

**Ravenglass Estuary:
Basic Characteristics and Evaluation
of Restoration Options**

Restoration Strategies for Radioactively Contaminated
Sites and their Close Surroundings
RESTRAT - WP 1.4

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Executive Summary

TD.12: Ravenglass Estuary: Basic Characteristics and Evaluation of Restoration Options

The aim of this report is to demonstrate the applicability of the decision-making approach, proposed by RESTRAT, for selecting restoration techniques to treat estuaries in the vicinity of nuclear facilities. The Ravenglass Estuary has been used as an example of such a site.

The decision-making procedure is based on multi-attribute utility analysis. This is superior to convention cost-benefit analysis because criteria can be incorporated without the need to convert them into common units of measurement. For the Ravenglass Estuary the attributes considered are radiation-induced health effects, monetary costs and social factors. These may be further broken down into a series of sub-attributes and weighting factors. A probabilistic approach was adopted to take account of the uncertainties with the utility functions and weighting factors.

The report provides a description of the steps followed in the decision-making process. These were as follows:

- Characterisation of the site in terms of:
 - description of the area in terms of geography and topography, geology and hydrogeology, pedology, meteorology, hydrology and demography;
 - physico-chemical characterisation of the radionuclides in the water column and in the sediment;
 - characterisation of the sources of contamination and the distribution of contamination throughout the site.
- Identification and characterisation of restoration options which are applicable to the site in terms of
 - effectiveness against contaminants (radionuclides);
 - cost of applying the restoration technique;
 - the exposure times for restoration workers during restoration.
- Determination of the radiological impact of the site through:
 - the development of compartment model to describe the site;
 - the quantification of processes, exposure pathways and exposure groups;
 - the calculation of radiological doses to the public and restoration workers.
- Ranking of the restoration options through the calculation of the attributes, utilities and weighting factors needed to perform a multi-attribute utility analysis on the various restoration options.

The Ravenglass Estuary which encompasses the tidal reaches of the Rivers Esk, Irt and Mite and occupies an area of 5.6 km² of which 86% is intertidal. The area represents a highly dynamic environment with sediments undergoing reworking and erosion. The estuary has been contaminated by the discharge of radionuclides from the nearby Sellafield plant. Principal radionuclides are plutonium, americium and caesium. The reworking of sediment has meant that radionuclides are distributed throughout the site. The locality is agricultural and sparsely populated. There is no commercial fishing in the estuary. The 500 year collective dose for the untreated site was calculated from the RESTRAT model to be 29 manSv, with the consumption of animal produce being the most important exposure pathway.

Restoration techniques which were applicable to this type of site were identified as source removal, sediment washing and chemical solubilisation of the radionuclides. Multi-attribute utility analysis showed that whilst source removal presented the best restoration option for the site, the option of not remediating the site gave a significantly higher score. This suggests that for the Ravenglass Estuary the best option would be to leave the site as it is with on-going programme of monitoring.

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1. Terms of reference

This report is submitted as Technical Deliverable 'TD12' against the requirements of the Work Package 1.4 for the Restoration Strategies for Radioactive Sites and their Close Surroundings (RESTRAT) project.

The RESTRAT project, which is funded by the European Commission under the Nuclear Fission Safety Programme, has an overall objective of developing generic methodologies for ranking restoration techniques as a function of contamination and site characteristics. The development of this generic methodology is based on an analysis of existing remediation methodologies and contaminated sites, and is structured in the following steps:

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

This Work Package, which is jointly funded by British Nuclear Fuels plc, is concerned with the characterisation of the Ravenglass Estuary and the identification of appropriate restoration techniques for this site. The aims of this Work Package are as follows:

1. describe the main geographic, hydrographic and geological features of the site;
2. describe the local human activities;
3. describe the major sources of radioactive contamination.
4. The identification of available restoration options for the site.
5. assess the impacts of the radioactive contamination and the effect of each restoration option;
6. evaluate and rank each remediation option, in terms of the suitability for the Ravenglass site.

2. Introduction

The 'Ravenglass estuary' is a local name for an area which encompasses the tidal reaches of the Rivers Esk, Irt and Mite. The estuary is located on the Cumbrian coast in the north-west of England (see Figure 1).

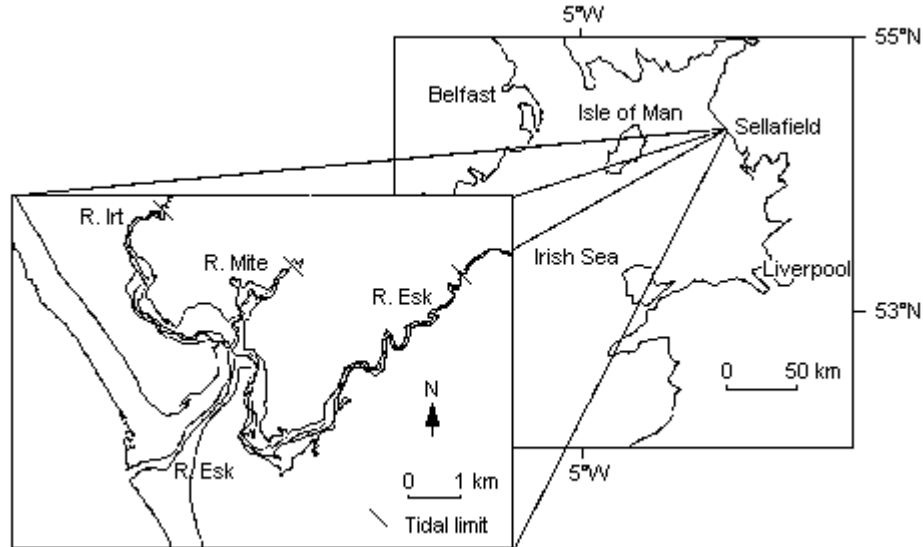


Figure 1 Geographical location of the confluence of the rivers Irt, Esk and Mite - the 'Ravenglass' estuary.

For the purposes of this report, the term Ravenglass estuary is used to designate the area of confluence of the three rivers. Where information is considered specific to one or other of the rivers and their estuaries, the specific river name is employed.

The estuary lies near to the point of authorised discharges of radioactive wastes into the Irish Sea from the British Nuclear Fuels plc (BNFL) nuclear fuel reprocessing plant at Sellafield. In addition, a stream from the Drigg low level waste disposal site discharges low levels of radionuclides into the River Irt and thus into the estuary. From various lines of evidence, including their mineralogy, the principal source of the sediment, as well as its radioactive contaminants, is considered to be the sea, with the rivers contributing relatively little from their catchments.

The association between the radionuclides and the sediment means that their accumulation and subsequent fate will be determined by the estuarine sedimentary system of physical, chemical and biological processes.

The Ravenglass estuary lies within the Lake District National Park which has open public access. To the north lies a nature reserve and the Drigg low level waste disposal site and to the south a military artillery range. The region is essentially rural. However, there are pockets of local industry, such as Sellafield. It is a sparsely populated area. Only recreational fishing presently takes place within the estuary.

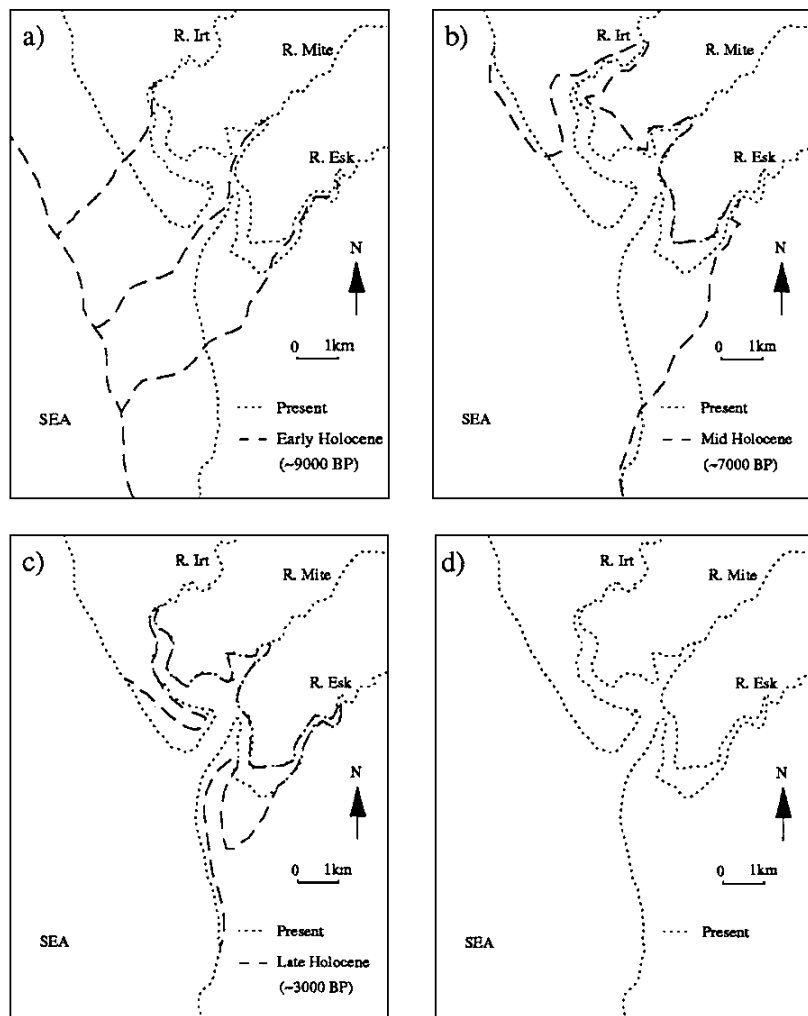
3. General site characterisation

3.1 Description of the area

3.1.1 Geography and topography

The Ravenglass estuary occupies an area of 5.6 km², of which 86% is intertidal.

The geological evolution of the estuary has determined its present morphology, and owes its origin to the world-wide eustatic rise in sea level following melting of the ice after the last glaciation. In north-west England the transgression of the sea reached within 4 m of its present position about 7,600 years ago (see Figure 2) and an associated rise in the water table led to the development of coastal swamps and fens. It is possible that some of the clays and organic deposits in the Ravenglass estuary represent that stage of coastal evolution (Tooley, 1974).



a) ~9000 BP; b) ~7000 BP; c) ~3000 BP; d) present.

From Kelly and Emptage (1992)

Figure 2 Reconstruction of the post-glacial evolution of the Esk estuary.

In the ensuing period of relatively stable sea levels, the coast of the area has evolved by a combination of normal marine processes and isostatic uplift. As sand beaches have developed in the area, so too has a coastal dune system.

Because of the recent anthropogenic influence, it is unlikely that the estuarine sediment system is at equilibrium. This, together with the long term geological changes affecting estuaries, makes it near impossible to predict the future evolution of the estuarine system.

3.1.2 Geology and hydrogeology

a) *Sediment facies*

The sediments of the Ravenglass estuary can be subdivided into categories which reflect the environmental conditions under which they have formed (i.e. “facies”). Mapping of the facies in the estuary allows the distribution of sediment associated radionuclides and activity to be both explained and predicted. The significant aspects of sediment behaviour, which the facies express and which determine radionuclide behaviour, are: grain size fractionation, sedimentation rate, residence time and aspects of diagenesis (post-depositional modification). Correlated with these are the radionuclide properties of specific activity, deposition flux, inventory, residence time and vertical distribution.

Estuarine deposits occupy 77% of the total area of the Ravenglass estuary (see Table 1). The remainder comprises relic facies sediments, i.e. non-estuarine in origin (9%), and the low water channel (14%). The estuarine deposits belong to three major facies: channel, bank and erosional facies. These facies occur, in varying proportions, throughout the estuary, but with channel facies increasingly important towards the mouth and bank facies towards the head of the estuary.

Table 1 Measured areas of sediment facies for the Ravenglass estuary.

Location	Area of sediment facies (km ²)							
	Total	Low-water channel	Bank		Erosional	Channel		Relic
			Upper	Lower		Sand	Gravel	
Irt	1.9	0.2	0.8	0.1	0.6	0.1	-	<0.1
Mite	0.6	0.1	0.2	<0.1	0.2	<0.1	-	0.1
Esk	3.2	0.5	1.1	0.1	0.2	0.9	<0.1	0.4
Whole estuary	5.7	0.8	2.1	0.2	1.0	1.0	<0.1	0.5

Simplified from Kelly and Emptage (1992).

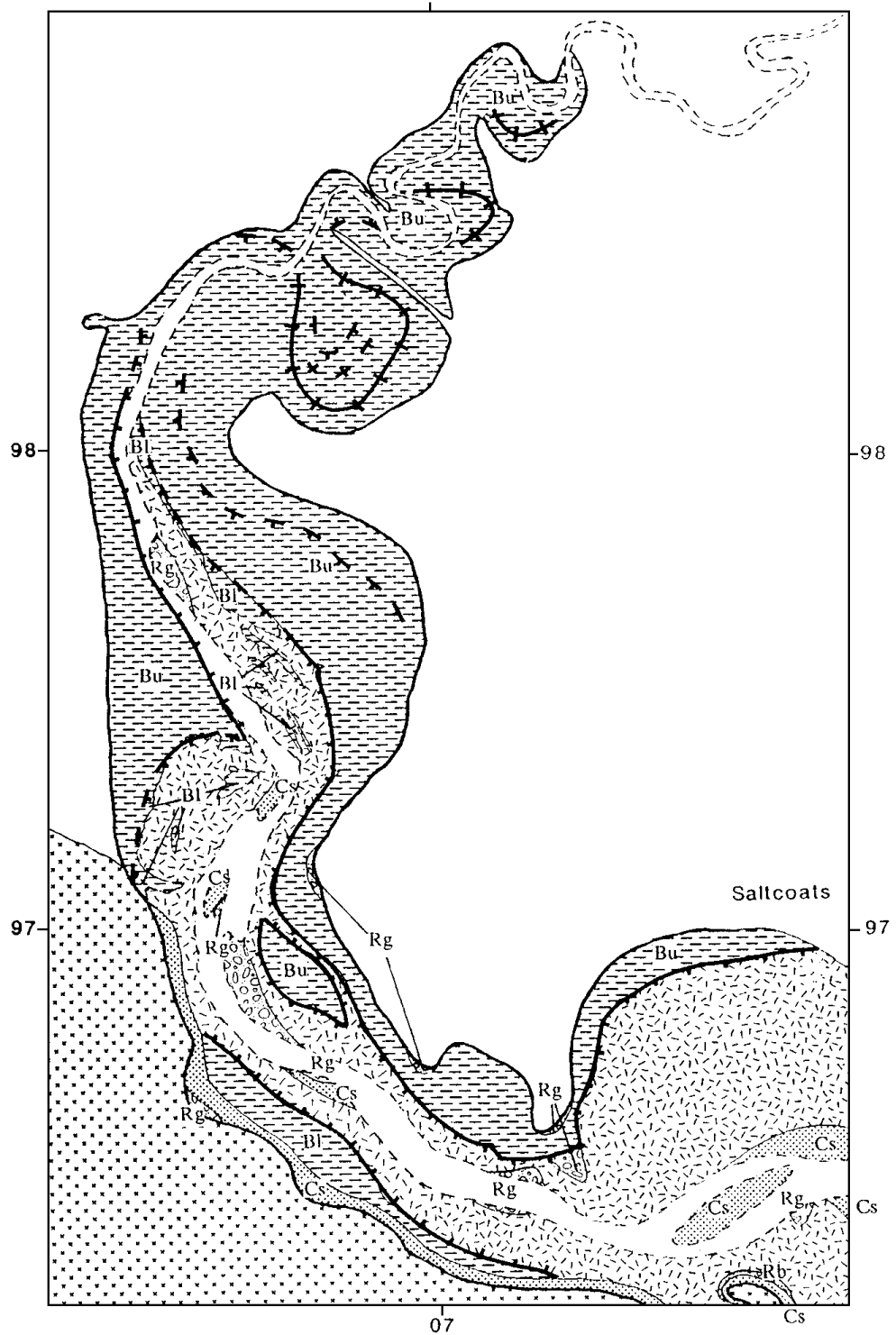
Relic facies include a wide range of sediments: clays, sands, gravels, till and organic deposits. These occurs where the currents are too strong for deposition of modern sediments.

Channel facies, which form 18% of the estuary, mainly comprise coarse grained sediments of sand grade which have been deposited from the bed load of the tidal currents in the estuary. Sand sub-facies are deposited in the main channel system of the estuary, where sediment transport and deposition is partly associated with mobile beds (as ripples, dunes and bars). The sand is supplied to the estuary by offshore and longshore currents to the mouth and by direct input from eroding subaerial material. Gravel dune bedforms, made up of shell rich fine/medium gravel, are found at bends in the river where the currents are strong due to confinement by the channel.

Bank facies deposits, comprising 40% of the estuary, consist mainly of fine grained sediments, of sandy silt and silt grade, i.e. deposits of a low energy environments. These deposits form the mudflats, saltmarshes and intertidal pastures of the estuary. These deposits constitute the main reservoir for sediment associated radioactivity in the estuary because of their sediment characteristics, and their extent. They are accumulating in the parts of the estuary which are covered only by low velocity water during the higher stages of the tide, i.e. towards the lateral margins and head of the estuary. Two sub-facies may be defined on the basis of their elevation. The lower bank facies includes unvegetated mudbanks, which are covered at almost every tide. The upper saltmarsh banks is covered with ungrazed saltmarsh vegetation.

The erosional facies occupies 18% of the estuary. This represents areas of estuarine sediments which are undergoing erosion and reworking, at least on a sporadic time scale, by the slow stripping of the sediment surface. They are characterised by a mixed grain size distribution of silty sand grade. It occurs extensively where large areas of bank facies and underlying channel facies sediments are, or have been, eroded.

Field mapping and aerial photograph interpretation (NERC, 1987) and subsequent direct measurement at a range of localities enabled Kelly and Emptage (1992) to produce detailed distributional maps of sediment facies throughout the area of the Ravenglass estuary (Figure 3a-3f).

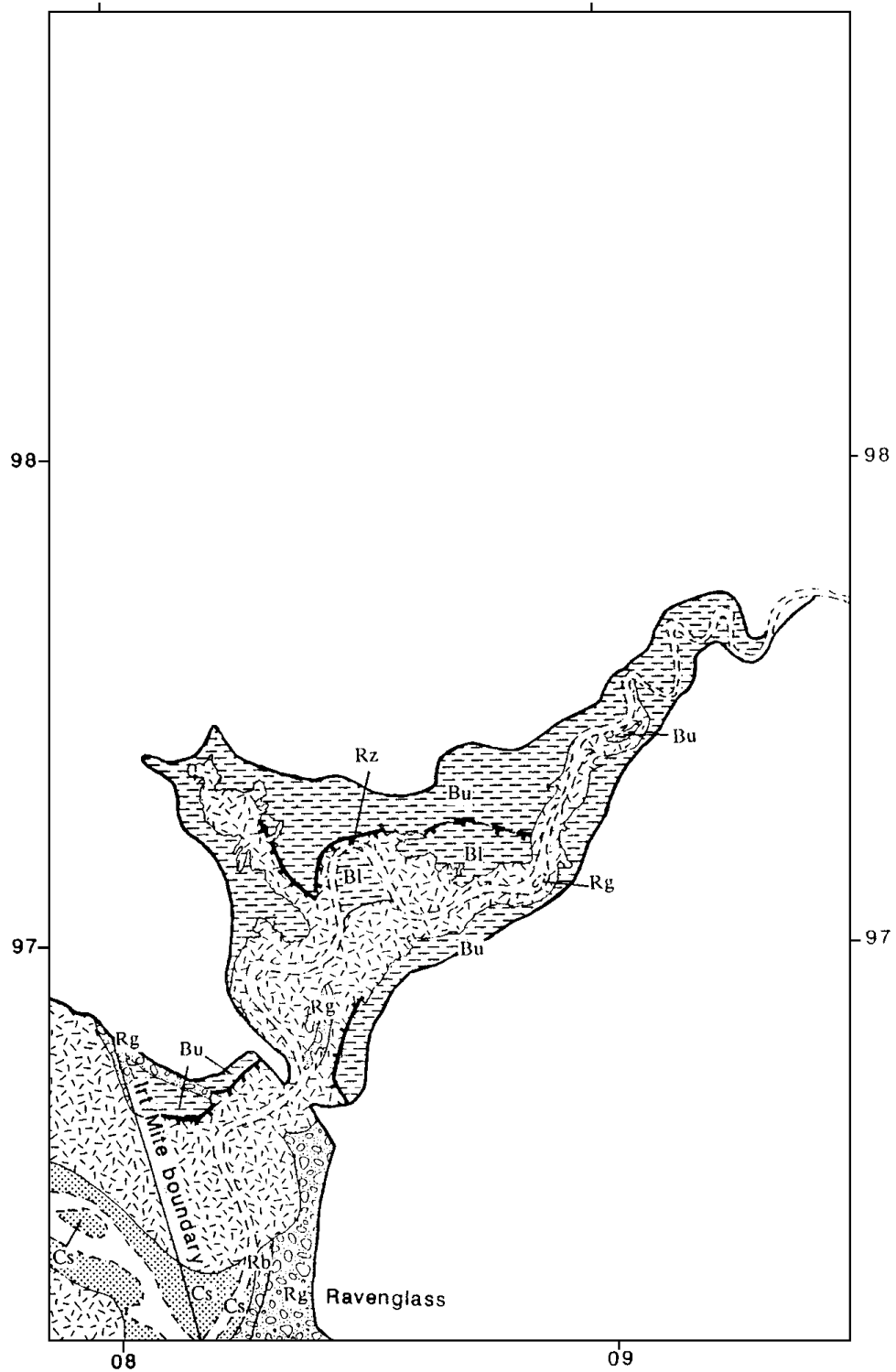


Key given with f.

From Kelly and Emptage (1992).

Figure 3 Sediment facies distribution in the Irt, Mite and Esk estuaries.

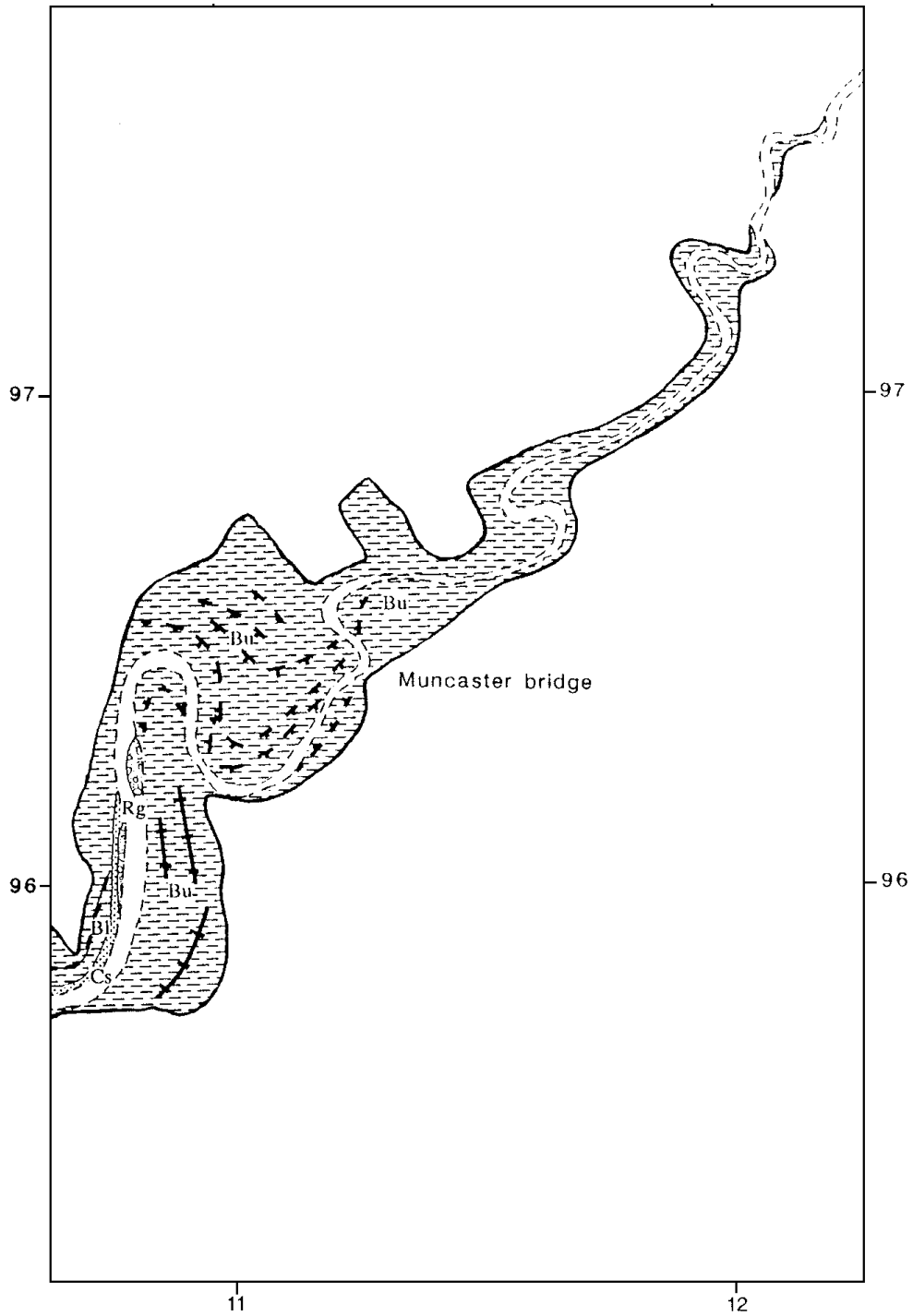
a) Sediment facies distribution in the Irt estuary.



Key given with f.

From Kelly and Emptage (1992).

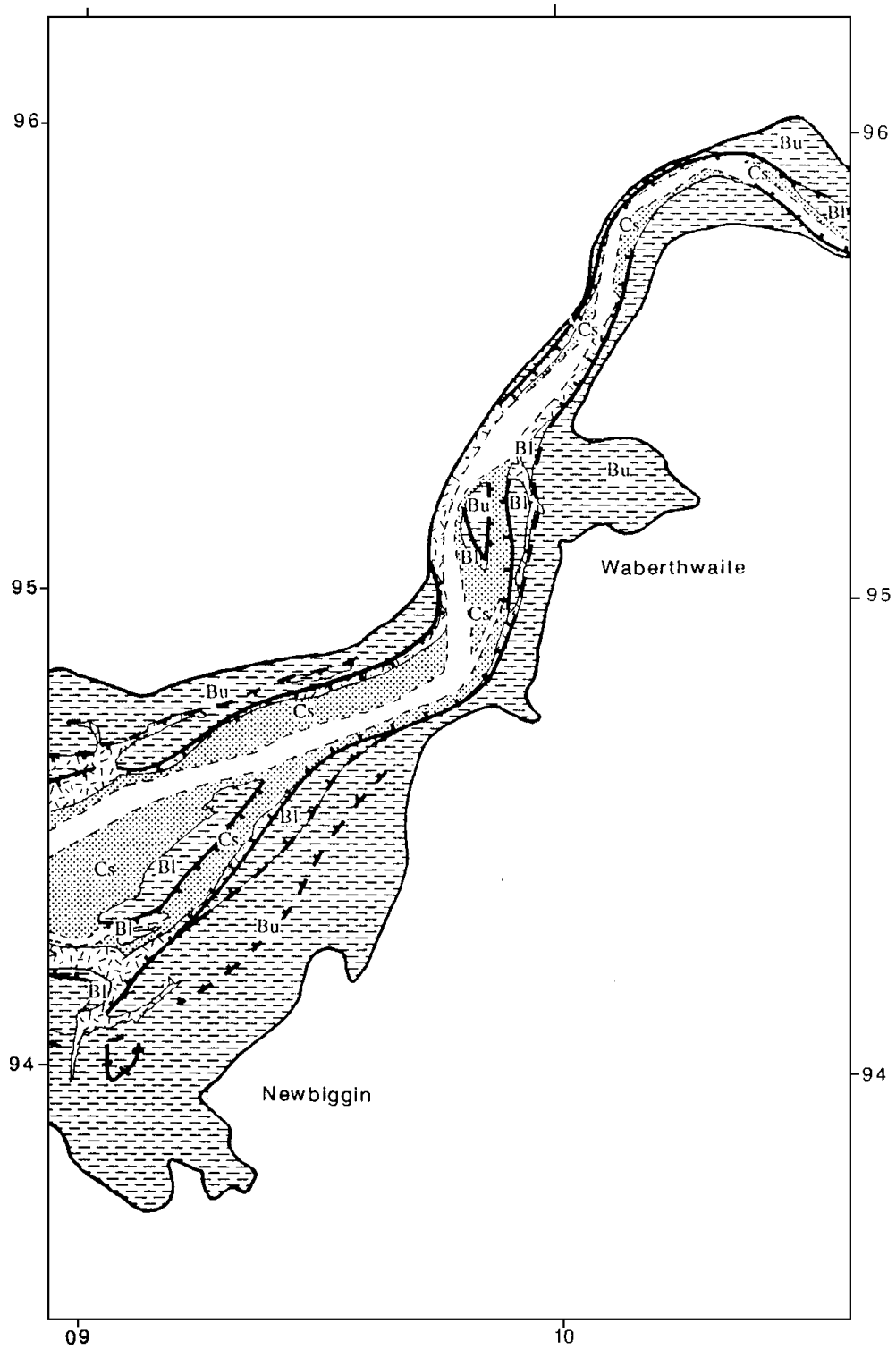
b) Sediment facies distribution in the Mite estuary.



Key given with f.

From Kelly and Emptage (1992).

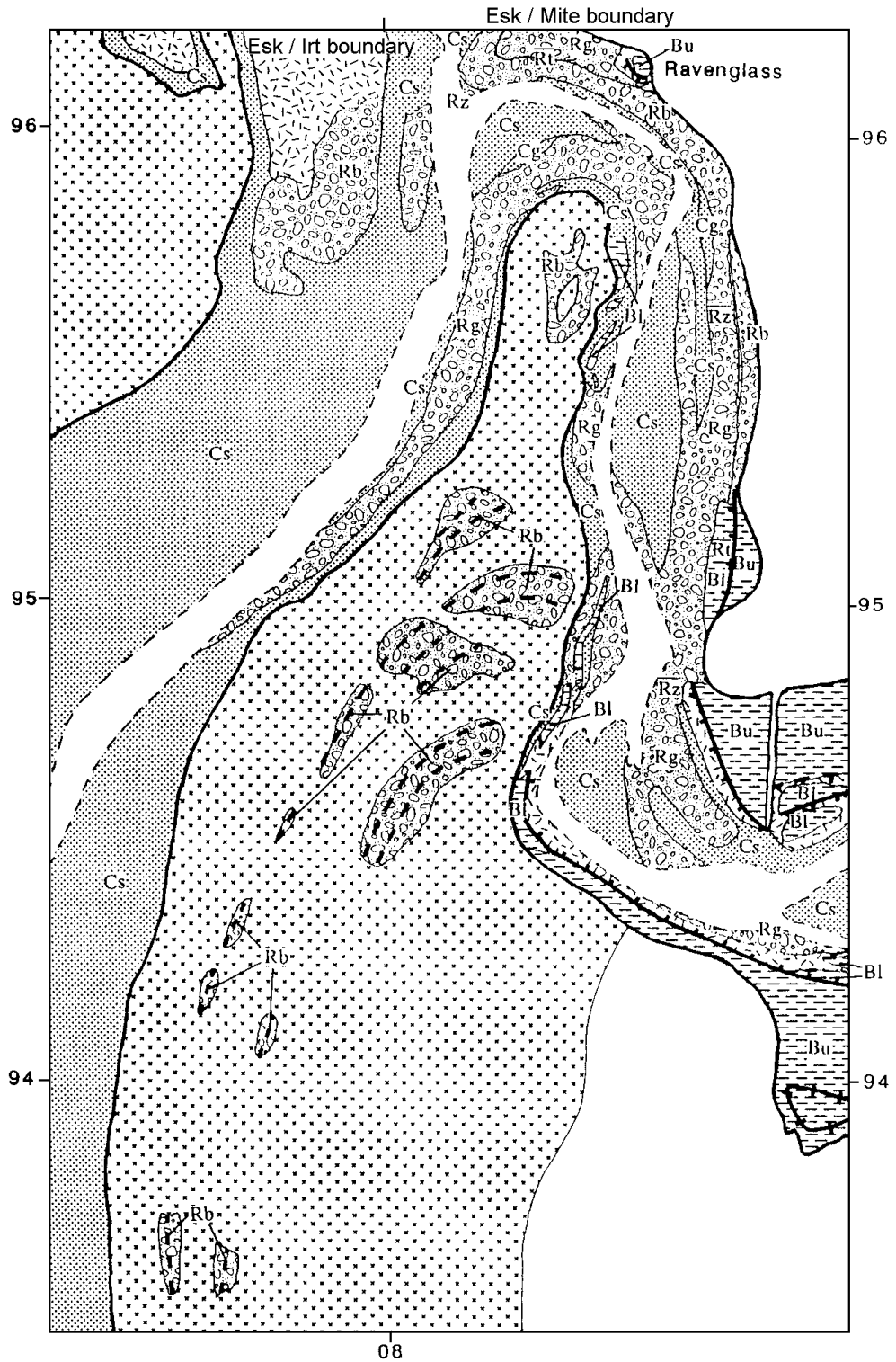
c) Sediment facies distribution in the upper Esk estuary.



Key given with f.

From Kelly and Emptage (1992).

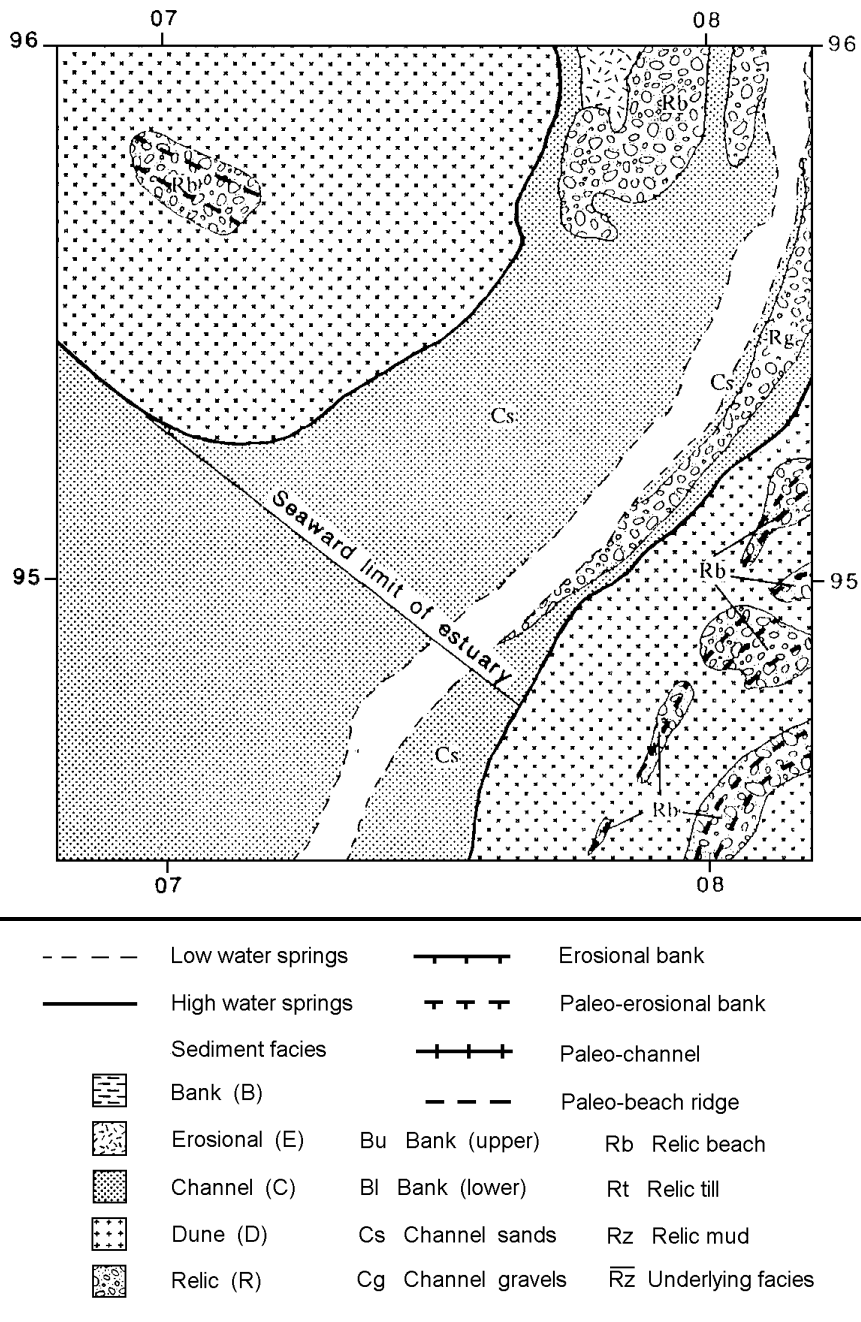
d) Sediment facies distribution in the middle Esk estuary.



Key given with f.

From Kelly and Emptage (1992).

e) Sediment facies distribution in the outer Esk estuary.



From Kelly and Emptage (1992).

f) Sediment facies distribution in the outer mouth of the Esk estuary.

b) Sediment grain size distribution

Grain size is an important property of sediment, which, together with shape and mineralogy, makes up its lithology. Sediment transport, deposition and erosion processes are highly sensitive to grain size and, consequently, sediments are partly or completely size fractionated during these processes.

The areas occupied by the different grain size classes are presented in Table 2. The major part of the estuary is intertidal (86%) and, of this, just under half is fine grained sediment (silts and sandy silts). The low water channel (which represents 14% of the total area) has not been surveyed directly but is thought to consist mainly of sands and gravels.

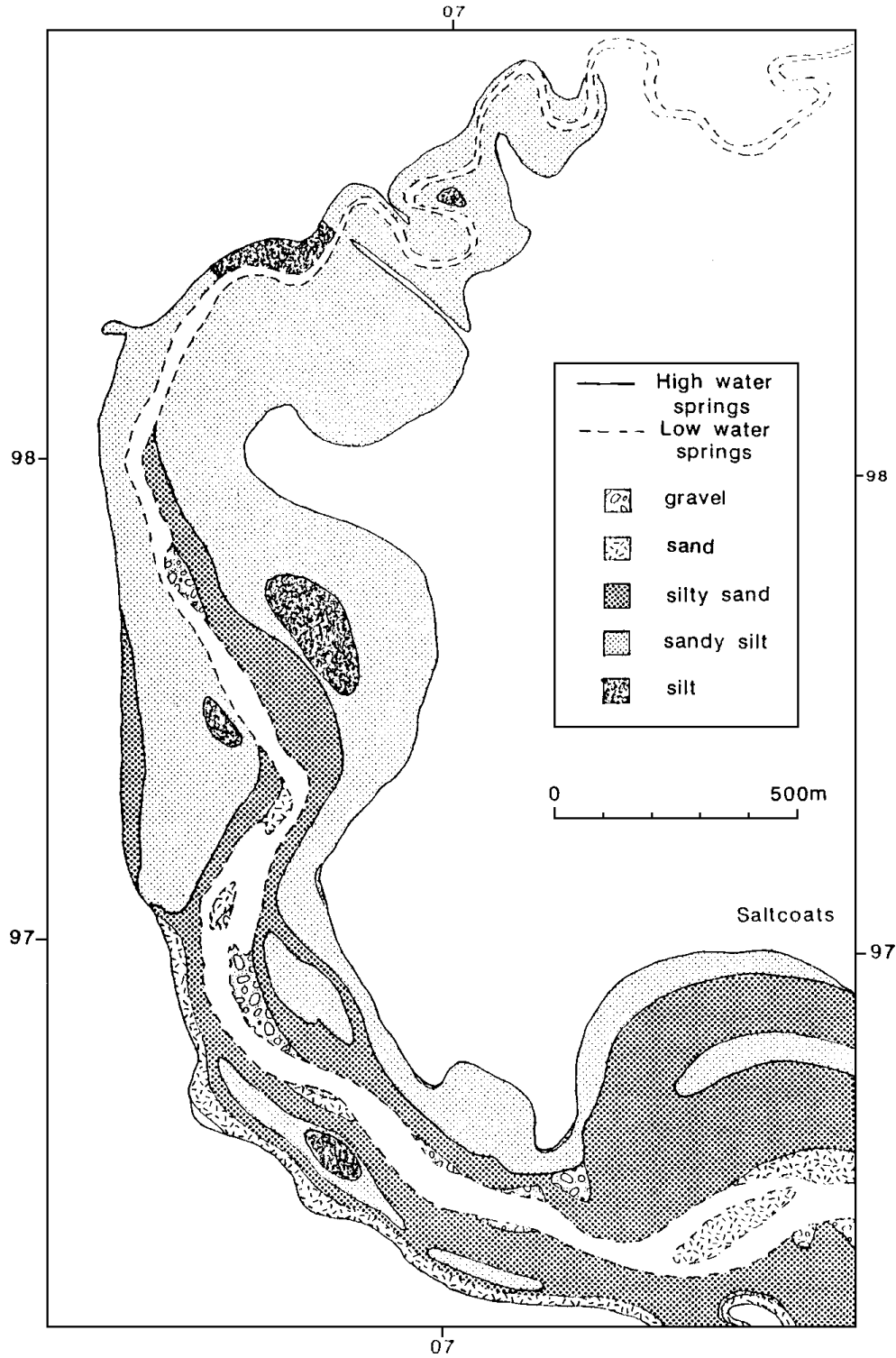
Table 2 Measured areas of sediment classes for the component parts of the Ravenglass Estuary.

Location	Area (km ²)						
	Total	Low-water channel	Silt	Sandy Silt	Silty sand	Sand	Gravel
Irt	1.9	0.2	0.1	0.8	0.6	0.1	<0.1
Mite	0.6	0.1	<0.1	0.3	0.2	<0.1	<0.1
Esk	3.2	0.5	0.1	1.0	0.4	0.7	0.4
Whole estuary	5.7	0.8	0.2	2.1	1.1	0.9	0.5

Simplified from Kelly and Emptage (1992).

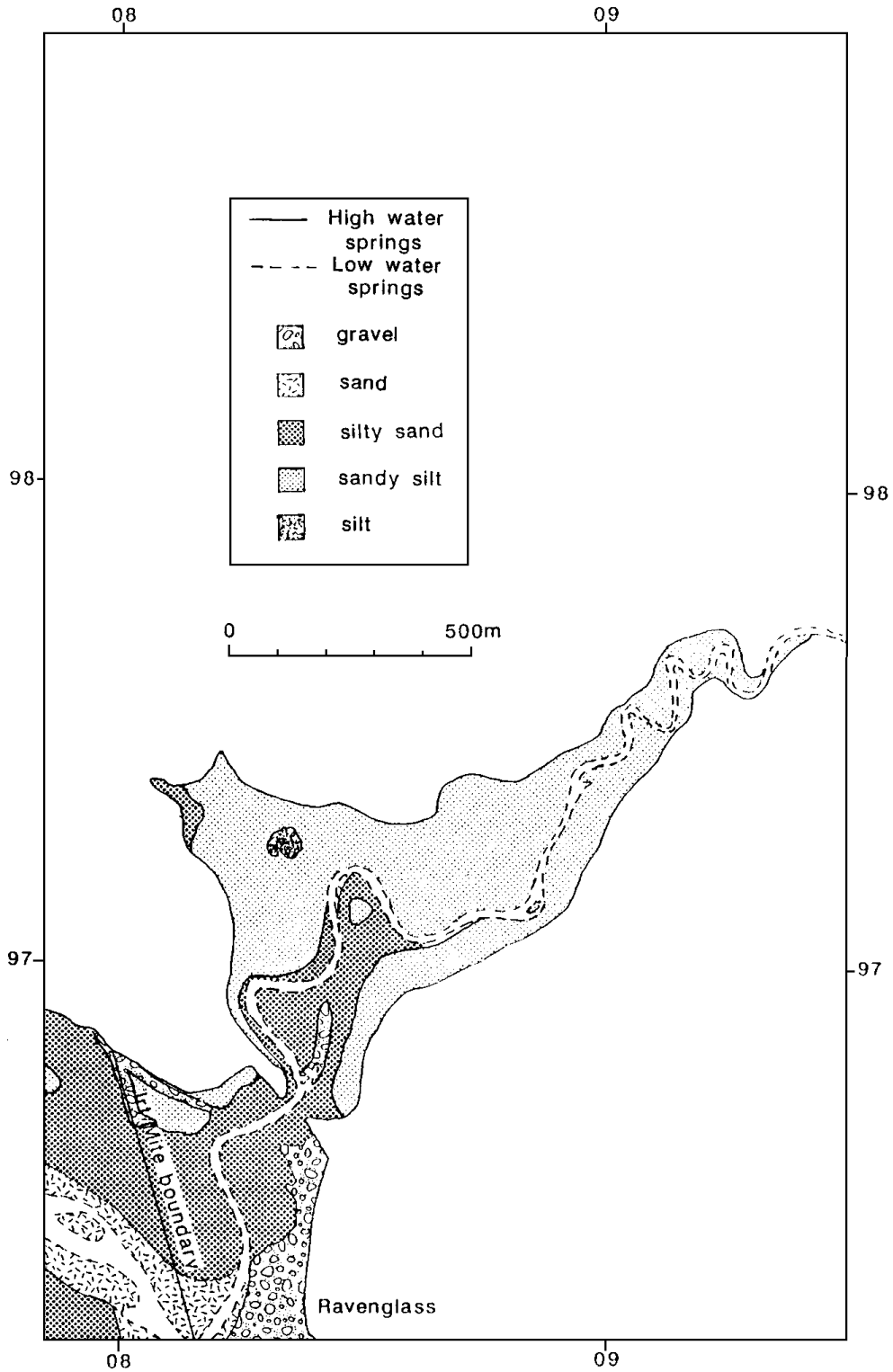
From field mapping and aerial photograph interpretation (NERC, 1987) and subsequent direct measurement at a range of localities, Kelly and Emptage (1992) have produced a distributional map of grain size throughout the area of the Ravenglass estuary (see Figure 4). The sediments generally have a silt-clay ratio of greater than 2 (Hetherington and Jefferies, 1974; Carr and Backley, 1987; Kelly and Emptage, 1992) which is consistent with sediments from the north-eastern Irish Sea (Pantin, 1978).

As a generalisation, the grain size of the estuarine sediments decreases toward the head of the estuary and, laterally, away from the low water channel. Relatively coarse sediments (sands and gravels) dominate the surface sediments in the lower reaches., extending upstream as far as the Newbiggin viaduct on the Esk, the Mite viaduct and Saltcoats on the Irt. In the middle reaches, coarse sediments in and adjacent to the low water mark are bordered by broad lateral banks of fine grained deposits. In the upper reaches, these fine grained sediments occupy most of the area.



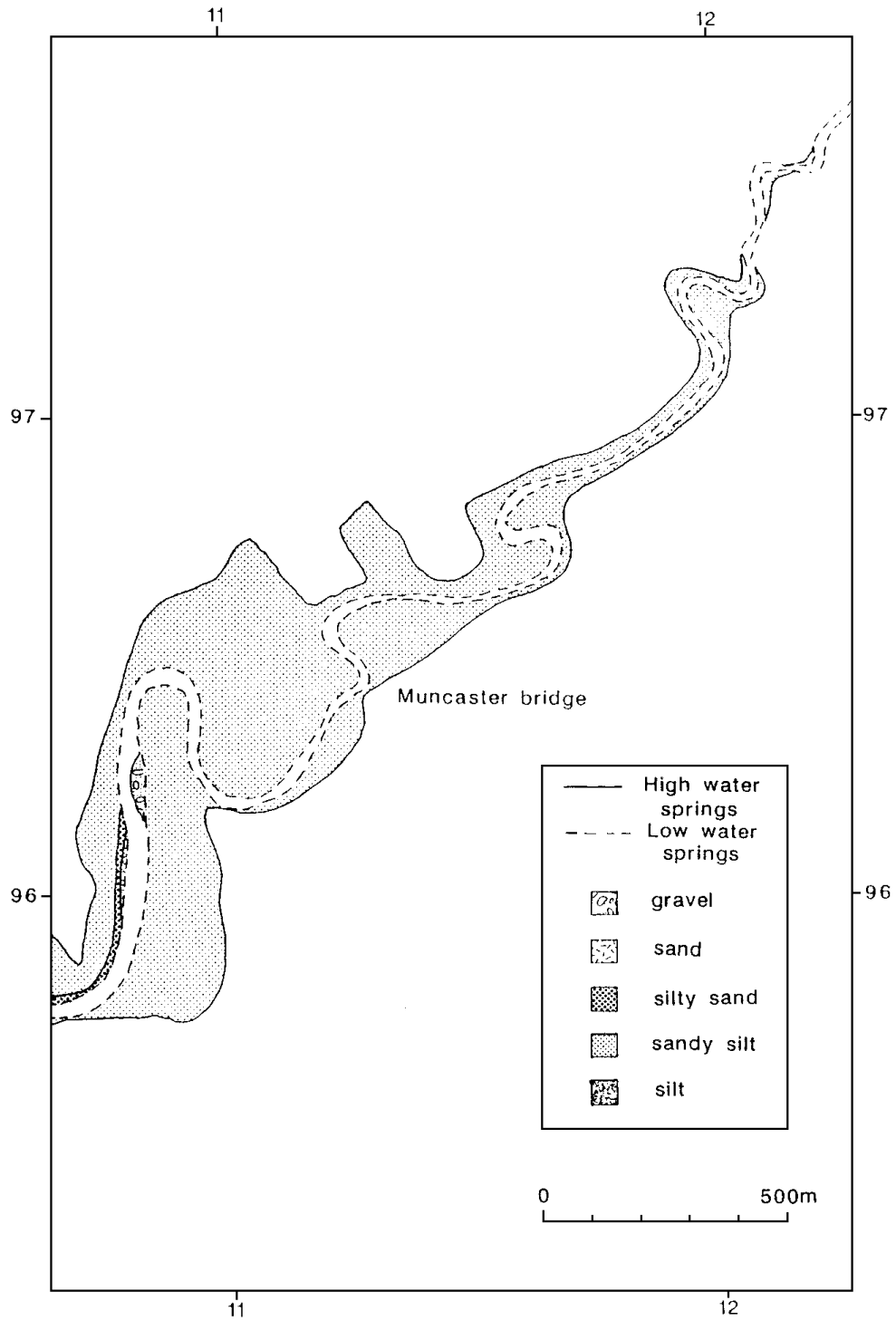
From Kelly and Emptage (1992).

Figure 4 Grain size distribution in the Irt, Mite and Esk estuaries.
a) Grain size distribution for the Irt estuary.



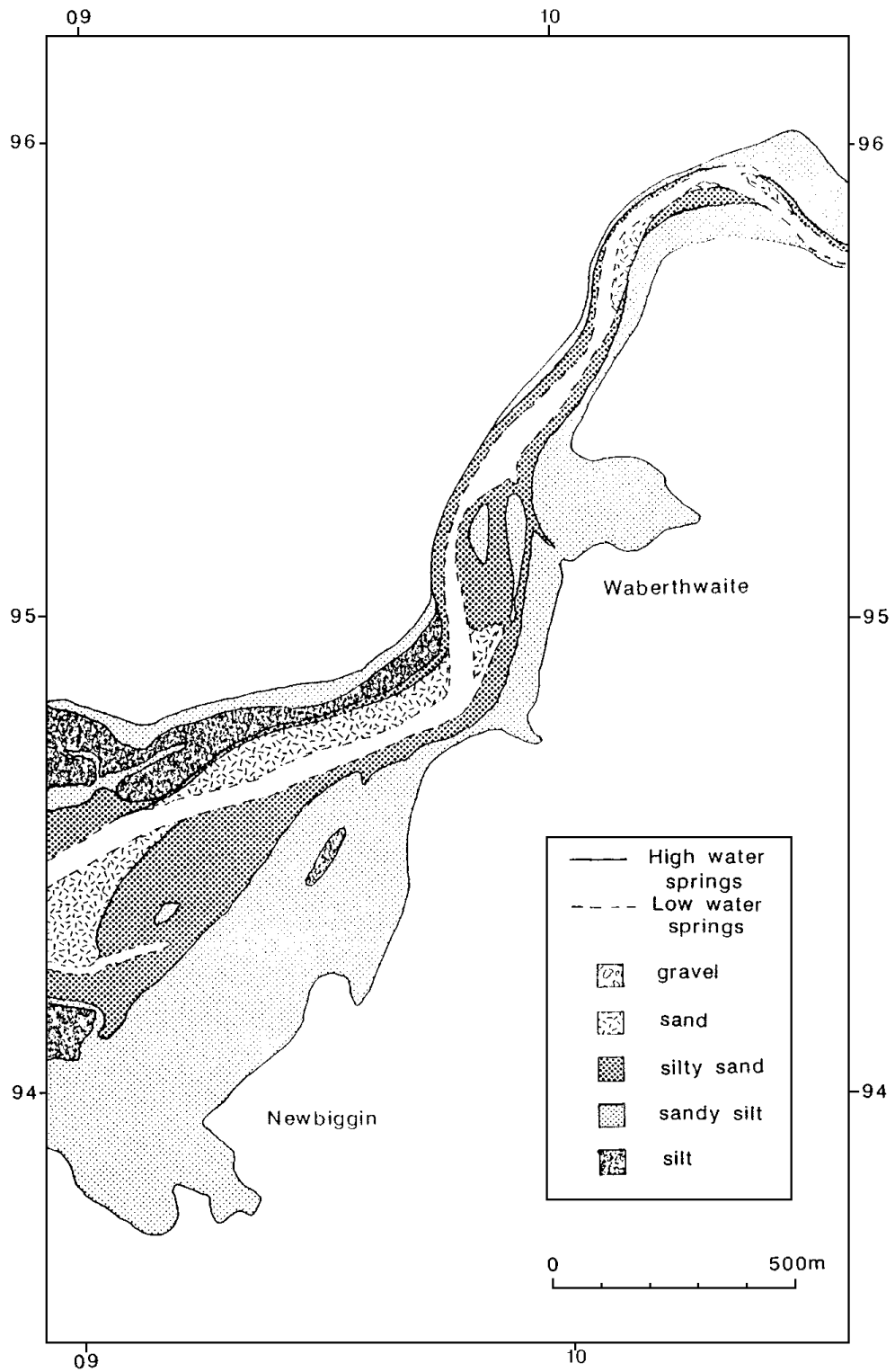
From Kelly and Emptage (1992).

b) Grain size distribution for the Mite estuary.



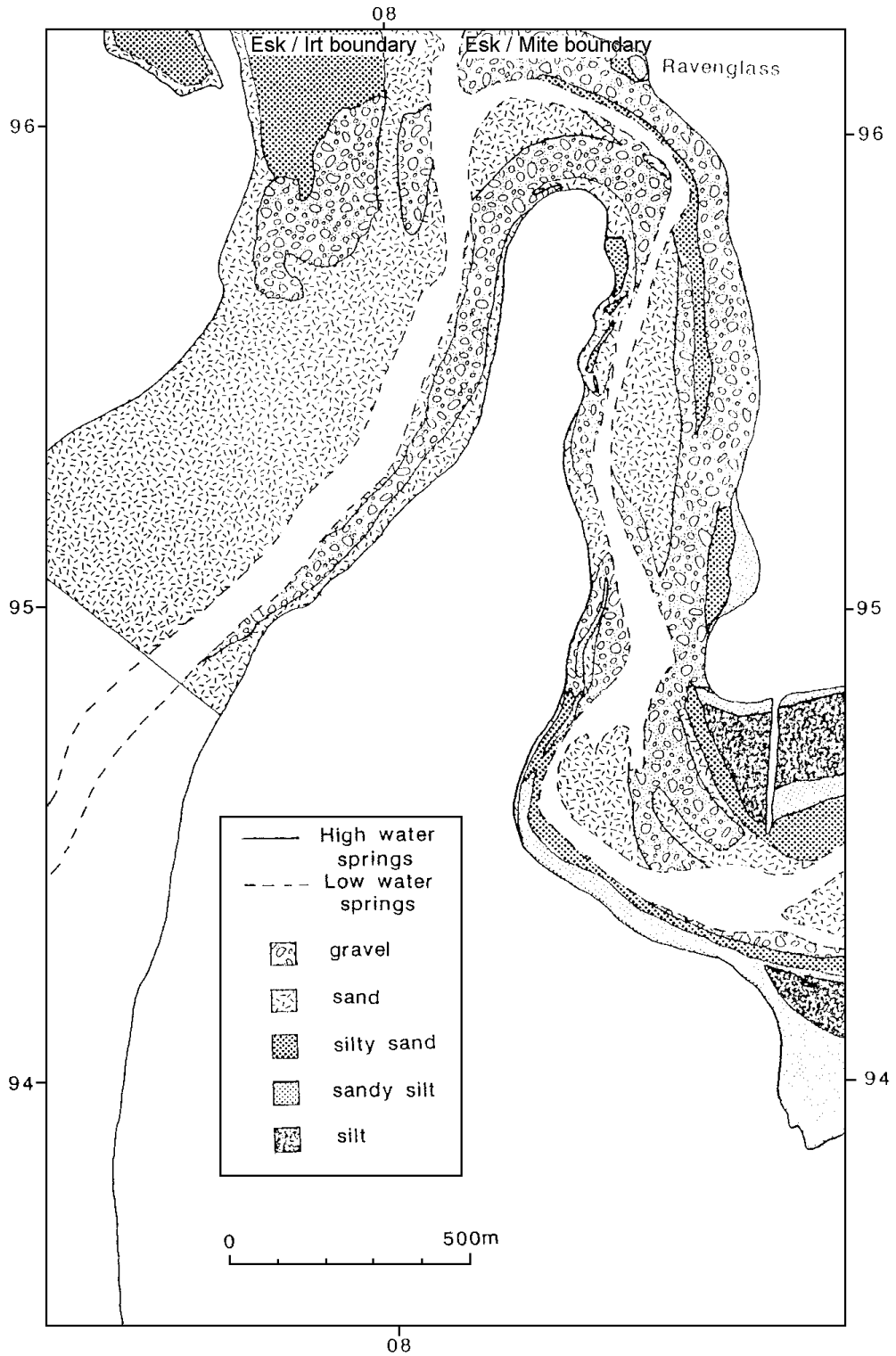
From Kelly and Emptage (1992).

c) Grain size distribution for the upper Esk estuary.



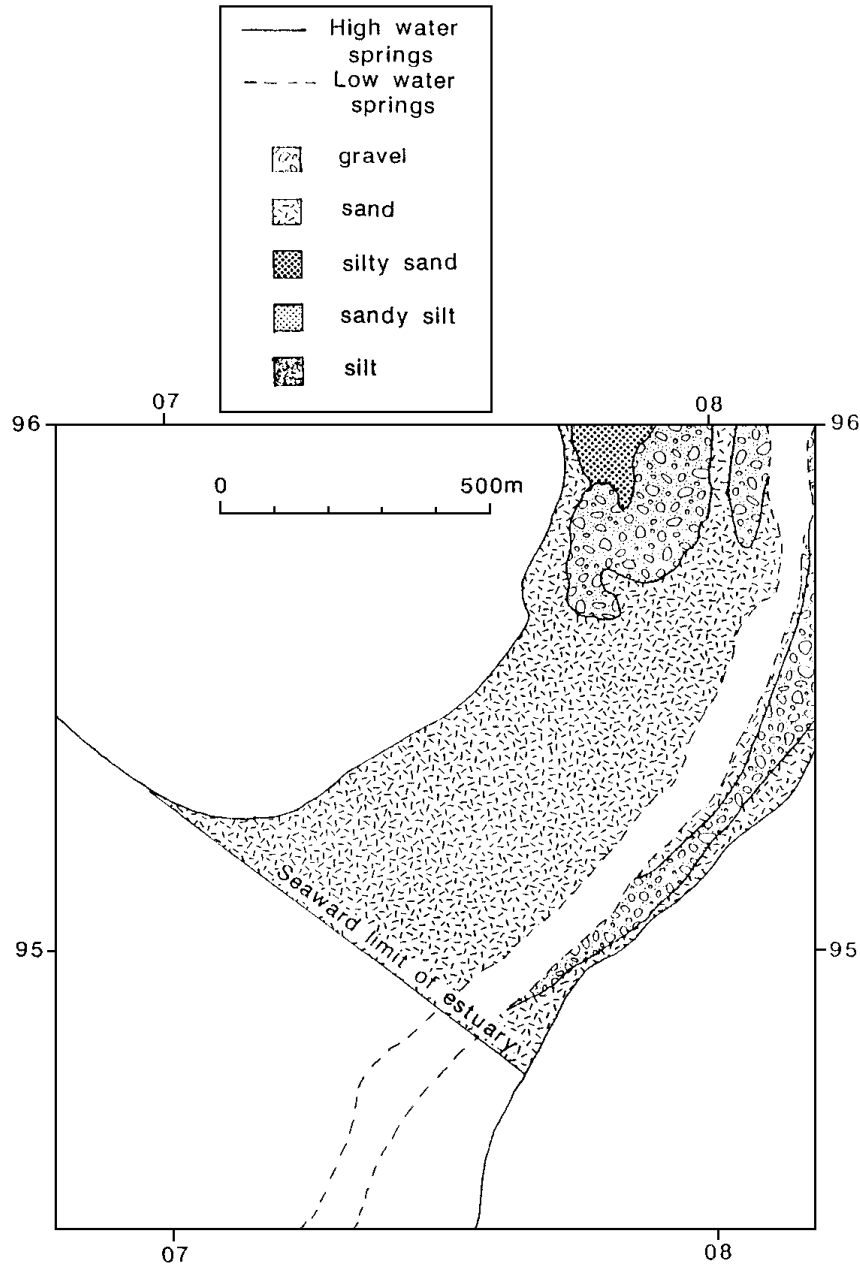
From Kelly and Emptage (1992).

d) Grain size distribution for the middle Esk estuary.



From Kelly and Emptage (1992).

e) Grain size distribution for the lower Esk estuary.



From Kelly and Emptage (1992).

f) Grain size distribution at the mouth of the Esk estuary.

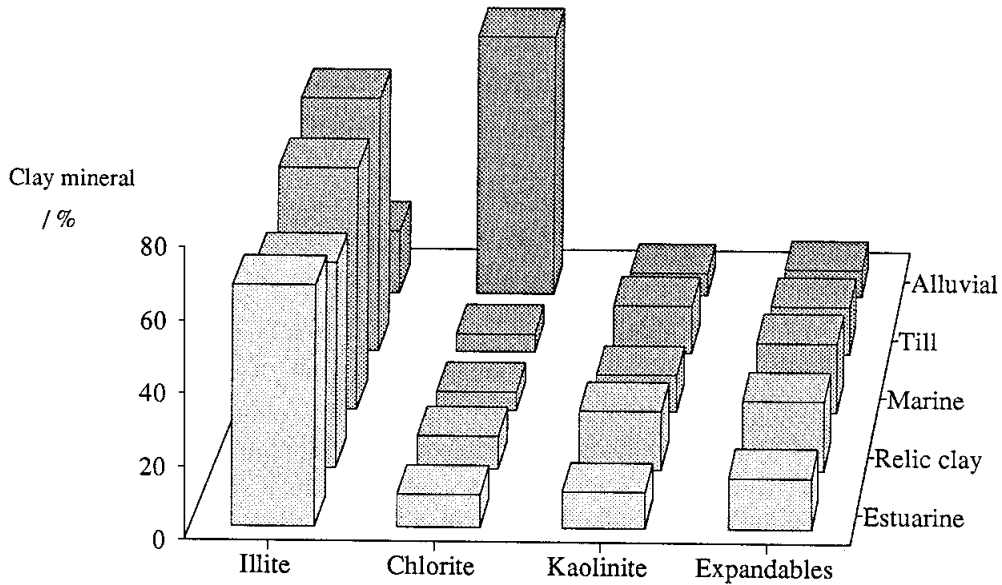
c) Mineralogy

The mineralogy of the sediment has an influence on the grain size characteristics, as well as providing an indication of their source or sources. Amongst the potential sources for the sediments of the Ravenglass estuary are the rocks and sediments on the sea bed, around the estuary margin and on the river catchments.

Qualitative analysis shows the dominant minerals in sands and silts to be quartz, with smaller amounts of feldspars and clay/micas (Kelly and Emptage, 1992). In estuarine sediments, calcite is a common minor mineral, whereas amphiboles are a minor component in the river sediments. Both minerals are present in rock fragments in the sediment, especially the sand grain sizes.

Semi-quantitative analysis of the clay mineral/mica content of the <2 µm fraction of the sediment indicates a high content of illite and expandable clays, the latter mainly mixed lattice clays including

illite-smectite (see Table 7). This compares very closely with the clay mineralogy of offshore marine sediments.



From Kelly and Emptage (1992).

Figure 5 Semi-quantitative clay mineral analysis of Ravenglass estuary sediments.

Although glacial sediments form the floor of much of the estuary, the scope for direct erosion of these to supply material for the modern sediments is almost non-existent. Therefore, the principal source of sediment is thought to be the seas, derived from marine sediments and, ultimately, from the glacial sediments exposed on the sea floor and in coastal cliffs, which provide the source material for the marine sediments themselves. In contrast, sediment transport by the rivers appears to have a limited influence on the sedimentology of the estuary.

d) Estimated mean annual sedimentation rates

Sedimentation may be measured directly (e.g. using marker posts) or indirectly (e.g. from radioactivity profiles), and will clearly vary between areas. Kelly and Emptage (1992) extensively measured sediment accumulation throughout the Ravenglass estuary using these techniques. A summary of the mean sedimentation rates on different facies is given in Table 3.

Table 3 Sedimentation rates for sediment facies in the Ravenglass estuary.

	Sediment facies					
	Total	Relic	Channel	Erosional	Lower bank	Upper bank
Area / km ²	5.7	0.5	1.0	1.0	0.2	2.0
Sedimentation rates						
Direct / kg m ⁻² a ⁻¹	n/d	n/d	n/d	n/d	28 ± 23	5 ± 3
Direct / mm a ⁻¹	n/d	n/d	n/d	0 ± 2	28 ± 27	4 ± 3
¹³⁷ Cs derived / mm a ⁻¹	4.5	n/d	11 ± 4	4 ± 2	11 ± 7	4 ± 3
Direct / 10 ³ tonne a ⁻¹	n/d	n/d	n/d	n/d	6.0	9.5

Uncertainty corresponds to one standard deviation.

From Kelly and Emptage (1992).

Overall, the impression is that the Ravenglass Estuary area is relatively dynamic with locally important erosional processes (e.g. Saltcoats) and saltmarsh stability measured in tens of years, whilst elsewhere accretion is leading steadily to grazing pastures on time-scales of hundreds of years.

Certain low-lying mudbanks (erosional facies) may have negative or low-positive sedimentation rates. These are either sites which are beginning to be sites of bank facies accumulation or are relic areas of such sediments undergoing erosion. In both cases they are due to changing conditions within the estuary. These deposits are devoid of vegetation even when at an elevation where it would be expected. Kelly and Emptage (1992) measured sediment accumulation at two adjacent sites on the Newbiggin mudflat (see Figure 6). One site (0909E 9419N) was on the edge of an erosional channel and demonstrated long periods where such sites can remain relatively inactive. The rate of accretion ranged from essentially zero to around 3 mm a⁻¹. Previous studies indicate a range of erosional loss of 10 mm a⁻¹ to an accretion of 71 mm a⁻¹, with a suggested mean overall accretion of about 5 mm a⁻¹ (e.g. Carr and Blackley, 1985, 1987; Stanners and Aston 1981; Heaton and Hetherington, 1984; Hetherington, 1976, 1978; Aston and Stanners, 1979; Clifton and Hamilton, 1982; Hamilton and Clarke, 1984; Bennett *et al.*, 1988). As a comparison the second site (0909E 9418N) is located at the margin between an erosional and accumulating area, and display significantly higher sedimentation rates.

The channel facies exhibit a nett deposition, although this is considered to be highly sporadic: related to an irregularity of significant sand inputs and to the frequency of reworking in this high flow velocity environment. Limited data give a mean sedimentation rate of 11 mm a⁻¹ but this is likely to under-represent the areas of this facies which have low sedimentation rates.

Sedimentation rates in the bank facies also tend to be variable (<1-100 mm a⁻¹) and correlate with the relative elevation of the surface, which determines the frequency and duration of tidal inundation and the proximity to the channel margin. The total sediment accumulation rate for this facies is estimated at 15.5 × 10³ tonne a⁻¹, mainly from settling of the fine grained suspended sediment load of this water. Within this facies, the lower bank facies, which occupy a relatively small part of the estuary (4%), tend to exhibit high sedimentation rates (mean of 28 mm a⁻¹), which result in an annual accumulation rate of 6.0 × 10³ tonne a⁻¹. The rapid growth of these deposits results from their low elevation and proximity to the channel, which allows coarser sediment deposition.

The upper salt marshes and intertidal pastures have low sedimentation rates (mean 4 mm a⁻¹) which show a seasonal cycle, with peaks in spring and autumn; related to a similar cycle in tidal amplitudes and, hence, in frequency and duration of inundation. Grain sizes tend to be finer in these areas than elsewhere in the estuary.

Kelly and Emptage also expressed the mean sedimentation rate for the upper and lower bank facies in terms of the different sectors of the estuary (see Table 4).

Table 4 Estimated total annual sedimentation rates for bank facies for sectors of the Ravenglass estuary.

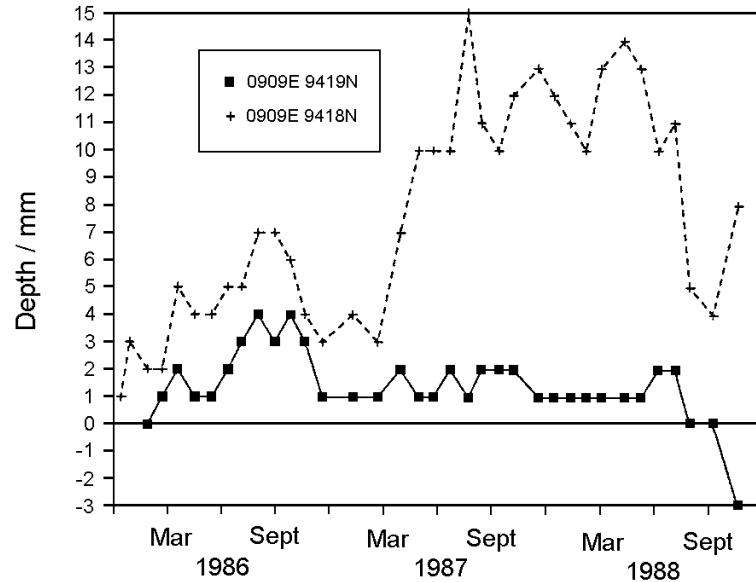
Sediment facies	Sedimentation rate / 10 ³ tonne a ⁻¹		
	Total	Upper bank	Lower bank
Region			
Irt estuary	5.5	3.5	2.0
Mite estuary	0.8	1.0	0.8
Esk estuary	8.2	5.0	3.2
Whole estuary	15.5	9.5	6.0

From Kelly and Emptage (1992).

The residence time of the sediment associated radionuclides in the estuary is related to the potential for reworking of the deposits of the different facies. This primarily occurs by lateral bank erosion initiated by the migration of the channels, where this is permitted by the geology and man's activities.

This may be followed by surface erosion, as in the erosional facies, where measured rates of erosion vary from 0 to 6 mm a⁻¹.

For the major reservoirs of activity, in bank facies sediments, it is estimated that those of the lower bank facies may be being reworked on time-scales of tens of years, whereas for the upper bank facies this is more probably hundreds of years. At present, bank erosion is very localised, although rates of bank retreat in these areas may be high, e.g. 14 m over 14 years.



From Kelly and Emptage (1992)

Figure 6 Sediment accretion/erosion characteristics on a mudflat, Newbiggin.

3.1.3 Pedology

The soil around Ravenglass has been described by Jarvis *et al.* (1984). The soils are part of the Sandwich association. This consists of deep calcareous and non-calcareous sandy soils on sand dunes, marine shingle and related beach deposits. The dunes, which rise to over 25 m above Ordnance Datum are unstable in places. The association contains a variety of soils. The area is exposed everywhere to the sea winds and soil development on the dunes is largely dependent upon the vegetation cover and the degree to which this has stabilised the dune system.

The most extensive soil is the Sandwich series (typical sand pararendzinas) found on sand dunes, usually over 100 years old and often considerably older. Here the vegetation is dominated by herbs, brambles, grasses and deciduous scrub.

Younger, unstable dunes, particularly those nearest the sea, are unvegetated or only thinly colonised by marram grass (*Ammophila arenaria*) and lyme grass (*Leymus arenarius*). Their shape constantly changes, as a result of wind erosion and deposition. Raw sands predominate here.

Soils of the Bechfoot series (typical sand-rankers) occur on stabilised non-calcareous dunes. Wind erosion ceases only when the land surface nearly reaches groundwater, at which stage small hollows (slacks) are formed. Larger elongated depressions result from the enclosure of lengths of beaches by a line of newly formed sand dunes.

Typical sandy gley soils of the Formby series occur in wet hollows among the dunes.

As sand is often blown on to fixed dunes, buried topsoils are widespread and it is common for several to occur in one profile where they form distinctive organic-rich layers.

3.1.4 Meteorology

Meteorological data was supplied by the Meteorological Office at the Eskmeals weather station (700E 402N). The mean monthly and annual statistics are summarised in Table 5.

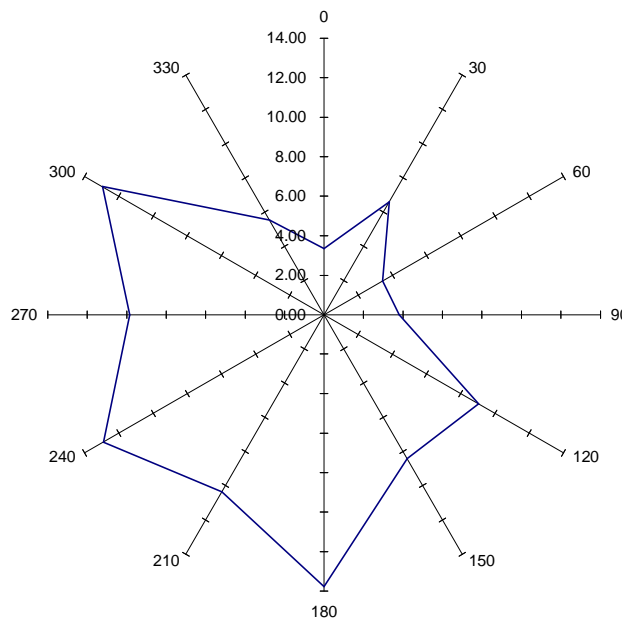
The differences between the monthly distributions of rainfall and potential evaporation, shown in Table 5, suggests that infiltration will occur in the period September to April.

Table 5 Meteorological Data for Ravenglass Estuary (1991-1995).

Month	Max Temp (°C)	Min Temp (°C)	Windspeed (m s ⁻¹)	Irradiation (MJ m ⁻² day ⁻¹)	Rainfall (mm month ⁻¹)	Total Potential Evaporation (mm month ⁻¹)
January	7.2	2.2	5.7	1.90	102.8	12.3
February	7.4	2.2	5.0	3.86	68.8	14.8
March	9.1	3.8	5.3	7.24	74.2	27.8
April	11.6	5.0	4.7	12.51	75.5	44.8
May	15.1	7.4	4.0	16.51	53.8	73.1
June	16.9	9.8	3.6	17.66	49.9	73.1
July	19.0	12.6	3.9	16.27	81.8	82.4
August	18.5	11.9	3.9	14.08	73.8	72.5
September	16.5	9.6	4.0	9.73	78.2	48.9
October	13.0	6.9	4.3	5.46	102.3	29.7
November	10.3	4.9	5.2	2.51	113.8	16.5
December	7.7	2.5	4.8	1.44	107.7	12.2
Annual	12.7	6.6	4.5	9.10	81.9	42.3

Data supplied by the Meteorological Office.

The wind rose, shown in Figure 7, shows that, predominantly, the winds originate from the south west.



Note: The wind rose indicates the direction from which the wind originated.

Data supplied by the meteorological Office.

Figure 7 Wind rose for the Ravenglass estuary.

3.1.5 Hydrology

a) Inputs from rivers

The sources of the three rivers which enter the Ravenglass estuary are located in the Lake District National Park. The Rivers Esk and Mite are primarily fed by small streams from the surrounding hills. However, the principle source of the River Irt is the Wastwater lake. There are no industries located on these rivers, but the rivers pass through agricultural land.

The Environment Agency has responsibility for monitoring the flow rates of the Rivers Irt, Mite and Esk. The flow rates of the Irt and Esk are monitored daily whilst the Mite is monitored periodically. The average monthly flow rates for the Irt and Esk are summarised in Table 6. Unfortunately, the data for the River Mite were too sparse to determine monthly values. However, the Mite rises in the same area as the Esk, and as such, is expected to show a very similar monthly trend to that of the Esk.

Table 6 shows that the Rivers Esk and Irt demonstrate slightly different seasonal patterns. Whilst the Esk is the larger river for most of the year, the Irt can become the larger river during the summer. This is a reflection of the different sources of the river. Wastwater, which feeds the Irt, is a more consistent source of water than the streams which feed the Esk.

Table 6 Mean monthly flow rates of the three main rivers into the Ravenglass Estuary.

	River Irt ¹			River Mite ²			River Esk ³		
	m ³ s ⁻¹			m ³ s ⁻¹			m ³ s ⁻¹		
	Mean	Max	Min	Mean	Max	Min	Mean	Max	Min
January	4.16	18.01	0.66	--	--	--	5.89	58.23	0.65
February	3.51	18.27	0.37	--	--	--	4.41	30.11	0.53
March	4.85	15.01	0.74	--	--	--	6.18	35.50	0.74
April	3.60	15.05	0.65	--	--	--	4.24	40.59	0.51
May	1.83	12.17	0.29	--	--	--	2.37	32.18	0.23
June	2.16	11.14	0.20	--	--	--	2.17	21.15	0.13
July	2.65	8.58	0.28	--	--	--	2.60	16.50	0.15
August	2.67	9.99	0.30	--	--	--	3.04	25.32	0.19
September	2.22	7.45	0.28	--	--	--	2.57	17.44	0.24
October	3.44	13.09	0.44	--	--	--	4.43	28.72	0.56
November	4.52	19.02	0.37	--	--	--	5.93	36.03	0.54
December	4.87	24.02	0.37	--	--	--	6.69	75.06	0.47
Mean	3.36	24.02	0.20	0.43	1.33	0.09	4.19	75.06	0.13

Data courtesy of Environment Agency

1. River Irt sampled daily at Galesyke (NGR NY 13560E 03820N) between 1 January 1991 and 31 December 1995.
2. River Mite occasionally sampled at Muncaster Hill (NGR SD 09401E 97704N) between 25 January 1987 and 15 September 1993.
3. River Esk sampled daily at Cropple How (NGR SD 13100E 97770N) between 1 January 1991 and 31 December 1995.

The water quality of the three rivers is also monitored by the Environment Agency. The agency monitors the compositions of the three rivers at approximately one month intervals. The water quality of the Rivers Esk and Irt are both measured at two locations.

Mean water quality data for the three rivers at each sampling location are summarised in Table 7.

The Holmrook sampling point on the River Irt and the Muncaster Mill Bridge on the River Mite were both tidal. Conductivities exceeding 20000 $\mu\text{S cm}^{-1}$ were sometimes reported. To ensure that the data reflected the composition of the river as opposed to that of the sea water, measurements where the conductivity exceeded 160 $\mu\text{S cm}^{-1}$ were discarded.

Table 7 Water quality of the three main river which flow into the Ravensglass Estuary.

Property	Units	River Irt					
		Santon Bridge NY 11015E 01607N			Holmrook* SD 08010E 99523N		
		Samples	Mean	Stand.dev.	Samples	Mean	Stand.dev.
pH		49	6.94	0.33	74	7.09	0.39
Temperature	°C	49	10.1	3.8	76	10.0	3.9
Suspended Solid	g m ⁻³ (at 105°C)	49	3.05	4.80	74	3.41	5.05
	g m ⁻³ (at 500°C)	49	5.76	3.48	73	4.43	2.90
Dissolved O ₂	%	18	106.4	6.7	21	102.0	6.2
	g m ⁻³ (as O)	20	11.7	1.1	21	11.3	1.3
BOD	g m ⁻³ (as O)	49	1.04	0.46	73	1.08	0.81
COD	g m ⁻³ (as O)	18	10.3	5.7	35	12.6	15.1
Conductivity	µS cm ⁻¹	49	66.0	12.6	74	93.3	26.2
Total hardness	g m ⁻³ (as CaCO ₃)	22	16.0	3.3	38	23.8	8.0
Alkalinity	g m ⁻³ (as CaCO ₃)	47	10.7	13.8	71	17.8	19.6
NH ₃	g m ⁻³ (as N)	49	0.033	0.040	65	0.069	0.294
NO ₃	g m ⁻³ (as N)	48	0.635	0.301	64	0.867	0.325
NO ₂	g m ⁻³ (as N)	49	0.005	0.004	66	0.009	0.012
PO ₄	g m ⁻³ (as P)	49	0.029	0.012	64	0.026	0.116
		River Mite					
		Muncaster Mill Bridge* SD 09401E 97704N					
		Samples	Mean	Stand.dev.			
pH		77	7.14	0.49			
Temperature	°C	78	10.0	3.9			
Suspended Solid	g m ⁻³ (at 105°C)	79	6.50	12.9			
	g m ⁻³ (at 500°C)	74	5.80	6.41			
Dissolved O ₂	%	20	100.0	6.7			
	g m ⁻³ (as O)	20	10.9	1.1			
BOD	g m ⁻³ (as O)	78	1.17	0.62			
COD	g m ⁻³ (as O)	33	12.1	10.6			
Conductivity	µS cm ⁻¹	77	133.4	30.4			
Total hardness	g m ⁻³ (as CaCO ₃)	36	40.0	60.1			
Alkalinity	g m ⁻³ (as CaCO ₃)	74	16.2	7.2			
NH ₃	g m ⁻³ (as N)	66	0.097	0.344			
NO ₃	g m ⁻³ (as N)	65	1.215	0.524			
NO ₂	g m ⁻³ (as N)	66	0.011	0.023			
PO ₄	g m ⁻³ (as P)	65	0.164	0.716			
		River Esk					
		Whahouse Bridge NY 20355E 00893N			Cropple How Gauging Station SD 13098E 97770N		
		Samples	Mean	Stand.dev.	Samples	Mean	Stand.dev.
pH		55	6.13	0.57	79	6.59	0.40
Temperature	°C	51	9.2	4.4	72	9.6	4.4
Suspended Solid	g m ⁻³ (at 105°C)	51	1.79	0.68	73	2.13	3.07
	g m ⁻³ (at 500°C)	51	5.03	2.05	73	4.06	2.72
Dissolved O ₂	%	18	98.7	3.4	18	97.3	4.2
	g m ⁻³ (as O)	20	11.1	1.5	20	10.9	1.6
BOD	g m ⁻³ (as O)	51	1.03	0.46	73	1.03	0.54
COD	g m ⁻³ (as O)	16	11.1	6.4	33	8.5	5.3
Conductivity	µS cm ⁻¹	55	41.8	22.8	79	58.3	17.4
Total hardness	g m ⁻³ (as CaCO ₃)	23	7.5	1.9	37	11.4	3.2
Alkalinity	g m ⁻³ (as CaCO ₃)	53	4.7	2.3	76	7.5	6.0
NH ₃	g m ⁻³ (as N)	55	0.030	0.036	78	0.026	0.033
NO ₃	g m ⁻³ (as N)	55	0.403	0.274	78	0.473	0.285
NO ₂	g m ⁻³ (as N)	55	0.005	0.004	78	0.006	0.003
PO ₄	g m ⁻³ (as P)	55	0.028	0.013	77	0.022	0.015

Data courtesy of Environment Agency

* This section of the river is tidal

b) Input from Drigg stream

The input from the Drigg stream was is 0.03-0.05 m³ s⁻¹. A detailed description of this stream is given in Technical Deliverable TD9.

c) Material balance for estuary

There have been two reported of attempts at balancing discharges into and out of the estuary (Tables 8 and 9). Both sets of data are given to illustrate the variations in these measurements.

Table 8 Mass balance of water, salt and sediment in the Ravenglass estuary (1985).

Component	Units	Flood tide	Ebb tide	Balance per 12 hr
Water	10 ⁶ m ³	3.9	4.6	-0.8
Salt	10 ⁶ kg	93	93	0
Sediment	10 ³ kg	100	140	-30

Data collected 24 June 1985, three days after a Spring tide near mouth of estuary. From Burton and Yarnold (1988).

Table 9 Mass balance of water, salt and sediment in the Ravenglass estuary (1987).

Component	Units	Flood tide	Ebb tide	Balance per tide
Water	10 ⁶ m ³	3.13	3.27	-0.14
Salt	10 ⁶ kg	93.4	100.8	-7.4
Sediment	10 ³ kg	106.4	88.8	+17.6

Data collected 6 October 1987, on a 9.23 m spring tide at five points at mouth of the River Esk branch of the estuary. Depth varied between 5.26 m and 0.65 m, width varied between 272 m and 37 m. From Mudge et al. (1989).

It is interesting to note that whilst there is reasonable agreement between the two sets of data for discharge of water and salt content, the sediment balances differ. This arises from differences for the ebb tide and may well be the result of turbulence at the time of one of the measurements. The first set of measurements were taken when there was a “stiff westerly breeze of 5-7 m s⁻¹ which produced an appreciable swell in the estuary around high tide”.

3.1.6 Population (characteristics)

a) Demography

The Ravenglass estuary lies in a sparsely populated area of Cumbria. The total population of the local authority area (Copeland Borough Council) is about 71,000 the major part of which reside in the three local towns of Whitehaven (27 km north of the estuary), Egremont (18 km north of the estuary) and Cleator Moor (21 km north-east of the estuary). To the south of of the estuary there are no significant centres of population until the town of Millom (18 km south-east of the estuary) with a population of about 7,200.

The Ravenglass estuary lies in the Parish of Drigg and Carlton (population of 425). Two other local parishes lie nearby. Muncaster Parish with a population of 325 and Waberthwaite with a population of 275 (Copeland Borough Council, 1998).

The village of Ravenglass which lies on the Ravenglass estuary has a population of about 200. A number of scattered farms and clusters of a few cottages are in the vicinity of the Ravenglass estuary, which may have a combined population of about 125.

b) Habits

Critical group characteristics for Sellafield are defined in Table 10. These groups may be applicable to Ravenglass for the purposes of calculating limiting doses which may be incurred, but it should be noted that most dose calculations in the vicinity of Ravenglass would, in practice, be dominated by Sellafield derived discharges.

Table 10 Critical group consumption and occupancy rates defined for Sellafield.

Foodstuff	Consumption rate (kg person ⁻¹ a ⁻¹)		
	Adults	Children	Infants
Cod & plaice ^a	34.75	Marine consumption pathways expressed on basis of adult consumption ⁱ	
Crab & lobster ^b	8.25		
Winkles	7.79		
Other molluscs ^c	2.16		
Milk	258.9		
Beef	29.3	14.7	7
Mutton	9.2	2.7	3
Sheep liver	1.6	0.9	2
Pig meat ^h	100	81.6	0
Green veg	22.2	14.4	30
Root veg ^e	48.5	30.2	50
Mushrooms ^h	0.8	0	0
Fruit ^f	11.0	5.6	1.2
Poultry	5.9	3.7	3
Eggs ^g	28.9	15.2	14.5
Honey ^j	6.72	n/a	
Venison ⁱ	134.4		
Other game ^j	17		
Occupancy pathways	Occupancy of area (h a ⁻¹)		
	Adult	Children	Infants
Whitehaven boat dweller	610	n/a	
Site perimeter	full occupancy		
Beach/leisure	1000		
Farmer (ploughing)	300		

Notes:

- a. A mix of 50% cod and 50% plaice is assumed.
- b. A mix of 70% crab and 30% lobster is assumed.
- c. Assumed to be an equal mix of cockles, mussels and limpet.
- d. Based on generalised NRPB (1987) recommendations
- e. Local potatoes only
- f. Soft fruits only
- g. Hens eggs only
- h. From Stewart et al. (1990), data for infants relate to 1-2 years.
- i. The critical group may include children 'scaled' to adults.
- j. From Fulker et al. (1996); maximum consumption rates reported

From BNFL (1995), except where otherwise stated

It should also be noted that critical group consumption rates are not simply additive. In fact, relatively little crossover between pathways is thought to occur. For dose calculation purposes it is usual to assume that marine pathway groups consume all seafoods at critical group rates, but these are not the same group as the Whitehaven boat dwellers. Critical group consumers of food do not generally eat elevated levels of fish etc. Caution should thus be exercised in calculating reasonable upper dose limits based on the critical group consumption rates given above.

Habits data applicable to the general population around Ravenglass can probably be modelled from UK national consumption data (e.g. Byrom *et al.*, 1995). A summary of consumption data for UK adults is presented in Table 11.

Table 11 Mean consumption of food by adults (16 to 64 years) in the UK.

Food group	Percentage of population as consumers	Mean consumption of consuming group (kg person ⁻¹ a ⁻¹)	97.5 th percentile consumption of consuming group (kg person ⁻¹ a ⁻¹)
Domestic fruit	76.3	20	50
Potatoes and root vegetables	100	50	95
Green and other domestic vegetables	96.6	15	35
Mushrooms	17.1	1.5	4.5
Honey	3.9	2	7.5
Pig meat	94.6	8.5	25
Cattle meat	96.9	15	30
Sheep meat	41.0	4	10
Offal	36.6	3	10
Poultry	68.0	5.5	15
Game	0.3	4	7.5 [†]
Milk	99.9	110	240
Butter, cheese and other milk products	98.3	15	45
Eggs	96.3	6.5	20
Fish	69.3	6	20
Shellfish	3.2	2.5	7

[†]. Based on a very small sample size (less than 40 consumers from the national survey). From Byrom *et al.* (1995).

c) Activities

The Ravenglass estuary is bounded by sand dunes, salt marsh/rough pasture with a nature reserve, the Drigg low level waste site to the north and a military firing range to the south. This land affords poor grazing to sheep only. To the north and east the land is more fertile, with mainly dairy cattle. Beef cattle and overwintering sheep may also be farmed in this area. A few root crops are grown for human consumption (mainly potatoes), although the majority of such crops (e.g. beets) are used as overwintering fodder for animals. Silage is also produced for overwintering, both as bale and clamp silage. Very limited cereal crops are grown anywhere near Drigg and such cereals are intended as animal fodder (e.g. some maize is grown approximately 10-12 km north-east of the site at Gosforth).

Very limited commercial fishing is undertaken around the estuary. One or more part time fishermen maintain boats at Ravenglass, but the main fishing ports are at Whitehaven, Workington and the Isle of Man. Mussel beds at Ravenglass have at various times been exploited commercially. Collection of shellfish (mussels, winkles, limpets, etc.) off the Cumbrian coast for personal consumption remains relatively common. However, there is no longer any commercial shellfish industry in the estuary. Scallop beds to the south and east of the Isle of Man are unlikely to receive any measurable input of radioactivity from the Drigg site, even in the absence of the influence of Sellafield discharges.

Commercial exploitation of the edible seaweed *Porphyra umbilicalis* ceased during the 1960's and no collection for personal consumption is known to occur. Seaweed derived fertilisers are not known to be utilised by local farmers.

For modelling purposes, the main potential source of contamination arising from the estuary is likely to be groundshine, and sheep meat consumption.

3.2 Physical-chemical characteristics

3.2.1 The water column

Nearly all compartments of the BIOPATH model (see Section 5) for the Ravensglass Estuary contain an aquatic phase, namely: the Irish Sea, the channel connecting the mixing zone of the three rivers (Irt, Mite, and Esk) with the open sea, the erosion zone, the lower banks and the upper banks. No analytical data specific for this location is available. Extrapolations, using data sets collected nearby, could be justified only for the composition of a typical water sample from the Irish Sea. In case of the estuary itself, however, a sampling campaign was necessary. Therefore, as part of the RESTRAT project, Westlakes Scientific Consulting took several samples between July 1996 and March 1997, which were assigned to the various compartments as follows:

- one sample of water from 50 m outside the mouth of the Ravensglass Estuary, representing the Irish Sea (to be combined with generic data);
- two samples of water from 50 m inside the mouth and from the barrier at the mouth, both representing the channel;
- one sample of water from the Eskmeal viaduct, representing the lower banks;
- one sample of water from the Drigg stream confluence of the River Irt, representing the upper banks.

Unfortunately, no sample could be obtained from the erosion zone. An additional sample was taken from the sediment of the estuary to allow analysis of the pore water. This uneven sample distribution also, to a certain degree, reflects the major changes that occurred in the BIOPATH compartment structure throughout most of the RESTRAT project. As anticipated, the Ravensglass Estuary turned out to be indeed the most complex test example case of the project.

The sampling procedure was in most cases accompanied by *in-situ* measurements of pH, temperature, redox potential Eh and oxygen content. All samples were then shipped to and analysed at the FZ Rossendorf. The data in Appendix C summarise the original analytical data. In a second step, these results were checked for consistency by means of chemical speciation modelling with EQ3/6 (Wolery, 1995).

Modelling gave a ionic strength of 0.57 mol L^{-1} for the Irish Sea, with a total charge imbalance of only 3.7%. The low redox potential value from the *in-situ* measurements could not be supported by the modelling, the water is expected to be much more oxidising.

As expected, the modelled ionic strength for the channel was similar with 0.56 mol L^{-1} ; close to the value of the open sea. Here, the measured low Eh can be explained by interactions with a reducing sediment. This would especially influence the speciation of plutonium.

Water from the lower bank, although sampled at high tide, already show a significant decrease in ionic strength (here 0.32 mol L^{-1}). The carbonate content was not measured, modelling of an equilibrium with the atmosphere gave the reasonable value of 1.33 mmol L^{-1} , corresponding to a charge imbalance of only 1.0%.

The sample from the upper banks, also collected at high tide had, at 0.061 mol L^{-1} , the lowest ionic strength. The carbonate content had also to be found through modelling. The value in equilibrium with air was $1.085 \times 10^{-4} \text{ mol L}^{-1}$, corresponding to a charge imbalance of only 1.6%.

The chemical composition of the main aqueous phases involved in the modelling clearly indicates, that the composition of the estuary water is nearly identical to that of the Irish Sea, due to the strong tidal effects. That means a high ionic strength due to the large amounts of sodium and magnesium chlorides and sulphates. In all cases, silica seems to be in equilibrium with either quartz or chalcedony.

The waters are supersaturated with respect to several manganese and iron minerals. However, these may not form due to kinetic hindrance. The pH becomes slightly more basic from the river through the estuary to the open sea.

In the case of the pore water sample from the mussel beds, reducing conditions were expected. The modelling yielded accordingly a redox potential of +218 mV. Contrary to all other compartments, silica is not in equilibrium with quartz, but strongly supersaturated. There are further supersaturations with respect to the solid phases magnesite (MgCO_3) and dawsonite ($\text{NaAl}(\text{OH})_2\text{CO}_3$). This may precipitate in the long time frame selected for the risk assessment in RESTRAT. The ionic strength was 0.41 mol L^{-1} , close to the high mineralisation values for marine waters. The quality of the chemical analyses which, for unknown reasons, were not as good as for the other samples, indicated by a charge imbalance of 9.4%.

The modelling results discussed above were combined with the experimental data to yield a set of selected best values. They are listed in Table 12.

3.2.2 Solid phases

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

A preliminary mean sedimentation rate for the channel facies is 11 mm a^{-1} , but deposition should be considered highly sporadic, partly in association with mobile beds. The main channel system is a high flow velocity environment. Lower bank facies have a relatively high sedimentation rate of 28 mm a^{-1} , whereas the upper bank facies (salt marshes and intertidal pastures) sediment at only 4 mm a^{-1} . The hydrology is strongly influenced by seasonal temperature changes and the circulation patterns of the Irish Sea.

The characterisation of the relevant solid phases is, by far, not as satisfactory that of the aqueous phase. There are only rather general geological investigations for this site published, and the capacities of the institutions taking part in the RESTRAT project did not allow a proper mineralogical characterisation. Based on the mud sample, an elementary analysis of the solid parts was performed (ICP-MS and F-AAS after addition of nitric acid and microwave digestion). The carbon and oxygen content was not determined, but the results for the other components showed a strong dominance of calcium (36.5 mol%), aluminium (27.6 mol%), magnesium (13.2 mol%), and iron (12.7 mol%) over silica and the alkaline cations, suggesting a very low content of quartz and silicates.

Analysis was complicated by a considerable content of organic matter of various origins and in different state of degradation. This reflects the need for further investigations to be performed to specify a more detailed the mineralogical background of the site.

Table 12 Selected best set of analytical values for the Ravenglass Estuary compartments.

Component	Compartment / Abbreviation												
	Upper Banks UB		Lower Banks LB		Channel CH		Sediment SD		Irish Sea IS				
	Mean mol L ⁻¹	STD mol L ⁻¹	Mean mol L ⁻¹	STD mol L ⁻¹	Mean mol L ⁻¹	STD mol L ⁻¹	Mean mol L ⁻¹	STD mol L ⁻¹	Mean mol L ⁻¹	STD mol L ⁻¹			
PO ₄ ³⁻	7.69 × 10 ⁻⁵	7.45 × 10 ⁻⁶	-	-	-	-	-	-	-	-	-	-	-
NO ₃ ⁻	5.65 × 10 ⁻⁵	6.84 × 10 ⁻⁶	-	-	-	-	-	-	-	-	-	-	-
SO ₄ ²⁻	2.64 × 10 ⁻³	1.47 × 10 ⁻⁵	1.34 × 10 ⁻²	5.80 × 10 ⁻⁴	2.45 × 10 ⁻²	7.36 × 10 ⁻⁴	1.17 × 10 ⁻²	1.03 × 10 ⁻³	2.67 × 10 ⁻²	5.89 × 10 ⁻⁴	1.03 × 10 ⁻³	2.67 × 10 ⁻²	5.89 × 10 ⁻⁴
CO ₃ ²⁻	1.09 × 10 ⁻⁴ †	-	1.33 × 10 ⁻³ †	-	2.07 × 10 ⁻³	3.48 × 10 ⁻⁵	2.00 × 10 ⁻²	2.55 × 10 ⁻³	1.20 × 10 ⁻³	1.18 × 10 ⁻³	2.55 × 10 ⁻³	1.20 × 10 ⁻³	1.18 × 10 ⁻³
Cl ⁻	4.823 × 10 ⁻²	3.99 × 10 ⁻⁴	2.67 × 10 ⁻¹	4.00 × 10 ⁻³	4.89 × 10 ⁻¹	1.40 × 10 ⁻²	3.82 × 10 ⁻¹	4.99 × 10 ⁻²	5.07 × 10 ⁻¹	7.98 × 10 ⁻⁴	4.99 × 10 ⁻²	5.07 × 10 ⁻¹	7.98 × 10 ⁻⁴
SiO ₂	4.93 × 10 ⁻⁵	5.79 × 10 ⁻⁶	3.06 × 10 ⁻⁵	7.71 × 10 ⁻⁶	3.37 × 10 ⁻⁵	8.98 × 10 ⁻⁶	3.89 × 10 ⁻⁴	8.48 × 10 ⁻⁵	3.59 × 10 ⁻⁵	1.12 × 10 ⁻⁵	8.48 × 10 ⁻⁵	3.59 × 10 ⁻⁵	1.12 × 10 ⁻⁵
K ⁺	9.85 × 10 ⁻⁴	3.62 × 10 ⁻⁵	4.89 × 10 ⁻³	1.94 × 10 ⁻⁴	9.42 × 10 ⁻³	3.55 × 10 ⁻⁴	6.42 × 10 ⁻³	9.04 × 10 ⁻⁴	9.43 × 10 ⁻³	2.74 × 10 ⁻⁴	9.04 × 10 ⁻⁴	9.43 × 10 ⁻³	2.74 × 10 ⁻⁴
Na ⁺	4.23 × 10 ⁻⁴	7.69 × 10 ⁻⁴	2.31 × 10 ⁻¹	1.92 × 10 ⁻³	4.14 × 10 ⁻¹	1.68 × 10 ⁻²	3.01 × 10 ⁻¹	2.77 × 10 ⁻²	4.19 × 10 ⁻¹	1.80 × 10 ⁻²	2.77 × 10 ⁻²	4.19 × 10 ⁻¹	1.80 × 10 ⁻²
Ca ²⁺	1.15 × 10 ⁻³	1.02 × 10 ⁻⁴	5.14 × 10 ⁻³	2.38 × 10 ⁻⁴	9.75 × 10 ⁻³	4.29 × 10 ⁻⁴	6.30 × 10 ⁻³	3.71 × 10 ⁻⁴	9.56 × 10 ⁻³	7.49 × 10 ⁻⁵	3.71 × 10 ⁻⁴	9.56 × 10 ⁻³	7.49 × 10 ⁻⁵
Mg ²⁺	4.57 × 10 ⁻³	2.33 × 10 ⁻⁴	2.58 × 10 ⁻²	2.00 × 10 ⁻³	4.69 × 10 ⁻²	9.12 × 10 ⁻⁴	3.47 × 10 ⁻²	3.84 × 10 ⁻³	4.80 × 10 ⁻²	1.86 × 10 ⁻³	3.84 × 10 ⁻³	4.80 × 10 ⁻²	1.86 × 10 ⁻³
Fe	-	-	-	-	2.39 × 10 ⁻⁶	1.45 × 10 ⁻⁶	5.30 × 10 ⁻⁵	6.94 × 10 ⁻⁵	2.51 × 10 ⁻⁶	-	6.94 × 10 ⁻⁵	2.51 × 10 ⁻⁶	-
Al ³⁺	-	-	-	-	3.78 × 10 ⁻⁶	5.15 × 10 ⁻⁶	4.43 × 10 ⁻⁷	5.64 × 10 ⁻⁷	2.08 × 10 ⁻⁶	-	5.64 × 10 ⁻⁷	2.08 × 10 ⁻⁶	-
Zn ²⁺	5.75 × 10 ⁻⁷	-	5.72 × 10 ⁻⁷	1.43 × 10 ⁻⁷	1.52 × 10 ⁻⁷	3.33 × 10 ⁻⁸	-	-	8.41 × 10 ⁻⁸	1.08 × 10 ⁻⁸	-	8.41 × 10 ⁻⁸	1.08 × 10 ⁻⁸
U	1.81 × 10 ⁻⁹	9.51 × 10 ⁻¹⁰	1.29 × 10 ⁻⁸	3.22 × 10 ⁻⁹	1.67 × 10 ⁻⁸	1.47 × 10 ⁻⁹	2.71 × 10 ⁻⁸	1.16 × 10 ⁻⁸	1.70 × 10 ⁻⁸	8.91 × 10 ⁻¹⁰	1.16 × 10 ⁻⁸	1.70 × 10 ⁻⁸	8.91 × 10 ⁻¹⁰
Pb ²⁺	-	-	-	-	3.62 × 10 ⁻⁹	3.29 × 10 ⁻⁹	1.45 × 10 ⁻⁹	6.83 × 10 ⁻¹⁰	2.90 × 10 ⁻⁹	2.73 × 10 ⁻⁹	6.83 × 10 ⁻¹⁰	2.90 × 10 ⁻⁹	2.73 × 10 ⁻⁹
Ni ²⁺	1.57 × 10 ⁻⁷	-	2.73 × 10 ⁻⁷	4.46 × 10 ⁻⁸	-	-	-	-	-	-	-	-	-
Mn ²⁺	8.71 × 10 ⁻⁷	1.18 × 10 ⁻⁶	3.60 × 10 ⁻⁷	6.56 × 10 ⁻⁸	1.26 × 10 ⁻⁷	1.35 × 10 ⁻⁷	3.01 × 10 ⁻⁴	5.54 × 10 ⁻⁵	1.01 × 10 ⁻⁷	1.08 × 10 ⁻⁷	5.54 × 10 ⁻⁵	1.01 × 10 ⁻⁷	1.08 × 10 ⁻⁷
Cd ²⁺	-	-	-	-	8.90 × 10 ⁻⁹	-	-	-	-	-	-	-	-
As	6.01 × 10 ⁻⁸	-	3.22 × 10 ⁻⁷	3.62 × 10 ⁻⁸	-	-	-	-	-	-	-	-	-
pH:	7.02	0.20	8.04	0.20	7.93	0.10	7.33	0.10	8.07	0.10	0.10	8.07	0.10
Eh / mV:	-	-	-	-	122.6	2.70	218.1	-	129.1	-	-	129.1	-

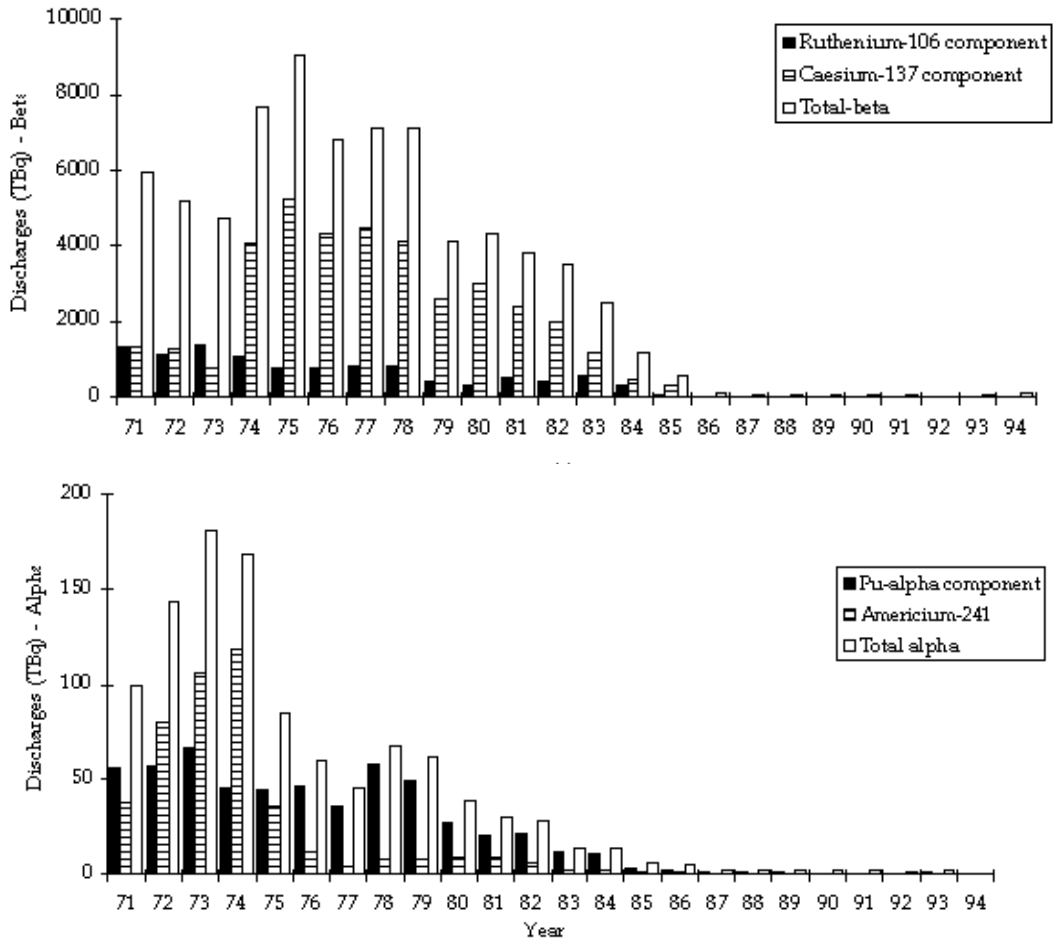
† values set according to the modelling results when no experimental information available. Average values (MEAN) and standard deviations (STD) were calculated assigning equal weights to all listed values. There are no values given for mercury, chromium, copper, and cobalt, because the available data was either too widely scattered, or there are hardly measurements of the values above their respective detection limits.

3.3 Radiological characteristics

The radioactive wastes discharged into the Irish Sea from the Sellafield nuclear fuel reprocessing plant are responsible for nearly all the inputs of artificial radioactivity into the Ravenglass estuary. Since the radionuclides in the discharges can react with sediment particles in the sea, transport into the estuary will occur in both the solution and particulate phases. Of the other potential sources of artificial radionuclides, only fallout from the Chernobyl accident has been identified. This input was in the form of direct fallout on the estuary as well as second only derived material from the catchment and, perhaps, the sea. This source is rapidly declining in significance.

3.3.1 Source term

The discharge levels of radioactivity from Sellafield since the mid 1980's are given in Figure 8. This shows that there has been decline in discharge levels since the introduction of SIXEP and the Salt Evaporator. Further reductions are anticipated with the operation of the Enhanced Actinide Removal Plant (EARP) which began active commissioning in March 1994 and the Segregated Effluent Treatment Plant (SETP) which replaced the old Sea Tanks in July 1994. The reductions in discharges arising from operation of these plants will be offset to some extent by the start up of the Thermal Oxide Reprocessing Plant (THORP), which will eventually reprocess up to 800 tonne a⁻¹ of oxide fuels. Contamination of storage pond water associated with oxide fuel is minimal compared to contamination from Magnox fuel, but discharges of certain isotopes (most notably ⁹⁹Tc) have increased dramatically (see Table 13).



*Total-alpha and total-beta are overall control measures which do not reproduce precisely the contribution of individual nuclides.
From BNFL (1994)*

Figure 8 Marine pipeline discharges from Sellafield.

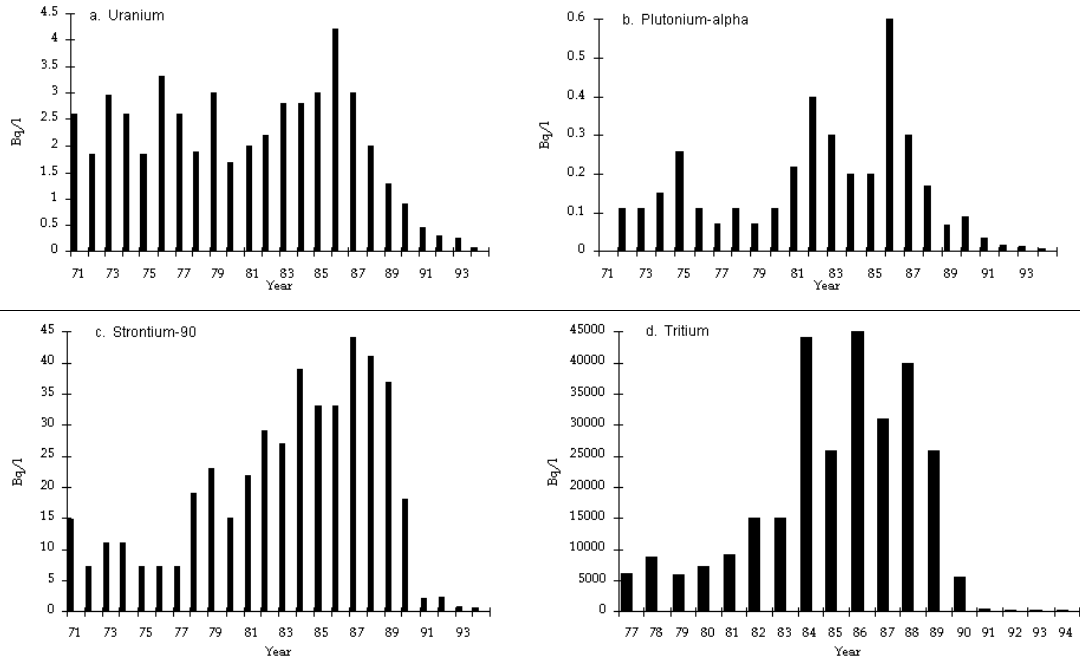
Table 13 Liquid Effluent discharges from BNFL Sellafield.

Nuclide	Annual Discharge (TBq)							
	1988	1989	1990	1991	1992	1993	1994	1995
Tritium	1724	2144	1699	1803	1199	2309	1680	2700
Carbon-14	3.0	2.0	2.0	2.4	0.80	2.0	8.2	12
Sulphur-35	1.0	1.3	1.1	0.96	1.1	0.82	0.39	0.65
Manganese-54	0.05	0.07	0.06	0.06	0.03	0.05	0.07	0.08
Iron-55	0.25	0.24	0.11	0.09	0.13	0.10	0.09	0.04
Cobalt-60	0.96	0.17	0.17	0.09	0.07	0.09	0.11	1.3
Nickel-63	3.2	2.5	0.06	0.12	0.08	0.13	0.40	0.41
Zinc-65	0.09	0.13	0.12	0.12	0.08	0.10	0.11	0.17
Strontium-89	0.21	0.37	0.30	0.18	0.14	0.22	0.33	0.38
Strontium-90	10.1	9.2	4.2	4.1	4.2	17.1	28.9	28
Zirconium-95	5.2	6.5	4.2	7.4	7.0	6.3	2.1	0.34
Niobium-95	4.6	4.6	2.6	5.0	3.3	3.4	1.2	0.40
Technetium-99	4.2	6.1	3.8	3.9	3.2	6.1	72.0	190
Ruthenium-103	0.42	0.54	0.44	0.43	0.33	0.30	0.21	0.19
Ruthenium-106	23.6	25.0	16.5	18.7	12.6	17.1	6.7	7.3
Silver-110m	0.07	0.10	0.09	0.31	0.21	0.24	0.18	0.12
Antimony-125	10.7	15.6	15.2	11.6	9.8	11.9	12.2	9.3
Iodine-129	0.13	0.17	0.11	0.16	0.07	0.16	0.16	0.25
Caesium-134	0.95	1.73	1.15	0.76	0.83	1.18	0.61	0.51
Caesium-137	13.3	28.6	23.5	15.6	15.3	21.9	13.8	12
Cerium-144	3.2	3.8	2.0	1.7	1.7	2.5	0.84	1.1
Promethium-147	2.3	3.6	1.7	1.3	1.2	1.7	0.54	0.61
Europium-152	0.08	0.09	0.08	0.07	0.04	0.05	0.22	0.18
Europium-154	0.08	0.10	0.05	0.05	0.04	0.04	0.11	0.14
Europium-155	0.08	0.10	0.06	0.05	0.04	0.04	0.11	0.076
Neptunium-237	0.28	0.40	0.28	0.29	0.18	0.39	0.33	0.18
Plutonium- α	1.4	1.2	1.14	1.08	0.94	1.33	0.66	0.31
Plutonium-241	36.1	30.2	31.6	29.5	25.3	37.5	14.4	7.7
Americium-241	0.75	1.06	0.75	0.74	0.54	0.87	0.38	0.11
Curium-242	0.06	0.07	0.04	0.03	0.03	0.03	0.009	0.031
Curium-243+244	0.01	0.02	0.01	0.01	0.003	0.004	0.003	0.008
Total- α [†]	2.1	2.7	2.2	2.1	1.6	2.6	1.0	0.40
Total- β [†]	101.0	101.0	70.9	62.3	57.2	97.0	125	190
Uranium (kg)	970	1212	853.0	860.5	633.0	653.7	1388	1300

†. Total- α and total- β are control measures relating to specified analytical determinations. They do not reproduce precisely the contributions from all individual isotopes. In particular, low energy beta emitters such as tritium or ²⁴¹Pu are not well represented, others such as ⁹⁹Tc are represented at higher efficiencies, but still not fully.

From BNFL (1988-1995).

The BNFL low level radioactive waste disposal site at Drigg is much closer to the Ravensglass estuary than Sellafield. Indeed the Drigg Stream flows into the estuarine reaches of the River Irt (see Technical Deliverable TD9 for detailed description). Until 1991 the collective leachates from the waste disposal trenches were discharged into the Drigg stream and contributed to the radioactive burden of the River Irt. Since 1991 the leachates have been diverted to a marine pipeline and form an insignificant contribution to the Irish Sea inventory compared with Sellafield discharges. In 1994, Drigg marine discharges were approximately 0.025 TBq beta and 0.001 TBq alpha compared with 125 TBq beta and 1 TBq alpha discharged from Sellafield (BNFL, 1995). Residual activity in the Drigg stream is now at very low levels (Figure 9) and no longer forms a significant input to the River Irt.



From BNFL (1994).

Figure 9 Radioactivity in Drigg stream.

3.3.2 Discharges/Effluents

a) Radionuclides outside of the estuary

The decrease in the discharge of radionuclides from Sellafield in recent years has led to a decrease in the level of contamination of the sediment in the vicinity of the Ravenglass estuary. Mass balance calculations suggest that most radionuclides such as americium and plutonium enter and leave the Ravenglass estuary as suspended particulate material. There have been two reported of attempts at balancing discharges into and out of the estuary (see Tables 14 and 15).

Table 14 Mass balance for radionuclides in the Ravenglass estuary (1985).

Component	Flood tide	Ebb tide
²³⁹⁺²⁴⁰ Pu(suspension) / MBq	155	274
²³⁹⁺²⁴⁰ Pu(solution) / MBq	44	56
²⁴¹ Am(suspension) / MBq	215	348
²⁴¹ Am(solution) / MBq	4.4	5.2

Data collected 24 June 1985, three days after a spring tide near mouth of estuary.

From Burton and Yarnold (1988).

Table 15 Mass balance for radionuclides in the Ravenglass estuary (1987).

Component	Flood tide	Ebb tide	Balance per tide
Pu(particulate) / MBq	254.6	169.7	+84.9
Pu(solution)			
Pu(III,IV) / MBq	7.3	7.4	-0.1
Pu(V,VI) / MBq	20.9	21.7	-0.8

Data collected 6 October 1987, on a 9.23 m spring tide at five points at mouth of the River Esk branch of the estuary. Depth varied between 5.26 m and 0.65 m, width varied between 272 m and 37 m. From Mudge et al. (1989).

Overall, more contaminated material is thought to be currently leaving the estuary than is entering it.

The fact that the radionuclide concentration in the sediment outside the estuary are changing presents a problem as this will affect the levels of input and output for the estuary. Recent data reported for sediments found immediately outside the estuary is for 1988 and is shown in Table 16. This measured the following levels of radionuclides in the top 5 cm of the sediment in the Irish Sea.

Table 16 Concentration of radionuclides in sediments at the mouth of the Ravenglass Estuary.

Radionuclide	Concentration range (Bq kg ⁻¹ dry wt)
⁶⁰ Co	<20
⁹⁵ Zr+ ⁹⁵ Nb	<40
¹⁰⁶ Ru	<40
¹²⁵ Sb	<20
¹³⁴ Cs	<40
¹³⁷ Cs	40-200
¹⁴⁴ Ce	<40
¹⁵⁴ Eu	<20
¹⁵⁵ Eu	<20
²³⁸ Pu	40-200
²³⁹⁺²⁴⁰ Pu	200-1000
²⁴¹ Am	200-1000
²⁴³⁺²⁴⁴ Cm	<1

From McCartney et al. (1994).

Given present trends it is likely that the current values are now at the lower ends of the given ranges.

b) Radionuclides within the estuary

A comprehensive survey was carried out on the radionuclide activity in water-sediment suspensions throughout the Ravenglass estuary in May 1981 (see Table 17).

Table 17 Concentration of radionuclides in the Ravenglass Estuary.

Nuclide	Soluble phase activity (kBq m ⁻³)			Particulate phase activity (kBq kg ⁻¹)		
	Mean	Samples	Range	Mean	Samples	Range
⁹⁵ Zr	--	--	<0.4	15.4	24	4.3-33.0
⁹⁵ Nb	--	--	<4.0	144.4	10	87.1-299.7
¹⁰⁶ Ru	1.1	52	<0.1-2.9	79.1	59	<1.6-120.3
¹³⁴ Cs	1.2	44	<0.03-2.6	0.7	51	0.04-1.8
¹³⁷ Cs	12.7	63	<0.05-33.3	16.3	63	0.1-44.9
¹⁴⁴ Ce	0.43	19	<0.2-0.9	11.6	50	0.4-43.1
²³⁸ Pu	0.006	171	<0.0002-0.007	3.4	155	0.15-10.6
²³⁹⁺²⁴⁰ Pu	0.019	171	0.0005-0.041	12.8	155	0.6-28.7
²⁴¹ Am	0.054	17	<0.03-0.097	22.2	62	<0.5-53.2

Particulate concentration in range 20 - 70 g m⁻³.
From Assinder *et al.* (1985).

Whilst the radionuclide levels given may not necessarily be representative of current conditions they do provide some indication of the levels likely to be encountered.

c) Distribution coefficients for the estuary

i) Plutonium

Plutonium can exhibit a number of oxidation states (Pu(III), Pu(IV), Pu(V) and Pu(VI)) in aqueous solutions in the Eh-pH-range of environmental interest (Allard and Rydberg, 1983). However, the analytical techniques normally employed are unable to distinguish between Pu(III) and Pu(IV) or Pu(V) and Pu(VI), and plutonium is usually described in terms of pairs of oxidation states i.e. Pu(III+IV) and Pu(V+VI). Pu(V) is believed to be the predominant oxidation state in the dissolved phase (Pentreath *et al.*, 1986; Orlandini *et al.*, 1986; Edgington and Nelson, 1984). It has been postulated from thermodynamic data that it is probably present as a range of ionic and neutral species, e.g. PuO₂⁺, PuO₂(OH)⁰, PuO₂(CO₃)⁻, and PuO₂(CO₃)₃⁵⁻ (Allard and Rydberg, 1983; Morse and Choppin, 1986; Mudge *et al.*, 1989). There is little evidence to suggest that organic material in the estuarine water will complex Pu(V+VI) (Mudge *et al.*, 1989). Pu(IV) is also present in the soluble phase, but at lower concentrations appears to be primarily present as anionic colloidal organic complexes (Mudge *et al.*, 1989).

Adsorbed Pu(III+IV) and Pu(V+VI) species can be further subdivided into 'labile' and 'non-labile' fractions. The labile fraction is an easily exchangeable fraction and is thought to be held as complexed species on the surface of sediments through ion-exchange reactions (rather than as aggregates). Surface complexation is dependent upon variables such as pH, ionic strength and relative ionic composition. The non-labile fraction of plutonium probably consists of the oxide, carbonates, organic and residual phases (Mudge *et al.*, 1989).

Both labile and non-labile fractions of plutonium can participate in interactions between sediments and solutions. The former fractions achieve equilibrium within an hour whereas the latter are slower reactions. The distribution of the plutonium fractions on the Ravenglass Estuary sediment was estimated to be those given in Table 18.

Table 18 Activity of different fractions of ²³⁹⁺²⁴⁰Pu in the surface sediment of the Ravenglass Estuary.

Fraction	Activity (Bq kg ⁻¹) (percentage of total)		
	Pu(III,IV)	Pu(V,VI)	Total Pu
Labile	26.4 (2.7)	21.0 (48)	47.0 (4.6)
Non-labile	950.8 (97.3)	22.7 (52)	973.9 (95.4)

From Mudge *et al.* (1989).

The K_d value obtained for the Ravenglass estuary is dependent upon the manner in which it is measured. It has been observed that K_d values obtained from adsorption experiments are significantly smaller (up to two orders of magnitude) than those obtained from desorption experiments (Burton, 1986; Eakins *et al.*, 1985; Mudge *et al.*, 1989).

An attempt was made to estimate the K_d values for each fraction from adsorption experiments (Mudge *et al.*, 1989). This used sediment from the Ravenglass estuary, seawater from the North Sea and water from the River Esk. The results are given in Table 19.

Table 19 Summary of K_d values for different plutonium fractions.

Fraction	Labile Pu		Non-labile Pu	
	III,IV	V,VI	III,IV	V,VI
K_d (m ³ kg ⁻¹)	~1	~10	>10 ³	10-10 ²

From Mudge *et al.* (1989).

The choice of appropriate K_d value may well depend upon what is being studied. However, for the Ravenglass estuary as a whole, it would be best to assume that metastable equilibria are attained throughout the estuary and to use an overall K_d value. A summary of overall K_d values obtained for the Ravenglass Estuary by different groups are given in Table 20. These were largely obtained through desorption experiments. The values are sufficiently consistent to suggest that the overall K_d value for plutonium within the estuary should be taken as 1×10^3 m³ kg⁻¹.

Table 20 Summary of overall K_d values for plutonium in the Ravenglass estuary.

Year	Location	K_d ($10^3 \text{ m}^3 \text{ kg}^{-1}$)		Notes	Reference
		Mean	Range		
1981	Throughout estuary during different tidal cycles	1.017±1.107	0.040-10.929	For ^{238}Pu . 155 samples	Assinder <i>et al.</i> (1984)
		1.085±1.056	0.040-10.340	For $^{239+240}\text{Pu}$. 155 samples	
1982	Sediment collected from east bank of the River Irt	1.1		Used North Sea water	Burton (1986)
1985	Samples collected from the mouth of the estuary over a full tidal cycle	0.13	0.11-0.14	Appears to reflect of both adsorption and desorption processes	Burton and Yarnold (1988)
1986-1989	Sediment collected from bank at Newbiggin	0.97		Used North Sea water	Mudge <i>et al.</i> (1989)
		1.2		Used water from River Esk	

ii) *Americium*

Am(III) and Am(IV) are likely to be the stable oxidation states for americium (^{241}Am) in estuarine waters (Aston and Stanners, 1982b). Of these Am(III) is likely to predominate (Schulz and Penneman, 1986).

The speciation of these oxidation states have not been particularly well characterised, but seems likely that these will largely consist of hydroxide and carbonate complexes. Organic complexes may also be possible. Americium is believed to be adsorbed onto iron/manganese oxide/hydroxide phase of the sediments (Assinder, 1983). It is thought that americium is adsorbed as a carbonate complex (Burton and Yarnold, 1988).

A summary of overall K_d values obtained for the Ravenglass estuary by different groups are given in Table 21. These were largely obtained through desorption experiments. The values are sufficiently consistent to suggest that the overall K_d value for americium within the estuary should be taken as approximately $1 \times 10^3 \text{ m}^3 \text{ kg}^{-1}$.

Table 21 Summary of overall K_d values for americium in the Ravenglass estuary.

Year	Location	K_d ($10^3 \text{ m}^3 \text{ kg}^{-1}$)		Notes	Reference
		Mean	Range		
1981	Throughout estuary during different tidal cycles	0.552	0.030-0.790	For 17 samples	Assinder <i>et al.</i> (1985)
1982	Sediment collected from east bank of the River Irt	2.0		Used North Sea water	Burton (1986)
1985	Samples collected from the mouth of the estuary over a full tidal cycle	1.1	0.8-1.2	For 4 samples	Burton and Yarnold (1988)

iii) Caesium

The chemistry of caesium makes it unlikely that much complex formation occurs in solution. However, it is known to be sorbed into minerals such as illite. Three types of sorption sites have been identified (Evans *et al.*, 1983); surface and planar sites where caesium is readily exchangeable, wedge sites where exchange is limited to cations with a similar size and charge, and interlayer sites where caesium is not exchangeable. There appears to be a trend that as the sediment ages (over years), the proportion of caesium in the interlayer sites increases. This would imply that, as was the case with plutonium and americium, the K_d value will be dependent on how the value is obtained.

iv) Ruthenium and cerium

The K_d values for ruthenium and cerium have also been measured in the Ravenglass estuary (Assinder *et al.*, 1985). The chemistry of these radionuclides is not very well characterised. It has been postulated that cerium exists in solution predominantly as Ce(III) and a small amount of Ce(IV) (Schoer and Förstner, 1985). The Ce(III) is believed to exist in seawater as CeCO_3^+ , Ce^{3+} , CeCl_2^+ and $\text{Ce}(\text{OH})_x^{(3-x)}$ and in river water as CeCO_3^+ , Ce^{3+} and CeSO_4^+ (Schoer and Förstner, 1985). Colloid formation is thought to be significant for both cerium(IV) and ruthenium.

The K_d values for caesium, cerium and ruthenium were obtained for samples collected throughout the Ravenglass estuary (Assinder *et al.*, 1985). These values are summarised in Table 22 where it can be seen that they are sufficiently consistent to allow the overall K_d values for caesium, ruthenium and cerium to be taken as $4 \text{ m}^3 \text{ kg}^{-1}$, $100 \text{ m}^3 \text{ kg}^{-1}$ and $30 \text{ m}^3 \text{ kg}^{-1}$, respectively.

Table 22 Summary of overall K_d values for radionuclides in the Ravenglass estuary in 1981.

Radionuclide	K_d ($m^3 kg^{-1}$)		Notes
	Mean	Range	
^{106}Ru	139	3-942	50 samples
^{134}Cs	1.2	0.2-10.5	55 samples
^{137}Cs	6.6	0.4-71.6	62 samples
^{144}Ce	33	12-108	14 samples

From Assinder *et al.* (1985).

3.3.3 Contamination

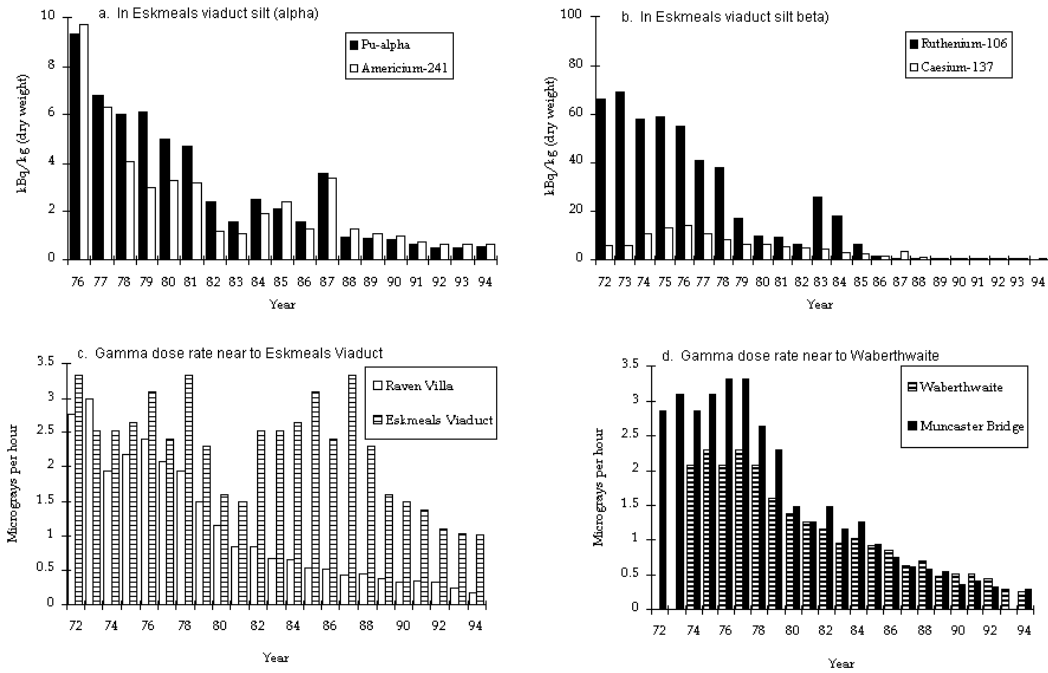
a) Specific activity in surface sediments

A considerable body of data has been published on the distribution of radioactivity in the Ravenglass estuary, most notably in the annual reports of BNFL (1976 to 1995) and MAFF (1979 to 1995). Additional datasets include surface sediment samples (e.g. Aston and Stanners, 1982a; Horrill, 1983, 1984), grab samples from broader areas (e.g. Aston and Stanners, 1981; Hamilton and Clarke, 1984; Aston *et al.*, 1985) and some vertical activity profiles (e.g. Heaton and Hetherington, 1984). Data obtained for political purposes are discounted here, as being of questionable objectivity (most notably Watts and Green, 1989). Prevailing levels of radioactivity in the Ravenglass estuary are given in Table 23. A time series of the dominant alpha and beta radionuclides is presented in Figure 10.

Table 23 Radionuclide concentrations in the Ravenglass estuary.

Location	Sediment type	$Bq kg^{-1}$ dry weight								
		^{60}Co	^{95}Zr	^{95}Nb	^{106}Ru	^{125}Sb	^{134}Cs	^{137}Cs	Pu- α	^{241}Am
RavenVilla	silt	5	18	27	190	<16	<5	290	400	510
Salmon Garth	silty sand	<3	<6	<6	<37	<9	<3	85	150	190
Boat area	silt	6	19	24	170	<20	<5	290	530	660
Eskmeals viaduct	silt	6	23	22	200	20	<5	290	530	660

From BNFL (1994).



From BNFL (1994).

Figure 10 Radioactivity levels in Ravenglass estuary silts.

Concentrations of all radionuclides in surface sediments (ca. 0 to 5 cm) have declined considerably over the past decade, particularly in the fine silt areas which tend to accumulate radionuclides to a greater degree than coarse sands. The decline in concentrations is matched by reducing external gamma doserates (see Figures 10c and 10d) and reflects the decline in discharges of radioactivity from Sellafield since the mid 1980's (see Figure 8).

Significant differences in total activity exist between sediments of differing types. From Table 23 it is clear that the slightly coarser grained silty sand at Salmon Garth contains a lower activity than the surrounding fine silt areas. Kelly and Emptage (1992) reported total alpha and beta concentrations in surface sediments classified according to type and grain size (see Table 24). Although restricted to gross measurements, a clear trend is apparent with increasing levels of activity in fine silts, with a parallel trend from terrestrial soils to saltmarsh banks. The latter trend almost certainly reflects grain size with the exception of the terrestrial areas where a lower source term is probable.

Table 24 Variation of total alpha and total beta activity concentrations in surface sediments of the Ravenglass estuary.

Sediment classification	n	Activity (Bq kg ⁻¹ d.w.)			
		Total alpha		Total beta	
		mean	range	mean	range
Terrestrial	10	410	201-798	981	502-2057
Sand dune	2	330	327-333	521	502-540
Relic gravel	4	570	438-850	853	540-1373
Channel sands	15	821	282-1954	686	383-1065
Erosional mudflat	7	6360	2293-7859	2407	968-3562
Lower saltmarsh bank	12	6557	2251-19566	4973	2194-13529
Upper saltmarsh bank	49	10267	1214-22566	8018	1851-17069
Gravel	4	570	438-850	853	540-1373
Sand	10	547	282-1455	576	383-944
Silty sand	11	4034	752-7859	1628	743-3233
Sandy silt	48	8464	327-19666	6636	502-16007
Silt	14	12603	5376-22566	9351	3466-17069

From Kelly and Emptage (1992).

Kelly and Emptage (1992) reported the distribution of ¹³⁷Cs as a function of the different sediment facies (see Table 25) and as a function of the three component estuaries (see Table 26).

Table 25 The distribution of ¹³⁷Cs in the sediment facies of the Ravenglass estuary

	Sediment facies					
	Total	Relic	Channel	Erosional	Lower bank	Upper bank
Area / km ²	5.7	0.5	1.0	1.0	0.2	2.0
Radioactivity						
¹³⁷ Cs inventory / MBq m ⁻²	0.8	n/d	0.70	0.50	3.97	1.18
Depth of contaminated layer / cm	28	n/d	42	21	61	21
Contaminated volume / 10 ⁶ m ³	1.2	n/d	0.43	0.22	0.13	0.42
¹³⁷ Cs inventory / TBq	4.5	n/d	0.66	0.56	0.85	2.41

From Kelly and Emptage (1992).

Table 26 ¹³⁷Cs inventories for the component estuaries of the Ravenglass estuary.

Sediment facies	¹³⁷ Cs Inventory / TBq (Percentage of total)				
	Total	Upper bank	Lower bank	Erosional	Channel (sediment)
Region					
Irt estuary	1.59 (36)	0.89 (20)	0.29 (6)	0.33 (7)	0.09 (2)
Mite estuary	0.49 (11)	0.25 (6)	0.11 (2)	0.12 (4)	0.01 (<1)
Esk estuary	2.38 (53)	1.27 (28)	0.45 (10)	0.10 (2)	0.57 (13)
Whole estuary	4.47 (100)	2.41 (54)	0.85 (19)	0.56 (12)	0.66 (15)

From Kelly and Emptage (1992).

Clearly, the correlation between grain size and activity content is not perfect and considerable variability within sediment types is apparent (exceeding an order of magnitude in some instances). Nonetheless, in broad terms, it is possible to characterise the specific activity of Ravenglass estuary sediments as a function of grain size.

Thus, the coarse grain size of the channel facies results in low specific activities, e.g. total alpha and total beta specific activities of 821 and 686 Bq kg⁻¹, and deposits of this facies contribute relatively little to the radionuclide inventories of the estuary, e.g. 0.66 TBq of ¹³⁷Cs, 15% of the estuary total.

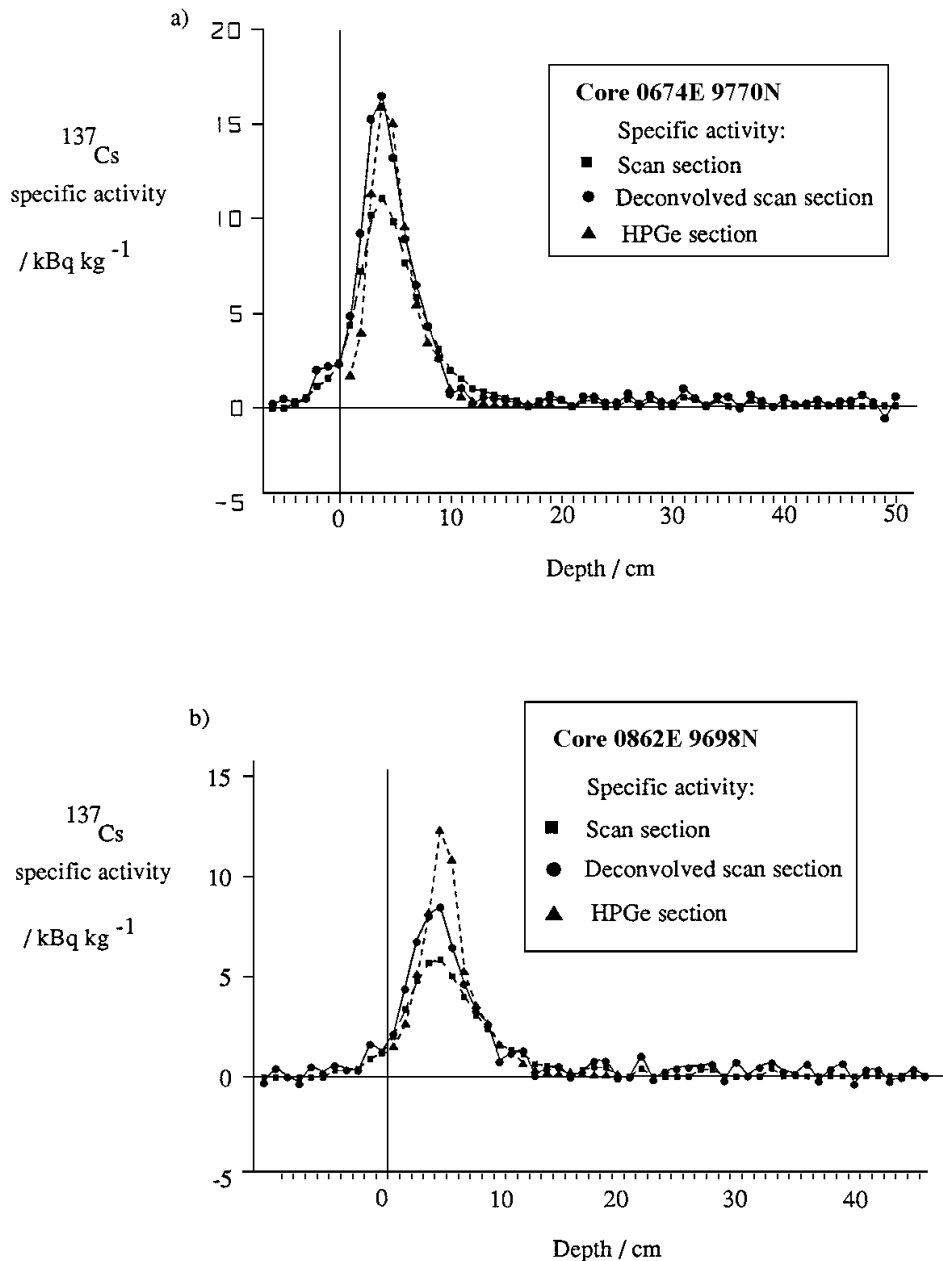
By contrast the bank facies have relatively high radionuclide specific activities, due to the fine grain size. This results in relatively large radionuclide inventories (i.e. 3.25 TBq of ¹³⁷Cs or 73% of the estuary total). The volume of such sediment, contaminated at more than trace levels, is estimated at 0.55 × 10⁶ m³. Of this, the lower bank facies contain 19% (0.85 TBq) of the total ¹³⁷Cs inventory, with a contaminated volume of 0.13 × 10⁶ m³. These areas are relatively unstable. Monthly measurements have shown that sediment mass flux and specific activity and, hence, radionuclide deposition flux are highly variable, partly due to grain size variations related to tidal amplitude, velocity and wave activity. In addition, there is a marked regular seasonal cycle in specific activities for many radionuclides in these areas, with peaks in late summer/early autumn and winter, which may be due to the influence of seasonal temperature change on the stability and circulation in the Irish Sea, on plankton productivity or on distribution coefficients. Sharp changes in the specific activity of short-lived radionuclides are considered to reflect short-term changes in discharges, or coastal circulation, and they demonstrate the ability of the estuary sediment system to react to the discharges on these time-scales, whether directly or indirectly. A spatial trend, of increasing levels of radiocaesium derived from the catchments is thought to occur through adsorption of caesium from solution in freshwater by sediments of marine origin, when they come into contact during the tidal cycle.

Sediment accumulation on saltmarshes is low, and hence the total depth of contamination is restricted (mean 20 cm). However, as a result of the low energy of this sedimentary environment, grain sizes tend to be finer than elsewhere and specific activities high, e.g. mean total alpha and total beta of 10.3 and 8.0 kBq respectively. The combination of low sedimentation rate and high specific activity means that this facies contributes proportionally less to the volume of contaminated sediment 35% (0.4 × 10⁶ m³) and more to the total inventories 54% (2.4 TBq) of ¹³⁷Cs.

Radionuclide inventories in erosional areas may be inherited from earlier depositional phases, or may be added from solution, and mixed by bioturbation. Sediments of this facies contain 12% (0.55 TBq) of the ^{137}Cs inventory of the estuary. The relatively high mean specific activities for surface sediments of this facies, e.g. 6.3 and 2.4 kBq kg⁻¹ for total alpha and total beta respectively, is due to the exposure at the surface of the higher levels of specific activity from the past, whereas these are buried in areas of net deposition.

b) Sediment core activity profiles

The peak of activity in sediments from the Ravenglass estuary generally occurs some distance below surface. Figure 11 shows ^{137}Cs activity profiles for two cores from the tidal reaches of the River Esk. These appear to be typical inasmuch as they indicate a single sub-surface peak with a relatively smooth activity profile indicating detectable contamination to a depth of about three times the peak depth. In these cases the peak occurs at about 5 cm below the surface, although a number of surveys indicate that the activity peak may occur from about 3 cm to lower than 30 cm beneath the surface. The depth of the activity peak, in combination with the depth of "surface" sediment sampled in routine surveys, can be a major influence on the range and variability of radionuclide concentrations reported.



From Kelly and Emptage (1992)

Figure 11 ^{137}Cs activity profiles in two cores from the Esk estuary.

More complicated core activity profiles are also reported in the literature, particularly at the bank edges of the saltmarsh/mudflat interface and in the channel sands. Polymodal distributions may reflect deposition from distinct single storm events, particularly in the channel sands which have a highly variable sedimentation rate. Occasional activity horizons below the main peak of activity may reflect pore water migration channels. Poorly defined peaks may be due to bioturbation, especially due to the burrowing of *Corophium volutator* which occurs at high population densities in parts of the estuary.

A study of the inputs of Chernobyl fallout radionuclides to the sediment deposits of the bank facies showed that radionuclide fluxes occurred not only by direct surface deposition of contaminated sediment but also by rapid post-depositional labelling of the sediment from percolating active surface waters. In particular, this occurred down macropores formed by burrowing organisms (mainly the sand shrimp *Corophium volutator*), but could potentially occur down macropores of other origins such as shrinkage cracks. In addition, these studies

showed that where these organisms occurred, the bioturbation caused by burrowing activity resulted in the destruction of activity peaks in the sediment profile and, hence, in the smoothing out of the variations in the radionuclide inputs. High populations of these organisms, and their associated effects on radionuclide behaviour, are confined to the lower bank and erosional facies in the middle and lower reaches of the estuary.

The mean total thickness of the contaminated zone in the estuarine sediments is about 20 cm to 60 cm. Deeper contaminated zones, sometimes exceeding 1 m in thickness, occur in areas of high sediment accretion rates.

Based on estimates of areas covered by differing sediment classes (Table 1) and the mean total depths of contamination, an estimate of $1.2 \times 10^6 \text{ m}^3$ can be derived for the total volume of contaminated sediment. Using mean ^{137}Cs concentrations and activity profiles to derive inventories for each sediment type, Kelly and Emptage (1992) estimate the Ravenglass estuary system to be contaminated with 4.5 TBq of ^{137}Cs . Of this, more than half is in the top 20 cm of sediment. In a similar calculation Eakins *et al.* (1985) estimated the estuary to be further contaminated with 4 TBq of $^{239+240}\text{Pu}$ and 4 TBq of ^{241}Am .

c) Reworking of sediments and 'availability' of radioactivity

Bioturbation, sediment erosion and change of use of land can all contribute to sub-surface activity becoming available for pathways back to man, just as continued accretion will generally decrease the impact, given the current low discharge levels.

The rate of potential reappearance of the activity has to be considered in relation to its physical half-life. Thus, the decline in gamma dose rates observed in Figure 10d results from both physical decay and shielding due to continued sediment accretion. Hunt (1984) modelled absorbed dose in air over sediment as:

$$D = 4.389 \times 10^{-3} \times R \times E \times I$$

where D is the absorbed dose in air ($\mu\text{Gy h}^{-1}$);
E is the energy per decay (MeV);
R is a reduction factor taking into account the activity profile; and
I is the radionuclide inventory (Bq), derived from:

$$I = A S / \lambda$$

where A is the surface activity concentration (Bq m^{-2});
S is the sedimentation rate (m a^{-1}); and
 λ is the decay constant (a^{-1}).

A number of better models now exist, but the clear principal is that shielding due to sediment accretion will be reversed by erosion. Most of the short-lived gamma emitters ($^{95}\text{Zr} + ^{95}\text{Nb}$, ^{106}Ru , ^{144}Ce) have now decayed to very low levels, leaving ^{137}Cs (with a half-life of 30 years) as the principle dose rate contributor.

Kelly and Emptage (1992) estimated that more than half of the total ^{137}Cs inventory is in the upper saltmarshes and inter-tidal pastures. These areas are unlikely to be extensively reworked on time scales of less than 100's of years. Around 19% of the total inventory is associated with lower bank areas, which are regions of rapid accretion. The remaining activity (no more than 30% of the total) is present in areas which may be available for export from the estuary or for redistribution within the estuary.

Activity associated with the stable saltmarsh and pasture areas may be incorporated in food chains (principally via grazing sheep) back to man. The distribution of radionuclides in the

silt and sediment of salt marshes for the Ravenglass estuary have been reported by Horrill (1984) and is summarised in Table 27.

Table 27 Mean activities of radionuclides in the silts and vegetation around the Ravenglass estuary.

Nuclide	Silt (Bq kg ⁻¹)		Vegetation (Bq kg ⁻¹)	
	mean	sd	mean	sd
⁶⁰ Co	11	8	2	2
⁹⁵ Nb	347	279	284	281
⁹⁵ Zr	41	44	21	28
¹⁰⁶ Ru	1870	669	358	274
¹²⁵ Sb	42	22	n/d	--
¹³⁴ Cs	84	29	50	55
¹³⁷ Cs	1681	590	897	877
¹⁴⁴ Ce	492	187	108	80
¹⁵⁴ Eu	44	32	7	8
¹⁵⁵ Eu	62	25	7	8
²³⁸ Pu	269	107	47	21
²³⁹⁺²⁴⁰ Pu	1075	434	176	83
²⁴¹ Am	565	215	66	49

n/d = not determined

For plutonium isotopes n = 27, for all others n = 100.

From Horrill (1984).

d) External gamma dose rates

The gamma dose rates in air in the estuary vary from 70 to 608 nGy h⁻¹. Their distribution is now largely related to the distribution of ¹³⁷Cs in the estuary. This is largely due to the decreases in the discharge of short-lived radionuclides from Sellafield. The dose rate distribution can be related to the combined effects of grain size and sedimentation rates within the estuary. The former influences the distribution of radionuclide specific activities and the latter the distribution of activity with depth, in particular determining the depth of burial of the more active sediments from the 1970's.

4. Restoration Options

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

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

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

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

The remediation techniques which are available for treating the site are described in Technical Deliverable 3+4 (Zeevaert and Bousher, 1998). Of the techniques described the ones considered appropriate to this site are as follows:

- source removal: excavation (Option B)
- physical separation: soil washing (Option C1)
- chemical separation (Option D1)

It is assumed that all technologies should be engineered to meet the current standards.

Technical Deliverable 3+4 gave ranges of values for the cost, performance and number of man hours required to remediate the waste. These ranges were generic, with the expectation that they would be used to derive site-specific parameters for each remediation technique. The site-specific values used for the Ravenglass estuary are summarised in Tables 27 to 30.

The values selected for a number of factors including: the type of waste material, the radionuclides present, their accessibility, the ease of excavation and the location of the site with respect to a suitable waste storage repository.

For the purposes of these calculations it is assumed that external operations will be conducted at the Drigg depository site (5 km from estuary).

Table 28 Site-specific values for the performance of remediation techniques appropriate to the Ravenglass estuary.

Remediation Technique	Unit [†]	Value
Removal of sources Excavation	DF	5
Physical separation (<i>ex-situ</i>) Soil washing	DF RF	5 80%
Chemical separation	DF DF	20 (Cs) 3.3 (Am, Pu)

[†] DF = decontamination factor; RF = waste reduction factor.

Table 29 Site-specific values for the costs of remediation techniques appropriate to the Ravenglass estuary.

Remediation Technique	Cost
<u>Source Removal</u> Soil Excavation Excavation costs (including transport) Excavation costs (including transport and RCRA disposal) Disposal cost for radioactive material (including transport)	 100 EUR m ⁻³ 700 EUR m ⁻³ 2500 EUR m ⁻³
<u>Physical Separation (ex-situ)</u> Soil washing Soil washing costs Cost of excavation and transport (prior to washing) Cost of disposal of radioactive residue (including transport)	 300 EUR m ⁻³ 100 EUR m ⁻³ 2500 EUR m ⁻³ (residue)
<u>Chemical Separation (ex-situ)</u> Chemical solubilisation Separation costs Cost of excavation and transport (prior to solubilisation) Cost of disposal of radioactive residue (including transport)	 450 EUR m ⁻³ 100 EUR m ⁻³ 2500 EUR m ⁻³ (residue)

Table 30 Site-specific values for the exposure times (restoration workers) of remediation techniques appropriate to the Ravenglass estuary.

Remediation Technique	Exposure Time (Restoration Workers)
Removal of sources Excavation	1 manh m ⁻³
Physical separation (ex-situ) Soil washing	1.1 manh m ⁻³
Chemical separation	2.5 manh m ⁻³

The performance values and costs associated with each each techniques may be used to calculate the cost of treating the material in Trenches 1 to 7 and also the cost of disposal of any residue from the remediation procedure. These calculations are described in detail in Appendix A. The results of these calculations are summarised in Table 31.

Table 31 The cost of applying remediation technologies to the Ravenglass estuary.

Remediation strategy	Monetary costs of remediation [EUR]	
	Remediation (incl. labour)	Waste disposal (incl. transport)
Drigg		
Source Removal	1.3 × 10 ⁸	7.8 × 10 ⁸
Soil Washing	5.2 × 10 ⁸	6.5 × 10 ⁸
Chemical Solubilisation	7.2 × 10 ⁸	1.1 × 10 ⁹

5. Radiological Impact Assessments

5.1 Compartment scheme

The Ravenglass Estuary may be described by the compartment scheme shown in Figure 12. Here the Cumbrain coast represents the Irish Sea in the scheme. Its volumes and turn over are taken from Jefferies *et al.* (1989). It is assumed that all particulate material in the water mass comes from the Irish Sea.

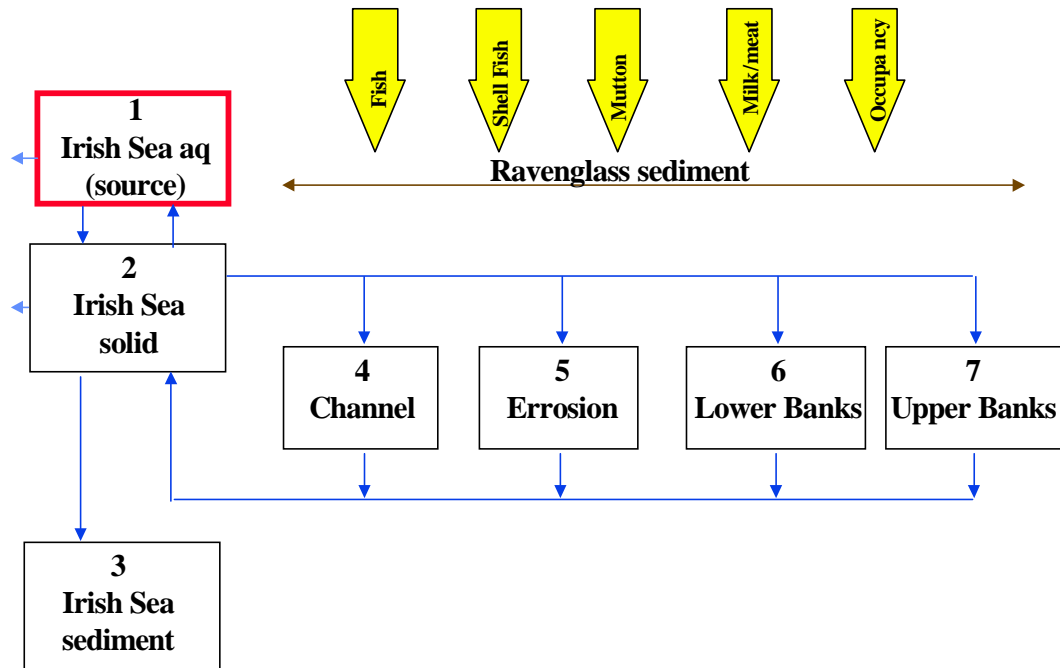


Figure 12 Compartment scheme for the Ravenglass estuary.

The main radionuclides under consideration are caesium-137, plutonium-239 and americium-241.

It should be noted that the half-life of caesium-137 is approximately 28 years. Consequently, over periods of 100 and 500 years it is the case that due to radioactive decay, the contribution of caesium-137 to the overall activity of a site will naturally decline. However, as a first approximation, for the purposes of the present calculations this was not taken into account.

5.2 Processes, exposure pathways and exposure groups

Each transfer coefficient can represent a number of complex processes. The processes are considered for the turnover of radionuclides in each major compartment of the model are given in Table 32. The equations and data used to describe these are given in Stiglund and Nordlinder (1999).

Table 32 Processes occurring in major compartments of the Ravenglass estuary model.

Compartment	Processes
Water	Sedimentation of particles Outflow
Sediment	Re-suspension

The important exposure pathways for the critical group are indicated in the compartment scheme. The exposure pathways considered are:

- consumption of fish;
- consumption of shell fish;
- consumption of meat via the drinking water of the cattle and sheep;
- consumption of meat via the pasturage for the cattle and sheep;
- occupancy.

The critical group concerned contains local fishermen. The dose assessment considers doses due to consumption of milk and meat from cattle and meat from sheep grassing on the upper banks and drinking water from the river. Fish and shellfish is also included in the pathways.

The committed collective dose is based on production data.

The dose to the workers considers external exposure from the sediments only.

5.3 Doses to public/remediation workers

The doses calculated consist of individual and committed collective doses.

5.3.1 Individual doses to critical group: first year of the base case

The results of the dose calculations for the individual doses for the first year are shown in Table 33.

Table 33 Individual dose at the Ravenglass Estuary during the first year.

Exposure pathway	Dose (Sv a ⁻¹)			
	Cs-137	Pu-239	Am-241	Total
Fish	4.2×10^{-5}	2.6×10^{-5}	4.6×10^{-5}	1.1×10^{-4}
Shellfish	5.8×10^{-6}	2.0×10^{-4}	2.7×10^{-4}	4.7×10^{-4}
Cattle (meat and milk)	5.3×10^{-4}	1.1×10^{-6}	8.7×10^{-7}	5.3×10^{-4}
Sheep	3.8×10^{-4}	3.7×10^{-6}	5.9×10^{-6}	3.9×10^{-4}
External exposure	2.8×10^{-7}	2.5×10^{-10}	2.8×10^{-8}	3.0×10^{-7}
Total	9.1×10^{-4}	2.3×10^{-4}	3.2×10^{-4}	1.5×10^{-3}

The contribution to the total dose from the different pathways is shown in Figure 13.

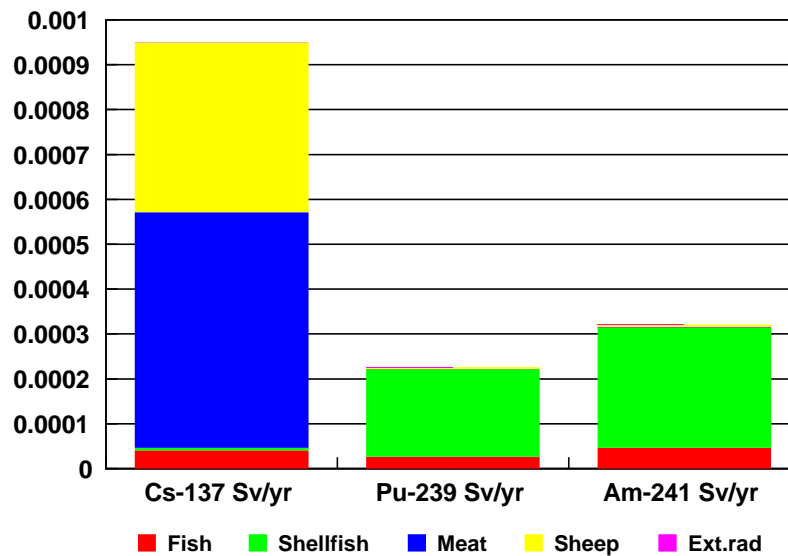


Figure 13 The contribution to the total dose from different pathways.

As can be seen in Figure 13, the overall dominating exposure pathway for caesium-137 is via the ingestion of meat both from sheep and cattle. For plutonium-239 and americium-241 the exposure via shellfish is the dominating one.

5.3.2 Collective doses to public and restoration workers over 100 years and 500 years

Relevant restoration options for the Ravenglass estuary have been considered in Bousher (1998). The base case, as the situation is without any restoration is called A. Only the sediments of the upper banks are considered for restoration.

Table 34 shows the restoration options considered for the Ravenglass estuary site.

Table 34 Restoration options considered for the Ravenglass estuary.

Case	Restoration option
B	Source removal
C1	Soil washing
D1	Chemical solubilisation

The results of the dose calculations for the collective intakes/doses at year 100 and 500 for the different restoration options are shown in Table 35.

Table 35 Collective doses at Ravenglass estuary at 100 years and 500 years.

Case	Year	Collective dose (manSv)			
		Cs-137	Pu-239	Am-241	Total
A	100	23	2.3	3.2	28
	500	23	2.6	3.6	29
B	100	11	1.6	2.2	15
	500	11	1.7	2.4	15
C1	100	18	2.1	3.0	23
	500	18	2.4	3.3	24
D1	100	2.8	2.1	2.8	7.7
	500	2.8	2.2	3.1	8.2

As can be seen in Table 35 the caesium-137 gives the highest exposure.

The restoration workers are assumed to only be exposed to sediment from the upper banks.

In Figure 14 the total collective dose for the critical group and the restoration workers are showed for caesium-137 for the different restoration options at 100 and 500 years.

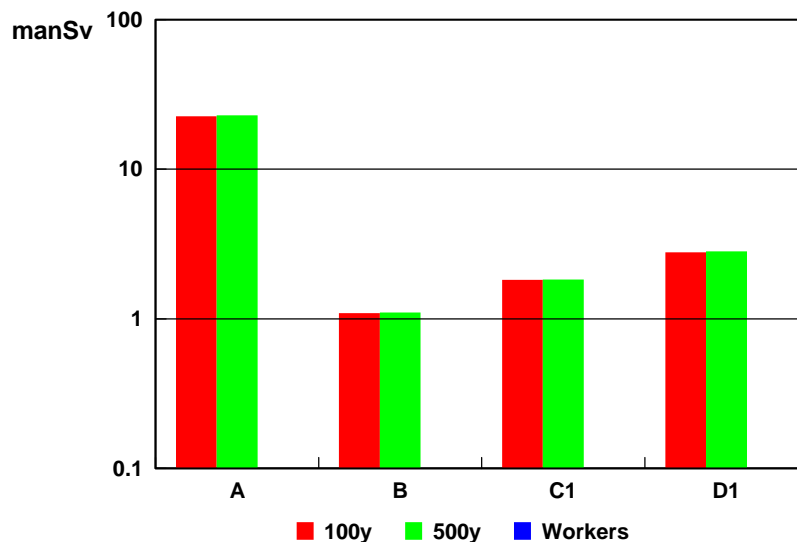


Figure 14 The total collective dose of caesium-137 for the different restoration options at 100 and 500 years.

As can be seen in Figure 14 the reduction of dose to the critical group for Option B is the best option when considering dose reduction. Option B stands for source removal which in this case means removal of the sediments in the estuary.

5.4 Uncertainty analysis

For caesium-137 the concentration factor from water to plants contribute the most to the uncertainty while for plutonium-239 and americium-241 suspended material in the estuary and the uptake factor from shellfish dominate.

6. Ranking of Restoration Options

Decisions on clean-up in long-lasting exposure situations may well go far beyond purely radiological protection considerations. Satisfying the justification principle requires that the overall effect of the actions involved should do more good than harm, taking account of relevant radiological and non-radiological factors. Most decisions require multiple criteria to be taken into account. The field of multiple criteria analysis offers a number of approaches which take explicit account of multiple criteria in providing structure and support to the decision making process. In case of restoration of contaminated sites there are several criteria or attributes that need to be considered when choosing an 'optimum' restoration strategy. When the performance and costs of all the protection options have been assessed, a comparison is needed to define the *optimum* protection option. When the optimum is not self evident, the comparison can be carried using a quantitative decision aiding technique. Of the different techniques available, cost-benefit analysis has been used to evaluate if the remediation options are justified on economic grounds and multi-attribute utility (MAU) analysis for ranking different remediation options. Moreover, annual individual doses to critical groups before implementation of remedial measures have been assessed for comparison with the clean-up criteria recommended by IAEA.

6.1 Evaluation of remediation options (MAU analysis)

6.1.1 Attributes

The attributes that has been considered in this study includes:

- *Health attributes:*
 - collective doses to population;
 - doses to remediation workers;
 - non-radiological health factors.
- *Economic attributes:*
 - costs of remedial actions (including costs of labour and monitoring);
 - costs of monitoring of remedial options;
 - costs of disposal of generated waste (in broad categories);
 - loss/gain of taxes due to loss/gain of income.
- *Social attributes:*
 - reassurance of the public;
 - discomfort, disturbance and anxiety from the remedial action;
 - loss/gain of income.

The restoration options included for the Ravenglass site have been identified in Chapter 4 (Table 28). The economic and radiological data for quantifying the various attributes for each of those options are shown in Table 36.

6.1.2 Utilities

Utility functions for the attributes *monetary costs* and *radiation doses* have been calculated from the values in Table 36 on monetary cost components and residual collective doses after remediation. Linear (risk neutral) utility functions have been used.

Table 36 Remediation costs and collective doses to population and workers for different restoration strategies at the Ravenglass site.

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

a) Utility functions for monetary costs

Utility functions have been determined for remediation costs (including labour costs), waste disposal costs (including transport costs), monitoring costs and loss of taxes due to loss of income:

$$u_{\text{remedia}}(x) = 100 \cdot \left(1 - \frac{x}{720,000} \right) \text{ for } 0 \leq x \leq 720,000 \text{ kECU}$$

$$u_{\text{waste}}(x) = 100 \cdot \left(1 - \frac{x}{1,100,000} \right) \text{ for } 0 \leq x \leq 1,100,000 \text{ kECU}$$

$$u_{\text{monitor}}(x) = 100 \cdot \left(1 + \frac{3,000 - x}{52,500 - 3,000} \right) \text{ for } 3,000 \leq x \leq 52,500 \text{ kECU over 500 y}$$

b) Utility functions for health factors

The following utility functions for the radiological health components have been determined for the exposed population and workers implementing the remedial actions. Only radiological health factors are considered for the Ravenglass site as no heavy metals are found.

$$u_{\text{dose, pop,100}}(x) = 100 \cdot \left(1 + \frac{7.7 - x}{28 - 7.7} \right) \text{ for } 7.7 \leq x \leq 28 \text{ man Sv}$$

$$u_{\text{dose, pop,500}}(x) = 100 \cdot \left(1 + \frac{8.2 - x}{29 - 8.2} \right) \text{ for } 8.2 \leq x \leq 29 \text{ man Sv}$$

$$u_{\text{dose, work}}(x) = 100 \cdot \left(1 - \frac{x}{2.29} \right) \text{ for } 0 \leq x \leq 2.29 \text{ man Sv}$$

c) Utility functions for social factors

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 has left all the activity on site in a contained form (capping, surface barriers, etc.) might give a substantial dose reduction and thus a low value of the residual doses. Detailed information on how social factors like reassurance are linked with individual doses and activity concentration on site is not available. Therefore, utility functions for 100 years and 500 years integration time have been proposed which gives a low value only when both sub-utilities have low values:

$$u_{\text{reas},100}(x, y) = 100 \cdot \left(\frac{1}{2} \cdot \left(1 + \frac{7.7 - x}{28 - 7.7} \right)_{\text{dose}} + \frac{1}{2} \cdot \left(1 + \frac{0.05 - y}{1.0 - 0.05} \right)_{\text{activity}} \right)$$

for $7.7 \leq x \leq 28$ man Sv and $0.05 \leq y \leq 1$

$$u_{\text{reas},500}(x, y) = 100 \cdot \left(\frac{1}{2} \cdot \left(1 + \frac{8.2 - x}{29 - 8.2} \right)_{\text{dose}} + \frac{1}{2} \cdot \left(1 + \frac{0.05 - y}{1.0 - 0.05} \right)_{\text{activity}} \right)$$

for $8.2 \leq x \leq 29$ man Sv and $0.05 \leq y \leq 1$

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} is 100 for a residual dose of 7.7 (8.2) manSv and a remaining fraction of the initial activity of 0.05 (best strategy) and 0 for a residual dose of 28 (29) manSv and a remaining activity fraction of 1.0 (worst strategy).

The utility function u_{distur} for disturbance has been related to the volume of soil and sediment waste to be transported to the waste disposal site:

$$u_{\text{distur}}(x) = 100 \cdot \left(1 - \frac{x}{430,000} \right) \text{ for } 0 \leq x \leq 430,000 \text{ m}^3$$

6.1.3 Weighting factors

Weighting factors are scaling factors that reflect the relative importance of each of the attributes. The weighting factors assigned to the different attributes at the same hierarchy level can be either the ratio of value ranges, R , of the attributes or by assigning values to the ratio of the weighting factors at that level. Both methods have been used here.

a) Weighting factors for major attributes

The major weighting factors considered in this study include those for monetary costs, health and social factors. The sum of these weighting factors should respect the following conditions:

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

The assessment of the weighting factors is discussed in Hedemann Jensen (1999) where conversion/scaling constants between weighting factors has been expressed as:

$$C_1 = \frac{w_{\text{economic}}}{w_{\text{health}}} \cong \frac{w_{\text{economic}}}{w_{\text{dose, pop}}} = \frac{R_{\text{economic}}}{\alpha \cdot R_{\text{dose, pop}}}$$

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

C_1 can be determined for a 100 and 500 years integration time for the collective dose from the values given in Table 36. The value of C_2 is more difficult to assess but a value of 0.2 - 0.3 has been argued for in Hedemann Jensen (1999). From these values of C_1 and C_2 the

weighting factors for health, economic and social factors have been calculated as shown in Table 36.

b) Weighting factors for health sub-attributes

The weighting factors for health sub-attributes include those of radiation induced stochastic health effects to the affected population and workers and non-radiation induced stochastic health effects to the affected population. The sum of these weighting factors should respect the following conditions:

$$w_{dose, pop} + w_{dose, work} + w_{non-rad} = 1$$

The conversion/scaling constant, C , for the health sub-attributes can according to Hedemann Jensen (1999) be expressed as:

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

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

$$w_{non-rad, pop} = C \cdot R_{non-rad, pop} \cdot l \cdot r_{non-rad}$$

Exposure to heavy metals is not relevant for the Ravenglass site and $R_{non-rad}$ is therefore zero. The value of C is given by (Hedemann Jensen, 1999):

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

From the calculated values of C_{100} and C_{500} (for 100 and 500 years integrating time for the collective dose to the population) the weighting factors for collective population and worker doses have been calculated as shown in Table 36.

c) Weighting factors for economic sub-attributes

The weighting factors for economic sub-attributes include those for cost of remediation, cost of waste disposal and costs of monitoring. The sum of these weighting factors should respect the following conditions:

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

The conversion/scaling constants for the economic sub-attributes can according to Hedemann Jensen (1999) be expressed as:

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

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

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

The conversion/scaling constant, C , for the economic sub-attributes can be determined from the cost ranges in Table 36 and the weighting factors for remediation costs, waste disposal costs and monitoring costs have been calculated as shown in Table 36.

d) Weighting factors for social sub-attributes

The weighting factors include those for reassurance and disturbance. The sum of these weighting factors should respect the following conditions:

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

The conversion/scaling constants for the social sub-attributes can according to Hedemann Jensen (1999) be expressed as:

$$C_1 = \frac{w_{reas}}{w_{distur}}$$

In Hedemann Jensen (1999) it is argued that $w_{reas} > w_{distur}$ and that $C_1 \approx 5 - 7$. From these values the weighting factors for disturbance and reassurance have been calculated as shown in Table 36.

Table 36 Weighting factors for attributes and sub-attributes applied in the optimisation of remediation of the Ravenglass site.

Health factors			Economic factors		Social factors	
1.08×10^{-3}		1.11×10^{-3}	0.999	0.999	2.71×10^{-4}	2.78×10^{-4}
Dose population	0.90	0.90	Remediation costs	0.385	Reassurance	0.86
Dose workers	0.10	0.10	Waste disposal costs	0.588	Disturbance	0.14
Non-radiation	-	-	Monitoring costs	0.026	Loss/gain of income	-

Note: The values in the left of the double columns are for an integration time of 100 years and in the right column for an integration time of 500 years.

It should be emphasised that value setting of weighting factors is the crucial issue of any optimisation because subjective judgements inevitably will enter the process.

6.2 Results

IAEA has proposed clean-up criteria in terms of individual doses. The individual doses assessed at the Ravenglass Estuary are of the order of 1.5 mSv a^{-1} at the time of decision to introduce remediation (year 1). According to the IAEA criteria, clean-up is almost always needed for an individual dose range of $1-10 \text{ mSv a}^{-1}$ if a constraint for controlled practices is applied. Even without the application of a constraint, IAEA suggests that for individual doses of $1-10 \text{ mSv a}^{-1}$ clean-up would usually be needed. Based on these recommendations it can therefore be concluded that some kind of remediation would almost always be justified for the Ravenglass site.

In a cost-benefit approach, the monetary costs, X , of the remediation strategies are compared with the benefit of the collective dose reduction, ΔS . The net benefit, ΔB , is given as:

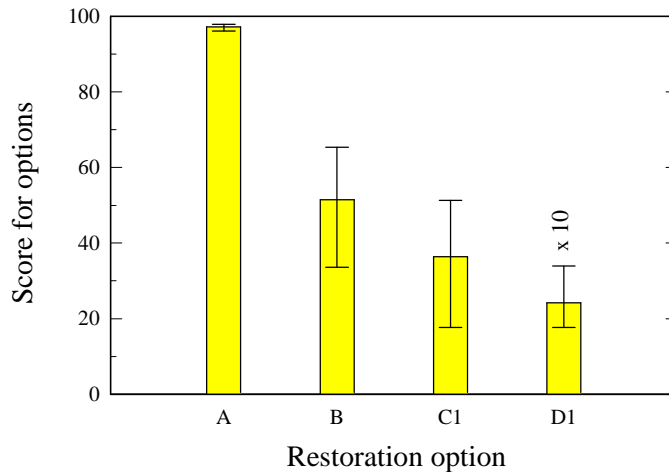
$$\Delta B = \alpha \cdot \Delta S - (\alpha \cdot S_{work} + X_{remedia} + X_{waste} + X_{monitor})$$

None of the remedial options are justified on economic grounds alone when only the central estimates of collective dose are used together with an α -value of $100,000 \text{ EUR manSv}^{-1}$. Not even with a higher value of α (e.g. $200,000 \text{ EUR manSv}^{-1}$) and more extreme values from the calculated collective dose distribution (e.g. the 95th percentile) would make any of the options economically justified, for any of the integration times for the collective doses.

In the multi-attribute approach, overall scores, U_i , of the remediation options i has been determined from the weighted sum of utilities for each of the attributes considered:

$$\begin{aligned}
 U_i &= \sum_{j=1}^3 w_j \cdot u_{ij} \\
 &= w_{health} \cdot (w_{dose,pop} \cdot u_{dose,pop} + w_{dose,work} \cdot u_{dose,work}) \\
 &+ w_{economic} \cdot (w_{waste} \cdot u_{waste} + w_{remedia} \cdot u_{remedia} + w_{monitor} \cdot u_{monitor}) \\
 &+ w_{social} \cdot (w_{distur} \cdot u_{distur} + w_{reas} \cdot u_{reas})
 \end{aligned}$$

The weighting factors above have all been sampled in a triangular distribution between 0.67-1.5 times the most probable value given in Table 36. Similarly, the values of all the utilities, $u(x)$, are determined from the utility functions in which the values of x are sampled in a triangular distribution between 0.67 - 1.5 times the central values of x given in Table 36. Negative correlation between collective doses and remediation costs has been applied with a correlation coefficient of -0.8 . The evaluation of the different strategies has been made with the forecasting and risk analysis program CRYSTAL BALL. Latin Hypercube Sampling technique was used and the number of trials were 10,000. The results for the scores, U_i , for the Options A - D1 are shown in Figure 15. The error bars represent the 5th and 95th percentiles of the distributions of U_i .



Note: Identical scores are found for an integration time of 100 years.

Figure 15 Overall evaluation of scores for different remediation strategies for the Ravenglass site for an integration time of 500 years for the collective dose.

It appears from Figure 15 that the Option A (no remediation) has the highest score. The scores for Option B (source removal) and C1 (soil washing) are both significantly lower than for Option A. Due to the highest total costs for Option D1 (chemical solubilisation) this option has the lowest score. The 'no remediation' option A is thus the best solution for the Ravenglass site and also the cheapest.

The ranking of the different remedial measures using multi-attribute utility analyses allows the inclusion of factors that are not easy to quantify in monetary terms as is required in cost-benefit analysis. The weighting factors assigned to the different attributes have been determined by use of scaling factors in terms of weighting factor ratios, and their values were sampled around a most probable value. Notwithstanding this advantage of the multi-attribute method there are difficulties with the determination of weighting factors for the different attributes. Without any terms of reference for the weighting between attributes, value settings by a decision maker could lead to 'optimised' results that might be useless because of a subjective bias of the decision maker in the selection of weighting factors. Therefore, the

outcome of any multi-attribute analysis, including the present study, should be judged very carefully in the light of the values assigned to the weighting factors before any firm conclusions could be drawn.

7. Conclusions

The purpose of this technical deliverable has been to provide an example site to demonstrate the application of the decision-making procedure, developed by RESTRAT, to remediate the Ravenglass estuary. Each step of the proposed procedure has been carried out. The site was characterised through a review of the available data, and a number of areas of contamination were identified along with pathways through which man can be exposed to these contaminants. Characterisation enabled the doses, arising from the site, to be quantified through the development of a model. Restoration techniques which were appropriate to this site were identified and the impact of these doses on the locality were calculated from the model. These performances of the techniques were combined with the economic and social costs in a multi-attribute utility analysis to rank each restoration technique.

For the Ravenglass Estuary, the procedure predicts that the 'no remediation' option is the preferable course of action. The application of restoration techniques for this particular example site cannot be justified. This outcome was to be expected given the size of the estuary, the small local population, the small production of foodstuff and the low concentration of radionuclides. Indeed, restoration of the site has not previously been considered necessary.

Although the results from the multi-attribute utility analysis show that the 'no remediation' option was clearly the best option for this example site, the high level of uncertainties meant that there was significant overlap in the ranking of the next two restoration techniques (sediment removal and sediment washing). The uncertainty arose both from the lack of precision in characterising the site and from the uncertainties associated with the restoration techniques themselves.

The uncertainties associated with the parameters of the site reflect the fact that the estuary is a highly dynamic environment where conditions change rapidly through tidal processes and the reworking of the sediment. In addition, the radionuclide discharges from the nearby Sellafield plant, which was the original source of the radionuclides in the estuary, has varied over the years. This leads to variable levels of radionuclides within the estuarine sediment. Therefore, the data concerning radionuclide distribution becomes less precise as it becomes older. This in turn leads to a lack of precision with regard to defining the performance of each restoration method. These can only be defined in terms of a range of values which generates the high levels of uncertainty in the multi-attribute utility analysis.

In spite of the problems presented by the lack of precision of the characterisation of the site, the multi-attribute utility analysis demonstrates that it is a fairly robust approach which, for this site, produces a clear-cut outcome.

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Appendix A Calculations of the volumes of waste and residual activity fractions after remediation of the Ravenglass Estuary

A1 Dimensions of site

The Ravenglass Estuary contains an estimated 4.5 TBq of ^{137}Cs , 4 TBq of $^{239+240}\text{Pu}$ and 4 TBq of ^{241}Am (Kelly and Emptage, 1992; Eakins, Burton, Humphreys and Lally, 1985).

It was estimated through a summation of the data given by Kelly and Emptage (1992), assuming specific depths of contamination for different sediment facies that the total volume of contaminated sediment for Ravenglass was $1.3 \times 10^6 \text{ m}^3$.

A2 Volume of waste and residual activity arising from remediation process

Using data from Zeevaert and Bousher (1998), in TD3+4, the specific performance data for remediation techniques which are appropriate to the Ravenglass Estuary may be estimated. These are summarised in Table A1. This data may be used to estimate the volume of waste generated through the remediation of the site and the fraction of the activity remaining on-site after remediation. These are summarised in Table A2. The calculations used to arrive at these values are described in the following sections.

A3.1 Source Removal

Assuming that the remediation process is the excavation of the site. Then,

$$\text{Volume of waste} = 1.3 \times 10^6 \text{ m}^3$$

The decontamination factor for the site is taken to be 5 (see Table A1).

Consequently,

$$\text{Residual activity fraction} = 1 / 20 = 0.05$$

A3.2 Soil Washing

Waste reduction is 80% (see Table A1). Therefore,

$$\text{Volume of waste} = 1.3 \times 10^6 \times (1 - 0.2) = 2.6 \times 10^5 \text{ m}^3.$$

A common decontamination factor of 5 is used for all radionuclides.

Consequently,

$$\text{Residual activity fraction} = 1 / 5 = 0.2$$

A3.3 Chemical Solubilisation

The extraction and processing of the contaminated sediment is readily calculated. However, there is little information concerning the fate of the solubilised radionuclides. In all probability it would be treated further to reduce the volume. Here it is assumed that the liquid waste will be stored in containers off-site. Any additional treatment will thus occur off-site.

A further problem is that there is no defined concentration of radionuclides in the liquid arising from the separation. For the purposes of this calculation, an arbitrary concentration of 8.3 MBq m^{-3} has been assumed. However, it should be noted that the higher the concentration of radionuclides in the liquid, the smaller the volume and thus, the lower the disposal cost.

The decontamination factor is radionuclide specific (see Table A1). For the purposes of these calculations, as a first approximation, an single value is assumed for all decontamination factors (5). Hence, the process generates $4.5 \times 10^{12} \times (4/5) = 3.6 \times 10^{12}$ Bq in the liquid waste.

If the concentration is 8.3 MBq m^{-3} then,

$$\begin{aligned} \text{Volume of waste} &= 3.6 \times 10^{12} / 8.3 \times 10^6 = 4.3 \times 10^5 \text{ m}^3 \\ \text{Residual activity factor} &= 1 / 5 = 0.2 \end{aligned}$$

A4 References

Eakins J.D., Burton P., Humphreys D.G. and Lally A.E. (1985) The remobilisation of actinides contaminated intertidal sediments in the Ravenglass Estuary. In: *Seminar on the behaviour of radionuclides in estuaries*, Renesse, 17-21 September 1984, CEC.

Kelly M. and Emptage M. (1992) Distribution of radioactivity in the Esk estuary and its relationship to sedimentary processes. DoE Report No. DoE/HMIP/RR/92/015, DoE, London.

Zeevaert T. and Bousher A. (1998) Restoration techniques: characteristics and performances. RESTRAT Technical Deliverable TD3+4.

Table A1 The performance of remediation techniques applied to the Ravenglass site.

Remediation Technique	Unit [†]	Value
Source removal Soil excavation	DF	20
Physical separation (<i>ex-situ</i>) Soil washing	DF RF	5 80%
Chemical separation (<i>ex-situ</i>) Chemical solubilisation	DF	20 (Cs) 3.3 (U, Am, Pu)

[†] DF = decontamination factor; RF = waste reduction factor.

Table A2 The volume of waste produced and activity remaining after remediation of the Ravenglass Estuary.

Remediation strategy	Volume of waste removed from site m^3	Fraction of activity remaining after remediation
Source removal	1.3×10^6	0.05
Soil washing	2.6×10^5	0.2
Chemical solubilisation	4.3×10^5	0.2

Appendix B Ravenglass Estuary cost calculations

B1 Dimensions of the site

It was estimated through a summation of the data given by Kelly and Emptage (1992), assuming specific depths of contamination for different sediment facies that the total volume of contaminated sediment for Ravenglass was $1.3 \times 10^6 \text{ m}^3$.

B2 Cost of Remediation Process

Using the specific performance data given in Appendix A and the costs of remediation techniques given by Zeevaert and Bousher (1998), in TD3+4 it is possible to determine the remediation, waste disposal and monitoring costs which are specific to the remediation of the Ravenglass Estuary. The data from these sources are summarised in Tables B1 and B2. The costs arising from remediation of the site and disposing of the waste are summarised in Table A3. The calculations used to arrive at these values are described in the following sections.

Table A3 also gives estimates for the monitoring costs after remediation. For the purposes of these calculation it has been assumed that monitoring costs are linearly dependent upon the fraction of activity remaining on the site after remediation. In addition, the cost of monitoring the site, if no remediation has taken place, is assumed to be $5.25 \times 10^7 \text{ EUR}$.

B2.1 Source Removal

Excavation costs (including transport) are 100 EUR m^{-3} (see Table A1).

The costs of excavation and disposal at a Resource Conservation Recovery Act permitted facility (RCRA) (including transport) are 700 EUR m^{-3} (see Table A1).

RCRA disposal (plus transport) is, therefore, 600 EUR m^{-3} (assuming any common transport costs are negligible).

Consequently,

$$\begin{aligned} \text{Remediation cost} &= 100 \times 1.3 \times 10^6 &= 1.3 \times 10^8 \text{ EUR} \\ \text{Waste disposal cost} &= 600 \times 1.3 \times 10^6 &= 7.8 \times 10^8 \text{ EUR} \end{aligned}$$

The fraction of activity remaining on the site after remediation was 0.05. Consequently,

$$\text{Monitoring costs} = 5.25 \times 10^7 \times 0.05 = 2.6 \times 10^6 \text{ EUR}$$

B3.2 Soil Washing

Excavation, transport and soil washing costs are 400 EUR m^{-3} (see Table A1).

The volume of waste disposed is $2.6 \times 10^5 \text{ m}^{-3}$ (see Table A2).

Disposal costs of the radioactive wastes will be 2500 EUR m^{-3} (see Table A1).

Therefore,

$$\begin{aligned} \text{Remediation cost} &= 400 \times 1.3 \times 10^6 &= 5.2 \times 10^8 \text{ EUR} \\ \text{Waste disposal cost} &= 2500 \times 2.6 \times 10^5 &= 6.5 \times 10^8 \text{ EUR} \end{aligned}$$

The fraction of activity remaining on the site after remediation was 0.2. Consequently,

$$\text{Monitoring costs} = 5.25 \times 10^7 \times 0.2 = 1.05 \times 10^7 \text{ EUR}$$

B3.3 Chemical Solubilisation

Excavation, transport and treatment costs are 550 EUR m⁻³ (see Table A1).

There will be 4.3 × 10⁵ m³ of waste solution (see Table B2).

Disposal costs of the wastes will be 2500 EUR m⁻³ (assumed to be the same as that for solid waste, see Section A3.2).

Consequently,

$$\text{Remediation cost} = 550 \times 1.3 \times 10^6 = 7.2 \times 10^8 \text{ EUR}$$

$$\text{Waste disposal cost} = 2500 \times 4.3 \times 10^5 = 1.1 \times 10^9 \text{ EUR}$$

The fraction of activity remaining on the site after remediation was 0.2. Consequently,

$$\text{Monitoring costs} = 5.25 \times 10^7 \times 0.2 = 1.05 \times 10^7 \text{ EUR}$$

B4 References

Zeevaert T. and Bousher A. (1998) Restoration techniques: characteristics and performances. RESTRAT Technical Deliverable TD3+4.

Table B1 The costs of remediation techniques applied to the Ravenglass site.

Remediation Technique	Cost
<u>Source Removal</u>	
Soil excavation	
Excavation costs (including transport)	100 EUR m ⁻³
Excavation costs (including transport and RCRA disposal)	700 EUR m ⁻³
Disposal cost for radioactive material (including transport)	2500 EUR m ⁻³
<u>Physical Separation (ex-situ)</u>	
Soil washing	
Soil washing costs	300 EUR m ⁻³
Cost of excavation and transport (prior to washing)	100 EUR m ⁻³
Cost of disposal of radioactive residue (including transport)	2500 EUR m ⁻³ (residue)
<u>Chemical Separation (ex-situ)</u>	
Chemical solubilisation	
Separation costs	450 EUR m ⁻³
Cost of excavation and transport (prior to solubilization)	100 EUR m ⁻³
Cost of disposal of radioactive residue (including transport)	2500 EUR m ⁻³ (residue)

Table B2 The volume of waste produced and activity remaining after remediation of the Ravenglass Estuary.

Remediation strategy	Volume of waste removed from site m ³	Fraction of activity remaining after remediation
Source removal	1.3×10^6	0.05
Soil washing	2.6×10^5	0.2
Chemical solubilisation	4.3×10^5	0.2

Data from Appendix A.

Table B3 The costs of applying remediation technologies to the Ravenglass estuary.

Remediation strategy	Monetary costs of remediation [EUR]		
	Remediation (incl. labour)	Waste disposal (incl. transport)	Monitoring costs
Source removal	1.3×10^8	7.8×10^8	2.6×10^6
Soil washing	5.2×10^8	6.5×10^8	1.05×10^7
Chemical solubilisation	7.2×10^8	1.1×10^{10}	1.05×10^7

Appendix C The Water Column

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Samples were usually analysed twice: first without further treatment (Columns A), and second after filtering, in three steps for particle sizes of 450, 100, and 15 nm (Columns B). Columns labelled with MW give the molecular weight of the atoms or ionic units specified in the first column, "n.d." stands for "not determined". Upper limit values denote, that the analytical value was below the respective detection limit. The methods given in the last column are: IC = Ion Chromatography, ICP-MS = Inductively Coupled Plasma - Mass Spectrometry, F-AAS = Flame - Atom Adsorption Spectrometry, they all apply to the samples analysed by FZ Rossendorf. Radium (with ^{223}Ra and ^{224}Ra being neglected) and thorium (determined from equilibrium with ^{228}Ac , other thorium nuclides being neglected) were analysed with g - Spectrometry, and here the measured values are given in Bq kg^{-1} .

Table C1 Analysis of water from the Drigg stream confluence of the Ravenglass Estuary.

Component	A $\mu\text{g L}^{-1}$	B $\mu\text{g L}^{-1}$	MW g mol^{-1}	A mol L^{-1}	B mol L^{-1}	Method
PO_4^{3-}	7800	6800	94.971	8.21E-05	7.16E-05	IC
NO_3^-	3800	3200	62.005	6.13E-05	5.16E-05	IC
SO_4^{2-}	253000	255000	96.058	2.63E-03	2.66E-03	IC
HCO_3^-	<2000	<2000	61.017	<3.28E-05	<3.28E-05	IC
Cl^-	1700000	1720000	35.453	4.80E-02	4.85E-02	IC
Si	1270	1500	28.086	4.52E-05	5.34E-05	ICP-MS
K^+	39500	37500	39.098	1.01E-03	9.59E-04	F-AAS
Na^+	984000	959000	22.99	4.28E-02	4.17E-02	F-AAS
Ca^{2+}	43100	48900	40.08	1.08E-03	1.22E-03	ICP-MS
Mg^{2+}	107000	115000	24.305	4.40E-03	4.73E-03	F-AAS
Fe	<100	< 50	55.847	<1.79E-06	<8.95E-07	F-AAS
Zn^{2+}	n.d.	37.6	65.38		5.75E-07	ICP-MS
U	0.59	0.27	238.03	2.48E-09	1.13E-09	ICP-MS
Ni^{2+}	n.d.	9.2	58.7		1.57E-07	ICP-MS
Mn^{2+}	2	93.7	54.938	3.64E-08	1.71E-06	ICP-MS
As	n.d.	4.5	74.922		6.01E-08	ICP-MS
^{226}Ra	1.99	-	-	2.40E-13	-	GAMMA
^{232}Th	0.17	-	-	1.72E-07	-	GAMMA

Note: Suspended material was determined to be 6.5 mg L^{-1} , and the *in-situ* pH was 7.02.

Table C2 Analysis of water from the Eskmeal viaduct of the Ravenglass Estuary.

Component	A $\mu\text{g L}^{-1}$	B1 $\mu\text{g L}^{-1}$	B2 $\mu\text{g L}^{-1}$	MW g mol^{-1}	A mol L^{-1}	B1 mol L^{-1}	B2 mol L^{-1}	Method
SO ₄ ²⁻	1230000	1300000	1340000	96.058	1.28E-02	1.35E-02	1.40E-02	IC
HCO ₃ ⁻	<2000	<2000	<2000	61.017	<3.28E-05	<3.28E-05	<3.28E-05	IC
Cl ⁻	9340000	9440000	9620000	35.453	2.63E-01	2.66E-01	2.71E-01	IC
Si	630	890	1060	28.086	2.24E-05	3.17E-05	3.77E-05	ICP-MS
K ⁺	186000	188000	200000	39.098	4.76E-03	4.81E-03	5.115E-03	F-AAS
Na ⁺	5290000	5280000	5361000	22.99	2.30E-01	2.30E-01	2.33E-01	F-AAS
Ca ²⁺	196000	215000	207000	40.08	4.89E-03	5.36E-03	5.17E-03	ICP-MS
Mg ²⁺	572000	660000	652000	24.305	2.35E-02	2.72E-02	2.68E-02	F-AAS
Fe	<100	<50	<50	55.847	<1.79E-06	<8.95E-07	<8.95E-07	F-AAS
Zn ²⁺	n-d.	44	30.8	65.38	-	6.73E-07	4.71E-07	ICP-MS
U	2.22	3.7	3.3	238.03	9.33E-09	1.55E-08	1.39E-08	ICP-MS
Ni ²⁺	n-d.	17.9	14.2	58.7	-	3.05E-07	2.42E-07	ICP-MS
Mn ²⁺	n-d.	22.3	17.2	54.938	-	4.06E-07	3.13E-07	ICP-MS
As	21	25.6	25.8	74.922	2.80E-07	3.42E-07	3.44E-07	ICP-MS
²²⁶ Ra	0.95	-	-	-	1.21E-13	-	-	GAMMA
²³² Th	0.27	-	-	-	2.72E-07	-	-	GAMMA

Note: In these analyses the effects of aging were studied by dividing the filtered sample and allowing one part (B2) to age another 72 hours before analysis. No significant difference was observed with the sample (B1) which was analysed immediately. Suspended material was determined as 47 mg L⁻¹, and the pH, measured *in-situ*, was 8.04.

Table C3 Analysis of water from 50 m inside the mouth of Ravenglass Estuary.

Component	A $\mu\text{g L}^{-1}$	B $\mu\text{g L}^{-1}$	MW g mol^{-1}	A mol L^{-1}	B mol L^{-1}	Method
SO ₄ ²⁻		2.40E+06	96.058		2.50E-02	IC
HCO ₃ ⁻		125000	61.017		2.05E-03	IC
Cl ⁻		1.70E+07	35.453		4.80E-01	IC
Si	1320	782	28.086	4.70E-05	2.78E-05	ICP-MS
K ⁺	353000	380000	39.098	9.03E-03	9.72E-03	F-AAS
Na ⁺	9180000	9810000	22.99	3.99E-01	4.27E-01	F-AAS
Ca ²⁺	410000	400000	40.08	1.02E-02	9.98E-03	ICP-MS
Mg ²⁺	1110000	1160000	24.305	4.57E-02	4.77E-02	F-AAS
Fe	220	120	55.847	3.94E-06	2.15E-06	F-AAS
Al ³⁺	298	1.9	26.9815	1.10E-05	7.04E-08	ICP-MS
Zn ²⁺	12.4	11.1	65.38	1.90E-07	1.70E-07	ICP-MS
U	3.8	4.5	238.03	1.60E-08	1.89E-08	ICP-MS
Th	0.2	0.1	232.0381	8.62E-10	4.31E-10	ICP-MS
Pb ²⁺	1.6	0.2	207.2	7.72E-09	9.65E-10	ICP-MS
Ni ²⁺	<10	<10	58.7	<1.70E-07	<1.70E-07	ICP-MS
Mn ²⁺	16.5	1.25	54.938	3.00E-07	2.28E-08	ICP-MS
Cd ²⁺	1	<0.4	112.41	8.90E-09	<3.56E-09	ICP-MS

Note: Before filtering the pH was 7.94, and the redox potential Eh, which was determined *in-situ*, was +121.3 mV.

Table C4 Analysis of water at the barrier of the mouth of the Ravenglass Estuary.

Component	A µg L ⁻¹	B µg L ⁻¹	MW g mol ⁻¹	A mol L ⁻¹	B mol L ⁻¹	Method
SO ₄ ²⁻		2.30E+06	96.058		2.39E-02	IC
HCO ₃ ⁻		128000	61.017		2.10E-03	IC
Cl ⁻		1.77E+07	35.453		4.99E-01	IC
Si	816	862	28.086	2.91E-05	3.07E-05	ICP-MS
K ⁺	360000	380000	39.098	9.21E-03	9.72E-03	F-AAS
Na ⁺	9180000	9880000	22.99	3.99E-01	4.30E-01	F-AAS
Ca ²⁺	373000	380000	40.08	9.31E-03	9.48E-03	ICP-MS
Mg ²⁺	1130000	1150000	24.305	4.65E-02	4.73E-02	F-AAS
Fe	60	<50	55.847	1.07E-06	<8.59E-07	F-AAS
Al ³⁺	104	4.2	26.9815	3.85E-06	1.56E-07	ICP-MS
Zn ²⁺	8.3	7.9	65.38	1.27E-07	1.21E-07	ICP-MS
U	3.8	3.8	238.03	1.60E-08	1.60E-08	ICP-MS
Th	0.3	0.2	232.0381	1.29E-09	8.62E-10	ICP-MS
Pb ²⁺	1	0.2	207.2	4.83E-09	9.65E-10	ICP-MS
Ni ²⁺	<10	<10	58.7	<1.70E-07	<1.70E-07	ICP-MS
Mn ²⁺	9	0.86	54.938	1.64E-07	1.57E-08	ICP-MS
Cd ²⁺	<0.6	<0.4	112.41	<5.34E-09	<3.56E-09	ICP-MS

Note: Before filtering, the pH was 7.93, and the redox potential, Eh, *in-situ* was +124 mV.

Table C5 Analysis of water from 50 m outside the mouth of the Ravenglass Estuary.

Component	A µg L ⁻¹	B µg L ⁻¹	MW g mol ⁻¹	A mol L ⁻¹	B mol L ⁻¹	Method
SO ₄ ²⁻		2.60E+06	96.058		2.71E-02	IC
HCO ₃ ⁻		124000	61.017		2.03E-03	IC
Cl ⁻		1.80E+07	35.453		5.08E-01	IC
Si	785	1230	28.086	2.80E-05	4.38E-05	ICP-MS
K ⁺	357000	378000	39.098	9.13E-03	9.67E-03	F-AAS
Na ⁺	9180000	9690000	22.99	3.99E-01	4.22E-01	F-AAS
Ca ²⁺	380000	386000	40.08	9.48E-03	9.63E-03	ICP-MS
Mg ²⁺	1120000	1170000	24.305	4.61E-02	4.81E-02	F-AAS
Fe	140	<50	55.847	2.51E-06	<8.95E-07	F-AAS
Al ³⁺	108	4	26.9815	4.00E-06	1.48E-07	ICP-MS
Zn ²⁺	6	5	65.38	9.18E-08	7.65E-08	ICP-MS
U	3.9	4.2	238.03	1.64E-08	1.76E-08	ICP-MS
Th	0.3	0.3	232.0381	1.29E-09	1.29E-09	ICP-MS
Pb ²⁺	1	0.2	207.2	4.83E-09	9.65E-10	ICP-MS
Ni ²⁺	<10	<10	58.7	<1.70E-07	<1.70E-07	ICP-MS
Mn ²⁺	9.7	1.34	54.938	1.77E-07	2.44E-08	ICP-MS
Cd ²⁺	<0.5	<0.7	112.41	<4.45E-09	<6.22E-09	ICP-MS

Note: Before filtering, the pH was 7.94, and the redox potential, Eh, *in-situ* was +129.1 mV.

Table C6 Analysis of pore water from mud at the mussel beds of the Ravenglass Estuary.

Component	B1 $\mu\text{g L}^{-1}$	B2 $\mu\text{g L}^{-1}$	MW g mol^{-1}	B1 mol L^{-1}	B2 mol L^{-1}	Method
SO_4^{2-}	1050000	1190000	96.058	1.09E-02	1.24E-02	IC
HCO_3^-	1330000	1110000	61.017	2.18E-02	1.82E-02	IC
Cl^-	12300000	14800000	35.453	3.47E-01	4.18E-01	IC
Si	9230	12600	28.086	3.29E-04	4.49E-04	ICP-MS
K^+	226000	276000	39.098	5.78E-03	7.06E-03	F-AAS
Na^+	6460000	7360000	22.99	2.81E-01	3.20E-01	F-AAS
Ca^{2+}	242000	263000	40.08	6.04E-03	6.56E-03	ICP-MS
Mg^{2+}	778000	910000	24.305	3.20E-02	3.74E-02	F-AAS
Fe	220	5700	55.847	3.94E-06	1.02E-04	F-AAS
Al^{3+}	22.7	1.2	26.9815	8.41E-07	4.45E-08	ICP-MS
U	8.4	4.5	238.03	3.53E-08	1.89E-08	ICP-MS
Pb^{2+}	0.2	0.4	207.2	9.65E-10	1.93E-09	ICP-MS
Mn^{2+}	18700	14400	54.938	3.40E-04	2.62E-04	ICP-MS

Note: As the water was pore water from muddy sediments, analysis before filtering was not meaningful. The pH was measured only after filtering and was 7.94 (B1) and 7.29 (B2).

Table C7 Analysis of water from the open Irish Sea.

Component	R $\mu\text{g L}^{-1}$	MW g mol^{-1}	R mol L^{-1}
SO_4^{2-}	2520000	96.058	2.62E-02
HCO_3^-	22000	61.017	3.61E-04
Cl^-	17960000	35.453	5.07E-01
K^+	371000	39.098	9.49E-03
Na^+	10000000	22.99	4.35E-01
Ca^{2+}	383000	40.08	9.56E-03
Mg^{2+}	1210000	24.305	4.98E-02

Note: The typical pH of the Irish Sea is around 8.2.