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Investigation of a possible basis for a common approach with regard to the restoration of areas affected by lasting radiation exposure as a result of past or old practice or work activity

CARE

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Final Report

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Foreword

The scope of the CARE project (Common Approach for Restoration of contaminated sites) is to develop a basis for policies in view of restoring areas contaminated as a result of past practices and work activities involving naturally occurring radionuclides (NOR).

Until recently, regulators have paid little attention to the radiation protection issues associated with such sites. The new Basic Safety Standards¹, for the health protection of the general public and workers against the dangers of ionizing radiation, include provisions for situations leading to lasting exposure under Title IX – Intervention.

The present document is the final report of a study conducted by a consultant (SCK-CEN, contract 96-ET-006), comprising an identification of the areas of concern, a methodology for radiological assessment, a characterisation of different restoration options in terms of performance and cost, and a decision aiding framework using multi-attribute utility functions as well as action levels for intervention referring to international guidance.

The outcome of the study was thoroughly discussed by a working party of the Group of Experts established under Article 31 of the Euratom Treaty, thus also providing quality assurance and editorial improvements. The study will further be used as a working document in view of the establishment of general guidance to Member States on approaches for dealing with lasting exposure situations. This may extend to situations other than those resulting from work activities involving NOR, in particular in the event of areas contaminated as a result of a radiological emergency.

S. Kaiser

¹ Directive 96/29/EURATOM

Abstract

The scope of the CARE project (Common Approach for Restoration of contaminated sites) is to develop a basis for policies in view of restoring areas contaminated as a result of past practices and work activities involving naturally occurring radionuclides (NOR).

The present document is the final report of a study conducted by a consultant (SCK-CEN, contract 96-ET-006), comprising an identification of the areas of concern, a methodology for radiological assessment, a characterisation of different restoration options in terms of performance and cost, and a decision aiding framework using multi-attribute utility functions as well as action levels for intervention referring to international guidance.

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Table of Contents

1.	SCOPE AND LIMITATIONS OF THE STUDY	1
1.1	Scope	1
1.2	Limitations	1
2.	EXECUTIVE SUMMARY	2
2.1	Identification of areas of concern	2
2.2	Assessment	3
2.3	Remediation options	3
2.4	Derivation of criteria and selection of remediation options	4
3.	DEALING WITH NOR CONTAMINATED SITES	5
4.	DESCRIPTION AND INVENTORY OF INDUSTRIES EXTRACTING AND PROCESSING NORM	6
4.1	Occurrence of enhanced levels of naturally occurring radioactive materials (NORM)	6
4.2	Categorisation of NOR-contaminated sites	8
4.2.1	Uranium mining and milling	9
4.2.2	Metal mining and smelting	10
4.2.3	Phosphate industry	10
4.2.4	Coal mining and power production from coal	11
4.2.5	Oil and gas drilling	11
4.2.6	Rare earth and titanium oxide industries	11
4.2.7	Zirconium and ceramics industries	12
4.2.8	Disposal of building materials	12
4.2.9	Application of radium and thorium	12
4.3	References	16
5.	GENERIC RADIOLOGICAL ASSESSMENT APPLIED TO A PHOSPHATE GYPSUM DUMP AS EXAMPLE CASE	17
5.1	Introduction	17
5.2	Summary description of AMCARE - the assessment model for CARE	18
5.3	Assessment for Tessenderlo	19
5.3.1	Site description	19
5.3.2	Model parameters	20
5.3.3	Local residency critical group exposure	26
5.3.4	Intrusion scenario critical group exposure	29
5.3.5	Workforce exposure	31
5.3.6	Collective dose	34
5.4	Discussion	37
5.4.1	Radionuclides and pathways of major importance	37
5.4.2	Effect of parameter uncertainty	37

5.5	Conclusions	38
5.6	References	38
6.	REMEDICATION OPTIONS	39
6.1	Introduction	39
6.2	Remediation options	40
6.3	Selection of remediation technologies for a contaminated site	44
6.4	Application of remediation technologies to a contaminated site	44
6.4.1	Remediation effects on the critical group dose at Tessenderlo	45
6.4.2	Remediation effects on the collective dose at Tessenderlo	45
6.4.3	Workforce dose during remediation activity	46
6.4.4	Cost of remediation based on the Tessenderlo site	48
6.4.5	Fractional removal of contaminant material	48
6.5	Discussion	49
6.6	References	50
7.	DERIVATION OF CRITERIA AND SELECTION OF REMEDIATION OPTIONS	51
7.1	Introduction	51
7.2	Methodology for selection of remedial measures	51
7.2.1	Normalised Action Levels for remedial measures	51
7.2.2	Optimisation of remedial measures	54
7.3	Assessments of remediation strategies for a selected European site	56
7.3.1	Action Levels for a phosphate industry site	57
7.3.2	Optimisation of remedial measures for a phosphate industry site	60
7.4	Conclusions	66
7.5	References	68
ANNEX A1 DESCRIPTION OF INDUSTRIES DEALING WITH NORMS: GENERAL BACKGROUND, INDUSTRIAL PROCESSES, CONTAMINATION LEVELS, MAJOR RELEASE ROUTES AND PATHWAYS		69
ANNEX A2 OVERVIEW OF CONTAMINATED SITES IN EUROPE		105
ANNEX B1 AN OVERVIEW OF COMPUTER-BASED MODELLING APPROACHES FOR RADIOACTIVE WASTE SITES		133
ANNEX B2 A TECHNICAL DESCRIPTION OF AMCARE		153
ANNEX B3 INTERCOMPARISON OF THE DOSE PREDICTIONS OBTAINED WITH THE GENERIC MODEL, AMCARE, WITH THOSE OBTAINED FROM A MODEL SPECIFIC TO THE TESSENDERLO SITE		177
ANNEX C1 OVERVIEW OF REMEDIATION TECHNOLOGIES		183
ANNEX D1 OVERVIEW OF REMEDIATION CRITERIA FROM INTERNATIONAL ORGANISATIONS		195

ANNEX D2 OVERVIEW OF CRITERIA FOR REMEDIAL MEASURES IN DIFFERENT COUNTRIES	207
ANNEX D3 JUSTIFICATION AND OPTIMISATION OF PROTECTION	219
ANNEX D4 METHODOLOGY FOR DETERMINING ACTION LEVELS FOR REMEDIAL MEASURES	230

1. SCOPE AND LIMITATIONS OF THE STUDY

1.1 Scope

The scope of the CARE project (Common Approach for REstoration of contaminated sites) is to develop a basis for a common approach to restoring areas affected by lasting radiation exposure from natural radionuclides. The sites considered are areas contaminated as a result of past practices or work activities. These may include activities which may not have been classified as 'of radiological concern' or which may be subject to site licensing for disposal of radioactive materials based on contemporary criteria.

Enhanced levels of naturally occurring radionuclides (NORs) may be associated with abandoned waste dumps, installations and surroundings from certain industries, involved in the extraction or processing of raw materials containing NORs. This can result in considerable exposure to the public. Until recently, regulators have paid little attention to the radiation protection issues associated with these sites. This study provides a basis for conceptualising and quantifying the extent of the radiation problem caused by residues from past activities of NOR-extraction and processing industries at a European scale, and also provides a basis for the relevant national and European authorities to set directives and regulations for the protection of the general public to radiation exposure of this origin.

The study is subdivided into four main parts:

- Identification of areas of concern:
 - category of industries handling NOR-containing material;
 - overview of sites in Europe contaminated with residues from the industries considered.
- Radiological assessment:
 - elaboration of an appropriate assessment methodology for existing conditions and with extrapolation to future normal evolution and intrusion scenarios.
- Restoration options:
 - characterisation and evaluation of remediation techniques in terms of performance, costs and social implications using a cost-effectiveness and multi-attribute utility decision aiding framework.
- Intervention levels:
 - overview of restoration criteria;
 - proposition of methodology for deriving action levels and criteria.

1.2 Limitations

There are a large number of non-radiological contaminants which can occur in the uranium tailings and other NOR-containing wastes which can be mobilised under acidic conditions and appear in seepage, including heavy metals, rare earths, salts and nutrients. Since the non-radiological pollutants in some of the wastes can present as great a hazard to the environment as the radiological pollutants, both must be taken into account and factored into the safe management of waste. Consideration of the non-radiological hazards is, however, outside the scope of the CARE project and is not dealt with in the general assessment and the multicriteria utility analysis.

Voluntary future occupation of the contaminated site (e.g. for the implantation of industries) was not considered. For voluntary occupation dose limits and criteria may be different.

2. EXECUTIVE SUMMARY

The objective of the CARE project is to develop a basis for a common approach to restoring areas affected by lasting radiation exposure from naturally occurring radionuclides (NORs). This can arise from past, or old, practices or work activities. This objective is met through considering, in detail, four principal areas of work:

- identification of areas of concern
- assessment of the radiological impact
- inventory of and methodology for selection of remediation options
- derivation of remediation criteria and action levels

2.1 Identification of areas of concern

Nine important categories of industries involving the extraction and processing of materials which contain enhanced levels of NORs have been identified:

- uranium mining and milling
- metal mining and smelting
- phosphate industry
- coal mining and power production from coal
- oil and gas drilling
- rare earth and titanium oxide industry
- zirconium and ceramics industry
- building materials
- application of radium and thorium

To determine the extent of the radiological problems related to these industrial activities, information has been collated on the industrial processes and the resulting levels of NORs in feed-stock, waste and (by)-products. Emphasis has been placed on the issues of past, and old, practices.

The most contaminating widely distributed industries are uranium mining and milling (mainly due to atmospheric exposure to ^{230}Th and ^{222}Rn in the vicinity of the tailings and exposure to ^{226}Ra through aquatic pathways), metal mining and smelting (^{210}Pb and ^{210}Po in the vicinity of smelters and ^{232}Th inhalation in the vicinity of the deposit) and the phosphate industry (radon and ^{226}Ra).

The levels of ^{226}Ra and ^{228}Ra in scales from the oil and gas extraction industry and levels of ^{232}Th and ^{238}U in feed material, products and wastes of the rare earth and zirconium and ceramics industries may be particularly elevated. However, given the limited extent of these industries and concomitant limited waste streams, the radiation problem for the general public is overall small.

Impact on the public from coal mining and power production from coal is commonly considered low. Specific activities in building materials are very low. No typical values are available for contaminant levels in materials, buildings and surroundings of radium extraction and luminising plants, nor for thorium extraction and processing plants.

An attempt to give an overview of sites in Europe contaminated with NOR, due to past activities, partially failed since information was often limited or was not available. One prominent European case of environmental contamination, due to past mining practices, is in the districts of Saxony, Thuringia and Saxony-Anhalt, in former East Germany. Here, exceptional radiological situations have developed as the result of centuries of uranium, coal and precious metal mining.

2.2 Assessment

The impact of waste from the extraction and processing of NOR-containing material, the consequential public concern and the need for decisions on restoration and remediation of radioactively contaminated sites, require systematic investigation and objective evaluation of the existing (and prospective) radiological situation.

A substantial site characterisation has to be performed before any assessment can be initiated. This will include radiological and physico-chemical characterisation of waste and surroundings, site geology and hydrology, demography, etc.

A model, AMCARE, has been developed under CARE to assess the individual doses to an average member of the critical group for current conditions and to assess the maximal dose occurring in a period of 10000 years. A local collective dose to the population living within a 20 km radius from the site is assessed for 100 and 500 years. Both 'normal evolution' and 'intrusion' scenarios are considered.

To illustrate the AMCARE model, the abandoned phosphogypsum dumps from Tessenderlo (Belgium) was selected. This site was chosen since sufficiently detailed information could be collected, and , secondly, since the phosphate industry is one of the most important contaminating industries, due to NOR-levels in the ore and wastes and its spreading in Europe. Although for the different categories, the wastes originate from a number of different industrial processes, many of the features of these wastes are common to all sites. This has allowed a generic modelling approach to be developed which can be applied on a site-by-site basis. The important exposure pathways for the radionuclides of major concern are indentified, and dose conversion factors calculated for the different scenarios.

The dominant dose to almost all exposure groups arises from inhalation of radon gas which arises from the parent inventory of ^{226}Ra . To be able to compare the dose estimates for the different sites, doses incurred per unit concentration of the dominant radionuclides were calculated.

Doses in excess of 1 mSv a^{-1} were obtained for external radiation and food ingestion pathways for the intrusion scenario only. However, the doses obtained from these pathways were always more than two orders of magnitude lower than that from the radon inhalation dose.

The main factors determining the uncertainty in the dose estimates, other than the waste inventory itself, relate to the rate of emission of radon gas to the atmosphere (emanation factor and surface layer thickness) and, to a lesser extent, to the shielding from external radiation. Radon exhalation in turn is very sensitive to changes in soil moisture and structure.

The model also predicts the impact of different remediation strategies on the different scenarios for each site (including the dose to the workforce). The uncertainties, associated with such strategies, are reflected in this model.

2.3 Remediation options

Remediation technologies are techniques (or measures) which prevent (or reduce) the radiological impact (or risk) to the population from a contaminant source. A wide variety of remediation technologies are available. However, techniques considered for the CARE project were limited to those which are well-established and require little maintenance. Remediation technologies may be divided in four categories:

1. removal of sources
2. containment (capping and subsurface barriers)
3. immobilisation (cement and chemical based solidification)
4. separation combined with removal (soil washing, flotation and chemical/solvent extraction)

The remediation technologies considered are discussed in terms of their impact on the emission of radon, the mobility of radionuclides and radiation effects. Each technology is assessed in terms of the cost of implementation, performance, service life and workforce exposure during remediation.

Since it is indicated that the main exposure pathway arises from radon emanation, actions which reduce the rate of emanation may thus supposed, *a priori*, to represent the most likely cost-effective approaches in site remediation measures.

2.4 Derivation of criteria and selection of remediation options

An overview is given of remediation criteria from international organisations. The ongoing work on criteria for protection of the public against chronic exposure, within ICRP and IAEA working groups, is not yet fully complete. The two approaches from IAEA and ICRP are in fact similar as they both operate with a generic individual dose level of about 10 mSv a⁻¹ as a dividing line between situations that might be considered as 'normal' and situations where some remedial or protective actions should be considered. Criteria for remedial measures at contaminated sites in Europe and other countries around the world are, with a few exceptions, not fully developed. The rationale for deriving remediation criteria in different countries is not very clear and conceptual differences, between existing national criteria, exist. So far no common approach, or common numerical guidance, has emerged.

In the context of remediation of contaminated sites, remediation actions should be justified and, hence, subjected to an optimisation process for selecting the best strategy of remedial measures.

A specific remediation option is justified when there is a positive net benefit between the total cost of the remediation, including the equivalent cost of the collective dose to the workers implementing the measure, and the equivalent monetary value of the dose reduction to the affected population. For the selected site, operational remediation criteria in terms of Action Levels (AL), e.g. the activity concentration within the contaminated media, have been derived from justification calculations in which site-specific parameters, such as cost, efficiency of the remedial measure and averted dose to the affected population, play an important role. An Action Level is the level of dose rate or activity concentration above which remedial or protective actions should be considered. An Action Level is not a limit but can be used as a screening tool to determine if a remediation is justified on economic grounds. In this study the Action Level concept has been restricted to a pure cost-benefit expression.

The optimum remediation option is selected by means of multi-attribute utility analysis (MAUA), which allows for the inclusion of factors which are not easy to quantify in monetary terms. The attributes which have been considered by CARE are:

1. health attributes (collective radiation dose to members of the public)
2. cost attributes (cost of remedial actions, including disposal costs of generated waste) and equivalent monetary costs of collective dose to contractual workers
3. social attributes (reassurance of the public)

The MAUA required the selection of appropriate weighing factors for the different attributes. These weighing factors are used in the derivation of scores for the different attributes for the different remediation options. The option with the highest score is considered the optimal

option. The outcome of any MAUA should, however, be judged carefully in the light of the attributes selected and the values assigned to the corresponding weighing factors before any firm conclusions can be drawn.

3. DEALING WITH NOR CONTAMINATED SITES

The scope of the CARE project is to develop a basis for a common approach to restoring areas contaminated by natural occurring radionuclides due to past practices. For situations where there is existing exposure of a population from sites contaminated with the residues of past practices or work activities, the principles of protection for *intervention* are applicable. As recommended by the ICRP (1990) dose limits do not apply to intervention situations. Levels for intervention and the application of a remediation strategy should be based on an optimisation of protection of the affected population. However, in most regulations, national levels for intervention are based on long-term exposures from the residues of old practices or events which have been judged to be unacceptably higher than the dose limit for ongoing practices. This view is *not* in accordance with the recommendations from international organisations. Further development and guidance is therefore needed to aid decisions on implementation of remedial measures at sites contaminated by past or old practices or work activities.

Decisions on remedial measures for a contaminated site will almost always be based on historical information and on radiation measurements. The decision process can be considered as iterative and interactive phases of planning, survey, assessment, and decision. Four phases can be characterised:

- The *planning phase* includes defining the problem, using the available information to estimate the kind and scope of the problem, and determining the kind, quality and quantity of measurements needed to make a decision.
- Several types of *surveys* may be necessary and have different objectives, e.g. historical site assessment, scoping measurement survey, detailed site characterisation surveys, surveys during clean-up operations, and surveys to confirm that the clean-up reached the final levels as planned.
- *Assessment* of the radionuclide levels in the environment based on measurements and verified with extra measurements, to ascertain that interpretations are valid. Assessment of the radiation exposure should hence be done for different scenarios (e.g. normal evolution, intrusion), including the assessment of the effect of remediation options.
- *Decisions* with respect to the measurements depend on verification that the data were interpretable as planned, that records document the findings, and that the basis for decisions is explicitly stated.

Occurrence of NOR-containing material and a description of the different industrial activities which involve extraction and processing of NORM is presented in Chapter 4 and Annex A1. Details on the assessment of radiological impact are presented in Chapter 5. The impact of remedial measures are given in Chapter 6. In Chapter 7, a methodology is proposed on the derivation of criteria and the selection of remediation options.

4. DESCRIPTION AND INVENTORY OF INDUSTRIES EXTRACTING AND PROCESSING NORM

4.1 Occurrence of enhanced levels of naturally occurring radioactive materials (NORM)

Naturally occurring radionuclides (NORs) can be subdivided into cosmogenic radionuclides (e.g. ^{14}C), which are continuously formed in the outer atmosphere by cosmic irradiation, and primordial radionuclides (e.g. ^{40}K , ^{232}Th , ^{235}U , ^{238}U), present since the formation of the Earth. The primordial radionuclides ^{238}U ($t_{1/2}$ 4.5×10^9 a, abundance 99.27%), ^{235}U ($t_{1/2}$ 7.1×10^8 a, abundance 0.72%) and ^{232}Th ($t_{1/2}$ 4.5×10^{10} a, abundance 100%) are followed by a series of nuclear decays, displayed in Figure 4.1. Uranium and thorium are present in the Earth's crust at average concentrations of 4.2 and 12.5 mg kg $^{-1}$, respectively (corresponding to 50 Bq kg $^{-1}$ for each of the nuclides). However, levels of up to a few percent are found in particular ores [Dixon, 1984].

Apart from the obvious occurrence of naturally occurring radioactive materials (NORM) in uranium deposits, a wide range of uranium- and thorium-bearing minerals (and daughters) are being mined and processed commercially.

In most minerals, natural levels of radionuclides are very low. In others, e.g. zircon and rare earths, the concentration of ^{238}U and ^{232}Th may be considerably elevated. The activity in zircon is principally due to thorium, which can reach levels of about 10 kBq kg $^{-1}$. The activity of thorium in monazite, which is mainly cerium phosphate, can exceed 350 kBq kg $^{-1}$. The size of the calcium ion is also similar to that of the tetravalent uranium and thorium ions, so the overall activity content of minerals, such as apatite, can reach levels of 2 kBq kg $^{-1}$.

^{238}U series		^{235}U series		^{232}Th series	
Nuclide	Half-life	Nuclide	Half-life	Nuclide	Half-life
^{238}U	4.51×10^9 a	^{235}U	7.1×10^8 a	^{232}Th	1.41×10^{10} a
↓		↓		↓	
^{234}Th	24.1 d	^{231}Th	25.6 h	^{228}Ra	5.8 a
↓		↓		↓	
^{234}Pa	1.17 min	^{231}Pa	3.4×10^4 a	^{228}Ac	6.13 h
↓		↓		↓	
^{234}U	2.47×10^5 a	^{227}Ac	21.6 a	^{228}Th	1.910 a
↓		↓		↓	
^{230}Th	8.0×10^4 a	^{227}Th	18.6 d	^{224}Ra	3.64 d
↓		↓		↓	
^{226}Ra	1602 a	^{223}Ra	11.7 d	^{220}Rn	55 s
↓		↓		↓	
^{222}Rn	3.823 d	^{219}Rn	3.9 s	^{216}Po	0.15 s
↓		↓		↓	
^{218}Po	3.05 min	^{215}Po	1.8×10^{-3} s	^{212}Pb	10.64 h
↓		↓		↓	
^{214}Bi	19.7 min	^{211}Pb	36 min	^{212}Bi	60.6 min
↓		↓		↓	
^{210}Pb	21 a	^{211}Bi	2.2 min	^{208}Pb	Stable
↓		↓			
^{210}Bi	5.01 d	^{211}Pb	0.5 s		
↓		↓			
^{210}Po	138.4 d	^{207}Pb	Stable		
↓					
^{206}Pb	Stable				

Figure 4.1 Decay series of ^{238}U , ^{235}U and ^{232}Th

The activity of minerals containing elements with different ionic sizes from uranium and thorium can also be enhanced if higher activity minerals are present as accessories. Zircon often occurs as an accessory in tin and copper deposits while phosphate can contain apatite. Activity levels from accessory material are difficult to predict, as their occurrence is uneven.

The potential for increased human exposure from ^{40}K is low. In addition, whilst there are a number of other radionuclides of primary origin, there is no evidence that they are of radiological significance. They are not considered in this study.

Where people are employed in mineral processing, they might receive substantial radiation doses. When wastes from mining, processing or construction are disposed off, people may also be exposed to radiation burden. Legislation regarding the protection of the public and workers against natural sources of radiation is rather vague. Title 7 of the Council Directive 96/29/EURATOM obliges the different member states to investigate the problem of natural sources of radiation, engage in an extensive monitoring programme and take adequate measures in case of increased exposure. No strict guidelines are given about measures to be taken by each of the member states. They may be guided by the criteria applicable for artificial radionuclides. Alternatively, the member states could be guided by the exemption values specified in the Basic Safety Standards [Council Directive, 96/29/EURATOM] for the quantities and concentrations of natural radionuclides. It should be stressed, however, that the application of exemption to natural radionuclides, is limited to the incorporation of naturally occurring radionuclides into consumer products or their use as radioactive source (e.g. ^{226}Ra , ^{210}Po) or for their elemental properties (e.g. thorium, uranium)" These exemption levels are hence not applicable for most of the waste the CARE project is referring to.

For the discussion on criteria for dose limits, the reader is referred to Chapter 7.

Table 4.1 Exemption values as specified in Council Directive 96/29/EURATOM.

Nuclide	Quantity (Bq)	Specific activity (Bq kg ⁻¹)
<i>²³⁸U decay chain</i>		
²³⁸ Usec	10 ³	10 ³
²³⁸ U+	10 ⁴	10 ⁴
²³⁴ Th+	10 ⁵	10 ⁶
²³⁴ U	10 ⁴	10 ⁴
²³⁰ Th	10 ⁴	10 ³
²²⁶ Ra+	10 ⁴	10 ⁴
²²² Rn+	10 ⁸	10 ⁴
²¹⁰ Pb+	10 ⁴	10 ⁴
²¹⁰ Bi	10 ⁶	10 ⁶
²¹⁰ Po	10 ⁴	10 ⁴
<i>²³⁵U decay chain</i>		
²³⁵ U+	10 ⁴	10 ⁴
²³¹ Th	10 ⁷	10 ⁶
²³¹ Pa	10 ³	10 ³
²²³ Ra	10 ⁵	10 ⁵
<i>²³²Th decay chain</i>		
²³² Thsec	10 ³	10 ³
²²⁸ Ra+	10 ⁵	10 ⁴
²²⁸ Ac	10 ⁶	10 ⁴
²²⁸ Th+	10 ⁴	10 ³
²²⁴ Ra+	10 ⁵	10 ⁴
²²⁰ Rn+	10 ⁷	10 ⁸
²¹² Pb+	10 ⁵	10 ⁴
²¹² Bi+	10 ⁵	10 ⁴

Note: 'Sec' means in secular equilibrium, '+' means with only short-lived daughters in equilibrium.

4.2 Categorisation of NOR-contaminated sites

For the purposes of CARE, industrial activities can be broadly subdivided in three groups.

- Past controlled practices:
 - uranium mining and milling.
- Practices not under radiological control:
 - metal mining and smelting;
 - phosphate industry;
 - coal mining and power generation from coal;
 - oil and gas drilling;
 - rare earths and titanium oxide industry;
 - zirconium and ceramics' industry;
 - disposal of building materials.
- Practices which would now be under control:
 - applications of the natural radionuclides radium and thorium.

The different categories were defined on the basis of the probability of occurrence of high levels of NORM in the mother material, products, by-products and wastes and also, on the importance of the industry (and hence possible extent of the problem). Some processes and activities, products and wastes, which contain potentially elevated levels of NORM, but for which, either, information is lacking or, the scale of the process is small, are not discussed. They include: extraction of geothermal water, glassware and glass enamel, porcelain teeth, ophthalmic and optical glass, gemstones and chemicals. Some of these categories are briefly described by Martin *et al.* (1996) who also concluded that insufficient information was available to allow conclusions to be drawn on their radiological impact.

Most categories have not previously been considered as industries with potential radiological impacts on man and environment and, until now, have not been under radiological control. In the industrial processes associated with the extraction and processing of geochemical materials, the hazard from radiation is generally small compared to that from other chemical substances and so radiation has not been systematically monitored. Since CARE deals with past practices, occupational exposure is outside the scope of the project but the interested reader is referred to a number of reports where attempts have been made to assess the occupational exposure in the extraction and processing of geochemical materials [Harvey *et al.*, 1994; Martin *et al.*, 1996; Scholten, 1996; Van Weers *et al.*, 1996; Mustonen *et al.*, 1997; Penfold *et al.*, 1997].

The present chapter overviews the industrial activities which involve the extraction and processing of materials which may contain enhanced levels of naturally occurring radioactive materials (NORM). Information about the processes, scale of the industry and levels of naturally occurring radionuclides (NORs) in feed materials, and waste and by-products was collated from literature and issues regarding past practices were emphasised. Radionuclides from the ^{238}U and ^{232}Th decay series were mainly considered.

The most important information concerning the extent of each industrial category and specific activity levels and most important pathways are presented below and summarised in Table 4.2.

A more in-depth description of each industrial category, in terms of general background, industrial processes and source term and major release routes and pathways can be found in Annex A1, of which this chapter is a summary. All reference material on which this summary is based can be found in Annex A1.

It is an objective of CARE to give an overview of all sites in Europe contaminated with NORM arising from past practices. The inventory of available information in terms of both category and country can be found in Annex A2. No detailed and complete information is (accessibly) available on the number and types of extraction industries in each European country or on the radiological impact of the industry on man and the environment. Furthermore, given the scope

of this report, only past activities were considered when describing and assessing the different categories. It was beyond the scope of this project to notify all relevant authorities or institutes in each European country to obtain information on the importance of the respective industry in a country and an indication of all past liabilities in each category. However, to obtain general information, all the participants to the IAEA TECDOC-865 [IAEA, 1996a,b,c] and some other sources were notified and asked for information on categories for their country (occurrence and importance of industry, indication of all liabilities in that category, status, i.e. operational-closed-under remediation-remediated, and, if possible, radionuclide levels, or information on a specific case study). Some addressees responded to the questionnaire and are referred to as 'personal communication'.

When no country-specific information was found for a category within the scope of the CARE project, the category was nevertheless described, the production data for Europe given (mostly only for the EU-12) and radiological hazards and pathways and possible extent of the problem indicated to provide the reader with an idea of the radiological importance of each category.

4.2.1 Uranium mining and milling

The cumulative uranium production in the western world to the end of 1990 was around 1 Mt. European uranium mining and milling was mainly carried out in Germany, Czech Republic, France, Bulgaria, Romania, Hungary and Spain. Most mining and milling sites in France and Spain are closed and remediated or remediation is in progress. There are also many abandoned sites in Eastern Europe; however none are remediated. Many of these sites pose serious problems because preservation and protection of the environment has often been neglected and also because they are, in many cases, situated close to human populations.

The most prominent case of environmental contamination is due to mining activities in former East Germany. The barren rock (3-30 t per t ore for open pit mines) contains on average 15-25 mg kg⁻¹ uranium with a total activity of 2-5 kBq kg⁻¹. Cut-off grade rocks generated in comparable amounts as the ore contain, on average, 150 mg kg⁻¹ uranium with a total activity of 30 kBq kg⁻¹. For the static and dynamic leaching residues, the uranium concentration and total activity are 20 mg kg⁻¹ uranium and <40 kBq kg⁻¹ and 50 mg kg⁻¹ uranium and 300 kBq kg⁻¹, respectively. The sands which contain 20% of the initial radium (1-4 kBq kg⁻¹ ²²⁶Ra) contain on average 0.004-0.01% U₃O₈ and 2-22 kBq kg⁻¹ ²³⁰Th. The fines have a similar ²³⁰Th content but contain 80% of initial ²²⁶Ra (5-20 kBq kg⁻¹) and 0.016-0.04% U₃O₈.

The major atmospheric exposure in the vicinity of tailings, comes from the inhalation of ²³⁰Th and ²²²Rn and ²²⁶Ra, ²³⁸U, ²¹⁰Po and ²³²Th on dust. ²²⁶Ra is generally the nuclide of major concern in the *aquatic pathway*. Doses to the public can result from consuming contaminated aquatic biota or water or through ingestion of plants irrigated with contaminated water. Where fish is a major source of food, this pathway can dominate. Since radium migration through groundwater is retarded by soils, exposure through this pathway is generally unimportant. Doses from radionuclides such as ²¹⁰Pb, ²¹⁰Po, uranium and thorium are usually less than those from ²²⁶Ra. However, under some circumstances, the release of these radionuclides can be of the same order of magnitude as or even higher than for ²²⁶Ra (e.g. U-migration under oxidising conditions) and should be considered separately.

4.2.2 Metal mining and smelting

Not much information is available on historic metal mining and smelting sites. This is particularly the case for radiological data. One area with a legacy of metal mining and smelting is the Mansfeld area of Germany. All sites have been abandoned since 1990. NOR levels are higher than the average for metal mining and smelting activities because the metal ore was uranium bearing.

For the different metal industries considered (aluminium, copper, iron and steel, lead, niobium, tin and zinc), NOR activities in feed material for metal smelting are generally low. The same holds for slags and other wastes. Ore and waste products for the tin industry are around the exemption limits. The pyrochlore feedstock for niobium production has activity levels of 10-80 Bq kg⁻¹ and niobium slag has activity levels comparable to the mother material. The ²¹⁰Pb and ²¹⁰Po activities in the dust and sludges can reach 10² to 10⁵ Bq kg⁻¹. However, given the relatively short half-life of these radionuclides, the hazard is limited in time. The ²¹⁰Pb and ²¹⁰Po from the stack of metal smelters for the Mansfeld area in Germany, has contaminated the area in a 3 km radius since no appropriate filters are used.

Public exposure from landfill disposal of tin slag was assessed to be in the range of 10 μSv a⁻¹ for a single site. The dominant radionuclide is ²³²Th and the exposure is mainly through inhalation of ²³²Th after re-suspension of material deposited on arable land.

4.2.3 Phosphate industry

Phosphate ore is not mined in Europe, yet phosphate ore processing plants are present in all European countries. Of the ~30 Mt imported annually, about 55-60% is consumed by the EC countries, the balance being made by the EFTA countries and the eastern European countries. Moroccan ore, which is the major feedstock for the European phosphate industry, has uranium and radium concentrations of between 1500 and 1700 Bq kg⁻¹. Fertilisers become enriched in uranium to about 100-150% of the initial ore concentration whereas most of the radium goes to the phosphogypsum (5 t per t phosphoric acid) (<3000 Bq kg⁻¹). Radionuclide concentration in the slag of the thermal process for converting ore to elemental phosphorus are also maximally a few thousand Bq kg⁻¹.

Little information was found on closed phosphate processing plants. However, the radiological impact on the environment can be deduced from an assessment of the impact of closed disposal sites. The exposure from a disposal site is mainly due to exposure from radon and from ground- and surface water contamination with radium (0.02-1.1 mSv a⁻¹ depending on pathway).

4.2.4 Coal mining and power production from coal

The total consumption of coal, brown coal and peat for electricity production, in the EU-12, was about 300 Mt in 1992. This results in ~30 Mt of ash, of which 10-25% was bottom ash and 75-90% fly ash.

The average specific activity in coal is generally around 20 Bq kg⁻¹ for both ²³⁸U and ²³²Th. An extreme case is reported in Freital where specific uranium activities of 15000 Bq kg⁻¹ were recorded. In general, the radionuclide enhancement factor for uranium and thorium in the ash is about 10.

Specific activities in lignite and peat are generally much lower than in coal. However, in Greece, activity levels in lignite comparable to coal have been reported.

The impact of coal mining on the public is considered to be low, except on occasions where the critical group lives close to the ash disposal site (re-suspension of ash onto agricultural land: up to 250 μSv a⁻¹).

4.2.5 Oil and gas drilling

The total production of natural oil and gas in the EU-12 is 140 Mt and 0.23 × 10¹² m³, respectively. This gives an annual sludge production of 10000 m³.

The dominating radionuclides in scales and other precipitates are ²²⁶Ra and ²²⁸Ra, with specific activities in the order of 10³-10⁵ Bq kg⁻¹. The activities in sludges are typically a factor 100

lower. However, the ^{210}Po and ^{210}Pb concentrations in sludges and scales can vary between 20 and 10^5 Bq kg^{-1} . ^{238}U and ^{232}Th activities are generally low.

There is a lack of data concerning public exposure to scales and sludges, although the main pathway would arise from waste disposal and storage processes. The (future) decommissioning of land- and sea-based extraction plants should be given attention with respect to volume and contamination levels (contaminated pipelines).

4.2.6 Rare earth and titanium oxide industries

Monazite and bastnasite are the most important rare earth-containing minerals. However, no mining is carried out in Europe. Rare earths are used in polishes, in catalysts, as burnable poisons in nuclear reactors and as additives in steels and special glasses.

The radionuclide concentrations in the mother material are around 1000 Bq kg^{-1} for the ^{238}U decay series and between $6000\text{-}10000 \text{ Bq kg}^{-1}$ limit for the ^{232}Th decay chain. Barium sulphate from the extraction process may contain 3000 and 450 Bq kg^{-1} of ^{228}Ra and ^{226}Ra , respectively. However, in general, the activity concentrations in the wastes are comparable to the concentrations in the feeding material. The same applies to the activity concentration in the end products.

The radionuclides of particular concern are ^{232}Th and ^{238}U . The individual dose from the migration of radionuclides from a landfill is $\sim 0.1 \mu\text{Sv a}^{-1}$.

Only 2 t of the $\sim 1 \text{ Mt}$ titanium ore processed is mined in Europe. The radiological hazards from TiO_2 production vary with the type and source of ore. The ore (mainly rutile, ilmenite, monazite) activity concentration of ^{238}U and ^{232}Th decay chains may vary between 30 to 600 Bq kg^{-1} and 35 to 6000 Bq kg^{-1} , respectively. In the presence of high sulphate concentrations radium is found in the solid residues as an insoluble sulphate compound. Specific activities of such radium precipitates in pigments or scale, formed in the heat exchanger, have been reported to be about $4 \times 10^5 \text{ Bq kg}^{-1} \text{ }^{226}\text{Ra}$.

Dose rates measured at the surface of some vessels, pipes and wet filters, in a Yugoslavian production unit, varied between background and $50 \mu\text{Sv h}^{-1}$. Care must thus be taken when these facilities are decommissioned.

4.2.7 Zirconium and ceramics industries

Zirconium is an abundant element whose principal sources are zircon in silicate sands and zirconia in baddleyite ore. Zircon and zirconia are used in the manufacture of refractory products, glazes, glasses and ceramics and as additives in alloys. They are not mined in Europe. The average activity levels in zircon and zirconia are 600 and 300 Bq kg^{-1} for ^{232}Th and 3000 and 7000 Bq kg^{-1} for ^{238}U , respectively. Except for refractory bricks, where ^{238}U activity levels of 10000 Bq kg^{-1} are reported, the activity concentrations in the products are comparable to the concentrations in the feeding material.

Exposure from the rare earths' industry, mainly occurs during the operational phase. Waste streams are limited but the products are eventually disposed of in non-radioactive landfill sites. The re-development of a disposal site where substantial quantities of refractory bricks were landfilled may result in individual doses of up to $100 \mu\text{Sv a}^{-1}$.

4.2.8 Disposal of building materials

Industrial by-products with possibly enhanced levels of NORs are used as raw materials for building materials. Phosphogypsum is the most widely used, but slags and mining wastes are also used. Specific activities in building materials are typically around 50 Bq kg^{-1} both for ^{238}U and ^{232}Th .

Re-development of a dump site containing building material 300 years after closure is calculated to result in an external exposure of 3 mSv a^{-1} . This is mainly due to ^{238}U . The radon exposure, during residence on the site, was 1.4 mSv a^{-1} .

4.2.9 Application of radium and thorium

The radium concentration in ores is very low. Almost half of the 4.5 kg of radium produced in the world since 1898 has been extracted in Belgium. Radium was used as luminising agent, in medicine and in material structure research. No typical values are available the materials, buildings and surroundings of radium extraction and luminising plants. High contamination levels were recorded in soil in the surrounding of luminising works in London with between 400 and 400000 Bq kg^{-1} ^{226}Ra with hot spots of 4 MBq kg^{-1} . Surface dose rates ranged from background ($0.1 \text{ } \mu\text{Sv h}^{-1}$) to $100 \text{ } \mu\text{Sv h}^{-1}$. Similar doses and concentrations were found in the vicinity of a watch factory (Dieppe, France).

The dose rate for a radium extraction plant disposal site in Olen, Belgium was 2.8 (0.1-1000) $\text{ } \mu\text{Sv h}^{-1}$. Dose rates in the vicinity of the plant are between 0.1 and $100 \text{ } \mu\text{Sv h}^{-1}$.

Thorium is extracted from the same minerals used for rare earth extraction. The specific activity of the feed material is in the range of 1000-10000 Bq kg^{-1} . The activity levels in the products (gas mantles, thoriated glass, Tungsten) are typically a factor of 100 higher. Some information on past thorium practices in France was found.

Table 4.2 Summary of the scale of industry and contamination levels in feed material, products and waste for the different industrial categories

CATEGORY 1. Feed material 2. Product 3. Wastes and by-product	Scale of extraction, production and waste generation	Specific activities of principal radionuclides and enhancement factors	Principal pathways from wastes and dose to public
URANIUM MINING & MILLING 1. Uranium ore 2. Yellow cake 3. a. barren rock b. cut-off grade rock c. static leaching residues d. dynamic leaching residues e. sands } f. slimes } constituents of c & d g. liquids }	1. World: 1000 t U a ⁻¹ ; cumulative U-prod. until 1990: ~1.7 Mt U. 3. a. open pit: 3-30 t (t ore) ⁻¹ ; underground: 1-2 t (t ore) ⁻¹ . 3. b. open pit: 1-2 t (t ore) ⁻¹ ; underground: 0.1-0.2 t (t ore) ⁻¹ . 3. c. 1 t (t ore) ⁻¹ . 3. d. 1.1 t (t ore) ⁻¹ .	1. 0.04-2% U ₃ O ₈ . 2. 90-95% of U and 15% of total activity. 3. a. 15-25 mg kg ⁻¹ U, 2-5 kBq kg ⁻¹ . 3. b. 150 mg kg ⁻¹ U, <30 kBq kg ⁻¹ . 3. c. 20 mg kg ⁻¹ U, <40 kBq kg ⁻¹ . 3. d. 50 mg kg ⁻¹ U, 300 kBq kg ⁻¹ . 3. e. 0.004-0.01% U ₃ O ₈ , 2-22 kBq kg ⁻¹ ²³⁰ Th 20% of initial ²²⁶ Ra = 1.4 kBq kg ⁻¹ . 3. f. 0.016-0.04% U ₃ O ₈ , 2-22 kBq kg ⁻¹ ²³⁰ Th 80% of initial ²²⁶ Ra = 5-20 kBq kg ⁻¹ . 3. g. 0.001-0.01% U ₃ O ₈ , 75-800 Bq L ⁻¹ ²³⁰ Th 0.5-280 Bq L ⁻¹ ²²⁶ Ra.	<i>Atmospheric pathway:</i> main exposure from gaseous ²²² Rn, and solid daughters attached to aerosols and air-born particulates containing, ²³⁰ Th, ²²⁶ Ra, ²¹⁰ Pb in immediate vicinity of old tailings: ~old ICRP dose limits; >0.5 km: no problem. <i>Aquatic pathway:</i> ²²⁶ Ra.
METAL MINING AND SMELTING 1. Metal ores 2. Metals, alloys, coatings, ... 3. a. waste rock b. dust/sludge c. slag	All data for EC: 1. 1 Mt a ⁻¹ Al & 5.8 Mt a ⁻¹ Fe ore, 4.2 kt a ⁻¹ Sn, 19 kt a ⁻¹ Cu & 176 kt a ⁻¹ Pb ore. 2. 132 Mt a ⁻¹ steel, 2.1 Mt a ⁻¹ Al, 1.3 Mt a ⁻¹ Cu, 1.3 Mt a ⁻¹ Pb, 5.6 kt a ⁻¹ Sn.	1. All ores between ~10 & ~1000 Bq kg ⁻¹ , except for pyrochlore (niobium production): 10 kBq kg ⁻¹ ²³⁸ U & 80 kBq kg ⁻¹ ²³² Th. 2. Bismuth metal and some tin alloys: 100-200 kBq kg ⁻¹ ²¹⁰ Po and 100-200 kBq kg ⁻¹ ²¹⁰ Po and ²¹⁰ Pb, respectively. 3. a. See ore. 3. b. ²¹⁰ Pb & ²¹⁰ Po : ~10 ² ~10 ⁵ Bq kg ⁻¹ . 3. c. ²³⁸ U and ²³² Th: ~100~1000 Bq kg ⁻¹ ; Nb-slag: 10 kBq kg ⁻¹ ²³⁸ U & 80 kBq kg ⁻¹ ²³² Th.	~10 μSv a ⁻¹ from single site Dominant radionuclide is ²³² Th and exposure is mainly due to inhalation of re-suspended dust on arable land. Special case: Cu-mine tailings in Mansfeld: 100-1000 μSv h ⁻¹ on local waste piles
PHOSPHATE INDUSTRY 1. Phosphate rock 2. Fertilisers (detergents, ...) 3. a. Phosphogypsum (PG) b. CaCl ₂ (CaF ₂) c. furnace slag and dust	1. P ₂ O ₅ production in EC: 2.23 Mt a ⁻¹ . 3. a. 1.3-1.7 t PG (t ore) ⁻¹ . 3. b. 90-100% of ore. 3. c. dust: 1% of ore; slag: 85% of ore.	1. 1500-1700 Bq kg ⁻¹ ²³⁸ U and ²²⁶ Ra & 10-200 Bq kg ⁻¹ ²³² Th in Moroccan ore. 2. phosphoric acid: 1200-1500 Bq kg ⁻¹ ²³⁸ U and 300 Bq kg ⁻¹ ²²⁶ Ra. 3. a. 80% Ra (<3000 Bq kg ⁻¹), 30% Th (10 Bq kg ⁻¹) and 14% U (500 Bq kg ⁻¹) in PG. 3. b. 100% of Ra (20 Bq L ⁻¹), U and Th, 80% of ²¹⁰ Pb. 3. c. furnace slag: 1000 Bq kg ⁻¹ ²³⁸ U and ²²⁶ Ra; dust 1000 Bq kg ⁻¹ ²¹⁰ Pb.	Collective dose from disposal site: 2.4 man-Sv from radon exposure, 2 man-Sv from groundwater contaminated with radium. Doses to member of critical group from ground and surface water contamination (major: ingestion of ²¹⁰ Po, daughter of ²²⁶ Ra) 0.02-1.1 mSv a ⁻¹ : depending on pathway

CATEGORY 1. Feed material 2. Product 3. Wastes and by-product	Scale of extraction, production and waste generation	Specific activities of principal radionuclides and enhancement factors	Principal pathways from wastes and dose to public
COAL MINING AND POWER PRODUCTION FROM COAL 1. Coal, lignite 3. a. bottom ash b. fly ash c. gypsum	1. EU total consumption of coal, lignite, peat was 300 Mt in 1992. 3. a,b 5-20% ash of which 25% bottom ash and 75% fly ash, 30 Mt a ⁻¹ . 3. c. 0-2 Mt per GWy.	1. Coal: 5-350 (20) Bq kg ⁻¹ ²³⁸ U and 5-230 (22) Bq kg ⁻¹ ²³² Th (Exception: Freital: 15000 Bq kg ⁻¹ ²³⁸ U). 3. a,b. enhancement factors ~10, specific act. 500 Bq kg ⁻¹ . 3. c. 20 Bq kg ⁻¹ ²²⁶ Ra & ²³² Th.	Re-suspension of ashes from disposal sites (²³⁰ Th, ²³² Th, ²¹⁰ Po, ²¹⁰ Pb): all pathways 250 μSv a ⁻¹ .
OIL AND GAS DRILLING 1. Natural oil and gas 2. Purified oil and gas 3. a. sludges b. scale	1. EU, 1993: Crude oil: 119 Mt; gas 2.5 EJ 3.a. in EU 10000 m ³ a ⁻¹ 3.b. 1 m ³ a ⁻¹ per plant	1. 1-70 Bq kg ⁻¹ ²³⁸ U and 6-70 Bq kg ⁻¹ ²³² Th 2. 337 (10-50000 Bq m ⁻³ ²²² Rn) 3.a. 5-80000 ²²⁶ Ra, 10-1.3*10 ⁶ ²¹⁰ Pb, 4-16000 ²¹⁰ Po, 500-50000 ²²⁸ Th (all conc. in Bq kg ⁻¹) 3.b. 200 ²²⁶ Ra, 50 ²¹⁰ Pb, 50 ²¹⁰ Po, 100 ²²⁸ Ra (in Bq kg ⁻¹)	No data on public exposure; little contact between public and industry. Main pathway will arise from disposal.
RARE EARTH & TITANIUM OXIDE INDUSTRIES 1. a. Rare earth bearing minerals 1. b. Titanium ores 2. a. Polishes, catalysts, special glass,... 2. b. Alloys, pigments, rubber,... 3. Waste	1. a. No rare earth mining in Europe. World production 30000 t a ⁻¹ . 1. b. Only 2 t of ~1 Mt titanium ore processed in Europe also extracted. 3. a. Almost no waste production. 3. b. Radium concentrates in scales and pigments	1. a. 1000 Bq kg ⁻¹ ²³⁸ U, ²¹⁰ Pb, ²²⁶ Ra, ²¹⁰ Pb; 6000-10000 Bq kg ⁻¹ ²³² Th, ²²⁸ Ra, ²²⁸ Th. 1. b. 30-700 Bq kg ⁻¹ ²³⁸ U, 30-6000 Bq kg ⁻¹ ²³² Th 2. a. glass: 1000 Bq kg ⁻¹ ²³⁸ U, ²²⁶ Ra, ²¹⁰ Pb; 3000 Bq kg ⁻¹ ²³² Th, ²²⁸ Ra, 5000 Bq kg ⁻¹ ²²⁸ Th. 2. b. Pigment: 4 ×10 ⁵ Bq kg ⁻¹ ²²⁶ Ra 3. a. Specific act. comparable with feeding material. 3. b. Scale: ~10 ⁶ Bq kg ⁻¹ ²²⁶ Ra and ²²⁸ Th.	a. ²³² Th and ²³⁸ U radionuclides of major concern. Individual doses from migration from landfill: 0.1 μSv a ⁻¹ due to different pathways. b. ~no data; BG-50 μSv h ⁻¹ at surface of piping & vessels of plant. a.b: overall, small risk.
ZIRCONIUM AND CERAMICS INDUSTRIES 1. Zircon, zirconia, clay 2. Refractory materials, glazes, glasses, ceramics 3. a. Chlorinator residue b. sludge	1. No mining in Europe: world prod. 70 kt a ⁻¹ ; import in Europe: 135 t a ⁻¹ .	1. Zircon: 400-40000 (600) Bq kg ⁻¹ ²³² Th and 200-74000 (3000) Bq kg ⁻¹ ²³⁸ U. Zirconia 300 Bq kg ⁻¹ ²³² Th and 3000 Bq kg ⁻¹ ²³⁸ U. 2. Glazes: 1000 Bq kg ⁻¹ ²³⁸ U, 20000 Bq kg ⁻¹ ²²⁶ Ra and ²¹⁰ Pb, 400 Bq kg ⁻¹ ²³² Th, 9000 Bq kg ⁻¹ ²²⁸ Ra, 5000 Bq kg ⁻¹ ²²⁸ Th. Refractories: 10000 Bq kg ⁻¹ ²³⁸ U, 10000 Bq kg ⁻¹ ²²⁶ Ra and ²¹⁰ Pb; 1000 Bq kg ⁻¹ ²³² Th, 2000 Bq kg ⁻¹ ²²⁸ Ra and ²²⁸ Th. 3. a. ~20 ×10 ⁶ Bq kg ⁻¹ ²²⁶ Ra. 3. b. ~10 ⁶ Bq kg ⁻¹ ²²⁶ Ra.	Overall small risk, given limited extent. Care is needed with the disposal of end products and some wastes since the exceed exemption limit

CATEGORY 1. Feed material 2. Product 3. Wastes and by-product	Scale of extraction, production and waste generation	Specific activities of principal radionuclides and enhancement factors	Principal pathways from wastes and dose to public
DISPOSAL OF BUILDING MATERIALS 1. a. Feedstock b. By-products/wastes 2. Bricks, concrete, cement	1. 15% phosphogypsum is recycled. 2. 20% slags used for concrete and cement production.	1. a. <100 Bq kg ⁻¹ ²³² Th and ²²⁶ Ra. 1. b. 100-1500 Bq kg ⁻¹ ²²⁶ Ra and <120 Bq kg ⁻¹ ²³² Th. 2. <100 Bq kg ⁻¹ ²³² Th and ²²⁶ Ra.	All material finally land-filled; low leachability of material. Major pathway from Rn and external radiation from ²³⁸ U: 1.4 and 3 μSv a ⁻¹ .
APPLICATIONS OF Ra & Th 1. a. U-ore b. monazite, thorite, thorianite 2. a. Luminising material b. gas mantles, alloy prod., ... 3. (old facilities, products)	1. a. 10 t U rich ore needed for production of 1 g radium; World production: 4.5 kg, half of it in Belgium between 1922 and 1969. 1. b. mainly from monazite (may contain 2.5-28% thorium oxide).	2. a. 400-400000 Bq kg ⁻¹ ²²⁶ Ra in soil surrounding radium luminising works. Hot spots 4 MBq kg ⁻¹ . 2. b. Gas mantles: 1000 kBq ²³² Th kg ⁻¹ ; thoriated glass: 400 kBq ²³² Th kg ⁻¹ ; thoriated tungsten: 100 kBq ²³² Th kg ⁻¹ ; titanium oxide: 50 kBq ²³² Th kg ⁻¹ .	2. a. Dominant radionuclides ²²⁶ Ra (int. ext. radiation) and ²²² Rn. Luminising works: Doses from 0.1-100 μSv h ⁻¹ . Radium extraction plant: disposal site: 0.1-1000 μSv h ⁻¹ ; surroundings: 0.1-100 μSv h ⁻¹ . 2. b. In States: annual collective dose is 86 man-Sv from 25 million gas mantles.

4.3 References

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5. GENERIC RADIOLOGICAL ASSESSMENT APPLIED TO A PHOSPHATE GYPSUM DUMP AS EXAMPLE CASE

5.1 Introduction

In order to assess the requirement for remediation at any particular site, and to quantify the benefit of intervention in terms of dose avoidance, it is necessary to establish the current site impact. This requires an adequate degree of site characterisation, together with suitably defined generic parameters. As an example to the approach, the abandoned phospho-gypsum dumps at Tessenderlo (Belgium) was modelled. This site was chosen since sufficiently detailed information could be collected, and, secondly, since the phosphate industry is one of the most important contaminating industries, due to NOR-levels in the ore and wastes, the extent of industry and its spread in Europe. Although for the different categories described in Chapter 4, the wastes originate from a number of different industrial processes, many of the features of these wastes are common to all sites. This has allowed a generic modelling approach to be developed (AMCARE) which may be applied on a site-by-site basis, if site-specific parameters are available. AMCARE (a flexible Assessment Model for CARE sites), as most generic models, may not be adequate for all sites. The generic results will only give a first impression of the radiological situation and these results should not be used for the actual decision making. This requires site specific models which have to be validated using actual measurements from the site. A detailed description of AMCARE is given in Annex B2.

For the example site considered here, site characterisation has been undertaken from a review of available information. Data obtained have then been used to assess the overall dose impact to members of the public under two scenarios:

- i) a 'normal evolution' case, where farmers are assumed to reside and work in close proximity to the site, representing a *status quo* in both the site and local population characteristics;
- ii) an intrusion case, where a hypothetical group is assumed to inhabit houses constructed on the contaminated site, in intimate contact with the main waste bearing area.

For both the intrusion and normal evolution scenarios, individual critical group doses are calculated and, for the normal evolution scenario only, a local collective dose is calculated for a population within a specified radial area of the site, including the critical group. Further details of the collective group assumptions are presented in Annex B2. As for the critical group, the larger collective dose group exhibits habits considered typical across Europe.

An additional 'site workforce' dose is also calculated. Members of this group are assumed to work on the site and to reside locally, but derive most of their food from non-local sources. The work undertaken by this group is not specified (e.g. whether operational, remediation or other construction work). Where comparison of remediation options is undertaken (see Chapter 6), the workforce habits are specific to each approach.

A further illustrative assessment for the Tessenderlo phosphate dump was undertaken using AMCARE to determine the relative ranking of available remediation options which may be considered in the restoration of sites which differ substantially in physical characteristics and types of waste.

Results for the assessment are presented below, together with generic and site specific parameters. Since for this common approach to restoration, generic recommendations on remediation approaches are required, undue site specificity in modelling impacts is avoided and ranges in parameters are incorporated. The presence of an existing, temporary, cap at Tessenderlo is incorporated in the dose estimates.

The dominant dose to all exposure groups at Tessenderlo arises from inhalation of radon (^{222}Rn) gas, which is supported by the parent inventory of ^{226}Ra . The dose to the local off-site critical group of farmers is estimated to be around $1.42 \times 10^{-4} \text{ Sv a}^{-1}$ per $\text{Bq } ^{226}\text{Ra kg}^{-1}$ in the waste material or 0.5 mSv a^{-1} overall. The only other pathway of significance arises from external irradiation of workers on the site, as a result of close proximity to the waste-bearing area. Actions to reduce the rate of radon emanation or, for workers, to enhance shielding may thus be supposed, *a priori*, to represent the approaches most likely to deliver cost effective dose reduction and site remediation measures.

With respect to the hypothetical intrusion scenario, much higher doses may be incurred where the existing cap is assumed to be penetrated, with a modal value of 365 mSv a^{-1} for the best estimate inventory. Where the cap is assumed to remain undisturbed, the intrusion dose declines to 25 mSv a^{-1} . In either case, radon inhalation is again the dominant pathway, although both ingestion doses and external irradiation potentially deliver doses in excess of 1 mSv a^{-1} . This might imply that, for the period post institutional control of the site, a range of remediation options would be considered which could not be justified for the off-site exposure group alone.

The main factors determining uncertainty in dose estimates, other than the waste inventory itself, relate to the rate of emission of radon gas to atmosphere and, to a lesser extent, to shielding from external irradiation.

5.2 Summary description of AMCARE - the assessment model for CARE

A number of approaches to modelling the dose impact of contaminated areas and waste disposal sites are possible, each of which may be equally valid and which may have specific advantages for particular types of sites. An overview of models currently in use for such assessments is presented in Annex B1. For the purposes of a comparative assessment of impacts and remediation effects, a more generic approach is followed here. A detailed description and conceptual diagram of the model developed (AMCARE - Assessment Model for a Common Approach to REstoration of contaminated sites) is presented in Annex B2, and summarised briefly below.

In essence, AMCARE is a flexible assessment tool, to determine the relative ranking of available remediation options which may be considered in the restoration of sites which differ substantially in physical characteristics and types of waste. Since generic recommendations on remediation approaches are required, undue site specificity in modelling impacts is avoided. Thus, a site may have a low impact solely because there is no local critical group of residents, whilst a similar site situated in a densely populated area may have a considerable impact. Consequently, a reasonably uniform approach to modelling pathways of concern is incorporated, illustrated in Figure 5.1.

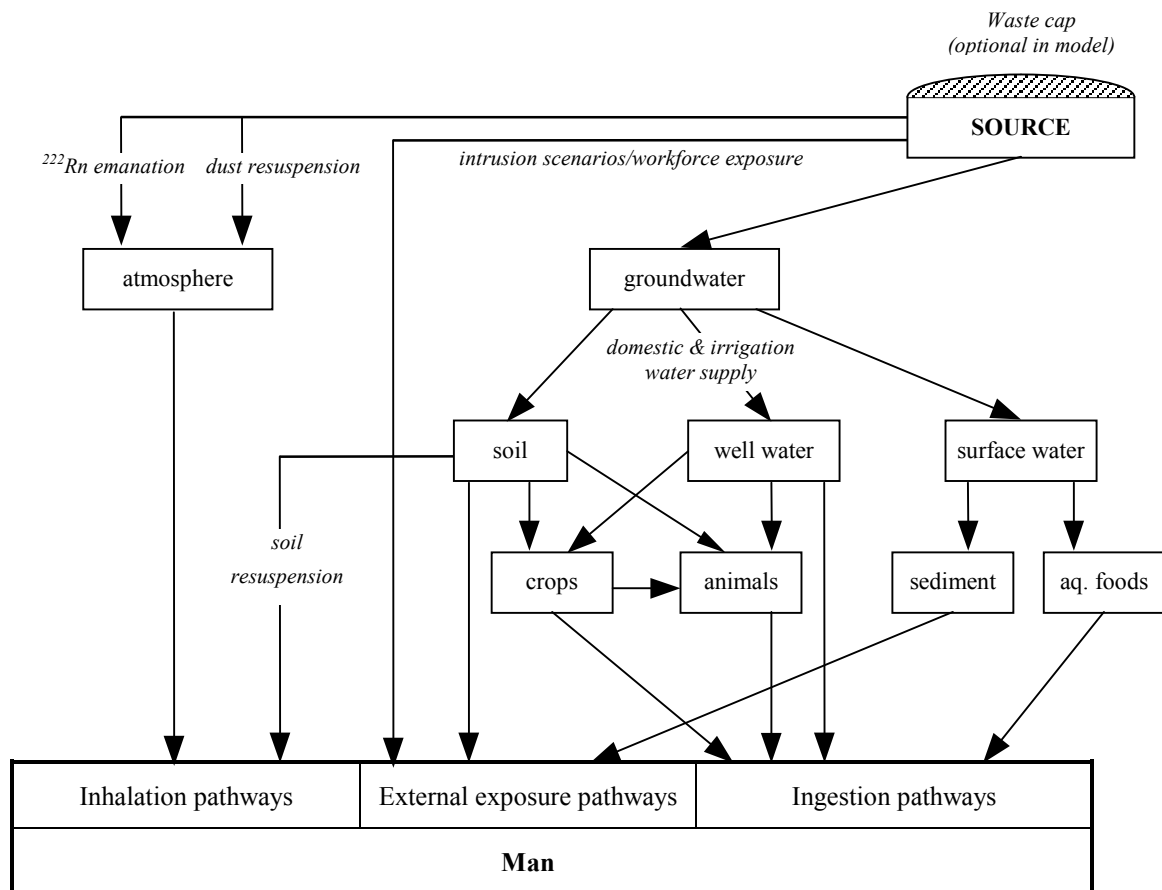


Figure 5.1 Exposure pathways from contaminated waste to man incorporated in AMCARE

The principal pathways of exposure are considered to be via ingestion of food and water, inhalation of radon or resuspended dust particles from the waste area and external exposure to the waste or contaminated soils or sediments. It is clear that many more pathways could be included, such as the use of surface fresh-water for irrigation, accidental ingestion of soil by man, resuspension of riverine bank sediment and subsequent inhalation by man, etc. In principal, these pathways exist within AMCARE, but for simplicity are not included for consideration here. Furthermore, a number of generic assumptions are introduced regarding population habits and proximity to sites, in order to allow a common approach to comparing differing remediation options. Again, these assumptions can be varied within AMCARE if required. In any case, AMCARE has been run here to incorporate a number of the most important site specific features (e.g. type of waste, radionuclide inventory, waste disposal area/volume, proximity to nearest surface water, etc.).

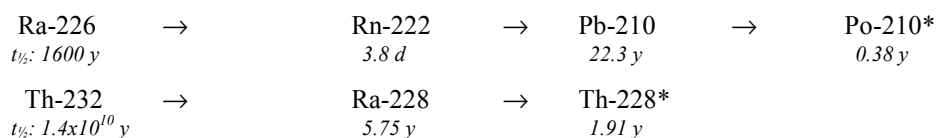
Details of the site and nuclide specific parameters assumed here, and the generic values used to provide a commonality between sites in assessing remediation options, are summarised in Section 5.3, following. Since many of the parameter values are subject to considerable uncertainty, based on literature reviews of best estimate values and associated ranges, a range on dose assessments is presented in Section 5.4 together with best estimate values.

5.3 Assessment for Tessenderlo

5.3.1 Site description

The Tessenderlo phosphate processing site in Belgium is operated by Tessenderlo Chemie and has been described in general terms in Annex A2. Since the facility is still in operation, the wastes should not be considered in an intervention framework. But since the disposal site is

abandoned and just for the sake of analysing, assessing and discussing a well-characterised site we made abstraction of this. As a result of past practices, a volume of about $9 \times 10^6 \text{ m}^3$ sludge has been disposed of, containing primarily ^{226}Ra and ^{232}Th . By decay, this also gives rise to radon gas, lead and polonium:



* Short lived members of the decay chains omitted.

The waste disposal area has been capped with soil. This covering is assumed to be temporary and is included as part of the ‘no treatment’ scenario, except in one specific instance (housing intrusion, Section 5.3.2.2) where this covering is assumed to be breached. A river flows approximately 50 m from the nearest site boundary.

5.3.2 Model parameters

5.3.2.1 Site specific and nuclide specific parameters

Reasonably detailed information exists to characterise the Tessenderlo site with respect to size, prevailing meteorological conditions and waste deposits. These parameters are summarised in Table 5.1.

Table 5.1 Site characterisation data for Tessenderlo

Wind velocity		$1.4 \times 10^8 \text{ m a}^{-1}$
Ground water	Cross sectional area	$2.6 \times 10^3 \text{ m}^2$
	Flow rate	10 m a^{-1}
Discharge compartment	Surface area	$5.3 \times 10^5 \text{ m}^2$
	Volume	$9.0 \times 10^6 \text{ m}^3$
	Depth	17 m
	Barrier thickness	1.5 m

Based on the total waste volume presented in Table 5.1, site radionuclide inventory estimates are presented in Table 5.2 encompassing a range on uncertainty around the best estimate values. A waste density of $1500 \text{ kg dry weight m}^{-3}$ is assumed (see Section 5.3.2.2).

Table 5.2 Summary of waste inventory for Tessenderlo

Nuclide	Best estimate	Lower estimate	Upper estimate
	<u>Reported concentration in waste (Bq kg^{-1})</u>		
^{226}Ra	3500	2100	4100
^{232}Th	40	33	47
	<u>Estimated total inventory (TBq)</u>		
^{226}Ra	47	28	55
^{232}Th	0.54	0.45	0.63

Estimating the inventory using the concentration will give rise to larger than expected uncertainties in the estimate for the inventory. A better approach would have been to estimate the inventory from the arisings. However, this information was not available and it was necessary to assume that the waste is both chemically and physically homogenous.

Nuclide specific distribution coefficients (K_d 's) for radium, thorium and daughter products are listed in Table 5.3. From the range of data tabulated the inherent uncertainty for this parameter is obvious. It is assumed that soil on and around the Tessenderlo site is primarily sandy, although it is known that the discharge is of a silty nature. This assumption is likely to be

conservative, in that the elements considered here will generally leach more freely through sandy soils than through clay layers or organic soils. However, the chemical, structural and physical composition of any spoil heap or waste disposal site is likely to be complex and substantially modified by comparison to the prevailing natural soil/rock types. It is also likely to be substantially fractured, unless it has been previously treated (see Chapter 6), which will tend to enhance flow rates of infiltrating water. Further site specific refinements to the impact assessments may be introduced, however, where local soils, or the waste material itself, are well characterised or where site specific K_d 's have been derived.

Table 5.3 Nuclide specific distribution coefficients and half lives

Nuclide	Half life (years)	Gross average distribution coefficient (K_d) values ($m^3 kg^{-1}$)			
		sandy soils		aqueous system sediments	
		best estimate	range	best estimate	range
Po-210	0.38	0.15	0.006 - 3.6	0.15	0.006 - 3.6
Pb-210	22.3	0.27	0.0027 - 27	0.27	0.0027 - 27
Ra-226	1600	0.49	0.00082 - 300	0.5	0.1 - 1
Ra-228	5.75	0.49	0.00082 - 300	0.5	0.1 - 1
Th-228	1.91	3.0	0.045 - 200	10	1 - 1000
Th-232	1.41×10^{10}	3.0	0.045 - 200	10	1 - 1000

Source: IAEA-IUR (1994). Data for aqueous systems assume oxidising conditions. No data for Po or Pb in aqueous systems are available and the value for sandy soils is assumed to apply for both systems.

The Tessenderlo waste disposal site is bounded by farmland, and the distances to the nearest water body, agricultural holdings and local population etc. are summarised in Table 5.4.

Table 5.4 Proximity of surface water bodies, agricultural land and local population data

Nearest agricultural land	Distance from tailings*	100 m
	Surface area*	20000 m ²
	Depth*	0.3 m
	Infiltration rate	0.1 m a ⁻¹
Nearest river	Distance from tailings	50 m
	Surface area*	3000 m ²
	Depth*	2 m
	Exchange/flow rate of water	$1.8 \times 10^7 m^3 a^{-1}$
Area affected	20 km radius from site	1257 km ²
Population affected	Population density c.150 persons km ⁻²	200000 persons

Note: * No site-specific information available, therefore 'most reasonable' estimates are presented.

In the case of Tessenderlo, the nearest surface water body is a river flowing near to the site. For coastal sites, a marine compartment within AMCARE can be employed instead.

5.3.2.2 Generic assumptions common to all CARE sites

a) Physical constants, animal intake rates and emanation/diffusion coefficients

A large number of parameters incorporated within the AMCARE database are assumed to be common to all sites under consideration. In principle, most of these parameters are user-definable, such that a much greater degree of site specificity can be introduced where required. At a practical level, however, a number of the parameters are unlikely to be available on a site specific basis and generic values incorporating a range on uncertainty may provide the best substantiated approach. In other cases, common assumptions are introduced specifically to allow comparison of results between sites. Table 5.5 lists a number of parameters which may be defined as physical constants (e.g. density of water) or which are assumed to be constant for the purposes of generic assessment (e.g. dust loading in air). It will be noted from the physical

characteristics of the surface cap (which is optional on a site-specific basis) that the covering material is essentially soil or a soil/clay mix.

Table 5.5 Physical constants and assumed rate coefficients common to all sites

Soil characteristics	
Density of water	1000 kg m ⁻³
Density of dry soil*	1500 kg m ⁻³ typical for sandy soils
Porosity	0.3 typical for sandy soils
Radon emanation fraction	0.23
Radon diffusion coefficient**	15.8 m ² a ⁻¹
Waste cap characteristics (if present)	
Density of cap material*	1500 kg m ⁻³ suitable for a range of materials
Radon diffusion coefficient**	15.8 m ² a ⁻¹
External exposure	
Dust loading in air [†]	1.0 × 10 ⁻⁷ kg m ⁻³
Shielding factor (outdoor) [‡]	0.75
Indoor exposure	
Area of room [†]	100 m ²
Ventilation rate [†]	6.6 × 10 ⁵ m ³ a ⁻¹ total turnover in 4 hours
Shielding factor (indoor) [‡]	0.25

Source: * UNSCEAR (1993), IAEA-IUR (1994); ** Evans (1997); [†] best estimate, no reference data available; [‡] Oztunali et al. (1981).

The short term dynamics of radionuclide transport through agricultural foodchain pathways are of little concern for waste disposal sites, since dose implications assessed for local exposure groups are generally over time periods greatly in excess of any growing season. Thus, within CARE reference time periods of 100 years, 500 years and 10,000 years are considered. Accordingly, the simplest approach to modelling the uptake of activity through the environment is to assume that equilibrium is established rapidly between radionuclide concentrations in soils, plants and animal products. Transfer factors are presented in Tables 5.6 to 5.8 as soil to plant concentration ratios (CR_{ag}), uptake to meat (F_f) and milk (F_m) and water to aquatic organism concentration ratios (CR_{aq}).

Table 5.6 Transfer factors pasture grass and agricultural crops

Element	Transfer factors (and ranges) for pasture grass and agricultural crops Bq kg ⁻¹ (dry plant) per Bq kg ⁻¹ (dry soil)			
	Grass	Leafy vegetables	Roots vegetables	Grain
Po	9.0 × 10 ⁻² (9.0 × 10 ⁻³ - 9 × 10 ⁻²)	1.2 × 10 ⁻³ (1.2 × 10 ⁻⁴ - 1.2 × 10 ⁻³)	7.0 × 10 ⁻³ (7.0 × 10 ⁻⁴ - 7.0 × 10 ⁻³)	2.3 × 10 ⁻³ (2.3 × 10 ⁻⁴ - 2.3 × 10 ⁻³)
Pb	1.1 × 10 ⁻³ (1.1 × 10 ⁻⁴ - 1.1 × 10 ⁻²)	1.0 × 10 ⁻² (5 × 10 ⁻⁴ - 2 × 10 ⁻¹)	2.0 × 10 ⁻² (2.0 × 10 ⁻⁴ - 2.0 × 10 ⁻¹)	4.7 × 10 ⁻³ (4.7 × 10 ⁻⁴ - 4.7 × 10 ⁻²)
Ra	8.0 × 10 ⁻² (1.6 × 10 ⁻² - 4.0 × 10 ⁻¹)	4.9 × 10 ⁻² (2.5 × 10 ⁻³ - 9.8 × 10 ⁻¹)	1.1 × 10 ⁻³ (2.2 × 10 ⁻⁴ - 1.1 × 10 ⁻¹)	1.2 × 10 ⁻³ (2.4 × 10 ⁻⁴ - 6.0 × 10 ⁻³)
Th	1.1 × 10 ⁻² (1.1 × 10 ⁻³ - 1.1 × 10 ⁻¹)	1.8 × 10 ⁻³ (4.5 × 10 ⁻⁵ - 7.2 × 10 ⁻²)	5.6 × 10 ⁻⁵ (5.6 × 10 ⁻⁶ - 3.9 × 10 ⁻¹)	3.4 × 10 ⁻⁵ (3.4 × 10 ⁻⁶ - 8.5 × 10 ⁻⁴)

Note: Best estimates and ranges are derived from Jackson (1984) and IAEA-IUR (1994). A default range of ± one order of magnitude is assumed where no other information is available. Values for Po were derived from field data and stated not to be corrected for foliar interception. No range on values is presented by IAEA-IUR (1994) but it is indicated that actual values may be 2-10 times lower than the best estimate. A range is assumed accordingly here.

Soil to plant concentration ratios will vary between plant/crop species, with soil type and with meteorological conditions (especially rainfall). Even within crop types, or within species of pasture grass, considerable variability in uptake of radionuclides may be evident. Values presented in Table 5.6 are taken to be broadly applicable across a range of conditions.

Similar variability in uptake of radionuclides to agricultural animal products and aquatic organisms may also be evident. The values presented in Tables 5.7 and 5.8 are regarded as broadly representative for temperate environments and are therefore applicable to most European locations.

Table 5.7 *Transfer factors for animal foodstuffs*

Element	Transfer factors (and ranges) for animal foodstuffs Bq kg ⁻¹ or L ⁻¹ (animal foodstuff) per Bq kg ⁻¹ (dry food) per kg day ⁻¹ (dry food intake)		
	<i>Beef</i>	<i>Milk</i>	<i>Pork</i>
Po	5.0 × 10 ⁻³ (6 × 10 ⁻⁴ - 5 × 10 ⁻³)	3.4 × 10 ⁻⁴ (n/a)	2.5 × 10 ⁻² (n/a)
Pb	4.0 × 10 ⁻⁴ (1 × 10 ⁻⁴ - 7 × 10 ⁻⁴)	3.4 × 10 ⁻⁴ (n/a)	2.0 × 10 ⁻³ (n/a)
Ra	9.0 × 10 ⁻⁴ (5 × 10 ⁻⁴ - 5 × 10 ⁻³)	1.3 × 10 ⁻³ (1 × 10 ⁻⁴ - 1.3 × 10 ⁻³)	4.0 × 10 ⁻³ (n/a)
Th	2.7 × 10 ⁻³ (n/a)	5.0 × 10 ⁻⁴ (n/a)	2.0 × 10 ⁻² (n/a)

Source: IAEA-IUR (1994). The data are sparse in some cases, especially for swine. A range of values of ×10 around the mean may be assumed where no data are available.

Table 5.8 *Transfer factors for aquatic foodstuffs*

Element	Transfer factors (and ranges) for aquatic foodstuffs Bq kg ⁻¹ per Bq m ⁻³ (water)			
	<i>Freshwater fish</i>	<i>Marine fish</i>	<i>Crustacea</i>	<i>Molluscs</i>
Po	0.05 (0.01 - 0.5)	2.0 (n/a)	50.0 (n/a)	10.0 (n/a)
Pb	0.30 (0.1 - 0.3)	0.20 (n/a)	1.00 (n/a)	1.00 (n/a)
Ra	0.05 (0.01 - 0.2)	0.50 (n/a)	0.10 (n/a)	1.00 (n/a)
Th	0.10 (0.01 - 10)	0.60 (n/a)	1.00 (n/a)	1.00 (n/a)

Note: Transfer factors and ranges for freshwater fish derived from IAEA-IUR (1994). Transfer factors for marine organisms derived from stable element concentrations in Bowen (1966). The data are sparse in some cases and a range of values of ×10 around the mean may be assumed where no data are available.

Intake rates of water and foodstuffs by animals will clearly vary during the animal's lifetime and between breeds. Lower milk yield cattle will generally require less water than higher milk yield breeds. Data presented in Table 5.9 are considered broadly applicable to the types of animals, and sizes at slaughter where relevant, likely to be reared across Europe. Local and national differences may be pronounced. In the UK, relatively few farms maintain free range pigs, whereas sheep are widespread, particularly on upland areas. AMCARE can be modified appropriately but, for simplicity, only uptakes through bovine and porcine pathways are considered here.

Table 5.9 Typical annual intakes by animals

Consumption	Dairy cattle (milk yield: 10-20 L d ⁻¹)		Beef cattle (500 kg)		Swine (110 kg)	
	best estimate	range	best estimate	range	best estimate	Range
Water m ³ d ⁻¹	0.075	0.050-0.100	0.040	0.020-0.060	0.008	0.006-0.010
Soil* kg d ⁻¹	0.64	0.4-1.0	0.3	0.2-0.4	0.48	0.4-0.6
cereal kg d ⁻¹ dw	0	0	0	0	2.4	2.0-3.0
Grass kg d ⁻¹ dw	16.1	10-25	7.2	5-10	0	0

Source: IAEA-IUR (1994). * Soil consumption represents 4% of fodder intake by dry weight for cattle and 20% for swine - no reference values are available.

Other agricultural conditions and practices are hard to define as ‘typical’ across Europe. Crop types vary with growing region and, in particular, cereal crops are not grown extensively in much of northern Europe. Requirements for irrigation also vary markedly. For the purposes of this comparative study, a number of simplifying assumptions are made. Primarily, it is assumed that agriculture local to each site produces sufficient cereal, root crops and leafy green vegetables to support the resident population, and that irrigation of agricultural fields is undertaken at a constant rate. Relevant parameters are defined in Table 5.10.

Table 5.10 Assumed agricultural constants for arable crops

Water content of root vegetables*	0.8 (potato)	} as fraction of total weight
Water content of cereals*	0.3 (average for maize and other cereals)	
Water content of leafy vegetables*	0.9	
Irrigation rate**	0.01 m ³ m ⁻² d ⁻¹	
Days of irrigation**	100 d a ⁻¹	

Source: * IAEA (1994); ** BIOMOV5 (1996).

b) Local population habits

For the purposes of establishing common points of comparison between sites and between remediation options, local exposure groups are considered to be similar at all sites. The local residency farming group is assumed to spend 1600 hours per year on the agricultural land nearest to the waste site (all of which time is outdoors), exposed to dust and radon, and to spend a further 250 hours each year fishing, exposed to river sediments. The rest of their time they live at the nearest identified habitations to the site, where they are exposed to radon and spend 67% of their time indoors. All of their agricultural food is derived from the field on which they work. Fish in the diet is derived from the local river, and all drinking water is supplied from a well sited on the field which also supplies food. These assumptions are clearly conservative and, in reality, would almost certainly be insupportable. For Tessenderlo, the nearest field is 100 m from the waste site and the nearest point of habitation is 1 km away.

A second exposure group is considered to consist of individuals who, at some time in the future, reside in dwellings constructed on the waste site. During construction, the house foundations are assumed to penetrate any layers covering the waste material and the residents are assumed to spend all of their time on the waste site, of which 67% of time is spent indoors. All ‘agricultural’ food is assumed to be grown on the spoil heap and drinking water is drawn from a well which penetrates the waste matrix. Fish are consumed from the nearest local watercourse (or coastal waters), but no time is spent off-site engaged in fishing activities etc.

A critical group of workers on the waste disposal site is also identified. The work undertaken by this group is not defined explicitly (i.e. whether operational, introducing remediation measures or undertaking other construction work), but they are assumed to spend 1600 hours per year on the spoil heap, of which 33% is outdoors. In this case, time spent ‘indoors’ encompasses time spent within buildings of whatever type, and time spent inside vehicle cabs. Whilst on the site, they are exposed to dust (outdoors only) and radon (which is assumed to be

the same indoors and outdoors). They are assumed to reside 1 km away, where they are exposed to dispersed radon. It is assumed that the workforce group, employed at the site, does not overlap with the local residency farming group (i.e. they do not also work part time on the local fields) and they derive only 25% of their food from the field 100 m away. Members of the workforce group do not eat local fish and derive their drinking water from a distant location (i.e. non-contaminated).

These characteristics are summarised in Table 5.11. As noted in Section 5.1, where comparisons between remediation options are undertaken (see Chapter 6), remediation-specific workforce habits are defined.

Table 5.11 Summary of critical group characteristics

	Local critical group	Intrusion	Workforce
Habitation	village 1km away	house on waste	village 1 km away
Time spent at habitation (h a ⁻¹)	6190	8760	7160
Outdoor occupancy at habitation (%)	33%	33%	33%
Workplace	field 100 m away	field on waste	waste
Time spent at work (h a ⁻¹)	1600	N/A	1600
Outdoor occupancy at workplace (%)	100%	N/A	33%
Terrestrial foodstuff source	field 100 m away	field on waste	field 100 m away
Local food intake (% of total food intake)	100%	100%	25%
Aquatic foodstuff source	nearest surface water	nearest surface water	none
Time spent fishing (h a ⁻¹)	250	none	none
Drinking water source	well 100 m away	well on waste	none
Irrigation water source	well 100 m away	well on waste	N/A
Animal drinking water source	well 100 m away	well on waste	N/A

In each case, consumption and inhalation rates for members of the exposure groups are taken to be typical for European adults (Table 5.12). Locally, critical groups may include children and may cover pathways not considered here. However, for comparative purposes these assumptions are considered appropriate and, in view of the inherent conservatism elsewhere, it is unlikely that the assumptions here will under-estimate the actual doses incurred around each of the sites.

Table 5.12 Typical population ingestion and inhalation habits assumed

Habits pathway		
Ingestion rates:	Drinking water	0.6 m ³ a ⁻¹
	Roots	81 kg a ⁻¹
	Cereals	84 kg a ⁻¹
	Leafy vegetables	110 kg a ⁻¹
	Beef	23 kg a ⁻¹
	Milk	124 kg a ⁻¹
	Pork	37 kg a ⁻¹
	Fish (fw or sw)	13 kg a ⁻¹
	Shellfish	0.6 kg a ⁻¹
Inhalation rate:		1 m ³ hr ⁻¹

Source: Simmonds et al. (1995).

c) Dose factors

The most recent dosimetric advice of the ICRP (1996) is followed with respect to dose per unit intake values (Table 5.13). Doses derived from cloud immersion or external exposure from

waste bearing materials are derived principally from the CEC-sponsored methodology for assessing the radiological consequences of releases of radionuclides to the environment [Simmonds *et al.*, 1995], as implemented in PC CREAM [Mayall *et al.*, 1997].

Table 5.13 Dose rate factors for adults

Nuclide	Inhalation (Sv Bq ⁻¹)	Ingestion (Sv Bq ⁻¹)	External (Sv a ⁻¹ per Bq m ⁻³)
²¹⁰ Po	3.30 × 10 ⁻⁶	1.20 × 10 ⁻⁶	1.47 × 10 ⁻¹⁴
²¹⁰ Pb	1.10 × 10 ⁻⁶	6.90 × 10 ⁻⁷	2.09 × 10 ⁻¹²
²²⁶ Ra	3.50 × 10 ⁻⁶	2.80 × 10 ⁻⁷	3.20 × 10 ⁻⁹
²²⁸ Ra	2.60 × 10 ⁻⁶	6.90 × 10 ⁻⁷	8.00 × 10 ⁻¹⁰
²²⁸ Th	4.00 × 10 ⁻⁵	7.20 × 10 ⁻⁸	4.90 × 10 ⁻¹⁰
²³² Th	2.50 × 10 ⁻⁵	2.30 × 10 ⁻⁷	5.90 × 10 ⁻¹³

Note: Inhalation and ingestion dose per unit intake factors from ICRP Publication 72 (1996). External dose factors derived from various sources (principally Mayall *et al.*, 1997) as CED. ²²²Rn inhalation doses are calculated using a factor of 5 × 10⁻⁵ Sv a⁻¹ per Bq m⁻³.

5.3.3 Local residency critical group exposure

The critical group of residential farmers on the periphery of the waste site (defined in Section 5.3.2.2) will be exposed to varying levels of radiation over time as material migrates away from the site in groundwater, as radon diffuses through the waste into atmosphere and as the radionuclides in the waste matrix decay and disperse. In practice, since the parent nuclides at Tessengerlo have long half-lives, the major factor in determining changes in exposure patterns over time will be the rate of migration of radionuclides in groundwater. Consequently, for this group, exposure pathways are assessed over the short term (i.e. within 500 years) and up to 10000 years in the future. These doses are expressed as ‘current’ and ‘peak’ exposures in Table 5.14. The peak exposure is simply the maximal dose estimated from all pathways combined which occurs at any time over the next 10000 years.

Table 5.14 Dose to local residents, calculated by AMCARE, for pathway and nuclide (best estimate inventory)

Radionuclide	Dose (mSv a ⁻¹)							
	External irradiation		Total (dust + radon) inhalation		Ingestion		Total	
	current	peak	current	peak	current	peak	current	peak
Radium and daughter nuclides								
²²⁶ Ra	7.14E-26	5.30E-07	3.20E-35	4.51E-10	2.53E-25	3.06E-07	3.24E-25	8.37E-07
²²² Rn	-	-	5.00E-1	5.00E-01	-	-	5.00E-1	5.00E-01
²¹⁰ Pb	5.64E-27	1.29E-11	6.50E-35	4.63E-13	8.38E-22	1.17E-07	8.38E-22	1.17E-07
²¹⁰ Po	4.16E-29	7.59E-15	6.65E-34	4.98E-12	4.58E-22	5.61E-08	4.58E-22	5.61E-08
Thorium and daughter nuclides								
²³² Th	5.44E-63	1.88E-18	5.91E-71	1.85E-18	8.58E-60	2.90E-15	8.58E-60	2.90E-15
²²⁸ Ra	3.00E-49	8.04E-16	3.25E-62	2.43E-22	1.06E-47	2.81E-14	1.08E-47	2.89E-14
²²⁸ Th	6.01E-49	1.61E-15	2.71E-62	2.03E-22	3.58E-49	9.57E-16	9.58E-49	2.57E-15
Total	7.70E-26	5.30E-07	5.00E-1	5.00E-01	1.30E-21	4.80E-07	5.00E-1	5.00E-01

Table 5.14 shows that doses, calculated by AMCARE, for a number of radionuclides and pathways are extremely small. These are considered to be too small to be meaningful and thus be disregarded.

A summary of estimated doses, assuming a range on estimates of the waste inventory, is presented in Table 5.15.

Table 5.15 Summary of doses, calculated by AMCARE, to local residents (upper and lower estimate inventory)

		Dose incurred by pathway (mSv a ⁻¹)				
		External exposure	Radon inhalation	Dust inhalation	Ingestion	Total
Best estimate inventory	current	7.70E-26	5.00E-1	7.62E-34	1.30E-21	5.00E-1
	peak	5.30E-7	5.00E-1	4.56E-10	4.80E-7	5.00E-1
Upper estimate inventory	current	9.02E-26	5.86E-1	8.93E-34	1.52E-21	5.86E-1
	peak	6.21E-7	5.85E-1	5.34E-10	5.62E-7	5.86E-1
Lower estimate Inventory	current	4.62E-26	3.00E-1	4.57E-34	7.78E-22	3.00E-1
	peak	3.18E-7	3.00E-1	2.74E-10	2.88E-7	3.00E-1

A dose for the best estimate inventory at Tessenderlo of around 0.5 mSv a⁻¹ (500 μSv a⁻¹) is predicted, largely determined by the radon inhalation pathway. The important difference in radon dose and dose due to dust inhalation can be explained by the fact that for the local critical group the dust inhalation pathway is derived from the resuspension of the agricultural soil on the field during time spent on the field. Radionuclide concentrations in the field are low because they get accumulated there via the groundwater. The dose from other pathways increases drastically over time as groundwater becomes contaminated and migrates to the field distance assumed for agricultural produce and drinking water. This determines both internal (ingestion and dust inhalation) pathways and external irradiation. However, these (modelled) pathways do not become significant by comparison to radon inhalation, at least over 10000 years (Table 5.15).

Considerable uncertainty attaches to the central dose estimate, due to potential variability around many of the best estimate parameter values assumed. Using Crystal Ball (a commercially available probabilistic modelling tool, Decisioneering Inc. (1996)) as a post-processor to AMCARE (see Annex B2), a range of dose estimates can be obtained. Ranges on parameter estimates are, in each case, assumed to have a triangular distribution, with the best estimate as the modal value within the distribution. On this basis, Figure 5.2 summarises the distribution of current dose estimates (i.e. relevant to conditions within 500 years) for the local residency critical group at Tessenderlo, when each parameter is allowed to vary randomly over the range described in Sections 5.3.2.1 and 5.3.2.2. The dose distribution described applies to the best estimate inventory scenario only.

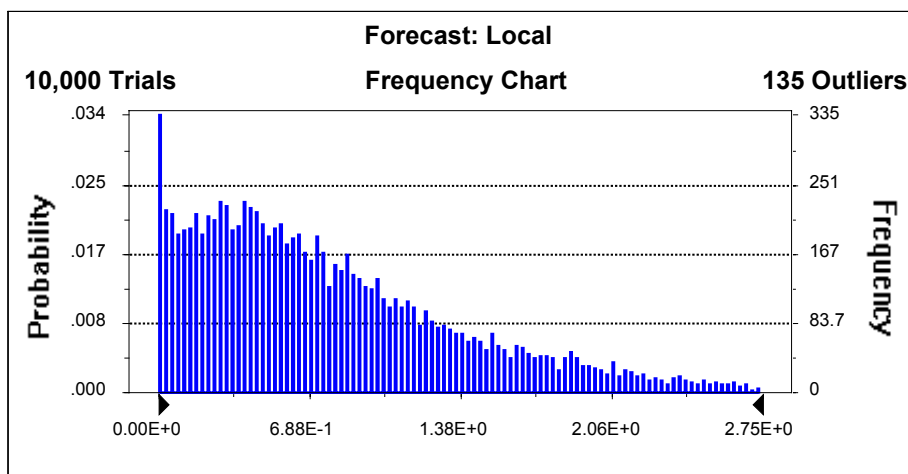


Figure 5.2 Probabilistic range on dose estimates to local residents (best estimate inventory)

While an annual dose of 0.5 mSv accurately describes the modal dose estimate, there is a highly skewed distribution from about 0.01 mSv a⁻¹ to 2.75 mSv a⁻¹. The upper and lower 2.5th centile values for dose are excluded from Figure 5.2 (which comprise the 135 outliers). A tabulation of the centile dose estimate values is presented in Table 5.16.

Table 5.16 Centile dose estimates, calculated by AMCARE, to local residents (best estimate inventory)

Percentile	Value
0.0%	9.75E-10
2.5%	1.78E-02
5.0%	5.15E-02
50.0%	6.53E-01
95.0%	2.16E+00
97.5%	2.45E+00
100.0%	4.48E+00

A sensitivity analysis of the pathways and parameters contributing to the local residency group dose indicates that the cap diffusion coefficient assumed (which determines the rate of radon evolution from the waste matrix to atmosphere) is the single most important factor (Figure 5.3). In turn, the rate of radon emanation from the waste and the cap thickness also influence the dose. Allowing for ±10% variability on the radon dose factor influences the dose potentially incurred in an almost linear fashion. This emphasises the dominant dose contribution from radon inhalation.

Factors affecting other pathways can be seen from Figure 5.3 to exert a much smaller influence on total dose. It should be noted that the ranges for K_d (presented in Table 5.3) and transfer factors (presented in Tables 5.6 to 5.8) exceed, in some cases, five orders of magnitude, indicating that very large variations in these parameters are of relatively minor consequence at Tessengerlo.

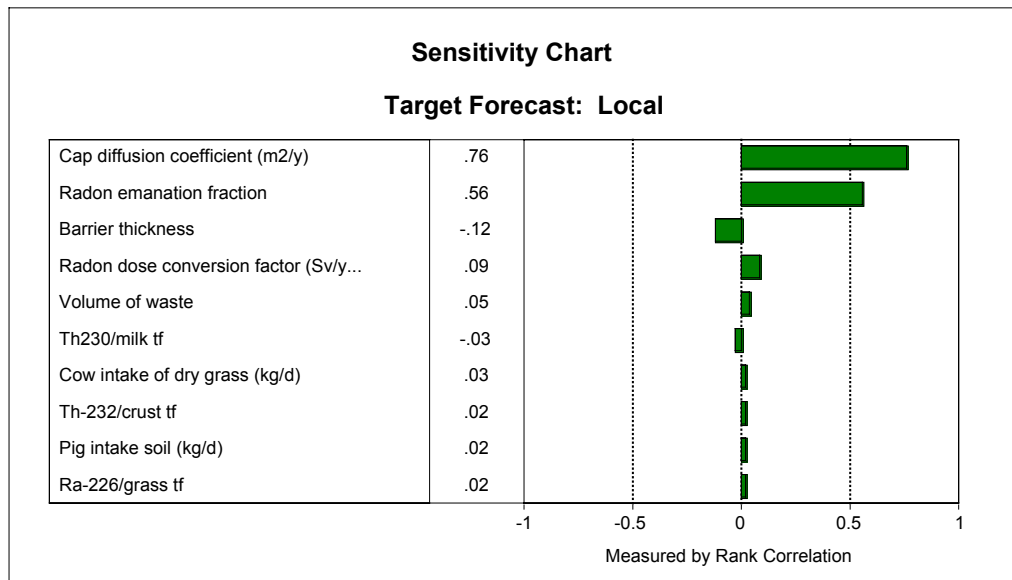


Figure 5.3 Key parameters determining dose to local residents (best estimate inventory)

In comparing dose estimates obtained for Tessengerlo with doses derived for other sites, the total waste inventories may be the main differentiating factor. As a simplified approach to comparing the relative impact of differing sites, a rough approximation of dose incurred per unit concentration of radionuclide *i* in the waste can be derived ($G_{i,j}$), such that:

$$G_{ij} = (\sum \text{dose}_{i,j} + \text{dose}_{\text{idtr}1,j} + \text{dose}_{\text{idtr}2,j} + \dots) / \text{concentration}_i$$

where: $\text{dose}_{i,j}$ is the total dose from all pathways (mSv a^{-1}) due to radionuclide i at location j , defined by distance from the site;

$\text{dose}_{\text{idtr}1,j}$ is the total dose from all pathways (mSv a^{-1}) due to the daughter nuclide 1 of radionuclide i at location j ;

$\text{dose}_{\text{idtr}2,j}$ is the total dose from all pathways (mSv a^{-1}) due to the daughter nuclide 2 of radionuclide i at location j , etc.;

Concentration_i is the average concentration of nuclide, i , in the source inventory (Bq kg^{-1}).

In the case of Tessenderlo, the radium decay chain supports both the main radon inhalation dose (^{222}Rn) and the ^{210}Po and ^{210}Pb contributions to food ingestion pathways. By comparison, the dose due to thorium and its daughters is much smaller. This is illustrated in Table 5.17, which indicates the annual critical group dose incurred for unit parent radionuclide concentration.

Table 5.17 Ratio of dose incurred per unit radionuclide concentration

Radionuclide i	Concentration _{i} (Bq kg^{-1})	Dose at location j (mSv a^{-1})	Ratio of dose to concentration ($G_{i,j}$) (mSv a^{-1} per Bq kg^{-1})
Ra-226 + daughters	3500	0.5	1.42E-4
Th-232 + daughters	40	1.18E-47	2.95E-49

The dose deriving from ^{226}Ra is more than 40 orders of magnitude greater than that from ^{232}Th , per unit concentration in the waste.

5.3.4 Intrusion scenario critical group exposure

It was noted in Section 5.3.2.2 that the intrusion exposure group is assumed, hypothetically to reside in dwellings constructed on the main waste bearing area. Such a group can only be conceived as a future scenario and should, rightly, be expressed as a probabilistic event since it is by no means certain that such intrusion will occur. Neither can the style of dwelling nor the population group potentially affected be defined with any certainty. Given these reservations, the simplest assumption is that intrusion occurs (i.e. probability of 1) and that the radionuclide inventory within the waste has not decayed appreciably. In effect, it is assumed that intrusion occurs shortly after the end of the institutional control period. Consequently, there is no distinction to be made between short-term and peak dose estimates since the peak dose will occur at the time of highest radionuclide inventory (i.e. the short term case).

Calculated doses for the best estimate inventory, by pathway and nuclide, are presented in Table 5.18. It is emphasised that this group is hypothetical only and that adopting a deterministic approach to dose calculation, rather than the more reasonable probabilistic approach, will err towards over-predicting the risk incurred.

Table 5.18 Dose to the intrusion scenario group, calculated by AMCARE, for pathway and nuclide (best estimate inventory)

Radionuclide	Dose (mSv a ⁻¹)				
	External irradiation	Radon inhalation	Dust inhalation	Ingestion	Total
Radium and daughter nuclides					
²²⁶ Ra	6.93E+00	-	3.54E-03	1.06E+00	7.99E+00
²²² Rn	-	3.57E+02	-	-	3.57E+02
²¹⁰ Pb	-	-	-	8.38E-22	8.38E-22
²¹⁰ Po	-	-	-	4.58E-22	4.58E-22
Thorium and daughter nuclides					
²³² Th	1.46E-05	-	2.89E-04	4.59E-03	4.89E-03
²²⁸ Ra	-	-	-	1.05E-47	1.05E-47
²²⁸ Th	-	-	-	3.58E-49	3.58E-49
Total	6.93E+00	3.57E+02	3.83E-03	1.06E+00	3.65E+02

A summary of estimated doses, assuming a range on estimates of the waste inventory, is presented in Table 5.19.

Table 5.19 Summary of doses, calculated by AMCARE, to the intrusion scenario group (upper and lower estimate inventory)

	Dose incurred by pathway (mSv a ⁻¹)				
	External exposure	Radon inhalation	Dust inhalation	Ingestion	Total
Best estimate inventory	6.93E+00	3.57E+02	3.83E-03	1.06E+00	3.65E+02
Upper estimate inventory	8.12E+00	4.18E+02	4.49E-03	1.24E+00	4.28E+02
Lower estimate inventory	4.16E+00	2.14E+02	2.36E+03	6.37E-01	2.19E+02

Doses incurred for this scenario (365 mSv a⁻¹) are clearly very much higher than for the off-site residential critical group (0.5 mSv a⁻¹). The principal dose pathway is again radon inhalation, but the ingestion doses and external exposure pathway doses are also very much elevated, as the radionuclides are available for immediate uptake to crops etc. Ranges on the radon emanation fraction and diffusion coefficient are the main parameters determining the range on total dose estimates (Figures 5.4 and 5.5).

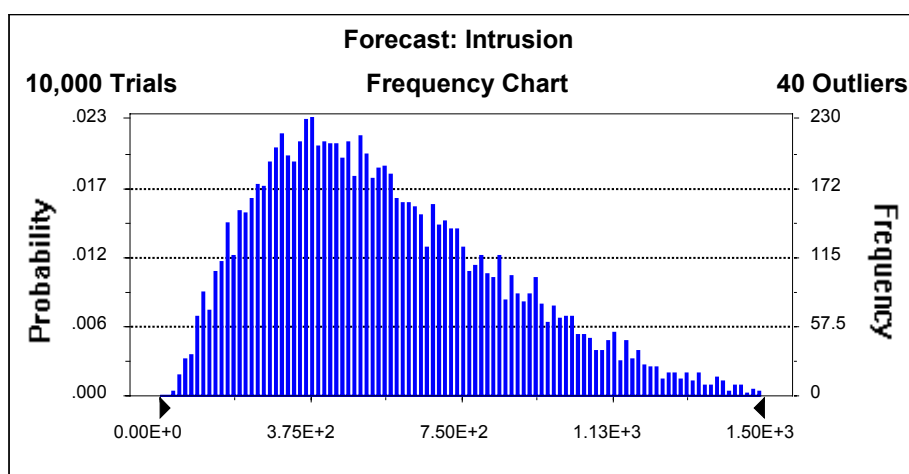


Figure 5.4 Probabilistic range on dose estimates to the intrusion scenario group (best estimate inventory)

The relatively high dose for the intrusion scenario group derives in part from the assumption that the house foundations penetrate the waste cap (Section 5.3.2.2) and that, consequently, the indoor radon emanation to air is not retarded. Furthermore, the doses have been calculated following a predominantly generic approach which considers, to only a limited extent, the site-specific characteristics. When the ratio of radon in the air of the houses to the radium concentration for local soil is taken into account, then the resulting dose decreases by about an order of magnitude. The reason for this lower value may be the presence of a foundation slab at the base of the houses and a lower emanation coefficient than the one used in the current study. Additional calculations with AMCARE have shown that when a house is effectively constructed on a raft foundation above the cap level the annual dose for an individual scenario group declines to 25 mSv.

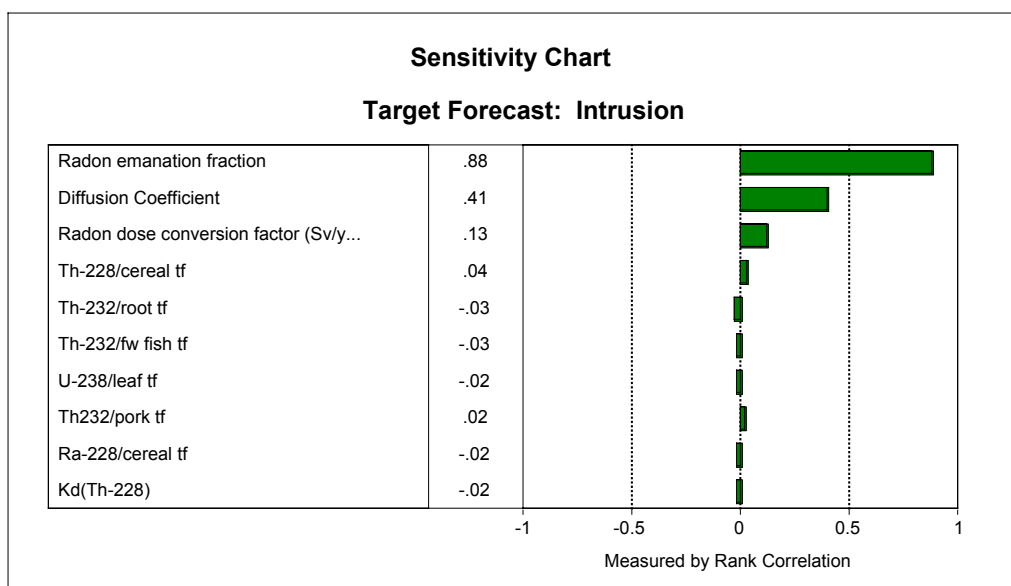


Figure 5.5 Key parameters determining dose to the intrusion scenario group (best estimate inventory)

The range on annual dose estimates is again very skewed, from about 50 mSv to 1800 mSv, and the centile distribution of doses is summarised in Table 5.20.

Table 5.20 Centile dose estimates, calculated by AMCARE, to the intrusion scenario critical group (best estimate inventory)

Percentile	Value
0.0%	3.12E+01
2.5%	1.21E+02
5.0%	1.62E+02
50.0%	5.16E+02
95.0%	1.13E+03
97.5%	1.24E+03
100.0%	1.77E+03

5.3.5 Workforce exposure

The workforce, in this case, is assumed to work at the Tessengerlo site and to live locally. Doses estimated to this workforce are presented in Table 5.21 by pathway and by nuclide. It is assumed that the workforce is currently present on the site, which represents the most

conservative case since no decay of the waste inventory will have occurred. Consequently, only the current dose is estimated.

It can be seen from Table 5.21 that the ingestion pathway is very much lower than for the previous exposure groups. This reflects the assumption that, for this group, only 25% of all food is locally derived and that all drinking water is obtained from a distant, non-contaminated, source.

Table 5.21 Dose to the workforce group, calculated by AMCARE, for pathway and nuclide (best estimate inventory)

Radionuclide	Dose (mSv a ⁻¹)				
	External irradiation	Radon inhalation	Dust inhalation	Ingestion	Total
Radium and daughter nuclides					
²²⁶ Ra	1.27E+00	-	6.47E-04	5.37E-33	1.27E+00
²²² Rn		6.34E-01	-	-	6.34E-01
²¹⁰ Pb	-	-	-	4.25E-31	4.25E-31
²¹⁰ Po	-	-	-	5.81E-32	5.81E-32
Thorium and daughter nuclides					
²³² Th	2.67E-06	-	5.28E-05	1.99E-70	5.55E-05
²²⁸ Ra	-	-	-	1.81E-59	1.81E-59
²²⁸ Th	-	-	-	1.79E-62	1.79E-62
Total	1.27E+00	6.34E-01	7.00E-04	4.89E-31	1.90E+00

A summary of estimated doses, assuming a range on estimates of the waste inventory, is presented in Table 5.22.

Table 5.22 Summary of doses, calculated by AMCARE, to the workforce group (upper and lower estimate inventory)

	Dose incurred by pathway (mSv a ⁻¹)				
	External exposure	Radon inhalation	Dust inhalation	Ingestion	Total
Best estimate inventory	1.27E+00	6.34E-01	7.00E-04	4.89E-31	1.90E+00
Upper estimate inventory	1.48E+00	7.42E-01	8.20E-04	5.72E-31	2.23E+00
Lower estimate inventory	7.59E-01	3.80E-01	4.32E-04	2.93E-31	1.14E+00

A range on total dose estimates is presented in Figure 5.6, based on the best estimate inventory. It can be seen that the dose range in this case has a very skewed distribution, with the modal value of 1.9 mSv a⁻¹ at the lower end of the estimates. However, the total range encompassed is relatively small, with an upper dose estimate around 5 mSv a⁻¹ (see also Table 5.23).

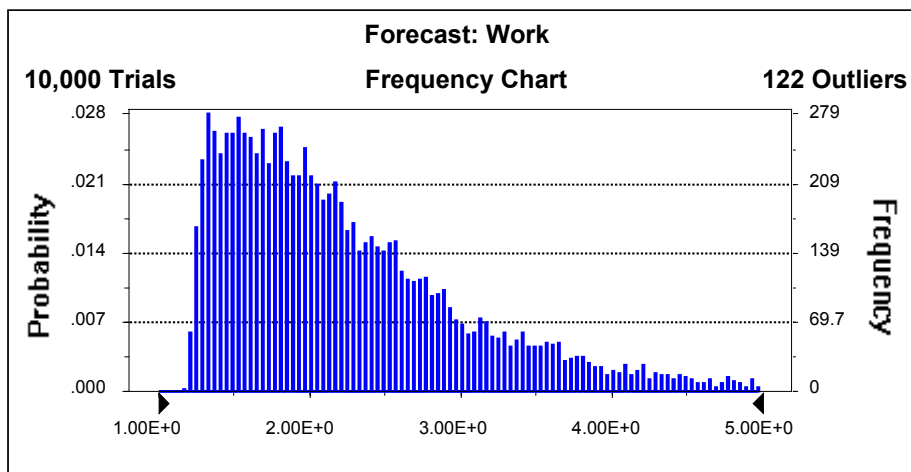


Figure 5.6 Probabilistic range on dose estimates to the workforce (best estimate inventory)

Table 5.23 Centile dose estimates, calculated by AMCARE, to the workforce (best estimate inventory)

Percentile	Value
0-0%	1.19E+00
2.5%	1.28E+00
5.0%	1.33E+00
50.0%	2.05E+00
95.0%	3.98E+00
97.5%	4.50E+00
100.0%	7.97E+00

Overall, the workforce dose (1.9 mSv a^{-1} for the best inventory estimate case) is rather higher than that estimated for the off-site residents (0.5 mSv a^{-1}). This is due markedly to increased external irradiation. As a consequence, a sensitivity analysis of factors determining variability on dose estimates (Figure 5.7) indicates that the cap characteristics and the assumed shielding factor for external irradiation have a higher importance than elsewhere.

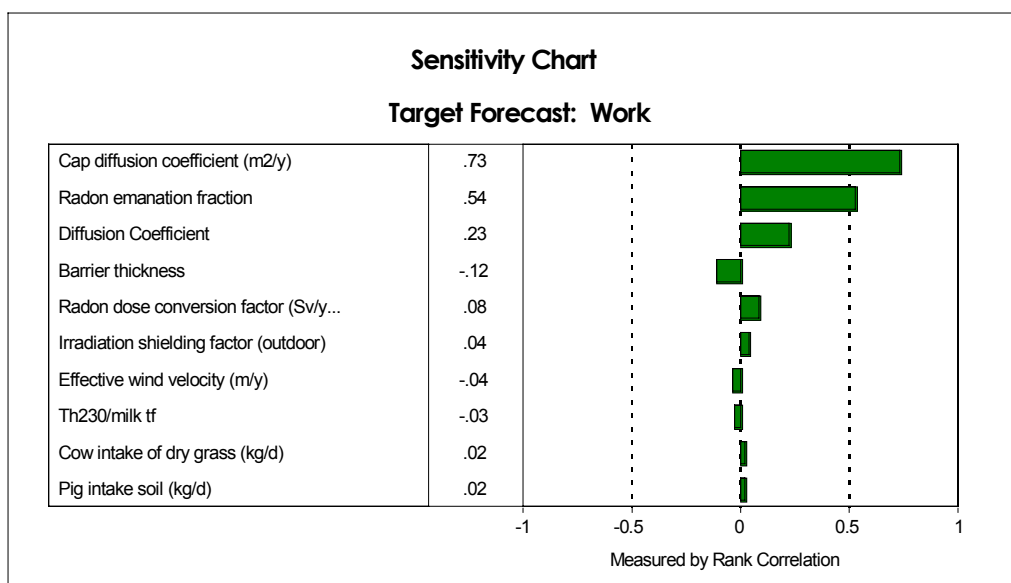


Figure 5.7 Key parameters determining dose to the workforce (best estimate inventory)

5.3.6 Collective dose

Collective doses are presented for the population in the vicinity of the Tessengerlo site, as described in Annex B2. This group is considered to reside within a radius of 20 km from the site, with a mean distance of 10 km, and includes the local critical group. The total population affected is taken to be 200000 individuals, equivalent to a population density of approximately 150 persons per km² which is considered to be representative of a semi-urban population. This is a very narrow definition of collective doses, and effectively limits the population of concern to those individuals potentially exposed to appreciably elevated levels of radon in air.

Doses are integrated over 100 years, which is commensurate with the foreseeable period of institutional control, and over the more commonly defined period of 500 years, allowing for inter-generational doses. For the best estimate inventory case, the collective dose so defined is around 20 to 110 manSv (see Table 5.24). All doses are expressed as the collective committed effective dose (collective CED).

Table 5.24 Collective dose to the population local to Tessengerlo, calculated by AMCARE, for pathway and nuclide (best estimate inventory)

Radionuclide	Dose (manSv)							
	External irradiation		Total inhalation		Ingestion		Total	
	100 years	500 years	100 years	500 years	100 years	500 years	100 years	500 years
Radium and daughter nuclides								
²²⁶ Ra	6.97E-43	1.07E-20	7.54E-52	4.80E-30	2.47E-42	3.79E-20	3.17E-42	4.86E-20
²²² Rn	-	-	2.28E+01	1.14E+02	-	-	2.28E+01	1.14E+02
²¹⁰ Pb	8.72E-39	8.46E-22	2.37E-47	9.75E-30	1.30E-33	1.26E-16	1.30E-33	1.26E-16
²¹⁰ Po	6.90E-41	6.24E-24	2.31E-46	9.97E-29	7.60E-34	6.87E-17	7.60E-34	6.87E-17
Thorium and daughter nuclides								
²³² Th	3.39E-81	8.15E-58	1.46E-89	8.87E-66	5.35E-78	1.29E-54	5.35E-78	1.29E-54
²²⁸ Ra	2.02E-54	4.51E-44	2.63E-68	4.87E-57	7.06E-53	1.57E-42	7.26E-53	1.62E-42
²²⁸ Th	3.19E-54	9.01E-44	1.72E-68	4.07E-57	1.90E-54	5.36E-44	5.08E-54	1.44E-43
Total	8.79E-39	1.16E-20	2.28E+01	1.14E+02	2.06E-33	1.94E-16	2.28E+01	1.14E+02

The dose received is proportional to the inventory assumed and, for completeness, upper and lower collective dose estimates are summarised in Table 5.25 for the upper and lower estimates of the radionuclide inventories in the waste. Doses arising from external exposure pathways for all off-site groups are negligible over the short term.

Table 5.25 Summary of collective doses, calculated by AMCARE, to the population local to Tessengerlo (upper and lower estimate inventory)

	Local collective dose by pathways (manSv)			
	external irradiation	Total (radon + dust) inhalation	ingestion	total
<i>Best estimate waste inventory</i>				
100 years	8.79E-39	22.8	2.06E-33	22.8
500 years	1.16E-20	114	1.94E-16	114
<i>Upper estimate waste inventory</i>				
100 years	1.03E-38	26.7	2.41E-33	26.7
500 years	1.35E-20	134	2.28E-16	134
<i>Lower estimate waste inventory</i>				
100 years	5.28E-39	13.7	1.23E-33	13.7
500 years	6.93E-20	68.4	1.17E-16	68.4

The collective dose for the best estimate inventory at Tessengerlo, of around 20 manSv over 100 years, or 110 manSv over 500 years, is largely determined by the radon inhalation pathway. As noted for the local residency farming group (Section 5.3.3), the dose from other pathways

increases substantially over time as groundwater becomes contaminated and migrates away from site, which affects both internal (ingestion and dust inhalation) pathways and external irradiation. However, the migration of nuclides in groundwater is relatively slow, determined largely by the assumed distribution coefficients (K_d 's), and these pathways do not become significant by comparison to radon inhalation over 500 years.

Again, considerable uncertainty attaches to the central dose estimate, due to potential variability of many of the parameter values assumed. Using Crystal Ball, a range on dose estimates is presented in Figure 5.8, relevant to the 500 year integral and the best estimate inventory scenario only.

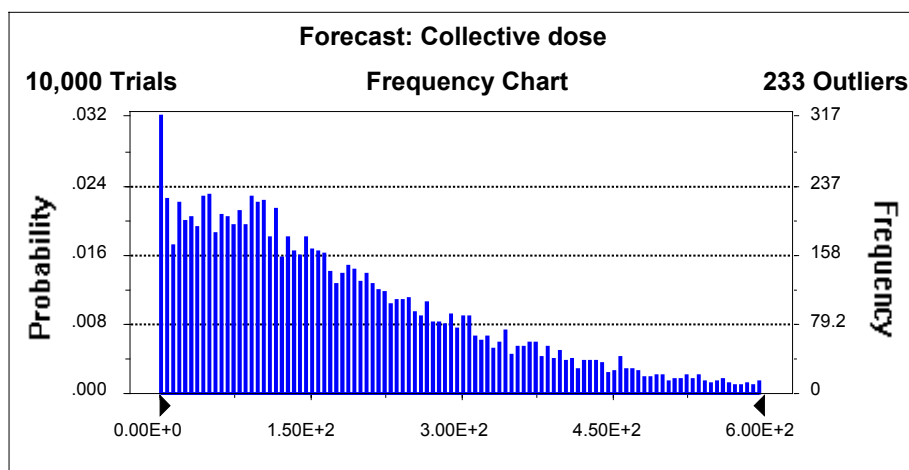


Figure 5.8 Probabilistic range on collective dose estimates to the population local to Tessenderlo (500 year integral, best estimate inventory)

It can be seen from Figure 5.8 that there is again a highly skewed collective dose distribution from about 5 manSv to more than 500 manSv. The upper and lower 2.5th centile values for dose are excluded from Figure 5.8 (which comprise the 233 outliers). A tabulation of the centile dose estimate values is presented in Table 5.26.

Table 5.26 Centile collective dose estimates, calculated by AMCARE, to the population local to Tessenderlo (500 year integral, best estimate inventory)

Percentile	Value
0.0%	5.2E-10
2.5%	3.94
5.0%	11.8
50.0%	150
95.0%	502
97.5%	592
100.0%	1050

A sensitivity analysis of the pathways and parameters contributing to the collective dose indicates that the radon cap diffusion coefficient assumed (which determines the fraction of radon evolution from the waste matrix to atmosphere) is the single most important factor (Figure 5.8), followed by the radon emanation rate and the cap thickness. A variability of $\pm 10\%$ on the radon dose factor influences the dose potentially incurred in an almost linear fashion, again emphasising the dominant dose contribution from radon inhalation.

Factors affecting other pathways can be seen from Figure 5.8 to exert a much smaller influence on total dose. As noted previously (Section 5.3.3) the ranges for K_d (presented in Table 5.3) and transfer factors (presented in Tables 5.6 to 5.8) exceed, in some cases, five orders of

magnitude, indicating that very large variations in these parameters are of relatively minor consequence at Tessengerlo.

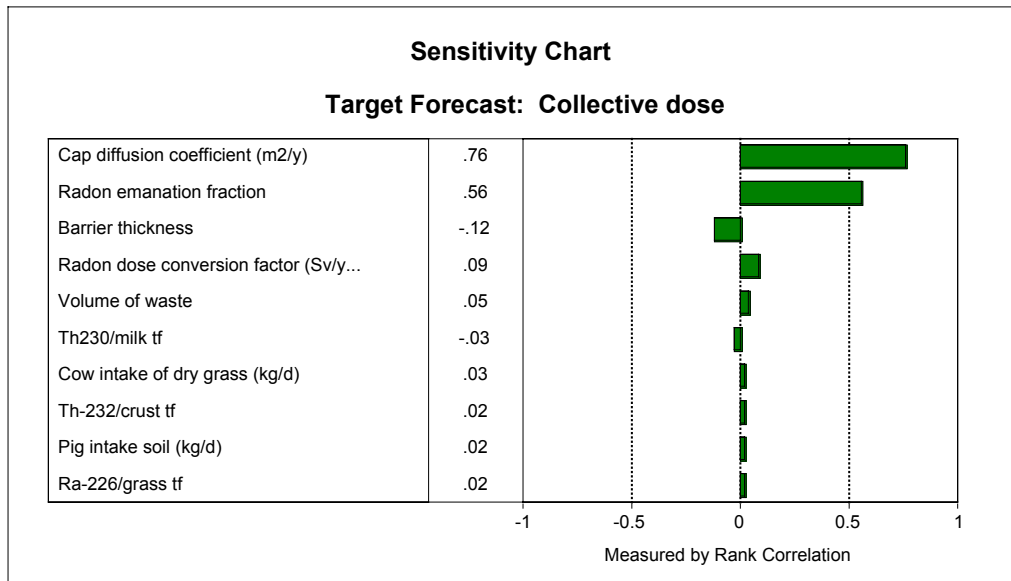


Figure 5.9 Key parameters determining collective dose to the population local to Tessengerlo (500 year integral, best estimate inventory)

It has been seen in Table 5.25 that the collective dose estimate is closely correlated to the estimated waste inventory. In order to compare collective dose estimates obtained for Tessengerlo with doses derived for other sites, a rough approximation of dose incurred per unit concentration of radionuclide i in the waste can be derived (${}^{\text{coll}}G_{i,jx}$), such that:

$${}^{\text{coll}}G_{i,jx} = (\sum {}^{\text{coll}}\text{dose}_{i,jx} + {}^{\text{coll}}\text{dose}_{\text{idtr}1,jx} + {}^{\text{coll}}\text{dose}_{\text{idtr}2,jx} + \dots) / \text{concentration}_{i0}$$

where: ${}^{\text{coll}}\text{dose}_{i,jx}$ is the total collective dose from all pathways (manSv) due to radionuclide i at mean distance j over time period x (years);

${}^{\text{coll}}\text{dose}_{\text{idtr}1,jx}$ is the total collective dose from all pathways (manSv) due to the daughter nuclide 1 of radionuclide i at location j over time period x (years);

${}^{\text{coll}}\text{dose}_{\text{idtr}2,jx}$ is the total collective dose from all pathways (manSv) due to the daughter nuclide 2 of radionuclide i at location j over time period x (years), etc.;

$\text{concentration}_{i0}$ is the average concentration of nuclide i in the source inventory (Bq kg^{-1}) at time zero (0 years).

It can be seen that the calculation of ${}^{\text{coll}}G_{i,jx}$ for collective doses is entirely analogous to the derivation of $G_{i,j}$ for individual critical group doses (Section 5.3.3).

In the case of Tessengerlo, the radium decay chain supports both the main radon inhalation dose (${}^{222}\text{Rn}$) and the ${}^{210}\text{Po}$ and ${}^{210}\text{Pb}$ contributions to food ingestion pathways. By comparison, the dose due to thorium and its daughters is much smaller. This is illustrated in Table 5.27, which indicates the total collective dose incurred to the local population for unit parent radionuclide concentration over 500 years (${}^{\text{coll}}G_{i,j500}$).

Table 5.27 Ratio of collective dose incurred per unit radionuclide concentration (best estimate inventory)

Radionuclide <i>i</i>	Concentration _{<i>i</i>} (Bq kg ⁻¹)	Dose at location <i>j</i> over 500 years (manSv)	Ratio of dose to concentration (^{coll} G _{<i>i,j</i>,500}) (manSv per Bq kg ⁻¹)
Ra-226	3500	114	3.25E-2
Th-232	40	1.76E-42	4.4E-44

It can be seen from Table 5.27 that ²²⁶Ra, and its daughter nuclides, have a more than forty orders of magnitude greater collective dose commitment to the local population per unit radioactivity than ²³²Th and its decay product daughter nuclides. This arises largely from the ²²²Rn dose (see Section 5.3.1 for simplified decay chains relevant to Tessenderlo).

5.4 Discussion

5.4.1 Radionuclides and pathways of major importance

The dominant dose to almost all exposure groups, arises from inhalation of radon (²²²Rn) gas, which is supported from the parent inventory of ²²⁶Ra. At Tessenderlo this results in a dose to the local off-site critical group of residential farm-workers of around 1.42×10^{-4} mSv a⁻¹ per Bq ²²⁶Ra kg⁻¹ in the waste material. The dose per unit parent radionuclide inventory, is a parameter which may be used to compare the relative impact of different sites or of a remediation strategy.

Doses in excess of 1 mSv a⁻¹ are calculated only for external irradiation and food ingestion pathways for the intrusion scenario only, although this is still two or more orders of magnitude lower than the radon inhalation dose for these groups. For the workforce groups, the external irradiation pathway contributes around 67% of the total dose at Tessenderlo. In general, the external dose component is dominated by ²²⁶Ra and its daughter nuclide chain.

Collective doses are again dominated by radon inhalation, although to some extent this may be an artefact since food consumption pathways etc. are included in a limited fashion. Nonetheless, a dose per unit parent nuclide inventory can again be calculated, and is 3.25×10^{-2} manSv a⁻¹ per ²²⁶Ra kg⁻¹ and 4.4×10^{-44} manSv a⁻¹ per Bq ²³²Th kg⁻¹.

5.4.2 Effect of parameter uncertainty

The main factors determining uncertainty in dose estimates, other than the waste inventory itself, relate to the rate of emission of radon gas to atmosphere and, to a lesser extent, to the shielding from external irradiation.

With respect to radon emission, cap diffusion, radon emanation fraction, the diffusion coefficient (which describes radon release from radium and hence the release of radon in the soil/waste pores) and the surface layer thickness of covering material are the principal determinants. The inclusion of a barrier of overlying material can be described with reasonable certainty, as this is a readily measured parameter. By comparison, the emanation coefficient is less readily defined and must be considered highly uncertain. This inevitably leads to a large range for predicted dose uptakes.

For external irradiation, the exposure group principally affected is the workforce on-site. In this case, shielding will largely be a function of cap thickness and relative spent indoors or outdoors. Both these quantities are inherently measurable and so ranges on uncertainty can be reduced for any specific site dose estimate.

5.5 Conclusions

This study indicates that for the example site, the main pathway of concern for nearby off-site residents arises from radon emanation. Actions to reduce the rate of emanation may thus be supposed, *a priori*, to represent the most likely approaches to cost effective dose reduction and site remediation measures. Since the estimated doses to this group are, in any case, relatively low ($\sim 0.5 \text{ mSv a}^{-1}$ at Tessengerlo) it would again be assumed, *a priori*, that site remediation measures would also be limited to relatively low cost options on a cost-benefit analysis.

With respect to the hypothetical intrusion scenario, much higher doses may be incurred. At Tessengerlo the modal value is 365 mSv a^{-1} for the best estimate inventory scenario (although this reduces to $\sim 25 \text{ mSv a}^{-1}$ if it is assumed that the waste cap remains *in-situ*). Radon inhalation is again the dominant pathway, although both ingestion doses and external irradiation potentially deliver doses in excess of 1 mSv a^{-1} . This might imply that, for the period post institutional control of the site, a range of remediation options would be considered which could not be justified for the off-site exposure group alone.

5.6 References

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6. REMEDIATION OPTIONS

6.1 Introduction

Sites containing radioactive materials may give rise to locally elevated levels of exposure. In particular, sites contaminated as a consequence of past activities, which may not have been subject to the authorisation and control procedures which currently apply, are more likely to result in local exposure pathways. This is discussed in Chapter 5 for a number of example cases. The purpose of applying any remediation technology, or combination of technologies, to a radioactively contaminated site is to reduce the level of public exposure to radioactivity on-site or, via radionuclide migration, off-site. In order to determine the requirement for remediation, and the options most likely to prove cost-effective, it is necessary to develop a common approach to categorising sites and types of remediation approaches. An overview of remediation techniques is presented in Annex C1. It is the purpose of this chapter to:

- i) summarise the remediation technologies available for treating radioactive contaminated waste;
- ii) identify where each technology may be appropriately applied;
- iii) examine the impact of applying each technology (and suitable combinations) on the waste at the Tessenderlo phosphate site in Belgium.

In general, the differing remediation technologies available fall broadly into four categories:

- bulk removal of waste from the site to a more appropriate location;
- installation of barriers to reduce on-site exposure or to restrict off-site migration of radionuclides;
- addition of materials to bind the radionuclides and, as a consequence, restrict their mobility;
- extraction of radionuclides from the waste and transfer to a more appropriate location.

An additional ‘institutional’ approach is available in principle, through placing restrictions on the use of the site. However, it is commonly accepted that such restrictions would be effective only over relatively short time-scales and this type of approach is not considered further here.

There are significant differences between each of the approaches considered here. In some cases the radionuclides will be removed from the site, in others they will be contained on the site. Such approaches will be appropriate to different types of contamination. The choice of which technology to apply to a particular site will depend on the location and physical characteristics of the site and the nature of the contamination.

Under some circumstances, a combination of technologies may be used to restrict exposure from the waste, especially where a number of pathways exist. Such a combination of technologies can, at times, be considerably more cost effective than applying a single, more costly technique. However, only certain combinations of technologies are logical. Similarly, the order in which different technologies are applied is important.

At the Tessenderlo example site, which contains a large volume of spoil with a relatively low concentration of ^{226}Ra , the principal dose exposure pathway is through inhalation of the short-lived daughter ^{222}Rn gas ($t_{1/2}$ 3.8 days). The most effective technologies for reducing the local dose are thus those which either remove the parent ^{226}Ra , or which reduce the rate of radon efflux. However, the cost of bulk removal of large volumes of waste can be prohibitive. It will be shown that it is likely that capping alone would be the preferred option at Tessenderlo considering dose reduction to the public, workforce dose during remediation work, potential costs and public reassurance or disturbance.

6.2 Remediation options

A number of reviews are available, which report various techniques and approaches as being effective in remediating contaminated land (e.g. Chilton and Pfuderer (1989); Gee and Wing (1994); Post (1994); Slate *et al.* (1994); Spectrum (1994, 1996); USEPA (1996); Van den Brink *et al.* (1995); and see Annex C1). This study, however, is restricted to those technologies (Table 6.1) which are well-established and have been successfully applied to treat radioactive waste sites, or show considerable promise in laboratory and/or field trials.

Table 6.1 Main approaches to remediation considered within CARE

Removal of Sources	Bulk removal Surface scraping Turf cutting
Containment	Capping Subsurface barriers
Immobilisation	Cement-based solidification (<i>in-situ</i> and <i>ex-situ</i>) Chemical immobilisation (<i>in-situ</i> and <i>ex-situ</i>)
Separation	Soil washing Flotation Chemical/solvent extraction

In determining the viability of applying any of the above technologies for a particular contaminated site, a number of factors must be taken into account including:

- the characteristics of the site;
- the risk to the public;
- the performance and cost of the technique to be applied; and,
- the exposure of the workforce during remediation work.

More subjectively, the degree of public reassurance arising from the chosen method may also be of concern. As far as possible, all above considerations have been quantified in Annex C1, through a review of published information and are summarised in Table 6.2.

AMCARE was run using the performance values, given in Table 6.2. The impact of the different categories of remediation technologies may be described as follows:

- removal of sources and separation technologies reduce the input of radionuclides into groundwater pathways and reduce the level of radon emanation by removing the parent nuclides with a proportionate decrease in the magnitude of all exposure pathways;
- immobilisation and containment (except capping) technologies reduce input to groundwater, and therefore have a significant impact on the off-site terrestrial and aquatic exposure pathways (ingestion and external irradiation), but have little impact on radon emanation;
- capping reduces the input of radionuclides into groundwater and also reduces radon emanation in a manner proportional to the thickness of the surface barrier.

Removal of source material and capping of the waste area reduce external irradiation both on-site and off-site: in the case of capping this presumes that the covering material remains in-situ. For extreme intrusion scenarios, where penetration of the cap may be assumed, the on-site external radiation pathway will not be reduced. Immobilisation and containment approaches, other than capping, tend to reduce off-site external irradiation, but may have little effect on-site.

It is also possible to use combinations of remediation technologies. This can be particularly useful if the combination reduces all of the major exposure pathways from the waste to man. However, only logical combinations should be considered and there may also be restrictions in

practice on the order in which combination approaches may be applied. Potentially useful combinations are summarised in Table 6.3.

Aside from enhancing the effectiveness of remediation, combining technologies may also cost less than the sum of the individual technologies. For example, the excavation of contaminated material is a requirement for a number of remediation approaches. Clearly, where two options are combined, each of which in isolation would require excavation of the waste, a considerable cost saving may accrue since a single 'pre-treatment' (i.e. excavation) only is required.

Table 6.2 Performance values of various remediation technologies

Technology	'Clean-up' Indicator	Cost (ECU)		Service life	Workforce exposure (manh)	Public reassurance
		Extraction (per m ³)	Disposal & transport (per m ³)			
Removal of Waste	<i>Decontamination factor</i>				(per m ³)	
Soil excavation	1 - 20	50-150	450-800	not applicable	0.2-1	high
Soil scraping	1 - 20	13-60	450-800	not applicable	0.2-1	high
Turf harvesting	1 - 20	9	450-800	not applicable	0.2-1	high
Separation & removal of waste	<i>Decontamination factor</i>	<i>Excavation & separation (per m³)</i>	<i>Disposal & transport (per m³)</i>		(per m ³)	
Soil washing	1- 10	150-500	50-150	not applicable	0.25-1.5	moderate-high
Flotation	1- 10	15-240*	50-150	not applicable	0.25-1.5	moderate-high
Chemical/solvent extraction	1 - 20	180-820		not applicable	1.2-3.5	moderate
Containment	<i>Resultant permeability (m s⁻¹)</i>	<i>Total (per m² surface area)</i>			(per m ² surface area)	
Capping	1 ×10 ⁻¹² - 1 ×10 ⁻⁹	30-45		1,000 y	0.03-0.3	low
Subsurface barrier						
a) slurry walls	1 ×10 ⁻¹² - 1 ×10 ⁻⁸	510-710		100-1,000 y	0.06-0.4	low
b) grout curtains	1 ×10 ⁻¹² - 1 ×10 ⁻⁸	310-420		100-1,000 y	0.06-0.4	low
Immobilisation	<i>Mobility reduction factor</i>	<i>Total (per m³)</i>			(per m ³)	
Cement-based solidification						
a) <i>ex-situ</i>	5 - 25	75-300		not known	0.25-1.5 (inc. excav ⁿ)	low
b) <i>in-situ</i>	5 - 25	50-310		not known	0.06-0.4	low
Chemical immobilisation						
a) <i>ex-situ</i>	5 - 50	110-570		not known	0.25-1.5 (inc. excav ⁿ)	low
b) <i>in-situ</i>	5 - 50	60-420		not known	0.06-0.4	low

Note: * Capital cost. A separately identified operating cost of 0.8-3.8 m⁻³ is also estimated.

Table 6.3 Combinations of remediation technologies considered appropriate

First Remediation Techn.	Second Remediation Technology									
	Bulk removal	Soil washing	Flotation	Chemical sep ^l /solvent extract ⁿ	Capping	Sub-surface barrier	Cement-based solidification (ex-situ)	Cement-based solidification (in-situ)	Chemical solidification (ex-situ)	Chemical solidification (in-situ)
Bulk removal		✓	✓	✓	✓		✓		✓	
Soil washing					✓	✓	✓		✓	
Flotation					✓	✓	✓		✓	
Chemical separation/solvent extraction					✓	✓	✓		✓	
Capping						✓				
Subsurface barrier					✓			✓		✓
Cement-based solidification (ex situ)	✓									
Cement-based solidification (in situ)					✓	✓				
Chemical-based solidification (ex situ)	✓									
Chemical-based solidification (in situ)					✓	✓				

6.3 Selection of remediation technologies for a contaminated site

The selection of appropriate remediation techniques will depend on the characteristics of the contaminated waste and the potential exposure pathways. Consequently, the choice is generally site specific. However, the characteristics of the CARE sites are sufficiently similar that some general considerations may be applied to the selection of remediation technologies.

- i) Volume: High volumes of contaminated waste will favour *in-situ* remediation technologies.
- ii) Accessibility: Inaccessible wastes, and waste sites, will favour *in-situ* technologies.
- iii) Local population: The presence of a large population near the site will favour removal of contaminant material from the site to a more secure location.
- iv) Exposure pathways:
 - a. sites where radon and dust emissions are high will be best treated by either removal of material from the site or the installation of a surface barrier above the waste;
 - b. sites where leaching and off-site migration of radionuclides is important will be best treated by technologies which reduce groundwater infiltration through the waste;
 - c. sites with high external irradiation levels will be best treated by surface barriers or removal of the radioactive material.

Most remediation techniques will, to some extent, reduce on-site and off-site exp inhalation of radon and contaminated dust, external exposure and consumption of contaminated food. However, *in-situ* and *ex-situ* solidification technologies will only have a minor impact on the external dose. In addition, the introduction of sub-surface barrier technologies will have no effect on on-site exposure pathways as they will not present a barrier to the occupants on the site.

A comment is in order regarding the fate of the large volumes of wastes containing natural radionuclides, which may be generated during the remediation action. Deep geological disposal is not a feasible option on cost grounds and seadumping is not feasible on political grounds, leaving surface/shallow land disposal as the only viable option. While it may be the case that the doses from 'normal' evolution of a properly designed and constructed repository could be lower than those arising

From leaving the wastes in situ, doses from intrusion scenarios will probably be the same in both cases. Therefore, there may be only marginal benefits to be gained from removing the waste and disposing of it, versus adopting in situ options such as capping. This may be an important point in deciding on the type of remedial actions that are viable.

6.4 Application of remediation technologies to a contaminated site

To illustrate the application of differing remediation technologies to a contaminated site, the Tessenderlo site in Belgium is considered. The site is located in a sparsely populated area surrounded by agricultural land and contains approximately $9 \times 10^6 \text{ m}^3$ of radioactive waste (3500 Bq kg^{-1} of ^{226}Ra and 40 Bq kg^{-1} of ^{232}Th) within a surface area of approximately $5.3 \times 10^5 \text{ m}^2$. The characteristics of the site are summarised in Chapter 5.

Given the large volume of waste, and relatively low concentration of radionuclides, it would be expected that on-site treatment would be the favoured solution. However, to illustrate the predictive approach arising from AMCARE (described in Chapter 5) the effects of all the remediation technologies are presented for this site and, in addition, a number of possible combinations of remediation technologies are considered. Effects are considered in terms of:

- dose avoidance to average members of the local critical group;

- collective dose avoidance to the local population (radius 20 km from site);
- dose incurred by the workforce during remediation; and,
- cost.

The fractional removal of waste from site is also considered separately, as this is considered to be linked to public assurance consequent to remediation work.

6.4.1 Remediation effects on the critical group dose at Tessenderlo

A detailed definition of the critical group is given in Appendix B2. Briefly, this is a group of 30 people who live 1 km away from the site. They grow their terrestrial food produce in a field 100 m from the site and get their fish from a river 50 m from the site. The drinking water comes from a well in the field 100 m away. The impact of various remediation options, applied singly or in combination, on the critical group dose is summarised in Table 6.4. It is assumed that all remediation options, and the ‘no treatment’ case, apply for current conditions rather than for ‘peak dose’ 10000 year conditions (see Chapter 5 for further discussion of current versus peak dose calculations).

Table 6.4 The impact of remediation technologies on the critical group dose for the Tessenderlo phosphate processing site (Belgium)

Remediation Technology	Critical Group Dose (mSv a ⁻¹) [†]	Ranking
No treatment	5.2 × 10 ⁻² - 2.2	--
Removal of source	5.2 × 10 ⁻³ - 0.49	8
Soil washing	8.3 × 10 ⁻³ - 0.73	9
Flotation	8.3 × 10 ⁻³ - 0.73	9
Chemical extraction	4.4 × 10 ⁻³ - 0.44	7
Capping	6.6 × 10 ⁻⁶ - 3.3 × 10 ⁻²	3
Sub-surface barrier (grout curtain)	5.1 × 10 ⁻² - 2.2	10
Cement-based solidification (<i>ex-situ</i> and <i>in-situ</i>)*	6.3 × 10 ⁻³ - 0.36	5
Chemical-based solidification (<i>ex-situ</i> and <i>in-situ</i>)*	6.1 × 10 ⁻³ - 0.37	6
Flotation + Capping	1.1 × 10 ⁻⁶ - 8.5 × 10 ⁻³	2
Cement-based solidification (<i>in-situ</i>)* + Capping	1.0 × 10 ⁻⁶ - 5.4 × 10 ⁻³	1
Chemical extraction + Cement-based solidification (<i>ex-situ</i>)*	6.5 × 10 ⁻⁴ - 7.1 × 10 ⁻²	4
Sub-surface barrier + Capping	6.6 × 10 ⁻⁶ - 3.3 × 10 ⁻²	3

Note: * Mobility factor used for all radionuclides including ²²²Rn.

† Range refers to the 5 to 95th percentile values.

These results may be explained by the fact that it is exposure pathways involving the influx of ²²²Rn from the Tessenderlo which are dominant. Consequently, remediation technologies which reduce the radon flux will cause the greatest reduction in the critical group dose. Capping gives rise to the greatest reduction in the radon flux and this is reflected in the fact that critical group dose which arises after the application of capping is about an order of magnitude lower than that obtained after the application of any other single technology. Combining capping with other technologies can lead to further reductions in the critical group dose. The application of a surface barrier to the Tessenderlo site will not reduce the radon flux and, as a consequence, has a negligible impact on the critical group dose.

6.4.2 Remediation effects on the collective dose at Tessenderlo

The collective dose is taken to be the sum of the total dose received by the critical group, over a defined period, plus the total dose received through radon inhalation by the remainder of the population within a 20 km radius of the site (assuming a mid-point distance of 10 km): see Annex B2 for a detailed description of the method. A uniform population density of c.150 people km⁻² is assumed (total population 200000) and the collective dose is calculated for

100 and 500 years. The effect of the remediation technologies on the collective dose, for both integration periods is given in Table 6.5.

Table 6.5 The impact of remediation technologies on collective dose (100 year and 500 year) for the Tessenderlo phosphate processing site (Belgium)

Remediation Technology	Collective Dose (manSv) [†]		Ranking
	100 year	500 year	
No treatment	2.4 - 100	11.8 - 502	--
Removal of source	0.24 - 22	1.2 - 111	9
Soil washing	0.40 - 33	2.0 - 16.6	6
Flotation	0.40 - 33	2.0 - 16.6	6
Chemical extraction	0.20 - 20	1.0 - 10.0	4
Capping	3.0×10^{-4} - 1.5	1.5×10^{-3} - 7.6	3
Sub-surface barrier (grout curtain)	2.3 - 100	11.6 - 502	10
Cement-based solidification(<i>ex-situ</i> and <i>in-situ</i>)*	0.29 - 16	1.4 - 82.0	7
Chemical-based solidification (<i>ex-situ</i> and <i>in-situ</i>)*	0.28 - 17	14.0 - 84.3	8
Flotation + capping	4.9×10^{-5} - 0.39	2.5×10^{-4} - 1.9	2
Cement-based solidification (<i>in-situ</i>)* + capping	4.7×10^{-5} - 0.25	2.4×10^{-4} - 1.2	1
Chemical extraction + cement-based solidification (<i>ex-situ</i>)*	3.0×10^{-2} - 3.2	0.15 - 16.2	5
Sub-surface barrier + capping	3.0×10^{-4} - 1.5	1.5×10^{-3} - 7.6	3

Note: * Mobility factor used for all radionuclides including ²²²Rn.

† Range refers to the 5 to 95th percentile values.

The dominance of exposure pathways involving ²²²Rn inhalation mean that remediation technologies which reduce the radon flux are most effective at reducing the collective dose (see Section 6.4.1). As was the case for the critical group dose technologies incorporating capping would have been most successful at reducing the collective dose. The installation of sub-surface barriers, which would not reduce the radon flux, would have had negligible impact.

6.4.3 Workforce dose during remediation activity

The workforce considered in this part of the study are taken to be contractors who will be employed to carry out the remediation of the site. For simplicity, it is assumed that during the period of remediation they live locally, but after the completion of the remediation they will leave the area.

Calculating the dose received by the workforce during remediation can be complex, as the workforce exposure will change over time, when and as the process of remediation itself proceeds. Depending on the technique applied, and the timing of actions, the workforce dose may decline gradually throughout the operations (e.g. with *in-situ* solidification), it may increase initially (e.g. if excavation of waste removes overlying soil and thus reduces shielding), or it may decrease rapidly (e.g. where a surface cap is applied). This is illustrated conceptually in Figure 6.1.

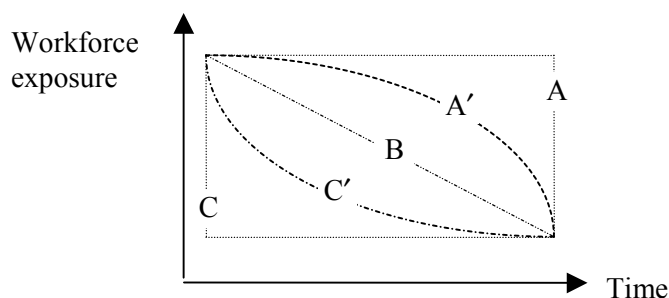


Figure 6.1 Possible changes in the levels of exposure to the workforce during remediation

In Case A and A', the level of exposure remains high until the end of the remediation, possibly corresponding to excavation of waste with enhanced external exposure and radon emanation (e.g. for the construction of sub-surface barriers). Case C or C' may occur if exposure to the source rapidly ceases (e.g. when a surface barrier is constructed with a soil covering).

In reality, doses may arise in a very much more complex fashion than indicated here. Case A is used in this study to calculate the exposure of the workforce, and is likely to represent a conservative simplification in most cases.

The workforce habits must also be defined. Generalised assumptions, which apply for each remediation option are summarised in Table 6.6, and are based on habits presented in Chapter 5 for a non-specific workforce. Whilst the inclusion into the model of factors such as off-site habitation and consumption of local foodstuffs will be of negligible importance, they are included to retain consistency with the other dose calculations.

Table 6.6 Summary of generalised workforce assumptions

	Workforce
Off-site place of habitation	village 1 km away
Time spent at habitation (h)	specific to remediation option
Outdoor occupancy at habitation (%)	33%
Workplace	waste site
Time spent at work (h)	specific to remediation option
Outdoor occupancy at workplace (%)	33%
Local source of terrestrial foodstuffs	field 100 m away from site
Local food intake (% of total food intake)	25%
Local source of aquatic foodstuff source	none
Time spent fishing (h)	none
Local source of drinking water	none

Much of the dose to the workforce is determined by the amount of time required to undertake a remediation technique. These are defined in Annex C1 and summarised in Table 6.2. It is assumed that for each hour occupied on-site, there is an associated off-site occupancy of 3.5 hours. This is based on a working week of 37 hours, which implies 131 hours spent off-site during the week, all of which is assumed to be at the place of habitation. In practice, contractors may work longer hours and spend less time at a place of local habitation. However, these present assumptions will probably not underestimate the total workforce dose.

The number of man hours needed for remediation may be calculated from the contents of Table 6.2 and the dimensions of the contaminated waste (see Section 6.4). The number of man hours needed to remediate the site may be used to calculate the period of occupancy using the method described above. AMCARE calculates the annual dose received by the workforce. The workforce exposure can, therefore, be calculated as the product of the annual dose and the occupancy.

The resultant doses arising from each remediation technology are summarised in Table 6.7.

It can be seen that capping, which has previously been identified as one of the most effective remediation options (see Tables 6.4 and 6.5), incurs one of the lowest workforce doses.

Table 6.7 Workforce exposures arising from different remediation technologies, for the Tessengerlo site

Remediation Technology	Dose to Workforce [†] (manSv)	Ranking
No treatment	0	--
Removal of source	0.88 - 14.0	6
Soil washing	1.09 - 21.0	7
Flotation	1.09 - 21.0	7
Chemical extraction	5.3 - 49.0	9
Capping	0.01 - 0.47	2
Sub-surface barrier (grout curtain)*	2×10^{-3} - 0.05	1
Cement-based solidification (<i>ex-situ</i>)	1.09 - 21.0	7
Cement-based solidification (<i>in-situ</i>)	0.26 - 5.6	4
Chemical-based solidification (<i>ex-situ</i>)	1.09 - 21.0	7
Chemical-based solidification (<i>in-situ</i>)	0.26 - 5.6	4
Flotation + Capping	1.10 - 21.5	8
Cement-based solidification (<i>in-situ</i>) + Capping	0.27 - 6.1	5
Chemical extraction + Cement-based solidification (<i>ex-situ</i>)**	5.5 - 56.0	10
Sub-surface barrier + Capping	0.01 - 0.52	3

Note: * The surface area of the barrier is taken to incorporate four sides of the waste and to be 20 m deep; ** A single excavation of the site is assumed; † Range refers to the 5 to 95th percentile values.

6.4.4 Cost of remediation based on the Tessengerlo site

The cost of applying each remediation technology may also be calculated using the data given in Table 6.2 and the dimensions of the waste at the Tessengerlo site. Costs are summarised in Table 6.8. It can be seen again that capping, previously noted as one of the more effective remediation options for Tessengerlo, and incurring one of the lowest workforce doses, is also the cheapest option.

Table 6.8 Illustrative costs arising to remediate the Tessengerlo Site

Remediation Technology	Cost (10 ⁶ ECU)	Ranking
No treatment	0	--
Removal of source	4050 - 13455	14
Soil washing	1620 - 6435	11
Flotation	533 - 3899	7
Chemical extraction	1458 - 8118	12
Capping	16 - 24	1
Sub-surface barrier (grout curtain)*	18 - 24	2
Cement-based solidification (<i>ex-situ</i>)	608 - 2970	4
Cement-based solidification (<i>in-situ</i>)	405 - 3069	5
Chemical-based solidification (<i>ex-situ</i>)	891 - 5643	10
Chemical-based solidification (<i>in-situ</i>)	486 - 4158	9
Flotation + Capping	549 - 3923	8
Cement-based solidification (<i>in-situ</i>) + Capping	421 - 3093	6
Chemical extraction + Cement-based solidification (<i>ex-situ</i>)	1616 - 9738	13
Sub-surface barrier + Capping	34 - 48	3

Note: * The surface area of the barrier is taken to incorporate four sides of the waste and to be 20 m deep. ** A single excavation of the site is assumed.

6.4.5 Fractional removal of contaminant material

It has been noted in Section 6.4 that public reassurance following remediation action at any site may be linked to the fractional removal of contaminant material. The degree of public reassurance is difficult to quantify and, in Annex C1, is ranked simply as high, moderate or

low. Table 6.9 indicates the fractional removal of contaminant and volume reduction of waste following each of the remediation options considered for Tessenderlo. In this case, capping and other containment measures would be deemed unlikely to secure high public reassurance as all the contaminant material remains on site.

Table 6.9 Fractional removal of contaminant material for remediation options at the Tessenderlo Site

Remediation Technology	Fraction of radioactivity removed from site	Fraction of waste volume removed from site
No treatment	0	0
Removal of source	0.09 - 0.95	0.50 - 0.90
Capping	0	0
Soil washing	0 - 0.90	0.02 - 0.50
Flotation	0 - 0.90	0.01 - 0.72
Chemical extraction	0 - 0.90	0
Sub-surface barrier (grout curtain)*	0	0
Cement-based solidification (<i>ex-situ</i>)	0	0
Cement-based solidification (<i>in-situ</i>)	0	0
Chemical-based solidification (<i>ex-situ</i>)	0	0
Chemical-based solidification (<i>in-situ</i>)	0	0
Flotation + Capping	0 - 0.90	0.02 - 0.50
Cement-based solidification (<i>in-situ</i>) + Capping	0	0
Chemical extraction + Cement-based solidification (<i>ex-situ</i>)	0 - 0.90	0
Sub-surface barrier + Capping	0	0

6.5 Discussion

Investigating the different remediation options shows that several technologies would be inappropriate for this particular site. Tessenderlo contains a relatively high total inventory of ²²⁶Ra, although the concentration is generally low, and those technologies which do not reduce ²²²Rn efflux (and associated ²²²Rn) are therefore largely ineffective. In this context, source term removal or surface capping are the preferred remediation options. Removal of the contaminant material clearly reduces the rate of radon generation (although the subsequent burial site may need further consideration) whereas capping acts to reduce the radon efflux rate. Nonetheless, other options, and combinations of options, have been considered here for illustrative purposes.

These options are ranked in terms of effectiveness in Table 6.10. The performance of options which include capping were found to be about an order of magnitude more effective than the other options.

Table 6.10 ranks the remediation options in terms of the cost of application. The costs of applying some of the remediation options are found to be prohibitively high. In particular, those which require excavation of the site. This is essentially due to the fact that Tessenderlo contains a very large volume of material, generally contaminated at low levels. This tends to disadvantage options involving source term removal. By contrast, public reassurance is likely to be highest for options which do include source term removal.

Considering dose reduction to the public, workforce dose during remediation work and potential costs, it is likely that capping alone would be the preferred option at Tessenderlo. A quantified cost-benefit analysis of options is presented in Chapter 7.

Table 6.10 Ranking of remediation technologies in terms of greatest reduction of critical dose and also in terms of lowest cost of applying the technology to the Tessenderlo site

Ranking of Impact of Remediation Options on Critical Dose (Most Effective First)		Ranking of Costs of Remediation Options (Lowest Cost First)	
1	Cement-based solidification (<i>in-situ</i>) + Capping	1	Capping
2	Flotation + Capping	2	Sub-surface barrier (grout curtain)
3	Capping	3	Sub-surface barrier + Capping
3	Sub-surface barrier (grout curtain) + Capping	4	Cement-based solidification (<i>ex-situ</i>)
4	Chemical extraction + Cement-based solidification (<i>ex-situ</i>)	5	Cement-based solidification (<i>in-situ</i>)
5	Cement-based solidification (<i>ex-situ</i>)	6	Cement-based solidification (<i>in-situ</i>) + Capping
5	Cement-based solidification (<i>in-situ</i>)	7	Flotation
6	Chemical-based solidification (<i>ex-situ</i>)	8	Flotation + Capping
6	Chemical-based solidification (<i>in-situ</i>)	9	Chemical-based solidification
7	Chemical extraction	10	Chemical-based solidification (<i>ex-situ</i>)
8	Removal of source	11	Soil washing
9	Soil washing	12	Chemical extraction
9	Flotation	13	Chemical extraction + Cement-based solidification (<i>ex-situ</i>)
10	Sub-surface barrier (grout curtain)	14	Removal of source

As was alluded upon in Chapter 5, the aim of CARE is not to engage in site-specific assessments. AMCARE is a generic assessment model, flexible enough to incorporate site specific parameters and also the estimates for the dose reduction potential of the different options are generic. The aim of CARE was to give generic illustrations which may be used as a first, coarse screen of options. A more detailed site specific approach would be required as the final selection of and decision on remediation options is to be made.

6.6 References

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7. DERIVATION OF CRITERIA AND SELECTION OF REMEDIATION OPTIONS

7.1 Introduction

The distinction between *practices* and *interventions*, as recommended by international radiation protection organisations, may not always be clear for clean-up of land that has been contaminated with radioactive materials. However, in cases where there is existing exposure of a population from sites contaminated with the residues of past practices or work activities, the principles of protection for *intervention* are applicable. In the context of remediation of such 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. The optimisation process for selecting the best strategy of remedial measures should, in addition to averted radiation detriment and monetary costs, include considerations on how measures can gain reassurance (reduce anxiety) of the affected population and achieve - to the extent practicable - conditions of *return to normality* without any restrictions associated with the residual contamination to the extent practicable.

7.2 Methodology for selection of remedial measures

The formulation of the principle of optimisation of protection for a practice and an intervention differ. However, the practical implementation of the optimisation of remedial measures for contaminated sites is essentially the same process, whether it is considered in the context of the continuing operation of a practice, as part of decommissioning of a practice, or for intervention. In all cases, it includes the identification of the remediation options available and how the exposures might be reduced, and also choosing that remedial action which results in the greatest net benefit, considering all of the relevant factors that influence costs and benefits. These costs and benefits may include populations directly affected by the measures, both now and in the future, as well as to other parts of society. Decisions on remediation may go far beyond purely radiological protection considerations but can, however, often be limited to considerations of whether or not any of the range of possible remedial actions will themselves result in a net benefit. In reaching such decisions it is important to consider, carefully, the benefits and disadvantages of the remedial actions because some actions can significantly disrupt the affected population or have serious impact on the environment.

As recommended by the ICRP (1990) dose limits do not apply to intervention situations. Many national action levels for remedial measures for contaminated sites are set at an annual effective dose of 1 mSv, which is numerically equal to the limit of population exposure from all practices. It seems, therefore, that national levels for intervention are not always based on an optimisation of protection of the affected population but rather on long-term exposures from the residues of old practices or events, which have been judged - not fully in accordance with international guidance - to be unacceptably higher than the dose limit for ongoing practices. Further development and guidance is therefore needed to aid decisions on implementation of remedial measures at sites contaminated by past or old practices or work activities.

7.2.1 Normalised Action Levels for remedial measures

For practical purposes measurable (operational) quantities such as radionuclide concentration, or dose rate, are needed to evaluate the effect of remedial measures in relation to radiological protection criteria. Such quantities are named *action levels* and they are related to the primary criterion, *e.g.* avertable dose, by suitable models for dose assessment from all relevant exposure pathways. Compliance with the action level would thus, ensure compliance with the primary criterion.

Action Levels can be used as a *screening tool* to determine if a remediation would be justified. They can be derived from generic values of environmental conditions and typical values of efficiency of the remedial measure. Action Levels correspond to dose levels and they are derived by mathematical modelling of all the significant pathways of exposure and the projected relevant behaviour of the average member of the affected population. To calculate an Action Level above which a specified remedial measure is to be taken requires a detailed understanding on a number of site specific factors. The most important of these factors include:

- the total costs of the remedial measure, X ;
- the equivalent cost of the collective dose to the workers implementing the measure;
- the number of people exposed by the contamination, N ;
- the reduction factor of the collective dose to the exposed population, f ; and
- the dose conversion factors for the radionuclides present in the contamination.

The procedure for deriving an Action Level is explained in Annex D4.

The minimum value of an Action Level, above which a remedial measure is justified, varies considerably with the site-specific parameters and the cost of the remedial measure. If the Action Level is normalised to the dose conversion factor, the following expression can be used to calculate a generic value of the normalised Action Level ($\text{man}\cdot\text{Sv}\cdot\text{a}^{-1}$) for typical values of the cost of the remedial measures, X , and the dose conversion factor, $\sum r_i \cdot G_i$ (see Annex D4):

$$AL_{norm} = AL \cdot \left(\sum_i r_i \cdot G_i \right) = \left(\frac{f}{f-1} \right) \cdot \left(\frac{X}{\alpha \cdot T} \right)$$

The normalised Action Level, AL_{norm} , has the unit of an annual collective dose as it arises from the product of activity concentration in the contaminated media (Bq kg^{-1}) and the annual collective dose per unit concentration ($\text{man}\cdot\text{Sv}\cdot\text{a}^{-1} / \text{Bq}\cdot\text{kg}^{-1}$) of the dominating radionuclide.

The parameters for calculating the Action Level have been assigned the following generic values:

α	75000 - 200000 ECU Sv^{-1} (uniform distribution)
T	100 - 500 years (uniform distribution)
f	1 - 50, most probable value 10 (triangular distribution)
X	0.67 - 1.5 times the value in a generic range of 10^4 - 10^{10} ECU (uniform distribution)

Generic values of the normalised Action Level, $(AL) \cdot (\sum r_i \cdot G_i)$, have been calculated by Latin Hypercube sampling and the results are shown in Figure 7.1 as a function of the monetary cost of the remedial measure, X .

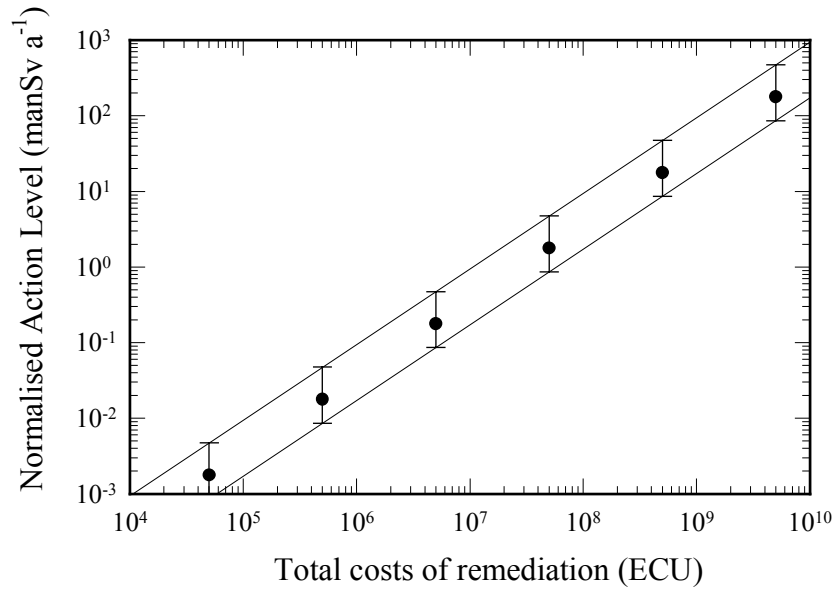


Figure 7.1 Normalised generic Action Levels for remedial measures as a function of the total costs of these measures

The error bands in Figure 7.1 represent the 5% - 95% percentiles in the calculated distributions. For the assumed value distributions of the parameters, given above the sensitivity of these on the calculated normalised Action Levels have been determined as:

- integration time, T : - 83%
- equivalent monetary value of a unit collective dose, α : - 38%
- normalised cost of remedial measure, X : + 36%
- dose reduction factor, f : + 30%

The normalised Action Levels can be used to calculate real Action Levels (AL) for screening the different remediation options at the considered site:

$$AL = \frac{AL_{norm}}{\sum_i r_i \cdot G_i}$$

The application of normalised Action Levels is presented in Figure 7.2.

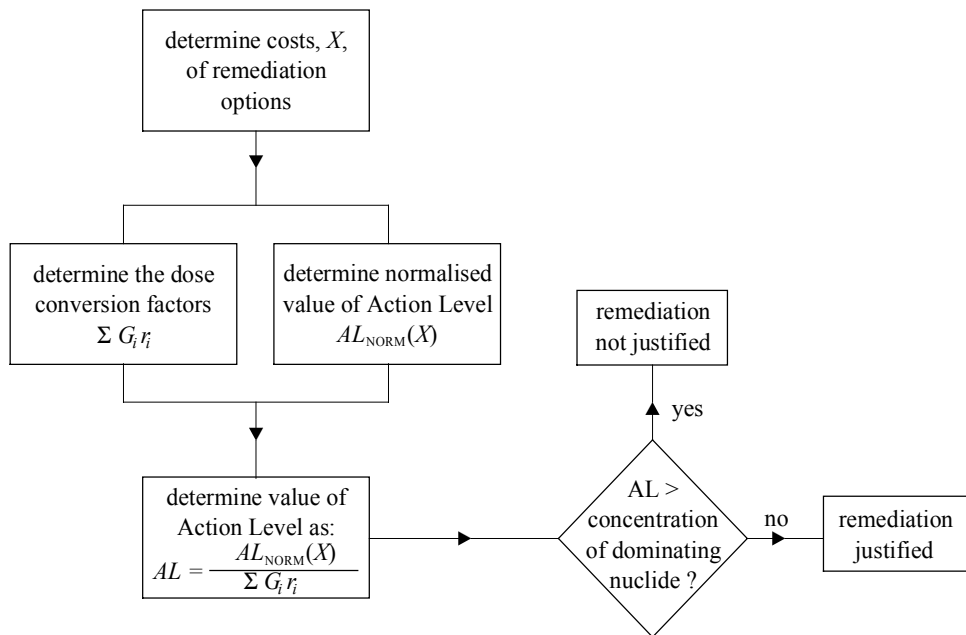


Figure 7.2 Flow chart for the application of generic Action Levels in situations where remedial measures are to be applied at contaminated sites

When detailed information of different remedial measures can be found, *e.g.* the costs and the efficiencies, specific calculations of an Action Level for each remedial measurement can be made and more precise conclusions, on justified and unjustified measures, can be drawn.

7.2.2 Optimisation of remedial measures

The consequences of the impact of a contaminated site not only include the increased risk of stochastic radiation effects attributable to the exposure from contamination, but also the increase within psychological strain in the affected population. There can also be mental distress and anxiety associated with the contamination, regardless of whether an actual radiation dose has been received or not. This is attributable to the perception of risk which depends, in part, on whether people have confidence that the authorities are competent and trustworthy. The quantification of such social factors is not an easy task and the comparison and trade-off between radiological protection factors and social factors is, therefore, extremely difficult.

Radiological protection factors are defined as those that are related to the level of radiological protection achieved. They include those factors which describe the dose distribution averted and those which describe the costs and other disadvantages incurred in averting the doses. *Social factors* are defined as those, which are not related to the level of radiological protection achieved by the remedial measures. They can have an important or even overriding influence on the decisions taken. Many remedial measures can be disruptive to normal social and economic life. However, the absence of protective measures can also cause anxiety, which is often exacerbated by a lack of objective information.

The overall health protection of people should be based on an optimised remediation strategy, which includes both radiation protection factors and social factors. The overall health consequences include the increased stochastic risks directly attributable to the contamination. They also include the perception of the hazard posed by radioactive materials dispersed into the environment and enforced changes of lifestyle which lead to increases in psychological strain in the affected population. Such increases may in turn lead directly, or indirectly, to increased illness.

Decisions on the introduction of remedial measures for long-lasting exposure situations can often be limited to considerations of whether or not any of the possible remedial actions will result in a net benefit. If so, the optimum measure can be taken as the one having the largest net benefit. In reaching such decisions it is important to consider carefully the benefits and disadvantages because some remedial actions can significantly disrupt the exposed population.

The analysis should address both radiological and non-radiological issues. Examination of the former will, in principle, be straightforward since it involves only the radiation detriment to be averted and the costs associated with the remedial action (including both the direct cost of the action and costs to affected parties). Examination of the latter issues involves, in addition to consideration of other hazards (such as those associated with chemical contaminants), economic and social considerations, some of which are beyond the scope of radiation protection. If it is determined that some remediation is justified on either of the above grounds, then the next step is to optimise the proposed remedial action.

Depending on the characteristics of a radioactively contaminated site, different remediation techniques will be appropriate for restoration of that site. Several factors (attributes) have to be considered in the selection of an optimum remediation strategy, e.g. *effectiveness* of the remediation with respect to *dose reduction*, *monetary* and *social costs*, *impact on the environment*, *acceptability* of the public, *personnel safety* etc. The field of multiple attribute analysis offers a number of approaches to provide structure and support to the decision-making process. In the case of restoration of a contaminated site, there are several attributes that need to be considered when choosing an 'optimum' restoration strategy. The attributes that have been considered in this study include both radiological protection factors and social aspects.

- *Health attributes*
 - collective radiation doses to members of the public
- *Cost attributes*²
 - costs of remedial actions, including costs of disposal of generated waste
 - equivalent monetary costs of collective dose to workers
- *Social attributes*
 - reassurance of the public

The attribute hierarchy to be used for selection of an optimum restoration strategy can be structured as shown in Figure 7.3.

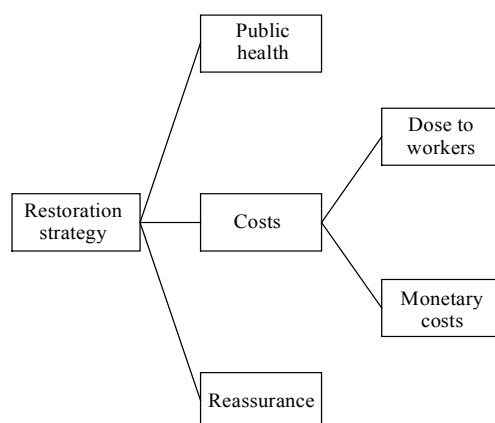


Figure 7.3 Attribute hierarchy for restoration of a contaminated site

It is assumed in this study that reassurance and risk of psychological harm are related. If the population is fully reassured there is no risk of psychological harm whereas a decreasing reassurance can be taken as an increasing risk of psychological harm. The risk per unit residual

² The attribute 'costs' are in this report considered as 'detriment' and includes doses to workers

dose has been assumed to be linked to the level of residual dose, i.e. the larger the residual dose the larger the risk of psychological harm in the affected population. This approach has been used in the optimisation of remedial measures for a selected European site (see Section 7.3.2).

A utility, u , or utility function, $u(x)$, expresses the score or utility of a given attribute with value, x , for a given protective option. A risk neutral utility function (see Annex D3) 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 utilities, u , and the weighting factors, w , can be expressed in an additive form to give the overall evaluation of each of the strategies, i , or options (see Annex D3):

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

U_i is here the overall evaluation of alternative i , w_j is the weight assigned to attribute j and u_{ij} is the score or utilities of the n factors associated with each of the alternative i on attribute j . The higher the figure of merit, U_i , the better the overall ranking of the option. Non-radiological health effects, e.g. from exposure of non-radioactive contaminants can be included in the analysis. The attributes for both radiation and non-radiation exposure would, however, have to be expressed on a risk scale rather than on a dose scale in order to determine the total expected detriment from the different types of exposures. 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.

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

Several software systems for uncertainty analysis and decision making between competing options are on the market. One of these systems is V•I•S•A from the company Visual Thinking [Visual Thinking International Limited, 1995]. This software system can be used to support the decision-making process. 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 priorities and values. Another system is Crystal Ball from the company Decisioneering [Decisioneering, 1996]. It has the advantage of working on spreadsheets enabling the development of rather complex models; uncertainties can be assigned to model parameters and correlations made between them. Crystal Ball provides a statistical picture of the range of possibilities inherent in the parameter assumptions. Crystal Ball uses either a Monte Carlo or a Latin Hypercube sampling method to generate random numbers within the assigned parameter distributions. The forecast is calculated with its own distribution from a set of, e.g. 5,000 - 10,000 simulations from which descriptive statistics can be interpreted. Also the sensitivity of the forecast to the different parameters can be analysed. In this study the Crystal Ball system has been used.

7.3 Assessments of remediation strategies for a selected European site

One of the nine main categories of industries extracting or processing material containing naturally occurring radionuclides is that of the phosphate industry. The phosphate industry in

Europe processes ores originating mainly from North Africa and the USA. The ores are used for production of phosphoric acid with the generation of phosphor-gypsum or CaF_2 as by-products. Phosphoric acid production plants within the EU are located mainly in France, Belgium and Spain. Assessments have been made of the best remediation strategies for a selected site of this type, using site-specific data on dose estimates, monetary costs and social costs. Tessenderlo Chemie processes Moroccan ores at Tessenderlo and Ham in Belgium. The process results in production of waste containing predominantly CaF_2 . The waste contains ^{226}Ra in a concentration of 3,000 - 4,000 Bq kg^{-1} . One of the dump sites consists of three separate disposal sites of which one has an area of 25 ha. Operational criteria in terms of Action Levels above which remedial measures would be justified have been derived for a number of remedial measures for this site based on generic data. To aid the selection of the best option amongst those being justified multi-attribute analyses have been used.

7.3.1 Action Levels for a phosphate industry site

A site contaminated with radionuclides might cause exposure of the surrounding population and the activity of the radionuclides present will determine the individual doses to the population. Remedial measures to be applied at the site can reduce this exposure but will give rise to both monetary costs and equivalent monetary costs of the collective dose to the workers implementing the remedial measures. These characteristic parameters can be used to derive normalised Action Levels for screening purposes. Table 7.1 shows the total costs and dose reduction factors for different remediation options for the Tessenderlo site in Belgium. The average reduction factor for a specific remedial measure, f , has been taken as the ratio of the geometric mean of the 500 years collective dose with no remediation and with the remedial measure considered and the range as 0.67 - 1.5 times the average value.

Table 7.1 *Costs and dose reduction factors for different remediation options to be applied at the Tessenderlo site in Belgium*

Remediation option	Total costs, X (MECU)	Reduction factor f
No remediation (A)	0	1
Removal of source (B)	4050 - 13455	4.5 - 10
Capping (C)	16 - 24	480 - 1080
Sub-surface barrier (grout curtain) (D)	18 - 24	1 - 1.1
Cement-based solidification (<i>in-situ</i>) (E)	405 - 3069	4.8 - 11
Cement-based solidification (<i>ex-situ</i>) (F)	608 - 2970	4.8 - 11
Chemical-based solidification (<i>in-situ</i>) (G)	486 - 4158	1.5 - 3.4
Chemical-based solidification (<i>ex-situ</i>) (H)	891 - 5643	1.5 - 3.4
Soil washing (I)	1620 - 6435	8.8 - 20
Flotation (J)	533 - 3899	8.8 - 20
Chemical extraction (K)	1458 - 8118	5 - 12
Flotation + Capping (L)	539 - 3923	3300 - 7400
Cement-based solidification (<i>in-situ</i>) + Capping (M)	421 - 3093	3300 - 7400
Chemical extraction + Cement-based solidification (<i>ex-situ</i>) (N)	1616 - 9738	33 - 75
Sub-surface barrier + Capping (O)	34 - 48	480 - 1080

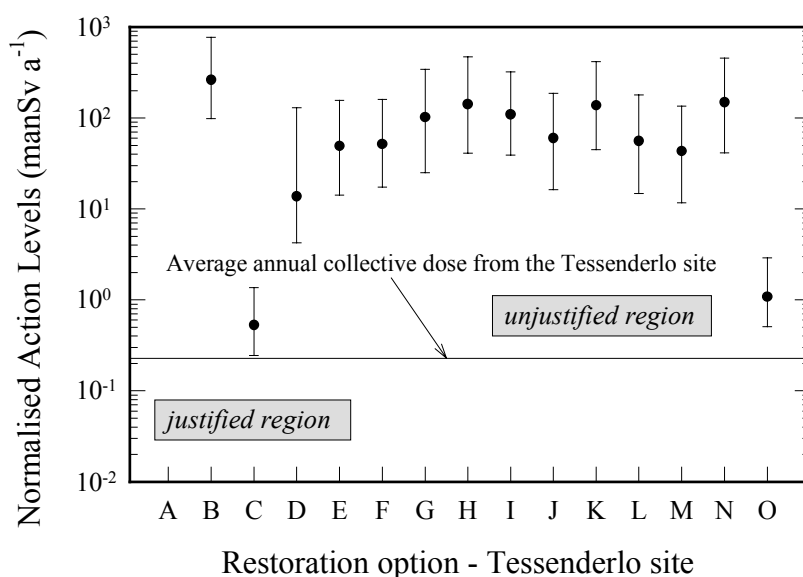
Note: *Costs are exclusively monetary costs as the equivalent costs of the collective dose to workers engaged in the remediation options are only marginal compared to the monetary costs.*

The generic estimates and assessments (Chapter 5 and 6) have been used in this study as basis for decisions on remediation activities. In reality, selection and optimisation of remediation options should not be based a generic approach, yet on site-specific assessments. The following example is presented to demonstrate the approach through an illustrative case.

The normalised Action Levels have been determined for the different remediation options for the Tessenderlo site, given in Table 7.1, according to:

$$AL \cdot \left(\sum_i r_i \cdot G_i \right) = \left(\frac{f}{f-1} \right) \cdot \left(\frac{X}{\alpha \cdot T} \right)$$

The calculated normalised Action Levels for different remediation strategies at the Tessenderlo site are shown in Figure 7.4.³

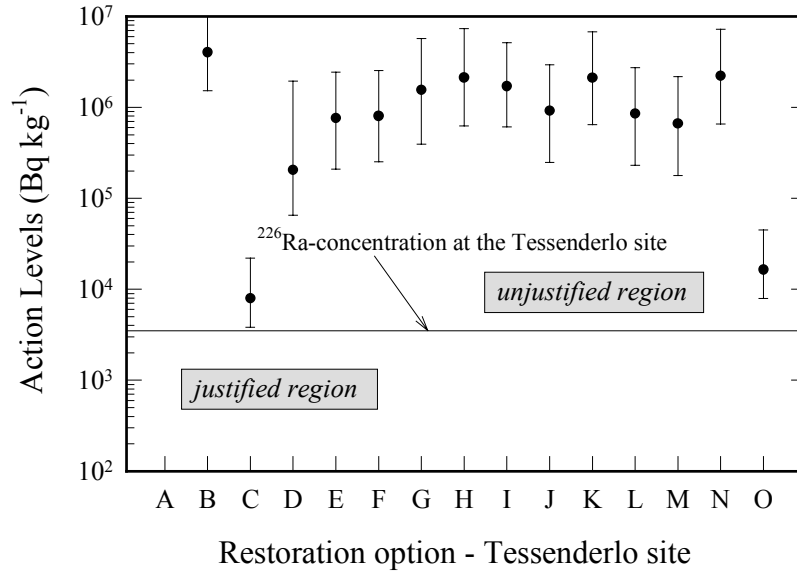


Note: Normalised Action Levels above the average annual collective dose to the affected population represent unjustified remedial measures. Normalised Action Levels below the average annual collective dose to the affected population represent justified remedial measures.

Figure 7.4 Calculated normalised Action Levels for remediation options at the Tessenderlo site in Belgium

The radiation exposure from the contamination at the site can be described by the individual doses to the surrounding population per unit concentration of the radionuclides present in the contaminated media. For the Tessenderlo site a generic value of the dose conversion factor of about 6.51×10^{-5} manSv a⁻¹ per Bq kg⁻¹ has been used (see Chapter 5).

³ The Action Level concept has been used in this study as a simple screening tool based solely on monetary costs and averted collective doses. This cost-benefit method should be seen as a justification process to indicate which of the remedial options are justified on purely economic grounds. The multi-attribute analysis method is applied in this study for the ranking/optimisation process in which also other factors, e.g. social factors, are included. Therefore, there is not a full consistency between the applied justification and optimisation procedures. However, these other factors could also be included in the cost-benefit derived Action Levels.



Note: Action levels above the actual radionuclide concentration represent unjustified remedial measures. Action levels below the actual radionuclide concentration represent justified remedial measures.

Figure 7.5 Calculated Action Levels for remediation options at the Tessenderlo site in Belgium

With the normalised Action Level and the dose conversion factor, given above, an Action Level, in terms of nuclide concentration, can be determined as shown in Figure 7.5. The average concentration of ²²⁶Ra in the contaminated media at the site has been determined as 3500 Bq kg⁻¹. As the Action Level for all options are higher than 3500 Bq kg⁻¹ these options appear to be unjustified from purely economic considerations.

An Action Level should be seen only as a screening tool to select those options having a positive net benefit on purely economic grounds. In the process of a multi-attribute optimisation, non-justified options on economic grounds may have come out with high scores because other factors that are impossible to include in the Action Level concept as defined here might be more significant than economic factors. Similarly, in situations with low collective doses to the affected population the 'do nothing' option can often have a high score in the multi-attribute optimisation. This option cannot be included in the Action Level concept because of lack of both avertable dose and monetary costs. However, in the optimisation process it has always the highest value of the cost utility (100), and this may be a decisive influence when comparing the total score of this option with those of the other options.

The individual doses would, on average, be of the order of 500 μSv a⁻¹ (see Chapter 5) at the time of decision to introduce remediation. IAEA has proposed clean-up criteria in terms of individual dose [IAEA, 1997]. For an individual dose range of 100 - 1,000 μSv a⁻¹, clean-up is usually needed if a constraint for controlled practices is applied. Without the application of a constraint IAEA suggests that, for individual doses of 100 - 1,000 μSv a⁻¹, clean-up would sometimes be needed. It can therefore be concluded that, based on the IAEA recommendations, some remediation would probably be justified for the Tessenderlo site. The ICRP Task Group considerations [ICRP Task Group, 1999] imply that an individual annual dose of about 10 mSv a⁻¹ is a non-concern level for allaying individual anxieties about residual exposures. It is meant to be a trigger level for consideration of dose reduction. If remedial actions are justified at dose levels below about 10 mSv a⁻¹ the appropriate dose reduction should be found by optimisation.

7.3.2 Optimisation of remedial measures for a phosphate industry site

Several remediation options are available for reducing the exposure of the affected population. The remediation options are summarised in Table 7.2 together with information of monetary costs of the remediation and their efficiencies for collective dose reduction.

Table 7.2 Data for different remediation options for the Tessenderlo site in Belgium

Remediation option	Collective dose to population [man Sv]		Collective dose to workers [manSv]	Monetary costs [MECU]	Fraction remaining on site (most probable)
	100 a	500 a			
No remediation (A)	2.4 - 100	11.8 - 502	0	0	1
Removal of source (B)	0.24 - 22	1.2 - 111	0.88 - 14	4050 - 13455	0.05 - 0.91 (0.10)
Capping (C)	0.0003 - 1.5	0.0015 - 7.6	0.01 - 0.47	16 - 24	1
Sub-surface barrier (D)	2.3 - 100	11.6 - 502	0.002 - 0.05	18 - 24	1
Cement-based solidification (in-situ) (E)	0.29 - 16	1.4 - 82	0.26 - 5.6	405 - 3069	1
Cement-based solidification (ex-situ) (F)	0.29 - 16	1.4 - 82	1.1 - 21	608 - 2970	1
Chemical-based solidification (in-situ) (G)	0.28 - 17	14 - 84	0.26 - 5.6	486 - 4158	1
Chemical-based solidification (ex-situ) (H)	0.28 - 17	14 - 84	1.1 - 21	891 - 5643	1
Soil washing (I)	0.40 - 33	2.0 - 17	1.1 - 21	1620 - 6435	0.1 - 1 (0.33)
Flotation (J)	0.40 - 33	2.0 - 17	1.1 - 21	533 - 3899	0.1 - 1 (0.33)
Chemical extraction (K)	0.20 - 20	1.0 - 100	5.3 - 49	1458 - 8118	0.1 - 1 (0.20)
Flotation + Capping (L)	0.00005 - 0.39	0.0003 - 1.9	1.1 - 22	539 - 3923	0.1 - 1 (0.33)
Cement-based solidification (in-situ) + Capping (M)	0.00005 - 0.25	0.0002 - 1.2	0.27 - 6.1	421 - 3093	1
Chemical extraction + Cement-based solidification (ex-situ) (N)	0.03 - 3.2	0.15 - 16	5.5 - 56	1616 - 9738	0.1 - 1 (0.20)
Sub-surface barrier + Capping (O)	0.0003 - 1.5	0.002 - 7.6	0.01 - 0.52	34 - 48	1

7.3.2.1 Utility functions

In a multi-attribute utility analysis all relevant factors can be included (see Annex D3) by the use of utility value functions. The utility function for radiation induced health effects can be expressed by use of the collective doses to the population (accumulated over 100 years) and are given in Table 7.2 as:

$$u_{\text{dose, pop}}(x) = 100 \cdot \left(1 + \frac{5 \cdot 10^{-5} - x}{100 - 5 \cdot 10^{-5}} \right) \text{ for } 5 \cdot 10^{-5} \leq x \leq 100 \text{ man Sv (100 years)}$$

$$u_{\text{dose, pop}}(x) = 100 \cdot \left(1 + \frac{2.5 \cdot 10^{-4} - x}{502 - 2.5 \cdot 10^{-4}} \right) \text{ for } 2.5 \cdot 10^{-4} \leq x \leq 502 \text{ man Sv (500 years)}$$

The value of the utility function $u_{\text{dose, pop}}$ will be 100 for a collective dose of 5×10^{-5} manSv and 2.5×10^{-4} manSv (for 100 and 500 years, respectively). The value of the utility function $u_{\text{dose, pop}}$

will be 0 for a collective dose of 100 manSv and 502 manSv (for 100 and 500 years, respectively).

The utility function for the monetary costs can be expressed by use of the costs given in Table 7.2 as:

$$u_{\text{money}}(x) = 100 \cdot \left(1 - \frac{x}{13,455}\right) \text{ for } 0 \leq x \leq 13,455 \text{ MECU}$$

The value of the utility function u_{cost} will be 100 for a cost of 0 and 0 for a cost of 13455 MECU.

The utility function for the collective dose to workers can be expressed by use of the values given in Table 7.2 as:

$$u_{\text{dose, work}}(x) = 100 \cdot \left(1 - \frac{x}{56}\right) \text{ for } 0 \leq x \leq 56 \text{ man Sv}$$

The value of the utility function u_{cost} will be 100 for a collective dose of 0 and 0 for a collective dose of 56 manSv.

The utility function u_{reas} for reassurance would be linked to both the residual dose and the fraction of activity remaining on the site after the remedial measure has been implemented. However, the residual dose and remaining activity are not necessarily correlated. A remedial measure that leaves all the activity on site in a contained form (capping, surface barriers etc.) might give a substantial dose reduction and thus a low value of the residual doses. Detailed information on how social factors like reassurance are linked with individual doses and activity concentration on site is not available. Therefore, a utility function has been proposed which gives a low value only when both sub-utilities have low values:

$$u_{\text{reas}}(x, y) = 100 \cdot \left(\frac{1}{2} \cdot \left(1 + \frac{5 \cdot 10^{-5} - x}{100 - 5 \cdot 10^{-5}}\right)_{\text{dose}} + \frac{1}{2} \cdot \left(1 + \frac{0.1 - y}{1.0 - 0.1}\right)_{\text{activity}} \right)$$

for $5 \cdot 10^{-5} \leq x \leq 100 \text{ man Sv}$ and $0.1 \leq y \leq 1$ (100 years)

$$u_{\text{reas}}(x, y) = 100 \cdot \left(\frac{1}{2} \cdot \left(1 + \frac{2.5 \cdot 10^{-4} - x}{502 - 2.5 \cdot 10^{-4}}\right)_{\text{dose}} + \frac{1}{2} \cdot \left(1 + \frac{0.1 - y}{1.0 - 0.1}\right)_{\text{activity}} \right)$$

for $2.5 \cdot 10^{-4} \leq x \leq 502 \text{ man Sv}$ and $0.1 \leq y \leq 1$ (500 years)

where y is the fraction of activity remaining on site after the remedial measures has been implemented. The value of the utility function u_{reas} will be 100 for a residual dose of $5 \times 10^{-5} \text{ manSv}$ and $2.5 \cdot 10^{-4} \text{ manSv}$ and a remaining fraction of the initial activity 0.1 (for 100 and 500 years, respectively). The value of the utility function u_{reas} will be 0 for a residual dose of 100 manSv and 502 manSv and a remaining activity fraction of 1.0 (for 100 and 500 years, respectively).

7.3.2.2 Weighting factors

The overall evaluation of the score U_i of the remediation options i has been determined from the weighted sum of utilities of each of the attributes considered as shown in Figure 7.2:

$$U_i = \sum_{j=1}^3 w_j u_{ij} = w_{\text{dose, pop}} \cdot u_{\text{dose, pop, } i} + w_{\text{cost}} \cdot (w_{\text{dose, work}} \cdot u_{\text{dose, work, } i} + w_{\text{money}} \cdot u_{\text{money, } i}) + w_{\text{reas}} \cdot u_{\text{reas, } i}$$

Determination of weighting factors is the crucial point of any multi-attribute analysis because subjective judgements will inevitably enter the process.

Weighting factors for major attributes

The primary or major attributes are the *economical*, the *health related* and the *reassurance* attributes, which are difficult to determine and compare, as they are 'measured' in different units. The methodology used here is to establish conversion/scaling constants between the weighting factors expressed as:

$$\frac{w_{cost}}{w_{dose,pop}} = C_1 \quad \text{and} \quad \frac{w_{reas}}{w_{dose,pop}} = C_2$$

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

$$w_{dose,pop} + w_{cost} + w_{reas} = 1$$

which would determine the weighting factors as:

$$w_{dose,pop} = \frac{1}{1 + C_1 + C_2} \quad \text{and} \quad w_{cost} = \frac{C_1}{1 + C_1 + C_2} \quad \text{and} \quad w_{reas} = \frac{C_2}{1 + C_1 + C_2}$$

The value of C_1 can be determined from the following ratio:

$$C_1 = \frac{w_{cost}}{w_{dose,pop}} = \frac{R_{cost}}{\alpha \cdot R_{dose,pop}} = \frac{R_{money} + \alpha \cdot R_{dose,work}}{\alpha \cdot R_{dose,pop}}$$

where R_{cost} and denote the overall ranges of monetary costs, including the equivalent cost of the collective dose to workers ($R_{dose,work}$) engaged in remediation and the collective doses to the affected population, respectively. The values of R for can be found from Table 7.2 for the monetary costs and collective dose to population and workers engaged in the remediation. The value α is the monetary value of avoiding a unit collective dose (set to 100,000 ECU Sv⁻¹ for the example given below).

The weight on reassurance of the population can be determined in a qualitative way as discussed in Section 7.2.2. A *decreasing* reassurance is here interpreted as an *increasing* anxiety and thus an *increasing* risk of psychological harm. A *decreasing* dose level can also be taken to result in an *increasing* reassurance and the risk of psychological harm would be *proportional* to the level of residual dose, *i.e.* the larger the residual dose the larger the risk of psychological harm in the affected population. The risk of radiation-induced stochastic health effects, r_{rad} , is proportional to the residual dose (0.05 Sv⁻¹). If it were possible to determine the risk of psychological effects per unit residual dose, r_{psy} , in terms of loss of life expectancy the scaling factor, C_2 , could be determined as:

$$C_2 = \frac{w_{reas}}{w_{dose,pop}} = \frac{r_{psy}}{r_{rad}}$$

Intuitively, the value of C_2 would be expected to be less than one and probably significantly less than one. However, the experience gained after the Chernobyl accident was that socio-psychological factors were given much higher weight than radiation factors, which indicates that the value of C_2 would be higher than one. However, this value judgement will depend on the specific situation. In a non-accidental situation like remediation of the selected sites with small exposures of the affected population, the social factors would probably be given far less weight than in a major accidental situation like Chernobyl. Consequently, the value of the scaling factor C_2 is in this study assumed to be less than one, *e.g.* 0.2 - 0.3.

Scaling factor C_1 can be determined for a 100 and 500 years integration time for the collective dose from the values given in Table 7.2:

$$C_{1,100} = \frac{1.34 \cdot 10^{10} + 10^5 \cdot 56}{10^5 \cdot 100} = 1341 \quad \text{and} \quad C_{1,500} = \frac{1.34 \cdot 10^{10} + 10^5 \cdot 56}{10^5 \cdot 502} = 267$$

The weighting factors can thus be determined as:

$$w_{dose,pop,100} = \frac{1}{1+1341+0.25} = \underline{7.5 \cdot 10^{-4}} \quad \text{and} \quad w_{dose,pop,500} = \frac{1}{1+267+0.25} = \underline{3.7 \cdot 10^{-3}}$$

$$w_{cost,100} = \frac{1341}{1+1341+0.25} \cong \underline{0.9991} \quad \text{and} \quad w_{cost,500} = \frac{267}{1+267+0.25} = \underline{0.9953}$$

$$w_{reas,100} = \frac{0.25}{1+1341+0.25} = \underline{1.9 \cdot 10^{-4}} \quad \text{and} \quad w_{reas,500} = \frac{0.25}{1+267+0.25} = \underline{9.3 \cdot 10^{-4}}$$

Weighting factors for economic sub-attributes

The economic sub-attributes considered in this study include the *monetary costs of the remediation operation*, which includes labour costs, and the equivalent *monetary costs of collective dose to workers*. The economic sub-attributes are given in the same units and the conversion/scaling constant for these sub-attributes can, therefore, be expressed as:

$$\frac{w_{money}}{R_{money}} = \frac{w_{dose,work}}{\alpha \cdot R_{dose,work}} = C$$

where R_{money} and $R_{dose,work}$ are the ranges of monetary costs and collective dose to workers, respectively, over all the different remediation options. The sum of the weighting factors for the health sub-attributes should be 1:

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

which would determine the scaling constant, C , as:

$$C = \frac{1}{R_{money} + \alpha \cdot R_{dose,work}}$$

The weighting factors can then be determined as:

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

$$w_{dose,work} = C \cdot \alpha \cdot R_{dose,work}$$

C can be determined from the values given in Table 7.2:

$$C = \frac{1}{1.34 \cdot 10^{10} + 10^5 \cdot 56} = 7.46 \cdot 10^{-11}$$

which would give the following values of w_{money} and $w_{dose,work}$:

$$w_{dose,work} = 7.46 \cdot 10^{-11} \cdot 56 \cdot 10^5 = \underline{4.2 \cdot 10^{-4}} \quad \text{and} \quad w_{money} = 7.46 \cdot 10^{-11} \cdot 1.34 \cdot 10^{10} = \underline{0.9996}$$

The weighting factors, given above, have all been sampled within a triangular distribution with the central values as calculated above and a variation of 0.67 - 1.5 times these central values (maximum value not greater than 1). Similarly, the values of all the utilities, $u(x)$, are determined from the utility functions and triangular distributions with central values of x given as the average value of the ranges given in Table 7.2.

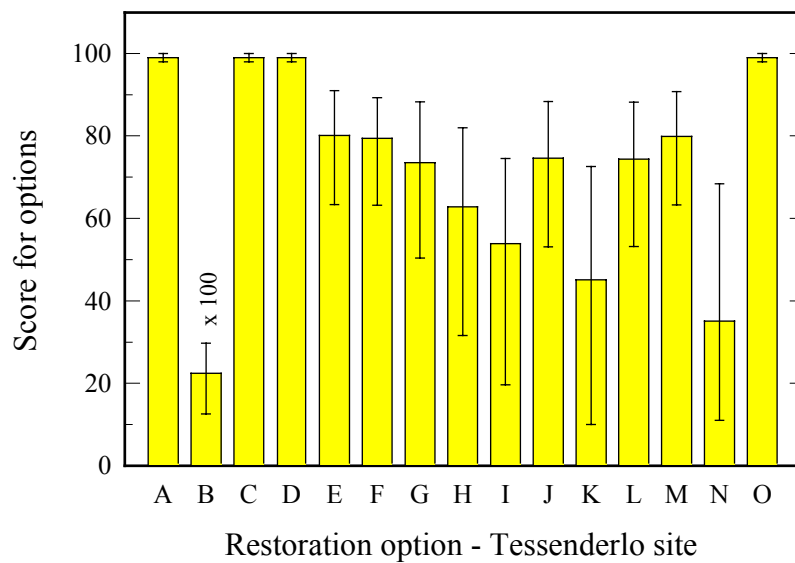
7.3.2.3 Scores for remediation options

The evaluation of the different strategies has been made with the software system CRYSTAL BALL by Latin Hypercube sampling. The distributions of the parameters in the ranking calculations have been assigned the following values:

Collective dose to population, S	Ranges as given in Table 7.2, average value as most probable value (triangular distribution)
Collective dose to workers, S_{worker}	Ranges as given in Table 7.2, average value as most probable value (triangular distribution)
Monetary costs of remediation, X_{remedial}	Ranges as given in Table 7.2, average value as most probable value (triangular distribution)
Fraction of activity left on site	Ranges and most probable value as given in Table 7.2 (triangular distribution)

In the sampling calculations the collective doses to population have been negatively correlated with the monetary costs of the remedial measures ($r = -0.8$).

The results for the scores, U_i for the Options A - O are shown at Figure 7.6. The error bars represent the 5% and 95% percentiles of the distributions of U_i . None of the options are justified from an economic point of view.



Note: An integration time of 500 years has been used in the calculation of collective doses.

Figure 7.6. Overall evaluation of scores for remediation strategies at the Tessenderlo site

The uncertainties on the different scores (5 - 95 percentiles) can be represented by normal distributions. These distributions are wider for the options with a high remediation costs than for the options with low remediation costs because the dominating weight given to the costs attributes.

The ranking has been based solely on the central values of U . Options A, C, D and O all have an equal score close to 100. The ranking of the options from best to worst has been found to be:

1. No remediation (A)
2. Capping (C)
3. Sub-surface barrier (grout curtain) (D)
4. Sub-surface barrier + Capping (O)
5. Cement-based solidification (*in-situ*) (E)
6. Cement-based solidification (*in-situ*) + Capping (M)
7. Cement-based solidification (*ex-situ*) (F)
8. Flotation (J)
9. Flotation + Capping (L)
10. Chemical-based solidification (*in-situ*) (G)
11. Chemical-based solidification (*ex-situ*) (H)
12. Soil washing (I)
13. Chemical extraction (K)
14. Chemical extraction + Cement-based solidification (*ex-situ*) (N)
15. Removal of source (B)

To determine which of the Options A, C, D or O is the optimum strategy another set of ranking calculations only for those four options are needed. The ranges of the collective doses, the monetary costs and the remaining activity fraction on site, which would determine utility functions and weighting factors, can be determined from Table 7.2 for the options A, C, D and O:

$$R_{dose, pop}: 3 \cdot 10^{-4} - 100 \text{ manSv (100 years integration time)}$$

$$R_{dose, pop}: 1.5 \cdot 10^{-3} - 502 \text{ manSv (500 years integration time)}$$

$$R_{dose, work}: 0 - 0.52 \text{ manSv}$$

$$R_{cost}: 0 - 48 \text{ MECU}$$

$$R_{frac}: 0.95 - 1$$

Based on these value ranges the weighting factors for a 500 years integrating time for the collective dose have been calculated as:

$$w_{dose, pop}: 0.453$$

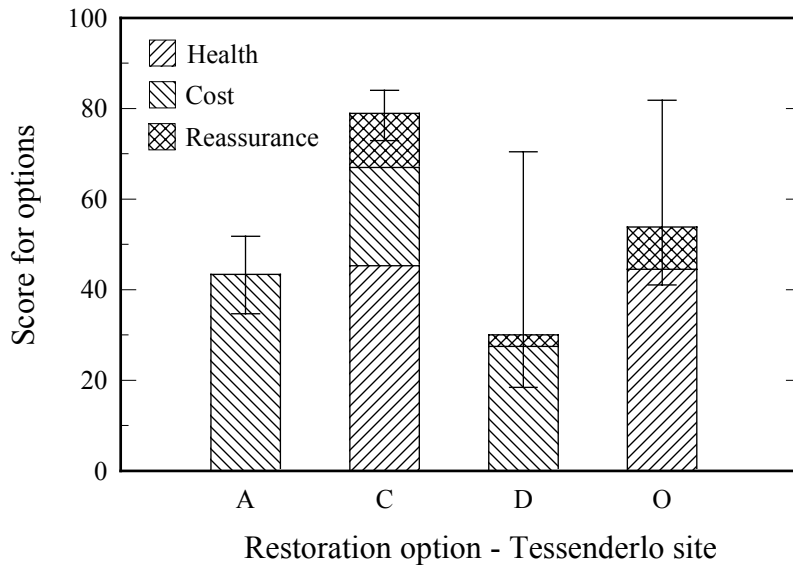
$$w_{cost}: 0.434$$

$$w_{reas}: 0.113$$

$$w_{dose, work}: 0.0011$$

$$w_{money}: 0.9989$$

The scores for the remediation Options A, C, D and O have been determined in the same way as that for the total number of options, *i.e.* with Latin Hypercube sampling within the ranges of weighting factors, w , and utility values, x , in triangular distributions. The results for the scores, U_i are shown in Figure 7.7.



Note: An integration time of 500 years has been used in the calculation of collective doses.

Figure 7.7. Overall evaluation of scores for the remediation strategies A, C, D and O at the Tessenderlo site

The remediation Option C has the highest score as indicated in Figure 7.7. The contribution of each of the attributes health, costs and reassurance is indicated for central values of the weighting factors and utility functions. The value of the utility for both reassurance and health (dose) is zero for Option A. Therefore, cost is the only attribute that contributes to the score for Option A. Similarly, the value of the utilities for the attributes health and cost is zero for Options D and O, respectively. These attributes do therefore not contribute to the score for Options D and O. The error bars represent the 5% and 95% percentiles of the distributions of the total score U_i .

7.4 Conclusions

Multi-attribute utility analyses have been used in this study to illustrate how an optimum remediation option among a number of different options can be selected. The attributes that have been used include monetary costs of the remedial measures, the collective dose to the clean-up workers, the collective dose to the affected population averted by the remedial measures, and reassurance of the population affected by these measures. Linear utility functions, so-called risk-neutral utility functions, have been used and uncertainties included as value distributions of the attributes. Two different methods have been used to determine the weighting factors, w . For attributes at the same hierarchy level given in the same unit, e.g. monetary costs, the weighting between the different attributes have been related to their value ranges, R , by the relation $(w/R)_1 = (w/R)_2 = (w/R)_3 = \dots C$. The weighting of attributes at the same hierarchy level for which the units are different, as they are for the social attributes, has been determined by assigning a value to the ratio of their weighting factors as $w_2/w_1 = C_1$, $w_3/w_1 = C_2$, $w_n/w_1 = C_{n-1}$.

For situations where both the collective dose to the affected population is relatively small (as they are for the sites considered in this study) and the monetary costs of the remedial measures are considerable, the no-remediation option would come out with a high score, sometimes even as the best score. This is due to the fact that the value of the monetary cost utility would have its maximum value for the no-remediation option and because the weight of social factors would be only marginal when doses are small. In this study a first set of ranking calculations for the Tessenderlo site in Belgium have been used to exclude those options with the lowest

scores. A repeat set of ranking calculations for the options with the highest scores were carried out and Option C (capping) was shown to have the highest score thus being the optimum remedial measure for the Tessenderlo site.

Optimisation of remedial measures using multi-attribute analysis with utility functions allows the inclusion of factors that are not easy to quantify in monetary terms. 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 useless because of a subjective bias of the decision-maker in the selection of weighting factors. Therefore, the outcome of any multi-attribute analysis should be judged very carefully in the light of the values assigned to the weighting factors before any firm conclusions could be drawn.

Justified Action Levels start at the minimum value of the avertable individual dose at which the remedial action is just beginning to do more good than harm. The Action Level can thus be defined as the lowest level at which remedial actions to reduce doses is justified. An equivalent definition is that the Action Level corresponds to the maximum level of dose attributable to the contamination without any protective actions being justified because the avertable dose by the specified action is not worthwhile in terms of the overall costs of the action.

The derivation of generic Action Levels for remedial measures would be influenced by the scale of the contamination and lead to either less restrictive (higher) or more restrictive (lower) action levels. In extreme cases, *e.g.* if high levels of contamination affects a significant fraction of the area of the country, or if remediation costs are significant compared to the Gross National Product, the remediation options available may be limited and the Action Level could be expected to be necessarily rather high. In other cases, when relatively small areas are contaminated to a moderate degree there may be significant 'socio-political' factors driving the decisions on remediation towards a complete removal of the contamination, irrespective of a strict cost-effectiveness. High values of the Action Level would emerge for sites contaminated with less toxic radionuclides, low values of the equivalent monetary value of avoiding a unit collective dose and for more expensive remedial measures. Alternatively, low values of the Action Level would emerge for sites contaminated with more toxic radionuclides, high values of the equivalent monetary value of avoiding a unit collective dose and for less expensive remedial measures.

In this study the Action Level concept has been restricted to a pure cost-benefit expression although other factors like social factors could be included in the derivation of Action Levels. For the Tessenderlo site in Belgium, Action Levels have been derived in terms of ^{226}Ra -activity concentration in the contaminated media. These levels are derived from costs of the remedial measures, efficiency of these measures, the equivalent monetary value of avoiding a unit collective dose and the averted collective dose for each measure. They should be seen as screening levels indicating that options having an Action Level below the actual activity concentration would be economically justified. The Action Levels are all above the actual ^{226}Ra -concentration on site, which means that none of the options would be justified from a pure economic point of view. A somewhat higher value of the equivalent monetary value of avoiding a unit collective dose would probably have resulted in Action Levels at which the Options C and O would have been economically justified.

International guidance from the IAEA and ICRP has not yet been completed. Both IAEA (1997) and ICRP (1999) have proposed a generic reference dose level as a dividing line between situations that might be considered as "normal" and situations where some remedial measures should normally be considered (see Annex D1). This reference dose level has emerged from analogy considerations to (a) world-wide variation in annual natural background dose, (b) action levels for radon in buildings, and (c) criteria for resettlement of populations after a nuclear accident. It is concluded that above a reference dose level of around 10 mSv a^{-1} , remedial measures should normally be considered, with an increasing need for such measures

to be taken with increasing dose above the reference dose level. At an annual dose level of about a hundred mSv, mandatory action should almost always be taken.

An overview of restoration criteria in Europe and other countries has been given (see Annex D2) based on requests to the radiation protection authorities in the respective countries. From this overview it can be concluded that except for a few countries, criteria for remediation of contaminated sites are not fully developed. The rationale for deriving remediation criteria in different countries is not very clear and conceptual differences between existing national criteria do exist.

7.5 References

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ANNEX A1
DESCRIPTION OF INDUSTRIES DEALING WITH NORMS:
GENERAL BACKGROUND, INDUSTRIAL PROCESSES,
CONTAMINATION LEVELS, MAJOR RELEASE ROUTES AND
PATHWAYS

Table of Contents

A1.1	Introduction	70
A1.2	Uranium mining and milling	70
A1.3	Metal mining and melting	78
A1.4	Phosphate industry	83
A1.5	Coal Mining and Power Production from Coal	86
A1.6	Oil and gas drilling	88
A1.7	Rare earth and titanium dioxide industries	91
A1.8	Zirconium and ceramics industry	94
A1.9	Disposal of building materials	96
A1.10	Applications of natural radionuclides radium and thorium	98
A1.11	References	100

A1.1 Introduction

It is one of the objectives of CARE to give an overview of all sites in Europe contaminated with NORM due to past practices. In this annex, the different categories will be described in terms of general background, industrial processes, source term and major release routes and pathways.

However, as commented in Chapter 4, the information available is often limited and the different categories will be described as far as this information was available to us. The inventory from available information per category and per country can be found in Annex A2.

The radiological hazards are generally more important for the uranium mining and milling industry than for the other industrial categories. Furthermore, remediation assessment or implementation of the remediation of historic sites resulting from the extraction or processing of NORM containing material, often comes down to remediating disposal sites and land surrounding the facility contaminated with similar radionuclides. Therefore, possible release mechanisms and pathways are more extensively described under the heading uranium mining and milling and to a lesser extent for the other industrial categories.

A1.2 Uranium mining and milling

[The major part of the information is extracted from IAEA (1992) and Hébert *et al.* (1996)].

A1.2.1 Category description

The purpose of the uranium mining industry is to produce material which may be used directly in manufacturing nuclear fuel after a five-step process in which the ore is extracted from the deposit (mining) to produce a high grade concentrate (ore processing), then converted into a product of nuclear purity and, in a suitable chemical form, isotopically enriched before the nuclear fuel manufacturing.

At present, twenty three countries are reported to have mining and milling activities and eight countries have a production capacity of at least 1000 tonnes of uranium per annum. Annually, 20 million additional tonnes of tailings are produced. A historical overview of the major uranium producing countries is presented in Vels and Ruhrmann (1994). The cumulative uranium production to the end of 1990 was 989×10^3 and 706×10^3 tonnes uranium for the western world and the CIS, China and Eastern Europe, respectively.

Mining operations may be carried out in open pits, underground or through *in situ* leaching. *Open pit mining* is appropriate for surface deposits and the upper layers from deep deposits. More often, access is via dip drifts with slopes of the order of 10-15%. Choosing between open-pit mining and *underground mining* is mainly determined by the number of cubic metres of barren rocks to be removed in order to extract a cubic metre of ore. *In situ leaching* usually requires lower investment and operating costs but certain site characteristics have to be met: the mineralised body must be located between waterproof layers and a thorough hydrogeological knowledge of the field and the surroundings is required. Leaching solutions (sulphuric acid or ammonium or sodium carbonate solutions) and sometimes oxidising agents are injected through wells in the mineralised zone and the leached ore is extracted through recovery wells. *In situ* leaching has the advantage of avoiding radionuclide discharge to the atmosphere but runs the risk that the solutions may migrate outside of the controlled area and into nearby aquifers.

After crushing and grinding, the ore is leached by acid (mostly applied) or alkaline treatment. The liquor containing the uranium is separated from the barren solids (e.g. countercurrent decantation, filtration) the clarified solution is then purified (ion exchange) and the uranium concentrated in stirred tanks. The uranium is precipitated from the solution using basic salts, the slurry is filtered and dried and the resulting yellow cake is sent to the refinery. Besides the

dynamic leaching processes, described above, others exist where only the crushing stage can be performed. The chemical processes remain the same, but the dissolution rate is smaller. Static leaching can be applied for poor ores or for medium or even high grade ore from small and/or isolated reserves. Static leaching methods can be divided into pile or heap leaching, in sealed "sumps" or "vats" leaching (flooding) or *in situ* leaching (injection).

A1.2.2 General description of source term.

For each 0.1% of uranium present in the undisturbed ore, the virgin ore has about 12.4 Bq of each member of the ^{238}U decay series or about 174 Bq of total radioactivity per gram.

There are two major categories of wastes produced: mining wastes and process residues.

The mining wastes consist of barren rock and cut-off grade rocks. Waste rocks having minor radioactivity levels below regulatory concern can be disposed of, used to refill the mines or to construct embankments, impoundments, roads, etc. on the site. Waste rock with levels of radioactive (or other) contaminants which could be of regulatory concern, and leached rock (from heap or vats leaching) should be disposed of or treated to meet the same requirements as mill tailings. Given their better physical properties, they are easier to cover than mill tailings. If the mill is adjacent to the mine, the integration of tailings' disposal with waste rock disposal may be advantageous [Hébert *et al.*, 1996]

Uranium mill tailings are the ground solid residues and the associated liquids resulting from the ore processing. They typically consist of a fine fraction (slimes or fines) largely made up of clayey minerals and impregnated with iron hydroxides. The fines contain 80% of the initial radium. The sands (>1.5 mm), the heavier coarser particles, contain about 20% of the initial radium.

Both slimes and sands contain chemical residues and precipitates from the mill process and a variety of heavy metal contaminants. The radioactivity remaining in the tailings depends on the ore grade, the percentage of uranium recovered and the radioactive daughter products present. Normally, about 90-95% of the uranium and about 15% of the total radioactivity of the ore is removed in the yellow cake concentrate, the rest remains in the tailings. The initial 85% of the activity decreases quickly to 70% as short-lived daughters, such as ^{234}Th , decay. ^{226}Ra is often considered as the most important radiotoxic decay product in the ^{238}U decay series (see Figure 3.1, Chapter 3). Furthermore it produces ^{222}Rn , a radioactive inert gas, which may contribute to an inhalation dose. ^{210}Pb is also an important decay product, because of its radiotoxicity and because of the high mobility of its daughter, ^{210}Po . In some pathways ^{210}Pb and ^{210}Po can contribute to a similar or even a higher dose than ^{226}Ra . The ^{226}Ra concentration in the tailings solution before treatment may vary between 10 and 200 Bq L⁻¹. In acidic solutions more than 50% of the initial ^{230}Th in the ore may be dissolved and could be of equal radiological importance since ^{226}Ra is a decay product. Neutralisation of the tailings will reduce the ^{230}Th concentration significantly [IAEA, 1992].

Large quantities of water result from the mining operations, from seepage and, to a lesser extent, from its use in drilling and dust control. This water may become contaminated with radioactive (or other toxic) materials and should be treated before discharge.

A general overview of the different mining residues is presented in Table A1.1. Table A1.2 presents typical characteristics for the uranium mill tailings.

Table A1.1 Characteristics of different mining residues

Residue	Uranium content mg kg ⁻¹		Mean total specific activity Bq kg ⁻¹	Quantity with respect to the ore	
	Max.	Mean		open-pit mine	underground mine
Barren rock	50	15-25	2000-5000	×3-30	×1-2
Cut-off grade rock	300	150	<30000	×1	×0.1-0.2
Static leaching residues	100	20	<40000	×1	×1
Dynamic leaching residues	300	50	300000	×1.1	×1.1

Source: Adapted from Hébert et al. (1996).

Table A1.2 Typical characteristics of uranium mill tailings

Tailings component	Part. size mm	Chemical composition		Radioactivity characteristics	
Sands	75-500	SiO ₂ , <1% Complex silicates of Al, Fe, Mg, Ca, Na, K, Se, Mn, Ni, Mo, Zn, U, V Some metal oxides		0.004-0.01% U ₃ O ₈ <u>Acid leaching</u> <u>Alkaline leaching</u> 1 - 4 kBq ²²⁶ Ra kg ⁻¹ 2 - 22 kBq ²³⁰ Th kg ⁻¹ No separate data	
Slimes	45-75	Little SiO ₂ Mostly very complex clay-like silicates of Na, Ca, Mg, Mn, Al, Fe Some metal oxides		U ₃ O ₈ and ²²⁶ Ra ~4 × conc. of in sands <u>Acid leaching</u> <u>Alkaline leaching</u> 5 - 15 kBq ²²⁶ Ra kg ⁻¹ 2 - 22 kBq ²³⁰ Th kg ⁻¹ No separate data	
Slimes/sands				<u>Alkaline leaching</u> ~22 kBq ²²⁶ Ra kg ⁻¹ ~22 kBq ²³⁰ Th kg ⁻¹	
Liquids	not applicable	<u>Acid leaching</u> pH: 1-2-2 Na ⁺ , NH ₄ ⁺ , SO ₄ ²⁻ , Cl ⁻ , PO ₄ ³⁻ Dissolved solids: <1%	<u>Alkaline leaching</u> pH: 10-10.5 CO ₃ ²⁻ , HCO ₃ ⁻ ~10%	<u>Acid leaching</u> 0.001-0.01% U 0.5-280 Bq ²²⁶ Ra L ⁻¹ 75-800 Bq ²³⁰ Th L ⁻¹	<u>Alkaline leaching</u> 8 Bq ²²⁶ Ra L ⁻¹ essentially no Th

Source: Adapted from ORNL (1994).

Tailings and to a lesser extent low grade rock, also contain significant amounts of non-radioactive pollutants which can be mobilised under acidic conditions (acid generation takes place when the metal sulphides present are oxidised and produce sulphuric acid) and appear in seepage, including heavy metals, rare earth's, salts and nutrients. Frequently, contaminants such as sulphates and chlorides will precede the radioactive constituents in the plume. A list of elements and compounds common to uranium tailings and piles is given by IAEA (1992). Commenting on hazardous substances other than radionuclides is outside the scope of this project. However, given the possible problems related with the presence of heavy metals and organic substances, they should be considered in the remediation planning. By way of example, the elemental composition of alum shale (from which uranium was extracted by acid leaching) and the mill tailings of the closed-down uranium mine in Randstad, Sweden is presented in Table A1.3.

Table A1.3 Elemental composition of the alum shale and the mill tailings of the uranium mine in Randstad, Sweden

Element	Alum shale (mg kg ⁻¹)	Mill tailings (mg kg ⁻¹)	Element	Alum shale (mg kg ⁻¹)	Mill tailings (mg kg ⁻¹)
²²⁶ Ra	(3500 Bq kg ⁻¹)				
U	300	64	Hg	0.3	0.3
Mo	340	330	Mn	250	110
V	750	650	Ni	200	130
Al	66000	60000	Pb	14	13
Fe	60000	56000	Sb	5	5
K	40000	38000	Ti	3800	3800
Na	2100	2000	Zn	130	100
Mg	4900	3700	C (organic)	151000	150000
Ca	9000	9000	S (total)	70000	74000
As	106	102	SiO ₂	45000	450000
Cd	2.2	0.6	CO ₃	13000	
Cr	320	300	PO ₄	2500	500
Cu	110	110	SO ₄		15000

Source: Adapted from Sundblad (1996).

A1.2.3 Radiological impact (releases and pathways)

A1.2.3.1 Releases

Important release mechanism for pollutants to the environment from tailings and/or tailings themselves or, from disposals in general, are:

- erosion of the cover or embankments
- radon emanation
- structural failure of tailings embankments
- controlled release of contaminated water
- seepage
- unauthorised removal

The principal erosion processes are water and wind erosion. Once the impoundment cover has been stabilised, wind erosion becomes less important, except in extremely arid areas. The amount of tailings dispersed by wind is a function of the physical characteristics of the tailings and the meteorological conditions and can be estimated using a soil loss equation. Water can erode through surface run-off or through sub-surface seepage.

The percentage of radon emanated from the waste or tailings' pile is determined by a variety of factors including the emanation coefficient, the self-confinement factor, adsorption, moisture, atmospheric pressure, thickness of cover and surface vegetation. The emanation factor (percentage of radon released: generally between 0.03 and 0.5) largely depends on the moisture content of the deposit and on the particle size. Tailings from alkaline extraction processes generally have higher emanation factors since the particles have to be crushed finer. The self-containment factor (radon fraction that emanates without decay) is a function of the tailings depth and density.

The probability of embankment failure will be small if the embankments are appropriately designed and constructed and when materials are employed according to the specific foundation

conditions, the seismicity and the characteristics of the impounded materials. A high stability can be expected when dams are low, gently sloped and when under-drained.

In areas with a low net evaporation, the natural water balance may be such that a controlled release to the environment is possible. However, in the majority of scenarios, if release water is not treated to remove pollutants, it could become an important release mechanism for such pollutants.

Another important release mechanism is the uncontrolled seepage of impounded liquids through the tailings (waste) dam walls and the substratum to contaminate the groundwater. Seepage flow is determined by the hydrogeological properties of the substrata (including gradients, water table and soil characteristics, such as permeabilities, porosities, fracture densities, sorption characteristics), the atmospheric conditions, structure and geographical location of the area, the infiltration potential and the long term stability of the cover over the tailings

Superficially, the coarser fractions of tailings look like clean, good quality sand. In the past, there have been cases where tailings were removed from a disposal area and used as materials for landfill and road or building construction.

A1.2.3.2 Pathways

Exposure pathways may be generalised as follows:

- atmospheric pathways which lead to irradiation by inhalation of radon and its daughters, inhalation of airborne radioactive particulates and external irradiation;
- atmospheric and terrestrial pathways which can cause doses due to ingestion of contaminated foodstuffs and external irradiation;
- aquatic pathways which can result in ingestion of contaminated water, ingestion of food produced using irrigation, fish and other aquatic biota and external irradiation.

The critical pathways at any particular site are dependent on the local environment and habits. They have to be determined for the specific site after a thorough survey of the local conditions.

The radionuclides of most concern for the *atmospheric pathways* are gaseous ^{222}Rn and its solid daughters which attach to aerosols, and airborne particulates containing ^{230}Th , ^{226}Ra and ^{210}Pb as well as uranium. The exposure resulting from airborne particulates is primarily through the inhalation of respirable particles and/or eating food which has become contaminated by ^{230}Th , ^{226}Ra and ^{210}Pb .

Deposition on soil of material released from the disposal may also lead to enhanced downwind radiation fields. The deposited material may be re-suspended and give rise to inhalation exposure or enhance concentrations in surface water when the material lands on water.

^{226}Ra is generally the nuclide of major concern in the *aquatic pathway*. Doses to the public can result from consuming contaminated aquatic biota or water or through ingestion of plants irrigated with contaminated water. Where fish is a major source of food, this pathway can dominate. Since radium migration through groundwater is retarded by soils, exposure through this pathway is generally unimportant. Doses from radionuclides such as ^{210}Pb , ^{210}Po , uranium and thorium are usually less than those from ^{226}Ra . However, under some circumstances, the release of these radionuclides can be of the same order of magnitude as or even higher than for ^{226}Ra (e.g. U-migration) and should be considered separately.

A1.2.4 Occurrence in European countries.

In Europe uranium mining has been mainly carried out in Germany, Czech Republic, France, Bulgaria, Romania, Hungary and Spain. Many of these mining and milling sites are now

abandoned and pose serious environmental problems. In Europe, 9 mining and milling sites are still in operation: France (2), Spain (1), Romania (3), Czech Republic (2) and Hungary (1).

Following the political changes in the former Eastern Bloc (Central and Eastern Europe, CEE), including former Eastern Germany, new environmental problems have been acknowledged. In many areas, the preservation and protection of the environment was often neglected in the process of uranium extraction and processing.

In the CEE countries, the following categories of conditions and scale of industry for basic environmental restoration can be identified [Toro, 1997]:

- countries with limited development of the uranium industry having small amounts of mining and milling wastes and few contaminated sites (e.g. Poland, Slovenia);
- countries with a more developed uranium industry (e.g. Hungary, Romania);
- countries with a fully developed uranium industry having many mills and mines and several impacted resources (e.g. Czech Republic, Bulgaria, former East Germany).

The problems associated with past practices in CEE include: radon release, ground water contamination, proximity of contamination to populations, lack of resources to conduct the restoration, misuse or removal of wastes for construction, absence of legislation and responsible operators, large inventories and high aerial dispersion.

The most prominent European case of environmental contamination due to mining activities is in the districts of Saxony, Thuringia and Saxony-Anhalt in former East Germany. Exceptional radiological situations have developed resulting from centuries of coal and precious metal mining. These natural resources were often found in paragenesis with uranium minerals. The legacy of the past mining activities worsened after 1945, following intense uranium exploitation by the Soviet and Soviet-German Joint-Stock Companies, SAG and SDAG WISMUT, respectively. Numerous mines, shafts, mills and other facilities were in operation and waste rock piles and tailings ponds of enormous dimensions were set up. Radioactive contaminated substances were released into the environment, resulting in environmental contamination and radiation exposure to the public. Many small mines and mills were closed down in the 1950s and 1960s with little attention to provisions other than mining safety. The production of uranium was abandoned for commercial and other reasons in 1990. After the reunification, WISMUT changed into a company of western corporate law and the federal government has committed to fund the costs and to conduct the rehabilitation of the former WISMUT sites. The emergency remedial actions have been completed and a complete rehabilitation plan is under implementation [Rönsch and Ettenhuber, 1994; Biesold *et al.*, 1994; Lange, 1996]. More information on the WISMUT sites can be found in Annex A2.1.

In former East Germany the quantity of abandoned properties and their dimensions are in general greater than the mining relics of other European countries. Further, the East German mining districts are densely populated and intensively used for industrial and agricultural activities and for recreation. Therefore, restoration and remediation of these sites is of special importance [ICRP, 1992].

The obvious consequences of traditional mining and the uranium industry, public concern and the need for decisions on restoration and remediation of radioactively contaminated sites require systematic investigations and objective evaluations of the existing radiological situation. With this intention, the comprehensive Federal Project "Registration, Investigation and the Radiological Assessment of Mining Residues" was launched and subsidised by the Federal Ministry for Environment, Natural Protection and Nuclear Safety (BMU) in 1991. The responsibility for this project was imposed on the Federal Office for Radiation Protection (Bundesamt für Strahlenschutz, BfS). In the framework of this project, a multi-stage procedure of investigation and radiological assessment had been developed and the criteria required for radiological assessment were developed [G. Henze, BfS, Pers. Comm.]. The programme for the

remediation of radioactively contaminated sites due to mining, milling and ore processing has two parts: the decommissioning and the remediation programme for the WISMUT sites and the investigation of the historical sites. In the first step "suspicious areas" have been spatially defined, resulting in 34 areas covering 1500 km². The most relevant information about NORM content of residues is collected under a Federal Project under the direction of the BfS [Henze and Weiss, 1995]. In the second phase the information was verified. Necessary additional measurements are being executed in a third phase. One of the tools being developed is the data bank ALASKA [BfS, 1992, and 1994].

In Tables A1.4, A1.5 and A1.6 information is given about the different mining liabilities and their status, the amount of ore processed and number and extent of waste dumps and tailings ponds resulting from the mining and milling operations and their radiological characteristics. Depending on information available, liabilities in all European countries are described.

Table A1.4 Uranium production and number and volume of waste rocks, ore dumps and tailings ponds and the extent of ISL-mines in Western and Eastern Europe

Country	U-production tonne	Waste rocks and ore dumps		ISL		Tailing ponds	
		No.	Volume m ³	No.	Area km ²	No.	Volume m ³
Albania ^{1,*}	0	122	77 280	0		0	0
Bulgaria ¹	confidential	551 (298) ³	13 123 846 (8 300 000) ³	13 (15 ³ 17 ⁴)	25.73 (0.6) ³	3	18 500 000
Czech Republic ¹	118 615 (103 000) ⁴	349 (544) ⁴	56 898 000 (80 000 000) ⁴	2	6.52	21 (20) ⁴	42 822 000 (32 000 000) ⁴
Estonia ¹	100 822	1	30 000	0		1	8 000 000
Finland ⁷	Experimental			0		1	0.03 Mt
France ^{4,6}	78 730			0			47.332 Mt
Germany ^{4,8,9} -former East	220 000	48	311 000 000	?	55 × 10 ⁶ m ³ rock/aquifer affected	14	161 000 000
Hungary ¹	20 100	9	9 870 000	1	<0.001	2	15 700 000
Poland ¹	695	38	1 303 600	0		1	113 800
Portugal ¹⁰				0		?	3.5 Mt
Romania ¹	16 580	46 (161) ⁵	4 412 084 (5 376 187) ⁵	0		1 (2) ⁵	3 580 000 (1 750 000) ⁵
Slovakia ¹	201	26	1 151 000	0		0	0
Slovenia ¹	383 (452) ²	5 (3) ²	1 010 000	0		1	420 000 (0.6 or 0.7 Mt) ²
Spain ^{11,12}	4462			?			1.628 Mt
Sweden ^{13,14}	215			0		1	1 000 000

Source: 1. Most information for non-EU countries comes from Pers. Comm. Uranerzbergbau (1997), unless otherwise mentioned; 2. Logar (1996), Krizman (1996), Krizman and Logar (1996); 3. Vapirev et al. (1996); 4. Hébert et al. (1996); 5. Sandru (1996a,b,c); 6. Daroussin and Pfiffelmann (1996); Crochon and Daroussin (1996); 7. Markkanen and Annanmäki (1994); 8. BMWi (1993); 9. Ettenhuber (1996); 10. IAEA (1992); 11. Pérez Estévez and Sanchez Delgado (1996a,b); 12. Santiago (1994); 13. Ehdwall (1996), Sundblad (1996); 14. Sundblad and Stiglund (1994). * In Albania only exploratory mining

Table A1.5 Number of open-pit, underground and in-situ leaching (ISL) mines and mills and their status for Western and Eastern Europe.

Country*	Underground mines (No.)	Open pit mines (No.)	ISL (No#)	Mills (No.) Between () operational	Process
Albania	0	0	0	1	
Bulgaria	23	4	17	2	
Czech Republic	99	16	2	8(1) ¹ (2) ²	
Estonia	several small	0	0	1	
Finland	0	1	0	1	experimental
France	>200			9(2)	acid treatm./leaching
Germany -former East	4 (regions)	1	1	2 (0)	acid (Seelingstadt)
-former West	2	0	0	1	alkaline (Crossing)
Greece	2 sites				
Hungary	5(3)	0	<1	(1)	acid & alkaline leaching
Poland	5	0	0	0	
Portugal	Not clearly specified			1 (?)	
Romania	21	1 (?)	0	1 (1) (3) ²	
Slovakia	~ 0				
Slovenia	1	0	0	0, 1 ²	
Spain	Not clearly specified			4(3)	
Sweden	0	1	0	1	acid treatment

Source: *: Most information from Uranerzbergbau (1997) verified with references for the respective countries in Annex A2.1 to which we refer for additional information.

Table A1.6 Radiological characteristics of the mining residues for Western and Eastern Europe

Country*	Low grade ore stored		Fine residues		Coarse residues	
	U (t)	²²⁶ Ra (TBq)	U (t)	²²⁶ Ra (TBq)	U (t)	²²⁶ Ra (TBq)
Albania						
Bulgaria			50 TBq U and ~ 1600 TBq ²²⁶ Ra, 850 t ²³⁰ Th			
Czech Republic						
Estonia			1830 t U and 0.300 TBq ²²⁶ Ra			
Finland			300 TBq Ra			
France	632	78	3595	834	1530	63
Germany -former East -former West			WISMUT tailings: 15000 t U and 2000 TBq ²²⁶ Ra			
Greece						
Hungary	90 (?)		1330 t U and 286 TBq Ra			
Poland						
Portugal						
Romania						
Slovakia						
Slovenia	84 (?)		67 t U			
Spain			182.5	15.7	1489	67.2
Sweden			100 t U and 5 TBq Ra			

Sources: * For the references we refer to the references of the respective countries in Annex A2.1.

A1.3 Metal mining and melting

The main sources of information for this section are Harvey *et al.* (1994), Martin *et al.* (1996), Scholten (1996) and Penfold *et al.* (1997).

The following sections present production figures for each industrial category. When no other data are available present production information (if available) are provided in order to give an idea of the extent of the problem associated with each category.

A1.3.1 Category description

A1.3.1.1 Aluminium

[Main source: Penfold *et al.* (1997)].

Aluminium is the most abundant metal in the Earth's crust and the most important aluminium bearing mineral is bauxite [(Al,Fe)₂O₃], which can obtain up to 55% alumina. The commercial deposits of bauxite are gibbsite (Al₂O₃·3H₂O) and boehmite (Al₂O₃·H₂O), of which the former is preferred given its higher solubility. All bauxite is extracted by open pit mining. The mineral is found in Greece, France, Italy, Luxembourg and Hungary. Bauxite is first refined to produce alumina (digestion with caustic soda and resulting hydrate is crystallised in a kiln in the Bayer process) which is then reduced to metallic aluminium (e.g. by electrolysis). Aluminium production in the EU-12 amounted to 8.3 Mt in 1987 of which 1 Mt was mined and 1.3 Mt recovered. Important by-products of the aluminium production are vanadium and gallium.

A1.3.1.2 Copper

[Sources: NUKEM (1995); and Wichterley (1994)].

Metal processing begins with the separation of the ore from the dead rock. The dead rock is of two basic types: white (low activity) and black rock (higher activity) which are more bituminous and contains more uranium. At the Mansfeld area, ore was roasted resulting in crude copper, slag and raw gas. According to Penfold *et al.* (1997), the copper ore is ground, washed, melted and cast after which reduction takes place. Oxides and carbonates can be leached with sulphuric acid and the copper can be electrolysed from this solution.

Over 75% of the copper output is used in the electrical industry. In the EU-12, copper is mined in Portugal, Spain and France. Copper production was 3.4 Mt in the EU-12 of which 0.02 Mt was mined, 0.98 Mt recycled and the rest imported.

A1.3.1.3 Iron and steel

[Sources: Martin *et al.* (1996); Scholten (1996); and Penfold *et al.* (1997)].

In preparation for the blast furnace, the ore is pre-processed in sintering (at 1300-1400°C) and pelleting plants, resulting in dust emissions containing the volatile radionuclides ²¹⁰Pb and ²¹⁰Po. Also, coal is converted into coke with the ²¹⁰Pb (~100 Bq kg⁻¹) and ²¹⁰Po (~300 Bq kg⁻¹) finally residing in the tar produced. In a blast furnace iron is reduced and coke oxidised, resulting in the production of pig iron, slag and blast furnace ash. Mainly of sludge with the highest amount of activity comes from the blast furnaces. In sinter factories, only low radioactivity levels have been measured. Sludges and slag are normally dumped at the site. With the exception of bismuth [Harvey *et al.*, 1994] metal/alloy products have low activity concentrations.

Iron ore is mined in the EU-12 (France, Spain, Germany and the UK). 149 Mt of iron is produced in the EU-12 of which 5.8 Mt is mined and 52 Mt is recovered. The pig iron production in the EU-12, in 1993, was 77 Mt and the crude steel production 132 Mt.

A1.3.1.4 Lead

[Source: Penfold *et al.* (1997)].

Lead ores are found in many parts of the world. The richest ore is galena (lead sulphide), which is the main commercial lead source. Lead is separated from the ore by dry crushing, wet grinding (to produce a slurry), gravity classification and flotation. The liberated lead minerals are smelted by a three stage process and the final blast furnace liquid is then refined by the removal of copper, arsenic, antimony etc.

About 40% of lead is used as metal, 25% in alloys and 35% in chemical compounds. The lead production in the EU-12 is 1.6 Mt of which 0.18 Mt is mined and 0.54 Mt is recovered.

A1.3.1.5 Niobium

[Sources: Harvey *et al.* (1994); Martin *et al.* (1996); and Penfold *et al.* (1997)].

Niobium is found with other elements such as titanium, zirconium, tungsten, thorium and uranium in ores such as tantalite, columbite, pyrochlore. The largest deposits are in Australia and Nigeria. Treatment of the ore includes melting with KOH or NaOH, dissolving in HCl and processing with chlorine at 750-800°C. The powdered metallic niobium is then purified by reduction and the pure metal is obtained by heating at 2000°C. Two factories in the UK were known to make ferroniobium alloy by mixing and then furnacing a mixture of pyrochlore, aluminium powder, aluminium wire and iron chippings. Both factories are no longer in operation.

Niobium is used in the electrovacuum and fine-electricity industry. Ferroniobium is used in thermonuclear appliances. In 1987 the EU-12 niobium production was 3.8 kt.

A1.3.1.6 Tin

[Sources: Harvey *et al.* (1994); Martin *et al.* (1996); and Penfold *et al.* (1997)].

Feedstocks in the form of natural ores or tin rich residues from other processes are smelted to produce a molten metal stream. This is then separated into various compounds particularly tin, lead, bismuth and lead/bismuth alloy.

Tin is used in alloys and as protective coating for other metals. Tin consumption in the EU-12 was 54.5 kt in 1987 of which 4.2 kt is mined and 14.5 kt recovered, the remaining 35.8 kt is imported.

A1.3.1.7 Zinc

[Source: Penfold *et al.* (1997)].

Zinc is widely distributed and its sulphide form, sphalerite is the source of 90% of metallic zinc and contains iron and cadmium as impurities. The ore is crushed, ground and upgraded by flotation and after roasting the product is further refined by smelting, distillation or electronic refining.

Zinc is used for galvanising metals against corrosion, as dyecast, for the manufacturing of roofing, etc.

A1.3.2 General description of source term

Bauxite ores for aluminium production contain between about 41 and 500 Bq kg⁻¹ ²³²Th and similar levels of uranium.

Three types of residues from copper production are waste rock (black and white), dust/sludge and slag. The gas produced contains fine dust particles of high ²¹⁰Pb and ²¹⁰Po content, which is partly removed by air filters. The gas may then be sent to an intensive purification scrubber unit where the waste is left as sludge (contains also ²¹⁰Po and ²¹⁰Pb: is called Theisen-sludge). The smelting processes impact the environment through atmospheric pathways.

The levels of NORM in iron ore are not high. Problems with steel are limited to blast furnace plants. The amounts of contaminated dust is in the order of 2000 tonnes per million tonnes of steel, with ²¹⁰Pb activities of 30-100 kBq kg⁻¹.

Only the slag from the blast furnace in lead production contains substantial amounts of NORM (265 Bq kg⁻¹ ²²⁶Ra).

The activities in the pyrochlore for ferroniobium production were 10000 Bq kg⁻¹ for ²³⁸U and 80000 Bq kg⁻¹ for ²³²Th. The production of slag was of the same rate as the consumption of feed stock with a similar NORM content. The slag is used as landfill.

The ore for tin manufacturing typically contains 1000 Bq kg⁻¹ ²³⁸U and 300 Bq kg⁻¹ ²³²Th. The slag contains 4000 Bq kg⁻¹ of both ²³²Th and ²²⁸Ra and 1000 Bq kg⁻¹ of both ²³⁸U and ²²⁶Ra.

The slag and sludge of many mining and smelting of metal ores is often used as building material and for road construction (see Section A1.8).

Martin *et al.* (1996) reported on some typical activity concentrations in feed materials of metal smelters which are summarised in Table A1.7.

Table A1.7 Typical activity concentrations in feed materials of metal smelters

Feed Material	²³² Th (Bq kg ⁻¹)	²³⁸ U (Bq kg ⁻¹)
Iron ore	<5	<5
Coal/coke	20	20
Tin ore	300	1 000
Pyrochlore (niobium)	80 000	10 000
Copper ore (Mansfeld, G)		1 000
Ilmenite (titanium)	1 000	1 000
Rutile (titanium)	200	350
Bauxite (aluminium)	300	300

Source: Adapted from Martin *et al.* (1996).

Typical activities in wastes from these industries are given in Table A1.8.

Table A1.8 Typical activity concentrations for ores and waste products in the metal mining and smelting industry.

Material	Activity concentration Bq kg ⁻¹	Radionuclides
Aluminium [Source: Penfold <i>et al.</i> (1997)]		
Bauxite	37-530 41-527	²³⁸ U ²³² Th
Red sludge	250-496 252-537 122-310	²³⁸ U ²³² Th ²²⁶ Ra
Copper [Sources: NUKEM (1995); Wichterey (1994); Martin <i>et al.</i> (1996)]		
Slags (old: 200-500 yr.)	800-1370 500 660-1550 (670 -1 420) 50	²²⁶ Ra ⁴⁰ K ²¹⁰ Pb, ²¹⁰ Po ²³² Th
Slags (new: since 1880)	270 -1040 400 120-680 (100-670) 50	²²⁶ Ra ⁴⁰ K ²¹⁰ Pb, ²¹⁰ Po ²³² Th
Dead rock (white)	10 -140 40 -110	²²⁶ Ra ²¹⁰ Pb
Dead rock (black)	250-810 220-760	²²⁶ Ra ²¹⁰ Pb
Sludge	140-740 200 3620-21540 (7900 - 21900) 20	²²⁶ Ra ⁴⁰ K ²¹⁰ Pb, ²¹⁰ Po ²³² Th
Roast product	170-440 2330 -18190 (2750 -19630)	²²⁶ Ra ²¹⁰ Pb, ²¹⁰ Po
Iron/steel [Source: Martin <i>et al.</i> (1996)]		
Slags	150 150	²³⁸ U to ²¹⁰ Pb sub-chain ²³² Th decay chain
Sludge	30 000 -100 000	Mainly ²¹⁰ Pb
Coal tar	110 140	²¹⁰ Pb ²¹⁰ Po
Dust scales	<200 000	²¹⁰ Pb, ²¹⁰ Po
Fumes	10	²¹⁰ Po
Lead [Source: Penfold <i>et al.</i> (1997)]		
Mining waste	<19 48 <15	²³⁸ U ²²⁶ Ra ²¹⁰ Pb
Furnace slag	36 265	²³² Th ²²⁶ Ra
Niobium [Sources: Harvey <i>et al.</i> (1994); Martin <i>et al.</i> (1996)]		
Ore/Slag	10 000 80 000	²³⁸ U ²³² Th
Tin [Harvey <i>et al.</i> (1994) (a); Martin <i>et al.</i> (1996) (b)]		
Ore	1000 (a) 300 (a)	²³⁸ U ²³² Th
Slag	1000 (a) 4000 (a); 300 (230-340) (b) 1000 (a) 10 000 (b)	²³⁸ U ²³² Th ²²⁶ Ra Mainly ²¹⁰ Pb and ²¹⁰ Po
Fumes	200 000 (b)	Mainly ²¹⁰ Po
Tellurium dross	20 000 (b)	²¹⁰ Po
Zinc [Source: Penfold <i>et al.</i> (1997)]		
Mining wastes	<19 7 <15	²³⁸ U ²²⁶ Ra ²¹⁰ Pb
Electrolysis waste	<6 8 8 96	²³⁸ U Th-230 ²²⁶ Ra ²¹⁰ Pb
Slag	33 30 44	²³⁸ U ²²⁶ Ra ²¹⁰ Pb

It can be concluded, from Tables A1.7 and A1.8, that the concentrations of NORs in ores of tin, aluminium and titanium and black rock for copper are generally around the exemption limits (see Table 4.1). Pyrochlore has much higher activity concentrations due to its intrinsic geochemical properties which make it rich in lead, bismuth and other heavy metals. Slag from copper and steel furnacing and red sludge produced in aluminium production are around the exemption limits. Slag from tin and niobium production have much higher NOR-levels. As noted previously, these exemption limits should be considered with care since they do not apply for bulk amounts of materials but only for small release sources. For bulk amounts of materials it is necessary for the national authorities have to decide which exemption values apply.

A1.3.3 Radiological impact (releases and pathways)

The prime hazards to the public come from the wastes rather than from the feedstock and the metal products themselves. Good data are available on the radiological impacts to workers and the public from tin smelting and niobium steel [Harvey *et al.*, 1994]. However, information for iron and other metal industries is lacking.

The disposal of slags from tin processing into shallow land fill sites opens up routes for potential exposure via the migration of radionuclides and the accidental development of these sites. The dose received by a member of the public (redevelopment, 10^5 year post site closure), was estimated to be $10 \mu\text{Sv a}^{-1}$. The dominant radionuclide is ^{232}Th [Harvey *et al.*, 1994].

The landfill disposal of slag from niobium steel production can lead to exposure to the public due to natural radionuclide migration and as a result of development of the site. For an undisturbed site, doses received by the critical group, living 500 m from the site are reported as $\sim 6 \mu\text{Sv a}^{-1}$ which is mainly the result of inhalation of re-suspended dust residing on arable land. Redevelopment workers face hazards mostly related to ^{232}Th while residents on the site will have high levels of radon in their houses. All details of the assessment scenario can be traced back in Harvey *et al.* (1994).

In the Mansfeld area (copper mining and smelting) exposure is mainly due to external irradiation from residues (average per pile $0.3 \mu\text{Sv h}^{-1}$; range $0.06\text{--}4590 \mu\text{Sv h}^{-1}$; average per sludge pool: $>300 \mu\text{Sv h}^{-1}$), inhalation of contaminated dust and the ingestion of contaminated food. Soil contamination is mainly due to ^{210}Pb from the smelting plants. The most important exposure pathway is the airborne spread of radionuclides from sludge and dust but can be minimised by covering the sites with a layer of soil. The release of ^{222}Rn is not significant since the ^{226}Ra in dust and sludges is rather low ($\sim 1000 \text{Bq kg}^{-1}$).

A1.3.4 Occurrence in European countries

Only limited information is available in literature on past practices related to metal mining and smelting. Also, very limited information is available on the radiological characterisation of the waste residues.

One of the more (radio)contaminated sites in Europe, from the metal extraction and processing industry, is the Mansfeld area in former East Germany. Here, ores of non-ferrous metals, predominantly copper slate, have been mined and smelted since the 12th century. The last works closed in 1990. The relics of the mining and smelting (hundreds of waste rock and slag piles) are found throughout the area. In the Mansfeld deposit, copper ores are frequently accompanied by uranium minerals. The environmental contamination resulted from the dust produced during smelting, which was contaminated with ^{210}Pb and ^{210}Po , and distributed through the atmospheric pathway. The release of radon from the relics is not significant. However, a lot of slag and slag bricks were used, on a large scale, for road and building construction. The Federal Office is investigating the situation (see also Section A1.1.4). More information can be found in Annex A2.2 [Wichterey, 1994; NUKEM, 1995].

Table A1.9 lists the European sites for which information has been found. These are described in some more detail in Annex A2.2. Also, in Annex A2.2, information is provided on the present extraction and production levels for the different metal industries.

Table A1.9 List of European metal extraction and processing plants for which information was found

Country	Al	Cu	Iron & Steel	Pb	Hg	Nb	Sn	Zn
Albania	1 plant closed							
Bulgaria		4.7 × 10 ⁶ m ³ mining waste dumped on coast						
Finland				Mining & milling; closed; 0.76 Mt waste; 36 kt milled ore				Vihanti zinc mine closed
Germany		Mansfeld 1253 waste piles, 10 ⁸ m ³ , 2.9 × 10 ⁶ m ³ sludge						
Netherlands			1 oper. factory					
Slovenia	ilmenite & bauxite proc.				Idri mining district; mines & mills closed			
UK			1 oper. factory			2 closed factories	1 closed factory	

Sources: See Annex A2.2.

A1.4 Phosphate industry

[The main sources of information on this section are: Baetslé (1991, 1994); Cancio *et al.* (1994); Scholten (1996); Schmidt *et al.* (1995); Saler *et al.* (1996); Sandru (1996a,b); Logar (1996); Subasic *et al.* (1996a,b); Krizman and Logar (1996); and Martin *et al.* (1996)].

A1.4.1 Category description

The phosphate industry is very important for the European agricultural and chemical sectors. The world production is about 130 Mt a⁻¹, of which 50 Mt is exported. The ores are shipped to Europe and processed in various countries. Europe constitutes the major importer on the world market.

Of the ~30 Mt a⁻¹ imported, about 55-60% is consumed by the EU-12 countries, with the balance being made by the EFTA countries and the East European countries. The phosphate ores processed in the EU mainly originate from Africa (Morocco 43%, Togo 7%, others 20%), USA (19%) and Israel (12%) [Baetslé, 1991] and is mainly used for fertiliser production. Smaller amounts are used for the chemical industry (detergents etc.) and the food industry. The EU-12 has a fertiliser production capacity of 4.2 Mt P₂O₅ a⁻¹ [Baetslé, 1991].

The phosphate in minable quantities is concentrated by sedimentary, igneous, weathering and biological processes. Uranium may be incorporated in sedimentary phosphorite ores through ionic substitution into the carbonate-fluoroapatite crystals. Igneous phosphorite contains less uranium but more thorium. High phosphate contents usually correspond to high uranium contents (50-300 mg kg⁻¹). The mean uranium content in the ore from Moroccan origin is 125 mg kg⁻¹ (1500-1700 Bq kg⁻¹ ²³⁸U; 1500-1700 Bq kg⁻¹ ²²⁶Ra; 10-200 Bq kg⁻¹ ²³²Th) [Martin *et al.*, 1996].

The apatite ores are particularly insoluble and the primary process for the production of phosphoric acid is by leaching the phosphate from the rock with strong acids. In 90% of the

cases, ore is treated with sulphuric acid to produce phosphoric acid and gypsum. The gypsum precipitates and is filtered out, washed and if necessary neutralised for disposal. The obtained phosphoric acid is very impure (30% P_2O_5) and further processing is generally carried out to obtain concentrations of 50% and more. Uranium and thorium become enriched in the fertiliser to about 150% of their original concentrations and radium reduced to 10% of the original concentration. About 80% of the ^{226}Ra , 30% of the ^{232}Th and 14% of the ^{238}U is left in the phosphogypsum.

In terms of rounded figures, the production of 1 tonne of phosphate requires the extraction of 3 tonnes of ore. When processed, this results in the generation of 4 to 5 tonnes of phosphogypsum. A reference 1000 t ore d⁻¹ plant produces about 240000 tonnes of phosphogypsum per year with a mean ^{226}Ra content of 800-1250 Bq kg⁻¹. The local dump sites, containing TBq of radium, are often unprotected from rainfall and are hydraulically connected to surface waters and to the shallow aquifers. The ^{226}Ra present is fairly insoluble but, given the high concentration of calcium, it can be solubilised.

If hydrochloric acid (HCl) is available as by-product from other chemical processes it may be used for leaching. The chemical reaction is similar but the waste products are different. Also, the total inventory of thorium and uranium are dissolved and enter the liquid $CaCl_2$ (with excess calcium, CaF_2 is formed) waste streams, resulting in fertilisers and chemicals free from radioactivity. A recently developed leaching process uses nitric acid. Details of the radionuclide distribution between the products and by-products are uncertain.

Alternatively, the rock can be melted in a furnace (1400°C) with sand, iron oxide and coal for the direct production of elemental phosphorus. The dust generated during the sintering process, on which the radionuclides (mainly ^{210}Pb), heavy metals and volatile organics condense, is trapped in the electrostatic precipitators and is several times recycled. However, part of this precipitator dust has to be removed to control the contaminant levels in the precipitator cycle and amounts to less than 1% of the of the raw phosphate ore [Erkens, 1997]. The quantity of slag amounts to 85% of the raw phosphate ore and contains the major fraction (93%) of ^{238}U and ^{226}Ra [Baetslé, 1991 and 1994, Schmidt *et al.*, 1995]. It can be used for road construction. In general, the waste products are also used in by-products (road building, ballast in concrete, wall board).

A1.4.2 General description of source term

The only radioactive sources related to closed phosphate fertiliser plants are linked with the disposal sites for slag, from the thermal processing and the phosphogypsum piles from sulphuric acid processing. As already noted, about 80% of the ^{226}Ra , 30% of the ^{232}Th and 14% of the ^{238}U is left in the phosphogypsum and the production of 1 tonne of phosphate results in the generation of 4 to 5 tonnes of phosphogypsum. Schmidt *et al.* (1995) also reported that, on the average, radionuclide contents of phosphogypsum piles were: 1300 Bq kg⁻¹ ^{210}Po ; 900 Bq kg⁻¹ ^{210}Po ; 3000 Bq kg⁻¹ ^{226}Ra ; 10 Bq kg⁻¹ ^{232}Th ; and 500 Bq kg⁻¹ ^{238}U . An overview of typical activity concentration in ore and waste products of the phosphate industry is given in Table A1.10.

Table A1.10 Typical activity concentration in ore and waste products of the phosphate industry

Material	Activity concentration Bq kg ⁻¹	Radionuclides
Ore (Morocco)	1500-1700 (b) 10-200 (b) 1500-1700 (b)	²³⁸ U ²³² Th ²²⁶ Ra
Phosphogypsum	500 10 3000 (a); 700 (c) 1300 900	²³⁸ U ²³² Th ²²⁶ Ra ²¹⁰ Pb ²¹⁰ Po
Furnace slag	1000 (d) 1400-1500 (a); 1000 (c)	²³⁸ U+ ²²⁶ Ra
Calcined dust	1000 (d)	²¹⁰ Pb

Sources: (a) Schmidt *et al.* (1995); (b) Martin *et al.* (1996); (c) Scholten (1996); (d) Erkens (1997).

In the hydrochloric acid treatment, which leads to the formation of CaCl₂ as an effluent, all the ²²⁶Ra in the raw phosphate is released with the liquid effluent (20 Bq L⁻¹) and most of it will leave the dump site. The ²³⁸U released from the ore will be precipitated by lime addition and build up with CaF₂ on the dump site. Introducing a BaSO₄ precipitation step will effectively decontaminate the effluents but the accumulation of a ²²⁶Ra rich dump creates local environmental problems due to ²²²Rn emissions [Baetslé, 1991].

The quantity of dust produced with thermal processing is less than 1% of the raw phosphate ore and contains levels of ²¹⁰Pb of up to 1000 Bq kg⁻¹ [Erkens, 1997]. The ²¹⁰Pb can be immobilised with cement. The quantity of slag produced with thermal processing amounts to 85% of the raw phosphate ore and contains the 93% of the ²³⁸U and ²²⁶Ra. According to Baetslé (1991) the ²²⁶Ra concentration is 1400-1500 Bq kg⁻¹ slag. Erkens (1997) reports 1000 Bq kg⁻¹ ²³⁸U+ (daughters included). The amount of radium leached depends on the amount of water that passed through the pile, the solubility of radium sulphate and the leachability of the phosphogypsum crystals.

A1.4.3 Radiological impact (releases and pathways)

The slag from the thermal processing and the phosphogypsum piles from sulphuric acid processing causes elevated radon exposures to the public. Collective dose calculations for a population (150 inhabitants km⁻²) round a plant operated at 0.13 Mt a⁻¹, for a period of 20 years, resulted in a figure of 2.4 manSv within 400 km of the plant. Covering the deposits with a 0.3-1.5 m soil cover (Huelva, Spain [Cancio *et al.*, 1994]) resulted in a reduced radon release and, hence, in a dose reduction by a factor of 2-3. However, when examining the data presented, no effect could be deduced for radon. It can be concluded, according to Martin *et al.* (1996), that atmospheric discharges from (an operating plant including) solid waste piles do not seem to be of major radiological significance.

The release of radium ions into leachate, and surface run-off from phosphogypsum piles, is mainly determined by the solubility of the gypsum. For the above example, the collective dose received from drinking water, farm produce and freshwater fish, after contamination of surface or groundwater by water percolating through the pile, combined to give total of only 2 manSv per year. This indicates that, in general, these pathways are not of major concern. Schmidt *et al.* (1995) assessed that the major part of the dose for radium releases from a gypsum pile into ground water and surface water is caused by the ingestion of ²¹⁰Po. The effective dose equivalent for a member of the critical group varied from 0.02 and 1.1 mSv a⁻¹, depending on the pathway. In particular, the doses by freshwater fish are of regulatory concern.

Although much of the uranium and thorium remains in the fertiliser (330-2300 Bq kg⁻¹ ²³⁸U), fertiliser application does not contribute to the general dose (max. 2 µSv a⁻¹ for an individual). An aspect of importance is the use of phosphogypsum and calcium silicate slag in building materials, since this can lead to increased radiation exposure. This is discussed in Section A1.8. A potential area where unaccounted exposure may occur, is in the cleaning of the process equipment after the operational phase and the possible decommissioning of the buildings.

On the basis of the information accumulated over the last years on the radiological aspects of the phosphate industry, no insuperable problem of providing adequate protection for workers or public is apparent.

A1.4.4 Occurrence in European countries

Phosphoric acid production plants in the EU-12 are mainly located in France (30%, based on 1993 production figures), Belgium (14%) and Spain (16%).

No information was found in literature on closed phosphorus-producing factories. However, some data are available on the waste produced and its accumulation. Some of these phosphogypsum waste dumps are closed (Tessengerlo, Belgium; Huelva, Spain; Romania). Additional information on one phosphate mine situated in North Estonia and some more information on the other sites mentioned can be found in Annex A2.3.

In addition to providing the radioactivity concentration in phosphate rocks of different origin and some EU-12 phosphate ore importation data, Annex A2.3 also provides economic data on phosphate consumption in EU-12 countries. Given that the production of 5 tonnes of phosphogypsum waste per tonne of phosphate produced (or 1.5 tonne per tonne of rock) and, that in Europe 90% of the phosphate rock is treated by the sulphuric acid method, an estimate can be made of the annual amount of waste production (and, hence, the amount of waste to be dealt with in future). With the data on the activity concentration of phosphates of different origin and their relative importance, the contamination of the waste can be estimated.

In Table A1.11 a list of sites is given which are discussed in Annex A2.3. We also refer to this annex for the references.

Table A1.11 Phosphate industry liabilities for which information is available.

Country	Liabilities
Belgium	- Tessenderlo; still operational; closed CaF ₂ dump.
Croatia	- Ina-Petrokemija fertiliser plant; 3.5 × 10 ⁶ m ³ phosphogypsum - Waste in Port of Sibenik; ore import site
Estonia	- Open-pit phosphorus mine and fertiliser production in N. Estonia.
Romania	- 4 nitric acid and 4 sulphuric acid plants. Latter 4, high deposits (each 20-40 ha with 5 × 10 ⁶ t deposit)
Spain	- At Huelva: gypsum deposit on the banks of River Tinto: 425 ha, 4-6 m high

A1.5 Coal Mining and Power Production from Coal

A1.5.1 Category description

The specific activity in coal is, in general, not high. It is of the same order of magnitude as mean concentrations in the Earth's crust, but may vary widely between different mines.

The greatest consumption of coal is for electricity production, where coal is used for underfiring steam boilers [Scholten, 1996]. Combustion of coal results in 80-95% mass

reduction, thus yielding 5-20% ash which contains most of the original radioactive material. This is typically concentrated by an order of magnitude. Bottom ash (~25%) remains in the furnace, whilst 99% of fly-ash (~75%) is recovered on filters and gaseous substances are carried by the gas stream to the stack [Martin *et al.*, 1996]. Scholten, (1996) reports 10% and 90%, bottom and fly ash, respectively. In the EU-12 the total consumption of coal, brown coal and peat for electricity production was about 300 Mt in 1992. This results in ~30 Mt of ash [Scholten, 1996]. The ash can be disposed of in landfill or in sea or re-used (dikes of landfill, road construction, substitute for cement, filling material for concrete).

Gypsum may be formed by desulphurisation of the flue gas from coal fired power plants. The production is about 200000 tonne per GWyear.

A1.5.2 General description of source term

The average global specific activities in coal (Table A1.12) are 20 (5-350) Bq kg⁻¹ ²³⁸U and 22 (5-230) Bq kg⁻¹ ²³²Th [Scholten, 1996]. For the EU-12, average values of 30 (7-185) Bq kg⁻¹ ²³⁸U and 15 (3-22) Bq kg⁻¹ ²³²Th are reported [Scholten *et al.*, 1993; Martin *et al.*, 1996]. An extreme case is reported for Freital, in the former German Democratic Republic, where coal and uranium occur together. Here, ²³⁸U concentrations in the coal of up to 1000 mg kg⁻¹ (corresponding to 15000 Bq kg⁻¹) are found. Coal ash arises in Europe at a rate of ~30 Mt a⁻¹.

Generally, the enhancement in concentration of natural radionuclides of coal ash, remaining in the plant after combustion, is about a factor of 10. In most cases the weighted specific activity of the collected fly ash is below 500 Bq kg⁻¹, thus below the reporting levels.

Results from a German study demonstrated that a specific activity of gypsum, from desulphurisation, was about 20 Bq kg⁻¹ for both ²²⁶Ra and ²³²Th. Thus its disposal is no problem [ref. 24 in Scholten (1996)].

In some German and Polish coal mines, radium-rich sewage waters are produced. Precipitates in the environment and installations of up to 200 kBq kg⁻¹ are reported [refs. 25 and 26 in Scholten (1996)]. A solution is precipitation with phosphogypsum, by which both products are stored in underground galleries.

Table A1.12 Specific activities in coal and fly ash and some radionuclide enhancement factors for small particulates in fly ash

Radionuclide	Specific activities (Bq kg ⁻¹)			Enhancement factors ²
	Coal (global) ¹	Coal (Europe) ²	Fly ash ²	
²³⁸ U	20 (5-350)	30 (7-185)	200	1-10
²³² Th	22 (5-230)	15 (3-22)		1-10
²¹⁰ Pb		20	2400	20-120
²¹⁰ Po		20	2000	30-200
²²⁶ Ra				5-15
⁴⁰ K		130 (1-300)		10-50

Sources: 1. Scholten *et al.* (1993); 2. Martin *et al.* (1996).

A1.5.3 Radiological impact (releases and pathways)

The leaching from fly ash is low and so, for landfill and road construction, there are hardly any constrictions. The use of fly-ash for buildings leads to an additional radiological burden by direct radiation and by exhalation of radon [Roelofs and Scholten, 1994]. When the level of 10 μSv a⁻¹ from a single source is introduced, then the use of fly-ash for building material may be subjected to restriction.

However, dumping may increase the radiation level around the dumping site. A Dutch study [ref. 20 in Scholten (1996)] estimated the dose to workers on a dry ash disposal site (100×100×2.5 m³) to be 0.12 mSv a⁻¹ from inhalation and direct irradiation from the ash. The most significant exposure route identified is from the re-suspension of ash from disposal sites with dose rates from associated agricultural pathways as high as 250 μSv a⁻¹ [ref. 3.11 in Martin *et al.* (1996)]. The radionuclides, ²³⁰Th, ²³²Th, ²¹⁰Po and ²¹⁰Pb, are the most significant in the ingestion and inhalation exposure pathways [Martin *et al.*, 1996].

The operation of coal-fired plants over several decades is reported to result in an increase in natural background of less than 1% [ref. 3.7 and 3.11 in Martin *et al.* (1996)].

A1.5.4 Occurrence in European countries

Information obtained is summarised in Table A1.13. However, no overall information was obtained on the distribution of the coal industry in Europe. In the UK and Belgium, for example, many coal mines were operative and some still are, however, since there is no central bureau for information on these industries, it is very cumbersome to find this information.

Table A1.13 List of European coal mines and coal power plants for which information is found

Country	Liabilities	Radiological characteristics (Bq kg ⁻¹)
Croatia ¹	Several coal mines, only one operational 2 coal fired plants discussed, one closed dump	dump: 10000 m ³ : 18640 ²³⁸ U and 6200 ²²⁶ Ra
Germany ²	Freital area: all mines closed	55 piles, 15 million m ³ coal: 1.24 g ²³⁸ U t ⁻¹ , 12.3 g ²³² Th t ⁻¹ , 987 g Ra (t U) ⁻¹
Greece ³	Several lignite/coal mines and power plants	lignite: 248 ²³⁸ U, 133 ²²⁶ Ra, 134 ²¹⁰ Pb, 18 ²²⁸ Ra fly ash: 517 ²³⁸ U, 366 ²²⁶ Ra, 275 ²¹⁰ Pb, 50 ²²⁸ Ra
Poland ⁴	still 66 active coal mines	brines: 400 kBq m ⁻³
Romania ⁵	3 old & 7 modern plants; all operational	80 ²³⁸ U, 126 ²²⁶ Ra, 210 ²¹⁰ Pb, 62 ²³² Th
Slovenia ⁶	Old coal mining district	Coal and coal ash: 400-2000 ²²⁶ Ra Outdoor Rn: 80 Bq m ⁻³ ; Indoor: several thousands

Sources: 1. Saler *et al.* (1996), Subasic *et al.* (1996a,b); 2. NUKEM (1995), Henze and Weiss (1995); 3. Manolopoulou and Papastefanou (1992); 4. Piestrynski *et al.* (1996); 5. Sandru (1996b); 6. Krizman (1996b).

A1.6 Oil and gas drilling

[The main sources on this subject are: Van Weers *et al.* (1996), Scholten (1996), Martin *et al.* (1996), Penfold *et al.* (1997), IAEA (1996a,b,c) and Jonkers and Knaepen. (1997)].

A1.6.1 Category description

Naturally occurring radionuclides are present in varying concentrations throughout hydrocarbon reservoirs. The type of rock formation largely determines the radionuclide content of the reservoir; sedimentary rocks having low radionuclide concentrations whereas phosphate rock types have high levels. The most important oil/gas source rocks are carbonates, limestones, shales and coal. After expulsion from the source rock, oil and gas migrate either to a reservoir formation (reservoir rock) or to the Earth's surface. The waters associated with the rock formations often contain barium, calcium, strontium and radium, which have higher solubilities than uranium and thorium. The activity in the connate water (afterwards production water) is mainly due to radium (0.002-1200 Bq L⁻¹ ²²⁶Ra and 0.3-180 Bq L⁻¹ ²²⁸Ra) with only minute quantities of thorium and uranium present [Jonkers *et al.*, 1997]. Sometimes connate waters have high concentrations of lead (>100 mg L⁻¹) and zinc (>300 mg L⁻¹).

In the process of oil and gas extraction, well fluids are transported, via tubings, to the well head. The flow from different well heads are commingled in the production manifold, after which it is led to separators for de-gasification and de-hydration. The same route is followed

for gas production until the production manifold. After separation from the associated fluids, the gas is heated and sent through absorbers to remove water and other products. Water (usually sea water or brine) is pumped into the well to maintain a high pressure to force the material to the surface. When this water is mixed with the geological formation water, there is often the formation of scale-forming solids due to the different water chemistries. Hard scales may be deposited in tubulars close to the perforation, at well heads and at the production manifold. Soft and medium hard scales are formed further downstream in a production installation (production piping, vessels, production water disposal/injection wells,...). Sludges, which maintain the majority of the radioactivity are deposited mainly in voluminous equipment (storage tanks, separator and dehydration vessels) and collected near the well where the initial purification is performed.

The occurrence of low specific activity scales and sludges is also associated with contamination of the inside of the equipment.

In gas production, ^{222}Rn ($10\text{-}50000\text{ Bq m}^{-3}$; weighed mean 337 Bq m^{-3}), and water with dissolved salts and solid material is transported with the gas. The water and solids are separated at the well head with an activity concentration generally between 100 and 200 kBq kg^{-1} .

The annual sludge production in the EU-12 from the gas and oil industry is presently $10000\text{ m}^3\text{ a}^{-1}$. This is reduced by a factor of 5 before disposal as waste. At UK platforms, scales are dumped in the sea. In Norway, scales with $>70000\text{ Bq kg}^{-1}$ are regarded as radioactive and are re-injected in the reservoir, or disposed off on-shore. In the Netherlands scales are currently stored on-shore.

Abandonment of several land-based production facilities in the EU has already taken place and also a few off-shore platforms on the continental shelf of the EU-countries have been removed [Van Weers *et al.*, 1996]. However, no information was found about closed oil and gas drilling sites. Van Weers *et al.* (1996) give an overview of current practice for dealing with NORM from oil and gas production in EU member states (mainly for UK, Netherlands, Norway).

The total production of natural gas in the EU amounts to $0.2 \times 10^{12}\text{ m}^3\text{ a}^{-1}$ of which about 43% is produced in the Netherlands and 35% in the UK. The total production of crude oil in the EU is 140 Mt a^{-1} of which about 80% is produced in the UK [Scholten, 1996].

A1.6.2 General description of source term

From the extensive literature review by Jonkers and Knaepen (1997) it is clear that both ^{238}U and ^{232}Th are only present in minute concentrations; mainly in production water and associated scale /sludge. The encounter of ^{238}U and ^{232}Th is strongly associated with suspended sand and clay particles. Hence, their concentration will be close to their concentration in reservoir rock ($<1\text{-}70\text{ Bq kg}^{-1}\text{ }^{238}\text{U}$ and $6\text{-}70\text{ Bq kg}^{-1}\text{ }^{232}\text{Th}$).

The dominating radionuclides in scales and other precipitates are ^{226}Ra and ^{228}Ra . Typically, the activity concentration in the scale is 100 times that of the reservoir rocks. Activity concentrations in the scale are (in Bq kg^{-1}): $2 \times 10^5\text{ Bq kg}^{-1}\text{ }^{226}\text{Ra}$; $5 \times 10^4\text{ Bq kg}^{-1}\text{ }^{210}\text{Pb}$; $5 \times 10^4\text{ Bq kg}^{-1}\text{ }^{210}\text{Po}$; and $10^5\text{ Bq kg}^{-1}\text{ }^{228}\text{Ra}$ [Scholten, 1996]. Hence, these are well above the reporting levels by factors of 20, 5, 5 and 10, respectively. An extensive literature review on activity levels in different (by)-products and wastes, from the oil and gas extraction and production industry, has been made by Jonkers *et al.* (1997). Some data on activity levels in waste products are presented in Table A1.14.

Table A1.14 Activity levels ($Bq\ kg^{-1}$) in waste products from oil and gas industry

Radionuclide	Physical form			
	Hard scales	Soft/medium scales	Sludges	Scrapings
^{238}U	1-500	1 - 50	5 - 10	
^{226}Ra	$100 - 15 \times 10^7 (2 \times 10^5)^*$	$80 - 4 \times 10^5$	$5 - 8 \times 10^5$	$10 - 7.5 \times 10^4$
^{210}Pb	$20 - 7.5 \times 10^4 (5 \times 10^4)^*$	$50 - 2 \times 10^6$	$10 - 1.3 \times 10^6$	$50 - 5 \times 10^4$
^{210}Po	$20 - 7.5 \times 10^4 (5 \times 10^4)^*$		$4 - 1.6 \times 10^4$	100 - 4000
^{232}Th	1 - 2	1 - 70	2 - 10	
^{228}Th	$50 - 2.8 \times 10^6 (10^5)^*$	$50 - 3 \times 10^5$	$500 - 5 \times 10^4$	$10 - 10^4$

Source: Adapted from Jonkers and KNaepen (1997). *: Averages according to Scholten (1996).

The decommissioning of oil and gas plants has not been investigated with respect to volume of contaminated material involved (contaminated steel pipe lines). In Croatia [Subasic *et al.*, 1996] $0.1\ Gy\ h^{-1}$ (with UNSCEAR conversion: $0.07\ Sv\ h^{-1}$) was recorded at the surface of pipelines. Jonkers *et al.* (1997) present some observed external radiation levels for the outside of processing facilities. The highest value recorded was $300\ \mu Sv\ h^{-1}$ at the surface of down hole tubings.

Additional NORM containing waste is generated at on-shore decontamination facilities, for components (pumps, valves, filters, tubulars and transport lines) [Van Weers *et al.*, 1996].

A1.6.3 Radiological impact (releases and pathways)

The main exposures to workers (within a factory, although similar values may be expected during remediation) are through external gamma irradiation from active deposits on equipment ($2\ mSv\ a^{-1}$: 200 h working in vicinity of scales will be $10\ \mu Sv\ h^{-1}$), ingestion and inhalation of airborne particles ($1\ mSv\ a^{-1}$). Wilson and Scott (1992) record radiation measurements on production piping of $0.01-10\ \mu Sv\ h^{-1}$. Problems are also encountered with land contamination resulting from the cleaning of scaled equipment. In localised areas of cleaning, ^{226}Ra levels as high as $3.2 \times 10^5\ Bq\ kg^{-1}$ were recorded. This compares to a local background of 37-260 $Bq\ kg^{-1}$. Fortunately, the radium scales are highly insoluble (max. 0.01% even in acidic environments) and once deposited the migration of the radium is thus virtually nil [Wilson and Scott, 1992].

There is lack of data concerning public exposure from scales and sludges. The main pathway would appear to arise from waste disposal and storage processes.

A1.6.4 Occurrence in European countries

The sites in Europe for which some radiological information was found are presented in Table A1.15. Further information is also given in Annex A2.5. Also some economic data for the oil and gas industry are presented in Annex A2.5. Apart from the general economic data presented in Annex A2.5.6, no information was found on the off-shore oil and gas industry in the UK.

Table A1.15 European gas and oil drilling sites for which information was available

Country	Liability	Waste characteristics (Bq kg ⁻¹)
Croatia ¹	3 gas and 5 oil fields	High active scales in pipelines
Germany ²	Operational Oil extraction	Hard scales: 59000 ²²⁶ Ra; 240000 ²²⁸ Ra
	Gas extraction	Soft scales: ²²⁶ Ra & ²²⁸ Ra: 100× less, ²²⁷ Ac: 2500 Hard scales: 850-10 ⁶ ²²⁶ Ra, 5900-1.2 ×10 ⁶ ²²⁸ Ra
Italy ^{2,3}	Operational plants Oil extraction	1126-2890 ²²⁶ Ra, <0.9 ²³⁸ U, <0.8 ²³² Th
	Gas collection	<2.7-30 ²²⁶ Ra, 24-54 ²³⁸ U, <0.8-19 ²³² Th
	Oil collection	110 ²²⁶ Ra, 11 ²³⁸ U, <0.8 ²³² Th
Netherlands ⁴	Gas extraction, abandoned in 1983	equipment: up to 3.3 ×10 ⁶ ²²⁶ Ra and 1.8 ×10 ⁶ Bq ²¹⁰ Pb
Norway ⁵	Only operational platforms discussed	Hard deposits: 4000-39000 ²²⁶ Ra Porous deposits: 300-24400 ²²⁶ Ra , 11300 ²²⁸ Ra Sludges: 100-4700 ²²⁶ Ra Sand: <limit ~21900 ²²⁶ Ra

Sources: 1. Saler *et al.* (1996), Subasic *et al.* (1996a,b); 2. Penfold *et al.* (1997); 3. Testa *et al.* (1994); 4. Scholten (1996), Eylander *et al.* (1997); 5. Lysebo and Strand (1997).

A1.7 Rare earth and titanium dioxide industries

[Most information was extracted from: Harvey *et al.*, 1994; Martin *et al.*, 1996; Scholten, 1996; Penfold *et al.*, 1997].

A1.7.1 Category description

The rare earths comprising the lanthanide series of minerals, occur in a variety of minerals, of which the most important are monazite [(Ce,La,Nd,Th)(PO₄)] and bastnasite [(Ce,La)(CO₃)F]. An overview of minerals, containing rare earths, is given by Martin *et al.* (1996). The monazite ores contain a low percentage (0.1-5%) of rare earth phosphates and a high percentage of other minerals (rutile, TiO₂; ilmenite, FeO·TiO₂ and zircon, ZrSiO₄), which are feed materials for the titanium industry.

Rare earths are used in polishes, in catalysts, as burnable poisons in nuclear reactors, in X-ray and CRT phosphors and as additives in steels, special glasses and incandescent glasses (see also Section A1.9).

Titanium is used as a pure metal, in alloys and in the form of various compounds. The bulk of titanium is used in iron and steel industry, for aircraft and rocket construction, in shipbuilding and for the construction of chemical plants [Penfold *et al.*, 1997]. Titanium oxide is used in the rubber, ceramics, paint and varnish industries.

Monazite ore concentrate is obtained from suitable sands by a gravimetric and electromagnetic sorting. Strong acids and alkaline solutions are then used to precipitate the lanthanides. The bastnasite ore concentrate obtained after washing and separation in water is washed with hydrochloric acid and calcined in order to produce a crude oxide containing 90% of lanthanum [Penfold *et al.*, 1997].

Martin *et al.* (1996) reports that, after pre-treatment, ores are heated to high temperatures followed by further conditioning to use rare earths for glass polishing. For the production of special glasses, the rare earths are mixed with the silica and then melted.

World production of rare earths is 30000 tonnes per year. None are mined in the EU. No figures were available regarding rare earth processing in Europe.

Industrial titanium production comprises the crushing and mixing with coal of titanium oxide, rutile and sometimes also ilmenite concentrate. Titanium tetrachloride is obtained by

chlorination in shaft furnaces to form a gas which is condensed and purified. The $TiCl_4$ is reduced to titanium sponge.

The total quantity of titanium dioxide use in Europe was 790000 t, of which only 2000 t was recovered from the ore in Europe.

A1.7.2 General description of source term

A1.7.2.1 Rare Earths

Typical concentrations of radionuclides in rare earth ores and (waste) products are depicted in Table A1.16.

Table A1.16 Typical concentrations of radionuclides in rare earth ores and (waste) products

Material	Activity concentration (Bq kg ⁻¹)						
	²³² Th	²²⁸ Ra	²²⁸ Th	²³⁸ U	²¹⁰ Pb	²²⁶ Ra	²¹⁰ Po
Rare earth	6000-10 ⁴	6000-10 ⁴	6000-10 ⁴	1000	1000	1000	1000
Monazite concentrate*	8000-3 × 10 ⁶	8000-3 × 10 ⁶	6000-4 × 10 ⁴	6000-4 × 10 ⁴	6000-4 × 10 ⁴	6000-4 × 10 ⁴	6000-4 × 10 ⁴
Glass	3000	3000	5000	1000		1000	1000

Sources: All data from Martin *et al.* (1996), except for those indicated with *, which are from Penfold *et al.* (1997).

It can be inferred from Table A1.16 that the radionuclide concentration in the feeding material are close to or higher than the exemption level (Table 4.1: consider remarks linked with table). The activity concentration is about a magnitude higher than in phosphate ores. However, with the annual production less than 1% of that of phosphate ore, the overall radiological impact of rare earths is substantially lower than for phosphate use.

During the extraction process, isotopes of radium are co-precipitated with barium sulphate to form a radium-bearing by-product (activity concentrations 3 × 10⁶ Bq kg⁻¹ ²²⁸Ra and 4.5 × 10⁵ Bq kg⁻¹ ²²⁶Ra). A second by-product, principally made up of calcium phosphate and ore residuals, has a typical activity concentration of 2 × 10⁴ and 3.5 × 10⁶ Bq kg⁻¹ of ²²⁸Ra and ²²⁶Ra, respectively [Penfold *et al.*, 1997].

The process of polish production generates minimal amounts of wastes. The end products (often containing a quarter of the initial activity concentration due to dilution of the feed stock) are non-radioactive and disposal in land-fill.

Faussat and Fernigie (1994) reports "low" specific activity levels of 10⁴ Bq kg⁻¹ for processed waste from rare earth production. However, given the large volumes of wastes involved, the overall activity is not negligible.

According to Martin *et al.* (1996) the radionuclide concentration in the products is diluted by a factor of 100 compared to the feed material. However, the data presented by the same authors, and reproduced in Table A1.16, indicates almost no dilution.

The refractory bricks (containing zirconium) in the furnace lining (see below) entail a solid waste stream.

A1.7.2.2 Titanium dioxide industry

The radiological hazards from TiO_2 production vary with the type and source of ore. The ore activity concentration of both ²³⁸U and ²³²Th decay chains may vary between 35 and 6000 Bq kg⁻¹. Specific activities in feeding materials in the titanium industry are presented in Table A1.17.

Table A1.17 Typical concentrations of radionuclides in titanium ores and products (Yugoslavia)

Material	Activity concentration (Bq kg ⁻¹)				
	²³² Th	²²⁸ Ra	²²⁸ Th	²³⁸ U	²²⁶ Ra
Ilmenite	600-6000 ¹ 35-90 ² 180 ³	180 ³	180 ³	<100-400 ¹ 31-71 ²	150 ³
Rutile	<600-4000 ¹ 64-90 ²			<100-250 ¹ 530-640 ²	
Titanium oxide	31 ³		9 ³		32 ³
Rubber coating		345 ³	200 ³		160 ³
Filters (Bq m ⁻²)		180-300 ³	64-88 ³		94-170 ³

Sources: Adapted from Penfold *et al.* (1997) except: 1. UNSCEAR (1993); 2. Johnston (1991); 3. Pucelj and Martincinic (1984) in Penfold *et al.* (1997).

In the presence of high sulphate concentrations, in both the plant feed liquor and the effluent, it was established that thorium remained in the solution but radium, which forms an insoluble sulphate compound, was found in the solid residues [UNSCEAR, 1988]. Specific activities of such radium precipitates in pigments or scale, formed in the heat exchanger, have been reported to be about 4×10^5 Bq kg⁻¹ ²²⁶Ra. The ²²⁸Th concentration in the heat exchanger scale was 1.5×10^6 Bq kg⁻¹. Table A1.17 shows the specific activity of several samples in the TiO₂ process from a Yugoslavian production plant. It is evident from this table that the chemical process concentrates radium from uranium decay.

A1.7.3 Radiological impact (releases and pathways)

A1.7.3.1 Rare Earths

No information was found on abandoned rare earth industries. Indications about the radiological problems to be encountered when an industry closes are the buildings (no specific information found) and the land fills. It may be inferred from Harvey *et al.* (1994) that the radionuclide concentration in the waste is comparable to the radionuclide concentration in the feed material. The radionuclides of major concern are ²³⁸U and ²³²Th. Individual doses arising from the migration of radionuclides from a landfill [Harvey *et al.*, 1994] are ~ 0.1 μ Sv a⁻¹ (radionuclide of largest impact being ²³⁸U). The collective effective dose to the public in the EU, from the solid waste stream, during rare earth furnacing, rises to 720 man-Sv (10⁶ years from a 50 year release) [Harvey *et al.*, 1994].

Overall, the radiological risk from the use of rare earth compounds is small.

A1.7.3.2 Titanium oxide industry

Dose rates measured at the surface of some vessels, pipes and wet filters in a Yugoslavian production unit varied between background and 50 μ Sv h⁻¹. Most of the radon measurements in the vicinity of the production facility were below 20 and 50 Bq m⁻³, except for those close to the mill, where concentrations of up to 800 Bq m⁻³ were measured.

A1.7.4 Occurrence in European countries

Rare earths are not mined in Europe. An example of rare earth processing is at Sillamäe, Estonia, where a plant, formerly used for the processing of uranium ore, switched, at the beginning of 1970, to the processing of loparite (from the Kola peninsula). This is a mineral rich in niobium, tantalum and other rare earths [Putnik *et al.*, 1994]. Although many more facilities must exist, only one operative facility in the UK, producing polish, and one,

producing special glass, were identified [Martin *et al.*, 1996]. Further, Andra (1998) reports monazite was treated by several French industries. One of those industries, ORFLAM PLAST, produced refractory bricks.

Titanium ores are only mined to a small extent in Europe; only 2000 tonnes compared to the 771000 tonnes annually imported. Of the total titanium usage of 790000 tonnes material, 558000 tonnes is consumed and the rest exported [Eurostat, 1995].

A1.8 Zirconium and ceramics industry

[Main sources: Scholten (1996); Harvey *et al.* (1994); Penfold *et al.* (1997); and Martin *et al.* (1996)].

A1.8.1 Category description

Zirconium is an abundant element, whose principal sources are zircon ($ZrSiO_4$), in silicate sands, and zirconia (ZrO_2) in baddeleyite ore. Zircon and zirconia are used in the manufacturing of refractory materials, in glazes, glasses and ceramics and in the manufacturing of dielectric materials and as additive in special metal alloys. World production of zircon is 700000 t a^{-1} . It is not mined in Europe, although Penfold *et al.* (1997) reports the existence of commercially useful deposits in Italy. Europe imports 350000 tonnes of zircon annually. 136 t a^{-1} zirconium is used in Europe.

Typically the sand is pre-processed, in very large quantities, by gravimetric and electromagnetic sorting. This separates the mineral salts which include monazite and bastnasite. In the zirconium metal extraction process, the zircon ore is then dressed with coke in a mill and processed in a chlorinating chamber. A by-product of this chlorinating process is crude chlorinate residue. The zirconium hydroxide, resulting from the subsequent zirconium-hafnium separation step (producing sludge), is fired and the resulting zirconium oxide is sent to a purifying chlorinating chamber to produce pure zirconium chloride. This is reduced by a magnesium reduction process to give metallic zirconium (used in nuclear power generation).

Refractory components are manufactured by mixing zircon sand with alumina and sodium carbonate and melting at high temperature. Zircon is used only in brick manufacturing whereas zirconia is used for bricks and nozzles.

The basic material for ceramics is clay. After shaping the moist emulsion, a glaze (containing zirconia) is applied and the product fired. The zirconia is fixed in the glaze, preventing dust generation.

A1.8.2 General description of source term

Activity concentrations in ores and products of the zirconium industry are presented in Table A1.18. According to reference 162 in Penfold *et al.* (1997), the main radiological concern, associated with the zirconium extraction process, is the fact that chlorinator residues contain significant amounts of soluble radium. The largest volume (quantities not specified) of waste generated is the sludge from the zirconium-hafnium separation process.

Table A1.18 Activity concentrations in ores and products of the zirconium industry

Material	Activity concentration (Bq kg ⁻¹)						
	²³² Th	²²⁸ Ra	²²⁸ Th	²³⁸ U	²¹⁰ Pb	²²⁶ Ra	²¹⁰ Po
Zircon	600 (400-40000)*	600	600	3000 (200-74000)*	3000	-	3000
Zircon casting	600	600	600	3000	3000	-	3000
Baddeleyite ore	300	600	2000	7000	7000	-	7000
Zirconia	300	600	2000	7000	-	-	-
Glazes	400	9000	5000	1000	-	20000	20000
Refractories	1000	2000	2000	10000	-	10000	20000
Glass	3000	3000	5000	1000	-	1000	1000

Sources: All data from Martin *et al.* (1996), except those indicate with * are from Penfold *et al.* (1997).

During the production of refractory bricks about 20 tonnes of dust is collected by the filters per 2000 tonnes of refractory materials, which initially contains 2×10^5 Bq kg⁻¹ ²¹⁰Pb and 6×10^5 Bq kg⁻¹ ²¹⁰Po. It can be inferred from Table A1.18, that refractory bricks can contain up to 1×10^4 Bq kg⁻¹ ²³⁸U.

Ceramics typically consist of 5% zircon. This results in a final product with activity concentrations of 200-300 Bq kg⁻¹ for both thorium and radium. Glazes can contain up to 6000 Bq kg⁻¹ thorium and radium (Table A1.18).

Table A1.19 presents data on the radium concentrations in some zirconium extraction materials and by- and waste-products for an English facility. According to Boothe (1980), the main radiological concern associated with the zirconium extraction process is the fact that chlorinator residues contain significant concentrations of soluble radium.

Table A1.19 Radium concentrations in zirconium extraction materials (Boothe, 1980)

Radium concentration	²²⁶ Ra (Bq kg ⁻¹)	Use or production rate (t a ⁻¹)
Zircon sand	3×10^6 - 3.7×10^6	16500
Crude chlorinator residue	-	500
Pure chlorinator residue	18.6×10^6 - 22.3×10^6	100
Chlorinator residue pile	2.8×10^6 - 4.8×10^6	-
Sludge (dry)	2.6×10^5 - 7.4×10^6	3500

A1.8.3 Radiological impact (releases and pathways)

As was the case for the rare earth industry, major exposure occurs during the operational phase (exposure to worker, which is outside the scope of this document). However, the overall the risk from the use of zirconium compounds is rather small.

The zircon products (glazes and refractories) and wastes have concentrations above the exemption limits (Table 4.1 and remarks). Fine ceramics generally do not breach reporting levels. However, some of the wastes may require attention.

The disposal of waste can occasionally result in increased radiological risk to the general public if it is not well planned. For example, when the refractory bricks (may contain up to 10000 Bq kg⁻¹ ²³⁸U), used in glass furnaces, are often disposed off (or replaced for the facility in operation) in large quantities in a single landfill. Individual doses of up to 100 μSv a⁻¹ have been estimated in redevelopment scenarios.

Harvey *et al.* (1994) calculated the collective doses to the public in the EU-12 (10⁶ years from a 50 year release: a remark is in place that 10⁶ years is a fairly long time!) from the waste streams after landfilling. The figures are presented below in Table A1.20 on an annual basis and clearly indicate that the risk to the general public is below normal levels of concern.

Table A1.20 Collective doses to the public in the from some waste streams following landfilling

Origin of waste	Collective effective dose (Sv a ⁻¹)
Conversion of baddleyite	1×10^{-1}
Refractory brick manufacture	1.1×10^{-2}
Refractory nozzle manufacture	2.9×10^{-3}
Glass furnace linings	7.2×10^{-5}

In an Australian study [Johnston, 1991] it was concluded that long-lived radioactive dusts constitute the greatest source of potential radiation hazard in a sand processing plant (in operation!). The main dose is attributable to the thorium in the dust. This heavy dependence of dose on thorium content arises because of the high standard metabolic uptake data used in the ICRP dosimetry models. Even for the lowest solubility class, the intake will depend more on the sand matrix metabolism than on the radionuclide compounds.

Landa and Councell (1992) studied the uranium leaching from glass and ceramic foodware and decorative items and showed that most items did not release uranium even not when treated with $1 \text{ mol L}^{-1} \text{ HNO}_3$. However, for "Fiesta" ceramic ware and Mexican ceramics with an orange-red uranium bearing glaze, significant leaching was observed, particularly with acidic solutions.

A1.8.4 Occurrence in European countries

According to Scholten (1996) about 350000 t zircon is yearly imported in the EU-12. Not much information is found on zirconium/zircon milling and processing facilities in Europe. Scholten (1996) briefly discusses a dry milling facility in the Netherlands and the Cookson Matthey group with dry and wet mills in England (HQ), Germany, Italy and Spain. In their discussion on radiological risks of non-nuclear industries, Pruppers *et al.* (1997) reports two mineral sands and ~75 ceramic factories in the Netherlands alone. Between 1932 and 1987, ORFLAM PLAST, in France, extracted cerium from monazite for the production of refractory bricks and this has resulted in several contaminated spots and 16 barrels containing ²³²Th [Andra, 1998]. Rhône-Poulenc, also situated in France, separates and produces rare earths from monazite for the production of television screens, micro-computers, catalysators of cars. The resulting historic radioactive contamination consists of 8 kt containing 1.15% ²³²Th (48 Bq g⁻¹) and 0.05% ²³⁸U (6 Bq g⁻¹). After 1994, the monazite was deactivated before import to France.

A1.9 Disposal of building materials

A1.9.1 Category description

Industrial by-products are widely used as raw materials in the building material industry because of the economic benefits. Concern arose when it was found that some of these waste materials can contain elevated levels of natural radioactivity. Fly ash, from coal-burning is used in concrete production. Bottom ash, from coal-burning, and slag, from electrical furnaces, is used for road construction. In addition, 15% of phosphogypsum is now being recycled.

Within the scope of the present report, the issue on the re-use of industrial waste materials mainly relates to the problem (dose to public) of disposing of old building materials in landfill after the abandonment of buildings and facilities or constructions.

Bricks are autoclaved from a mixture of sand and lime at moderate temperatures. The activity in the raw material is low. Fly ash can also be added to the mix, significantly influencing the

radionuclide concentrations. *Light-weight building blocks* are made by sintering a mixture of iron slag and fly-ash. The basic material for *cement* is marl and/or blast-furnace slag. After mixing in additives, grinding and heating, the clinker obtained is then mixed with gypsum and fly-ash as ballast. *Phosphogypsum* products are used in road building and as ballast in concrete and wall-board. *Slags* from the metal industry are crushed and used for road construction and foundation, as a concrete additive (up to 20%), a cement additive (25% max. content) and brick production additive.

A1.9.2 General description of source term

In Table A1.21 presents typical activity concentrations for feed-stock, by-products and for building materials. The concentration of ^{226}Ra is considered to be a more useful measure of activity than ^{238}U , because ^{226}Ra has a half-life comparable to the life of buildings and is a major gamma emitter. Its daughter is ^{222}Rn , which also has important implications for building materials. Most data are reproduced from Martin *et al.* (1996).

Table A1.21 Typical activity concentrations from feed-stocks, by-products and building materials

Material	^{226}Ra Bq kg ⁻¹	^{232}Th Bq kg ⁻¹
<i>Feedstock</i>		
Natural gypsum*	20 (10, 10)*	10 (10, 10)
Granite	100	80
Natural building stones*	(60, 30)*	(60, 20)*
Clay	40	20
Sand and gravel	15	20
<i>By-products/wastes</i>		
Fly-ash	100 (180, 140)	60 (20, 20)*
Phosphogypsum*	500 (390, 340)*	20 (20, 20)*
Blast-furnace slag	35 (270, 70)*	120 (70, 20)*
Slags (from Cu, Ni, steel)	up to 1500	up to 100
<i>Building material</i>		
Ordinary cement	40	30
Cement with 20% fly-ash	55	40
Ordinary concrete	40	40
Aerated and light-weight concrete*	(60, 60)*	(40, 30)*
Concrete with 20% Cu-slag	140	35
Wallboard with natural gypsum	20	10
Wallboard with phosphogypsum	500	20
Tiles (floor)	60	55
Brick	50	60
Clay (red) bricks*	(50, 50)*	(50, 40)*
Sand (white) bricks*	(10)	(10)

Sources: All data from Martin *et al.* (1996) except for * from Mustonen *et al.* (1997). *: Mean concentrations as average of national means weighed by the population of different states. The first figure between brackets is for EU-12 member states, the second for European non-member states.

A1.9.3 Radiological impact (releases and pathways)

The radiation dose, due to the natural radionuclide content of mineral building materials, is an important component of the overall dose to the public since most people spend the majority of their time indoors exposed to both external irradiation and radon emanation. However, the issue of radiation exposure in and from houses is considered outside the scope of this project.

Similarly, the radiation exposure to the public from the construction of roads with NOR-containing waste products is outside the scope of this project. However, it should be noted that the construction of roads using phosphogypsum and calcium silicate slags was found to not to

significantly enhance the radionuclide concentration in nearby air, soil and groundwater, although an external dose of 0.20-0.35 mSv a⁻¹ can be incurred through occupying roads constantly [Martin *et al.*, 1996].

Scholten (1996) comments that the leaching of radionuclides and heavy metals or other toxicants from fly-ash is low, so that there would be hardly any restrictions for landfill.

Harvey *et al.* (1994) discuss and estimate public doses (individual and collective) arising from two exposure pathways from the disposal of old building materials (brick and concrete with pulverised coal ash). The generic properties of this landfill site close to the River Thames (London) catchment system are fully outlined in Harvey *et al.* (1994). In the migration scenario the fate of the radionuclides is assessed from release through the waste, through the immediate geosphere and into the biosphere (rivers, seas, foodchain). In the redevelopment scenario it is assumed that the landfill site has been closed for 300 years and that all records have been lost. Dose estimates during redevelopment consider the investigation, construction and residing phase. The calculations are based on data from the brick and concrete industry in the UK which are considered to be representative for whole Europe. They assume a landfill of 15000 t waste diluted with 300000 t mixed waste. The specific activity of the major nuclides are 300 Bq kg⁻¹ ²³⁸U, 14 Bq kg⁻¹ ²³⁵U and 100 Bq kg⁻¹ ²³²Th. All radionuclides are in equilibrium with their respective daughters. Results of the dose calculations are presented in Table A1.22.

Table A1.22 Doses and dominant nuclide and pathway from disposal of building material

Scenario	Individual dose (mSv a ⁻¹)	Dominant nuclide and pathway
Migration (Dose rate at 10 ⁶ a after site closure)	9.2 × 10 ⁻⁴	²³⁵ U, arable resuspension
Redevelopment (300 y after closure)		
- investigation	6.4 × 10 ⁻⁴	²³² Th, inhalation
- construction	3.0	²³⁸ U, external
- residence	1.4	²²² Rn, radon

Source: Adapted from Harvey *et al.* (1994).

A1.10 Applications of natural radionuclides radium and thorium

A1.10.1 Category description

The **radium** concentration in ores is very low. In 1920, 300 to 400 tonnes of uranium ore were required to produce one gram of radium. Only 10 t uranium rich ore (average 50% uranium oxide) was needed for the production of 1 g radium in Olen, Belgium. The world production of radium is estimated at 4.5 kg since 1898, of which half was produced in Olen, Belgium, between 1922 and 1969. The second most important producer, since 1938, has been Canada. Interest in radium decreased with the development of nuclear reactors and accelerators, through which more reliable and less dangerous and expensive radionuclides became available.

Many items of equipment were luminised in the 1940's, 50's and early 60's with radium luminising agents. Radium was also used in medicine and in material structure research.

Thorium oxide is a constituent of many minerals of which the most important are monazite (which can contain between 2.5 and 28% thorium oxide mixed with rare earths), thorite and thorianite. Thorium is obtained by mining and concentrating the minerals. The concentrate is decomposed by acids to obtain thorium salts which are the raw materials for metallic thorium. According to Penfold *et al.* (1997) European thorium dioxide is produced in Austria (Treibacher) and France (Rhône-Poulenc, La Rochelle).

Originally, thorium's main industrial application was as thorium nitrate in the manufacturing of gas mantles. It is also used in alloy production in e.g. as hardener in magnesium alloys for casting. The alloying of thorium at about 2% in tungsten electrodes, used in inert gas welding in many industries, improves the kindling of the initial arc at a lower voltage. It plays an important role in the nuclear industry, in vacuum technology and in the chemical, electronic and engineering industries.

Incandescent gas mantles without any radioactive substances are now available with similar properties. As such, the use of thoriated gas mantles is expected to decline.

A1.10.2 General description of source term

No typical values are available for the radionuclide concentration in waste dumps of radium extraction plants. Buildings or workshops where ²²⁶Ra luminising or refurbishing took place were typically contaminated in the following areas: workbenches, walls and floors, sinks etc. inside buildings and external walls near windows and burial sites next to the building. The main hazard is ²²⁶Ra, representing both an internal and external radiation hazard and its daughter ²²²Rn. Drury (1994) recorded contamination levels in the soil surrounding of luminising works to be 400 to 4×10^5 Bq kg⁻¹ ²²⁶Ra with hot spots of 4 MBq kg⁻¹. Surface dose rates ranged from background (0.1 µSv h⁻¹) to 100 µSv h⁻¹. Similar doses and concentrations were reported in the vicinity of a watch factory in Dieppe (France) [Andra 1995, 1996].

The dose rate for a radium extraction plant disposal site in Olen (Belgium) was 2.8 (0.1-1000) µSv h⁻¹. Dose rates in the vicinity of the plant were between 0.1 and 100 µSv h⁻¹ [Cottens *et al.*, 1994].

Dalheimer and Hendrichs (1994) give some specific activity concentrations of thorium containing products. These are presented in Table A1.23.

Table A1.23 Activity levels of products containing thorium

Product	Activity Bq kg ⁻¹
Gas mantles	1 000 000 (1000-4000 Bq per piece)
Discharge lamps	100-1000 Bq per piece
Thoriated glass	400 000
Thoriated tungsten, welding rods	100 000
Titanium oxide (pigment)	50 000

Source: Adapted from Dalheimer and Hendrichs (1994).

A1.10.3 Radiological impact (releases and pathways)

Exposure data during production of thorium-containing products are presented in the Penfold *et al.* (1997) study. However, no data were found on past practice radiological hazards. Martin *et al.* (1996) report the results of a study where the dose to firemen in the event of a warehouse fire involving 50000 welding rods was estimated at 1 Sv. Careful changing of gas mantles results in a mean inhalation of 0.5 Bq; careless exchanging can lead to the inhalation of 18 Bq. In the USA it is estimated that the annual collective dose to the population is 86 man-Sv by the distribution and use of 25 million gas mantles a year.

A1.10.4 Occurrence in European countries

Information was found for only for a few example cases. These are presented in Table A1.24. For additional information about these cases, the reader is referred to Annex A2.9.

Table A1.24 Available information of sites related to radium industry in Europe

Country	Type of liability	Site described
Belgium Olen	Radium extraction	Old disposal site
France Dieppe	Watch factory	60 m ³ soil excavated
UK East London	* Radium/thorium factory - extraction of Th and rare earths from monazite - radium luminising facility - production of chemical compounds	13 ha of land, contamination buried 6 m deep
RAF Carlisle Dalgety Bay	* Radium luminising facility * Military airfield and dismantling works	Burial site, 92 hot spots largely associated with clinker material located 0.3-0.6 m below surface of beach.

Penfold *et al.* (1997) report a 'UK-supplier' of thorium alloys as hardener for magnesium alloys which ceased operation 10 year ago. Except for the industrial site in East London, described above, no other information was found about specific workshops for thorium applications. According to a German study reported in Penfold *et al.* (1997), the two European producers of thorium oxide are located in Austria (Treibacher) and France (Rhône-Poulenc in La Rochelle). Andra (1998) reports seven sites in France with potential contamination due to radium extraction, six sites contaminated with radium or tritium and more than 50 sites where radium was used for chemical purposes. Thorium was produced by Rhône-Poulenc in La Rochelle and by SMIF in Clichy.

A1.11 References

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ANNEX A2
OVERVIEW OF CONTAMINATED SITES IN EUROPE

Table of Contents

A2.1	Uranium mining and milling	106
A2.2	Metal mining and smelting	112
A2.3	Phosphate industry	115
A2.4	Coal mining and power production from coal	118
A2.5	Oil and gas drilling	121
A2.6	Rare earth and titanium oxide industries	124
A2.7	Zirconium and ceramics industries	124
A2.8	Wastes for building materials	124
A2.9	Applications of natural radionuclides radium and thorium	125
A2.10	References	127

A2.1 Uranium mining and milling

A2.1.1 Albania

[Source: Uranerzbergbau, 1997].

There was no uranium production in Albania. Exploratory work, however, resulted in 127 adits and 77·280 m³ waste rock.

A2.1.2 Bulgaria

[Sources: Vapirev *et al.* (1994); Vapirev *et al.* (1996); Dimitrov and Vapirev (1996); Dimitrov *et al.* (1996); and Hébert *et al.* (1996)].

After World War II, the first uranium mines were established in the deposits near Buchovo and Streltsha. The ore was shipped to the USSR until a first processing factory was built in the Buchovo region in 1947, close to living areas. Following a countrywide exploration, numerous, low grade (<0·1%) deposits were discovered. Most of the ~40 mined deposits are located in the Western Balkan and the South of Bulgaria. To process the ore from the southern deposits, a new mill was built in Eleshnitsa in 1966. This plant did not produce the final product (yellow cake). This was dried, ground and packed in Buchovo. Up until 1979 underground mining predominated. For a more effective exploitation of low grade ore deposits (0·006 - 0·03%), investigation into *in-situ* acid leaching (ISL) began. During the whole period of uranium industry in Bulgaria, 23 underground and 4 open pit mines as well as 17 ISL sites existed (13 according to Uranerzbergbau 1997). Following a government decision, the uranium industry was closed in 1992. According to Vapirev *et al.* (1994), there are 298 waste heaps covering an area of 85 ha and 8·3 × 10⁶ m³ (13·7 Mt). There are three tailings ponds, representing 10 × 10⁶ m³ [18·5 × 10⁶ m³ according to Uranerzbergbau (1997)]. The total area contaminated by the uranium industry is approximately 20 km², including 4 km² of forest. The major part is agricultural land along the Rivers Maritsa, Tundzha and Struma, which have been lent for ISL [Vapirev *et al.*, 1996].

No clear data are available about the amounts of uranium that have so far been produced in Bulgaria. However, a production of 22000 tonnes of uranium can be estimated [Hébert *et al.* 1996; Vels and Ruhrmann, 1994].

Around the Buchovo mill (situated 15 km from Sofia), the major radioactive hazards are re-suspension of dust from the filled and dried tailings pond, radon emanation from the milled rock and radium contamination of agricultural land (up to 10 kBq kg⁻¹, corresponding to a dose of 10 μSv h⁻¹) of which a certain fraction is in villages. Around the milling plant in Eleshnitsa, the main hazard is radon emanation. The environmental impact of the underground mines is determined by the conditions of the mine water. The main radiation hazard is due to radium in the surface water, which has a radium content between 50 and 800 mBq L⁻¹ (limit 150 mBq L⁻¹). The waste heaps from the underground mines (approximately 130 × 10⁶ m³) have an average specific mass activity of 6·3 kBq kg⁻¹ and can be considered as non-radioactive, but with enhanced levels of NORM. The ion exchange resins used for ISL are enriched to 1000-1300 kBq kg⁻¹, surpassing the limit of 7 kBq kg⁻¹ for alpha-emitters. The most serious radioactive hazard is the possible contamination of groundwater which reaches the region of the ore. However, the radiation hazard is less important than the chemical and acid contamination. The quantity of sulphuric acid is estimated at 2·5 million tonnes.

Some proposals for remediation of the mining and milling sites have been made. These are described by Dimitrov *et al.* (1996).

A2.1.3 Croatia

[Source: Saler *et al.* (1996)].

No uranium mining has taken place in Croatia.

A2.1.4 Czech Republic

[Sources: Andel and Priban (1996a,b,c); Hébert *et al.* (1996); and Uranerzbergbau (1997)].

The mineable uranium deposits of former Czechoslovakia are situated, without exception, in the present Czech Republic. With an uranium production of 103 kt [118 kt according to Uranerzbergbau (1997)] between 1946 and 1992, it was the sixth largest uranium producer world-wide. The countrywide uranium exploration programme after World War II led to the discovery of many deposits, the most important of which are the vein deposits around Pribram (40% of total uranium production; exhausted in 1992). There are a total of 106 uranium mining sites with 16 open pit and 99 underground mines. Four mechanical processing sites were in operation until 1960 and, one until 1991. There were four hydrometallic processing sites, of which only one was still in operation in 1994 [Hébert *et al.*, 1996]. ISL has only been carried out in one ore zone [two ISL mines according to Uranerzbergbau (1997)] and there is still restricted production. Hébert *et al.* (1996) report 544 waste dumps covering 450 ha and comprising of 121 Mt ($\sim 80 \times 10^6 \text{ m}^3$) and 20 tailing ponds, covering 473 ha and containing 49 Mt ($\sim 32 \times 10^6 \text{ m}^3$). According to Uranerzbergbau (1997) there are 349 dump sites containing $57 \times 10^6 \text{ m}^3$ and 21 tailing ponds amounting to $43 \times 10^6 \text{ m}^3$.

Except for some data on gamma-irradiation and uranium contents of a river [Andel and Priban, 1996a], no data are available on radiocontamination levels. Planning for environmental restoration in the river have been described by Andel and Priban (1996b) and technologies and implementation of water decontamination techniques in Andel and Priban (1996c).

A2.1.5 Estonia

[Sources: Putnik *et al.* (1994); Putnik (1996); Hébert *et al.* (1996); Uranerzbergbau, (1997); and E.K.K. Rajamäe (Personal Communication)].

Estonia does not have important uranium deposits. However, since 1948, a large facility for treatment and production of uranium for civil and military purposes has been operating at Sillamäe, a town with 20000 inhabitants in the Gulf of Finland. Initially it processed low grade (0.03%) local alum-shale but, later on, it became the processing centre for ores (up to 1%) from different Eastern European countries. The total amount processed was 4 million tonnes of ore with an uranium production of 101 kt [Uranerzbergbau, 1997]. In the beginning of 1970, the plant switched to the processing of loparite, rich in niobium, tantalum and other rare earths. In addition, loparite contains elevated concentrations of uranium (about 0.03%) and, in particular, thorium (0.6%). Since 1977, no uranium ore has been processed and the plant is only operating at 15-20% of its capacity. The mill tailings deposit covers 33 ha and contains 12.7 Mt of tailings. Approximately half of it is residue from uranium ore processing (in Bq kg⁻¹: 1500-23000 ²³⁸U; 1000-220000 ²²⁶Ra; 25-330 ²³²Th) and half from loparite processing and oil shale ash processing (in Bq kg⁻¹: 180-3600 ²³⁸U; 130-2300 ²²⁶Ra; 200-4000 ²³²Th). The estimated total amounts are 1830 t uranium [1200 t according to Putnik (1996)], 850 t thorium and 7.8 kg radium [Rajamäe, Personal Communication]. Several international research projects are dedicated to the evaluation of the environmental hazard.

A2.1.6 Finland

[Source: Markkanen and Annanmäki (1994)].

Uranium was experimentally mined and processed in Paukkajanvaara (east) between 1958 and 1961. The mining and processing wastes were untouched until 1992, when the remediation of the area begun. The 30 kt of processing wastes have an average ^{226}Ra concentration of 10 MBq kg^{-1} .

A2.1.7 France

[Sources: Daroussin and Pfiffelmann (1996); Crochon and Daroussin (1996); Faussat and Fernique (1994); and Hébert *et al.* (1996)].

CEA began to extract uranium ore in France about 45 years ago and then COGEMA took over in 1976. More than 50 Mt of ore have been processed and 70 kt of uranium were produced as concentrates. Years of uranium mining by COGEMA and subsidiaries have led to more than 200 mining sites in France, 75% of them being more than one hectare. The mines were either open pit or underground and each has an associated waste dump. There are eleven industrial sites, where operations were mill or heap leaching, and 22 storage sites for the residues of ore treatment residues.

Only two mills are still operating, four are closed and remediated and for three site remediation is still in operation. Sites have closed due to exhaustion of deposits or due to lack of provisions.

Due to the very low grade of uranium ores which were treated in France (average 0.15-0.23% uranium, if heap leaching is excluded), more than 99% goes to waste and important tonnages have to be dealt with (nearly 50 Mt by the end of 1993). Dismantling of the mills leads to several thousand tonnes of slightly contaminated concrete debris; the most contaminated equipments being those for attack and resin extraction. Of major concern are the residues coming from the treatment of uranium ore. Heap-leaching waste contains 20-40% of the original uranium content of the ore (0.015-0.06%) and 100% of the original weight. The total inventory as given by Hébert *et al.* (1996) as 632 t uranium in 849 kt low grade ore; 3595 t uranium and 830 TBq ^{226}Ra in 29 Mt fine residues and 1530 t uranium and 63 TBq ^{226}Ra in the coarse residues. The ^{226}Ra activity is $2000\text{-}7000 \text{ Bq kg}^{-1}$ and the total mass activity about 40000 Bq kg^{-1} . For the mill tailings, 3-10% of the original ore grade (0.1-1%) is residual uranium, being present in ~110% of the original weight (32 Mt in 1992) and with an average ^{226}Ra and total activity of 20000 and $300000 \text{ Bq kg}^{-1}$ respectively [Daroussin and Pfiffelmann, 1996].

Tailings storage sites are between 50 and 1000 m from the closest population and the average population density of the district is between 10 and 370 inhabitants per m^2 .

A2.1.8 Germany

[Sources: Ettenhuber (1996); Rönsch and Ettenhuber (1994); Biesold *et al.* (1994); Ettenhuber *et al.* (1996); Ettenhuber and Rönsch (1996); Hébert *et al.* (1996); and BMWi (1993)].

Between 1946 and 1990 geological-geophysical explorations of mining ore were carried out by the WISMUT AG in the Southern part of the former GDR over a total area of 55000 km^2 . Within the framework of geological expeditions, 386000 surface drillings were made. The mining activities of WISMUT AG covered an area of 240 km^2 of which 37 km^2 are, today, in need of remediation. Ore production (average grade 0.08-0.1%) took place in nine mining enterprises with 240 shafts (in 1990 there were still 56 shafts within 110 km^2) and five open-cast mines of which 5 were used as tailing ponds. Altogether, 14 tailing ponds existed with a total area of 720 ha ($160 \times 10^6 \text{ m}^3$). Waste rock piles and dumps number about 48, covering an area of 1500 ha ($311 \times 10^6 \text{ m}^3$). There are numerous underground mining works (1395 km

extent) and one open pit mine (1.6 km² with 84 × 10⁶ m³ open). Milling was predominantly carried out in Seeligenstadt (acid pressure leaching, alkaline leaching from October 1989) and Crossen (alkaline leaching). There were also some small local installations. Between 1946 and 1990 the WISMUT AG produced 220 kt of uranium and was the world's third largest uranium producer after the USA (333 kt) and Canada (240 kt) [Vels and Ruhrmann, 1994].

It should be noted that information differs for different sources: most information is from BMWi (1993), some from Hébert *et al.* (1996) and Ettenhuber (1996). All sites are closed and an extensive monitoring programme (with establishment of database ALASKA) and remediation has begun (flooding of mine; backfilling of the open pit, stabilisation of tailings, covering of the waste rocks, decommissioning of the facilities). For more information the reader is referred to Ettenhuber (1996). Some indicative values for river water quality, and air quality (gamma radiation and radon concentrations) can be found in Hébert *et al.* (1996). The data given in Tables A1.4-A1.6 of Annex A1, for Germany, only comprise of the WISMUT sites.

Besides of the WISMUT sites, there are numerous sites in former East Germany, where uranium and non-uranium (silver, cobalt, copper) ore and coal mining are abandoned by operators who are not available or legally responsible anymore. Many of these sites are in commercial or public use. As such, 34 areas of suspicion have been identified, covering 1500 km². About 8000 mining relics (73 km² and about 350 × 10⁶ m³) of different types (waste rock piles, tailings ponds, slag deposits) have been identified and registered in the data bank ALASKA by BfS [BfS, 1992 and 1994].

The areas in Saxony and Thuringia are densely populated (250-2300 inhabitants km⁻²). This creates a potential risk to the population through water and airborne pollutants, through toxic elements (bismuth, arsenic, acidity) and heavy metals and natural radionuclides.

Hébert *et al.* (1996) also report two mining areas in Western Germany. In Mähring (Bavaria), mining took place from 1968 until 1982, with uranium extraction through heap leaching or bacterial leaching. In Ellweiler open-pit mining took place from 1961 until 1972 and dynamic leaching until 1989. For both sites, 793 kt of tailings were produced containing 9.1 TBq ²²⁶Ra. About 15.4 t uranium was mined.

A2.1.9 Greece

[Source: C. Papastefanou (Personal Communication)].

Uranium deposits are in the North of Greece in the area of Paranesti, Drama at the Fteroto and Morfovouni sites.

A2.1.10 Hungary

[Sources: Hébert *et al.* (1996); Juhasz and Erdi-Krausz, (1996); Juhasz *et al.* (1996a,b); and Uranerzbergbau (1997)].

Hungary has only one uranium mining site in the southern part of the country nearby Pecs (5 km from site, 170000 inhabitants). The uranium is produced from a low-grade, underground sandstone ore (0.07-0.12% uranium) [Juhasz and Erdi-Krausz, 1996]. Mining started in two mines in 1958. The total mining complex consists of five mines of which five are now exhausted. Until 1962 the ores were only ground and sorted and transported to the former USSR; later the milling was completed (acid and alkaline leaching).

The total amount of uranium production from 1958 and 1993 was estimated to be between 17100 and 19970 t [Hébert *et al.*, 1996] [20100 according to Uranerzbergbau (1997)]. The present annual ore production is 400-600 kt a⁻¹.

The total amount of waste rocks disposed of near the mines is 18 Mt [450 ha, 90 t uranium, specific activity 8000 Bq kg⁻¹; according to Juhasz and Erdi-Krausz (1996): seven dumps; according to Hébert *et al.* (1996): nine ore dumps and ten additional dumps: five from waste and from heap leaching. The mill tailings have been placed in two retention ponds (one out of operation) and contain 19 Mt solids and 7 Mt solution. The amount of ore heaps for alkaline extraction (operating from 1965 until 1990) process is 7 Mt (60 ha, 560 t uranium). According to Uranerzbergbau (1997) there was some *in situ* leaching. Radiation levels on mining and milling area vary from 10 to 100 times background.

Since the uranium mining and milling activities are close to shut down, a radiological impact analysis of the mining and milling area has been carried out [Juhasz *et al.*, 1996a] and remediation of some of the waste rock piles has been performed [Juhasz *et al.*, 1996b]. Uranium, in the form of dust and radon, as a gas, are the main sources of air pollution. In conclusion, 90% of the radiation burden is the result of inhalation of the airborne activity and the ingestion of contaminated food. The critical radionuclide is ²²⁶Ra, the critical pathway is leafy vegetables (contaminated with uranium dust) and the critical group is a one year old child. After restoration work (covering) the critical radionuclide is still ²²⁶Ra, the critical pathway is contaminated tuberous vegetable and the critical group is a one year old child. Restoration of one of the major waste rock piles have been performed and resulted in an about five-fold reduction in external exposure and radon inhalation. More detailed information can be found in [Juhasz *et al.*, 1996b].

A2.1.11 Poland

[Sources: Hébert *et al.* (1996); Piestrynski *et al.* (1996); and Uranerzbergbau (1997)].

In the early 1950s about 100 locations containing uranium minerals were found in the lower Silesia (SW). Extraction of uranium ores took place at three sites and had probably finished before 1960. In the small uranium deposits, ore grades of 0.8% were reported. Uranium production is estimated to total 1000 t uranium [Hébert *et al.*, 1996] [695 t uranium according to Uranerzbergbau (1997)]. At Kowary there are 11 dumps (two disappeared few years ago and are used for building), at Radoniow there is one (2.5 ha, 75% of former volume use for building) and, at Kletno there are 24 (two with elevated radiation) [Hébert *et al.*, 1996] [a total of 35 dumps according to Uranerzbergbau (1997)].

No information is available about uranium contents, reserves and total tonnage of the extracted ore or in the waste piles. Piestrynski *et al.* (1996) provide some gamma-radiation measurements, performed in 1971 and 1991. Uranium pollution seems to be limited to the dumps and their nearest vicinity. All dumps are unprotected and accessible to the public.

A2.1.12 Portugal

[Source: IAEA (1992)].

In Portugal, there is one small uranium mill, with 3.5 tonnes of tailings. No additional information was found.

A2.1.13 Romania

[Sources: Ghilea *et al.* (1994); Hébert *et al.* (1996); Sandru (1996a,b,c); and Uranerzbergbau, (1997)].

Following an exploration campaign, several deposits were identified in the Apenusi mountains (up to 1.2% uranium content in the Baita Bihor area) and the Banat Mountains. Further deposits were discovered, mainly in the eastern Carpathians, in the period up to 1989. Uranium extraction began in the Baita Bihor deposit in 1952 and in the Banat Mountains, in 1956. All

the ore was shipped to Estonia for extraction. 14500 t uranium were produced between 1952 and 1961. Uranium exploitation ceased from 1962 until 1978, when a processing plant was built in Fedioara. Between 1980 and 1992, 2350 t uranium were extracted. Due to depletion or for economical reasons many mines closed and at present it is believed that only four of the original 22 mines are in production [Hébert *et al.*, 1996]. Most of the mines are underground mines.

According to Sandru (1996a,c), there are about 173 contaminated sites containing about 5.35 Mt (1382 ha, $5.4 \times 10^6 \text{ m}^3$, 161 piles) of waste rocks and 30400 t (1.85 ha, 30004 m^3 , 10 piles) of low grade uranium ore (average 0.02%). There are two tailing ponds covering 35 ha and containing $1.75 \times 10^6 \text{ m}^3$. So far no remediation has been carried out. The estimated funds needed are \$1.5 million.

A2.1.14 Slovakia

[Sources: Hébert *et al.* (1996); and Uranerzbergbau (1997)].

Although Hébert *et al.* (1996) report that all minable uranium deposits of the former Czechoslovakia are without exception situated in the Czech republic, Uranerzbergbau (1997) reports an uranium production of 201 t in Slovakia, with 26 waste rock and/or ore dumps containing $1.15 \times 10^6 \text{ m}^3$ of material.

A2.1.15 Slovenia

[Sources: Logar (1996); Krizman (1996), Krizman and Logar (1996); Hébert *et al.* (1996); and Uranerzbergbau (1997)].

Several uranium ore deposits were discovered following an intensive exploration programme. However, only the Zirovski deposit was of economic value. Underground mining began in 1982 and uranium concentrate production, by acid leaching, in 1985. The mine stopped its regular operation in 1990, mainly for economic reasons. In 1992, the permanent close-out of the mine was decided and the environmental restoration of the area and decommissioning initiated.

During the mining operation 540000 t of ore were excavated [0.084% U_3O_8 according to Hébert *et al.* (1996), 1.2% U_3O_8 according to Logar (1996)]. The uranium concentrate production amounted to 452 t [383 t according to Uranerzbergbau (1997)]. According to Hébert *et al.* (1996) there are three dump sites (3.4 ha, 300000 t) and one tailings pile on a hill slope (4.3 ha, 375000 m^3 , 100000 t). Logar (1996) reports 1.5 Mt mine waste and 0.7 Mt tailings (120 mg kg^{-1} uranium). Krizman (1996) also reports a deposit of some thousand tons of uranium ore. The mine waste has an average uranium content of 70 mg kg^{-1} and consists of mine waste material under 300 mg kg^{-1} U_3O_8 , mill raffinated neutralisation filter cake (red mud; 40000 t; high ^{230}Th content) and some building construction material. The mill waste tailings pile represents a serious reclamation problems because of the area is sliding. The moving mass is 7 Mt. A third main radioactive contaminant source is run-off mine water (60-90 $\text{m}^3 \text{ h}^{-1}$). From the environmental and dose assessment study [Krizman, 1996] it was concluded that the main radiological impact to the critical group appears to originate from inhalation of radon and its short-lived daughters.

A2.1.16 Spain

[Sources: Pérez Estévez and Sanchez Delgado (1996a,b); and Santiago (1994)].

In many parts of Spain, uranium exploration and mining began in 1950 (ore purity between 0.06 and 0.23%). Mining was in open pits or underground. There were four mills [Salamanca (2), Andujar, Badajoz] of which one was still operating in 1996. At three of the four mill sites,

heap leaching was performed. Many mines were associated with each mill, for example, 24 mining sites with the Andujar mill. Many of the mining sites were abandoned in the 1970s due to lack of interest or exhaustion of the mine. Of the three closed mill sites, the Andujar site is remediated while for the other two, remediation is in operation.

A2.1.17 Sweden

[Sources: Ehdwall (1996); Sundblad (1996); Sundblad and Stiglund (1994 and 1996)].

In Sweden, large, but low grade (0.025-0.033%), uranium ore reserves are found in the district of Västergötland. At Randstad, an uranium mill (acid leaching) was operating between 1965 and 1969, during which time 1.5 Mt of alum scale were mined (open pit) and 215 t of uranium had been produced. Tailings cover an area of 25 ha (volume of material $1 \times 10^6 \text{ m}^3$). Restoration was mainly performed during 1990 and 1992.

A2.2 Metal mining and smelting

A2.2.1 Aluminium industry

A2.2.1.1 Croatia

Subasic *et al.* (1996a) report a light metal plant in Sibenik. Given the supposed thorium content in sludge, generated in the aluminium industry during the treatment of hydrated alumina, attention should be paid to the possible radiological consequences.

A2.2.2 Copper industry

A2.2.2.1 Bulgaria

During the period of 1954 and 1977, the waste from the polymetallic mine "Rosen" (Burgas Copper mines), which is ~30 km east of the town Burgas on the Black Sea, was dumped in the Bay of Vromos. The slime was predominantly rock and 2-6% polymetallic ore. The total waste is estimated to be approximately 8 Mt. On the coast, the waste is 1300 m long, 120 m wide and 2-3 m thick. The exposure rate varies between $1.2-1.5 \mu\text{Sv h}^{-1}$ in the central part and $0.3-0.4 \mu\text{Sv h}^{-1}$ at the edge. In 1985, an experimental project was started, re-extracting 10×10^3 tonnes of ore and the re-processing proved economically feasible [Vapirev *et al.*, 1994].

A2.2.2.2 Germany

Within the EU, mineral deposits in eastern Germany are known to contain some of the highest concentrations of naturally occurring radioactive material. Only copper mining and melting in the Mansfeld area will be considered here because, for the other metal mining and milling activities, there is a lack of sound data and they are of lesser importance. Copper mining and smelting in Mansfeld area began in the 12th century and was halted in 1990. Originally, there were numerous small near-surface pits and dumps of waste rock and slags. Later on, the mining and smelting industry was concentrated at a few sites only and shafts were dug to depths of 800 m. Since 1969, further amounts of copper ore from the adjoining district of Sangerhausen were smelted in the Mansfeld region where ore deposits were exhausted. Mining smelting, metal-processing and metal-working industries were closed down in 1990 for economic reasons [Wichterey, 1994].

In the Mansfeld deposits, copper ores were frequently accompanied by uranium minerals. Unfortunately, the numerical data available concerning the radionuclide concentration are

incomplete and often inconsistent [NUKEM, 1995]. A total of 1253 waste piles of various sizes were created amounting to about $110 \times 10^6 \text{ m}^3$. These piles consist mainly from dead rock (>90%) from the copper ore segregation, slags, roast products and other non-copper related materials. Additionally $2.9 \times 10^6 \text{ m}^3$ sludge has been dumped into the area of which, roughly, 25% can be allocated to Theisen sludge.

From radiation protection point of view, sites of copper smelting works are of special importance since the dusts and sludges from the process of smelting are contaminated by the volatile ^{210}Pb and ^{210}Po which can be widely distributed by atmospheric pathways (dust from piles and from the smelters) over distances of 3 km. Certain smelters, used for reprocessing lead-containing dusts, are so heavily contaminated that extensive and complete remediation is unavoidable prior to any other use. Slag piles present no problem of radiological protection with the exception of an increased external exposure in cases of direct stay on the sites (e.g. houses built on former slag). The slag material should not be used for the building of houses. "Black" waste rocks are insignificant with respect to radiation exposure as long as they are not used for building purposes. For "white" waste rocks the radionuclide concentration is below 200 Bq kg^{-1} and so no licensing is required for use.

The atmospheric pathway is the critical pathway for inhalation as well as ingestion of contaminated products. Therefore investigations in the surroundings of smelting sites have to be carried out.

A2.2.3 Iron and steel industry

A2.2.3.1 General, Netherlands and UK

Scholten (1996) provides information about the steel industry in the EU. Crude steel production in the EU-12 was 130000 million tonnes (Netherlands ~6000, Belgium, Luxembourg and Spain ~13000, UK and France ~17000, Italy ~26000 and Germany ~37500). Natural radioactivity levels in the ore of $<50 \text{ Bq kg}^{-1}$ for the activity of the ^{238}U and ^{232}Th chains. About 2000 tonnes of potentially contaminated dust is produced per million tonnes of steel. For the British site, the total activity of the sludge is $<10 \text{ Bq g}^{-1}$. For the Dutch industry $\sim 100 \text{ Bq g}^{-1}$. Both sites are still operational.

A2.2.4 Lead industry

A2.2.4.1 Finland

Between 1961 and 1972 lead was mined and processed in Kosnäs, on the west Coast of Finland. The average uranium and thorium concentrations of the 760000 t waste are estimated to be 700 and 250 Bq kg^{-1} , respectively. Also, about 36000 t of milled ore containing 1500-4500 Bq kg^{-1} uranium and 1000-1500 Bq kg^{-1} thorium is stored at the mining area. The Kosnäs area is fenced. If the area is used for recreation then the estimated dose will be 0.3 mSv a^{-1} . Remediation is needed to fulfil the safety requirements of 0.1 mSv a^{-1} [Markkanen and Annanmäki, 1994].

A2.2.5 Mercury industry

A2.2.5.1 Slovenia

Logar (1996), Krizman (1996) and Krizman and Logar (1996) only briefly mention the existence of some mercury ore wastes but do not discuss them since they are of minor radiological importance compared to wastes from uranium mining and milling. In the town of Idria, the mercury mining district, residues of ignited mercury ore and the mine vents (in

closing phase but still in operation) cause high outdoor concentrations of radon in the town (50-100 Bq m⁻³). Indoor radon concentrations can reach up to 1500 Bq m⁻³). Krizman and Logar (1996) also refer to ore processing factories (ilmenite and bauxite).

A2.2.6 Niobium industry

A2.2.6.1 United Kingdom

Two closed UK-factories are reported having produced ferroniobium from pyrochlore (one closed in 1980, the other in 1992). A mixture of pyrochlore, aluminium powder and aluminium wires and iron chippings are melted. The activity concentrations of the pyrochlore were 10000 Bq kg⁻¹ ²³⁸U and 80000 Bq kg⁻¹ ²³²Th. Slag production is about the same as ore use and is mainly disposed of as shallow land burials [Harvey *et al.*, 1994; Martin *et al.*, 1996].

A2.2.7 Tin manufacturing

A2.2.7.1 United Kingdom

Harvey *et al.* (1994) describe a tin manufacturing factory in the UK which was operational between 1971 and 1992. The ore typically contained 1000 Bq kg⁻¹ ²³⁸U and 300 Bq kg⁻¹ ²³²Th is smelted and then separated into components containing tin, lead, bismuth/lead alloy. During metal production, contaminated fume was produced containing 200000 Bq kg⁻¹ ²¹⁰Po but little ²¹⁰Pb. Solid wastes were mostly produced as furnace slags. These were produced at a rate of 55 kt a⁻¹ and contained 4000 Bq kg⁻¹ ²³²Th and ²²⁸Ra and 1000 Bq kg⁻¹ ²³⁸U and ²²⁶Ra. In the case studied, the slag was sold to a neighbouring factory for road construction. A tellurium dross (metal surface oxide) which was heavily contaminated with ²¹⁰Po was also produced at the rate of 600 kg a⁻¹. It was stored until the polonium concentration decayed to 20000 Bq kg⁻¹ and then sold.

A2.2.8 Zinc industry

A2.2.8.1 Finland

At the Vihanti Zinc mine which closed in 1992, wastes contained, on average, 400 Bq kg⁻¹ of uranium. The area has been covered with a soil layer, which, together with the vegetation, will prevent dust dispersion and reduce gamma irradiation. These measures were assessed to adequately to fulfil the safety requirements of the area [Markkanen and Annanmäki, 1994].

A2.2.8.2 Sweden

In Sweden there are many mines and mills but none which have caused contamination [H. Ehdwall, Swedish Radiation Protection Institute, Personal Communication].

A2.2.9 Economic data

[Source: Eurostat (1995)].

Economic data are summarised in Tables A2.1 and A2.2.

Table A2.1 Aluminium, copper and lead production in the EU for 1993 (1000 t metal content)

Country	Aluminium ore	Primary Aluminium	Copper ore	Refined Copper	Lead ore	Refined Lead
Belgium	-	-	-	298.9	-	108.0
Denmark	-	-	-	-	-	-
Germany	-	551.9	-	632.2	-	334.6
Greece	1684.9	147.7	-	-	26.4	-
France	104.0	462.2	0.1	59.2	-	258.7
Ireland	-	-	-	-	44.6	12.0
Italy	98.0	155.7	-	88.0	7.4	188.1
Luxembourg	90.1	-	-	-	-	-
Netherlands	-	231.8	-	-	-	24.2
Portugal	-	-	150.4	-	-	4.0
Spain	-	355.9	3.5	179.2	25.3	54.1
United Kingdom	-	239.1	-	46.6	1.0	364.0
EU-12: Production	2976.0	2144.3	154.0	1304.1	104.7	1347.7
EU-12 (1987), Mining	1052		19		176	
Recovering	1314		981		540	
Consumption	6432		2811		1322	

Table A2.2 Iron and steel, niobium and tin production in the EU for 1993

Country	Iron ore (1000 t) (metal content)	Pig iron (1000 t)	Crude steel (1000 t)	Niobium (1000 t)	Refined Tin (1000 t) (metal content)
Belgium	-	8178	10178		0.2
Denmark	-	-	604		-
Germany	109	26970	37625		0.1
Greece	-	-	980		0.2
France	3542	12665	17107		-
Ireland	-	-	329		-
Italy	-	11200	25817		-
Luxembourg	-	2411	3292		-
Netherlands	-	5406	6001		0.2
Portugal	-	397	775		0.1
Spain	2080	5394	12960		2.0
United Kingdom	29	11600	16707		2.8
EU-12: Production	5760	77301	132375		5.6
EU-12 (1987) Mining	5826			-	4.2
Recovering	52282			0.03	14.5
Consumption	118359			3.3	54.5

A2.3 Phosphate industry

A2.3.1 Belgium

[Sources: Baetslé (1991); and Internal SCK•CEN reports].

Tessenderlo Chemie treats Moroccan ore at Tessenderlo and Ham. They use the hydrochloric acid process which results in the production of a waste which consists predominantly of CaF₂. Between 30-40% of the radium from the ore (~1500 Bq kg⁻¹) remains in the CaF₂-waste (3000-4000 Bq ²²⁶Ra kg⁻¹ on dry matter). There are several dump sites. One of those sites

consists of three separate disposal sites of which one old sludge disposal site (in use from 1963-1986) covers an area of 25 ha and was not lined at the bottom. This disposal site was fully characterised. More information can be found under the description of the selected cases.

A2.3.2 Croatia

[Sources: Saler *et al.* (1996); Subasic *et al.* (1996a,b)].

Saler *et al.* (1996) report two sites containing phosphates and phosphogypsum, remaining from the fertiliser industry: 1) The INA-PETROKEMIJA, fertiliser plant in Kutina and 2) the Port of Sibenik, through which large quantities of phosphate rock were imported. At the Port of Sibenik, where phosphate imports stopped due to the war in this region, no data on possible pollution are available.

At INA-PETROKIMIJA, the phosphogypsum is transported 5 km from the facility through pipelines. There are four pools (landfills) organised in the flood plains of the Sava river. The total surface is 136 ha and the capacity is $20 \times 10^6 \text{ m}^3$ of which only $3.5 \times 10^6 \text{ m}^3$ have been used to store phosphogypsum. The mean radioactivity in the deposit (sampled in 1988) is $\sim 110 \text{ Bq kg}^{-1} \text{ K}$, $22 \text{ Bq kg}^{-1} \text{ }^{228}\text{Ra}$, $650 \text{ Bq kg}^{-1} \text{ }^{226}\text{Ra}$, $560 \text{ Bq kg}^{-1} \text{ }^{238}\text{U}$ and $26 \text{ Bq kg}^{-1} \text{ }^{235}\text{U}$ [Saler *et al.*, 1996]. In 1994, radium measurements estimated a concentration of 537 Bq kg^{-1} [Subasic *et al.*, 1996a]. This would correspond in a total ^{226}Ra activity of $1.99 \text{ TBq }^{226}\text{Ra}$. Additional measurements have been carried out in ground water ($80 \text{ Bq m}^{-3} \text{ }^{226}\text{Ra}$), in well-water ($40 \text{ Bq m}^{-3} \text{ }^{226}\text{Ra}$) and waste water ($50 \text{ Bq m}^{-3} \text{ }^{226}\text{Ra}$). Exposure at the nearest house to the facility (2.5 m) was 1.26 mSv a^{-1} [Subasic *et al.*, 1996a].

A2.3.3 Estonia

[Source: R. Rajamae, Estonian Radiation Protection Centre (Personal Communication)].

There are wastes with enhanced concentrations of NORM, resulting from phosphate mining and fertiliser production at Maardu, near Tallinn (North Estonia). The total open pit mine area is 1055 ha which are mainly covered with spoil heaps about 20-25 m thick. These contain in the mixture reworked carbonate rocks and about 75 Mt of alum scales. The latter is characterised with an uranium content of 35 mg kg^{-1} . The thorium concentration is close to the Clark value. The alum shale also contains 15% organic matter which causes spontaneous combustion of the heap. Due to this, a lot of heavy metals can be released and result in groundwater contamination. Some small characterisation projects are being carried out but the environment contaminant problem is not yet fully quantified.

A2.3.4 Greece

[Source: C. Papastefanou, Aristotle University of Thessaloniki (Personal Communication)].

There are two phosphate industries: one in Thessaloniki, one in Nea Karvali, near Kaval (North Greece). No further information is available.

A2.3.5 Netherlands

[Source: Pruppers *et al.* (1997)].

According to Pruppers *et al.* (1997), there are two phosphoric acid plants and one elemental phosphorous plant operational in the Netherlands.

A2.3.6 Romania

[Source: Sandru (1996a,b,c)].

About 85% of the imported phosphate ores are of sedimentary origin with uranium contents between 0.008 and 0.015%. The phosphate industry is located in eight large plants, processing $\sim 3 \text{ Mt a}^{-1}$ phosphate, with a total of 200 t a^{-1} uranium. Nitric acid is used to treat 50% of the phosphate ore, leaving almost no activity in the resulting calcium carbonate. The 50% is treated with sulphuric acid, resulting in an annual production of about 2 Mt phosphogypsum. To date, nothing has been done with the huge deposits, which have an average radium content of $500\text{-}1000 \text{ Bq kg}^{-1}$ and are situated close to human settlements (within 1-2 km range). All four sites [Calugareasca (60 km north of Bucharest), Navodari (250 km south), Bacau (300 km northeast) and Turnu-Magurele (100 km southwest)] contain about 5 Mt of deposits spread over an area of 20 to 40 ha. The mean activity is between $500\text{-}1000 \text{ Bq kg}^{-1}$. No site-specific remedial action plan has been prepared so far.

A2.3.7 Slovenia

[Sources: Logar (1996); Krizman (1996), Logar and Krizman (1996)].

Local disposal sites are identified near ore processing facilities (e.g. phosphate industry). However, to date, all effort has gone into the restoration of the former uranium mine which is a more urgent matter.

A2.3.8 Spain

[Source: Cancio *et al.* (1994)].

Since the middle of the 1960s, an important phosphate industry has been operating in an industrial area close to Huelva, a city with 130000 inhabitants, located on the Atlantic coast in the southwest of Spain. The exact location, Punta del Sebo, is the marshland formed by the estuary of two rivers. Approximately $8 \times 10^6 \text{ m}^3$ of liquid effluents, containing around 0.4 Mt of phosphogypsum were annually discharged into the rivers. The gypsum deposit, on the banks of the River Tinto, reach a thickness of 4 to 6 m and their total surface covers about 425 ha. To date, 10 Mt of phosphogypsum have been deposited. Vegetation is totally absent from the pile. The ^{226}Ra content varies between 400 and 1000 Bq kg^{-1} . The liquid and atmospheric discharges are not radiologically evaluated here because these are present practices.

Before remediation, the measured ^{222}Rn emanation was $\sim 0.04 \text{ Bq m}^{-2} \text{ s}^{-1}$. This resulted in an exhalation dose, to the nearest inhabitant in Huelva, of 0.015 mSv a^{-1} . An average dose rate, for gamma exposure, of $0.3 \mu\text{Sv h}^{-1}$ was measured at a height of 1 m. After restoration (30 cm base, 1.2 m top soil, vegetation) ^{222}Rn exhalation rate varied between 0.005 and 0.026 on the crests (total 1.5 m soil) and between 0.005 and 0.002 in the flat areas (0.3 m soil). The gamma exposure was $0.1 \mu\text{Sv}$ or lower at both locations.

A2.3.9 Sweden

[Source: H. Ehdwall, Swedish Radiation Protection Institute (Personal Communication)].

There is a phosphorus treatment plant in Helsingborg. However, given the low uranium and thorium contents in the ore there appears to be no contamination problem.

A2.3.10 Economic data

[Source: Eurostat (1995)].

Table A2.3 P_2O_5 by EU countries for 1993

Country	P_2O_5 (1000 t)
Belgium/Luxembourg	318
Denmark	60
Germany	165
Greece	122
France	697
Ireland	-
Italy	252
Netherlands	256
Portugal	30
Spain	365
United Kingdom	81
EU-12: Production	2238

Adapted from Penfold *et al.* (1997).

A2.4 Coal mining and power production from coal

A2.4.1 Belgium

Dusar (1996) described the coal production in the North of Belgium. From 1906 and 1992, 440 million tonnes of coal were excavated in seven mining districts. There was also substantial coal mining in the South of Belgium. No radiological characterisation was carried out.

A2.4.2 Croatia

[Sources: Saler *et al.* (1996); and Subasic *et al.* (1996a,b)].

Saler *et al.* (1996) report the existence of several coal mines in Croatia. These are situated in the carbonate (krast) area of Dinarides. Exploitation of most coal mines was uneconomic and they have been shut down, with exception of the coal mine at Rasa, supplying the power plant Plomin (Istria). Considerable amounts of the radionuclides ^{238}U and ^{226}Ra have been identified, especially in the Rasa/Labin mines at Istria; also at Siritovci and at Bubravice. The boiler rooms of many large buildings, schools and hospitals operate using this coal.

There are two coal-fired power plants (one at the INA-VINYL PVC factory and the coal-fired power plant PLOMIN). Both facilities are still in operation, but one dumping site at INA-VINYL in Castle Sucurac is closed. This is discussed here. The 10000 m³ dump is situated 5 km north of Split (200000 inhabitants). Dumping of coal ash and slag was carried out between 1950 and 1970. The ^{238}U and ^{226}Ra activity in the coal-slag and ash are up to 18640 and 6200 Bq kg⁻¹, respectively. Some attention should be paid to domestic houses whose foundations were, in some cases, filled by contaminated material (some health problems of locally living people have been reported).

A2.4.3 Germany

[Sources: Henze and Weiss (1995); and NUKEM (1995)].

In the NUKEM report (1995), the radiological hazards related with lignite (brown coal burning) were considered because of the large quantities extracted in Germany (~400 Mt in 1983

decreasing to 200 Mt in 1993). However, given the low NORM content in lignite (between 1.5 and 26 Bq kg⁻¹ for ²³⁸U, 1.5 and 11 Bq kg⁻¹ for ²³²Th), and a low concentration factor in the ashes (between 2 and 5), lignite mining and combustion is of no radiological concern.

The hard coal mining activities are concentrated in the states of Sachsen, Thüringen and Sachsen-Anhalt. The most prominent are those carried out in the Freital area, where the ore is associated with uranium bearing ore strata. Hard coal in this area has been in use for about 400 years. It has been exploited on an industrial scale for the last 130 years, during which time, between 150 and 180 individual coal mines have been in operation. The hard coal was used as an energy source for glass and porcelain works, the steel industry and also as a source for domestic fuel. The ashes and slags were used by the residents as additives for building materials. Production was shut down in 1989.

After World War II, the Soviet-German stock company, Wismut, used the uranium-bearing hard coal for uranium production. Between 1955 and 1989, 3132 t of uranium was extracted from 3.5 Mt coal (0.09 wt %). The amounts of coal produced are not known.

The recorded NORM content in the Freital coal is 1.24 g ²³⁸U t⁻¹, 12.3 g ²³²Th t⁻¹ and 978 g Ra (t U)⁻¹. The calculated specific activities is 15000 Bq kg⁻¹ for ²³⁸U. The activity related to ²²⁶Ra in dead waste rock piles is between 400-4000 Bq kg⁻¹. In the ashes uranium and thorium are concentrated on average by a factor of 10, radium by a factor of 15.

As a result of the various mining activities, a lot of relics still exist in the Freital region. These may cause levels of radiation exposure to the public that are higher than those of natural radiation. The following types of objects have to be considered as sources of radiation exposure of the public:

- sites of abandoned uranium mills that had not been decontaminated but had been used for other industrial purposes;
- areas open to the public where mining residues and mill tailings have been dumped carelessly;
- areas, paths or roads covered by coal ashes or slags;
- waste rock piles, especially deposited slags and other combustion residues.

The investigations have shown that only a small part of the investigated area should be considered as being seriously contaminated. The external exposure to people living on radioactively contaminated ground was found to be the major pathway in the Freital region. All other exposure pathways, including exhalation of radon from mining relics and inhalation of radon and radon daughter products, can be neglected [Henze and Weiss, 1995]. The result of the first radiological assessment of mining relics in the Freital region have shown that from the total amount of 163 relics, 13 can be classified as "radiologically not relevant", 70 as "possibly radiologically relevant", 68 without classification (criteria not applicable) and 12 not classified (fault of measuring values).

Those relics classified as being "possibly radiologically relevant" were investigated with increasing profundity and these will be subjected to the final radiological assessment. Relics being finally assessed as "radiologically relevant" will be investigated in the framework for site-specific programmes to make a decision whether remedial actions are necessary or not. All liabilities in the Freital area are recorded in the ALASKA data base which is being established under the Federal Project (see also Annex A1.4.1).

A2.4.4 Greece

[Sources: C. Papastefanou (Personal Communication); and Manolopoulou and Papastefanou (1992)].

Coal mine and deposits are mainly situated in the North of Greece: in the Ptolemais region, in Ahlada and Vevi (Florina), in Serres and at Melivia and Kotili (Xanthi); one in the centre, Aliveri (Evia); and one in the South in Megalopolis (Peloponisos). There are in total 16 coal fired power plants. No indication is given whether any have already been abandoned. Manolopoulou and Papastefanou (1992) give some information on contamination levels in lignite and fly ashes from four regions (coal-fired power plants). Calculated averages are presented in Table A2.4.

Table A2.4 Average specific activities in lignite and fly ashes from mining districts in Greece

Radionuclides (Bq kg ⁻¹)	²³⁸ U	²²⁶ Ra	²¹⁰ Pb	²²⁸ Ra
Lignites range	117-399	44-206	59-205	9-41
average	243	133	134	18
Fly ash range	236-950	142-605	133-428	27-68
average	517	366	275	50

Adapted from Manolopoulou and Papastefanou (1992).

The average specific activities of the lignites are higher than the averages reported by Martin *et al.* (1996) for Europe, i.e. 30 (7-185) Bq kg⁻¹ ²³⁸U and 15 (3-22) Bq kg⁻¹ ²³²Th. Martin *et al.* (1996) and Scholten (1996) report a 10-fold concentration factor in fly ash, but, for Greek fly ash, only a concentration factor of 2-3 was reported. This is possibly due to the high ash content (28.8-41.6%). In addition, it is remarkable that the specific radioactivity of lignites (brown coal) and hard coal are comparable [Papastefanou and Charalambous, 1979].

A2.4.5 Poland

[Source: Piestrynski *et al.* (1996)].

In the Upper Silesian Coal Basin (USCB), deep regions of carboniferous coal formations contain brines with ²²⁶Ra concentrations reaching 400 kBq m⁻³. The problem in Poland is very pronounced since the USCB covers about 4500 km² and has 66 active coal mines. The mines pump out the saline waters, containing natural radioactive elements, and discharge them into surface waters. The saline waters (containing 200 kg m⁻³ salt) contain barium. Radium is precipitated from these brines. Near mine water discharge sites, as well as in active sediments of small rivers, up to 460 Bq kg⁻¹ ²²⁸Ra and 1430 Bq kg⁻¹ ²²⁶Ra are present. At least some mines are still operational, although this is not specified.

A2.4.6 Romania

[Source: Sandru (1996a,b)].

In Romania there are three old coal-fired power plants and seven modern plants. It is not clear which power plants are still in operation. The total ash production is 0.72 Mt a⁻¹ for the old stations; 12.2 Mt for the new ones. The average (and maximum) radionuclide contents of the ashes are: 80 (415) Bq kg⁻¹ ²³⁸U; 126 (557) Bq kg⁻¹ ²²⁶Ra; 210 (510) Bq kg⁻¹ ²¹⁰Pb; 262 (580) Bq kg⁻¹ ²¹⁰Po; and 62 (170) Bq kg⁻¹ ²³²Th. In Table A2.5 doses at old and new coal-fired power plants in Romania are presented.

Table A2.5 Doses at old and new coal fired power plants in Romania

Coal fired power plants		Internal dose due to inhalation manSv GW ⁻¹	External dose manSv GW ⁻¹
Modern	Romania	0.17 - 2.31	0.009 - 0.101
	World	0.26	0.015
Old	Romania	7.2 - 31.0	2.4 - 10.5
	World	1.35	0.3

Adapted from Sandru (1996a).

A2.4.7 Slovenia

[Source: Krizman (1996)].

In the old coal mining district of Kocevje, daily average outdoor radon levels were recorded of 80 Bq m⁻³ with hourly maxima of 150 Bq m⁻³. These high values were probably related with the large amounts (millions of cubic meters) of radioactive coal and coal ash (400-2000 Bq kg⁻¹ ²²⁶Ra) deposited on an area of 1.5 km² at the edge of the town. Indoor radon levels were up to some thousands of Bq m⁻³. Cancer statistics show the the highest incidence of lung cancer in Slovenia occurs in this town and the town of Idria, which is in the mercury mining district.

A2.4.8 Sweden

[Source: H. Ehdwall (Personal Communication)].

There are no coal mines in Sweden.

A2.5 Oil and gas drilling

A2.5.1 Croatia

[Sources: Saler *et al.* (1996); and Subasic *et al.* (1996a,b)].

There are a number of gas (>3) and oil fields (>5) being exploited but their production is low (2.5 Mt of oil per year). Almost all are situated in the Pannonian Basin (interior of the country). There are two oil refineries in Croatia, in Rijeka and Sisak (refinery capacities are 7-8 Mt for each). The scale formed during gas and oil drillings, which accumulates in the pipelines, results in a dose rate of 0.1 Sv h⁻¹ (i.e. 10⁷ Sv h⁻¹ higher than background) at the surface of pipelines.

A2.5.2 Germany

[Source: Penfold *et al.* (1997)].

Penfold *et al.* (1997) report the results of a German study (see Table A2.6). The median radium concentration in the German (Hannover) brines was 1.5 Bq L⁻¹ (4 mBq- 80 Bq L⁻¹). Oil field scales contain 59000 Bq kg⁻¹ ²²⁶Ra and 240000 Bq kg⁻¹ ²²⁸Ra. The recorded gas field scales contain 850-10⁶ Bq kg⁻¹ ²²⁶Ra and 5900-1.2 × 10⁶ Bq kg⁻¹ ²²⁸Ra. The specific activity of CaCO₃ scales is usually a factor 100 lower, but may contain ²²⁷Ac as major radionuclide (2500 Bq kg⁻¹). The radionuclide ²²⁶Ra dominates in most gas fields, whilst ²²⁸Ra dominates in most oil fields.

Table A2.6 Specific activities ($Bq\ kg^{-1}$) of some German oil and gas field scales and other precipitates

Origin	Oil field	Gas field	Gas field	Gas field	Gas field	Gas field
Physical form	Scale	Scale	Scale	Deposition	Scale	Scale
Chemical form	Ba/SrSO ₄ , PbS	Ba/SrSO ₄	Pb, Ba/SrSO ₄	SiO ₂ , PbS, Hg		CaCO ₃
²²⁶ Ra	59 000	350 000	160 000	7 400	1 000 000	850
²¹⁰ Pb	-	-	30 000	70 000	22 000	1 400
²²⁸ Ra	240 000	7 400	120 000	5 900	<10 000	2 500

A2.5.3 Italy

[Sources: Testa *et al.* (1994); Penfold *et al.* (1997)].

Penfold *et al.* (1997) report the results of an Italian survey [Testa *et al.*, 1994] on specific activities in scales present in Italian gaseous and liquid hydrocarbon plants. Table A2.7 is an adaption of these results. The main results show that concentrations of ²²⁶Ra are higher in oil extraction plants than in other plants (contrary to German case). It is remarkable that the radium activity in the scale is about two orders of magnitude lower than average. The radionuclides ²³⁸U and ²³²Th are either undetectable or low.

Table A2.7 Specific activity of some in scales present in Italian gaseous and liquid hydrocarbon plants ($Bq\ kg^{-1}$)

Plant	Extraction	Extraction	Extraction	Collection	Collection	Collection
Origin	Oil	Oil	Mixed	Gaseous	Gaseous	Oil
Depth (km)	6	5	5	-	-	-
²²⁶ Ra	2890 ±578	1126 ±225	120 ±24	30 ±6	<2.7	110 ±22
²³⁸ U	<0.9	<0.9	<0.9	23.8 ±4.3	53.8 ±10.8	11.3 ±2.3
²³² Th	<0.8	<0.8	<0.8	18.9 ±3.8	<0.8	<0.8

Adapted from Testa *et al.* (1994) and Penfold *et al.*, (1997).

A2.5.4 Netherlands

[Sources: Scholten (1996); and Eylander *et al.* (1997)].

Scholten (1996) briefly reports the waste production for a productive plant (NAM) for on-shore and off-shore gas and oil extraction. For gas extraction and production, 5 m³ a⁻¹ of waste are produced on-shore and eight times as much off-shore. Besides the sludge, 1 m³ of scales are produced. For an oil well, sludge production is 10 times greater than for a gas well.

Eylander *et al.* (1997) report a gas extraction unit in the northeast of the Netherlands where production started in 1975 (3000 m deep well, Zechstein carbonate) and had to be suspended in 1983. The final abandonment of the site was investigated in 1995-96. The NORM is not uniformly distributed inside the equipment and the presence of NORM with specific activities of up to 3.3 × 10⁶ Bq kg⁻¹ ²²⁶Ra and 1.8 × 10⁶ Bq kg⁻¹ ²¹⁰Pb were established. Radium isotopes were encountered in the scales and ²¹⁰Pb was encountered in deposits of metallic lead, galena and hydrocerussite. No indication was given concerning the amount of scale and sludge produced nor on their respective activities.

Trials were performed to demonstrate the technical feasibility of on-site decontamination of the equipment, using scale dissolver solutions (based on chelating capacity, no further specification was available). The cost was estimated at 350 000 Nfl, which is small compared to the cost of ~1.25 million Nfl, necessary for abandonment by conventional means (dismantling, transportation to licensed decontamination facility).

A2.5.5 Norway

[Source: Lysebo and Strand (1997)].

Lysebo and Strand (1997) give some data on activity levels in production water and deposits from operational platforms (taken during revision stop in the summer of 1995). The mean activity in the *production water* was 4.1 Bq L⁻¹ for ²²⁶Ra and 2.1 Bq L⁻¹ for ²²⁸Ra. The activity concentration of ²²⁶Ra in the *hard deposits* varied between 4000-39000 Bq kg⁻¹ (75% of the samples were above the exemption limit of 10000 Bq kg⁻¹, Table 4.1 and remarks). The ²²⁶Ra in the *porous deposits* varied between 300 and 24400 Bq kg⁻¹ (25% above 10000 Bq kg⁻¹). The overall mean ²²⁸Ra activity is 11300 Bq kg⁻¹.

The ²²⁶Ra activity in the *sludges* varied between 100 and 4700 Bq kg⁻¹ and is, therefore, below exemption limit. In the *sand*, the ²²⁶Ra activity varied between below detection limit and 21900 Bq kg⁻¹ (only 2 of the 26 samples had activities higher than 10000 Bq kg⁻¹).

The activity concentration of ²¹⁰Pb was low in all samples (maximum 700 Bq kg⁻¹).

The mean activity concentration of ²²⁶Ra in wastes from on-shore decontamination of tubulars was estimated at 25000 Bq kg⁻¹ (range 1000-100000 Bq kg⁻¹).

The total amount of wastes stored at different locations in Norway was 130 tonnes by the end of 1996.

A2.5.6 Economic data

[Source: Eurostat (1995)].

Table A2.8 Natural gas and oil production in EU countries for 1993

Country	Crude oil 1000 t	Natural gas 1000 TJ
Belgium	-	-
Denmark	8285	174.8
Germany	3066	633.4
Greece	562	4.4
France	2754	131.9
Ireland	-	100.2
Italy	4584	764.5
Luxembourg	-	-
Netherlands	3285	2914.6
Portugal	-	-
Spain	875	27.4
United Kingdom	95226	2524.5
EU-12: Production	118637	7257.8

A2.6 Rare earth and titanium oxide industries

A2.6.1 Rare Earths

A2.6.1.1 Estonia

[Source: Putnik *et al.* (1994)].

At Sillamäe, Estonia, a plant formerly used for the processing of uranium ore, switched at the beginning of 1970 to the processing of loparite (from the Kola peninsula). This mineral is rich in niobium, tantalum and other rare earths. In addition, loparite contains elevated concentrations of uranium (0.03%) and thorium (about 0.6%). In 1992, the plant was operating only at 15-20% of its capacity due to the shortage of raw material and chemicals for the process. The concentration in the presently produced waste are: 15000-26000 Bq kg⁻¹ ²³²Th, 1500-2200 Bq kg⁻¹ ²³⁸U and 1400-1800 Bq kg⁻¹ ²²⁶Ra. More information on Sillamäe can be found in Annex A2.1.

A2.6.1.2 France

[Source: ANDRA (1998)].

ORFLAM PLAST, in France, extracted from 1932-1987 cerium from monazite for the production of refractory bricks, resulting in several contaminated spots and 16 barrels containing ²³²Th. Rhône-Poulenc, also situated in France, separates and produces rare earths from monazite for the production of television screens, micro-computers and catalytic-converters for cars. The resulting historic radioactive contamination consists of 8 kt containing 1.15% ²³²Th (48 Bq g⁻¹) and 0.05% ²³⁸U (6 Bq g⁻¹). After 1994, the monazite was deactivated before import in France.

A2.6.2 Titanium oxide

Titanium ores are only mined to a small extent in Europe. Only 2000 t of the 771000 t annually imported. Of the total titanium usage of 790000 t material, 558000 t is consumed internally and the remainder is exported [Eurostat, 1995].

Penfold *et al.* (1997) report a Yugoslavian production plant. However, information on other European sites was not found.

A2.7 Zirconium and ceramics industries

Not much information is found on zirconium/zircon milling and processing facilities in Europe. Scholten (1996) briefly discusses a dry milling facility in the Netherlands and the Cookson Matthey group, with dry and wet mills, in England (HQ), Germany, Italy and Spain. In their discussion on radiological risks of non-nuclear industries, Pruppers *et al.* (1997) report two mineral sands and ~75 ceramic factories in the Netherlands alone.

A2.7.1 Economic data

According to Eurostat (1995), 136 t of zirconium was used in Europe in 1987. No zircon is mined or recovered in the EU. 350000 t zirconium ore is imported annually in Europe (Scholten, 1996).

A2.8 Wastes for building materials

No specific sites in Europe found.

A2.9 Applications of natural radionuclides radium and thorium

A2.1.9 Radium industry

A2.9.1.1 Belgium

[Source: Cottens *et al.* (1994).]

One of the world's largest radium extraction plants was operated, from 1922 until 1969, in Olen. The presence of a radium contamination on a dumping ground, owned by the plant and adjacent farmland, was already documented in the 1960s. In the early 1970s and mid-1980s more detailed inventories were made. Estimation of the exposure to the local population required complementary information.

The most important contaminated sites are:

- the D1-waste deposit, which is 10 ha of radium-containing waste and chemical waste products and 3 m depth. The mean dose rate is $2.8 \mu\text{Sv h}^{-1}$, with arange of $0.1\text{-}1000 \mu\text{Sv h}^{-1}$ (a homogeneous soil with a radium concentration of 1000Bq kg^{-1} , results in an additional dose of $0.49 \mu\text{Sv h}^{-1}$) [Deworm, 1988].
- the river Bankloop, of which the banks and the bottom sediments are contaminated with radium and heavy metals over a distance of 1400 m and a depth of 1 m. The dose rate is $0.5 (0.15\text{-}100) \mu\text{Sv h}^{-1}$.
- the 3 ha of farmland, where the River Bankloop enters the River Kleine Nete, contaminated through frequent inundations over a depth of 1 m. The dose rate is $0.5 (0.1\text{-}5) \mu\text{Sv h}^{-1}$.
- streets with spots of surface contamination, up to 30 cm deep. The dose rate is $0.5 (0.2\text{-}10) \mu\text{Sv h}^{-1}$.
- a residential building, with contaminated material under the veranda [$0.3 (0.1\text{-}0.6) \mu\text{Sv h}^{-1}$] and, an adjacent farmland with radium contamination at between 0.5 and 2m ($0.1 \mu\text{Sv h}^{-1}$).

Radon exposure in a house near the D1 disposal site results in a dose rate of 4mSv a^{-1} .

The action level for remediation is 0.2 and $0.4 \mu\text{Sv h}^{-1}$, within and below the first metert respectively. A covering with 1 m soil is proposed.

The external exposure from the banks of the River Bankloop and streets are some $100 \mu\text{Sv a}^{-1}$ but this figure depends on exposure time and level. The action levels are $0.15 \mu\text{Sv h}^{-1}$ for the Bankloop and $0.2 \mu\text{Sv h}^{-1}$ for the streets.

The internal exposure due to the consumption of contaminated food is estimated at a few tens of $\mu\text{Sv a}^{-1}$. Removing the contaminated soil at the mouth of the River Bankloop is presently considered unnecessary.

The radon exposure in the house with radium containing material under the veranda is estimated at 5.1mSv a^{-1} (for some years, radon was pumped away). The soil under the house and on the adjacent land has to be removed to reduce radon to background levels.

A2.9.1.2 France

[Sources: Andra (1995, 1996, 1998)].

ANDRA executed a radiological survey in 1993 in Dieppe, close to a watch factory. The highest gamma dose rate recorded was 10mSv h^{-1} , corresponding to ^{226}Ra concentrations of more than 1MBq kg^{-1} . About 60m^3 soil was excavated with a mean radium concentration of

330 kBq kg⁻¹. This preliminary intervention was followed by a more thorough remediation based on following criteria: a 2000 Bq intervention level; rest activities (and dose rate) in buildings and surroundings of 1000 Bq kg⁻¹ (0.2 μSv h⁻¹) and 5000 Bq kg⁻¹ (1 μSv h⁻¹), respectively. Some 1000 m³ of waste was conditioned, with mean ²²⁶Ra concentrations of 29000 Bq kg⁻¹.

The total cost of the remediation was 9.3 million French Francs, excluding burial of waste. Given a remaining radium activity of 5000 Bq kg⁻¹ the site is not open for the public.

Andra (1998) synthesised the different categories of radium liabilities in France. Six sites were linked with radium production. At Nogent-sur-Marne, the expected radium production was 15 g. Not much information on these potentially contaminated sites is provided. Furthermore, Andra (1998) reports more than 50 places where radium was used for medical purposes. Between 1983 and 1991, 27 g radium was collected corresponding to more than 1000 needles and tubes. Finally, the watch industry has used radium-containing paints and, by the end of 1998, five more liabilities (excluding the one in Dieppe) were identified. No information on the extent of contamination or contamination levels is given.

A2.9.1.3 United Kingdom

East London

[Source: Drury (1994)].

A 130000 m² area of land in east London was contaminated due to different industrial activities over the past 90 years. The chemical works, which was located on this land, comprised of: 1) the extraction and processing of thorium and rare earths from monazite sands; 2) a radium luminising facility; and, 3) the production of organic and inorganic chemical compounds. As a result of the operations under 1 and 2, radioactive wastes were produced and buried at the site to depths of 6 m. This was adjacent to a school, a factory and residential premises. The plant was demolished in 1970. Before clearance for new industrial activities, the site had to be remediated. Therefore, 200000 m³ of contaminated material had to be removed, of which, ~4000 m³ was radioactive with activities between 400 and 4 × 10⁵ Bq kg⁻¹ for thorium (2500 m³) and between 400 and 3.2 × 10⁵ Bq kg⁻¹ for radium (1200 m³). In addition, hot spots of 4 MBq kg⁻¹ were identified. Surface dose rates varied between background (0.1 μSv h⁻¹) and 100 μSv h⁻¹ and discrete sources of radium were detected, total 2 GBq.

The result of an assessment study showed that direct external exposure would be the critical pathway. The end remediation criteria were: 40 Bq kg⁻¹ thorium and 80 Bq kg⁻¹ radium within the top first metre, and 300 Bq kg⁻¹ thorium and 3000 Bq kg⁻¹ radium and below this. Remediation comprised in essence the removal of the contaminated material and the area was levelled and covered with 0.7 m soil. The disposal of the waste following criteria for ²²⁶Ra: <370 Bq kg⁻¹ *de minimis*, 370-49000 Bq kg⁻¹ exempt waste: special local waste dump, 49000 Bq kg⁻¹-4 MBq kg⁻¹ low level waste (Drigg site) and >4 MBq kg⁻¹ medium level waste, brought to AEAT. Specific information of the remediation actions can be found in Drury (1994).

Dalgety Bay

[Sources: Wilson (1990); Milton (1991); Tilly (1991, 1992a,b, 1993a,b, 1994)].

Dalgety Bay is a shallow inlet on the north coast of the River Forth. During World War II and until about 1955, the area to the north and the east of the Bay was a military airfield. During 1946, a large number of aircraft were dismantled and an aircraft salvage section was operated. Unsalvaged material was probably burnt and the resulting clinker and other residues from incineration were used as in-fill for tips or quarries. Anecdotal evidence indicates that the western headland in front of the Sailing's Club has been built up with clinker and ash and later removed during excavations for the club house. The excavated material was relocated along

the shore at Sealstrand and the general area has more recently been used for residential purposes.

In 1990 a monitoring team from the Rosyth Naval Dockyard found that the bay was contaminated with ^{226}Ra , probably from the luminous paint used on articles which were dismantled at the salvage site. Since the discovery, there have been a number of surveys of the area.

RAF Carlisle

[Source: Gibbs (1994)].

The ^{226}Ra contamination at the Site 5, RAF Carlisle, Cumbria, originated from pre- and post-war luminising equipment buried in the ground. For security reasons, redundant material was routinely incinerated and it is believed that ashes were raked out and buried over de green field site in the 1950s. The incinerator and its immediate surroundings cover an area of $\sim 2000\text{ m}^2$.

Ninety-two individual hot spots were identified. The highest contact dose rate was $4\ \mu\text{Sv h}^{-1}$, with a dose rate of $0.8\ \mu\text{Sv h}^{-1}$ at 1 m above surface. On average contact dose rates above hot spots were $0.2\text{-}3\ \mu\text{Sv h}^{-1}$ with a dose rate of $0.2\text{-}0.6\ \mu\text{Sv h}^{-1}$ at 1 m above surface. Burnt luminous dials contained 6 to 880 kBq of ^{226}Ra .

Readings with a scintillation detector at 1 cm above surface of 1 counts per second above background corresponded with an activity concentration of 0.37 Bq g^{-1} (clearance level for ^{226}Ra). If the reading exceeded 1 counts per second, the soil was excavated. In total, 140 m^3 of ^{226}Ra contaminated earth, ash and other items were removed containing 350 MBq ^{226}Ra . The material was transferred to standard 200 L drums. On completion of the clearance survey, the Site 5, RAF Carlisle was made available for unrestricted use.

A2.9.2 Thorium industry

Penfold *et al.* (1997) report a 'UK-supplier' of thorium alloys as hardener for magnesium alloys which ceased operation 10 year ago. Except for the industrial site in East London, described above, no other information was found about specific workshops for thorium applications. According to a German study reported in Penfold *et al.* (1997), the two European producers of thorium oxide are located in Austria (Treibacher) and France (Rhône-Poulenc in La Rochelle).

According to Andra (1998), thorium was produced by Rhône-Poulenc, in Rochelle, and by SMIF in Clichy. Until 1994, Rhône-Poulenc separated and produced rare earths and thorium directly from monazite for the production of TV-screens, micro-computers, HIFI and catalytic converters for cars. From 1994 onwards deactivated monazite was imported. The 'historic' residue from the older process consists of 8025 t (50% moisture) containing 1.5% ^{232}Th and 0.05% ^{238}U . The average activity concentrations are 48000 Bq kg^{-1} ^{232}Th and 6000 Bq kg^{-1} for ^{238}U . The total activity is 217 GBq. At Clichy, thorium was extracted for the production of incandescent gas mantles. At present, the site is occupied by another industry. Two hot spots are situated on the site.

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ANNEX B1
AN OVERVIEW OF COMPUTER-BASED MODELLING
APPROACHES FOR RADIOACTIVE WASTE SITES

Table of Contents

B1.1	Introduction	134
B1.2	Computer-based assessment models	135
B1.3	General description	142
B1.4	Atmospheric migration	142
B1.5	Groundwater migration	143
B1.6	Biosphere processes	145
B1.7	Dose calculations	147
B1.8	Probabilistic modelling	147
B1.9	Conclusions	148
B1.10	References	151

B1.1 Introduction

In the context of developing a decision-aiding framework for remediation strategies for contaminated sites, it is necessary to determine both the current site impact and the comparative effects of differing remediation options. For convenience, in the initial stage of an investigation, assessments may be undertaken using conceptual or mathematical models of the key processes governing impact pathways. A detailed description of a comparative assessment tool developed specifically for the CARE project, AMCARE (Assessment Model for developing a Common Approach to REstoration of contaminated sites), is presented in Annex B2. However, there are a number of approaches to modelling the impact of contaminated areas and waste disposal sites, each of which may be equally valid and which may have specific advantages for different types of site. Indeed, if models are defined broadly as ‘any device that represents an approximation of a field situation’ [Anderson and Woessner, 1992] then laboratory simulations of particular processes may also be included. This review is limited to the following computer-based assessment models, developed to deal with situations similar to those encountered within the CARE example sites:

- ECOSR (Environmental Contamination from Surface Repositories);
- GEOS/ABRICOT;
- IMPACT (Integrated Model for the Probabilistic Assessment of Containment Transport);
- INTAKE;
- JAERI (Japan Atomic Energy Research Institute Model);
- MEPAS (Multimedia Environmental Pollutant Assessment System);
- RESRAD (Residual Radioactive Material Model);
- SACO (Safety Assessment Comparison);
- SONS (State Office for Nuclear Safety Model).

In each case the models address radiological assessments of sites and determine the impact of the radioactive waste inventory on the population. The following factors are common to all models:

- the composition of the waste;
- the location of the waste;
- the nature of the surrounding environment;
- the mechanisms and pathways through which radionuclides from the waste are released into the environment;
- the off-site locations where migrating radionuclides may accumulate to form secondary sources of contamination;
- uptake into the food-chain;
- the habits of the population.

Clearly, many of these factors will be specific to individual sites. However, the underlying approach to quantifying these factors is often common to a number of sites. Key points of commonality found by this review are that generally:

1. waste is treated as a point source;
2. the migration of radionuclides through atmosphere is modelled assuming a Gaussian plume dispersion;
3. groundwater is modelled using a one-dimensional advection-dispersion equation;
4. the biosphere incorporates compartments to describe the distribution of radionuclides in soil, livestock, vegetables and aquatic foods and is modelled by assuming that transfers within the system are in equilibrium.

In all cases, standard dose conversion factors are assumed. In most cases, radioactive decay and in-growth of daughters are included. This is considered essential where the time periods of interest are relatively long by comparison to the physical processes involved.

B1.2 Computer-based assessment models

The function of computer-based models falls broadly into three categories:

Predictive: Where the model is used to predict the future consequences of a proposed action.

Interpretative: Where the model is used to gain an insight into the controlling parameters in a site-specific setting and/or as a framework in assembling and organising field data.

Generic: Where the model is broadly applicable to analysing processes for a range of sites. The predictive capability of this type of model makes it a useful tool in framing regulatory guidelines for a specific region or for screening the suitability of a region for a proposed action.

Mathematical models may be solved analytically or numerically. In general, analytical solutions are appropriate to situations where empirical data or assumptions are applied to solve what would otherwise be a complex mathematical problem. Numeric solutions solve these problems directly by a mathematical procedure.

The dose assessment models reviewed here were chosen because they have been involved in comparative trials and applied to calculate doses arising from a uranium tailings pile [BIOMOVS II.4, 1995; BIOMOVS II.5, 1996].

- BIOMOVS II.4 describes a 1000 year atmospheric release with end points in an agricultural and a forested area. In addition, there is a 1000 year release of radionuclides to groundwater, which migrates to a small river and, in turn, flows into a lake. The groundwater is abstracted through a well. The lake contains fish which are consumed locally. Leafy vegetables are grown on agricultural land which is irrigated with lake water. Cattle graze on local pasture which is not irrigated. Both well water and lake water are used for domestic consumption. Data such as radionuclide fluxes, transfer factors and human and animal habits are given. Deterministic calculations of the doses are undertaken.
- BIOMOVS II.5 describes the 10000 year atmospheric release of radionuclides to an agricultural area and a 10000 year release of leachate into groundwater. The groundwater is abstracted through a well which is used for irrigation and domestic purposes. The agricultural land is used for growing leafy vegetables which are irrigated. Cattle graze on local pasture which is not irrigated. Data such as radionuclide fluxes, transfer factors and human and animal habits are provided. Both deterministic and probabilistic calculations are undertaken.

These scenarios provide a degree of overlap with those encountered within the CARE example sites. Therefore, it is possible to make direct comparisons of the structure of these models and to examine their approaches to defining features, events and processes which influence the calculated doses. Such comparisons may identify the important features of a model which can be applied to the CARE example sites.

The models reviewed are listed in Table B1.1. These are all mathematically-based predictive generic models which have been applied to the one or both of the scenarios described as BIOMOVS II.4 or BIOMOVS II.5.

Table B1.1 Models reviewed

Model	Development Organisation	Reference
ECOSR	Institute of Public Health and Medical Research, Romania	Tora (1993)
GEOS/ABRICOT	Institut de Protection et du Sûreté Nucleaire, Commissariat à l'Energie Atomique (CEA), France	Santucci (1995) Ferry (1995)
IMPACT	Developed jointly by Beak Consultants Ltd, Canada, and the Atomic Energy Control Board (AECB), Canada	Beak Consultants (1995)
INTAKE	SENES Consultants Ltd, Canada	SENES Consultants (1985, 1986, 1987)
JAERI	Japan Atomic Energy Research Institute	Sakamoto and Tanaka (1990)
MEPAS	US Department of Energy's Pacific Northwest National Laboratories (PNNL), United States of America	Droppo and Buck (1996), Streile (1996), Strenge and Chamberlain (1995), Wheelan and McDonald (1996).
RESRAD	US Department of Energy's Argonne National Laboratory, United States of America	Yu (1993)
SACO	Jointly by Intera Information Technologies and IMA/CIEMAT	Bergström (1982), IMA/CIEMAT (1993) Robles <i>et al.</i> (1993), Yuan (1993)
SONS	State Office for Nuclear Safety, Czech Republic	BIOMOVS II (1995) and BIOMOVS II (1996)

It is important to recognise that models are, often, updated continually and that the descriptions given below may have been superseded by later versions of the models.

B1.2.1 ECOSR

ECOSR is a time-dependent, multi-compartmental model, developed for BIOMOVS II as an exercise for research purposes only. Groundwater and atmospheric dispersions are treated separately.

B1.2.2 GEOS/ABRICOT

The GEOS/ABRICOT model employs two codes. The GEOS V2.0 code is used to calculate the transfer of radionuclides through the aquifer, and the ABRICOT code to calculate the transfers into the biosphere (see Figure B1.1). Inputs from atmospheric release are calculated separately.

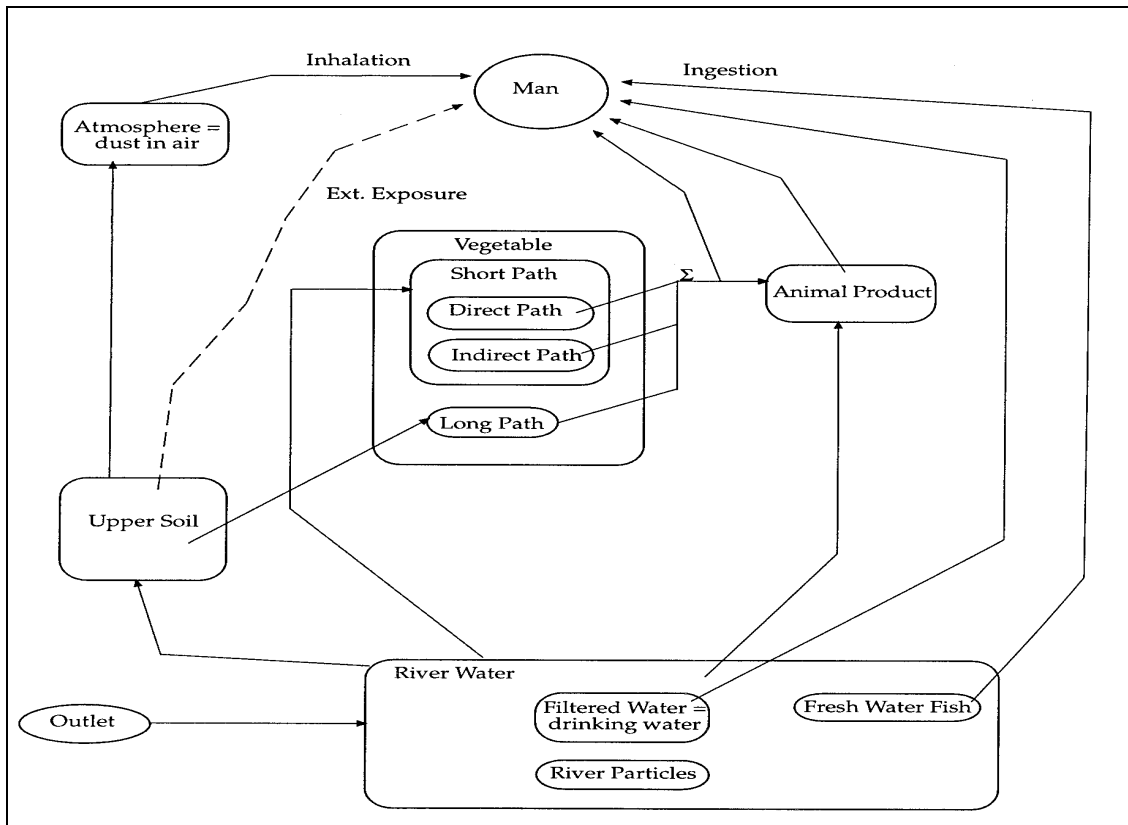


Figure B1.1 ABRICOT v.2.0 compartments and pathways

B1.2.3 IMPACT

The IMPACT model simulates multi-media radionuclide fate and transport, calculates radionuclide uptake and transfer through the food chain, and estimates dose and risk to man and other biota. It accepts time-varying contaminant fluxes to air, water, groundwater, soil or sediment at multiple locations, and predicts exposure concentrations, dose and risk to multiple receptors at specified locations.

B1.2.4 INTAKE

The INTAKE model used by SENES was derived from components of the environmental pathways portion of the UTAP (Uranium Tailings Assessment Program) model. The original model was developed by SENES for the National Uranium Tailings Program (NUPT), in Canada, to assess environmental transfer, food uptake and the risks associated with a typical reference tailings sites. The transfer and exposure pathways are summarised in Figure B1.2.

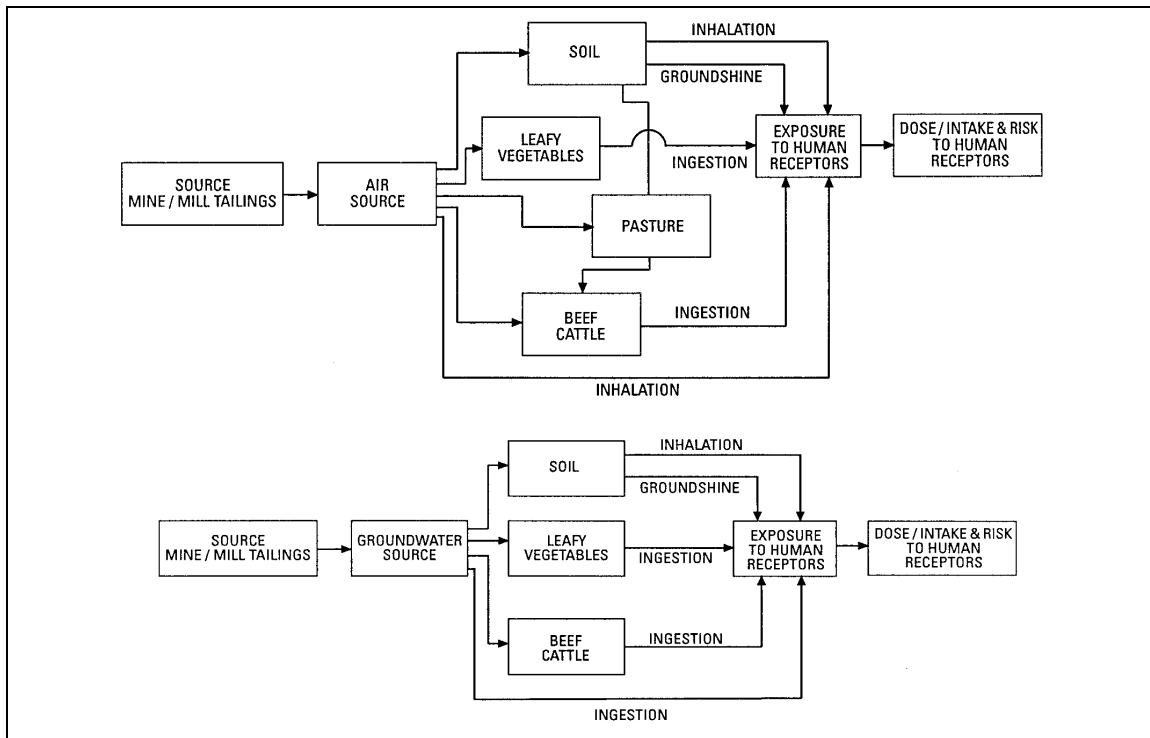


Figure B1.2 Atmospheric and groundwater source exposure pathways in INTAKE

B1.2.5 JAERI

JAERI is a single computer code which deals with exposure pathways arising from both atmospheric and groundwater release. The model can be applied to exposures arising from both terrestrial systems and to aquatic systems. The atmospheric and groundwater pathways are shown in Figure B1.3.

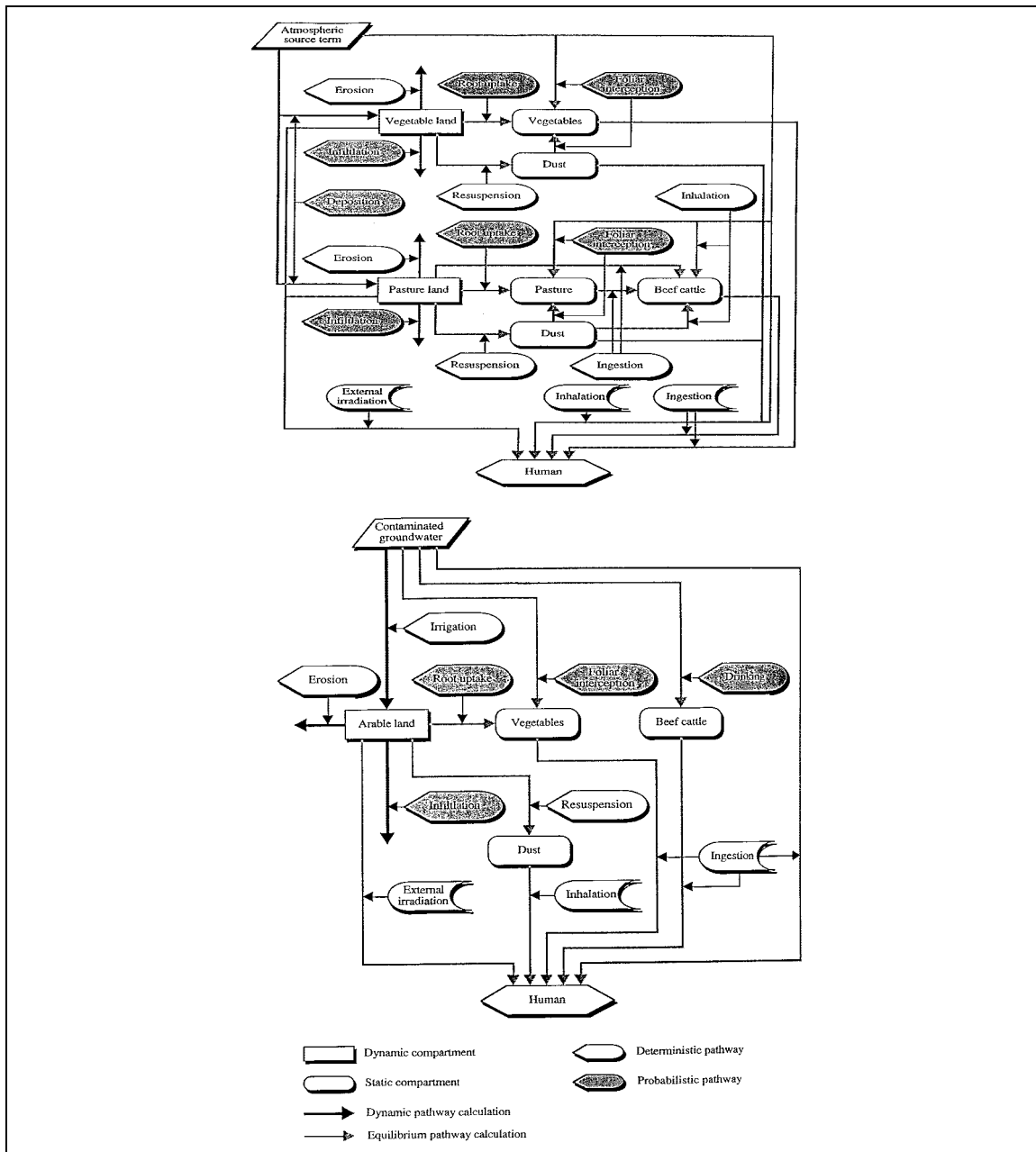


Figure B1.3 Atmospheric and groundwater exposure pathways in the JAERI model

B1.2.6 MEPAS

MEPAS is a risk assessment computational tool that evaluates impacts to exposed individuals and to surrounding populations. It is an integrated system of analytical, semi-analytical, and empirically based mathematical models which simulate source-term release rates, transport processes, exposure and uptake and human-health effects.

B1.2.7 RESRAD

RESRAD is a computer code developed to calculate site-specific radioactive materials guidelines and radiological dose/risk to an individual. The transfer pathways used by RESRAD are shown in Figure B1.4.

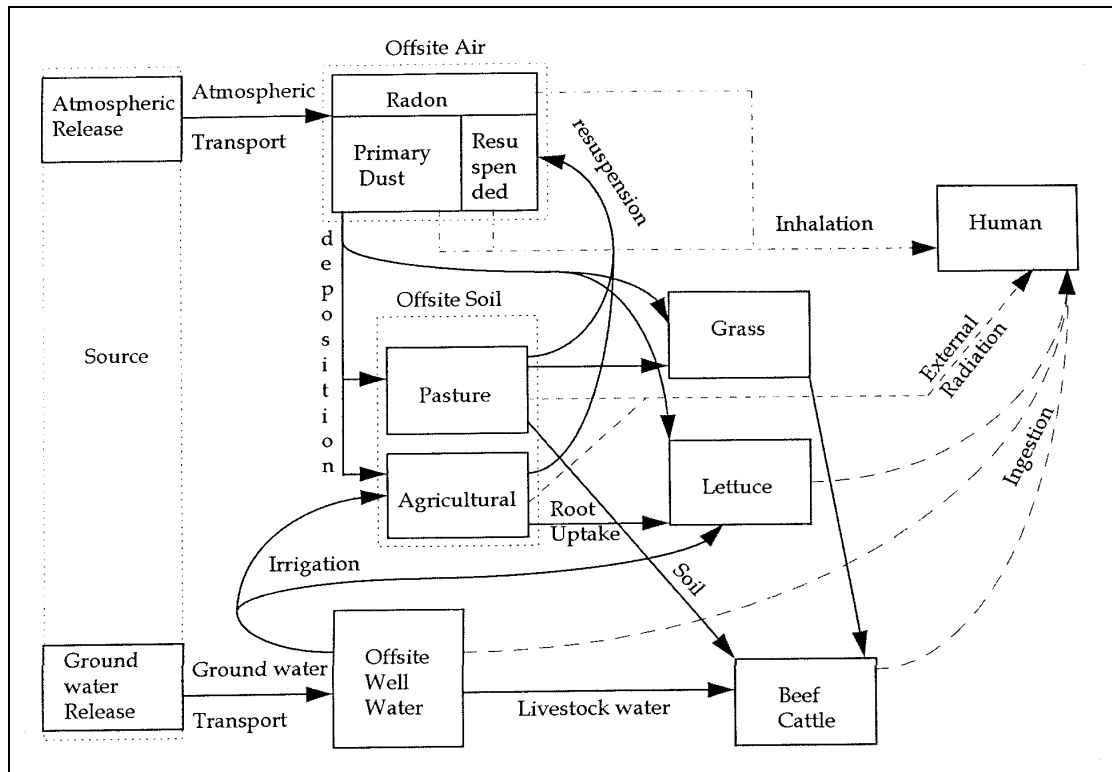


Figure B1.4 Contaminant transfer and exposure pathways used by RESRAD

B1.2.8 SACO

The SACO model has a modular structure involving a set of codes for various components of a system and it is designed to model environmental impacts from all types of wastes (toxic, radioactive and mixed wastes). SACO can deal with both shallow and deep disposal facilities and deal with varying levels of data availability. The atmospheric and groundwater pathways for the SACO model are shown in Figure B1.5.

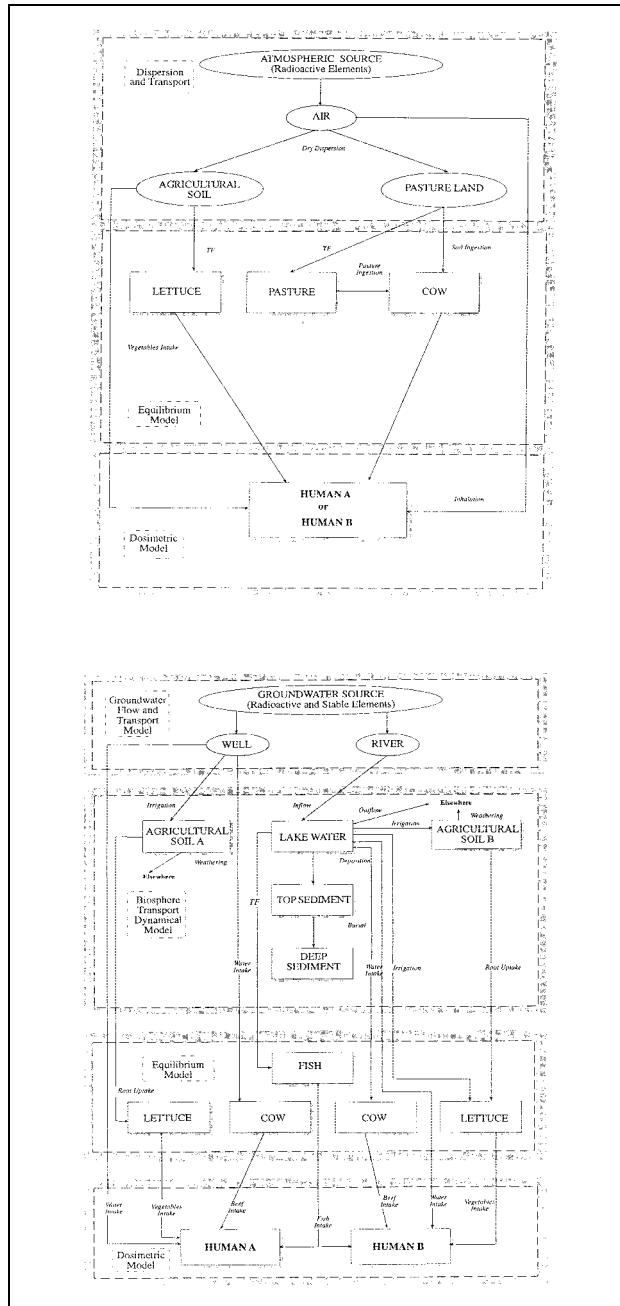


Figure B1.5 SACO conceptual model for atmospheric and groundwater release

B1.2.9 SONS

The SONS model employs three separate computer codes to model atmospheric dispersion, groundwater contamination and contamination of foodstuff. The exposure pathways through the food chain are shown in Figure B1.6.

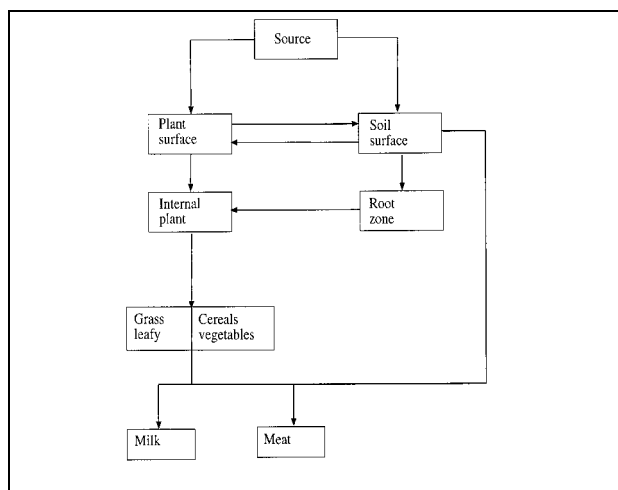


Figure B1.6 Flow chart of the terrestrial food chain in the SONS model

B1.3 General description

The models reviewed were chosen because they had been shown to be applicable to similar sites to those encountered at the CARE example sites. The basic scenario for these sites is summarised in Figure B1.7.

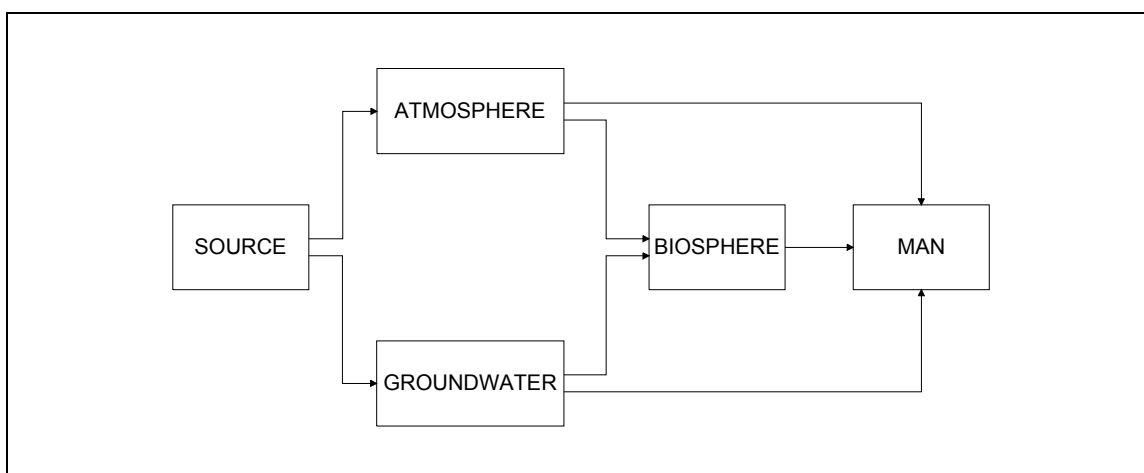


Figure B1.7 General description of exposure pathways for contaminated sites

Given that the basic scenario is similar for all the assessment models considered here, those 'key factors' or approaches identified as being in common to several of the methods may also be appropriate to assess the CARE sites.

B1.4 Atmospheric migration

Release into the atmosphere, and the subsequent distribution of radioactive gases and dust particles, presents a major pathway through which radionuclides can migrate from the primary source to secondary locations (i.e. the biosphere). Quantifying this pathway depends on a number of site-specific factors. These include the geometry of the waste, its composition, topography of the surrounding area and local meteorological parameters (wind velocity, wind direction, turbulence and rainfall). The degree to which these factors can be defined will influence the accuracy of the calculation.

All of the models reviewed treat the radioactive waste as a point source (although the SONS model incorporates a correction factor to allow for the source dimensions). This suggests a

consensus view that this, admittedly simple approach to defining the waste source, provides an adequate representation of the situation. Similarly, most of the models calculate atmospheric dispersion using a Gaussian plume approach. This involves the division of the area around the source into sectors to allow for differing wind direction frequencies between the sectors. Changing weather conditions are characterised by 'stability categories'. Again, it may be inferred that the consensus view is that the Gaussian plume provides a 'fit-for-purpose' model of atmospheric dispersion.

Most models incorporate terms to correct for dry deposition from the plume and to allow for plume depletion (although the RESRAD model conservatively discounts the effects of plume depletion). Only the SONS model allows for wet deposition from the plume, although this may be an important feature for a number of nuclides. The IMPACT and SONS models also incorporate corrections for radioactive decay and in-growth within the plume, which are clearly of importance where the transit time is long by comparison to the half-lives of the nuclides of concern. In addition, the MEPAS model allows the modelling of different particle sizes, although this level of detailed information is not likely to be available in the preliminary stages of an investigation.

B1.4.1 Discussion of atmospheric migration approaches

Overall, BIOMOV5 II.5 (1996) shows that there is good agreement between the results obtained with these models. Treating the atmospheric distribution by assuming a Gaussian plume dispersion from a point source appears to provide an adequate method for describing the migration of radon and dust emissions to locations off-site. Factors such as radioactive decay and in-growth of daughter nuclides in the plume should be incorporated. In addition, dry deposition would also be an important factor when calculating the migration of dust.

B1.5 Groundwater migration

Migration of radionuclides through the movement of groundwater provides a major route through which radionuclides are transported to locations off-site. The process may be simplified to two basic considerations: leaching of radionuclides from the primary location (the waste) and transportation to a secondary location (the biosphere). Both are considered below.

B1.5.1 Leaching from waste

The rate of leaching from a primary source is normally based on a series of crude assumptions which can give rise to serious uncertainties. However, this reflects the fact that waste sources are themselves often poorly defined in terms of contact geometry, composition and infiltration of water. It is common to assume a simple geometry and to further assume that the source is homogenous. An average leaching coefficient for each radionuclide is generally assumed to be appropriate and these are either obtained empirically for the site, or general values from the literature are used for the waste/soil type in question. For any simple approach, it is also necessary to assume that the radionuclide content in the infiltrating water rapidly equilibrates with the radionuclide content in the solid waste. This forms the basis of the approaches used by MEPAS and SONS.

B1.5.2 Groundwater transport

The migration of the radionuclides in groundwater is controlled by two processes: advection (movement due to groundwater flow) and dispersion (movement due to mechanical mixing and molecular diffusion). This migration can be described mathematically by an advection-dispersion equation. Equations may be derived to describe one-, two- or three dimensional flow scenarios [Freeze and Cherry, 1979]. Clearly, an equation which describes three-dimensional

flow to take account of lateral and vertical migration along with longitudinal migration will provide a more rigorous treatment of the situation. However, most of the models listed above describe the migration of radionuclides in terms of the one-dimensional expression. This suggests that, on the basis of the most common (or ‘consensus’) approach, the one-dimensional expression is considered adequate for the purposes of calculating the migration of radionuclides to a specified location.

Exceptions to this include RESRAD, which takes account of lateral flow (i.e. a two-dimensional model). The ECOSR model also takes account of lateral dispersion but by an indirect method. In this case a one-dimensional lateral flow is assumed, which has a flow velocity 1000 times lower than the longitudinal flow.

Most models assume that retention of the radionuclides, relative to that of the groundwater, can be represented by linear, reversible sorption processes. Quantification of this retention factor is, in most cases, based on the effective porosity of the rock, its density and the radionuclide specific distribution coefficient (K_d). This may be defined as follows:

$$K_d = \frac{C(X)_{\text{solid}}}{C(X)_{\text{gw}}}$$

where: K_d is the distribution coefficient for radionuclide X ($\text{m}^3 \text{kg}^{-1}$);
 $C(X)_{\text{solid}}$ is the concentration of radionuclide X in the solid phase (Bq kg^{-1}); and
 $C(X)_{\text{gw}}$ is the concentration of radionuclide X in the groundwater (Bq m^{-3}).

All the models reviewed assume that migration of radionuclides through the groundwater is a non-conservative process and that radioactive decay and in-growth need to be taken into account. However, a number of approaches are employed for this purpose. Most use a numeric method, assuming transfer through a series of compartments between the source and the eventual destination. GEOS/ABRICOT, JAERI and MEPAS models, however, utilise analytic methods with the numerical method to evaluate convolution integrals over time. RESRAD uses an analytic solution only.

The GEOS/ABRICOT model deals with the in-growth of daughter radionuclides indirectly, as its solution to the transport equation can only account for the radioactive decay of the parent radionuclide in a homogenous medium. Within the model, this limitation is overcome by dividing the path through the aquifer into a series of intermediate compartments. The amount of daughter produced during the passage through a compartment is then estimated and this estimate is applied as an input for the following compartment.

The JAERI model solves the transport equation by dividing the aquifer into three regions: a region upstream of the source (Region 1); a region between the source and the point of release into the biosphere (Region 2); and a region downstream of the point of release (Region 3). A condition of zero flow is assumed above Region 1 and a condition of zero gradient is assumed downstream of Region 3. The migration lengths of Regions 1 and 3 are chosen such that these conditions have no effect on the output values at the point of release.

Most models assume that the migration is from a point source. The MEPAS model however, assumes a source of defined dimensions.

B1.5.3 Discussion of groundwater migration approaches

The BIOMOVs studies show good agreement in the results obtained by each model for this pathway and most of the models all base their calculations of groundwater migration on a one-dimensional advection-diffusion equation. It is general practice, and endorsed here, that radionuclide decay and in-growth of daughter nuclides should be taken into account.

B1.6 Biosphere processes

The biosphere represents the environment in which biological organisms (plant or animal) are exposed to radiological contamination. The uptake of radionuclides by these organisms and, consequently, their incorporation into the food-chain, allows radionuclides to become distributed throughout the biosphere. This distribution can be treated in terms of a steady-state model (e.g. ECOSR), or as a linear dynamic model (e.g. JAERI and SONS). The differential equations generated by the latter approach are solved by the Runge-Kutta method.

The biosphere can be divided broadly into a number of sub-categories, into which inputs from the atmosphere and groundwater migration of radionuclides are made. The division of these sub-categories tends to vary between the models and is discussed below.

B1.6.1 Radionuclides in soil

The input of radionuclides into a soil compartment can be from the migration of radionuclides through groundwater, deposition of atmospheric releases and through irrigation with either well water or river water. Radionuclide input from groundwater is normally calculated on the basis of defining a compartmental volume into which the radionuclides migrate at a rate defined by the groundwater flow. Atmospheric deposition is normally a function of the atmospheric concentration of each radionuclide and the rate of particle deposition. Irrigation is defined with respect to human practice (frequency and volume) and source (well or river). All of the models listed take these factors into consideration.

The loss of radionuclides from the soil can include leaching, radioactive decay (and in-growth) soil erosion and the re-suspension of dust particles. Most of the models examined include pathways to take account of these terms, although most are based on generic data rather than site-specific information. Exceptions include JAERI which does not model radionuclide in-growth and MEPAS which omits erosion.

In some cases the soil compartment is sub-divided to allow for different inputs. For example, the SONS model uses a soil surface compartment and a root zone compartment. The soil surface compartment allows for the input of irrigation water and the deposition of atmospheric material. The root zone compartment will only be affected by these factors through interaction with the soil surface. The SACO model also sub-divides the soil compartment to allow for separate irrigation practices involving either river water or well-water.

B1.6.2 Radionuclides in vegetation

The incorporation of radionuclides into vegetation can be the result of direct stomatal uptake of gases, foliar interception of contaminated atmospheric particulates, foliar interception of irrigation water, and root uptake of radionuclides from the soil. Most of the models considered tend to assume that radionuclides deposited on the external surface of vegetables do not interact with those taken up through the roots. The SONS model is an exception to this.

Foliar interception (and direct stomatal uptake) is time dependent. Consequently, uptake of radionuclides incorporated in particles depends on the rate of deposition of the particles and uptake from water is a function of the irrigation practices and the composition of the water. All models include a loss term to take account of the rate of loss due to weathering.

Root uptake by plants from soil is assumed by all models to be equilibrium based and a set of generic soil to plant concentration factors is used to calculate the distribution of the radionuclides in the plant. Therefore, the plant content is taken to be a simple function of the radionuclide content of the soil. The transfer factor to describe this may be defined as follows:

$$TF_{\text{soil/plant}} = \frac{C(X)_{\text{plant}}}{C(X)_{\text{soil}}}$$

where: $TF_{\text{soil/plant}}$ is the transfer coefficient for radionuclide X from soil to plant (Bq kg⁻¹ plant/Bq kg⁻¹ soil);
 $C(X)_{\text{plant}}$ is the concentration of radionuclide X in the plant (Bq kg⁻¹ plant); and
 $C(X)_{\text{soil}}$ is the concentration of radionuclide X in the soil (Bq kg⁻¹ soil).

B1.6.3 Radionuclides in livestock

The uptake of radionuclides by livestock derives principally from ingestion of contaminated vegetable matter, water and soil. All of the models take these pathways into account. Additional pathways are considered by some of the models. For example, dust inhalation is considered within IMPACT, JAERI and INTAKE. The IMPACT model also includes a term for radon inhalation. The radionuclide content in animal foodstuffs (meat or milk) is calculated as an equilibrium term, which is a function of daily total radionuclide intake by the animal. The transfer factor to describe this may be defined as follows:

$$TF_{\text{fodder/livestock}} = \frac{C(X)_{\text{livestock}}}{C(X)_{\text{fodder}} \text{ CON}_{\text{fodder}}}$$

where: $Tf_{\text{fodder/livestock}}$ is the transfer coefficient for radionuclide X from a given source to livestock (Bq kg⁻¹ /Bq kg⁻¹ fodder per kg d⁻¹ fodder intake);
 $C(X)_{\text{livestock}}$ is the concentration of radionuclide X in the livestock (Bq kg⁻¹ livestock);
 $C(X)_{\text{fodder}}$ is the concentration of radionuclide X in the fodder (Bq kg⁻¹ fodder);
and
CON is the daily fodder intake by livestock (kg d⁻¹).

B1.6.4 Radionuclides in well water

All models take the radionuclide content of the well water to be the same as that of the local groundwater.

B1.6.5 Radionuclides in surface water

All the models examined treat the input of radionuclides into a surface water system in a similar manner. The radionuclide content of the groundwater at the point of entry into the surface water is calculated and then a dilution factor applied to take account of the volume of the surface water. Transference into bottom aquatic sediment is calculated from the concentration in the surface water and assuming steady state mixing between the sediment and water. Generically derived distribution coefficients are used. These are defined as follows:

$$K_d = \frac{C(X)_{\text{sediment}}}{C(X)_{\text{aq}}}$$

where: K_d is the distribution coefficient for radionuclide X (m³ kg⁻¹);
 $C(X)_{\text{sediment}}$ is the concentration of radionuclide X in the aquatic sediment (Bq kg⁻¹);
and
 $C(X)_{\text{aq}}$ is the concentration of radionuclide X in the surface water (Bq m⁻³).

Some models, such as the IMPACT and SACO models, assume an upper and lower sediment compartment to allow for burial of radionuclides, considering radionuclide decay and in-growth.

B1.6.6 Radionuclides in aquatic food

All the models calculate the radionuclide contents of aquatic food using generic transfer factors and the radionuclide concentration in the surface water system. The transfer factor to describe this may be defined as follows:

$$TF_{aq} = \frac{C(X)_{aq}}{C(X)_{sw}}$$

where: TF_{aq} is the transfer coefficient for radionuclide X from surface water to aquatic organism ($Bq\ kg^{-1} / Bq\ m^{-3}$ surface water);
 $C(X)_{aq}$ is the concentration of radionuclide X in the aquatic organism ($Bq\ kg^{-1}$);
and
 $C(X)_{sw}$ is the concentration of radionuclide X in the surface water ($Bq\ m^{-3}$ surface water).

B1.6.7 Discussion of approaches to modelling radionuclides in the biosphere

BIOMOVs II.4 and BIOMOVs II.5 showed good agreement in these calculations. Some variations in the results were attributed to the level of sophistication of the models for the biosphere. However, the simple approach of applying transfer factors to calculate the distribution of radionuclides throughout the biosphere was adequate for describing the transfer of radionuclides throughout the biosphere.

B1.7 Dose calculations

The dose to man, arising from interaction with the radiologically contaminated biosphere, is quantified by all of the models reviewed. The major exposure pathways are assumed to be ingestion of food and water, inhalation of radon and dust, and external irradiation. Quantification of the doses arising from these pathways is based on a series of generic values which describe human habits, such as food and water consumption, inhalation rates and occupancy of the area, and standard dose conversion factors. Therefore, the dose to an individual of defined habits can be calculated. The collective dose was not calculated by these models.

Both IMPACT and MEPAS incorporate more rigorous assessments which take account of additional factors such dermal adsorption, external irradiation by air and water immersion (e.g. when swimming).

B1.8 Probabilistic modelling

The models reviewed have facilities to carry out both deterministic and probabilistic calculations. In most cases the probabilistic facility was represented by an additional module to the deterministic model.

The models use one of two sampling approaches for probabilistic modelling; Monte Carlo sampling and Latin-Hypercube sampling. Monte Carlo procedures apply randomly sampled input variables to deterministic processes. These values are selected by assuming specified distribution about deterministic input values. The sampling procedure is carried out for a sufficient number of iterations (usually greater than 1000) to allow uncertainties associated with the output from the model to be defined. Latin Hypercube sampling assumes that the probability distribution about the deterministic values can be divided into intervals of equal probability. An assumption value can be obtained for each interval according to the interval's probability distribution. Applying these values to the model provides a more precise sampling procedure which covers the entire sample range is sampled. The Latin Hypercube approach is

more efficient as a smaller number of samples are required. However, the Latin Hypercube approach requires greater computer memory than the Monte Carlo approach.

Monte Carlo sampling for uncertainty distributions is employed by the IMPACT, INTAKE, MEPAS and SONS models. Maximum and minimum parameter values are used as cut-off points for the sampled distribution.

Latin Hypercube sampling is employed by the GEOS/ABRICOT, JAERI and RESRAD models. The GEOS/ABRICOT and JAERI models specify maximum and minimum parameter values as cut-off points. The RESRAD model specifies a cut-off defined by 3.1 times the standard deviation.

Probabilistic results from all models deviated within an order of magnitude of one another. Any discrepancies were taken to be a reflection of different choice on the relative importance of exposure pathways. It would appear that the choice of Monte Carlo or Latin Hypercube sampling procedures made little difference to the results. The application of a probabilistic module to a deterministic model appear to provide a satisfactory method for determining the uncertainty to be associated with the results from the models.

B1.9 Conclusions

This review highlights the factors which are important for describing the CARE example sites. The models reviewed take a broadly similar approach to calculating doses. However, they are of varying degrees of sophistication. These are summarised in Table B1.2. Comparative trials where the models were applied to calculate doses for the same scenario showed there to be good agreement between the results. The small variations between the results were attributed to variations in the importance of minor exposure pathways rather than major pathways [BIOMOVS II.4, 1995; BIOMOVS II.5, 1996].

Table B1.2 Summary of the characteristics of the models considered

	Model								
	ECOSR	GEOS/ABRICOT	IMPACT	INTAKE	JAERI	MEPAS	RESRAD	SACO	SONS
	<u>Atmospheric Migration Features</u>								
Point source term	✓	✓	✓	✓	✓	✓	✓	✓	✗
Gaussian plume	✓	✓	✓	✓	✓	✓	✓	✓	✓
Dry deposition	✓	✓	✓	✓	✓	✓	✓	✓	✓
Wet deposition	✓	✗	✗	✗	✗	✗	✗	✗	✓
Plume depletion	✓	✓	✓	✓	✗	✓	✗	✓	✓
Decay/ingrowth of plume	✓	✗	✓	✗	✗	✗	✗	✓	✓
Radon release modelled	unknown	✓	✓	✓	✓	✗	✓	unknown	✗
	<u>Groundwater Migration Features</u>								
Solution method	Numerical	Numerical	Numerical	Analytic/numerical	Numerical	Analytic/numerical	Analytic		Numerical
1 D advective flow	✓	✓	✓	✓	✓	✓	✓	✓	✓
Longitudinal dispersion	✗	✓	✓	✓	✓	✓	✓	✗	✓
Lateral dispersion	✓	✗	✗	✗	✗	✗	✗	✗	✗
Molecular diffusion	✗	✗	✓	✗	✗	✓	✗	✗	✗
Decay	✓	✓	✓	✓	✓	✓	✓	✓	✓
In-growth	✓	✓	✓	✓	✓	✓	✓	✓	✓
Retardation	✓	✓	✓	✓	✓	✓	✓	✓	✓
	<u>Biosphere Processes</u>								
Plant									
Foliar interception (dust)	✓	✓	✓	✓	✓	✓	✓	✓	✓
Foliar interception (water)	✓	✓	✓	✓	✓	✓	✓	✓	✓
Weathering	✓	✓	✓	✓	✓	✓	✓	✓	✓
Root uptake	✓	✓	✓	✓	✓	✓	✓	✓	✓
Soil									
Resuspension	✗	✗	✓	✓	✓	✓	✓	✗	✓
Erosion	✗	✓	✓	✓	✓	✗	✓	✗	✓
Leaching	✓	✓	✓	✓	✓	✓	✓	✓	✓
Decay	unknown	✓	✓	✓	✓	✓	✓	unknown	✓
Ingrowth	unknown	✓	✓	✓	✓	✓	✓	unknown	✗
Animal									
Fodder ingestion	✓	✓	✓	✓	✓	✓	✓	✓	✓
Water ingestion	✓	✓	✓	✓	✓	✓	✓	✓	✓
Soil ingestion	✓	✓	✓	✓	✓	✓	✓	✓	✓
Dust inhalation	✗	✗	✓	✓	✓	✗	✗	✓	✗
Radon inhalation	✓	✗	✓	✗	✗	✗	✗	✓	✗

Note: ✓ Characteristic included in model. ✗ Characteristic not included in model.

Table B1.2 Summary of the characteristics of the models considered (cont.)

	Model								
	ECOSR	GEOS/ABRICOT	IMPACT	INTAKE	JAERI	MEPAS	RESRAD	SACO	SONS
	<u>Exposure Pathways</u>								
Meat ingestion	✓	✓	✓	✓	✓	✓	✓	✓	✓
Leafy vegetable ingestion	✓	✓	✓	✓	✓	✓	✓	✓	✓
Water ingestion (G)	✓	✓	✓	✓	✓	✓	✓	✓	✓
Dust inhalation	*	✓	✓	✓	✓	✓	✓	unknown	✓
Radon inhalation (A)	unknown	✓	✓	✓	✓	✓	✓	unknown	✓
Soil external irradiation	*	✓	✓	✓	✓	✓	✓	✓	✓
	<u>Dose Calculations</u>								
Individual dose	✓	✓	✓	✓	✓	✓	✓	✓	✓
Collective dose	*	*	*	*	*	*	*	*	*
	<u>Probabilistic Calculations</u>								
Sampling technique	unknown	Latin Hypercube	Monte Carlo	Monte Carlo	Latin Hypercube	unknown	Latin Hypercube	unknown	Monte Carlo
Sampling range	unknown	Max. / min.	Max. / min.	Max. / min.	Max. / min.	unknown	3-1 × st. dev.	unknown	Max. / min.

Note: ✓ Characteristic included in model. * Characteristic not included in model.

A number of conclusions may be drawn from this review concerning the features which are required of a model for treating example sites in the CARE project. These are as follows:

1. The treatment of the waste as a point source can often be an adequate approximation within a model. However, this depends on the distribution and distance from the waste.
2. The migration of radionuclides through the atmosphere can be adequately modelled assuming a Gaussian plume dispersion. The model should take account of the decay and in-growth of short-lived radionuclides by assuming that these are in equilibrium.
3. Groundwater can be adequately modelled with the one-dimensional advection-dispersion equation. The model should include corrections for radionuclide decay and in-growth.
4. The biosphere can be adequately modelled by assuming that transfers within the system are in equilibrium. Generic transfer coefficients can be used to calculate the distribution of radionuclides between compartments. The biosphere should incorporate compartments to describe the distribution of radionuclides in soil, livestock and vegetables and also aquatic food obtained from local surface waters.
5. Probabilistic modelling could be adequately performed by applying Monte Carlo sampling procedures to a deterministic model.

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ANNEX B2
A TECHNICAL DESCRIPTION OF AMCARE

Table of Contents

B2.1	Introduction	154
B2.2	Site characterisation for inclusion in AMCARE	155
B2.3	Exposure Pathways	164
B2.4	Collective doses	174
B2.5	Incorporation of remediation techniques	174
B2.6	References	175

B2.1 Introduction

AMCARE is the collective name for a modular approach to the generic assessment of remediation options which may be applied to sites contaminated through differing processes and with differing physical characteristics. As an assessment tool, AMCARE has been developed primarily to present a common approach to the relative ranking of differing remediation options rather than to assess in detail the individual characteristics of particular sites and the exposure of local critical groups. Consequently, AMCARE is generally conservative for all transport pathways and critical group habits are assumed to be uniform at all sites, based on European-wide generic habits surveys. Similarly, all foodstuffs are generally assumed to be produced at the point of habitation. Nonetheless, AMCARE is sufficiently flexible to allow important site specific features to be taken into account (e.g. type of waste, radionuclide inventory, waste disposal area/volume, proximity to nearest surface water etc.) and the model has been designed to meet a number of requirements.

- i) To predict local critical group doses for each site over time, with or without remediation measures being introduced. In general, results are expressed for short term dose impacts, but peak dose, and therefore peak dose avoidance, over time-scales up to 10000 years may be derived.
- ii) To predict doses to workers on the site, whether engaged in remediation or other construction work.
- iii) To predict doses to a hypothetical group who inhabit houses constructed on the contaminated site in intimate contact with the main waste bearing area.
- iv) To provide an approximate local collective dose estimate over the period of ‘institutional control’ (100 years) and over a longer inter-generational period (500 years).
- v) To reflect inherent uncertainties in the accuracy (or applicability) of data used to define key parameters, through the incorporation of ranges on values allowing dose impacts to be expressed as ‘best estimates’ within a defined distribution.

It is clear from the descriptions of sites considered (Chapter 5) that the principal pathways for exposure are via ingestion of food and water; inhalation of radon or contaminated dust (from the waste and/or secondary contaminated sources); and external exposure to the contaminated waste or to secondary contaminated sources derived from the waste. In this case, ‘secondary contaminated sources’ are defined as any geosphere receptor (e.g. soil, surface water etc.) or biosphere receptor (e.g. meat or arable produce) for radionuclides migrating from the waste.

The modular construction of AMCARE (see Figure B2.1) breaks down the components of groundwater migration of radionuclides contained within the wastes (GWAM), the atmospheric dispersion of radon emanating from the waste (GASAM) and the calculation of resultant doses (DOSEAM). A separate, proprietary model (CRYSTAL BALL) is used to derive uncertainty distributions.

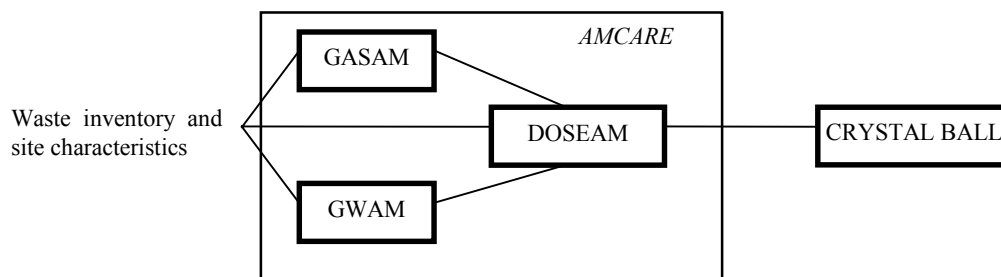


Figure B2.1 The modular construction of AMCARE

GWAM is a one dimensional groundwater pathway model which represents dissolved phase radionuclide migration through soil from the source to a location, which may then act as a

secondary source, over time. GASAM is a Gaussian plume atmospheric dispersion model, based on the characteristics defined by the UK Working Group on Atmospheric Dispersion [NRPB, 1979]. Output from both GWAM and GASAM is used to define the local contamination of soil, groundwater, surface water and air as inputs to the dose assessment module (DOSEAM). Since direct intrusion is also included as a potential pathway for exposure, the waste characteristics of the site can also be used as an input to define local conditions for DOSEAM. Both GWAM and GASAM are FORTRAN-based modules. DOSEAM is a relatively simple equilibrium-based spreadsheet model (using MS EXCEL) developed for the CARE project. The bases of these modules are described in more detail in the following sections. CRYSTAL BALL is a commercially available module for uncertainty and sensitivity analysis [Decisioneering Inc., 1996], and is not described further here.

B2.2 Site characterisation for inclusion in AMCARE

B2.2.1 Characterisation of the waste

The physical characteristics of each site and, in particular, of the waste disposal area, have been derived from maps and local knowledge, as described in Chapter 5. Such sources have been used to derive the surface area (or ‘footprint’) of the contaminated waste. Information relating to total waste inventory and mean concentrations of activity can then be used to yield estimates of the volume of waste. For simplicity, a monolithic slab of waste is assumed so that from the known surface area and volume of waste a uniform depth is derived (see Figure B2.2). For the purposes of off-site transport mechanisms, the waste is treated as an area source (i.e. a multiple point source).

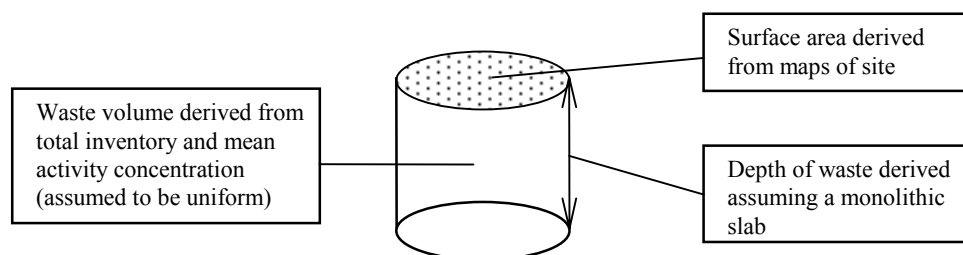
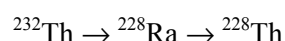


Figure B2.2 Physical characterisation of waste for input to AMCARE

At distances large by comparison to the source (e.g. $>3\times$ the waste surface diameter) consideration of the waste site can be simplified to a single point source.

B2.2.1.1 Radionuclide inventories

The radionuclides considered most relevant to the industrial processes included within CARE are those from the natural decay series of ^{238}U and ^{232}Th , which may be truncated to those nuclides of greatest radiological significance, i.e.,



In principle, other nuclides may be included for input to AMCARE, but are not considered here.

Correct characterisation of the waste is clearly fundamental to subsequent impact assessments. Since for many sites contaminated as a result of historic practices such characterisation may be relatively poor, a range of waste inventories can be used as input. Within this uncertainty, it is considered that all daughter nuclides exist in secular equilibrium with their parents and that

depletion of ^{238}U ($t_{1/2}$ 4.5×10^9 years) and ^{232}Th ($t_{1/2}$ 1.4×10^{10} years) is not significant over timescales of the order of hundreds of years.

B2.2.1.2 Leachate

Leaching of material from waste is determined by the geometry of the waste, radionuclide inventory (and chemical forms) and the physical characteristics of the disposal area (soil type, local and regional groundwater aquifers, waste porosity and water infiltration rates etc.). Where site specific values are not available, or are poorly characterised, it is assumed that the waste resembles a sandy soil with density 1500 kg m^{-3} , soil water content 0.3 and a vertical infiltration velocity of 0.1 m a^{-1} . It is further assumed that equilibrium is rapidly established between the concentration of radionuclides in leachate and solid waste, defined by a nuclide-specific solid:water distribution coefficient (K_d):

$$K_d(X) = \frac{C(X)_{\text{solid}}}{C(X)_{\text{leach}}}$$

where: $K_d(X)$ is the distribution coefficient for radionuclide X ($\text{m}^3 \text{ kg}^{-1}$);
 $C(X)_{\text{solid}}$ is the concentration of radionuclide X in the solid waste (Bq kg^{-1}); and
 $C(X)_{\text{leach}}$ is the concentration of radionuclide X in the leachate (Bq m^{-3}).

In principle, the solid:water K_d for any nuclide is also dependent on the soil (or sediment) type and the chemistry of the infiltrating water (e.g. calcium concentration). Since these latter characteristics may be poorly defined, however, a range of K_d values may be used as input to AMCARE to encompass such uncertainty. Nuclide specific 'best estimates' and ranges for K_d are presented in Table B2.1.

Table B2.1 Nuclide specific distribution coefficients and half lives

Nuclide	Half life (years)	Gross average distribution coefficient (K_d) values ($\text{m}^3 \text{ kg}^{-1}$)			
		sandy soils		aqueous system sediments	
		best estimate	range	best estimate	range
Po-210	3.80×10^{-1}	0.15	0.006 - 3.6	0.15*	0.006 - 3.6
Pb-210	2.23×10^1	0.27	0.0027 - 27	0.27*	0.0027 - 27
Ra-226	1.60×10^3	0.49	0.00082 - 300	0.5	0.1 - 1
Ra-228	5.75	0.49	0.00082 - 300	0.5	0.1 - 1
Th-228	1.91	3.0	0.045 - 200	10	1 - 1000
Th-230	7.54×10^4	3.0	0.045 - 200	10	1 - 1000
Th-232	1.41×10^{10}	3.0	0.045 - 200	10	1 - 1000
U-234	2.45×10^5	0.033	0.000055 - 20	0.05	0.02 - 1
U-238	4.47×10^9	0.033	0.000055 - 20	0.05	0.02 - 1

Source: Tuli (1985), IAEA-IUR (1994). Data for aqueous systems assume oxidising conditions. * No data for Po or Pb in aqueous systems are available and the value for sandy soils is assumed to apply for both systems.

B2.2.1.3 Radon emissions

The production of ^{222}Rn arises from the presence of ^{238}U , and its decay chain radionuclides, in the waste inventory and from naturally occurring radioactivity in the surrounding area. For many of the sites considered in this study the radioactivity present is essentially of natural origin, albeit subject to 'technological enhancement', hence it is not practical to distinguish the contribution of other local sources of activity from the waste inventory. In any case, such sources probably represent a minor contribution and the amount of ^{222}Rn generated is thus calculated simply from the total amount of ^{226}Ra estimated to be present in the waste. Radon so produced diffuses through the waste to the atmosphere, and may be retarded by a number of factors including the geometry and physical state of the waste or by the presence of soil or other covering layers. Such retardation may be important given the relatively short half-life of ^{222}Rn

(3.8 days) and it is likely that very little of the radon generated deep within the waste will be emitted to atmosphere. The exhalation rate of radon only contains a diffusion contribution. Though convective transport may contribute importantly to radon release, it is only prevailing for a limited number of cases.

If the waste/soil matrix is regarded as a porous mass of homogeneous material, semi-infinite in extent, then the efflux of radon at the surface is given by the expression [UNSCEAR, 1993]:

$$E(^{222}\text{Rn})_{\text{soil}} = C(^{226}\text{Ra}) \lambda(^{222}\text{Rn}) f \rho A [D_e/(\lambda(^{222}\text{Rn}) \epsilon)]^{0.5}$$

where: $E(^{222}\text{Rn})_{\text{soil}}$ is the ^{222}Rn efflux (Bq a^{-1}) from the waste/soil matrix;
 $C(^{226}\text{Ra})$ is the concentration of ^{226}Ra in the waste (Bq kg^{-1})
 $\lambda(^{222}\text{Rn})$ is the decay constant of ^{222}Rn (a^{-1});
 f is the emanation fraction for the waste/soil matrix;
 ρ is the density of waste (kg m^{-3});
 A is the surface area of the waste site (m^2);
 D_e is the effective diffusion coefficient through the waste/soil ($\text{m}^2 \text{a}^{-1}$); and
 ϵ is the porosity of the waste/soil.

In the case where a cap or cover is introduced over the waste bearing area, a further factor may be introduced. The cover is treated as a parallel-sided slab of thickness L (m), in which the production rate R of ^{222}Rn ($\text{Bq m}^{-2} \text{s}^{-1}$) is treated as a flux at the lower boundary of the cap. $C'(z)$ (Bq m^{-3}) is the ^{222}Rn concentration at height z (m) within the cap, and D_e' ($\text{m}^2 \text{s}^{-1}$) is the diffusion coefficient for ^{222}Rn in the cap. If the decay constant for ^{222}Rn is λ , then:

$$-D_e' \frac{dC'}{dz} \Big|_{z=0} = R \text{ (input flux boundary)}$$

$$C' \Big|_{z=L} = 0 \text{ (zero concentration above cap)}$$

$$D_e' \frac{d^2 C'}{dz^2} - \lambda (^{222}\text{Rn}) C' = 0 \quad 0 \leq z < L \text{ (diffusive transport)}$$

An appropriate solution of these equations within the cap is:

$$C'(z) = \frac{R}{(D_e' \lambda (^{222}\text{Rn}))^{0.5}} \exp \left[- \left(\frac{\lambda (^{222}\text{Rn})}{D_e'} \right)^{0.5} z \right]$$

The emitted flux at the top of the cap is given by:

$$R' = -D_e' \frac{dC'}{dz} \Big|_{z=L} = R \exp \left[- \left(\frac{\lambda (^{222}\text{Rn})}{D_e'} \right)^{0.5} L \right]$$

Thus, the efflux of radon at the surface in the presence of a cap is given by:

$$E(^{222}\text{Rn})_{\text{cap}} = E(^{222}\text{Rn})_{\text{soil}} \exp \left[- \left(\frac{\lambda(^{222}\text{Rn})}{D_e'} \right)^{0.5} L \right]$$

where: $E(^{222}\text{Rn})_{\text{cap}}$ is the ^{222}Rn efflux (Bq a^{-1}) from the waste if a cap is present
 $E(^{222}\text{Rn})_{\text{soil}}$ is the ^{222}Rn efflux (Bq a^{-1}) from the waste/soil matrix;
 $\lambda(^{222}\text{Rn})$ is the decay constant of ^{222}Rn (a^{-1});
 D_e' is the diffusion coefficient of radon in the cap ($\text{m}^2 \text{a}^{-1}$); and
 L is the thickness of the cap (m)

Suitable values for f , ρ , D_e and ϵ are presented by UNSCEAR (1993) and are summarised in Table B2.2. Site specific and generic default values for L are presented in Chapter 5. For the modelling the best estimates presented in Table B2.2 are used. To estimate the bias introduced by using a single value for these parameters, the probabilistic and sensitivity analyses of model parameters using Crystal Ball was carried out.

Table B2.2 Typical values for radon emanation, diffusion coefficient and bulk density

Parameter	Symbol	Best estimate	Range
the radon emanation fraction for waste/soil matrix	F	0.23	0.02 - 0.83
the density of waste/soil	ρ	1500 kg m^{-3}	
the effective radon diffusion coefficient through waste/soil	D_e	15.8 $\text{m}^2 \text{a}^{-1}$	0.00316 - 31.56
the effective radon diffusion coefficient for a cap	D_e'	15.8 $\text{m}^2 \text{a}^{-1}$	0.00316 - 31.56
the porosity of waste/soil	ϵ_l	0.3	0.01 - 0.5

B2.2.2 Characterisation of the surroundings

Over time, the impact of contaminated waste will not be confined to the site of disposal as radionuclides migrate slowly through the environment to soils, rivers, lakes and coastal areas in the region. Such sources can represent major exposure pathways to the local population.

Since the geology, topography and demography of regions surrounding waste sites may be poorly defined, a number of generic assumptions are employed. These include flat terrain, a uniform windrose, the presence of a nearby surface water body and of an arable field within 100 m of the site boundary. Such assumptions may not be representative, but allow a common approach to assessing the potential effects of remediation measures which may be introduced.

B2.2.2.1 Atmospheric transport of radon

The atmospheric transport model GASAM adopts a Gaussian plume approach, as defined in the recommendations of the UK Working Group on Atmospheric Dispersion [Clarke, 1979], for the short and medium range dispersion of radionuclides under different atmospheric conditions (wind speed, direction and atmospheric stability). In this approach the concentration of radon (C) for a number of point sources with the wind blowing into the sector considered, is given by:

$$C(^{222}\text{Rn})_j = \frac{F_j E(^{222}\text{Rn})}{(2\pi)^{1/2} \delta \Delta\theta v_j \sigma_{zj}} / 3.15 \times 10^7 \quad \text{where } \sigma_{zj} \leq H_j$$

where: $E(^{222}\text{Rn})$ is the annual flux of ^{222}Rn from the site (Bq a^{-1});
 δ is the downwind distance (m);
 $\Delta\theta$ is the included angle for each sector (radians);
 v_j is the mean wind speed during stability category j (m s^{-1});
 σ_{zj} is the vertical standard deviation of the plume at downwind distance at a defined distance from the waste during stability category j (m);
 F_j is a term to describe the vertical distribution of the plume during stability category j, defined below; and
 3.15×10^7 converts time units (s a^{-1}).

$$F_j = \exp\left(-\frac{(h_1 - h_0)^2}{2\sigma_{zj}^2}\right) + \exp\left(-\frac{(h_1 + h_0)^2}{2\sigma_{zj}^2}\right) + \exp\left(-\frac{(2H_j + h_1 + h_0)^2}{2\sigma_{zj}^2}\right) \\ + \exp\left(-\frac{(2H_j + h_1 - h_0)^2}{2\sigma_{zj}^2}\right) + \exp\left(-\frac{(2H_j - h_1 + h_0)^2}{2\sigma_{zj}^2}\right) + \exp\left(-\frac{(2H_j - h_1 - h_0)^2}{2\sigma_{zj}^2}\right)$$

where: h_0 is the height above the ground at the release point (m);
 h_1 is the height above the ground at the reception point (m);
 H_j is the atmospheric inversion height for stability category j (m); and
 σ_{zj} is the vertical standard deviation of the plume at the reception point for stability category j (m).

The method for calculating the vertical standard deviation of the plume for different atmospheric stability categories is described in Clarke (1979).

For longer distances, where the vertical extent of the plume approaches the height of the inversion ($\sigma_{zj} > H_j$), the plume is considered to be well mixed between the ground and the inversion layer, and a simpler approach is adopted:

$$C(^{222}\text{Rn})_j = \frac{E(^{222}\text{Rn})}{\delta \Delta\theta v_j H_j}$$

Finally, the annual average radon concentration at a point downwind of the waste depends on the annual probability of each stability category and of the wind direction into the sector considered, i.e.

$$C(^{222}\text{Rn}) = \frac{\Delta\theta}{2\Pi} \sum_j r_j C(^{222}\text{Rn})_j$$

where: $C(^{222}\text{Rn})$ is the mean annual concentration of ^{222}Rn at a given point (Bq m^{-3});
 r_j is the annual probability of stability category j;
 $\Delta\theta/2\Pi$ accounts for probability of wind blowing into the sector; and
 $C(^{222}\text{Rn})_j$ is the annual concentration of ^{222}Rn at a given point due to stability category j (Bq m^{-3}).

Stability categories in this case are defined by Smith (1973) based on the work of Pasquill (1961), and a detailed description of each 'Pasquill' stability category may be found in Clarke (1979).

In principle, any set of site-specific meteorological conditions can be defined within GASAM. However, for simplicity and to aid comparison of results, a series of default parameters are assumed for each site. These include a uniform wind rose and a constant distribution of Pasquill stability categories (A: 0-6%, B: 6%, C: 17%, D: 60%, E: 7%, F: 8% and G: 1-4%). Clearly, meteorological characteristics will vary across regions and countries and the derivation of frequency of stability categories, inversion heights and windspeeds by Simmonds *et al.* (1995) for Spain suggests that these assumptions will not be unreasonable for many parts of Europe. For non-uniform windroses, and where the population is unevenly distributed, a weighting factor can be introduced to emphasise the sector of highest exposure (e.g. predominantly upwind or downwind).

B2.2.2.2 Transport through groundwater

The transport of radionuclides through groundwater is modelled in GWAM by solving the standard advection-dispersion transport equation using a finite difference scheme developed by Crank-Nicolson, described in detail by Crank (1975), to calculate the rates of migration of radionuclides in the ground water, as follows.

$$D \frac{\delta^2 C}{\delta x^2} - v \frac{\delta C}{\delta x} - \lambda R_f C + (\lambda R_f C)_{parent} = \lambda R_f \frac{\delta C}{\delta t}$$

where: D is the diffusion coefficient ($\text{m}^2 \text{s}^{-1}$);
 C is the solute concentration (Bq m^{-3});
 x is the distance (m);
 v is the ground water velocity (m s^{-1});
 λ is the decay constant (s^{-1});
 R_f is the retardation factor; and
 $(\lambda R_f C)_{parent}$ has the same meaning, but uses parameters specific to the parent nuclide, and calculates the contribution arising from in-growth.

The retardation factor (R_f) is then calculated as:

$$R_f = \left(1 + \frac{\rho K_d}{\varepsilon} \right)$$

where: ρ is the density (kg m^{-3});
 K_d is the soil-water distribution coefficient ($\text{m}^3 \text{kg}^{-1}$); and
 ε is the porosity (-).

Water is assumed to infiltrate the waste from above and to leach radionuclides into a through-flowing groundwater body.

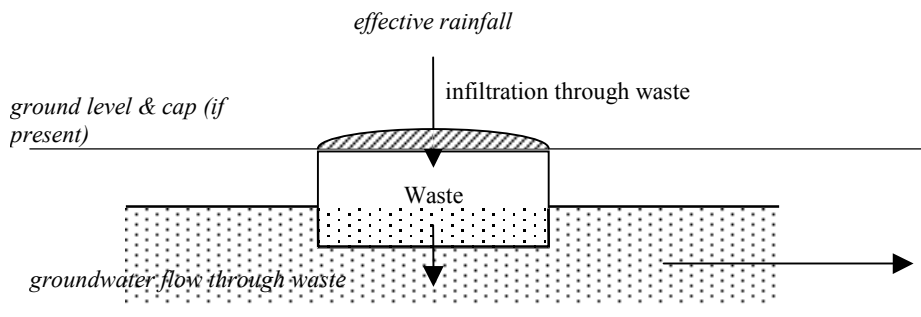


Figure B2.3 Conceptual model for infiltration of water through emplaced waste

The rate of input of each radionuclide into the groundwater is derived from the leachate flow through the waste and the concentration of radionuclides in the leachate (see Section B2.2.1.2). Values of the nuclide specific distribution coefficients are indicated in Table B2.1 and a generic

soil porosity for the aquifer flow of 0.3 is assumed. The cross sectional area of the flow is presented on a site specific basis in Chapter 5. The output from this calculation is the rate of input of a radionuclide (plus its daughter and granddaughter) into the soil or aqueous compartment at a defined distance from the waste for given intervals of time. These are considered separately below.

a) *Contamination of soil*

It is assumed that release of radionuclides from the groundwater is to a single well-mixed soil compartment via one or two pathways, (i) through groundwater migration, and (ii) through irrigation using surface water or well water. The concentration due to groundwater migration is calculated as:

$$C(X)_{soil} = \frac{\psi(X)_{soil-ground}}{\lambda(X)_{soil-loss} V_{soil}}$$

where: $C(X)_{soil}$ is the concentration of radionuclide X in the soil compartment ($Bq\ m^{-3}$);
 $\psi(X)_{soil,ground}$ is the rate of input of radionuclide X into the soil compartment from groundwater ($Bq\ a^{-1}$), i.e. the output from the GWAM module;
 $\lambda(X)_{soil,loss}$ is the rate of loss of radionuclide X from the soil compartment (a^{-1}); and
 V_{soil} is the soil compartment volume (m^3)

When ψ is defined as a flux (in $Bq\ m^{-2}\ a^{-1}$) then V may be replaced by d (depth), which is easier to determine, such that:

$$\lambda(X)_{soil-loss} = \frac{Infiltr}{d(\theta + \rho_{soil} K_d(X))} + \lambda(X)$$

where: Infiltr is the infiltration rate ($m\ a^{-1}$);
d is the depth of the terrestrial compartment (m);
 ρ_{soil} is the density of soil ($kg\ m^{-3}$);
 $K_d(X)$ is the distribution coefficient of radionuclide X ($m^3\ kg^{-1}$);
 θ is the soil water content at the rooting zone; and
 $\lambda(X)$ is the radioactive decay constant for radionuclide X (a^{-1}).

The concentration due to irrigation is calculated by:

$$C(X)_{soil-irrig} = \frac{C(X)_{water-irrig} t_{irrig} irr}{\lambda_{soil-loss} d_{soil}}$$

where: $C(X)_{water,irrig}$ is the concentration of radionuclide X in the irrigation water ($Bq\ m^{-3}$);
 t_{irrig} is the time period of irrigation ($d\ a^{-1}$);
irr is the irrigation rate ($m^3\ m^{-2}\ d^{-1}$);
 $\lambda(X)_{soil,loss}$ is the rate of loss of radionuclide X from the soil compartment (a^{-1});
and
 d_{soil} is the depth of the soil compartment (m).

The soil is assumed to be a slab of defined volume and depth. Site specific values for soil density, water content, K_d values etc. may be used where available. It is assumed that the soil on which arable crops are grown is irrigated with water abstracted from a well on that soil compartment. Pasture land is assumed not to be irrigated. Site specific data are presented in Chapter 5. A generic rate of irrigation for arable crops is applied at 0.01 m^3 water per m^2 soil per day, for 100 days over the year.

Decay of radionuclides within the soil matrix, and consequent in-growth of daughter nuclides is not explicitly incorporated within AMCARE or DOSEAM.

b) *Contamination of surface water*

As for soil, it is assumed that radionuclide transfer from groundwater is to a single well-mixed surface water compartment, in this case with a single outflow and the standard advection dispersion equation is again solved to calculate the input of radionuclides into the surface water. In practice, the distance of the surface water body from the source, and the size of the water body, must be defined on a site specific basis for realistic assessments (see Chapter 5), although default options may be adopted for comparative purposes. No default values are adopted here.

The input into this aquatic compartment is calculated as:

$$C(X)_{water} = \frac{\psi(X)_{water-input}}{\lambda(X)_{water-loss} V_{water}}$$

where: $C(X)_{water}$ is the concentration of radionuclide X in surface water ($Bq\ m^{-3}$);
 $\psi(X)_{water,input}$ is the rate of input of radionuclide X into surface water ($Bq\ a^{-1}$);
 $\lambda(X)_{water,loss}$ is the overall rate of loss of radionuclide X from surface water (a^{-1}); and
 V_{water} is the surface water compartment volume (m^3).

The loss of radionuclides from water can be further resolved into its component parts as:

$$\lambda(X)_{water-loss} = \frac{flow}{V_{water}} + \lambda(X)$$

where: flow is the water flow from the water compartment ($m^3\ a^{-1}$) - for rivers this can be defined as the flow through rate, for lakes as the outflow rate;
 V_{water} is the volume of the surface water compartment (m^3); and
 $\lambda(X)$ is the radioactive decay constant for radionuclide X (a^{-1}).

The volume of the surface water affected by the input of radionuclides is clearly site specific, but may be highly uncertain even where reasonable site characterisation is available and a range of values may be used within AMCARE.

For nuclides with a long half-life, the radioactive decay constant is small. Where $V_{water} \cdot \lambda(X)$ is also small by comparison to the flow rate (which is likely to be the case for most rivers, but may not apply to lakes), $C(X) = \psi(X)/flow$.

Upon entry to surface water, it is assumed that equilibrium is rapidly established between the radionuclides in the water and those in suspension or on bottom sediment or stream banks. Specific equilibria are defined by their distribution coefficients, $K_d(X)$, for which a range of values may be adopted (see Table B2.1).

B2.2.3 Local population characteristics

The distribution of future local populations is difficult to predict and this may, potentially, create problems in characterising both the critical group dose and collective dose estimates for the site. For simplicity, and to facilitate comparison of the generic effects of remediation options, a number of generic assumptions are adopted.

- i) The local critical group is taken to be a group of 30 people who reside 1 km distant from the site and work in a field 100 m from the site. The figure of 30 people for the critical group has been derived from the definition of a critical group by ICRP of consisting of a few tens of individuals. All the food and drinking water consumed by this group is assumed to be produced at the field 100 m from site.
- ii) In order to derive a collective dose, the area in which the population will be affected by the site is taken to be defined by a radius of 20 km from the site boundary, with a mean distance from site of 10 km. The population density is expressed on a site specific basis,

although it is rounded considerably to facilitate cross-comparison of results as far as possible.

- iii) Both the critical group and larger local residency group are assumed to remain constant in size and habits.

Where the local population is known to be significantly skewed from an even distribution, for instance due to the presence of a large town (see Figure B2.4), the radial area around the site can be divided into a number of concentric rings (e.g. *a*, *b* below) and the population of each area considered separately.

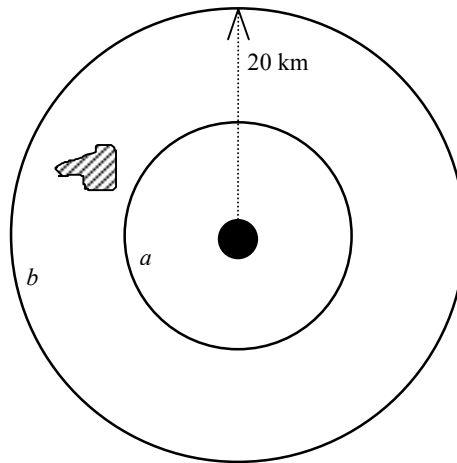


Figure B2.4 Construction of population characteristics

In addition to the scenario of local populations as defined above, an intrusion scenario is incorporated within the DOSEAM module, assuming that the waste site is developed as a housing estate and that people are thereby directly exposed to the radionuclides in the waste. Although in reality such a scenario can only be envisaged some considerable time after the operational period (100 - 200 years), the inventory of the important radionuclides will not have changed much due to the relatively long half-lives of the main waste nuclides of concern. For this scenario, it is assumed that food and vegetables are grown on the site and drinking water is derived from a local well, although this may be readily modified.

Workers on the site, engaged in remediation or other construction work, are assumed to reside at a more distant location and to consume 25% of the food produced locally. Their drinking water is assumed to originate from a distant (non-contaminated) source.

A summary of the critical group characteristics for the normal evolution case (local residency), the intrusion scenario and the workforce group is presented in Table B2.3.

Table B2.3 Summary of critical group characteristics

	Local critical group	Intrusion	Workforce
Habitation	village 1 km away	house on waste	village 1 km away
Time spent at habitation (h a ⁻¹)	6190	8760	7160
Outdoor occupancy at habitation (%)	33%	33%	33%
Workplace	field 100m away	field on waste	waste
Time spent at work (h a ⁻¹)	1600	N/A	1600
Outdoor occupancy at workplace (%)	100%	N/A	33%
Terrestrial foodstuff source	field 100 m away	field on waste	field 100 m away
Local food intake (% of total food intake)	100%	100%	25%
Aquatic foodstuff source	nearest surface water	nearest surface water	none
Time spent fishing (h a ⁻¹)	250	none	none
Drinking water source	well 100m away	well on waste	none
Irrigation water source	well 100m away	well on waste	N/A
Animal drinking water source	well 100m away	well on waste	N/A

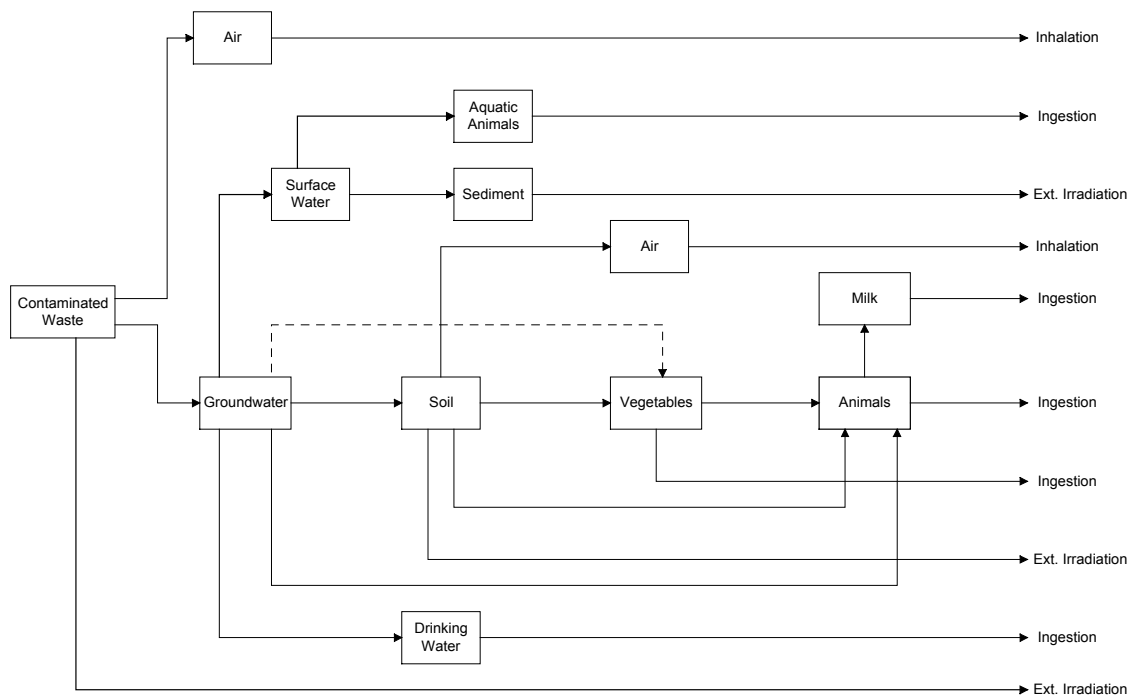
In all cases the habits of the local population and the workforce are defined by the average consumption and occupancy habits presented by Simmonds *et al.* (1995) for Europe and summarised in Table B2.4 below. Thus the ‘critical group’ is simply the group of people living closest to the site and who are the most exposed by virtue of local contaminant concentrations. The collective dose is then defined as the overall dose received by the entire local population in the defined area over 100 years (where the period of institutional control is of prime importance), or 500 years (for longer term projections; see Section B2.1). Within AMCARE, however, differing habits may be readily introduced where site specific data are available and the proportions of foods derived locally may also be introduced on a site specific basis.

Table B2.4 Typical population habits assumed

Habits pathway			Comments
Ingestion rates:	Drinking water	0.6 m ³ a ⁻¹	
	Roots	81 kg a ⁻¹	
	Cereals	84 kg a ⁻¹	
	Leafy vegetables	110 kg a ⁻¹	
	Beef	23 kg a ⁻¹	
	Milk	124 kg a ⁻¹	
	Pork	37 kg a ⁻¹	
	Fish (fw or sw)	13 kg a ⁻¹	
	Shellfish	0.6 kg a ⁻¹	
Inhalation rate:	1 m ³ h ⁻¹		
Occupancy:	Indoor	0.67	fraction of time spent at location
	Outdoor	0.33	fraction of time spent at location

B2.3 Exposure Pathways

Numerous pathways can be envisaged giving rise to potential exposure, and the relative importance of each pathway will vary from site to site. As a consequence, AMCARE incorporates a number of potentially relevant pathways (see Figure B2.5).



Note: Irrigation (-----) occurs only for arable crops and not for pasture land (see Section B2.2.2.2).

Figure B2.5 Exposure pathways from contaminated waste to man incorporated in AMCARE

In principal, other pathways may be included within AMCARE, which are not illustrated above. Accidental soil ingestion by children and infants, ingestion of soil with vegetables or irrigation by surface water instead of groundwater can be incorporated if this is likely to be of concern, as indicated in Figure B2.5.

Exposure due to uptake of radionuclides by the geosphere and biosphere is modelled assuming that equilibrium is rapidly achieved (Section B2.1). Generic values are presented in Tables B2.5-B2.7 for relevant transfer factors describing uptake of radionuclides to animal foodstuffs, pasture, agricultural crops and aquatic foodstuffs. Typical annual fodder intake rates for agricultural animals relevant to the assumed consumption groups are presented in Table B2.8.

Table B2.5 Transfer factors assumed for pasture grass and agricultural crops

Element	Transfer factors (and ranges) for pasture grass and agricultural crops Bq kg ⁻¹ (dry plant) per Bq kg ⁻¹ (dry soil)			
	Grass	Leafy vegetables	Roots vegetables	Grain
Po	9.0 × 10 ⁻² (9.0 × 10 ⁻³ - 9 × 10 ⁻²)	1.2 × 10 ⁻³ (1.2 × 10 ⁻⁴ - 1.2 × 10 ⁻³)	7.0 × 10 ⁻³ (7.0 × 10 ⁻⁴ - 7.0 × 10 ⁻³)	2.3 × 10 ⁻³ (2.3 × 10 ⁻⁴ - 2.3 × 10 ⁻³)
Pb	1.1 × 10 ⁻³ (1.1 × 10 ⁻⁴ - 1.1 × 10 ⁻²)	1.0 × 10 ⁻² (5 × 10 ⁻⁴ - 2 × 10 ⁻¹)	2.0 × 10 ⁻² (2.0 × 10 ⁻⁴ - 2.0 × 10 ⁻¹)	4.7 × 10 ⁻³ (4.7 × 10 ⁻⁴ - 4.7 × 10 ⁻²)
Ra	8.0 × 10 ⁻² (1.6 × 10 ⁻² - 4.0 × 10 ⁻¹)	4.9 × 10 ⁻² (2.5 × 10 ⁻³ - 9.8 × 10 ⁻¹)	1.1 × 10 ⁻³ (2.2 × 10 ⁻⁴ - 1.1 × 10 ⁻¹)	1.2 × 10 ⁻³ (2.4 × 10 ⁻⁴ - 6.0 × 10 ⁻³)
Th	1.1 × 10 ⁻² (1.1 × 10 ⁻³ - 1.1 × 10 ⁻¹)	1.8 × 10 ⁻³ (4.5 × 10 ⁻⁵ - 7.2 × 10 ⁻²)	5.6 × 10 ⁻⁵ (5.6 × 10 ⁻⁶ - 3.9 × 10 ⁻¹)	3.4 × 10 ⁻⁵ (3.4 × 10 ⁻⁶ - 8.5 × 10 ⁻⁴)
U	2.3 × 10 ⁻² (2.3 × 10 ⁻³ - 2.3 × 10 ⁻¹)	8.3 × 10 ⁻³ (8.3 × 10 ⁻³ - 8.3 × 10 ⁻²)	1.1 × 10 ⁻² (1.4 × 10 ⁻³ - 1.4 × 10 ⁻¹)	1.3 × 10 ⁻³ (1.3 × 10 ⁻⁴ - 1.3 × 10 ⁻²)

Note: Best estimates and ranges are derived from Jackson (1984) and IAEA-IUR (1994). A default range of ± one order of magnitude is assumed where no other information is available. Values for Po were derived from field data and stated not to be corrected for foliar interception. No range on values is presented in IAEA 364 [IAEA, 1994] but it is indicated that actual values may be 2-10 times lower than the best estimate. A range is assumed accordingly here.

Table B2.6 Transfer factors for animal foodstuffs

Element	Transfer factors (and ranges) for animal foodstuffs Bq kg ⁻¹ or l ⁻¹ (animal foodstuff) per Bq kg ⁻¹ (dry food) per kg day ⁻¹ (dry food intake)		
	Beef	Milk	Pork
Po	5.0 × 10 ⁻³ (6 × 10 ⁻⁴ - 5 × 10 ⁻³)	3.4 × 10 ⁻⁴ (n/a)	2.5 × 10 ⁻² (n/a)
Pb	4.0 × 10 ⁻⁴ (1 × 10 ⁻⁴ - 7 × 10 ⁻⁴)	3.4 × 10 ⁻⁴ (n/a)	2.0 × 10 ⁻³ (n/a)
Ra	9.0 × 10 ⁻⁴ (5 × 10 ⁻⁴ - 5 × 10 ⁻³)	1.3 × 10 ⁻³ (1 × 10 ⁻⁴ - 1.3 × 10 ⁻³)	4.0 × 10 ⁻³ (n/a)
Th	2.7 × 10 ⁻³ (n/a)	5.0 × 10 ⁻⁴ (n/a)	2.0 × 10 ⁻² (n/a)
U	3.0 × 10 ⁻⁴ (n/a)	4.0 × 10 ⁻⁴ (7.3 × 10 ⁻⁵ - 6.1 × 10 ⁻⁴)	6.2 × 10 ⁻² (n/a)

Note: Best estimate values and ranges are derived principally from IAEA-IUR (1994). The data are sparse in some cases, especially for swine. A range of values of ± one order of magnitude around the mean may be assumed where no data are available.

Table B2.7 Transfer factors for aquatic foodstuffs

Element	Transfer factors (and ranges) for aquatic foodstuffs Bq kg ⁻¹ (aquatic foodstuff) per Bq m ⁻³ (water)			
	Freshwater fish	Marine fish	Crustacea	Molluscs
Po	0.05 (0.01 - 0.5)	2.0 (n/a)	50.0 (n/a)	10.0 (n/a)
Pb	0.30 (0.1 - 0.3)	0.20 (n/a)	1.00 (n/a)	1.00 (n/a)
Ra	0.05 (0.01 - 0.2)	0.50 (n/a)	0.10 (n/a)	1.00 (n/a)
Th	0.10 (0.01 - 10)	0.60 (n/a)	1.00 (n/a)	1.00 (n/a)
U	0.01 (n/a)	0.001 (n/a)	0.01 (n/a)	0.03 (n/a)

Note: Transfer factors and ranges for freshwater fish derived from IAEA-IUR (1994). Transfer factors for marine organisms derived from IAEA (1985) and from stable element concentrations in Bowen (1966).

Table B2.8 Typical annual intakes by animals

Consumption	Dairy cattle (milk yield: 10-20 L d ⁻¹)		Beef cattle (500 kg)		Swine (110 kg)	
	best estimate	range	best estimate	range	best estimate	range
Water m ³ d ⁻¹	0.075	0.050-0.100	0.040	0.020-0.060	0.008	0.006-0.010
Soil* kg d ⁻¹	0.6	0.4-10	0.3	0.2-0.4	0.48	0.4-0.6
cereal kg d ⁻¹ dw	0	0	0	0	2.4	2.0-3.0
Grass kg d ⁻¹ dw	16.1	10-25	7.2	5-10	0	0

Source: IAEA-IUR (1994). * Soil consumption represents 4% of fodder intake by dw for cattle and 20% for swine.

Values for dose per unit intake of principal radionuclides, and their daughter products, are summarised in Table B2.9, for adults only. Relevant dose per unit intake values for children and infants, or for other nuclides, are available in ICRP Publication 72 [ICRP, 1996].

Table B2.9 Dose rate factors for adults

Nuclide	Inhalation (Sv Bq ⁻¹)	Ingestion (Sv Bq ⁻¹)	External (Sv a ⁻¹ per Bq m ⁻³)
²¹⁰ Po	3.30 × 10 ⁻⁶	1.20 × 10 ⁻⁶	1.47 × 10 ⁻¹⁴
²¹⁰ Pb	1.10 × 10 ⁻⁶	6.90 × 10 ⁻⁷	2.09 × 10 ⁻¹²
²²⁶ Ra	3.50 × 10 ⁻⁶	2.80 × 10 ⁻⁷	3.20 × 10 ⁻⁹
²²⁸ Ra	2.60 × 10 ⁻⁶	6.90 × 10 ⁻⁷	8.00 × 10 ⁻¹⁰
²²⁸ Th	4.00 × 10 ⁻⁵	7.20 × 10 ⁻⁸	4.90 × 10 ⁻¹⁰
²³⁰ Th	1.40 × 10 ⁻⁵	2.10 × 10 ⁻⁷	7.70 × 10 ⁻¹³
²³² Th	2.50 × 10 ⁻⁵	2.30 × 10 ⁻⁷	5.90 × 10 ⁻¹³
²³⁴ U	3.50 × 10 ⁻⁶	4.90 × 10 ⁻⁸	7.20 × 10 ⁻¹³
²³⁸ U	2.90 × 10 ⁻⁶	4.50 × 10 ⁻⁸	9.10 × 10 ⁻¹²

Note: Inhalation and ingestion dose per unit intake factors from ICRP 72 (1996). External dose factors derived from various sources as CED. ²²²Rn inhalation doses are calculated using a factor of 5 × 10⁻⁵ Sv a⁻¹ per Bq m⁻³.

B2.3.1 On-site exposure (intrusion scenario and workforce dose)

Six main exposure pathways are considered for on-site critical groups:

- i) dust inhalation;
- ii) external irradiation from the waste;
- iii) radon inhalation;
- iv) ingestion of well water abstracted on-site;
- v) ingestion of aquatic foodstuffs from nearest surface water body;
- vi) ingestion of food grown on-site.

Direct ingestion of waste is not included for adults, as discussed above. In the case of intrusion, where habitation occupancy is assumed on-site, all food is assumed to be grown on site and drinking water is derived from a well constructed on site. Workers engaged on remediation measures, or other construction work, are exposed during the time spent on the site but are otherwise assumed to be distantly located. Consequently, their drinking water is assumed to derive from a non-contaminated source. Workers, however, do derive 25% of their agricultural food (meat, milk, crops etc.) from a local source. Similarly, they derive their fish from a nearby surface water body, although this is assumed to be purchased and they do not spend time actually fishing in the area.

B2.3.1.1 Dust inhalation

In the absence of capping, or where the cap has been breached due to construction work, dust inhalation may arise from re-suspension of particles from the waste, and:

$$DINH(X) = C(X)_{\text{waste}} (DL_{\text{ext}} OCC_{\text{ext}} + DL_{\text{int}} OCC_{\text{int}}) BR DF(X)_{\text{INH}}$$

where: DINH(X) is the dose received from radionuclide X from inhaling dust (Sv a⁻¹);
 C(X)_{waste} is the concentration of radionuclide X in the waste (Bq kg⁻¹);
 DL_{ext} is the dust loading in air, outdoors (kg m⁻³);
 OCC_{ext} is the outdoor occupancy time (h a⁻¹);
 DL_{int} is the dust loading in air, indoors (kg m⁻³);
 OCC_{int} is the indoor occupancy time (h a⁻¹);
 BR is the inhalation rate (m³ h⁻¹); and
 DF(X)_{INH} is the dose factor for inhalation of radionuclide X (Sv Bq⁻¹).

In practice, for the residents or workforce on the site, the dust loading outdoors is likely to be higher than the dust loading indoors, although the respirable fraction of particles may be higher indoors. Consequently, in the absence of site specific information a single 'respirable' dust loading on the site is defined as 1 × 10⁻⁷ kg m⁻³ and this is taken to apply both indoors and

outdoors (but note that both the workforce and the intrusion scenario group of residents are exposed to dust outdoors only on site). This also simplifies the occupancy factor to be applied to residency on the site, since both the indoor and outdoor occupancy can be combined.

B2.3.1.2 External irradiation

The dose received from external irradiation is calculated from the expression:

$$DEXT(X) = C(X)_{\text{waste}} (OCC_{\text{ext}} s_{\text{ext}} + OCC_{\text{int}} s_{\text{int}}) DRF(X)_{\text{EXT}} \rho / 8760$$

where: $DEXT(X)$ is the dose received from radionuclide X by external irradiation (Sv a^{-1});
 $C(X)_{\text{waste}}$ is the concentration of radionuclide X in the waste (Bq kg^{-1});
 $OCC_{\text{ext/int}}$ is the occupancy time on the site outdoors or indoors (h a^{-1});
 $s_{\text{ext/int}}$ is a shielding factor for irradiation outdoors or indoors;
 $DRF(X)_{\text{EXT}}$ is the dose rate factor from external irradiation by radionuclide X (Sv a^{-1} per Bq m^{-3});
 ρ is the density of the waste (kg m^{-3}); and
 8760 is the number of hours in a year (h a^{-1}).

A generic factor for shielding is currently applied as 0.75 for s_{ext} and 0.25 for s_{int} [Oztunali *et al.*, 1981].

B2.3.1.3 Radon inhalation

DOSEAM calculates radon doses for both indoor and outdoor scenarios. A similar approach is used in both cases. A compartment is defined above the source and the radon input and output rate is calculated. The input rate is assumed to be the same as the efflux from the source (see Section B2.2.1.3).

The concentration of radon inside a building will be governed by the surface area of the building and its ventilation rate.

$$C_{\text{in}}(^{222}\text{Rn}) = \frac{E(^{222}\text{Rn})}{A(\text{site})} \frac{A(\text{building})}{\text{vent}}$$

where: $C_{\text{in}}(^{222}\text{Rn})$ is the indoor concentration of radon (Bq m^{-3});
 $E(^{222}\text{Rn})$ is the radon efflux (Bq a^{-1}) - see Section B2.2.1.3;
 $A(\text{site})$ is area of the waste site (m^2);
 $A(\text{building})$ is area of the building (m^2); and
 vent is the ventilation rate of the building ($\text{m}^3 \text{a}^{-1}$).

The indoor inhalation dose is then given by:

$$DINH_{\text{in}}(^{222}\text{Rn}) = C_{\text{in}}(^{222}\text{Rn}) DF_{\text{INH}} BR OCC_{\text{in}}$$

where: $DINH_{\text{in}}(^{222}\text{Rn})$ is the indoor inhalation dose (Sv a^{-1});
 $C_{\text{in}}(^{222}\text{Rn})$ is the indoor concentration of radon (Bq m^{-3});
 DF_{INH} is the dose factor for the inhalation of ^{222}Rn (Sv Bq^{-1});
 BR is the inhalation rate ($\text{m}^3 \text{h}^{-1}$);
 OCC_{in} is the indoor occupancy (h a^{-1});

The outdoor concentration of radon relevant to inhalation exposure is governed both by the assumed vertical cross sectional area above the source and the ratio between air concentration and emanation rate:

$$C_{out}({}^{222}\text{Rn}) = \frac{E({}^{222}\text{Rn})}{A(\text{site}) \quad v}$$

where: $C_{out}({}^{222}\text{Rn})$ is the outdoor concentration of radon (Bq m^{-3});
 $E({}^{222}\text{Rn})$ is the radon flux from the site (Bq a^{-1}) - see Section B2.2.1.3;
 $A(\text{site})$ is the area of the waste site (m^2); and
 v is the ratio between air concentration and emanation rate (taken to be $5.09 \times 10^{-4} \text{ m a}^{-1}$; Chamberlain, 1991).

The dose from inhalation can then be expressed as:

$$DINH_{out}({}^{222}\text{Rn}) = C_{out}({}^{222}\text{Rn})DF_{INH}BR \quad OCC_{out}$$

where: $DINH_{out}({}^{222}\text{Rn})$ is the indoor inhalation dose (Sv a^{-1});
 $C_{out}({}^{222}\text{Rn})$ is the indoor concentration of radon (Bq m^{-3});
 DF_{INH} is the dose factor for the inhalation of ${}^{222}\text{Rn}$ (Sv Bq^{-1});
 BR is the inhalation rate ($\text{m}^3 \text{ h}^{-1}$);
 OCC_{out} is the outdoor occupancy (h a^{-1}).

B2.3.1.4 Ingestion

a) Ingestion of aquatic foodstuffs

The ingestion of aquatic foods is calculated directly from the radionuclide concentrations in the contaminated surface water.

$$DING(\text{X})_{aq} = C(\text{X})_{aq} \text{TF}(\text{X})_{aq} \text{ING}_{aq} \text{DF}(\text{X})_{ING}$$

where: $DING(\text{X})_{aq}$ is the dose received from radionuclide X from ingesting a given aquatic foodstuff (Sv a^{-1});
 $C(\text{X})_{aq}$ is the concentration of radionuclide X in surface water (Bq m^{-3});
 $\text{TF}(\text{X})_{aq}$ is the transfer factor between radionuclide X from water to a given aquatic foodstuff ($\text{Bq kg}^{-1} \text{ food per Bq m}^{-3} \text{ water}$);
 ING_{aq} is the annual consumption of a given aquatic foodstuff (kg a^{-1}); and
 $\text{DF}(\text{X})_{ING}$ is the dose intake factor for ingestion of radionuclide X (Sv Bq^{-1}).

b) Ingestion of vegetables

The dose is calculated from the radionuclide content of the contaminated soil.

$$DING(\text{X})_{veg} = (C(\text{X})_{soil} + C(\text{X})_{soil,irrig}) \rho \text{TF}(\text{X})_{soil/veg} \text{ING}_{veg} \text{DF}(\text{X})_{ING}$$

where: $DING(\text{X})_{veg}$ is the dose received from radionuclide X from ingesting a given vegetable foodstuff (Sv a^{-1});
 $C(\text{X})_{soil}$ is the concentration of radionuclide X in soil ($\text{Bq m}^{-3} \text{ soil}$);
 $C(\text{X})_{soil,irrig}$ is the additional concentration of radionuclide X in soil due to irrigation ($\text{Bq m}^{-3} \text{ soil}$);
 ρ is the bulk density of soil (kg m^{-3});
 $\text{TF}(\text{X})_{soil/veg}$ is the crop specific soil-to-plant transfer factor of radionuclide X ($\text{Bq kg}^{-1} \text{ veg. per Bq kg}^{-1} \text{ soil}$);
 ING_{veg} is the annual consumption of a given vegetable (kg a^{-1});
 $\text{DF}(\text{X})_{ING}$ is the dose intake factor for ingestion of radionuclide X (Sv Bq^{-1}).

For all agricultural food consumption pathways, it is assumed that residents on the site (intrusion scenario) derive all their food from fields on the contaminated waste area. Therefore, the model assumes that, for on-site scenarios, the concentrations of radionuclides in the soil,

$C(X)_{\text{soil}}$, are taken to be those of the waste, $C(X)_{\text{waste}}$. For workers, 25% of food is derived from a nearby field, all other food is derived from a distant (non-contaminated) source.

c) *Ingestion of meat and milk*

Animals grazing pasture land, drinking contaminated well water and accidentally ingesting associated soil, may give rise to contaminated meat and milk which is then available for ingestion by man. This is calculated as follows within DOSEAM. No allowance is made for irrigation of pasture land (see Section B2.2.2.2 and Figure B2.5).

$$DING(X)_{\text{meat}} = [(C(X)_{\text{soil}} \rho TF(X)_{\text{soil/veg}} CON_{\text{veg}} TF(X)_{\text{veg/meat}} + (CON_{\text{soil}} C(X)_{\text{soil}} \rho TF(X)_{\text{soil/meat}}) + (CON_{\text{water}} C(X)_{\text{water}} TF(X)_{\text{water/meat}})] ING_{\text{meat}} DF(X)_{\text{ING}}$$

where: $DING(X)_{\text{meat}}$ is the dose received from radionuclide X from ingesting a given meat foodstuff (Sv a^{-1});
 $C(X)_{\text{soil}}$ is the concentration of radionuclide X in soil (Bq m^{-3} soil);
 ρ is the bulk density of soil (kg m^{-3});
 $TF(X)_{\text{soil/veg}}$ is the soil-to-pasture transfer factor for radionuclide X (Bq kg^{-1} veg. per Bq kg^{-1} soil);
 CON_{veg} is the daily fodder intake by animal (kg d^{-1});
 $TF(X)_{\text{veg/meat}}$ is the pasture-to-meat transfer factor for radionuclide X (Bq kg^{-1} meat per Bq kg^{-1} fodder per kg d^{-1} fodder intake);
 CON_{water} is the daily water intake by animal ($\text{m}^3 \text{d}^{-1}$);
 $TF(X)_{\text{water/meat}}$ is the water-to-meat transfer factor for radionuclide X (Bq kg^{-1} meat per Bq m^{-3} water per $\text{m}^3 \text{d}^{-1}$ water intake);
 CON_{soil} is the daily soil intake by animal (kg d^{-1});
 $TF(X)_{\text{soil/meat}}$ is the soil-to-meat transfer factor for radionuclide X (Bq kg^{-1} meat per Bq kg^{-1} soil per kg d^{-1} soil intake);
 ING_{meat} is the annual consumption of a given meat (kg a^{-1}); and
 $DF(X)_{\text{ING}}$ is the dose intake factor for the ingestion of radionuclide X (Sv Bq^{-1}).

Soil consumption by animals is generally derived as a percentage of dry fodder intake and typical values are presented in Table B2.6. For the purposes of these calculations the value of $TF(X)_{\text{soil/meat}}$ and $TF(X)_{\text{water/meat}}$ is taken to be the same as that for $TF(X)_{\text{veg/meat}}$. It is likely that this will overestimate resultant doses.

d) *Ingestion of drinking water*

The calculation of the dose from the ingestion of drinking water depends upon the source of the water. For habitation occupancy on-site (intrusion scenario), all drinking water is assumed to be derived from wells and radionuclides in the aqueous phase only are considered.

$$DING(X)_{\text{water}} = C(X)_{\text{water}} ING_{\text{water}} DF(X)_{\text{ING}}$$

where: $DING(X)_{\text{water}}$ is the dose received from radionuclide X from ingesting a given volume of drinking water (Sv a^{-1});
 $C(X)_{\text{water}}$ is the concentration of radionuclide X in drinking water (Bq m^{-3});
 ING_{water} is the annual consumption of contaminated drinking water ($\text{m}^3 \text{a}^{-1}$);
and
 $DF(X)_{\text{ING}}$ is the dose intake factor for the ingestion of radionuclide X (Sv Bq^{-1}).

Drinking water is not included as a pathway for workers on-site, who are assumed to derive their water from a distant source.

B2.3.2 Off-site exposure (local residency critical group)

The main indirect exposure pathways will be:

- i) inhalation of dust;

- ii) external exposure.
- iii) radon inhalation;
- iv) ingestion of food and water.

These exposure routes can arise from different secondary sources, as discussed below.

B2.3.2.1 Dust inhalation

Dust inhalation may arise from contaminated soil, calculated as follows.

$$DINH(X) = C(X)_{soil} (DL_{ext} OCC_{ext} + DL_{int} OCC_{int}) BR DCF(X)_{INH}$$

- where:
- $DINH(X)$ is the dose received from radionuclide X from inhaling dust ($Sv a^{-1}$);
 - $C(X)_{soil}$ is the concentration of radionuclide X in the soil, 100 m from source ($Bq kg^{-1}$);
 - DL_{ext} is the dust loading in air, outdoors ($kg m^{-3}$);
 - OCC_{ext} is the outdoor occupancy time ($h a^{-1}$); and
 - DL_{int} is the dust loading in air, indoors ($kg m^{-3}$);
 - OCC_{int} is the indoor occupancy time ($h a^{-1}$);
 - BR is the inhalation rate ($m^3 h^{-1}$); and
 - $DCF(X)_{INH}$ is the dose factor for inhalation of radionuclide X ($Sv Bq^{-1}$).

In practice, the dust loading outdoors is likely to be higher than the dust loading indoors, although the respirable fraction of particles may be higher indoors. In the absence of site specific information a single respirable dust loading value is defined as $1 \times 10^{-7} kg m^{-3}$ - see Section B2.3.1.1.

B2.3.2.2 External irradiation

The dose accruing from external irradiation can arise due to exposure to contaminated soil or to contaminated sediment on the banks of surface water bodies. The magnitude of these doses depends on the exposure time, for which a generic assumption is made.

$$DEXT(X) = C(X)_{soil} OCC DF(X)_{EXT} / 8760$$

- where:
- $DEXT(X)$ is the dose received from radionuclide, X, from exposure ($Sv a^{-1}$);
 - $C(X)_{soil}$ is the concentration of radionuclide, X, in soil, 100 m from source ($Bq m^{-3}$);
 - OCC is the exposure time to an external irradiation source ($h a^{-1}$);
 - $DF(X)_{EXT}$ is the dose rate factor for external irradiation by radionuclide, X ($Sv a^{-1}$ per $Bq m^{-3}$); and
 - 8760 is the number of hours in a year ($h a^{-1}$).

B2.3.2.3 Radon inhalation

At any distance (x) from the source, the dose due to inhalation is defined by the concentration of ^{222}Rn ($Bq m^{-3}$), the breathing rate of an individual ($m^3 h^{-1}$), occupancy time ($h a^{-1}$) and the dose per unit intake ($Sv Bq^{-1}$). For convenience, it is assumed that an average breathing rate applies and that ^{222}Rn concentrations are the same indoors and outdoors. Therefore, the overall occupancy for the area is used.

$$DINH_x(^{222}\text{Rn}) = C_x(^{222}\text{Rn})OCC_xDF(^{222}\text{Rn})$$

where: $DINH_x(^{222}\text{Rn})$ is the inhalation dose at distance x (Sv a^{-1});
 $C_x(^{222}\text{Rn})$ is the concentration of radon in air (Bq m^{-3}) at distance x - see Section B2.2.2.1;
 OCC_x is the occupancy at distance x (h a^{-1}); and
 $DF(^{222}\text{Rn})$ is the radon inhalation dose factor (assumed to be $5 \times 10^{-5} \text{ Sv h}^{-1}$ per Bq m^{-3}).

B2.3.2.4 Ingestion

Figure B2.5 indicates that ingestion of contaminated material may arise from a variety of pathways. In addition, local agricultural practices may lead to contamination being the result of more than one secondary source. Pathways considered in DOSEAM are discussed below. The sources of foodstuffs, and quantities consumed, are summarised in Tables B2.3 and B2.4.

a) Ingestion of aquatic foodstuffs

The ingestion of aquatic foods is calculated directly from the radionuclide concentrations in the contaminated surface water.

$$DING(X)_{\text{aq}} = C(X)_{\text{aq}} TF(X)_{\text{aq}} ING_{\text{aq}} DF(X)_{\text{ING}}$$

where: $DING(X)_{\text{aq}}$ is the dose received from radionuclide X from ingesting a given aquatic foodstuff (Sv a^{-1});
 $C(X)_{\text{aq}}$ is the concentration of radionuclide X in surface water (Bq m^{-3});
 $TF(X)_{\text{aq}}$ is the transfer factor between radionuclide X from water to a given aquatic foodstuff (Bq kg^{-1} food per Bq m^{-3} water);
 ING_{aq} is the annual consumption of a given aquatic foodstuff (kg a^{-1}); and
 $DF(X)_{\text{ING}}$ is the dose intake factor for ingestion of radionuclide X (Sv Bq^{-1}).

b) Ingestion of terrestrial vegetables

Dose arising from the ingestion of terrestrial vegetables is calculated from the radionuclide content of the contaminated soil. In addition, irrigation is taken into account. The source of the irrigation water is assumed to be from a well in the contaminated groundwater.

$$DING(X)_{\text{veg}} = (C(X)_{\text{soil}} + C(X)_{\text{soil,irrig}}) \rho TF(X)_{\text{soil/veg}} ING_{\text{veg}} DF(X)_{\text{ING}}$$

where: $DING(X)_{\text{veg}}$ is the foodstuff ingestion dose received from radionuclide X (Sv a^{-1});
 $C(X)_{\text{soil}}$ is the concentration of radionuclide X in soil, 100 m from source (Bq m^{-3});
 $C(X)_{\text{soil,irrig}}$ is the additional concentration of radionuclide X in soil due to irrigation (Bq m^{-3});
 ρ is the bulk density of soil (kg m^{-3});
 $TF(X)_{\text{soil/veg}}$ is the soil-to-plant transfer factor for radionuclide X (Bq kg^{-1} veg. per Bq kg^{-1} soil);
 ING_{veg} is the annual consumption of a given vegetable (kg a^{-1});
 $DF(X)_{\text{ING}}$ is the dose intake factor for the ingestion of radionuclide X (Sv Bq^{-1}).

c) Ingestion of meat and milk

Calculation of the dose arising from the consumption of contaminated meat and milk is determined by the soil-pasture-animal uptake pathway. Contamination of fodder takes place through root uptake. The animal can also consume contaminated water (well water or surface water) or contaminated soil. The significance of each of these pathways will be specific to a given site. For simplicity, it is assumed that the water consumed by animals is, in all cases, well water. No irrigation of pasture land is assumed.

$$DING(X)_{meat} = [(C(X)_{soil} \rho TF(X)_{soil/veg} CON_{veg} TF(X)_{veg/meat}) + (CON_{water} C(X)_{water} TF(X)_{water/meat}) + (CON_{soil} C(X)_{soil} \rho TF(X)_{soil/meat})] ING_{meat} DF(X)_{ING}$$

where: $DING(X)_{meat}$ is the dose received from radionuclide X from ingesting a given meat foodstuff ($Sv a^{-1}$);
 $C(X)_{soil}$ is the concentration of radionuclide X in soil, 100 m from source ($Bq m^{-3}$);
 ρ is the bulk density of soil ($kg m^{-3}$);
 $TF(X)_{soil/veg}$ is the soil-to-plant transfer factor for radionuclide X ($Bq kg^{-1} veg. per Bq kg^{-1} soil$);
 CON_{veg} is the daily fodder intake by animal ($kg d^{-1}$);
 $TF(X)_{veg/meat}$ is the fodder-to-meat transfer factor for radionuclide X ($Bq kg^{-1} meat per Bq kg^{-1} veg per kg d^{-1} fodder intake$);
 CON_{water} is the daily water intake by animal ($m^3 d^{-1}$);
 $C(X)_{water}$ is the concentration of radionuclide X in well water ($Bq m^{-3}$);
 $TF(X)_{water/meat}$ is the transfer factor for radionuclide X from water to meat ($Bq kg^{-1} meat per Bq m^{-3} water per m^3 d^{-1} water intake$);
 CON_{soil} is the daily soil intake by animal ($kg d^{-1}$);
 $TF(X)_{soil/meat}$ is the transfer factor for radionuclide X from soil to meat ($Bq kg^{-1} meat per Bq kg^{-1} soil per kg d^{-1} soil intake$);
 ING_{meat} is the annual consumption of a given meat ($kg a^{-1}$);
 $DF(X)_{ING}$ is the dose factor for the ingestion of radionuclide X ($Sv Bq^{-1}$).

For the purposes of these calculations the values of $TF(X)_{soil/meat}$ and $TF(X)_{water/meat}$ are taken to be the same as that for $TF(X)_{veg/meat}$. This will be generally conservative. Soil intake may be derived as a percentage of dry fodder intake. Generic guidance is presented in Table B2.6.

As is the case for the ingestion of vegetables, the model assumes that, for on-site scenarios, the concentration of radionuclides in the soil, $C(X)_{soil}$, is the same as those found in the waste, $C(X)_{waste}$.

d) *Ingestion of drinking water*

The calculation of the dose from the ingestion of drinking water depends upon the source of the water. In this case, all drinking water is assumed to be derived from wells and radionuclides in the aqueous phase only are considered (i.e. filtration of drinking water is assumed).

$$DING(X)_{water} = C(X)_{water} ING_{water} DF(X)_{ING}$$

where: $DING(X)_{water}$ is the dose received from radionuclide X from ingesting a given volume of drinking water ($Sv a^{-1}$);
 $C(X)_{water}$ is the concentration of radionuclide X in drinking water from well ($Bq m^{-3}$);
 ING_{water} is the annual consumption of contaminated drinking water ($m^3 a^{-1}$);
and
 $DF(X)_{ING}$ is the dose intake factor for the ingestion of radionuclide X ($Sv Bq^{-1}$).

B2.4 Collective doses

Collective dose is defined here within a very narrow context and applies only to the summated dose to the local population over a defined period of time. That is, within the local population radius (20 km - see Figure B2.5) the collective dose is taken to be:

$$\text{Coll} = \int_x \text{DINH}_{\text{IN}}(^{222}\text{Rn})_{10\text{km}} \Pi r^2 \text{POP}_{\text{den}} + \int_x \text{CR}_{\text{dose}} \text{CRPOP}$$

where: Coll is the collective dose to the local population (manSv);
 $\int_x \text{DINH}_{\text{IN}}(^{222}\text{Rn})_{10\text{km}}$ is the time integrated inhalation dose over x years (see below) from radon (previously defined) at a representative mid point distance, taken here as 10 km;
 Πr^2 defines the area with radius r , taken here as 20 km;
 POP_{den} is the population density per km^2 ;
 $\int_x \text{CR}_{\text{dose}}$ is the time integrated average dose, over x years (see below), to a member of the local residency critical group from all pathways (dust inhalation, external irradiation and all ingestion pathways other than radon inhalation); and
CRPOP is the population size of the local residency critical group.

The collective dose thus includes the critical group, but excludes any far distant population. A default assumption is made that the local critical group comprises 30 individuals. In fact, it can be seen that the dose for pathways other than that due to radon derives solely from the critical group. This reflects the very slow migration of radionuclides in groundwater such that beyond the first km or so, all dose pathways other than radon inhalation, are non-significant.

Radon inhalation will affect all members of the local population. The model assumes that the level of radon linearly declines from its source. As such, the average radon dose to a member of the local population may be calculated at the mid-point of the affected area, i.e. at 10 km.

Doses are summed over 100 years, or over a longer inter-generation period of 500 years. Long-term collective doses (e.g. to 10,000 years or more) are not presented.

Collective doses are derived only for the off-site exposure (normal evolution) scenario. Site intrusion is considered in the context of critical group doses only.

B2.5 Incorporation of remediation techniques

This is achieved by identifying the exposure routes affected by different techniques and estimating the likely magnitude of the effect. The techniques fall into several broad categories: removal of source, containment of source, immobilisation of the source and separation of radionuclides from the source. A detailed discussion of techniques, and their potential application, is presented in Chapter 6 and Annex B3.

B2.5.1 Bulk removal of waste

This may be interpreted as a reduction in the magnitude of the source. It will have an effect on both the level of radon generation and on the groundwater flux into the geosphere. The level of reduction will depend on the new geometry of the site. For the purposes of CARE the change is taken to be proportional to the reduction in the volume of the waste.

B2.5.2 Containment by capping

The presence of a cap will considerably reduce the level of radon emitted from the waste. This can be reflected in AMCARE by a reduction in the flux of radon from the waste (i.e. by varying the depth of cap (L), in Section B2.2.1.3).

Capping will also considerably reduce vertical infiltration from the waste into the groundwater. The model can reflect this by altering the input into the advection dispersion model through a reduced infiltration rate.

B2.5.3 Containment by subsurface barriers

These will primarily alter the flow of the groundwater through the waste. The model can reflect this by altering the input term into the groundwater.

These techniques will have minimal impact on the radon flux from the waste.

B2.5.4 Immobilisation

This will have an effect on the amount of radon emitted by the source and on the flux for other radionuclides into the groundwater. Changes to the radon emission are reflected by an alteration to the emanation fraction, f , in the model. The reduction of radionuclide flux into the groundwater can be represented by a reduction in radionuclide into the advection-dispersion model through an increased retardation rate and K_d value.

B2.5.5 Separation processes

These can be interpreted as a removal of source. They can affect both the radon flux and the groundwater release of other radionuclides. The model deals with these changes by altering the concentrations of radionuclides in the waste. However, the efficiency of a separation process will vary considerably depending on the choice of separation process and the radionuclide, which may be represented by a range in values.

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ANNEX B3
INTERCOMPARISON OF THE DOSE PREDICTIONS OBTAINED
WITH THE GENERIC MODEL, AMCARE, WITH THOSE
OBTAINED FROM A MODEL SPECIFIC TO THE TESSENDERLO
SITE

Table of Contents

B3.1	Introduction	178
B3.2	Normal evolution scenario	178
B3.3	Intrusion scenario	180
B3.4	Workforce scenario	181
B3.5	Conclusions	181
B3.6	References	182

B3.1 Introduction

AMCARE is a generic model which has been developed to predict the impact of remediation options for contaminated sites. A detailed description of the model may be found in Annex B2. AMCARE allows the impact of remediation options to be determined by first calculating the doses resulting from the current situation and then calculating the doses resulting from the application of a remediation technology. The model allows doses to be calculated both on the waste site (for the purposes of intrusion scenario calculations) and also at specified distances from the waste deposits (for normal evolution scenario calculations). However, it is necessary to verify this model in order to estimate the accuracy of these predictions.

AMCARE was used to calculate the doses arising from the wastes of a phosphate factory in Tessenderlo (Belgium). This site occupies an area of $5.3 \times 10^5 \text{ m}^2$ and contains a volume of $9 \times 10^6 \text{ m}^3$ of waste material. The waste contains 3500 Bq kg^{-1} of ^{226}Ra and 40 Bq kg^{-1} of ^{232}Th . The waste is covered by about 1.5 m of sand and the cover is drained. The doses arising from this waste have been calculated using a site-specific model developed by SCK•CEN. A comparison of the two sets of results was used to determine the likely accuracy of the AMCARE model.

The SCK•CEN model was designed to assess the maximum individual dose to an average member of the critical group. It considered two scenarios:

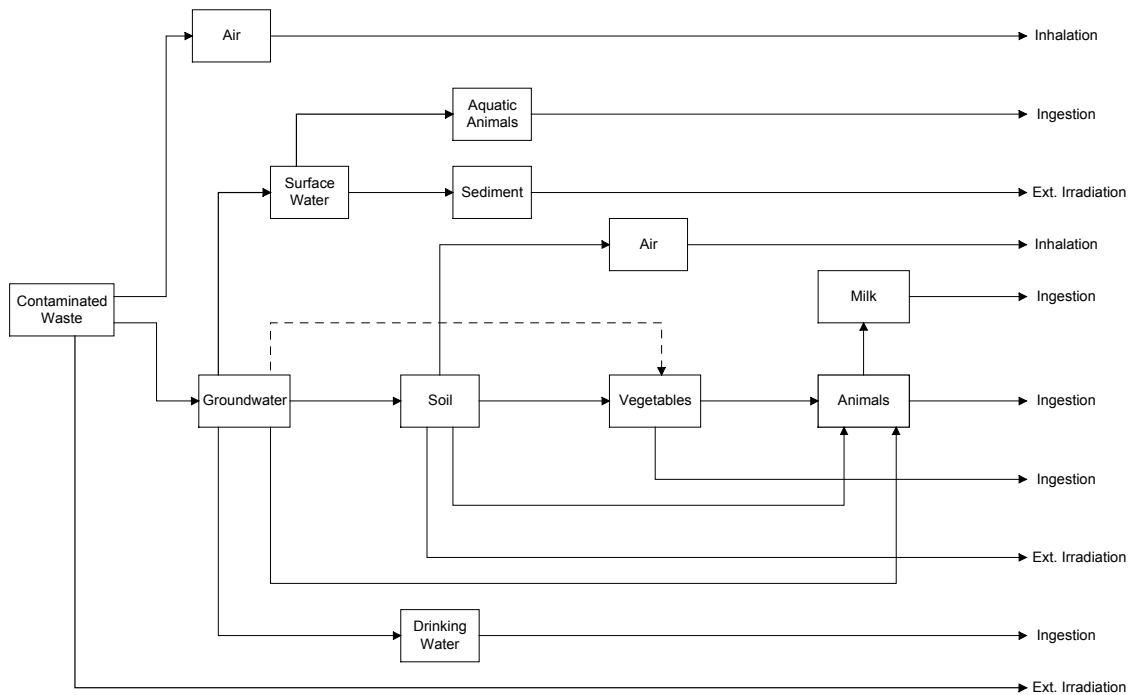
- normal evolution scenarios (well and river receptors);
- human intrusion scenarios (construction and residence).

These scenarios were of sufficient similarity to those of AMCARE to enable comparisons to be made.

This annex compares the results obtained with the two models. Variations in the two sets of results are discussed and differences in approaches identified. However, it was found that only the intrusion scenario provided an opportunity for a detailed comparison of the two models. The conclusion is that AMCARE can produce results which are comparable to those obtained by the SCK•CEN model. However, significant differences in doses can arise from differences in some of the assumptions made for the two models.

B3.2 Normal evolution scenario

AMCARE models the normal evolution scenario by assuming that two transport pathways exist for the migration of radionuclides from the waste to a specified population who live off-site. These pathways involve the emanation of ^{222}Rn and its subsequent transport from the radioactive waste and also the leaching of radionuclide from the waste into groundwater and subsequent migration through the aquifer into the local biosphere. Exposure of the population can arise through the inhalation of the radon, inhalation of contaminated dust, ingestion of contaminated foodstuff (and drinking water) and through external irradiation of contaminated soil. These exposure pathways are summarised in Figure B3.1.



Note: Irrigation (-----) occurs only for arable crops and not for pasture land.

Figure B3.1 Exposure pathways from contaminated waste to man incorporated in AMCARE (see Annex B2)

The groundwater pathways considered by AMCARE are very similar to those used by the SCK•CEN model whose groundwater pathways are summarised in Figure B3.2

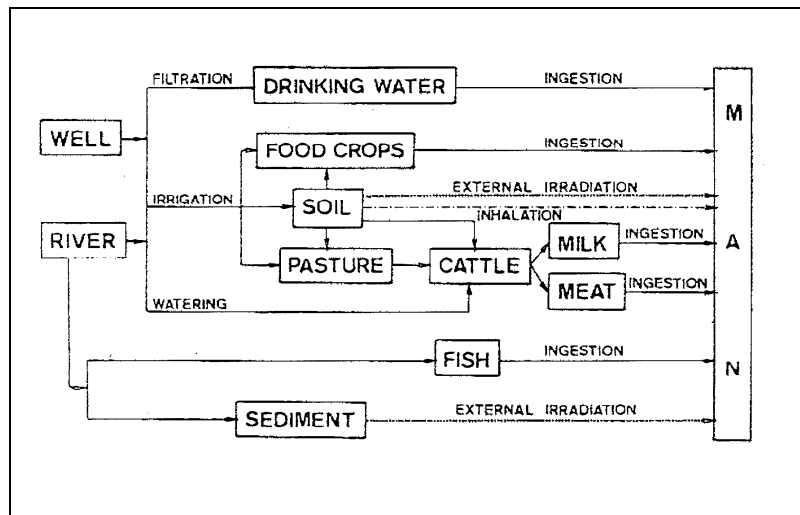


Figure B3.2 Exposure pathways from contaminated waste to man incorporated in SCK•CEN model for Tessenderlo

This SCK•CEN model does not take account of exposure to radon (the inclusion of radon exposure is through direct measurement). Performing a radiological assessment for the Tessenderlo site with the AMCARE model has shown that inhalation of radon is the most important pathway by which the local population is exposed to the contaminated waste. A direct comparison of the overall dose for a normal evolution scenario predicted by both models is therefore not possible.

However, a limited comparison may be made if the radon dose is ignored. Under these circumstances, both models will be dominated by exposure pathways involving ^{226}Ra . The migration of ^{226}Ra through groundwater will be greatly affected by the choice of K_d value for groundwater. AMCARE assumes the K_d value for radium in groundwater to be $0.5 \text{ m}^3 \text{ kg}^{-1}$ and predicts a peak dose (assuming a 10,000 year scenario) of $1 \times 10^{-6} \text{ mSv a}^{-1}$. The SCK•CEN model also presents a dose where the K_d value for radium is assumed to be $0.5 \text{ m}^3 \text{ kg}^{-1}$. This model predicts the peak dose to be $2.2 \times 10^{-6} \text{ mSv a}^{-1}$. These results show reasonable agreement and it may be concluded that differences will only result from the differences in the transfer parameters incorporated into the two models.

AMCARE is used to calculate the collective dose for the area. This is based on a number of assumptions concerning the local population and their habits. The SCK•CEN model calculates the maximum individual dose to an average member of the critical group only and, therefore, the collective dose calculations cannot be verified.

B3.3 Intrusion scenario

The intrusion scenarios of the two model are directly comparable although some of the assumptions made are slightly different. The most significant of these differences are as follows:

- AMCARE assumes individuals on the site spend 67% of the time indoors. The SCK•CEN model for the site assumes 7000 h a^{-1} indoor (80%) and 1800 h a^{-1} outdoors.
- The typical consumption habits used for the two models show some degree of variation (see Table B3.1). Food is assumed to come from the nearest food or the spoil heap.

Table B3.1 Comparison of population habits for AMCARE and SCK•CEN model

Habit Pathway			AMCARE	SCK•CEN model
Ingestion rates:	Drinking water	$\text{m}^3 \text{ a}^{-1}$	0.6	0.4
	Roots	kg a^{-1}	81	{ 170
	Cereals	kg a^{-1}	84	
	Leafy vegetables	kg a^{-1}	110	
	Beef	kg a^{-1}	23	
	Milk	kg a^{-1}	124	150
	Pork	kg a^{-1}	37	0
	Fish (fw or sw)	kg a^{-1}	13	5
	Shellfish	kg a^{-1}	0.6	0
Inhalation rate:	Indoor	$\text{m}^3 \text{ hr}^{-1}$	1	1
	Outdoor	$\text{m}^3 \text{ hr}^{-1}$	1	0.75

- The SCK•CEN model uses a set of standard transfer factors to account for the uptake of radionuclides by vegetables and animals. These transfer factors are used to determine the distribution of all radionuclides. The AMCARE model uses transfer factors which are radionuclide-specific (see Annex B2).
- The SCK•CEN model applies specific dust concentration values for indoor and outdoor situations. AMCARE only considers the concentration of dust for outdoor situations.
- The SCK•CEN model applies specific radon emanation rates to the inside and outside of buildings. AMCARE calculates these directly using generic data.
- The SCK•CEN model takes account of the building being on a foundation slab. AMCARE does not consider this.

It is clear from these points that there are differences between the two models. However, many of these differences are small and it is to be expected that the doses calculated by the two models should be sufficiently similar to allow the results to be compared. The doses calculated by the two models are given in Table B3.2.

Table B3.2 A comparison of the doses, calculated by AMCARE and the SCK•CEN model, arising from exposure pathways at the Tessenderlo site

Exposure pathway	AMCARE prediction mSv a ⁻¹	SCK•CEN model mSv a ⁻¹
Radon inhalation (indoor)	356	34
Radon inhalation (outdoor)	1.07	0.29
Dust inhalation	0.0003	0.002
Ingestion	1.1	1.6
External exposure	6.9	2.0
Total	365	38

It is clear from these values that there is good agreement in the dose calculated for the exposure pathways involving ingestion, external exposure and dust inhalation. However, significant differences appear to be the case for radon inhalation, particularly indoor inhalation.

The reason for the discrepancy involving the radon doses is unclear. The most probable explanation lies in manner by which radon efflux is calculated by the two models. The site-specific model calculates the radon dose by using conversion factors (²²⁶Ra to ²²²Rn) which are specific to the Tessenderlo site (and derived by direct measurement). In contrast AMCARE uses generic data to calculate the radon dose. In particular, the emanation constant (0.23) used by AMCARE is taken to be the arithmetic mean of a wide range of values (0.02 to 0.83). However, it may have been more appropriate to use lower values for the Tessenderlo site, e.g. 0.02 to 0.05 for sand [Penfold *et al.*, 1997]. This factor could well be responsible for the higher outdoor radon concentrations obtained with AMCARE.

A second factor which could have contributed to the higher indoor radon concentrations obtained by AMCARE arose from the fact that the foundations of the building were not taken into account. The reduction factor would be typically 2.7 for 10 cm of concrete [Zeevaart, 1998]. This factor combined with the difference in emanation factors, described above, would account for the differences between the two sets of values.

These assumptions are further supported by altering the parameters of the CARE model to calculate the dose due to radon if the building were constructed without penetrating the cap. In this case the radon dose would be approximately 25 mSv a⁻¹. This is close to the dose calculated by the SCK•CEN model (38 mSv a⁻¹).

B3.4 Workforce scenario

Both AMCARE and the SCK•CEN model calculate doses to the workforce. However, AMCARE deals with the workforce employed in remediating the site whilst the SCK•CEN model deals with workforces involved in the construction of buildings and roads on the site. The habits of these two groups will be significantly different and this will preclude any comparison.

B3.5 Conclusions

The comparison of AMCARE with the SCK•CEN model for the Tessenderlo site shows that, where the two models can be compared, there is generally good agreement. Both models have considered the same factors and, despite using slightly different parameters, appear to achieve

similar results. The major difference (radon inhalation in the intrusion scenario) can be explained in terms of different assumptions regarding the construction of buildings and differences in specific radon emanation coefficients.

It may be concluded that AMCARE can be partially verified with regard to the SCK•CEN model for the Tessenderlo site. However, a complete validation of AMCARE could not be performed due to the differences in parameter values used by the two models. It would also be questionable whether a realistic validation could be undertaken for AMCARE due to the fact that it is used to perform dose assessments for a wide variety of wastes.

B3.6 References

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ANNEX C1
OVERVIEW OF REMEDIATION TECHNOLOGIES
Table of Contents

C1.1	Introduction	184
C1.2	Removal of the source term	184
C1.3	Containment	185
C1.4	Immobilisation	187
C1.5	Separation	188
C1.6	References	191

C1.1 Introduction

Remediation technologies are defined for the purposes of this study as techniques or measures which reduce the radiological impact, and associated risks, to the population from contaminated sites and which may lessen the need for on-site or off-site environmental monitoring.

A wide variety of remediation technologies are available [USEPA, 1996]. Techniques most suited to the CARE project are those which are well-established, require little maintenance and which deal with wastes containing radionuclides which arise from the ^{238}U and ^{232}Th decay chains. In addition, techniques which are considered to be promising, based on laboratory or field trials, are also included for consideration here.

Remediation technologies may be divided into four major categories.

- i) 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 a subsequent separation procedure.
- ii) Containment - barriers which may be installed between contaminated and uncontaminated media to prevent the migration of contaminants include principally:
 - capping;
 - sub-surface barriers.
- iii) Immobilisation - materials added to the contaminated medium, in order to bind the contaminants and reduce their mobility, principally as:
 - cement-based solidification;
 - chemical immobilisation.
- iv) Separation - applicable to both contaminated soil and groundwater. Separation technologies, which can be carried out both *in-situ* and *ex-situ* (following excavation or removal of the contaminated medium), include:
 - soil washing;
 - flotation;
 - chemical/solvent extraction.

This review considers each of the above remediation technologies in terms of their impact on the emission of radon and contaminated dust, the mobility of radionuclides and external radiation effects. Each technology is further assessed in terms of the cost of implementation, performance, service life, workforce exposure during remediation and public reassurance.

C1.2 Removal of the source term

Conceptually, the most straightforward method to reduce the radiological impact of a site to the local population is to remove the medium containing the contaminants to a disposal site. This is appropriate in situations where contaminated material, from past industrial practices, has not been stored in a manner which meets current environmental standards. Transport to, and disposal at, a site which meets these standards will reduce overall public exposure, as well as local exposure, to the contamination.

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

Table C1.1 Summary of source removal characteristics

Applicability	Media which can be treated by these technologies include soil, vegetation, mud, tailings and buried waste. All radionuclide contaminants can be removed, including radon which will be reduced due to the removal of parent radionuclides.	
Effectiveness	Bulk removal	DF values: 1 - 20
	Soil scraping	DF values: 1 - 20
	Turf harvesting	DF values: 1 - 20
Service life	Permanent.	
Economic cost	Bulk removal	Excavation: 50 - 150 ECU m ⁻³ . Disposal (including transport): 450 - 800 ECU m ⁻³ .
	Soil scraping	Removal: 13 - 60 ECU m ⁻³ . Disposal (including transport): 450 - 800 ECU m ⁻³ .
	Turf harvesting	Removal: 9 ECU m ⁻³ . Disposal (including transport): 450 - 800 ECU m ⁻³ .
Technical feasibility	This is a mature, well-known technology. Materials and equipment are readily available	
Secondary effects	The safe disposal of potentially large volumes of radioactive contaminated waste could be difficult.	
Workforce exposure	i) inhalation of radon and fugitive dust emissions, ii) external irradiation. Estimated workforce exposure times: 0.2 - 1 manh m ⁻³ .	
Public reassurance	High. The contaminated material is removed to a more secure disposal site.	

The bulk removal of groundwater, or other contaminated liquids, is likely to be subject to many of the same considerations as bulk removal of solid material, although this option has been less well documented in the available literature. Removal of liquids is generally undertaken in more modest quantities, for a particular purpose (e.g. transfer of wastes for further treatment prior to disposal). Where continuous removal of contaminated groundwater is to be contemplated, it is likely that public reassurance will be less satisfactory since, in many instances, the source of contamination will not be removed. Furthermore the volumes to be handled, with subsequent costs, workforce dose, effectiveness etc., may prove hard to estimate. For these reasons, the bulk removal of liquid contaminated wastes is noted in principle, but is not included further within this project.

C1.3 Containment

C1.3.1 Capping

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

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

A cap usually consists of a combination of several layers of different materials. Most of the objectives of capping can be achieved with natural materials, such as layers of soil, gravel, rock or rip-rap. However, the prevention or restriction of infiltration of water is only properly achieved by introducing low-permeability layers. These may be naturally low-permeability soils such as clay [McGregor, 1994], or artificial materials such as plastics, geomembranes (e.g. PVC, HDPE, LLDPE, VLDPE, polypropylene, polyester, hypalon), geosynthetic clay liners

(GCL) (usually blankets of bentonite clay with geotextile), asphalt (asphalt-mix, asphalt concrete, asphalt rubber membrane) or cement (concrete).

Caps for radium contaminated sites are designed to delay the emission of gaseous radon until it has essentially decayed. If synthetic membrane liners are not used then the depth of cover necessary is about 1.5 to 4 m. Where radon is not of primary concern, a cap depth of 0.5-1 m will provide effective shielding for gamma radiation [USEPA, 1996]. A typical cap for containing radioactive media may thus consist of a compacted filler, a geomembrane, a layer of compacted clay, another geomembrane and several metres of top soil.

Table C1.2 Summary of capping option characteristics

Applicability	Capping can be used on a variety of solid materials including soil, mine tailings, sediment and bulk waste, and is applicable to the containment of all radionuclides including radon.
Effectiveness	Radon emissions, windblown dust emissions and external irradiation will be substantially reduced. Permeability factor: 1×10^{-12} and $1 \times 10^{-9} \text{ m s}^{-1}$.
Service life	Estimated to be in the order of 1,000 years.
Economic cost	Estimated to be between 30 to 45 ECU m^2 , mostly capital cost. Operating and maintenance costs will be low and result from monitoring and possible repair.
Technical feasibility	Most capping technologies are well developed and considered reliable.
Secondary effects	No important side effects.
Workforce exposure	Exposure to fugitive dust emissions during installation. Estimated workforce exposure times 0.03 - 0.3 manh m^2 (barrier).
Public reassurance	Low. All the contaminated material remains on the site.

C1.3.2 Sub-surface barriers

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

- *Slurry walls* consist of vertically excavated trenches that are filled with slurry which hydraulically shores the trench to prevent the collapse of the side walls during excavation and produces a barrier to groundwater flow. Slurry walls are generally a mix of bentonite and water, or Portland cement, bentonite and water [USEPA, 1996].
- *Grout curtains* may be constructed in two ways: permeation grouting or jet grouting by mixing. Permeation grouting, takes advantage of the soil's natural permeability by using pressure to inject the grout, which then flows into the soil. Jet grouting by mixing uses a rotating drill while injecting the grout. This fractures the soil and mixes it with the grout. The spacing of grout curtains (narrow, vertical, grout walls) is selected so that each "pillar" of grout intersects the next, thus forming a continuous wall or curtain. Typical grouts include Portland cement, alkali silicate grouts and organic polymers.

These technologies are primarily concerned with reducing the contamination of groundwater. They will not prevent the emission of radon and windblown dust nor will they provide shielding against gamma radiation. Consequently, sub-surface barriers are frequently used in conjunction with other technologies. In particular, they are used with capping since this would produce an essentially complete containment structure surrounding the waste mass. The summary of characteristics provided in Table C1.3 is restricted to vertical wall barriers. Where total land encapsulation is to be considered, the area to contain the waste must first be excavated, after which a liner or other impermeable material can be installed. This approach is therefore more

appropriate to primary repository design rather than to secondary remediation, and is not considered further in this project.

Table C1.3 Summary of sub-surface barrier option characteristics

Applicability	Applicable to soil, sediment, tailings and bulk waste for reducing migration of most soluble radionuclides into groundwater. However, sub-surface barriers are ineffective in preventing radon or dust emission and do not provide shielding against external radiation.
Effectiveness	Permeability factor: 1×10^{-12} - 1×10^{-8} m s ⁻¹
Service life	A reliability of between 100 and 1000 years.
Economic Cost	Slurry wall: 510 to 710 ECU m ² . Grout curtains: 310 to 420 ECU m ² . In both cases operation and maintenance costs will be low.
Technical feasibility	A well-established technology
Secondary effects	No important side effects.
Workforce exposure	Exposure to fugitive dust emissions during installation. Estimated workforce exposure times 0.06 - 0.4 manh m ⁻² (barrier).
Public reassurance	Low. The contaminated material remains on site.

C1.4 Immobilisation

C1.4.1 Cement-based solidification

Solidification processes reduce the mobility and solubility of contaminants by stabilising them into a solid matrix and may be accomplished *in-situ*, by injecting the solidifying agent directly into the contaminated material, or *ex-situ* by excavating the contaminated material and machine-mixing it with the solidifying agent. The objective is to limit the spread of contaminants via leaching and to slow the rate of emission of any radon within the solidified mass. The end product of solidification should be a monolithic block of high structural integrity. However, these technologies only offer partial shielding against external irradiation and the covering of the solidified mass with a layer of soil may be necessary [USEPA, 1996].

Cements generally used for soil stabilisation may be subdivided into three major types: i) Portland cements, ii) pozzolanic materials such as fly ash (non crystalline silica) or lime (calcium), and iii) blends of pozzolanic and Portland cement, with additives such as bentonite or other clays (to lower permeability), calcium chloride (as a setting agent and for freeze resistance during curing), lignosulfonates (as setting agents and viscosity modifiers) and gypsum (to increase strength and reduce permeability) [Malone and Lundquist, 1994].

Table C1.4 Summary of cement based solidification characteristics

Applicability	Applicable for all soil, sediment, sludge and refuse. Radon emissions will be reduced due to slowing considerably the rate of emanation.
Effectiveness	Mobility reduced by a factor of between 5 and 25.
Service life	Leach resistance is initially high, although the long term effects of weathering, groundwater infiltration and physical disturbance cannot be predicted.
Economic cost	<i>Ex-situ</i> : 75 to 300 ECU m ⁻³ (including excavation). <i>In-situ</i> : 50 to 310 ECU m ⁻³ .
Technical feasibility	A well-established technology.
Secondary effects	No important side effects although the total volume of material is increased.
Workforce exposure	i) inhalation due to fugitive dust and radon, ii) external irradiation. <i>Ex-situ</i> : estimated exposure time 0.25 to 1.5 manh m ⁻³ (including excavation). <i>In-situ</i> : estimated exposure time 0.06 to 0.4 manh m ⁻³ .
Public reassurance	Low. The contaminated material remains on site.

C1.4.2 Chemical immobilisation

Chemical immobilisation reduces the solubility of radionuclides by increasing their chemical stability through the addition (either *in-situ* or *ex-situ*) of a chemical binding agent and hence restricts migration into groundwater. However, its effect on radon and dust emissions will be variable and, in some cases, negligible. The process will not provide shielding from external irradiation and where this is of concern, it is likely that the treated waste will require capping [USEPA, 1996].

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

Ex-situ technologies require excavating the contaminated material and mechanically mixing it with the immobilising reagents. A full-scale polyethylene extruder can process material on a scale of 900 kg h⁻¹. The extruded material will cool and set within a few hours [USEPA, 1996] and the solidified material is then stored in containers, for disposal, or re-buried at the site under a sufficiently thick soil layer to reduce gamma radiation.

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

Table C1.5 Summary of chemical immobilisation characteristics

Applicability	Can be applied to most soluble radionuclides in soils, sediments, sludge and refuse: best suited to sites containing fine grain material. Chemical immobilisation is ineffective in preventing radon emission, dust emission or in providing shielding against external radiation.
Effectiveness	Mobility reduced by a factor of between 5 and 50.
Service life	The long-term effects of weathering, groundwater infiltration and physical disturbance cannot be predicted.
Economic cost	<i>Ex-situ</i> 110-570 ECU m ⁻³ (including excavation). <i>In-situ</i> 60 to 420 ECU m ⁻³ .
Technical feasibility	Studies have only been carried out at pilot scale.
Secondary effects	No important side effects.
Workforce exposure	i) inhalation of fugitive dust and radon and ii) external irradiation. <i>Ex-situ</i> Estimated exposure time 0.25 to 1.5 manh m ⁻³ (including excavation). <i>In-situ</i> Estimated exposure time 0.06 to 0.4 manh m ⁻³ .
Public reassurance	Low. The contaminated material remains on site.

C1.5 Separation

C1.5.1 Soil washing

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

- washing and rinsing [Bondar *et al.*, 1995; Bovendeur and Pruijn, 1994; Dworjanyn, 1996; Chilton and Pfuderer, 1989; USEPA, 1996];
- attrition scrubbing [Timpson *et al.*, 1994; Sadler and Krstich, 1994; Peng and Voss, 1994; USEPA, 1996; Groenendijk *et al.*, 1996];
- centrifugation [Goldberg *et al.*, 1994; Mathur *et al.*, 1996; Mista *et al.*, 1995];

- gravity separation [Elless *et al.*, 1994; Bovendeur and Pruijn, 1994; USEPA, 1996];
- hydrocycloning [Peng and Voss, 1994; Bovendeur and Pruijn, 1994; Mathur *et al.*, 1996; Nechaev and Projaev, 1996].

Where the contamination is largely associated with the fine particle fraction (e.g. uranium, thorium, lead) this process can be highly effective [USEPA, 1996].

Table C1.6 Summary of soil washing characteristics

Applicability	Applicable only where the contaminants are closely associated with fine particles. Can be used on soils, sediments and sludge, but humus soils (high organic content) can be difficult to clean, whereas sandy soils are much easier. Radon levels will be reduced due to the removal of parent radionuclides.
Effectiveness	DF values 1-10 depending on the matrix of the medium. RF values are reported to be between 50 and 98%.
Service life	Permanent.
Economic cost	150 to 500 ECU m ⁻³ for washing and separation, plus 50 to 150 ECU m ⁻³ excavation and transport costs
Technical feasibility	Not tested for radionuclide removal beyond demonstration levels.
Secondary effects	The residual soil fines and process/wash waters may contain levels of contamination requiring further treatment and/or disposal.
Workforce exposure	Excavation may lead to fugitive dust emissions. Estimated exposure times 0.25 to 1.5 manh m ⁻³ (including excavation but excluding transport).
Public Reassurance	Moderate/high. The volume of waste will be slightly reduced. Contaminated material will be removed from the site.

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

C1.5.2 Flotation

Like soil washing (Section C1.5.1, above), flotation is an *ex-situ* process to separate fine soil particles (silts and clays) from the large granular soil particles and gravel, in which the contaminated soil is first excavated then mixed with water to form a slurry. A flotation agent, which binds to the clay micelles to form a hydrophobic surface, is then added to the slurry. Small air bubbles are introduced in the solution and adhere to the hydrophobic particles and transport them to the surface. The foam is then removed from the surface for further treatment or disposal and the clean soil is returned to the excavation area.

Types of flotation reported to have been carried out, include froth-flotation [Buckley *et al.*, 1995; Bovendeur and Pruijn, 1994; Palmer *et al.*, 1995] tall column flotation and automated mechanical flotation [Mathur *et al.*, 1996]. Demonstration projects have shown that flotation is effective in removing uranium from sandstone ores containing uranium oxide and in reducing radium in uranium mill tailings [USEPA, 1996].

Table C1.7 Summary of flotation characteristics

Applicability	Applicable where radionuclides bind to fine particle materials within soils, tailings and sediments. Radon levels will be reduced due to the removal of parent radionuclides.
Effectiveness	DF values of 1 to 10, depending on the waste matrix and nuclide mix. RF values are reported to vary between 28 and 99%.
Service life	Permanent.
Economic cost	Capital cost 15 to 240 ECU m ⁻³ . Operation cost 0.8 to 3.8 ECU m ⁻³ , plus 50 to 150 ECU m ⁻³ for excavation and transport.
Technical feasibility	Studies with radionuclides have been carried out at bench scale.
Secondary effects	Residual foam generated by the flotation process may require further treatment or disposal.
Workforce exposure	Fugitive dust generation during excavation. Estimated exposure time 0.25 to 1.5 manh m ⁻³ (including excavation and transport).
Public reassurance	Moderate/high. The volume of waste will be slightly reduced. Contaminated material will be removed from the site.

C1.5.3 Chemical extraction technologies

The purpose of chemical extraction is to separate specific radionuclides from the waste matrix using a suitable solvent, thus collecting them as a concentrated solution. Separation of the solvent containing the dissolved radionuclide from the solid waste material can be an effective method for decontaminating wastes but can potentially be a complex and intensive process, requiring:

- a method for collecting the waste;
- a vessel for containing the waste;
- a means for delivering the solvent to the waste;
- a means for ensuring good contact between the solvent and the waste, and;
- a method for extracting and collecting the resulting contaminated solvent.

Most importantly, this approach is likely to be limited by the availability of a solvent which is effective for the specific radionuclide(s) of concern, the contaminated medium, and the concentration range present. Consequently, a varying degree of efficiencies may be encountered when treating a waste containing a group of radionuclides with a single solvent [USEPA, 1996]. This problem is simplified in cases where the waste contains high levels of only one radionuclide and the solvent is chosen to efficiently remove it. Therefore, the choice of solvent tends to be site-specific.

Factors which have a significant effect on the solubility of a radionuclide include:

- choice of solvent;
- adjustment to the pH of the solvent;
- addition of complexing agents;
- addition of ion exchange agents to replace the radionuclide in the solid medium;
- addition of oxidation and reduction agents.

Examples of these approaches show that inorganic salt extraction may be used to remove radium and thorium from uranium mill tailings; radium, thorium and uranium may be extracted from mineral ores with mineral acids; and EDTA can extract radium from soils and tailings [USEPA, 1996]. Similarly, lead may be extracted from spoil with an alkali solution [Palmer *et al.*, 1995].

Chemical solvent extraction reduces the radionuclide content in the waste by removing the soluble portions of each radionuclide. This will result in reduction of the flux of radionuclides into groundwater. The impact of chemical solvent extraction on radon emissions will depend on the amount of the parent radionuclides (in particular, radium) which are removed from the waste. The impact on dust emissions will be a reduced level of contaminants in the dust. External irradiation from the waste will also be reduced due to the smaller source term and, possibly, due to increased self shielding since the volume of inert waste *in-situ* will remain largely unaffected.

Table C1.8 Summary of chemical/solvent extraction characteristics

Applicability	Suitable for treating a range of radionuclides in dry soils, sediments and sludge. The resulting removal of radionuclides will reduce the flux of radionuclides into groundwater, levels of radon emissions, the level of radionuclides in fugitive dust waste and the level of gamma radiation.
Effectiveness	DF 1 - 20
Service life	Removal of radionuclides from the source will be permanent.
Economic cost	180 - 820 ECU m ⁻³ (including excavation).
Technical feasibility	This treatment of contaminated soils has been taken from bench-scale experiments to pilot-scale processes for radionuclides.
Secondary effects	Chemical extraction gives rise to liquid wastes containing high levels of radionuclides which require suitable disposal or further treatment. Where organic solvents are used, the volume may be minimised by recovering the solvent by distillation for reuse.
Workforce exposure	Excavations may lead to fugitive dust emissions. Estimated exposure times 1.2 - 3.5 manh m ⁻³ .
Public reassurance	Moderate. Some of the radionuclides will be removed from the site.

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

C1.6 References

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ANNEX D1
OVERVIEW OF REMEDIATION CRITERIA FROM
INTERNATIONAL ORGANISATIONS

Table of Contents

D1.1	Introduction	196
D1.2	The International Commission on Radiological Protection (ICRP)	197
D1.3	The International Atomic Energy Agency (IAEA)	201
D1.4	The ICRP and IAEA approaches	205
D1.5	References	206

D1.1 Introduction

So far, the recommendations from international radiation protection organisations have neither addressed radiation protection in chronic exposure situations (except for radon in dwellings) nor protection criteria for such situations, *e.g.* criteria for remediation of contaminated sites. However, recommendations on optimisation of protection and selection between protection options have been published for many years. Within the International Commission on Radiological Protection (ICRP) and the International Atomic Energy Agency (IAEA) guidance is being developed on protection of the public in chronic exposure situations and on clean-up of land contaminated with radioactive materials. The status of this work is briefly presented below. It should be emphasised that the statements found in Sections D1.1 and D1.2 are those given in the reports by the Task Group and Advisory Group.

Within the international framework for radiation protection, human activities that involve or could involve exposure to radiation can be dealt with either as practices or as intervention. A practice is defined as [ICRP, 1990]:

any human activity that introduces additional sources of exposure or exposure pathways or extends exposure to additional people or modifies the network of exposure pathways from existing sources, so as to increase the exposure or the likelihood of exposure of people or the number of people exposed.

In contrast, intervention assumes the introduction of exposures to radiation has already occurred or is presently occurring and is defined as [ICRP, 1990]:

any action intended to reduce or avert exposure or the likelihood of exposure to sources which are not part of a controlled practice or which are out of control as a consequence of an accident.

Situations involving remediation of contaminated areas may fall into either of these categories, and in some cases it may not be clear which is more appropriate. For example, the clean-up of a licensed nuclear site as part of decommissioning is clearly a part of that practice, and the clean-up of contaminated areas from a major nuclear accident would clearly be intervention. However, clean-up of contamination left behind from a previously discontinued practice may be controllable by the generator and this would be a practice.

The distinction between practices and intervention is fairly explicit, and can be summarised as follows. Any contaminated area would constitute a source. If this source, at the time when a decision on clean-up is being taken, is within an authorised practice, then any clean-up activities would be part of that authorised practice, and the radiological protection principles for practices would apply. If the source, *i.e.* the contaminated area, is not within an authorised practice, then any clean-up action will be classified as intervention, and the corresponding principles apply.

Contamination situations may be subdivided for convenience in considering the development and application of clean-up criteria into the following main categories or situations:

- (a) residual contamination post decommissioning of existing sites (existing practices, *e.g.* decommissioning of contaminated areas and installations for the nuclear industry)
- (b) residues from operations prior to regulation or under control inadequate from a present day point of view (past practices, *e.g.* contamination resulting from past uranium mining and milling operations)
- (c) residual long-term contamination following accidental release of radionuclides to the environment (accidents, *e.g.* contamination of the environment due to accidents in the nuclear industry)

With these definitions, clean-up or chronic exposure situations can readily be categorised: situations of type (a) would be part of the relevant practice, whereas those of type (b) and (c) (residues from past practices and accidents) would be intervention situations.

Chronic exposure situations that may need to be remediated can arise under a wide variety of circumstances, and the choice of the elements of the system of radiological protection that are appropriate for application to a specific decision on a remedial action will not always be straightforward. In some cases the principles of protection for practices would clearly apply; in some it will be the principles for intervention; but in others the choice will be ambiguous.

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 Publication 60, paragraph 100) [ICRP, 1990]. 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)

Task Groups and Advisory Groups within the ICRP and IAEA have since 1994 been developing guidance on radiation protection of the public in chronic exposure situations. Both the IAEA and the ICRP working groups have elaborated on the basic radiation protection principles in ICRP Publication 60.

D1.2 The International Commission on Radiological Protection (ICRP)

In 1994, on the recommendation of the ICRP Committee 4, the Commission appointed a Task Group “*to develop recommendations concerning: (a) the application and withdrawal of countermeasures in exposure situations arising from the long-term presence of radioactive materials in the environment, and (b) the management of residual exposures after the withdrawal of countermeasures*”. A new Task Group was appointed at the end of 1996 to continue the work “*to develop protection criteria for chronic public exposure covering: (a) situations where consideration is given to the suspension of countermeasures including situations where countermeasures were considered, but not applied, (b) situations of decontamination and reclamation of land that had become contaminated by past practices or past accidents, and (c) situations of unexpected high exposure to natural sources*”. The major ideas in this work are presented below.

D1.2.1 Individual- and source-related approaches

Although the main emphasis of the Commission’s System of Protection is on the source, its practical application involves a pragmatic combination of source-related assessments and individual-related assessments linked to a number of defined sources. For example, in the System of Protection for practices, the optimisation of protection involves the use of collective dose (a source related concept) supplemented by the use of dose constraints (an individual-related concept linked to a defined source). The system also includes individual dose limits; these apply to the total dose from the relevant sources, and not to the total dose from all sources. Exposures that are outside the scope of the System of Protection for practices and the deliberate medical exposures are excluded from the individual dose limitation.

However, purely individual-related approaches and consequential criteria for the total dose incurred by individuals as a result of the exposure to all sources may be deemed necessary for a number of purposes. One purpose could be to determine whether an individual dose approaches a threshold for deterministic effects or involves a too high probability of stochastic effects; in both cases radiation protection actions would seem to be required almost mandatory.

It should be noted, however, that there might be a practical problem for establishing acceptance criteria for this purpose: it may not be feasible to use total individual dose requirement through a formal regulatory system of protection. It is difficult to envisage how a source operator can control the dose delivered by other sources. Fortunately, high exposures that might approach the thresholds for deterministic effects and impose high individual risks are rare and would almost always arise from a single predominant source. Another very important purpose is to allay individual anxieties about residual exposures. Individual-related criteria should be based on total dose and *consequential* to the application of the Commission's System of Protection. They could be viewed as *complementary* to the System.

In fact, the current, fundamentally source-related, System of Protection for practices and interventions would imply a *consequential* criterion of an individual-related nature, namely the level of total annual individual dose that should not be of serious concern to the exposed individual. This criterion can be derived from the principles of the current System and somehow be viewed as *complementary* to those principles.

D1.2.2 Limitation of the total annual environmental dose to individuals

The ICRP had indicated that there would be some level of dose above which "intervention will always almost be justified" under any conceivable circumstances. The Commission's current recommendations associated this level with a risk of "serious deterministic effects" and it could also have also been linked with a very high risk of stochastic effects.

There is no direct human epidemiological data on deterministic effects from chronic exposures but information has been extrapolated from experience with protracted doses incurred in the course of radio-therapeutical procedures complemented by data from animal experiments. On the basis of the available information, the Commission has estimated the lower bound of dose-rate thresholds for a number of deterministic effects. They vary from over 400 mSv per year for a clinically significant depression of the blood-forming process, to somewhere above 150 mSv per year for opacities in the lens of the eye [ICRP, 1990; IAEA, 1996]. These estimations have been reflected in international standards on continuous annual doses for which intervention should be almost always justified.

Taking account of the presumed thresholds for deterministic effects and on the basis of the current system's principles for interventions, it would be obvious that individuals under almost no conceivable circumstances should be exposed to a total annual environmental dose that could cause deterministic effects. This would mean that the annual dose should be less *than about a hundred mSv*. Although at this level of dose deterministic effects should not be expected, the risk of stochastic effects to individuals exposed at these annual dose levels will not be acceptable.

D1.2.3 Acceptability of chronic exposures of no serious concern

Many chronic exposure situations are natural in origin or give rise to dose levels that are similar in magnitude to those experienced in many parts of the world. In fact, the *average* annual individual effective dose from natural sources including radon, in large areas of the world, are up to the order of 10 mSv if areas with elevated exposures are taken into account. This suggests that in such situations there are few grounds for concern at these *dose levels* provided they cannot reasonably be reduced or avoided.

Further information on radiation levels of concern can be gained from consideration of previous recommendations from the Commission and also from internationally agreed levels for undertaking protective action against radiation exposures in various situations. In this context, the following examples are important. The ICRP action level for radon in dwellings corresponds to a dose of 3-10 mSv in a year for simple remedial measures, while for more severe measures (*i.e.* permanent removal of people from their homes) the action levels should

be at least one order of magnitude higher. The intervention level for permanent resettlement due to exposure from deposited activity in the environment from a nuclear accident has been recommended by the ICRP (and established in international standards) to be 1 Sv in a lifetime, which corresponds to an annual average dose level of about 10-15 mSv.

From the above discussion it appears that a total environmental dose up to about some tens of mSv per annum should not represent a serious concern to an individual. Moreover, such a dose level could represent a kind of upper bound that might be to divide situations into two "classes":

- situations with total annual individual doses above this level should trigger investigations into the feasibility of reducing doses, and
- situations with total annual individual doses below this level could, depending on the situation, be considered as a normal situation of exposure to environmental "background" radiation.

The following consequential and complementary criteria could therefore be formulated:

a total individual environmental dose of about 10 mSv in a year is the highest that could be considered of no serious concern to individuals without further investigation, although annual doses up to this level may not be acceptable under all circumstances.

D1.2.4 Chronic exposure situations

A number of situations can be characterised on the basis of the major source of exposure, giving rise to chronic exposure and it is indicated below how they can be dealt with in the context of the Commission's recommendations.

(a) "Natural" sources of radiation

In a few parts of world, building materials with high concentrations of natural radionuclides have been used over generations. Dose rates from the resulting gamma radiation sometimes exceed 100 mSv per year. It is then necessary to consider how best to apply the Commission's System of Protection. The buildings already exist and therefore the exposure situation is extant and only intervention is available.

The application of the Commission's System of Protection to radon in buildings has been dealt with in Publication 65 [ICRP, 1994]. The Commission has emphasised that intervention should take place to protect the more highly exposed individuals in the population. The actions needed to reduce concentrations are usually fairly simple and only moderately expensive. The recommended range of annual effective dose from which an action level for intervention should be selected is 3-10 mSv.

(b) Residual environmental sources of radiation from past human activities

In the context of waste disposal, residues include deposits from the disposal on land of long-lived materials from previous operations such as mining and luminising works with radium compounds, and buildings that have been used for long-term storage of waste or for radium work and, subsequently, put to other uses. Residues have also been created by accidents in which radioactive materials have been dispersed in residential and agricultural areas.

Following a very severe nuclear reactor accident, significant quantities of long-lived radionuclides might be deposited in the environment leading to a long-term chronic exposure situation. However, such an accident would have invoked emergency countermeasures and in the cases where the protracted incremental annual doses was above around 10 mSv, this would have involved relocation of the affected population. In locations where countermeasures have been considered, but not taken, or have been taken and later withdrawn, the residual total environmental doses may well be higher than in normal situations.

If people are already living or working in a region of unusually high exposure, the first step is to consider the need for intervention. If the only form of intervention is the relocation of residents, it will usually be appropriate to accept moderately high exposures rather than to impose the social costs and disadvantages of relocation. It would then be inconsistent to prevent people from outside the affected area from moving in to take up residence or work. Guidance will be needed on return to and migration into an affected area. Return to the area can be treated as the withdrawal of a countermeasure. It is then a logical part of the System of Protection for intervention. The area should, however, *not* be treated as a practice on the grounds that the return in itself will result in an increase of exposure. This would introduce inconsistencies. The exposure of both returning and incoming populations should be regarded as being outside the scope of the System of Protection.

D1.2.5 Guidance on the management of chronic exposure situations

The basis for the ICRP Task Group work on developing guidance for protection of the public against chronic exposure is the System of Protection. The System would apply to (a) controlling the *increase* in the extant doses caused by the introduction (or continuation) of beneficial *practices* and to (b) determining the *reduction* of extant doses by the introduction of *intervention* with protective actions.

Most of the situations giving rise to chronic exposure are of no concern and require no further consideration. These situations include the great majority of the locations in which people live and work and in which the exposures are due to the normal range of the environmental background radiation. Exceptionally, there are locations in which the chronic exposures due to “natural” or “artificial” environmental sources are high enough to cause concern and may call for the application of the System of Protection. There are also many applications of the System of Protection to practices or intervention that leave residual sources of chronic exposure. Once the System of Protection has been fully applied, the Commission does *not* require any further action. This is because the system requires that all reasonable protection measures should already have been taken, either in the management of practices or by intervention. A total environmental dose level of about 10 mSv in a year is recommended as a level, below which, there would normally not be a need for intervention [ICRP Task Group, 1998].

Table D1.1 Existing total environmental dose levels at which intervention should be considered

Annual environmental dose level (mSv a ⁻¹)	Need for intervention
> about ten	intervention usually needed
< about ten	of no individual concern

The introduction of generic total environmental dose levels for individuals in terms of *an almost always justified level for undertaking protective actions* and a non-concern level for *allaying individual anxieties about residual exposures* does *not* imply, that such dose levels are automatically acceptable [ICRP Task Group, 1998]. The levels are meant as *trigger levels* for consideration of dose reductions. Consequently, if remedial actions are justified at dose levels below about 10 mSv in a year, the appropriate dose reduction should be found by optimisation. At dose levels of about 100 mSv in a year or higher intervention would almost always be justified [ICRP Task Group, 1998].

D1.3 The International Atomic Energy Agency (IAEA)

In 1994, the IAEA formed an Advisory Group to advise the Agency on the preparation of guidelines for developing clean-up criteria for contaminated land. Consultants have further developed this work which was published at the end of 1997 as an IAEA TECDOC [IAEA, 1997] for world-wide comments in Member States. A general framework for establishing clean-up criteria that does not differ depending on whether the situation is deemed to fall within the category of practices or of intervention has been suggested. This general framework includes the principles of radiation protection for practices and for intervention, but places them in a wider context of principles that also provide guidance for situations that do not fit well into either category. The work also addresses the problem of deciding whether the residual radioactivity after clean-up plus background may be redefined as normal. Return to normality does not necessarily imply a return to conditions before the contamination occurred - there would usually be an increased level of radiological risk. "Normality" or "normal conditions" means, in the IAEA work, that members of the public can live and/or work in the area under consideration without any restrictions associated with residual contamination. This means that no restrictions on behaviour or use of the area to control exposure, such as limiting access, preventing use of local foods, water, building materials or other resources, are necessary. When normality is redefined, the exposure to the residual contamination within the area is outside the scope of the System of Radiation Protection and should be considered as the new background.

D1.3.1 Bases for clean-up criteria

The basic radiation protection principles as recommended by ICRP in Publication 60 have been applied by the IAEA to determine whether clean-up is justified, and to optimise any clean-up actions, subject to any constraints that may be considered appropriate for the given situation. This process would determine the "end-point" for clean-up, which may be expressed in terms of the residual dose - the projected dose from use of the cleaned-up area, taking account of an appropriate range of uses. To provide some indication of the range of likely results of such detailed analyses, a set of generic criteria - referred to as clean-up levels - are proposed, based on general consideration of the justification/optimisation principles of protection, the need to protect individuals, and the acceptability of different levels of risk. Figure D1.1 shows the range of possible clean-up situations, divided into six sections or "bands", each covering approximately an order of magnitude in dose or risk. For easy reference, these are numbered from 1 (very low doses) to 6 (very high doses); each band covers approximately one order of magnitude of exposure.

Band	Need for clean-up actions if this is initial level	Acceptability of this level for release	Additional annual dose projected mSv a ⁻¹	Additional annual mortality risk a ⁻¹
6	Clean-up or prevent use	Not suitable for release (restrict use)	≈ 100	≈ 10 ⁻²
5	Clean-up or restrict use (Clean-up likely)	Release may be possible subject to regular review of situation	≈ 10	≈ 10 ⁻³
4	Clean-up decisions based on justification/optimisation	Release possible - situation may need occasional review	≈ 1	≈ 10 ⁻⁴
3	(Clean-up unlikely unless constrained)	Release likely - review only if a problem becomes apparent	≈ 0.1	≈ 10 ⁻⁵
2	Clean-up not likely to be necessary on the basis of radiological risk	Can be released without controls	≈ 0.01	≈ 10 ⁻⁶
1	Clean-up not necessary			≈ 10 ⁻⁷

Note: The doses and risks are those to an average member of the affected population, based on appropriate assumptions about use of the area, and exclude background. Exposures are assumed to be chronic, i.e. approximately constant over a period of at least a significant fraction of a lifetime. When doses are essentially due to radon, clean-up and subsequent release would normally fall above the three lower bands. The incremental lifetime risk can be found as the product of the annual risk and the number of years of exposure.

Figure D1.1 Proposed criteria by the IAEA for clean-up of contaminated areas

Band 1 with annual doses less than 10 µSv above background, represents risks that would be regarded as trivial in the vast majority of situations. Criteria for triviality of risks have been published in the context of exemption of practices and sources and clearance of materials from practices. The Basic Safety Standards specify a criterion for exemption and clearance of the order of 10 µSv a⁻¹ for the average member of the affected population.

Band 2 represents annual doses (typically tens of µSv a⁻¹ above background) in the range that would be considered acceptable as additional exposures imposed on members of the public as a result of a set of planned actions with an overall nett benefit to society, i.e. a justified practice.

Band 3 represents risks that might be considered tolerable as additional risks from a justified practice, provided that they were as low as reasonably achievable; the upper bound of Band 3 corresponds approximately to the ICRP dose limit for members of the public. Also, many national authorities have adopted dose constraints, typically between a hundred and some hundreds of µSv a⁻¹ to apply to new and/or existing practices, and international recommendations have been made about rationales for choosing constraints. These levels of

risk are low enough that they are considered acceptable in many other situations, *e.g.* occupational exposure, doses incurred over a year by "frequent fliers" from air travel.

Band 4 represents risks corresponding to doses of the order of a few mSv a^{-1} . These would not normally be considered acceptable if they were deliberately imposed on the public, but are low enough that they would be acceptable in a range of other situations, such as:

- (a) if the individuals are exposed voluntarily and receive a direct compensating benefit, *e.g.* radiation workers, then risks of this magnitude would be acceptable if they were as low as reasonably achievable;
- (b) radiation risks of this magnitude are routinely accepted from natural sources, and variations of this magnitude in levels of background radiation do not appear to influence people's behaviour.

Band 5 with doses of tens of mSv a^{-1} represents risks that would generally be regarded as unacceptable from any source (with the exception of necessary medical treatment) because the stochastic risks associated with exposures in this band are too high to be tolerated under normal circumstances.

Band 6 with doses of hundreds of mSv a^{-1} or more, represents risks (whether in terms of serious deterministic effects or a high probability of stochastic effects) that are clearly intolerable in all but the most exceptional circumstances (*e.g.* radiation therapy). Both the risk of serious deterministic effects and stochastic risks would always be so high as not to be tolerated under any circumstances.

D1.3.2 Considerations in setting criteria

In principle, consideration of situations in all bands would be based on the basic principles of protection. Methods such as cost-benefit analysis can be used to assist in seeking solutions that comply with the justification and optimisation principles. However, justification/optimisation studies based on cost-benefit analysis methods may omit certain factors that are of potentially great relevance to clean-up decisions, but are difficult to quantify in monetary terms, such as the social and political aspects of clean-up decisions. Therefore other methods, such as multi-attribute analyses, may be needed. Furthermore, considerations based on the protection of the individual may constrain optimisation (and perhaps even justification).

Each band in Figure D1.1 is categorised in two aspects - the need (or not) for clean-up if this level of exposure would result from the initial level of contamination, and the post-clean-up measures that would be implied if the situation were to be used as the end-point, indicating its possible suitability as a release level for "return to normality". The doses quoted are assumed to be chronic (*i.e.* persisting at a similar level over at least a significant fraction of a lifetime); where shorter term exposures are involved, it might be considered appropriate to use higher annual dose/risk criteria such that the lifetime risks are similar. The dose and risk levels refer to the dose/risk to an average member of the affected population, that is received additional to the level of doses from the regional natural background radiation.

The boundaries between the bands in Figure D1.1 are, to a large extent, arbitrary. In many cases existing criteria offer convenient points at which to set these boundaries, and in other cases (*e.g.* between Bands 1 and 2), the precise position of the boundary is not of great importance. However, the boundary between Bands 4 and 5 defines an important upper bound in two regards, namely that above this point:

- (a) some form of clean-up action or prohibition of use would almost certainly be required; and
- (b) the situation could never be regarded as "normal".

There is no obvious existing criterion with which to link this boundary, and therefore, it is necessary to form a judgement about an appropriate level. Internationally agreed levels for undertaking protective action against radiation exposures in various situations would here be an important reference and they are reproduced below.

The average annual individual effective doses from natural sources, including radon, are of the order of 2-4-10 mSv when areas with elevated exposures are included. The action level for radon in dwellings is recommended by the ICRP to be 3-10 mSv a⁻¹ for simple remedial measures [ICRP, 1994]. For more severe measures (*i.e.*, permanent removal of people from their homes) the ICRP recommends that the action level should be at least one order of magnitude higher [ICRP, 1994]. The exemption levels for activity in foodstuffs moving in international trade recommended by the World Health Organization/Food and Agriculture Organization Codex Alimentarius Commission [CAC, 1989] correspond to an annual committed effective individual dose of around 10 mSv a⁻¹. The assumption is here that the annual food basket contains a level of activity as recommended by the CAC or that the annual food product constituents have activity levels as recommended by the CAC. However, it is not realistic to anticipate that contamination of food would result in chronic exposure at such levels. The intervention level for permanent resettlement due to exposure from deposited activity in the environment from a nuclear accident has been recommended to be 1 Sv in a lifetime corresponding to an annual average dose level of about 10-15 mSv [IAEA, 1994; IAEA, 1996; ICRP, 1993; CEC, 1993].

From the above discussion it appears that an effective dose of about 10 mSv a⁻¹ represents an upper bound on levels that might be used as a generic maximum residual annual dose dividing contamination situations into two "classes". Situations with annual individual doses above this level would never be considered as normal, whereas some situations with annual doses below this level could, depending on the situation, be considered as normal. In cases where the residual dose is characterised as "normal" it would, henceforth, be considered "background".

Clean-up should almost always result in an annual residual dose less than the generic upper bound of 10 mSv a⁻¹. If measures leading to lower residual doses are justified, clean-up should be undertaken and the scale of the clean-up be determined by optimisation. It is conceivable that there may be rare situations, where clean-up is justified, but where the residual individual doses after clean-up are greater than 10 mSv a⁻¹. Such situations necessarily would be very severe, *e.g.* necessitating relocation of a large city or removal of all top soil (arable soil), thus stopping agricultural production.

D1.3.3 Proposed clean-up criteria

The approaches for implementation of the clean-up criteria are summarised in the form of clean-up levels shown in Table D1.2. The clean-up levels relate to the annual individual doses, to an average member of the affected population, additional to the regional level of background at the time when clean-up is considered. However, for Bands 5 and 6 (and possibly 4), the additional dose is usually large compared to this background, and so the criteria might reasonably be applied to the total dose including background if this is more convenient.

Table D1.2 Proposed clean-up levels from IAEA

Band No.	Range of annual doses (to the average member of the affected population)	Is clean-up needed?	
		With constraint	Without constraint
Band 6	>100 mSv a ⁻¹	always	always
Band 5	10-100 mSv a ⁻¹	always	almost always
Band 4	1-10 mSv a ⁻¹	almost always	usually
Band 3	0.1-1 mSv a ⁻¹	usually	sometimes
Band 2	10-100 mSv a ⁻¹	sometimes	rarely
Band 1	<10 mSv a ⁻¹	almost never	almost never

The annual doses dividing the bands can only be approximations in view of the uncertainties involved. Nevertheless, it is convenient to have single numbers to represent criteria, and considerable presentational problems may be expected if slightly different numbers be quoted in different situations. In this case, the most significant criterion is probably that dividing Bands 4 and 5. This represents a point above which clean-up would normally be expected to be undertaken in unconstrained situations, and therefore also represents a generic maximum level of residual dose that (apart from exceptional circumstances) might be considered acceptable as a new "background" level.

D1.4 The ICRP and IAEA approaches

The ongoing work on criteria for protection of the public against chronic exposure within ICRP and IAEA working groups is not yet fully complete (1998). The two approaches from IAEA and ICRP are in fact similar as they both operate with a generic individual dose level of about 10 mSv a⁻¹. This dose level should be considered as a dividing line between situations that might be considered as "normal" and situations where some remedial or protective actions should normally be considered.

The IAEA proposals on clean-up criteria in six individual dose (risk) bands can be considered as a standard on environmental quality. From analogy considerations to (a) world-wide variation in annual natural background dose, (b) action levels for radon in buildings, (c) criteria for resettlement of populations after a nuclear accident, and (d) doses implied by interdiction levels of activity in foodstuffs, it is concluded that dose levels of about 10 mSv a⁻¹ and above would normally call for consideration of clean-up. The IAEA work has been published as a TECDOC for world-wide comments in Member States [IAEA, 1997].

The ICRP work has brought some new ideas on *consequential* criteria, which should be considered as *complementary* to the current system of protection. An individual-related criterion for chronic exposure has been suggested in terms of an individual annual total dose of about ten mSv below which there should be no serious concern to the exposed people. Another criterion is suggested at which intervention almost always should be introduced at very high chronic dose levels to avoid deterministic effects.

D1.5 References

CODEX Alimentarius Commission (1989) *Proposed FAO/WHO levels for radionuclide contamination of food in international trade*. Joint FAO/WHO Food Standards Programme,, CX/FAC 89/17.

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ANNEX D2
OVERVIEW OF CRITERIA FOR REMEDIAL MEASURES IN
DIFFERENT COUNTRIES

Table of Contents

D2.1	Germany	208
D2.2	United Kingdom	210
D2.3	The United States of America	211
D2.4	Sweden	212
D2.5	Norway	212
D2.6	Denmark	213
D2.7	Finland	213
D2.8	Switzerland	213
D2.9	Netherlands	213
D2.10	Belgium	213
D2.11	Luxembourg	214
D2.12	France	214
D2.13	Spain	214
D2.14	Italy	214
D2.15	Russia	214
D2.16	Ukraine	215
D2.17	South Africa	215
D2.18	Australia	216
D2.19	Canada	217
D2.20	References	217

Criteria for remedial measures at contaminated sites in, Europe and other countries around the world, have been set although international guidance is not fully developed yet. So far, no common approach or common numerical guidance has emerged. However, national criteria have been proposed for remediating uranium mining and milling sites, sites contaminated from past practices and industrial processing of minerals where natural occurring radionuclides are concentrated in by-products.

D2.1 Germany

In certain regions of the new Federal Lands, Saxony, Thuringia and Saxony-Anhalt the levels of environmental activity, due to particular geological conditions, are above national averages. Ore mining activities conducted since the Middle Ages have caused additional contamination from waste rock and tailings. Residues from mining were heavily increased by the rigorous mining and milling of uranium ores following World War II. In addition, modern uranium and milling has been continued since the middle of the 1960s based upon post-war practices. These activities made a further contribution to the environmental exposures in densely populated and industrially used areas.

The areas, within which potentially contaminated sections or objects of former ore mining and uranium mining are situated, have been estimated to be of the order of 1,200 to 1,400 km². The Wismut GmbH is responsible for the decommissioning and the remediation of four uranium mining complexes covering an area of about 1,800 ha of which 800 ha contain several millions of tonnes of waste rock. They are also responsible for uranium mill complexes which cover an area of about 1,200 ha, of which 500 ha contain more than 130 million tonnes of tailings and a further 240 ha of waste rock dumps. It is expected that in the process of decommissioning and remediation more than 250,000 m³ concrete, 100,000 m³ stonework, 16,000 m³ wood and 60,000 tons of steel which might be contaminated and also several million tonnes of contaminated soil have to be removed.

The German Commission on Radiological Protection (GCRP) has prepared recommendations on the principles concerning the safeguard, use or release of contaminated residues from uranium mining [German Commission on Radiological Protection, 1991]. Recommendations are given for the following areas:

- (1) Release, for industrial use, of areas contaminated by uranium mining.
- (2) Use of areas contaminated by uranium mining for forest and agricultural purposes and as public parks and residential areas.
- (3) Safeguard and use of mine dumps.
- (4) Release of buildings, used for commercial or industrial purposes, for further commercial or industrial use; disposal of building debris from uranium mining and milling.
- (5) Release, for general use, of reusable equipment and installations from uranium mining.
- (6) Release of scrap from the shut-down of uranium mining plants.

The recommendations include considerations on radiological protection of the general public and workers. They are divided in *general principles* and *criteria* for release, subsequent use or safeguard.

D2.1.1 Release of areas for further use

The recommendations are applied to release of areas that have been contaminated by uranium mining. Tailing ponds and mine dumps as well as areas contaminated as a result of chemical ore treatment are explicitly excluded.

The GCRP recommends the effective dose of 1 mSv a^{-1} as a primary criterion for release of radioactively contaminated areas due to uranium mining.

The assessment of exposure should be done as realistically as possible, but be sufficiently conservative. This concerns the whole chain of assessment: relevant radionuclides, source characteristic, release/migration/dilution parameters, size of affected population groups, and possibilities of use. All relevant exposure pathways are included in these primary criteria with exception of radon exposure indoors.

A separate recommendation for indoor radon has to be regarded as a further primary criterion in this context. As a reference level for remedial measures in existing buildings or for planning of new ones, an annual average radon concentration of 250 Bq m^{-3} is recommended, which is the upper constraint of the nationally defined normal range.

Both primary criteria have to be regarded as "non-action" level. Below these levels remedial measures are not justified for radiation protection reasons, above these levels remedial measures shall be decided upon using a site-specific optimisation process.

For release of areas the GCRP recommends reference levels in terms of measurable quantities. GCRP recommends a release level for unrestricted use of contaminated areas of 0.2 Bq g^{-1} for ^{226}Ra as the essential single radionuclide of the ^{238}U decay series in radioactive equilibrium (in non-equilibrium situations the value of 0.2 Bq g^{-1} is related to the radionuclide with the highest specific activity in the soil). On the one hand, this value, without additional conditions, fulfils the dose criterion of 1 mSv a^{-1} for all exposure pathways with realistic assumptions about the possible use of such areas. On the other hand, this value is at the upper end of the range of specific activity naturally occurring in soils or building materials to avoid "remediation of nature".

Furthermore, the GCRP recommends a value of 1 Bq g^{-1} for ^{226}Ra for a release of contaminated areas for defined use, taking into account additional conditions. This value, with additional conditions, fulfils the dose criterion of 1 mSv a^{-1} taking into account only the exposure pathways for this defined use and realistic assumptions. Examples for such defined use are:

- industrial use with the restriction that the soil is covered in order to limit the local gamma dose rate to a maximum $0.3 \text{ } \mu\text{Sv h}^{-1}$; construction and use of factory-owned apartments, recreation centres and kindergartens are only permitted on those parts of the areas having a specific activity of ^{226}Ra below 0.2 Bq g^{-1} ;
- use as public garden (park) with the conditions, that the soil is covered in order to limit the local gamma dose rate to a maximum of $0.3 \text{ } \mu\text{Sv h}^{-1}$; sports grounds, playgrounds and other recreation centres are established only on those parts of the areas with a specific activity of ^{226}Ra below 0.2 Bq g^{-1} ;
- use as forestry or grassland without additional conditions.

Additionally, in case of the release of several contaminated sites within the feeding area of a single ground water horizon, the competent authority has to confirm that an annual effective dose of 0.5 mSv is not exceeded if the ground water downstream of the sites is continuously used as drinking water. For all essential radionuclides, reference levels of concentrations in water are given, corresponding to 0.5 mSv a^{-1} for drinking water for the critical group (7 Bq L^{-1} for natural uranium, 0.7 Bq L^{-1} for ^{226}Ra , 0.4 Bq L^{-1} for ^{210}Pb and 0.6 Bq L^{-1} for ^{210}Po). Since several radionuclides always occur in the water, a summation is necessary.

All new buildings on released sites have to be planned in a way which ensures, by appropriate construction, that the expected indoor radon concentration does not exceed 250 Bq m^{-3} .

The given values of soil activity can be regarded as result of a generic optimisation and below these values a release is therefore possible without dose assessment. Above these values a site-

specific dose assessment is necessary to decide whether, and if, in which way, remediation is an optimum solution and whether additional restrictions need be made.

D2.1.2 Safeguard and use of mine dumps

The recommendations are applied to mine dumps from uranium mining as well as from other mining operations. Mine dumps containing increased amounts of pyrite, leached low-grade ore dumps, tailing dumps from metallurgical and chemical processes and tailing ponds are explicitly excluded.

For any mine dumps with a specific activity for ^{226}Ra of less than 0.2 Bq g^{-1} , and for dumps of less than 1 Bq g^{-1} covering an area less than 1 ha or comprising less of then 10^5 m^3 deposited material, radiological protection measures are not required as a safeguard.

Regarding future use, these mine dumps must be given the same consideration as areas contaminated by uranium mining (see Section D2.1.1).

If the above indicated values are exceeded for the given specific activity, covered area or amount of deposited material, it should be determined which radiological protection measures should be applied, considering site-specific conditions, and which future uses are possible.

For mine dumps already being used for construction or other purposes, decisions on further procedure must be made and based on site-specific investigations of radiation exposure.

D2.1.3 Release of buildings and disposal of building material

The recommendations are applied to commercially or industrially used buildings which have been radioactively contaminated from mining activities and which should be directed to other commercial or industrial use. The recommendations are also applied to the release for conventional disposal of the debris produced when these buildings are pulled down.

The recommended criteria are based on an additional annual effective dose of 1 mSv due to uranium mining activities from all relevant exposure pathways, other than indoor radon levels. Additionally, the recommendation on indoor radon levels should be applied.

Buildings may be released for commercial or industrial use when the origin of contamination is only from waste rock or from uranium ore. When ceilings, walls and floors have been decontaminated (are cleaned so that soiling is no longer detectable, i.e. visible) and, when the local gamma dose rate in any room is less than $0.3 \mu\text{Sv h}^{-1}$, the building can be released.

Building debris can be released without restrictions for disposal if the activity concentration of the relevant radionuclide of the decay chain (*e.g.* ^{226}Ra) is less than 0.2 Bq g^{-1} . For values between 0.2 and 1 Bq g^{-1} the disposal of debris should be disposed at contaminated sites which are not intended for unrestricted release. For values greater than 1 Bq g^{-1} , the feasibility of disposal should be investigated for each individual case.

D2.2 United Kingdom

The National Radiological Protection Board has published Radiological Protection Objectives for Land Contaminated with Radionuclides [NRPB, 1998]. The publication describes Board advice on the radiological protection criteria for the change of use of land contaminated with radioactive materials from past practices. It supersedes the Consultative Document published by the Board in 1996. Although advice already exists on a number of related subjects, there is relatively little specific advice concerning the particular issue of land contamination.

Contaminated land situations which have so far been found to be of concern in the UK largely fall into of two categories.

- Sites used in the past for industrial processes (*e.g.* radium luminising, thorium extraction, mineral processing) which have caused contamination with enhanced concentrations of naturally occurring radionuclides. In some cases, the sites were contaminated decades ago and the organisations responsible have long since disappeared - others are much more recent. In some cases, the contamination is "re-discovered" by new owners or potential developers - in others it has always been known about.
- Sites, or parts of sites, that are currently licensed under the Nuclear Installations Act, but which are no longer needed by the licensee. This would include, in principle, the sites of decommissioned reactors or fuel cycle facilities or, of more topical interest, surplus areas of land owned by industrial or research facilities.

Besides the historical review of the sources of radioactive contaminated land in the UK, the new publication summarises the Board's reasons for issuing advice and gives guidance on the application of radiological protection principles and standards to contaminated land situations.

NRPB advice divides contaminated land situations into two categories: those where a change of use of the land is proposed, and those where the contamination is discovered when the exposed people and exposure pathways are already in place. The former would include the "release" of land from a licensed site (irrespective of its intended use in the public domain) as well as the more direct change of use in, for example, redeveloping industrial or derelict land. Situations in this category should be subject to the requirements applied to controlled practices; in particular the residual risks should not exceed a constraint of 10^{-5} a^{-1} , and should be ALARA (As Low As Reasonably Achievable). NRPB also proposes a lower residual risk figure of 10^{-6} a^{-1} , below which it is considered unlikely that any significant expense to reduce exposures further would be warranted. Note that in some situations, "no action" may be the optimum level of remediation.

For other situations, where the use of a site is already clearly established at the time when contamination is discovered, and hence where there is no planned action causing an increase in exposures, the cost of complying with practice criteria (in terms of money or other resources and/or the disruption to the lives of the people currently using the site) may well be so high as to do more harm than good at both a societal and individual level. Such situations need to be addressed as interventions, deciding whether there are measures that could reduce doses that would do more good than harm (*i.e.* that are justified) and, if so, applying these measures so as to do as much net good as possible (*i.e.* optimisation).

The intervention category covers a much greater range of potential doses than are allowed in 'normal practice' situations, and therefore it is more difficult to identify generic criteria, other than an "upper bound". Actions will almost certainly be justified if lifetime doses from the contamination would otherwise exceed 1 Sv. It is unlikely that options involving major resource or disruption would be appropriate unless it was expected that they would avert doses of at least several mSv in a year; less severe measures producing smaller dose savings might be justified.

D2.3 The United States of America

Radioactive materials have been produced, processed, used, and stored at thousands of sites throughout the United States. Many of these sites - ranging in size from Federal weapons-production facilities covering hundreds of square kilometres to the nuclear medicine departments of small hospitals - were, or are, presently contaminated with radioactive materials. It is estimated that about 5,000 sites in the US are contaminated with radioactive materials [Wolbarst *et al.*, 1996]. The US Environmental Agency (EPA) has recently issued a directive under CERCLA (the **C**omprehensive **E**nvironmental **R**estoration, **C**ompensation, and **L**iability

Act, otherwise known as Superfund) which deals with criteria for clean-up [Richardson, 1997; USEPA, 1997].

D2.3.1 Federal sites

Most Federal sites (mainly DOE and DoD sites) are now listed on the "National Priorities List" (NPL) which places them directly under the jurisdiction of the EPA and the States through CERCLA. CERCLA has developed general criteria that apply to all carcinogens, including radiation. In simplified form, those criteria provide that:

- sites should be cleaned up to a lifetime risk (for all reasonably plausible use scenarios) of no greater than approximately 10^{-4} (interpreted as $<3 \times 10^{-4}$), and in the case of radiation this upper bound is defined to be 0.15 mSv a^{-1} (based on $5 \times 10^{-2} \text{ Sv}^{-1}$ and an assumed 30 years typical exposure period based on anticipated maximum residence times);
- sites that cannot satisfy that criterion cannot be released for unrestricted use, but may be released under enforceable use restrictions that lead to satisfying the same risk criteria for a more circumscribed set of uses (*i.e.* an industrial or park site, under effective zoning restrictions, would not have to satisfy 0.15 mSv a^{-1} for permanent occupancy under residential use). There is an additional requirement that ground water that is an actual or potential drinking water source be cleaned up to satisfy drinking water standards, based on national groundwater policy.

In summary, as noted above, the EPA requires, as official policy, that the clean-up of radioactively contaminated sites under CERCLA meet the same requirements that all other carcinogens must meet.

D2.3.2 Non-federal sites

The situation for non-federal sites varies. For sites not licensed by the NRC, CERCLA applies if the site gets listed on the NPL, and the States have jurisdiction if it does not. This is the situation for all the radium, uranium mining, and phosphate sites, some of which are on the NPL and some not (depending on the degree of contamination). For NRC licensees the situation is at present uncertain. The NRC has recently issued a final standard for decommissioning that does not satisfy CERCLA criteria, and the EPA, up until then, had deferred to the NRC and declined to list NRC licensees on the NPL on the assumption that the NRC would manage any problem adequately. However, the NRC standard specifies 0.25 mSv a^{-1} with provisions permitting exceptions (release for unrestricted use) of up to 1 mSv a^{-1} . It also makes no provision for satisfying national groundwater protection policy. The EPA has notified the NRC that it will have to reconsider its policy of deferring to the NRC and exempting NRC sites from the NPL, but has delayed making a final decision for the time being.

D2.4 Sweden

In Sweden, no specific criteria for remedial measures at contaminated sites have been established.

D2.5 Norway

In Norway, no specific criteria for remedial measures at contaminated sites have been established.

D2.6 Denmark

In Denmark, no specific criteria for remedial measures at contaminated sites have been established.

D2.7 Finland

In Finland, activity indices have been defined for concentrations of radium, thorium, potassium and caesium in different materials in order to restrict external γ -doses. These indices have been derived for the following materials from dose limits for exposure of the population [Zeevaert *et al.*, 1997]:

- building materials: 1 mSv a^{-1}
- materials for streets and playgrounds: 0.1 mSv a^{-1}
- landfill: 0.1 mSv a^{-1}

Activity indices have also been derived for peat-ash, based on a dose limit of 1 mSv a^{-1} for workers.

D2.8 Switzerland

In Switzerland there are no specific criteria for restoration of contaminated land. However, according to the Swiss Legislation on Radiological Protection [Swiss Regulation, 1994] it is permitted for solid radioactive waste materials with specific activities less than defined exemption limits to be discharged into the environment with the approval of the licensing authority. A further condition is that the materials are blended with non-active materials in such a way as to ensure that local dose rate at 10 cm from the surface after subtraction of the natural background does not exceed $0.1 \mu\text{Sv h}^{-1}$. Examples on Swiss exemption limits are 400 Bq kg^{-1} for ^{238}U and 700 Bq kg^{-1} for ^{137}Cs [Swiss Regulation, 1994].

D2.9 Netherlands

In the Netherlands, no official criteria for remedial measures at contaminated sites have been established. However, a general approach is adopted based on a maximum permissible individual mortality risk of 10^{-6} a^{-1} for a particular activity or substance. Two sites with radioactive material in soil have been remediated. In both situations a value of 100 Bq kg^{-1} was used as a dividing line between contaminated and uncontaminated soil [Zuur, 1997].

D2.10 Belgium

Site-specific criteria have been proposed in a SCK•CEN study on restoration of the environmental contamination with ^{226}Ra in Olen and Geel. The study was undertaken on behalf of the Federal Government and the Service for Protection against Ionising Radiation. The criteria are expected to be accepted for those sites for which the action levels for remedial measures are given below [Vanmarcke, 1997]:

- disposal site: $0.2 \mu\text{Sv h}^{-1}$ for the first metre and $0.4 \mu\text{Sv h}^{-1}$ for deeper layers if covered by one meter of clean soil
- river bank: $0.15 \mu\text{Sv h}^{-1}$
- streets: $0.2 \mu\text{Sv h}^{-1}$
- houses and building grounds: removal until natural background levels

In total, about 135000 m^3 of material are expected to be remediated [Vanmarcke, 1997].

D2.11 Luxembourg

No criteria have been established for restoration of areas affected by lasting radiation exposure from historic practices [Feider, 1997]. Only one site during the past decade has been restored. This was a small industrial company using ferro-niobium ores and producing slag with a high ^{226}Ra content (up to $100,000 \text{ Bq kg}^{-1}$). Over 21,000 tonnes of this slag were deposited in a non-radioactive waste disposal site and covered with 2.5 metres of soil.

D2.12 France

According to French regulation (Decree 66450 of 20 June, 1966), it is the responsibility of the Ministry of Health to set levels, above which, a material is considered to comply with regulatory requirements on radiological protection (Article 30) [Sugier, 1998]. The approach is used on a case by case basis. In each case examined the Ministry have set a concentration level (not a dose level) including mandatory control conditions. Until now the concentration values selected were about $5,000 \text{ Bq kg}^{-1}$.

D2.13 Spain

The regulations and standards that govern the remediation activities at the Andujar mill tailings have been established by the Spanish Nuclear Safety Council (CSN). This takes into account the recommendations of international organisations, the standards given by the US EPA for the remediation of uranium mill tailings, and the Spanish regulations, especially those related to groundwater protection and the long-term disposal of radioactive wastes. These regulations can be summarised as [Cancio, 1998]:

- long-term radiation protection: achieve an effective equivalent dose to individuals, in the critical group below 0.1 mSv a^{-1} ;
- soil clean-up: reduce the residual concentration of ^{226}Ra on land, averaged over an area of 100 m^2 , so the background level is not exceeded by more than 0.2 Bq g^{-1} (averaged over the upper 15 cm soil) and is less than 0.6 Bq g^{-1} (averaged over 15 cm-thick layers of soil more than 15 cm below the surface).

The authorisation has been given by a ministerial order from the Ministerio de Industria y Energia, No. 3181, BOE 31 of February 1991.

D2.14 Italy

In Italy, no specific criteria for remedial measures at contaminated sites have been established [Belli, 1998].

D2.15 Russia

Criteria have been established in Russia for sites contaminated from nuclear accidents, nuclear weapons testing, nuclear weapons production and the nuclear fuel cycle. The areas include those contaminated by the Chernobyl accident, the Semipalatinsk site contaminated by nuclear weapons testing, and the sites contaminated by production of weapons plutonium by the Mayak enterprise. The following criteria have been proposed for remediation and restoration measures [Zeevaert, 1997]:

- remedial measures are justified if the monetary costs are less than $2 \times 10^4 \text{ roubles manSv}^{-1}$ ($4 \times 10^4 \text{ USD manSv}^{-1}$) and not justified if the monetary costs are greater than $4 \times 10^4 \text{ roubles manSv}^{-1}$ ($8 \times 10^4 \text{ USD manSv}^{-1}$). Additional data are needed if the monetary costs are within the interval $2 \times 10^4 - 4 \times 10^4 \text{ roubles manSv}^{-1}$ ($4 \times 10^4 - 8 \times 10^4 \text{ USD manSv}^{-1}$);

- after implementation of the remedial measures individual doses to members of the critical group should be less than 1 mSv a^{-1} ;
- for individual doses in the range of $1 - 50 \text{ mSv a}^{-1}$ voluntary relocation can be introduced.

The cost levels for justification of remedial measures as indicated above are based on an exchange rate between roubles and USD at the 1980-level, *i.e.* $2 \text{ USD} \approx 1 \text{ rouble}^{-1}$.

D2.16 Ukraine

In the Ukraine, criteria have been established for remedial measures in areas that were contaminated by the Chernobyl accident. If the annual effective doses to the population living in the contaminated areas exceed 1 mSv a^{-1} remedial measures should be introduced (Ukrainian law). The following scheme has been proposed by the Ministry [Zeevaert, 1997]:

- when individual doses, mainly from external exposure from deposited activity, are in the range of $1 - 5 \text{ mSv a}^{-1}$, decontamination of 'hot spots' should be introduced (*e.g.* soil removal);
- when individual doses, mainly from internal exposure from contaminated foodstuffs, are greater than 2.5 mSv a^{-1} , agricultural countermeasures should be introduced;
- when individual doses from both external and internal exposure are in the range of $1.5 - 2.5 \text{ mSv a}^{-1}$, decontamination of hot spots and agricultural countermeasures should be introduced;
- when the external dose rate in contaminated settlements exceed $1.5 - 2 \mu\text{Sv h}^{-1}$ decontamination of the settlements should be introduced.

The remedial measures include soil removal, washing, asphaltting, chemical fixation, filtering of milk, and change of agricultural production.

D2.17 South Africa

The Council for Nuclear Safety (CNS) in South Africa has prepared the Nuclear Energy Act (1993). This Act applies from 200 Bq kg^{-1} of each radionuclide, *i.e.* if the soil is contaminated at, or above 200 Bq kg^{-1} , the owner of the land must apply for a licence. This does not mean that a licence is necessary in all cases, since exemption may also be available. The CNS takes 80 Bq kg^{-1} as a reference background concentration level, which corresponds to an upper bound in the variation of the undisturbed background concentration [Petr, 1998].

D2.17.1 Unconditional clearance

If an existing licensed facility is being closed, *i.e.* the licence will not apply after closure, it is a requirement that all contamination above background be removed, as long as the background before starting the facility operation is known. If it is not known, the facility is requested to remove all contamination above the reference background. In some cases such a request may be associated with enormous costs and an optimisation must be applied. However, a constraint on the optimisation of 200 Bq kg^{-1} should be applied. Consequently, if a facility land is contaminated to, say, 400 Bq kg^{-1} , and a valid justification shows that it would not be viable to decontaminate to the background reference level of 80 Bq kg^{-1} , the land should be decontaminated to a level of at least 200 Bq kg^{-1} for satisfying the requirement for an unconditional clearance.

D2.17.2 Conditional clearance

If unconditional clearance is not possible, the land could be decontaminated to a level acceptable to conditional clearance. Such an approach could be accepted, *e.g.* for construction of a new identified facility, such as factory, store, shopping centre, etc. The land may also be permitted to be used for road or airport construction, parking lot, or similar use. However, the purpose to which the land is intended to be used, must be stated. The clean-up criteria appropriate to a conditional clearance are established on a case by case basis and derived from the magnitude of the individual and collective doses pertinent to the most conservative scenario within the envisaged application(s). The new use of land may typically involve public groups and workers.

If the new use of land involves the public, the maximum individual dose to a member of an identified group must not exceed $100 \mu\text{Sv a}^{-1}$ and the collective dose must not exceed 1 manSv a^{-1} . It should be emphasised that this incremental dose is due to the residues that remained after clean-up, *i.e.* the dose above the “old” (or reference) background level. If the new use of land involve workers, an individual dose limit of 1 mSv a^{-1} should be respected in the working area, where the public access is restricted. Conditional clearance of land automatically stipulates restriction on deed regarding future land use.

D2.18 Australia

Australia established criteria in 1990 for the rehabilitation of former British nuclear test sites in Australia [Lokan, 1998]. At two of these sites, Emu and the Monte Bello Islands, there was little need for remediation. However, at Maralinga, several locations were contaminated with plutonium that had been dispersed locally by chemical explosions.

Following extensive experimental studies at Maralinga, it was established that the inhalation of respirable plutonium contaminated dust by a critical group of Aborigines, living a semi-traditional life-style, was the dominant pathway for exposure in most cases. A second important pathway was the incorporation of plutonium, by way of wound contamination, at least in areas where many plutonium-contaminated fragments or particles were to be found.

The general criterion for the clean-up was to undertake remedial measures (interventions) to ensure that annual effective doses to the critical group under conditions of full-time occupancy should not exceed 5 mSv. The interventions took several forms.

- Removal of soil from areas where the ^{241}Am exceeded 40 kBq m^{-2} (*i.e.* about 300 kBq m^{-2} of plutonium as the plutonium to americium ratio is about 8), with a restriction on land use which prohibits camping but allows access for hunting or transit. This figure was based on observations of the likely proportion of time to be spent in the area on allowed activities. Approximately 2 km^2 of soil has been removed from the most contaminated areas with the required end state being that after clean-up:
 - the residual americium activity should not exceed 3 kBq m^{-2} , averaged over a hectare;
 - no particles or fragments should remain with activities greater than 100 kBq of americium;
 - not more than 0.1 particles per m^2 should remain with activities above 20 kBq of americium.
- An outer boundary, marked by heavy-duty galvanised steel posts at 50 m intervals, warns that camping is not permitted within the area. These warning signs generally follow the road system and contain all areas where continual occupancy would lead to doses in excess of 5 mSv a^{-1} .

- Twenty-one pits, containing unknown quantities of plutonium, but possibly with an overall content of about 2 kg, will be immobilised by *in-situ* vitrification.

The removed soil has been buried nearby in a 200 m × 100 m × 15 m pit with a 5 m cover of uncontaminated rock and soil.

The actual clean-up at Maralinga began with site preparations at the beginning of 1996, and is expected to be completed before the end of 1999.

The criteria and policies were developed in the period prior to the publication of ICRP-60 [ICRP, 1990] and are not, therefore, expressed in terms of an optimised intervention. The outcomes, though, are quite consistent with current ICRP intervention philosophy.

D2.19 Canada

The Atomic Energy Control Board (AECB) does not currently have any prescriptive values for clean-up and restoration of contaminated lands. In the recent past, the AECB has accepted values proposed on a site by site basis for the clean-up of lands contaminated by radium, uranium or both. As a matter of principle, the AECB target level for restoration of lands would be the background level for that site. In the absence of baseline data or in the event that background is not reasonably achievable, the owner of a contaminated site would need to justify the residual contamination scheduled to remain on site. This has in the past required a conceptual pathway analysis [Stenson, 1998].

In Canada restoration of areas affected by historic contamination is carried out by the Low-Level Radioactive Waste Management Office (LLRWMO). Many of the restoration projects undertaken by the LLRWMO are not licensed by the AECB. The contamination at these sites falls below the AECB licensing limits. At such, the level to which the clean-up is performed is dictated by the intended use of the site and the priorities of the involved parties (land-owners, local citizens, local governments, etc.)

If the site were licensed by the AECB, the clean-up criteria would need to be accepted and verified by the AECB before the license could be revoked. In the past these levels have been as shown in Table D2.1 below.

Table D2.1 Criteria used by AECB in Canada for contaminated sites

<i>Licensee</i>	<i>Contaminant</i>	<i>Criteria</i>
Eldorado Nuclear	²²⁶ Ra	0.2 Bq g ⁻¹
	Uranium	35 µg g ⁻¹
LLRWMO	²²⁶ Ra	0.1 Bq g ⁻¹
	Uranium	30 µg g ⁻¹

These levels are used as guidelines for current assessments, but are not prescribed values for new work. Canadian *Surface water Guidelines* for ²²⁶Ra and uranium are 1 Bq L⁻¹ and 0.02 mg L⁻¹, respectively.

D2.20 References

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ANNEX D3
JUSTIFICATION AND OPTIMISATION OF PROTECTION
Table of Contents

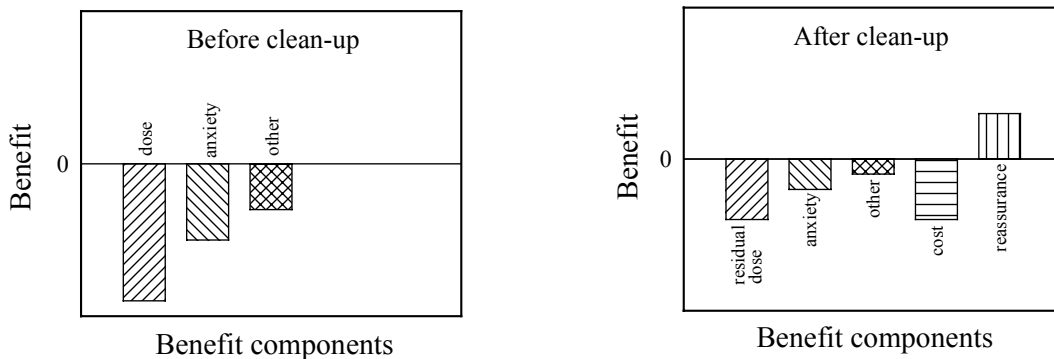
D3.1	Introduction	220
D3.2	Justification	220
D3.3	Optimisation	221
D3.4	Techniques for optimisation	222
D3.5	References	229

D3.1 Introduction

The principles of radiation protection are based on the so-called justification and optimisation principles. When the subject is protection of the public against radiation exposure from contaminated land the justification/optimisation procedure is applied to the remedial or protection action for reducing this exposure. A short review of the justification and optimisation principles is given below.

D3.2 Justification

Clean-up of contaminated land will introduce some benefit to the affected populations. The components of benefit will include, for example, averted dose and decrease in anxiety. Without any clean-up the benefit components will all be negative as shown in the left picture of Figure D3.1. After a clean-up has been implemented some of these negative benefits have been reduced or even removed but other negative benefits and positive benefits have been introduced as shown in the right picture of Figure D3.1.



Note: The left picture shows that the benefit components are all negative. The right picture shows that clean-up will reduce (or remove) some of the negative benefits, introduce new negative benefits (e.g. costs) and positive benefits (e.g. reassurance). The component 'other' includes negative benefit components such as social disruption.

Figure D3.1 Components of benefit of clean-up operations.

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

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

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

Some *decision-aiding* techniques available for carrying out decision analysis have been described in detail in ICRP Publication No. 55 [ICRP, 1989]. The primary objectives of these techniques are to identify the various factors influencing the decision, to quantify them if this is reasonable and desirable, and to systematically examine the trade-offs between them. This provides a process which be made open to the people responsible for making the decision and to public scrutiny.

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 the multi-attribute utility analysis. The factors to be used in such an analysis are quantifiable to a greater or lesser extent. The more quantifiable factors include the avertable individual and collective risks from exposure to radiation for the members of the public, the individual and collective physical risks to the public caused by the clean-up, the individual and collective risks to the workers carrying out the clean-up, and the monetary cost of the clean-up. The less quantifiable factors, including the reassurance provided by the clean-up (taken as a decrease in anxiety), and the resulting individual and social disruption, are also factors relevant to the decision.

In analysing the inputs to the decision, it is necessary to decide on the relative importance or weight of each factor. These judgements have to be made irrespective of the decision-aiding technique used. The resultant decision will be the same provided that the database is the same and the judgements are consistent. If multi-attribute utility analysis is the technique used, then all the relevant factors can be directly included in the analysis by deriving or assigning utility functions to them, but weights still need to be assigned.

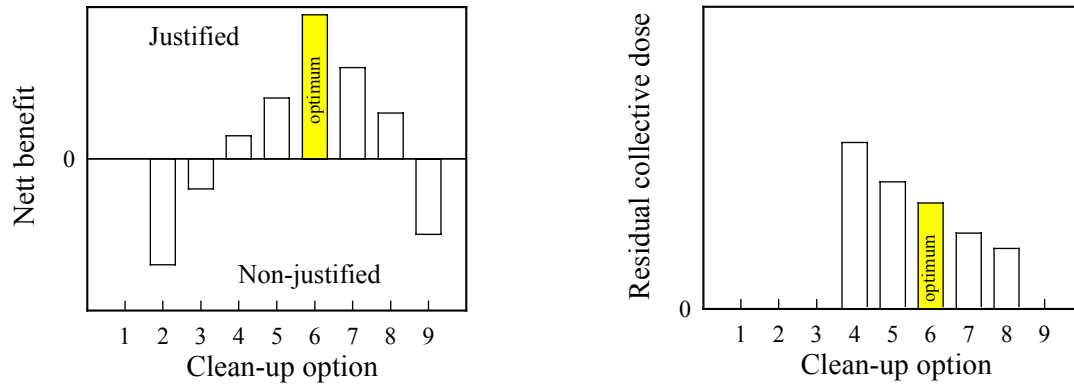
The nett benefit, ΔB , of a clean-up operation will depend on several factors (attributes), e.g. avertable collective dose, ΔS , individual dose, E , monetary costs of a clean-up operation, X , anxiety of the contamination, A , reassurance by the clean-up, R , etc. Thus the nett benefit, ΔB , is a function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

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

The individual dose, E , is often taken as the dose to the average member of the affected population. Depending on the clean-up option, collective dose may be reduced with or without changing the specified individual dose, E . Also, the affected population may change depending on the clean-up option. Thus it may be useful first to examine the effects of various levels of individual dose within a single option and among all options.

D3.3 Optimisation

Normally, there would be a range of justified clean-up options for which the nett benefit would be positive. The optimum clean-up option would be the one for which the nett benefit is maximised, as shown in the left picture of Figure D3.2. There might be justified options with a lower residual dose than at the optimum. This is due to the fact that some of the negative benefit components entering the optimisation process would have a higher weighting than averted dose. This is illustrated in Figure D3.2 where both Options 7 and 8 are justified but give a smaller nett benefit than the optimised Option 6. The optimum clean-up option is the one among all the *justified* options which has the largest nett benefit as shown in Figure D3.2.



Note: The left picture shows that there is a range of options, both justified and non-justified. The right picture shows the residual collective dose after clean-up for the justified options. The optimum solution is not necessarily the one with the lowest residual collective dose, because there are additional considerations for determining the nett benefit.

Figure D3.2 Nett benefit of different clean-up options and the corresponding residual collective dose after clean-up.

Clean-up is therefore optimised when the nett benefit is maximised, *i.e.* when $\sum \Delta b_i$ is maximum. Most of the methods used in optimisation of protection tend to emphasise the benefits and detriments to society and the whole exposed population. Optimisation of clean-up, whether it is considered in the context of a practice or for intervention, is essentially an identical process: choosing the course of action which results in the maximum nett benefit, considering all the relevant factors that influence the advantages and disadvantages of the clean-up operation.

For clean-up of contaminated land, the society usually requires that the same level of protection be provided regardless of the source of exposure. Therefore, clean-up criteria that do not differ depending on whether the situation is falling in the category of practices or interventions are desirable, but may in special cases be difficult.

The concept of optimisation of protection is practical in nature. Optimisation provides a basic framework of thinking - that it is proper to carry out some kind of balancing of the resources put into protection, and the level of protection obtained. The reduction in dose can only be achieved by the expenditure of some effort and by allocating additional resources. In such cases, it is necessary to decide whether the dose saving that is likely to result is worth the effort of achieving that saving. This is entirely consistent with the optimisation principle. In the optimisation process, two categories of radiological factors can be distinguished. The first category comprises the factors (attributes) that will always have to be included in the analytical procedure, particularly the cost of protection and the collective doses. The second category comprises the factors that may not always be necessary, such as the individual dose distribution, the time distribution of doses, the population receiving the doses, the possibility of options, etc. When all attributes that need to be considered have been specified, it may be that some of them cannot be appropriately quantified for inclusion in the analytical procedure. In this case, these factors will have to be assessed qualitatively, but the results of the qualitative analysis must be taken into account in reaching the optimum.

D3.4 Techniques for optimisation

When the performance and costs of all the protection options have been assessed, a comparison is needed to define the *optimum* protection option. When the optimum is not self evident, the comparison can be carried out using a quantitative decision-aiding technique. The result of the application of the quantitative techniques is known as the *analytical solution*. If there are non-

quantified, radiological protection factors to be taken into account, the analytical solution is *not* the optimum solution. The qualitative factors will have to be combined with the analytical solution to give the true optimum. Of the different techniques available three will be described below. These are cost-effectiveness analysis, cost-benefit analysis and multi-attribute utility analysis.

D3.4.1 Cost-effectiveness analysis

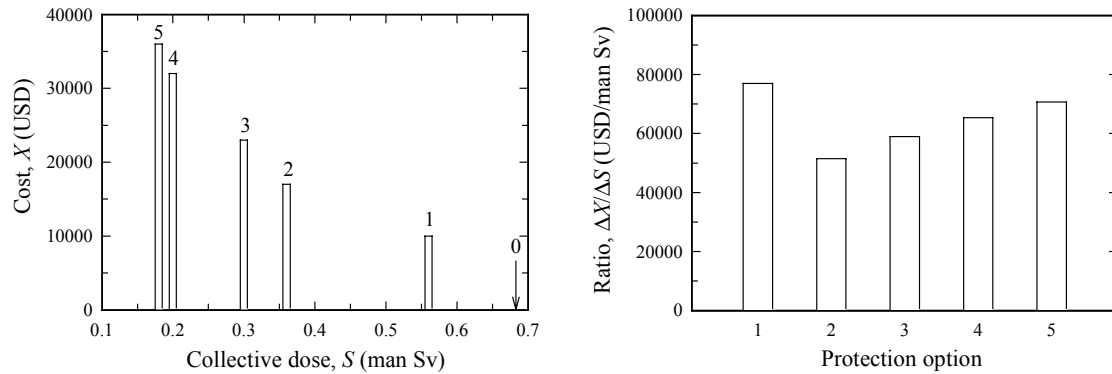
In cost-effective analyses, only two factors can be included in the quantitative analyses, namely monetary cost of the different protective measures and the individual and collective dose reduction from those measures. However, a cost-effectiveness analysis does not result in an optimisation of protection, since it does not involve the trade-off between protection costs and collective dose reduction. A cost-effectiveness analysis is rather a method to determine the best protection strategy obtainable from fixed resources. Cost-effective analyses are carried out when a specific dose reduction or when the amount of money available for radiation protection is fixed. In this case, the nett benefit, B , will be maximised by either varying the monetary costs, X , with the detriment costs, Y , as a constant, or varying Y , with X as a constant.

Cost-effectiveness analyses can, therefore, only define either the least costly way of achieving a specified reduction in exposure or the maximum reduction in exposure that can be attained for a fixed cost. It cannot optimise radiation protection. Cost-effectiveness analyses may, however, allow the *a priori* exclusion of available protection options and thus precede and simplify the formal optimisation analysis. For illustration of the cost-effectiveness methodology the data in Table D3.1 has been used.

Table D3.1 *Collective doses and costs of protection for five protection options and for the reference case without protection.*

Protection option	Monetary costs [USD]	Collective dose [manSv]
No protective measures	0	0.69
Option 1	10,000	0.56
Option 2	17,000	0.36
Option 3	23,000	0.30
Option 4	32,000	0.20
Option 5	36,000	0.18

It can be seen from the figures in Table D3.1 that the collective dose, S , decreases gradually when more efficient protection options with increasing cost, X , are being implemented. This can be seen in Figure D3.3 where the costs are plotted against collective dose (left-hand picture). The ratio $\Delta X/\Delta S$ is shown at the right-hand picture for each of the protection Options 1 - 5.



Note: The option marked "0" shown at the left-hand figure is the reference case without any protective action for which the residual collective dose is 0.69 manSv. The cost-effectiveness ratio $\Delta X/\Delta S$ is shown at the right-hand figure, where ΔX is the change in cost and ΔS the change in collective dose, both compared to the reference case. As $X_0 = 0$ it follows that $\Delta X_i = X_i$, and $\Delta S = S_0 - S_{residual}$.

Figure D3.3 Protection options in terms of monetary costs and residual collective dose.

It appears from Figure D3.3 that protection Option 2 is the most cost-effective because this option has the lowest monetary cost per collective dose reduction.

D3.4.2 Cost-benefit analysis

Cost-benefit analysis involves a balancing of costs in order to establish optimum levels of radiation protection. Optimisation of protection can be generally limited to the selection of the best available combination of cost of radiation protection, X , and cost of detriment, Y , by minimising the sum ($X + Y$) and thus maximising the nett benefit, B . The optimisation condition is fulfilled at a value of collective dose, S_{opt} , such that the increase in the cost of protection per unit collective dose balances the reduction of radiation detriment per unit collective dose:

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

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

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

$$l = R \cdot L \quad (\text{a} \cdot \text{Sv}^{-1})$$

giving a value of l of approximately 1 year per sievert.

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

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

For rich European countries the value of GNP is of the order of 25,000 USD a⁻¹, which would give a reference value of α between 25,000 USD Sv⁻¹ and 250,000 USD Sv⁻¹. The Nordic radiation protection authorities have recommended a maximum value of α of 100,000 USD Sv⁻¹. For illustration of the cost-benefit methodology the data in Table D3.2 have been used. The cost and collective dose data are identical to those used in the cost-effectiveness analysis.

Table D3.2 *Collective doses and costs of protection and radiation detriment for five protection options and for the reference case without protection.*

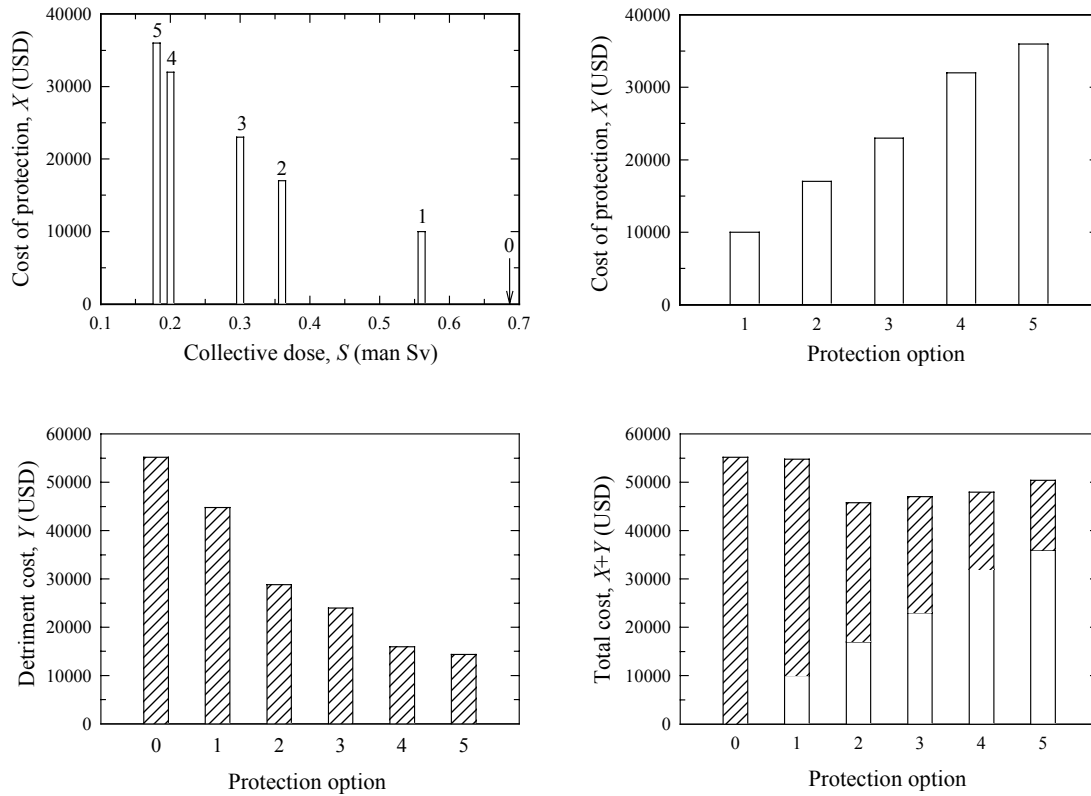
Protection option	Monetary costs [USD]	Collective dose [manSv]	Detriment costs [USD]
No protective measure	0	0.69	55,200
Option 1	10,000	0.56	44,800
Option 2	17,000	0.36	28,800
Option 3	23,000	0.30	24,000
Option 4	32,000	0.20	16,000
Option 5	36,000	0.18	14,400

In addition to the monetary costs for the different protection options the equivalent monetary cost of the detriment, Y , are presented in Table D3.2. This cost component is calculated as:

$$Y = \alpha \cdot S_{\text{residual}}$$

where α is the equivalent monetary cost of a unit collective dose.

The upper left-hand and right-hand pictures in Figure D3.4 show the protection costs as a function of the residual collective dose, S_{residual} , and for each of the protection options. The lower left-hand picture in Figure D3.4 shows the detriment costs, Y , for the protection options, including the reference case without protection. An α -value of 80000 USD manSv⁻¹ have been used for the calculation of detriment cost. The lower right-hand picture shows for each option the sum of the protection costs and the detriment costs. It appears that Option 2 has the lowest total cost and should therefore be considered as the optimum protection option. This conclusion can also be found by considering the differential cost, dX , per unit differential dose saving, dS , moving successively from Option i to Option $i + 1$ when the options are ranked in terms of increasing monetary costs. Calculating the value of dX/dS between successive options, the optimum option will be the one just before the numerical value of dX/dS equals or exceeds the value of α . In the example given in Table D3.2 the numerical value of dX/dS would exceed α when moving from Option 2 to 3 ($dX/dS = 100000$ USD manSv⁻¹), and Option 2 would therefore be the recommended option.



Note: The option marked “0” shown at the left figure is the reference case for which the residual collective dose is 0.69 manSv without any protective action. The protection costs, the detriment costs and the total costs are shown at the following figures for each of the protection options. The detriment cost, Y , is calculated as αS with a value of α equal to 80,000 USD per manSv.

Figure D3.4 Costs and residual collective dose for different protection options.

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

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

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

D3.4.3 Multi-attribute utility analysis

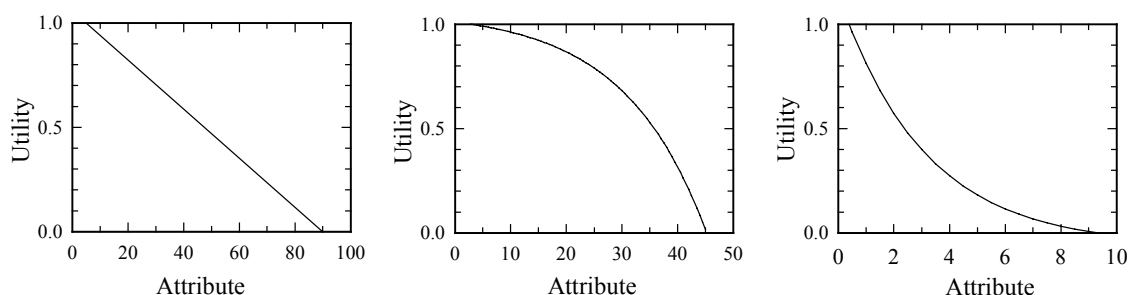
The essence of *multi-attribute utility analysis* is to use a scoring scheme (or multi-attribute utility function) for the relevant factors. If the score (or utility) is the same for two options there is no preference for one or the other and both options are, therefore, optimal. As a basis for comparison between options or alternative strategies, a *multi-attribute* value function approach can be used. There are two major components of such a value function: the evaluations of each of the alternative strategies with respect to the considered attributes, known as the *scores* or *utilities*, u and scaling factors which reflect the relative importance of each of the attributes, known as the *weights*, w . The use of utility functions allows introduction of factors which are not easy to quantify in monetary terms as is required in cost-benefit analysis. The utilities and weighting

factors can be expressed in an additive form to give an overall evaluation of each of the alternative strategies, i , or options:

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

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

The aim of scoring is to assign values to each alternative, reflecting the contribution to the overall evaluation from their performance on each end-attribute (sub-attribute). One way of defining the scores (utilities) is to assign a score of 100 to the alternative which performs best on a particular attribute and a score of 0 to the least appropriate alternative. 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 a third point (in addition to the points 0 and 100) is required to characterise the single utility function, $u(x)$. Depending on the relative position of the three points, the general shape can be determined as a linear, concave or convex function, either as increasing or decreasing functions. Functions of the decreasing type are shown in Figure D3.5 below.



Note: The left figure illustrates a risk neutral utility function (linear); the middle figure illustrates a risk averse utility function which decreases faster nearer the worst consequences being more sensitive to variation at the upper end of the range of consequences; the right figure illustrates a risk prone utility function which decreases slower at the upper end thus being less sensitive to variation at the upper end of the range of consequences.

Figure D3.5 Examples of utility functions of the decreasing type.

The data used for the cost-effectiveness analysis and the cost-benefit analysis regarding the monetary costs of protection, X , and for the collective doses, S , have also been used for the multi-attribute utility analysis. The attributes are shown in Figure D3.6.

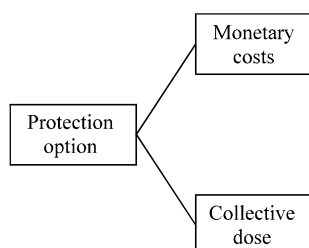


Figure D3.6 Attributes (criteria) hierarchy used in the multi-attribute utility analysis.

The utilities, u , for the attributes *monetary costs* and *collective dose* for each protection option have been determined from risk neutral utility functions, $u(x)$, where x describes the value of

the attributes for the different options. For the monetary costs and the collective dose the utility functions has been determined from:

- monetary costs: $u(0 \text{ USD})=1$ and $u(36,000 \text{ USD})=0$
- collective dose: $u(0.18 \text{ manSv})=1$ and $u(0.69 \text{ manSv})=0$

The utility functions, $u(x)$ can thus be expressed in the following way:

$$u_{\text{cost}}(x) = 1 - \frac{1}{36,000} \cdot x \quad \text{for } 0 \leq x \leq 36,000 \text{ USD}$$

$$u_{\text{dose}}(x) = 1 + \frac{0.18 - x}{0.69 - 0.18} \quad \text{for } 0.18 \leq x \leq 0.69 \text{ man Sv}$$

and they are shown in Figure D3.7.

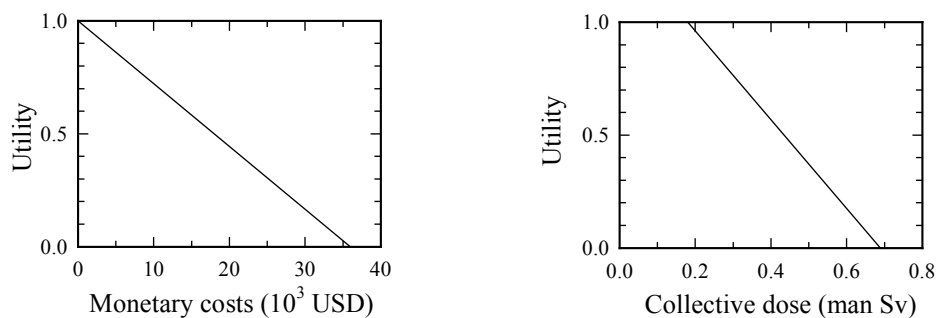


Figure D3.7 Utility functions for the attributes 'monetary costs' and 'collective dose'.

The utilities for each attribute and each option have been determined from the utility functions given above and the utilities are shown in Table D3.3.

Table D3.3 Utilities or scores, $u(x)$, for five protection options and for the reference case without any protection.

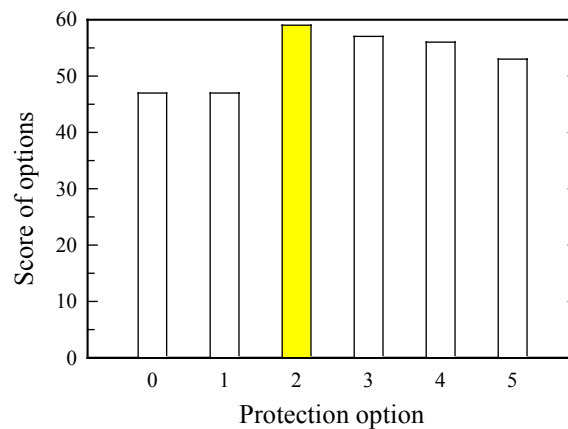
Protection option	Monetary costs	Collective dose
No protective options	1	0
Option 1	0.72	0.25
Option 2	0.53	0.65
Option 3	0.36	0.76
Option 4	0.11	0.96
Option 5	0	1

The weighting factors, w , have been determined in the following way. If the ranges of the monetary costs and collective dose are called $R(X)$ and $R(S)$, respectively, the weighting factors can be obtained by constraining them to the same imposed criterion as for the cost-effectiveness and cost-benefit analyses described in Sections D3.1 and D3.2 as:

$$\frac{w(X)}{w(S)} = \frac{R(X)}{\alpha \cdot R(S)}$$

and then normalising so that $w(X) + w(S) = 1$. This gives the values $w(S) = 0.53$ and $w(X) = 0.47$.

The overall score of the different protection options, U_i , has been calculated as the sum of the products of weighting factor and utility. The results are shown in Figure D3.8. It appears that Option 2 comes out with the highest score and this protection option would thus be the optimum solution.



Note: Values of utilities for the attributes 'monetary costs' and 'collective dose' as shown in Table D3.3 and weighting factors of 0.47 and 0.53 for these attributes, respectively.

Figure D3.8 Overall evaluation of the five different protection options

It should be emphasised that the specification of the values of the different factors and attributes entering the analysis determines the outcome, and *not* the technique used. Therefore, it is to be expected that the optimum results using different optimisation techniques would be the same if the same values of parameters are used in the analyses.

This important point can be verified by comparing the outcome from the example analyses given in the preceding sections. The outcome from the cost-benefit analysis and the multi-attribute utility analysis both indicates that protection Option 2 to be optimum. Although the cost-effectiveness technique does not present an optimised protection option, because it does not involve any trade-off between collective dose and protection cost, it is found that Option 2 is the most cost-effective giving the highest dose reduction per invested amount of money.

D3.5 References

ICRP, International Commission on Radiological Protection (1989) *Optimization and Decision-Making in Radiological Protection*. Publication 55, Pergamon Press, Oxford.

ANNEX D4
METHODOLOGY FOR DETERMINING ACTION LEVELS FOR
REMEDIAL MEASURES

Table of Contents

D4.1	Concept of an Action Level	231
D4.2	Calculation of Action Levels	232
D4.3	References	234

D4.1 Concept of an Action Level

Action Levels refer to different protective measures or strategies of actions like agricultural countermeasures or radon reducing measures in houses and they relate to the residual dose without remedial actions. The action level is defined as [IAEA, 1996]:

Action level is the level of dose rate or activity concentration above which remedial actions or protective actions should be carried out in chronic or emergency exposure situations

Action Levels are levels of dose rate, activity concentration, etc above which remedial actions are taken and below which they are not. An Action Level is set such that the dose averted by taking the remedial action is always worthwhile in terms of the costs and other disadvantages involved. Justified Action Levels would begin at the minimum value of the avertable dose at which the remedial action is just beginning to do more good than harm. The Action Level can thus be defined as the lowest level at which remedial actions to reduce doses is justified. An equivalent definition would be that the Action Level is equal to the maximum level of dose attributable to the contamination without any protective actions being justified, because the avertable dose for a specified action is not worthwhile in terms of the overall costs of the action.

If an Action Level is exceeded, it is indicated that some form of remedial action specific to the situation considered is likely to be appropriate. The concept of an Action Level is illustrated in Figure D4.1.

Effective dose E_0 in a particular exposure situation

Action Level



An Action Level is the lowest level at which a remedial measure to reduce doses is justified

Residual dose E_1 after implementing a remedial action

Background Dose Level

Note: The avertable dose is here given as $\Delta E = E_0 - E_1$.

Figure D4.1 The action level (AL) is the minimum residual dose level at which a remedial action is justified to reduce doses

The Action Level is *not* a limit but a reference level closely related to the reduction in individual doses to be achieved by a specific remedial action. The Action Level is directly derived from the pre-remediation dose level and the efficiency of the considered remedial action. If an Action Level were to be set in advance from any consideration of an acceptable pre-remediation dose, this would be in conflict with the basic radiation protection principles because such a pre-set level would in fact be a dose limit.

Action Levels would also be used for the reduction of radon in dwellings. Recommended Action Levels from ICRP for radon in dwellings are 3 - 10 mSv a⁻¹ [ICRP, 1994] corresponding to a radon concentration of 200 - 600 Bq m⁻³. The action level for remedial actions relating to chronic exposure situations involving radon in workplaces is a annual average concentration of ²²²Rn of 1,000 Bq m⁻³.

Different organs and tissues have a wide range of sensitivity for radiation induced deterministic effects. Threshold dose rates for deterministic effects under conditions of prolonged exposure over many years have been used as action levels for chronic exposure [IAEA, 1996]. The value for permanent sterility, for clinically significant depression of the blood-forming process and for opacities sufficient to cause impairment of vision is 0.2, 0.4 and 0.1 Gy a⁻¹, respectively [IAEA, 1996].

D4.2 Calculation of Action Levels

Action Levels are used as a screening tool to determine if a remedial measure is justified. Action Levels correspond to dose levels and are derived by mathematical modelling of all the significant pathways of exposure and the projected relevant behaviour of the average member of the affected population.

It is important to note that the Action Level is specified in terms related to the dose attributable to the contamination giving rise to the exposure; however, it is derived from considering the possibilities for reducing or averting exposures. Action Levels can also – as indicated above - be expressed in any surrogate quantity related to dose. Quantities like dose rate in air, surface contamination density, and activity concentration in air, water and foodstuffs can all be derived from suitable models. The resulting Action Levels derived from such models would depend on site and nuclide specific parameters. Action Levels specified in terms of naturally occurring radionuclides may include the local background level.

For situations where the exposed population is clearly identifiable, the Action Level can often be expressed as individual dose. In situations where the exposed individuals cannot be identified, as for exposure from contaminated food, the Action Level may be more usefully expressed as collective dose per unit mass of the foodstuff or as activity concentration in that foodstuff. In the latter case, the distribution of contamination in foodstuffs making up the human diet after the application of any protective actions would range from zero up to the Action Level.

Specific information on the nature of the contamination, which is expected to be available at a given site, involve characteristics of the source as well as environmental and demographic data. In this case, a more accurate and specific optimisation analysis may be carried out on the basis of actual data and the actual efficiency of the remedial measure resulting in specific operational quantities for the remediation. Some of the parameters, such as the extent of the contamination, may not become fully known until after remediation is in progress. Therefore, new information may require an adjustment of the calculated operational quantities as part of the iterative nature of the decision-making process. In those cases where the contamination comprises both radioactive and non-radioactive materials, planning for and confirmation of the remediation should take both kinds of contaminants into account.

Annual (committed) collective effective doses, S , to members of the affected population for a given exposure pathway, $path$, and radionuclide, i , with concentration, C_i , can (simplified) be calculated as:

$$S_{path,i} = C_i \cdot g_{path,i}(p_1, p_2, \dots, p_n)$$

The function, g_i , describes the annual collective effective dose for a specific pathway, $path$, and radionuclide, i , per unit activity concentration of that radionuclide. The parameters p_1, p_2, \dots, p_n denote transfer factors, migration factors, concentration profile, location factors, dose conversion factors etc. The annual collective effective dose, S_i , from all relevant pathways to the affected group of people would be:

$$S_i = C_i \cdot \sum_{path} g_{path,i}(p_1, p_2, \dots, p_n) = C_i \cdot G_i$$

The factor, G_i , for radionuclide i describes the ratio of annual (committed) collective effective dose from all pathways of radionuclide i per unit concentration of nuclide i . The total collective dose to the affected population from all radionuclides and all pathways over time, T , can be calculated as:

$$S = T \cdot \sum_i C_i \cdot G_i$$

It is here assumed that the half-life of the nuclides considered are large compared to the integration time, T . When this is not the case, radioactive decay has to be included in the time-integration of the collective dose.

When remediation of a contaminated site will reduce the total collective dose by a factor, f , (>1) the avertable collective dose, ΔS , can be calculated as:

$$\Delta S = \frac{f-1}{f} \cdot T \cdot \sum_i C_i \cdot G_i$$

If the total costs of the remediation is called X and the equivalent monetary value of avoiding a unit collective dose is called α , the specific remediation option is justified when the nett benefit is positive:

$$\alpha \cdot \Delta S - X \geq 0$$

which is equivalent to:

$$\sum_i C_i \cdot G_i \geq \frac{f}{f-1} \cdot \frac{X}{\alpha \cdot T}$$

The total costs would include the monetary costs of the remedial measures, X_{remedial} , and the equivalent cost of the collective dose, S_{worker} , to the workers who carry out the remedial measures:

$$X = X_{\text{remedial}} + \alpha \cdot S_{\text{worker}}$$

In the following the ratio of the concentration of the single nuclides, C_i , to the nuclide contributing mostly to the total collective dose, C_1 , is called r_i , and the monetary costs are called, X . A generic justified and normalised Action Level, AL_{norm} , in terms of annual collective dose to the affected population above which the remedial measure is justified can then be calculated as:

$$AL_{\text{norm}} = C_1 \cdot \sum_i r_i \cdot G_i \geq \left(\frac{f}{f-1} \right) \cdot \left(\frac{X}{\alpha \cdot T} \right)$$

Model calculations will determine the values of $G_i = \sum g_{\text{path}, i}(p_1, p_2, p_3, \dots, p_n)$.

Action Levels in terms of activity concentrations can be determined from the normalised values and the dose conversion factors as:

$$AL = \frac{AL_{\text{norm}}}{\sum_i r_i \cdot G_i}$$

The Action Level will express, for each of the specified remediation options, the concentration of the dominating radionuclide above which the specified option should be introduced.

D4.3 References

IAEA, International Atomic Energy Agency (1996) *International basic safety standards for protection against ionizing radiation and for the safety of radiation sources*. Safety Series No. 115, Vienna.

ICRP, International Commission on Radiological Protection (1994) *Protection against radon-222 at home and at work*. Publication 65, Pergamon Press.