T-6-4, P-5-343

Restoration Strategies for Radioactively Contaminated Sites and their Close Surroundings.

Th. Zeevaert[(1)], A. Bousher[(2)], V. Brendler[(3)], P. Hedemann Jensen[(4)], S. Nordlinder[(5)]
[(1)]SCK•CEN, Boeretang 200, 2400 Mol, Belgium
[(2)]Westlakes Scientific Consulting, Moor Row, Cumbria, CA24 3LN, UK
[(3)]Forschungszentrum Rossendorf, Institut für Radiochemie, 01314 Dresden, Germany
[(4)]Risoe National Laboratory, Dpt. of Nuclear Safety Research, 4000 Roskilde, Denmark
[(5)]Studsvik Eco & Safety AB, 61182 Nyköping, Sweden

INTRODUCTION

A number of nuclear facilities in Europe began operations in the 1950s and 1960s and, consequently, 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, consideration is also required for the restoration of sites where contamination has been dispersed or where radioactive residues are contained by methods which may be unreliable for long-term storage.

Restoration of such sites would seem advisable, both for the sake of the protection of the population and for relieving 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 these sites in both Eastern and Western Europe. The US experience shows that the application of alternative techniques, e.g. *in-situ* restoration is hampered by the lack of transparent risk assessments which consider the exposure of present and future populations and of restoration workers. Also, the lack of relevant data from on-site experiences is another shortcoming for decision-aiding in this domain.

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

An international project partly funded by the EU under the Fourth Framework of the Nuclear Fission Safety Programme, has been addressing this problem. In this project a transparent generic decision-aiding method has been established, capable of evaluating and ranking restoration strategies for radioactively contaminated sites and their close surroundings (RESTRAT). Partners to the project were: SCK•CEN (co-ordinating the work programme), Westlakes Scientific Consulting, Forschungszentrum Rossendorf, Studsvik Ecosafe and Risoe National Laboratory. The study has now been concluded and its findings are presented here. A manual (1) has been produced explaining the method and applying it to typical example cases.

Five typical example sites have been studied: the Drigg low-level waste disposal site, the Ranstad uranium tailing site, the Molse Nete river, the Ravenglass estuary and the Tranebärssjön lake. They were selected as being representatives of major classes of contaminated sites in Europe.

METHOD

The methodology of ranking restoration options has been based on the radiation protection principles of justification and optimisation, as recommended by ICRP 60 (2). The justification principle requires that the restoration (remediation) should do more good than harm, meaning that the net benefit (benefit of the reduction in radiation detriment less the harm and the cost of the restoration) should be positive. The optimisation principle requires that among the justified restoration options, the one with the highest net benefit should be selected for implementation. Dose limits do not apply as the restoration of sites contaminated with residues of past or old work activities may be considered as an intervention (when there is existing exposure of a population from these sites).

Criteria (attributes) that have to be taken into account in order to evaluate and calculate the net benefit include :

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

For performing radiological optimisation analyses, several quantitative decision-aiding techniques are available (3, 4). When factors are difficult to quantify (e.g. social factors), a multi-attribute utility (MAU) type of analysis is the most appropriate technique for dealing with them.

The major attributes are further divided into sub-attributes forming the hierarchical structure shown in Fig. 1 below.

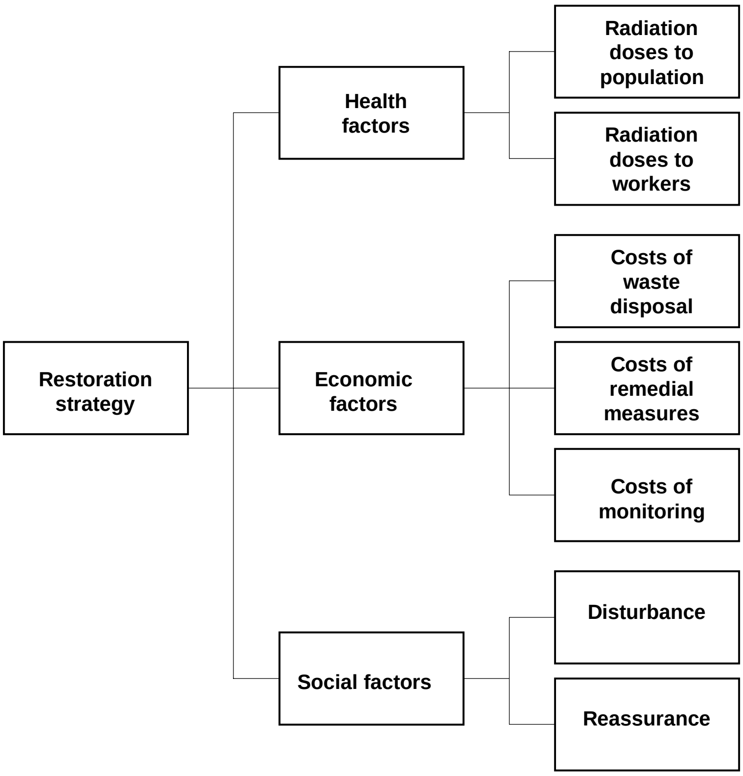


Figure 1 : Attribute hierarchy for restoration of a contaminated site.

The relative weights of the attributes and sub-attributes in each group must then be determined and, for each option, the utility value with respect to each of the attributes.

For the conversion from attribute values to utility values, various types of utility functions may be applied. In this study risk-neutral, linear utility functions are used. These are expressed as :

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

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

The alternative (option) which does best on a particular attribute is assigned a utility value of 100, the alternative which does worst a utility value of 0 for that particular attribute. Other alternatives are assigned intermediate utility values according to the utility function indicated above.

The determination of the weighting factors is a much more difficult task. A simple scaling method is proposed establishing conversion constants between the weighting factors of attributes of the same group (cf. Fig. 1), with the sum of the weighting factors set equal to 1.

For the attributes expressed in the same units these constants can be taken to be equal to the ratios of the ranges of the value of the attributes.

For the radiological health detriment and the economic cost the conversion constant is derived in the same way, adopting a monetary value of the man Sievert, taken here as 100 000 EUR/man Sv according to the Nordic Radiation Protection Authorities (5).

The conversion constant between the weighting factors of the radiological health attribute and the social factors are much more difficult to quantify. Intuitively it has been assumed to be less than 1 and for non-accidental situations like remediation of contaminated sites with small exposures, significantly less than 1. Here a value of 0.25 has been adopted.

For the social sub-attributes other considerations were taken into account (see below : "Social factors").

The results of the MAU analysis will be shown in detail for three example sites investigated:

- the uranium mining and milling site of Ranstad (Sweden);
- the Drigg site for the disposal of low-level radioactive waste (UK) as it was prior to remediation;
- the Molse Nete river which receives low-level liquid radioactive effluents from a waste treatment facility (Belgium).

A comprehensive literature review was carried out in order to identify potentially relevant restoration techniques and to determine the characteristics needed in the ranking methodology.

Four major categories of restoration technologies were distinguished :

- removal of sources : excavation of contaminated soil, sediment
- separation of contaminated fraction from uncontaminated fraction
- containment : providing barriers between contaminated and uncontaminated medium
- immobilisation : adding agents to the contaminated medium to bind the contaminants.

Values of performance characteristics and unit values (per unit volume or area of contaminated medium) of costs and exposure times of restoration workers were extracted from the literature. Uncertainty ranges are indicated in Table 1.

Table 1 : Performance, cost and workforce exposure values of various remediation techniques.

Technology (‡)	**Performance Indicator**		**Cost (EUR/m^3)**		**Workforce exposure (manh/m^3)**
Removal of Source (B)	*Decontamination factor*		*Extraction*	*Disposal & transport*	
Soil excavation	1 - 20		50 - 150	450 - 800	0·2 - 1
Physical separation (C)	*Decontaminat-ion factor*	*Waste reduction*	*Excavation & separation*	*Disposal & transport of residue*	
Soil washing (C1)	1 - 10	50 - 98%	200 - 650	2000 - 3000	0·25 - 1·5
	Decontamination factor		*Separation from liquid*		
Filtration (C2)	2 - >100		0·1 - 3·8	2000 - 3000	0·4 - 1·4
Chemical separation			*Excavation & separation*		
Chemical solubilisation (D1)	1 - 20		180 - 820	2000 - 3000	1·2 - 3·5
			Separation from liquid		
Ion exchange (D2)	3 - 100(U), 20 - 100(Cs)		1·3 - 2·5	2000 - 3000	0·4 - 1·4
Biological Separation					
Biosorption (D3)	2·5 - >100		1 - 3	2000 - 3000	0·4 - 1·4
Containment (E)	*Resultant permeability (m s^{-1})*		*Total (per m^2 surface area)*		*(per m^2 surface area)*
Capping (E1 + E2)	1×10^{-12} - 1×10^{-9}		30 - 45		0·03 - 0·3
Subsurface barrier (E3 + E4)					
a) slurry walls	1×10^{-12} - 1×10^{-8}		510 - 710		0·06 - 0·4
b) grout curtains	1×10^{-12} - 1×10^{-8}		310 - 420		0·06 - 0·4
Immobilisation	*Mobility reduction factor*		*Total*		
Cement-based solidification (F)					
a) *ex-situ* (F1)	5 - 25		75 - 300		0·25 - 1·5
b) *in-situ* (F2)	5 - 25		50 - 310		0·06 - 0·4
Chemical immobilisation (G)					
a) *ex-situ* (G1)	5 - 50		110 - 570		0·25 - 1·5
b) *in-situ* (G2)	5 - 50		60 - 420		0·06 - 0·4

Radiological health detriment

With respect to the radiological health attributes, doses to the public and to the restoration workers were assessed.

Collective doses to the public and to the restoration workers were considered as a measure for the radiological health detriment.

Maximum annual individual doses to the critical group (public) have been assessed with the view to compare them with the recommendations of ICRP (6) and IAEA (7) on clean-up of contaminated land.

Collective doses to the public were truncated at 100 and 500 years.

For the restoration workers, the doses can be calculated straightforwardly from the contamination levels at the site and the exposure times.

For the public a more complicated impact assessment model was needed.

A compartmental type of model was chosen, based on the BIOPATH model developed by Studsvik (8).

An example of a compartmental scheme for a uranium mill tailing (Ranstad) is shown in Fig. 2.

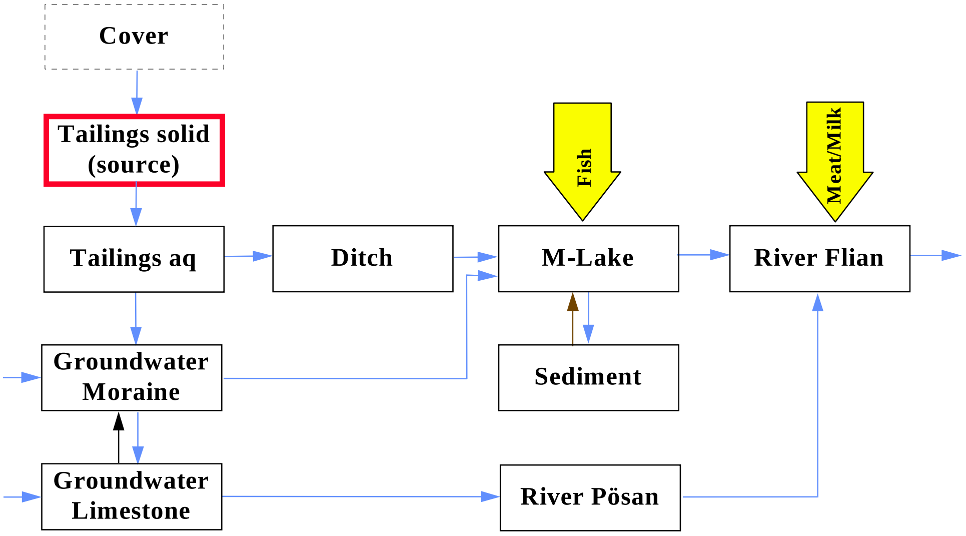


Figure 2 : Compartmental scheme for uranium mill tailings (Ranstad tailing site).

The implementation of the various restoration options considered was modelled by changing the source term (where there is a removal of sources whether or not it was followed by separation) or altering the transfer factors which were influenced by the restoration techniques (permeability k, distribution coefficient Kd).

Uncertainty and sensitivity analyses were carried out with respect to the collective doses to the public, using the PRISM code (9). Here a Latin Hypercube sampling technique is applied to generate random input parameter values, with which the model is to be run. The most influential parameters, contributing most to the uncertainty were identified, using the simple Pearson and Spearman rank correlation coefficients, between model parameters and model results.

The influence of physico-chemical phenomena on the behaviour of the contaminants (radionuclides) in the environment has been recognised to be of great importance. Most impact assessment models use values of distribution coefficients (Kd) taken directly from the literature, without paying much attention to basic physico-chemical phenomena (such as complexations, redox reactions, hydrolysis etc.) and associated physico-chemical parameters (such as Eh, pH, mineral composition etc.) influencing strongly these values. This brings about large uncertainties of Kd values, which have been shown to constitute very important sources of uncertainty for the dose results. A better strategy than using these single Kd values, is to unfold the Kd into a parameter vector, decomposing the Kd into its underlying basis processes.

From an overview of geochemical speciation modelling software, MINTEQA2 has been selected as an appropriate program, that could allow the unfolding of Kd values.

With this program site-specific values of the distribution coefficients have been calculated for a number of compartments and for the major radionuclides of the example cases. They are compared with the literature-based values in Table 2. The calculated Kd values show uncertainty ranges that are considerably smaller than those based on the literature values.

The speciation model MINTEQA2 has been incorporated into the impact assessment software (PRISM/BIOPATH). The way of incorporation has been kept flexible so that substituting the present speciation modules by another program would require comparatively little effort.

Economic costs

Economic costs constitute one of the major attributes in the optimisation analysis for the ranking of restoration options.

Important cost categories include:

- pure restoration costs
- waste disposal costs
- survey and monitoring costs.

Waste disposal costs, in particular, may be very high, when large quantities of contaminated media are to be disposed of.

Pure restoration costs consist of two important components:

- capital costs, generally incurred once;

- operation and maintenance cost, mainly including labour and consumable costs and usually evaluated on an annual basis.

As far as the restoration costs and waste disposal costs are concerned, values used in this study are derived from unit cost data collected from a literature survey.

Survey and monitoring costs cannot be derived from literature data. They have been estimated on a site-specific basis for each example site.

Table 2 : Distribution coefficients from literature and from site-specific calculations (m^3/kg).

Site and nuclide	Literature data			Site-specific data		
	Mean	Low	High	Mean	Log Mean	Std of log
River Molse Nete						
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)

Social factors

Social factors belong to the group of non-radiological protection factors that must be considered in an optimisation analysis.

Important social factors considered in this study, include:

- reassurance of the public by the implementation of remedial measures;
- disturbance caused by the remedial operations.

These attributes, being difficult to quantify, have been linked to quantities, considered important in determining those factors.

Reassurance is linked to the residual dose and the fraction of activity remaining on the site after the remediation; these being considered the major influences on public reassurance. Disturbance has been linked to the volume of waste to be transported to the waste disposal site, constituting the major cause of disturbance.

The utility values for reassurance have been taken to be inversely proportional to the residual dose and activity remaining on the site. Utility values for disturbance have been taken to be directly proportional to the volumes of waste transported.

The weighting factor for reassurance is assumed to be higher (5 to 7 times) than the weighting factor for disturbance because of its more permanent nature.

RESULTS

The restoration options considered for the three example cases mentioned above, are indicated in Table 3.

With respect to the assessment of the radiological impact at these sites, several very conservative assumptions have been made. As a consequence, the radiological doses assessed should not be considered as realistic values for these sites, but merely as hypothetical examples.

The exposure pathways taken into account at the example sites include:

- external irradiation on contaminated fields or river banks
- inhalation due to resuspension
- ingestion of contaminated drinking water
- ingestion of fish from contaminated surface water
- ingestion of food crops, contaminated through irrigation, or grown on soil contaminated through application of amendments (sediments)

- ingestion of milk, or meat contaminated through watering of the cattle, or through grazing of the cattle on contaminated pasture.

Table 3 : Restoration techniques considered for the example cases.

	Drigg site	Ranstad tailing site	Molse Nete river
A Basecase (No remediation)	X	X	X
B Removal of Sources (Soil/Sediment excavation)			X
C Physical Separation :			
Soil washing C1		X	X
Filtration C2	X		
D Chemical/Biological Separation:			
Solubilisation D1	X	X	X
Ion Exchange D2	X		
Biosorption D3	X		
E Containment : Capping E1&E2	X	X	X
Subsurface Barriers E3&E4	X		
F Physical Immobilisation : *ex*-situ F1	X		X
In-situ F2	X	X	X
G Chemical Immobilisation : *ex-situ* G1	X		X
in-situ G2	X	X	X

The collective doses at the example sites and for the restoration options mentioned above are given in Table 4 (Molse Nete river), Table 5 (Ranstad tailing site) and Table 6 (Drigg disposal site). The uncertainty ranges estimated from the parameter variability were rather restricted (5th and 95th percentiles within a factor of 2 to 5 from the mean values) at the Ranstad site.

Table 4 : Evaluation of attributes at the Molse Nete river.

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

Table 5 : Evaluation of attributes at the Ranstad tailing site.

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

Table 6 : Evaluation of attributes at the Drigg disposal site.

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

At the other two sites the 5th percentiles were 1 to 2 orders of magnitude lower than the mean. The most influential parameters were shown to be primarily the distribution coefficients in soil and sediment (especially for the Pu- and Am-doses) and also:

- the soil-to-plant concentration factors for the Molse Nete river
- the concentration factors to fish for the Ranstad site
- the water flow rates for the Drigg and Ranstad site.

Applying the calculated site-specific Kd values yielded a considerable reduction of the contribution from the Kd values to the uncertainty of the collective doses. For the Drigg site the contribution from the Kd in the drain was reduced from 69 % (for Pu-239) and 64 % (for Am-241) to 3 % and 15 % respectively.

For the Ranstad site the contribution from the Kd in soil (U-238) to the total uncertainty was reduced from 80 to 27 % (U-238 dose).

The data necessary for the evaluation and quantification of the economic and social attributes at the example sites are also indicated in Tables 4, 5 and 6. From these data, utility values and weighting factors for the attributes are derived, according to the procedure described above. The overall scores of the restoration options, determined as the weighted sum of the utilities for all attributes considered are shown in Fig. 3, 4 and 5. The left hand figures show the results for 100 year dose integration and the right hand figures show the results for 500 year dose integration. For the Drigg site, both results were identical.

Uncertainties have been taken into account applying Latin Hypercube Sampling technique on the values for weighting factors and attribute values, assuming a triangular distribution between 1.5^{-1} and 1.5 times the best-estimate value. A negative correlation between collective doses and remediation costs has been applied with a correlation coefficient of –0.8.

The error bars in Fig. 3, 4 and 5 represent the confidence intervals between the 5th and 95th percentiles of the distribution of the total scores, according to the uncertainties adopted.

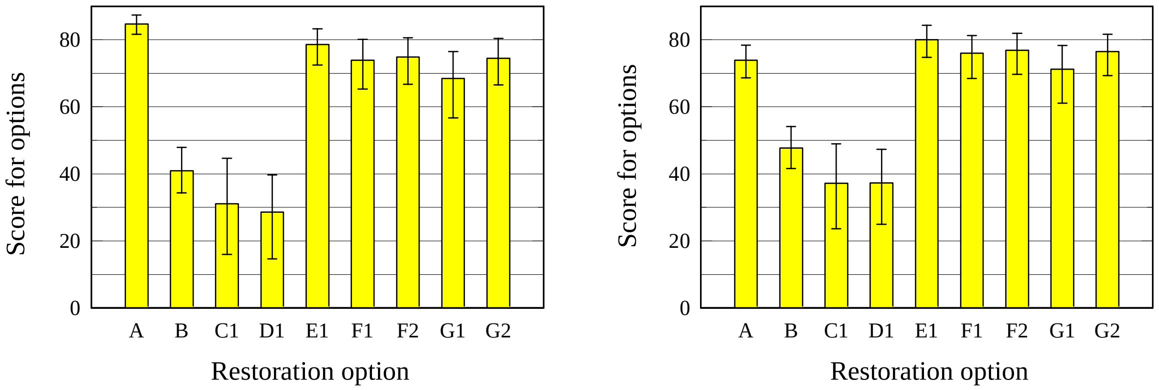
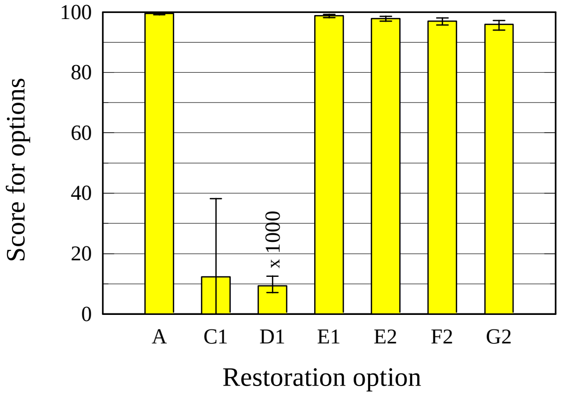


Figure 3 : Total scores for restoration options at the Molse Nete river (100y – 500y collective dose)



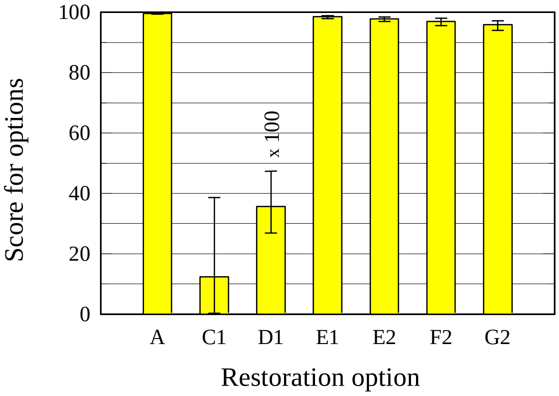
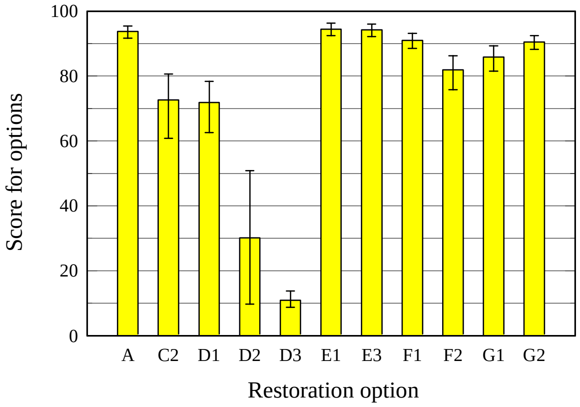


Figure 4 : Total scores for restoration options at the Ranstad tailing site (100y – 500y collective dose)
Figure 5 : Total scores for restoration options at the Drigg disposal site (100y – 500y collective dose)



When considering the best-estimate values in these figures, the no-remediation option seems to be the optimal solution, except for the Molse Nete river, when integrating the collective dose over 500 years and for the Drigg site, where capping seems to be optimal (Table 7). However, when the uncertainty ranges are taken into account, no significant difference is to be observed between the no-remediation, containment and (most of the) immobilisation options. These results reflect the importance of the economic costs accompanying the remediation and, in particular, of the waste disposal costs. These cause the removal and separation options to be never the optimal solution. However, we should bear in mind the premises underlying these results; very conservative dose assessments, generic estimations of the economic costs and a rather arbitrary determination of the weighting factors of the social attributes.

The maximum annual individual dose to the critical group, before remediation, has also been assessed for comparison with the IAEA criteria on clean-up of contaminated land (7). According to these criteria, the need for remediation ranges from "sometimes" (with constraint) or "rarely" (without constraint) for the Ranstad site to "almost always" or "usually needed" respectively for the Drigg site (Table 7).

Table 7 : Summary of results

Site	**Compliance with IAEA criteria**	**Optimised strategy**
Molse Nete river	Remediation usually needed (constraint) or sometimes needed (no constraint)	'No remediation' (100 years); Capping soil and sediment (500 years);
Drigg	Remediation almost always needed (constraint) or usually needed (no constraint)	Capping
Ravenglass estuary	Remediation almost always needed (constraint) or usually needed (no constraint)	'No remediation'
Ranstad tailing site	Remediation sometimes needed (constraint) or rarely needed (no constraint)	'No remediation'
Lake Tranebärssjön	Remediation sometimes needed (constraint) or rarely needed (no constraint)	'No remediation'

CONCLUSIONS

A method for the ranking of remediation options for contaminated sites has been elaborated, based on the optimisation (and justification) principle(s) of radiological protection and taking into account radiological health detriment, economic costs and social factors. Its usefulness has been demonstrated through the application to typical representatives of the major categories of contaminated sites.

The aim of the study has not been to produce some "universal" results for typical cases, but to deliver a method for allowing all attributes influencing the ranking of the restoration options, to be included in a clear and systematic manner, taking into account the uncertainty associated with their evaluation. Such a method is of use for all groups of stakeholders involved in the decision-aiding process with respect to the restoration of contaminated sites, introducing attributes and data reflecting their viewpoints.

REFERENCES

1. Th.Zeevaert A.Bousher, V.Brendler, S.Nordlinder and P.Hedemann Jensen, *RESTRAT Manual. RESTRAT :*

Restoration Strategies for radioactively contaminated sites and their close surroundings. SCK•CEN Report BLG-819. Mol : SCK•CEN (1999).

2. International Commission on Radiological Protection, *1990 Recommendations of the International Commission on Radiological Protection.* Oxford : Pergamon Press; ICRP Publication 60; Ann. ICRP 21(1-3) (1991).
3. International Commission on Radiological Protection, *Optimisation and Decision-Making in Radiological Protection.* Oxford : Pergamon Press; ICRP Publication 55; Ann. ICRP 20 (1) (1989).
4. P.J.Stokell, J.R.Croft., J.Lochard, and J. Lombard, *Alara. From theory towards practice.* Luxembourg : Commission of the European Communities; EUR 13796EN (1991).
5. Nordic Radiation Protection Authorities, *The monetary value of collective dose reduction (α-value).* Statement from the meeting at Reykjavik, 14 June, 1991. Strälskyddsnytt 4/91, Swedish Radiation Protection Institute (1991).
6. International Commission on Radiological Protection, *Protection of the public in situations of prolonged exposure.* Adopted by ICRP Main Commission; to be published (2000).
7. International Atomic Energy Agency, *Application of radiation protection principles to the clean-up of contaminated areas.* Vienna : IAEA – TECDOC – 987 (1997).
8. U.Bergström, O.Edlund, S.Evans and B.Röjder, *BIOPATH - A computer code for calculation of turnover of nuclides in the biosphere and resulting doses to man.* Studsvik AB, Sweden (STUDSVIK/NW-82/261) (1982).
9. R.H.Gardner, B.Röjder and U.Bergström, *PRISM - A systematic method for determining the effect of parameter uncertainty on model predictions.* Studsvik AB, Sweden (STUDSVIK/NW-83/555) (1983).