September 1998

Wragg S, Jackson D, Bousher A, Zeevaert Th, Stiglund Y, Nordlinder S, Brendler V and Hedemann Jensen P. Remediation strategies for radioactively contaminated sites and their close surroundings (RESTRAT). In proceedings Achievements & Challenges: Advancing Radiation Protection into the 21st Century, SRP Southport 99 International Symposium, 14-18 June 1999.

Further publications and presentations are foreseen:

at least one publication on the results of the project in an international journal with peer review (Health Physics or another one);

at least one presentation in an international conference or symposium (IRPA-10 or another one).