



FINAL REPORT

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ACRONYM : REMOTRANS

TITLE :Processes Regulating Remobilisation, Bioavailability and Translocation in Marine Sediments.

PROJECT CO-ORDINATOR : Department of Radiation Physics,
Lund University (ULUND) SE

PARTNERS : :

Institut de Radioprotection et de Sureté Nucléaire	(IRSN)	FR
Radiological Protection Institute of Ireland	(RPII)	IRL
Norwegian Radiation Protection Authority	(NRPA)	NO
Agricultural University of Norway	(AUN)	NO
Risoe National Laboratory	(RISOE)	DK
Institutt for Energiteknikk	(IFE)	NO
The Centre for Environment, Fisheries and Aquaculture Science	(CEFAS)	UK
National University of Ireland, University College Dublin	(NUI-UCD)	IRL
National Agency for Energy, new Technology and Environment	(ENEA)	I
Universidad de Sevilla	(USEV)	ESP
Centro de Investigaciones Energéticas, Medioambientales y Tecnológicas	(CIEMAT)	ESP

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1. Introduction.

Radioactive waste has been discharged into the Irish Sea as a consequence of reprocessing radioactive waste at Sellafield (formerly Windscale) since 1952. In the intervening period approximately 610 TBq of $^{239+240}\text{Pu}$ and 41 PBq of ^{137}Cs have been discharged. Together with these radionuclides there have been significant releases of ^{99}Tc during different periods and also radionuclides such as ^{237}Np and ^{129}I . Such more conservatively behaving radionuclides can be used as tracers for the water movements and mixing from the source. Releases from the French reprocessing facility, La Hague, has also contributed, but to a relatively smaller extent, to radioactivity in European waters.

As plutonium is a particle reactive substance much of it has been removed from the water column and has found its way to the fine grain and muddy sediments of the west coast of Britain. Other radionuclides, such as Tc and Np are expected to behave more conservatively in marine waters while radio-caesium especially in coastal waters can be scavenged associated with clay particles.

The Baltic sea is a marginal sea and was the sea which was most contaminated following the Chernobyl accident. The inventory of ^{137}Cs is estimated to 5 PBq. The Baltic Sea is especially vulnerable due to lower salinity, which causes higher concentration factors in biota. While caesium is substantially more conservatively than plutonium much of the activity in the shallow Baltic sea has also been deposited in the sediments. A significant amount, about 60 TBq per year, is leaving the Baltic sea to the North sea with the net amount of outgoing water corresponding to the outflow from rivers.

However such sediments cannot be considered a final repository for artificial radionuclides and in fact there is strong evidence that the contaminated sediments have become a source of re-mobilised radionuclides.

The deposition of Chernobyl radionuclides onto the Mediterranean Sea (about 2.5 PBq of ^{137}Cs) was very patchy and was highest in the northern and eastern basins. In general radionuclide levels in seawater returned to pre-Chernobyl values within a few years after the accident, mainly due to dilution with non-contaminated water coming from the southern and western basins and to convection processes that have efficiently transported them to depth. However the Northern Adriatic Sea, besides a significant atmospheric input of ^{137}Cs (around 1 PBq) in the period following the accident received a further contribution from the main Italian rivers, draining heavily contaminated areas (average deposition 10 kBq m^{-2}). For this reason, ^{137}Cs concentrations in the Adriatic have remained higher than in other Mediterranean basins until 1990, and considerable quantities of Chernobyl radionuclides have been found in sedimentary deposits near the river deltas. From the vertical profiles of natural and artificial radionuclides there is some evidence that these sedimentary deposits are not stable and that a significant fraction of the radionuclides initially stored in these sites can be re-distributed in a wider area.

Plutonium concentrations in the Mediterranean Sea are much higher than in the Baltic, because the Mediterranean is a deep sea with relatively low particulate load and scavenging processes, able to remove plutonium from the water column at short time scales, are mainly on the shelf (less than 20% of the total surfaces). There are only few data on plutonium inventories in sediments, particularly in deep basins, but it is evident that for water depths greater than 1000m the inventories are a few percent of the cumulative fallout deposition, while on the shelf they can reach values higher than twice this cumulative fallout deposition. This implies that lateral transport of plutonium towards the shelf and enhanced scavenging in this area are important mechanisms for the removal of this radionuclide from the water column of the hole basin.

Certain marine areas, such as particular fjords, have natural boundaries such as anoxic-oxic interfaces, ice-water interfaces (Arctic) an/or saline-freshwater interphases. This causes natural remobilisation and accumulation processes with higher levels in the water especially actinides, close to the bottom. Such fjords were used as natural laboratories for the project.

For executing the REMOTRANS project field work was necessary. The sampling programme of water, sediment and biota was carried out using national scientific ships regularly operating in the areas of interest. Existing agreements between research stations were used for joint expeditions. The parties within the consortium have a wide experience in and possess equipment for such expeditions including the Arctic.

For the subsequent analysis the participants used their well established techniques for the programmes that were carried out such as radiochemical separation, alpha, beta and gamma spectrometry, mass spectrometry, speciation studies. The analytical quality was guaranteed and controlled by participation in international intercalibration exercises.

Several of the laboratories have experience from radioecological modelling which was taken place under a specific work package. The data from the field and laboratory studies were made available to the modelling work package participants.

REMOTRANS consisted of 6 work packages which all had a work package leader. This structure was kept throughout the project.

Work-package 1 Inventories. Responsible RPII

Work-package 2 Time-series. Responsible RISOE

Work-package 3 Remobilisation. Responsible NUID-UCD

Work-package 4 Bioavailability. Responsible CEFAS

Work-package 5 Modelling. Responsible NRPA

Work-package 6 Reporting. Responsible ULUND (co-ordinator)

2. Objectives.

The aim of the project was to study remobilization of radiologically important radionuclides from sediments of different characteristics in different European environments. The radiological consequences and transboundary exposure were evaluated including uptake in biota and radioecological modelling. The results created a more precise knowledge of long-term effects on environment and man from past and present sources and a more informed debate on waste management of controlled releases of radioactivity in the marine environment.

Important objectives were to enhance European competence in the field of environmental radioactivity, encourage the transfer of skills, provide training and increase mobility for younger scientists.

The REMOTRANS project provided a basis for informing the public, the news media and politicians of current development and may prove to be of special value in emergency situations.

When dealing with the environmental transfer of radionuclides in the marine environment, assessment of source terms, transfer factors, models and committed doses has generally only considered the actual annual releases from nuclear facilities. Remobilization of radionuclides initially associated with sediments must be taken into consideration for realistic assessment especially in the long term.

The aim of the project was to study remobilization of radiologically important radionuclides from sediments of different characteristics in different European environments. The radiological consequences and transboundary exposure will be evaluated including uptake in biota and radioecological modelling. The results will create a more precise knowledge of long-term effects on environment and man from past and present sources and a more informed debate on waste management of controlled releases of radioactivity in the marine environment. The actual effects of reducing discharges to the marine environment (e.g. Sellafield and la Hague) or shutting down nuclear facilities has not necessarily resulted in the expected decrease in activity and doses. The study of the mechanisms for remobilization and of bio-availability of remobilised radionuclide species is an innovative approach which will lead to a more robust understanding of marine environmental radioactivity. The multinational collaboration makes it possible to reach and perform sampling in different European marine environments.

The overall objective of the project was achieved through a programme of work which follows the following steps:

- A sediment sampling and analysis programme leading to improved knowledge of inventories and quantifying potential sediment source terms.
- Perform time series measurements of radionuclides at key locations to quantify transport rates and transfer factors.
- Develop a protocol for and perform determination of physico-chemical forms of the radionuclides in order to assess their potential remobilization ability.
- Perform laboratory experiments to assess the impact of remobilization on bioavailability.
- Develop radioecological models taking remobilization processes into account.

Work performed. Improved radionuclide inventory estimates has been obtained for 5 principal marine European waters where contaminated sediments have been identified for study both in terms of inventory characterisation and remobilization studies. These areas include the Irish Sea, Skagerrak, and Kattegatt, the estuaries of the Odiel and Tinto rivers, the Rhone Delta and the Northern Adriatic. Depth distributions of americium and plutonium have been studied in the Irish Sea. Long-distant transport of plutonium to Skagerrak and Kattegatt has been demonstrated. Depth distributions of ^{129}I in Tinto river show additional contribution than from nuclear tests. The inventories at the Rhone Delta are studied by deep coring. In the northern Adriatic Sea similar studies were carried out at the mouths of Isonzo and Tagliamento rivers.

Study of the translocation in time and space of radionuclides (^{137}Cs , ^{99}Tc , ^{129}I , ^{239}Pu , ^{240}Pu , ^{237}Np) from the sources Sellafield and La Hague to key areas has been accomplished. Such key areas are the west coast of Ireland, the Swedish west coast, south and north Norwegian coast, and the Barents Sea. The releases of ^{99}Tc in 1994 reached the Arctic in 2000-2002. The translocation of remobilised sediments show transfer of plutonium from the eastern to the western Irish Sea. There is also an indication of remobilization of Chernobyl derived ^{137}Cs in the Baltic Sea.

A sequential extraction protocol for the studying the solid partitioning of plutonium has been developed. This protocol will be used for the future work within future projects. Oceanographic expeditions have been done in the Kongsfjord-Krossfjord system (Svalbard) in order to investigate sedimentation/resuspension processes at the glacier-sea interphase, the origin and pathways of radionuclides associated with suspended sedimentary particles and the stability of sedimentary deposits. The remobilization processes linked to organic colloidal matter are studied at permanently anoxic fjords. Results from the meromictic lake, Rørholt fjord, Norway show a remobilisation and an accumulation of radiocaesium in the saline layer. The influence of pore-water chemistry and the development of anoxic conditions on remobilisation of plutonium has been studied. The remobilisation of plutonium by resuspension has been demonstrated.

Mesocosm experiments to establish radionuclide uptake from contaminated sediments are carried out. Different controlled parameters such as turbulent diffusion, passive diffusion and bioturbation has been applied. Concentration ratios for a number of biota/ sea water samples at coastal stations in Norway are now available as well as the distribution of Tc in organs of fish.

The redissolution of Pu in the Irish Sea is studied using a 3D model that includes a hydrodynamic submodel, a suspended matter submodel and a dispersion model. A model for simulating the movement of Pu and radiocaesium released from Sellafield is applied to the Irish Sea. A compartment model is applied to European coastal waters, the Arctic and the North Atlantic describing the dispersion of radionuclides in time and space. A numerical model has been developed to study the redissolution of ^{226}Ra in the Odiel and Tinto estuaries. The models have been developed and verified for describing the sediment-water interactions and improving parameters with regards to remobilisation of radionuclides. Results show that remobilisation of radionuclides from sediments can be significant for an increasing impact of radioactivity to environment and population.

3. Progress and Results

Work Package 1:

Objectives

A sediment sampling and analysis programme leading to improved radionuclide inventory estimates in areas of interest and an overview of the contamination and comparison with earlier studies.

Participants: RPII, CEFAS, NRPA, USEV, ENEA, RISØ, IRSN, UCD, AUN

Specifically:

1. To assemble and evaluate existing inventory data in the Irish Sea; the Skagerrak and Kattegat; the estuaries of the Odiel and Tinto Rivers; the Rhone Delta and the northern Adriatic Sea.
2. To carry out sampling campaigns in the areas where gaps have been identified with a view to improving the radionuclide inventory estimates in these marine areas.

Progress and Results

Further details of the progress and results in WP 1 are provided in Deliverable 1.1 (Sediment Inventory Database), and in a number of publications (Ledgerwood *et al.*, 2001; Kershaw *et al.*, in prep; McMahon *et al.*, in prep; López-Gutiérrez *et al.*, in prep., Santez *et al.*, in prep.).

The field studies of the REMOTRANS project were concentrated in five principal marine zones in European waters where contaminated sediments have been identified for further study of both inventory characterisation and remobilisation.

Irish Sea: Radioactive waste has been discharged into the Irish Sea as a consequence of reprocessing activities at Sellafield since 1952. In the intervening period over 600 TBq of Pu(α) and more than 40 000 TBq of ^{137}Cs have been discharged. As plutonium is very particle-reactive, much of it has been removed from the water column and been deposited in the fine-grained and muddy sediments off the west coast of Britain and, to a lesser extent, in the mud patch in the western Irish Sea. Figure 1 shows the sedimentology of the Irish Sea. The three boxed areas are the main sampling areas addressed in the REMOTRANS project.

The sediments of the western Irish Sea are, thus, estimated to contain in the order of 120 TBq of ^{137}Cs and 20 TBq of $^{239,240}\text{Pu}$. $^{238}\text{Pu}/^{239,240}\text{Pu}$ isotopic ratios were 0.14-0.18, even in surface sediments, which is typical of historical (as opposed to contemporary) Sellafield waste discharges. Summary data for the Western Irish Sea are shown in Figure 2 as an example of the data generated. Full details are given in Kershaw *et al.* (in prep.), McMahon *et al.* (in prep.) and Ledgerwood *et al.* (2001), as well as in Deliverable 1.1 (final version).

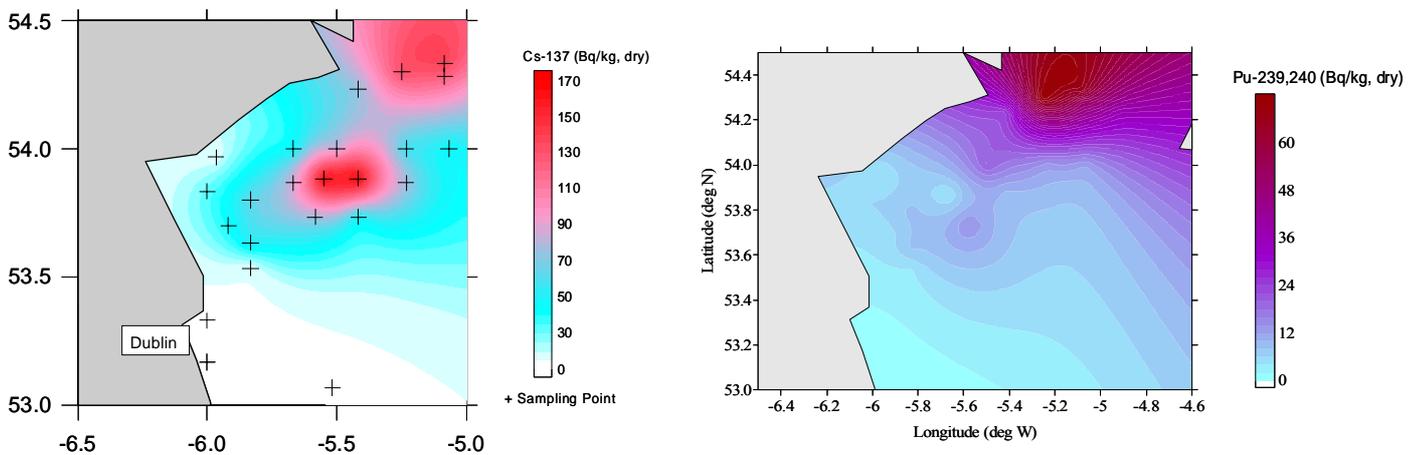
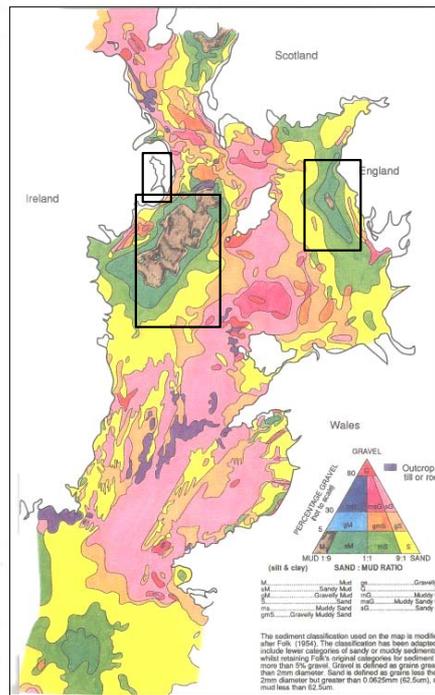


Figure 2. Distribution of ^{137}Cs and $^{239,240}\text{Pu}$ in Western Irish Sea Sediments

Skagerrak and Kattegat: The Baltic Sea was the marine zone most contaminated following the Chernobyl accident. The inventory in the Baltic Sea is estimated to be 5 PBq. This area is especially vulnerable to this contamination due to its lower salinity and associated increased concentration factors in biota. There is strong evidence to suggest that the contaminated sediments have already become a source of remobilised radionuclides.

A sampling campaign in the North Sea was carried out in November/December 2001 with the research vessel G.O. Sars of the Institute of Marine Research. Sediment cores, grab samples and surface samples were collected in the Skagerrak and Kattegat area, and a few also in other areas of the North Sea. A number of cores were assessed for low bioturbation based on ^{210}Pb data. This assessment was further supported by ^{234}Th data indicating no recent mixing and a clear Chernobyl- ^{137}Cs peak. The sedimentation rate was determined as $0.4 \text{ g cm}^{-2} \text{ yr}^{-1}$. Two

sediment cores were subsequently selected and collaboratively analysed for plutonium isotopes. $^{239,240}\text{Pu}$ concentrations appear to have remained constant in sediment layers deposited since the 1970's indicating a continuing source since that time. This indicates long-distance transport of remobilised plutonium. Plutonium ratios further point to European reprocessing as the source: $^{238}\text{Pu} / ^{239,240}\text{Pu}$ ratios increased gradually from global fallout levels (0.04) in the early 1970s to around 0.1 in the recent layers.

It has been demonstrated that the coastal deposits of fine-grained sediments at the mouths of Mediterranean rivers are efficient traps for contaminants transported from the drainage basins. These areas are characterised by high sediment deposition rates. Two Mediterranean river systems were studied under REMOTRANS.

Odiel/Tinto Rivers:

The estuary formed by these two rivers in the Gulf of Cadiz is shown in Figure 3. In order to obtain samples from the study areas, two sampling campaigns were carried out in 2001 and 2002 respectively. These sampling campaigns also provided samples for work carried out under WP2. About 20 sediment and water samples were collected at different points in the zone. Two artificial radionuclides have been studied in these sediments: ^{129}I and ^{36}Cl . The detected levels of ^{129}I and ^{36}Cl were typical of 'background zones' (i.e., those far removed from the main sources of these radionuclides). In the case of ^{36}Cl , very low levels were found.

This gave rise to difficulties in its analysis even using accelerator mass spectrometry (AMS). For ^{129}I , the measured levels at the Tinto and Odiel Rivers are comparable to others existing at other background zones, but, in this case, were easily detectable by AMS. The study of a



Figure 3. Sampling Locations in Odiel/Tinto Rivers

sediment core from the zone (Figure 4) showed that the increase of the ^{129}I concentration in the zone coincides with the time at which the atmospheric releases of ^{129}I at Sellafield and La

Hague first became significant. For this reason, these reprocessing plants are considered the main sources of the ^{129}I present in this zone. This set of measurements constitutes a very important contribution to the data library of ^{129}I environmental levels.

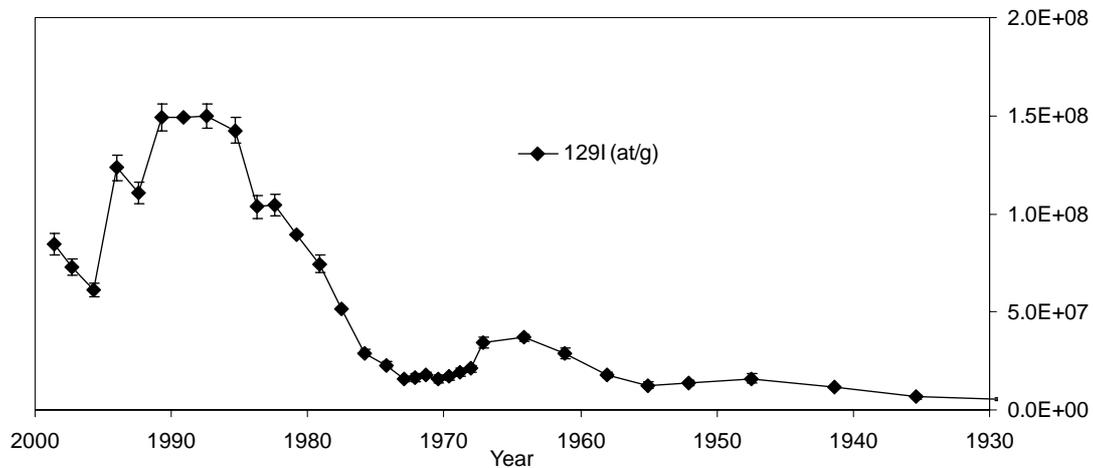


Figure 4. Depth profiles of ^{129}I in sediment core from Gulf of Cadiz.

Rhone Delta:

Data obtained in the 1990s were assembled and inventories calculated. Gamma-emitting radionuclides (including $^{210}\text{Pb}_{\text{ex}}$) and, in some samples, transuranics were analysed. Particular attention was paid to examining the consequences of the high river flow in 1994 on the remobilisation of sediments labelled by the radionuclides released at Marcoule. Two sampling campaigns were undertaken under WP1 in the Gulf of Lions (REMORA1 and REMORA2). REMORA1 was undertaken in March 2001, with 525 samples collected from 24 stations

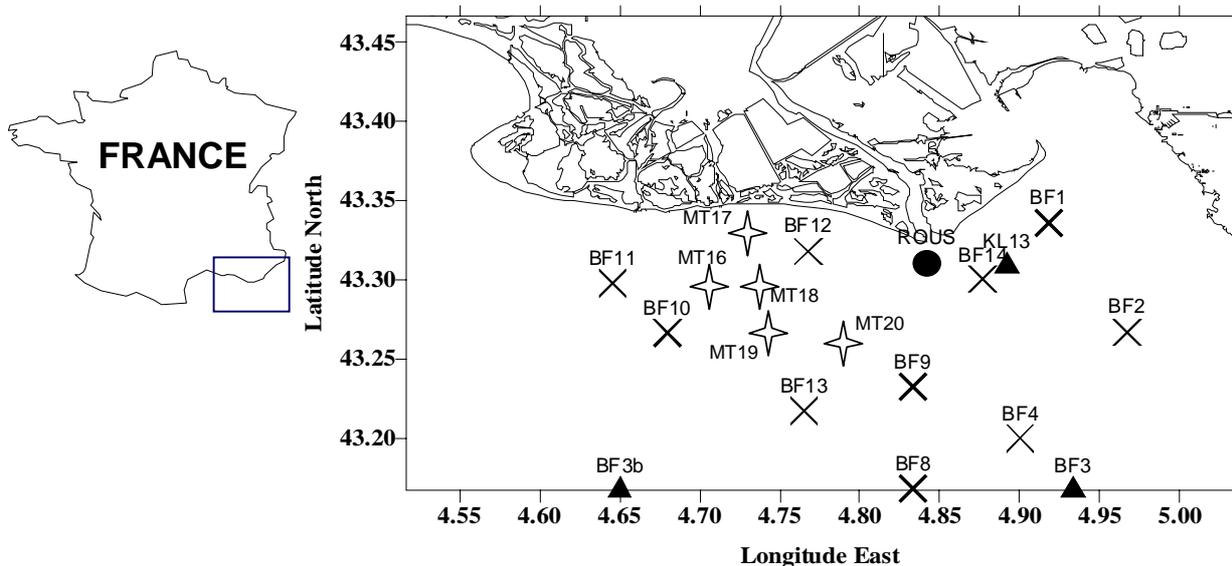


Figure 5. Sampling Locations in Rhone Delta

using a multi-tube corer, giving cores of 12-30 cm length. REMORA2 took place in November 2001 at a location very close to the river mouth. Despite very bad weather

conditions, 20 cores were successfully recovered using a Kullenberg corer, which yielded cores of 2-4.75 m length. Cs-137 inventories calculated for the stations shown in Figure 5 are presented in Table 1.

Table 1. Selected Inventory Data for Rhone Delta
(for location of sampling stations refer to Figure 1.6)

Cores	Depth	¹³⁷ Cs inventories (kBq m ⁻²)		^{239,240} Pu inventories (Bqm ⁻²)
		1990-1991	2001-2002	2001-2002
91KLRous/2001KLRous	30-40 m	>603	>185	57.1
90BF1/2001MT01	20 m	1.64	0.41	
90BF2/2001MT4	95 m	2.55	1.83	
91BF3/2001MT7	110 m	0.84	1.07	
91BF3b/2001MT12	80 m	1.91	2.37	
90BF4/2001MT6	104 m	2.69	2.86	
90BF8/2001MT8	102 m	3.79	1.78	
90BF9/2001MT09	95 m	9.17	>5.58	78.3
90BF10/2001MT14	56 m	7.71	>4.62	321.4
90BF11/2001MT15	30 m	3.96	0.93	62.2
90BF12/ 2002MT01Bis	23 m	8.85	>3.66	
90BF13/2001MT10	90 m	8.92	>6.28	85.2
2001MT17	26 m			135.5
2001MT19	75 m			65.9
2001MT20	85 m			65.7
2001MT16	41 m			79.4
2001MT18	41 m			62.3

Northern Adriatic Sea

The North Adriatic Sea was selected as one of the REMOTRANS study areas because, following the Chernobyl accident, this region received a significant direct atmospheric input of ¹³⁷Cs (estimated deposition onland: 5-15 kBq m⁻²) and a further contribution from some of the main Italian rivers (Po, Isonzo, Tagliamento, Adige, Piave), draining heavily contaminated areas. As a consequence, considerable amounts of Chernobyl radionuclides are accumulated in the sedimentary deposits near the river mouths. In 1987, the vertical profiles of natural and artificial radionuclides in sediments indicated that the sedimentary deposits near the mouths of the rivers Isonzo and Tagliamento were not stable and that a significant fraction of the radionuclides initially “stored” in these areas could be remobilised and distributed in a wider zone.

The historical data on the surface concentrations and vertical profiles of ¹³⁷Cs and ¹³⁴Cs in the Northern Adriatic area facing the mouths of the Isonzo and Tagliamento rivers are now stored in the ENEA-MERC Marine Information System:

www.santateresa.enea.it/www/siams/help2/mainpage/javaapplet_1.html.

The 1987 – 1989 data show that most Chernobyl radionuclides were associated to the fine sediments exported by the rivers and accumulated along shore, while the radionuclide levels were lowest off shore, where sediments are mainly constituted by residual sand. Inventories

were highest in the prodelta of the rivers (12 – 64 kBq m⁻²) and ranged between 2 and 6 kBq m⁻² in the shelf mud.

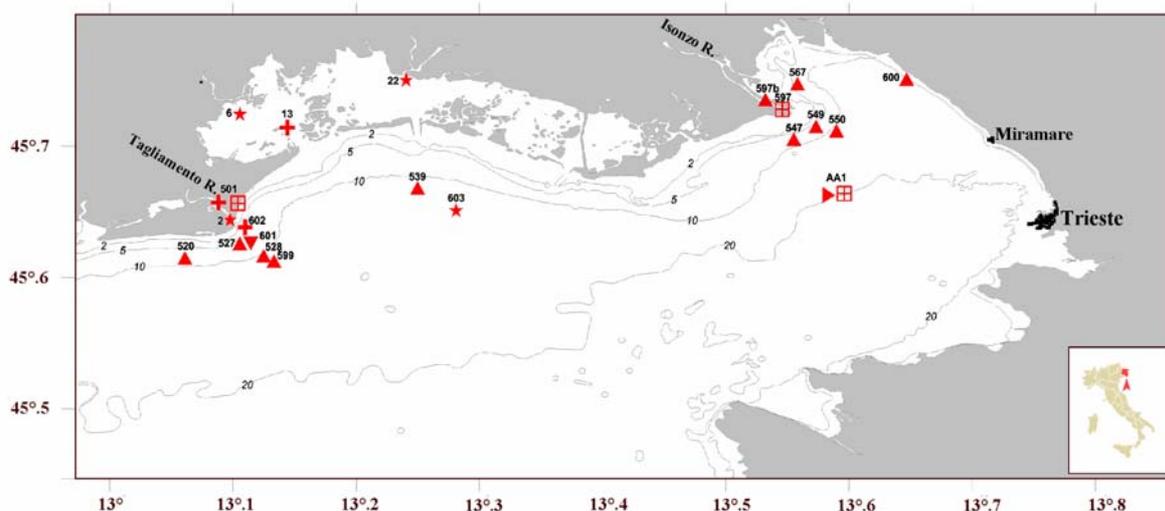


Figure 1.7. Sampling points in the Northern Adriatic Sea.

Table 2. Inventories of ¹³⁷Cs (kBq m⁻²) in sediment cores of the Northern Adriatic Sea, 1987 – 2001.

Station No.	June 1987	Nov. 1987	Nov. 1989	June 2001
Tagliamento Area				
527	12	15	30	5
528	2	-	-	7
599	4	-	-	6
Isonzo area				
547		> 64	> 21	> 17
549	> 4			> 13
550	> 6			7
567	6			6

New core samples (length 80 cm) were collected in the same area in 2001, to estimate the present inventories. Fig. 1.7 and Table 2 show sampling sites and temporal trend of the inventories of ¹³⁷Cs in the area.

The analysis of the data shows that in the protected gulf of Trieste (St. 550, 567) the inventories of ¹³⁷Cs (6 – 7 kBq m⁻²) remained constant in the period 1987 - 2001, as the new input compensated for the radioactive decay. Close to the Isonzo river mouth (St. 547, 549), the cores were too short to estimate the present inventories, but ¹³⁷Cs concentrations were five times lower than in 1987, yielding to a lower inventory in the upper 60 cm. Near the Tagliamento river mouth (Stns 527, 528, 599) present inventories range between 5 and 7 kBq m⁻². Cs-137 is detectable only in the upper 20 cm of sediment and present concentrations are one order of magnitude lower than in 1987.

In addition to the sampling campaigns for inventory estimation purposes, a number of sampling campaigns were also undertaken in support of other work-packages, e.g., DIAPLU-Irish Sea (2002), SIRRO -a joint Russian-German-Norwegian expedition to the Ob and Yenisey Rivers and the adjacent Kara Sea (2001) and a collaborative campaign in Rørholt Fjord in 2002.

Work Package 2:

Objectives

Quantification of long distant transport of remobilised

Participants: ULUND, IRSN, RPII, NRPA, RISOE, IFE, CEFAS, ENEA, USEV

Norwegian Coast

The brown alga *Fucus vesiculosus* has been collected annually by Institute for Energy Technology at more than ten sampling locations since 1980, mainly in August and September (Christensen, 1986; Christensen and Selnaes, 1995). At Utsira in Western Norway (59°17.9'N 04°54.1'E) monthly samples have been collected since January 1986. At the northernmost location Ingøy (71°05.2'N 24°03.3'E), close to the North Cape, monthly samples have been collected since August 1998.

In the context of Remotrans, historical samples from Utsira (1980-2001) were analysed for plutonium isotopes in order to trace any remobilisation from the Irish Sea. More recent samples (1995-2003) were analysed for gamma emitters, ^{99}Tc and iodine isotopes to continue the time series study and let relevant results be input to model work of Remotrans partners.

All *Fucus* samples were dried at 105 °C, homogenised and analysed for gamma emitters using high purity germanium gamma spectrometry and computerised spectrum analysis. Portions of the dried and homogenised *Fucus* samples were analysed for ^{99}Tc at the University of Lund, Sweden and for $^{127}\text{I}/^{129}\text{I}$ at Centre de Spectrométrie Nucléaire et de Spectrométrie de Masse, Orsay, France. The results for ^{99}Tc and $^{239,240}\text{Pu}$ in *Fucus vesiculosus* from Utsira are shown in Figs. 1 and 2.

The ^{99}Tc data set from Utsira have been used as input parameters to test both the CEFAS and the ARCTICMAR 2 (NRPA-box) models. The results from these tests are described elsewhere by these partners.

It has previously been shown (Christensen and Strålberg, 2000) that most of the ^{137}Cs along the Norwegian coast now originates from the Baltic Sea. A transit time for ^{137}Cs from Sellafield to Utsira has been estimated to 3-4 year and from the Baltic to Utsira to 2 years (Christensen and Strålberg, 2000).

The ^{99}Tc along the Norwegian coast now solely originates from Sellafield. The transit time from Sellafield to Ingøy has earlier been estimated to 4-5 years (Strålberg *et al.*, 2002). The transit time from Sellafield to Utsira, based on our newest data, has now been calculated to be 3.5-4 years. However, the discharge front was detected at Utsira already at the end of 1996, which is only 2-2.5 years from the first major new discharges in 1994 (March-April and September-October). The transfer factor for ^{99}Tc from Sellafield to Utsira, based on the above mentioned transit time of 3.5-4 years, is calculated to be 2.9 ± 0.9 (Bq/kg dw)(TBq/yr $^{-1}$) for *Fucus*. If we use a concentration factor of 10^{-5} , this TF corresponds to 29 ± 9 (Bq/m 3)(PBq/yr $^{-1}$) $^{-1}$ for seawater.

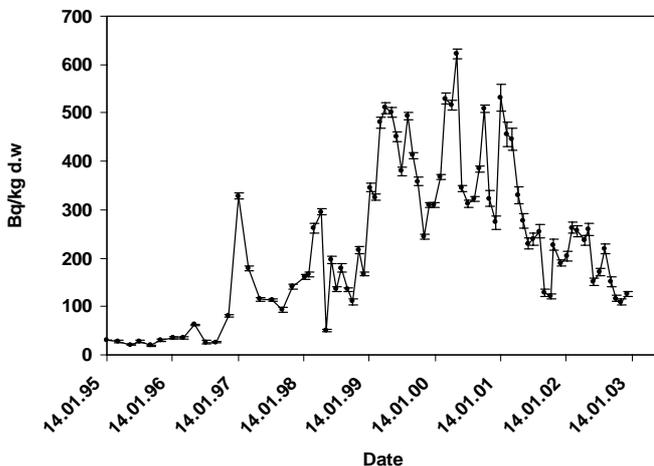


Figure 1. ^{99}Tc in *Fucus* collected at Utsira

$^{239,240}\text{Pu}$ in *Fucus vesiculosus* from Utsira shows a significant downward trend although the concentrations seem to vary significantly from year to year (Fig. 2). The reason for this variation is not understood. Obtained $^{238}\text{Pu}/^{239,240}\text{Pu}$ ratios, although infested with large uncertainties because of the low ^{238}Pu concentrations, show that the ratio is significantly higher than the ratio for global fallout (~ 0.03), with a decay-corrected average of 0.13, indicating a contribution of

remobilised Sellafield plutonium. The $^{238}\text{Pu}/^{239,240}\text{Pu}$ ratio in the Sellafield discharge was 0.35 in 1992. However, the ratios in the sediment and seawater have been measured to 0.20-0.27 and 0.19-0.24 (northern Irish Sea), respectively. This indicates that the dissolved plutonium originates from both the discharges and remobilisation processes. Our data also show that the remobilised plutonium can be transported over longer distances.

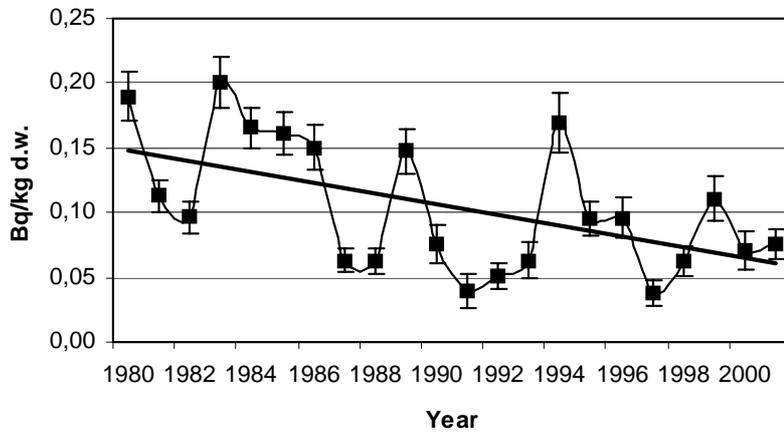


Figure 2. $^{239,240}\text{Pu}$ in selected samples (August/September) of *Fucus vesiculosus* collected at Utsira

NRPA collected samples of sea water and *Fucus vesiculosus* have been collected at Hillesøy coastal station in northern Norway each month. The highest annual average seawater concentrations of Tc-99 were observed in 1999 and 2000. In 2001, and 2002, average concentrations seem to decrease slightly. The highest average annual average concentrations of Tc-99 in seaweed were observed in 2000 and 2001. (Fig. 3 and 4) Elevated levels of Tc-99 discharge from Sellafield commenced in March 1994. The annual discharges peaked in 1995, and remained high in 1996. From 1997, annual discharges have been reduced by a factor ranging from ~2 to ~4 compared to discharges in 1995 and 1996. A transport time of 42 months (3.5 years) was estimated. An average transfer factor of $8,4 \text{ Bq m}^{-3}$ per PBq a^{-1} discharged Tc-99 was obtained for the 5.5 years of observation at Hillesøy (July 1997 to December 2002). The “transfer factor” for *Fucus vesiculosus* was estimated to 1810 Bq kg^{-1} per PBq a^{-1} . These estimates assumed a “background” (pre-EARP) concentration of Tc-99 at Hillesøy of 0.3 Bq m^{-3} in sea water and 30 Bq kg^{-1} (dw) in fucus.

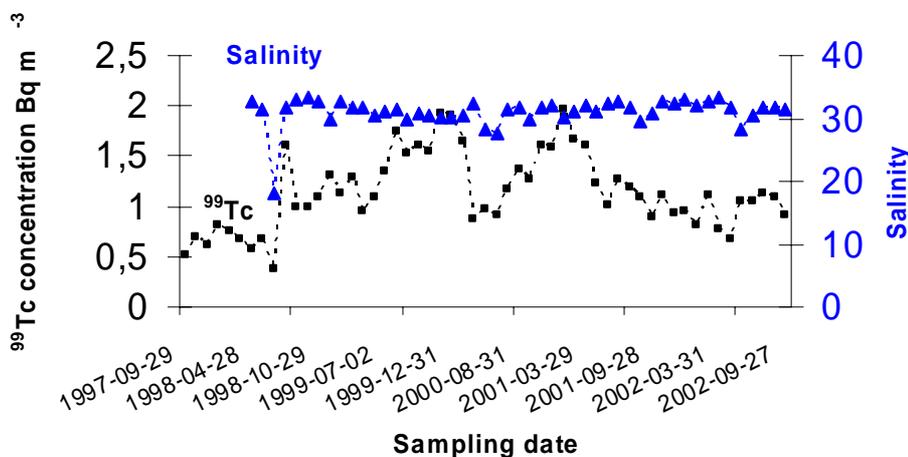


Fig. 3 Concentration of Tc-99 in sea water at Hillesøy

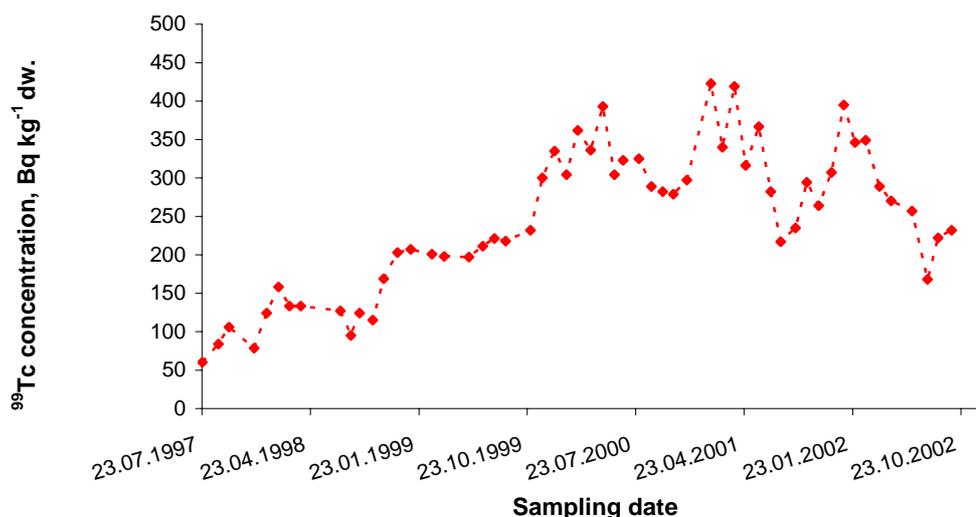


Fig. 4 Concentration of Tc-99 in *F. vesiculosus* (Bq/kg dry weight), Hillesøy

In the Fram Strait, special focus was set on the radionuclide levels in the main ocean current systems, the West Spitsbergen Current and the East Greenland Current. Levels of caesium, technetium and plutonium isotopes were investigated in these for the Arctic crucial currents. Levels of ⁹⁹Tc were observed in 2000 and 2001 to be significantly higher than earlier investigations showed, (Table 1.) indicating that traces of the enhanced discharges from the Sellafield nuclear reprocessing plant that begun in 1994 reached the West Spitsbergen Current. Deep water samples from the Fram Strait (~1000m) had very low concentrations of Tc-99.

Table 1. Mean annual concentrations of ⁹⁹Tc in seawater and seaweed (*Fucus vesiculosus*). The errors represent standard deviations of the mean values.

Year	Sea water		Seaweed	
	Number of samples	Mean activity concentration (Bq/m ³)	Number of samples	Mean activity concentration (Bq/kg d.w.)
1997*	6	0.61 ± 0.13	4	82 ± 16
1998	11	0.93 ± 0.36	10	138 ± 31
1999	10	1.46 ± 0.30	10	232 ± 47
2000	9	1.42 ± 0.34	11	318 ± 36
2001	12	1.25 ± 0.33	12	321 ± 68
2002	12	0.98 ± 0.15	11	264 ± 52
Mean Transfer factor		8,4 Bq m⁻³ per PBq a⁻¹		1810 Bq kg⁻¹ per PBq a⁻¹

* Sampling commenced July 1997

Monitoring of radioactivity in seawater and sediments are carried out at regional cruises in the open seas around Norway. Data on the translocation of Tc-99 and other radionuclides are available from a coastal cruise in November 2000 from Kirkenes to Ålesund, from the North Sea and Skagerrak/Kattegat in 2001 and from the Barents Sea in 2002. It is difficult to carry out calculations of transfer factors on the basis of these data, however, it provides important input to modelling of the dispersion of Sellafield discharges.

Changes in Tc-99 activity in *Laminaria* from the Norwegian coast.

Due to the increased discharges of ⁹⁹Tc from Sellafield in 1994, analysis of ⁹⁹Tc in the brown algae *Laminaria hyperborea* (Fig. 1) collected at 2 sites along the Norwegian coast (Fig. 2) has been performed since 1996. *Laminaria hyperborea* is large brown algae, which grows on rocky substrata up to 30 m below sea level. It consists of a stiff, tapering stipe up to 2 m long, and a broad, deeply divided blade. It can live for up to 15 years. The peak growing season occurs in winter, and the old blade is shed and replaced with the newly grown one in spring.

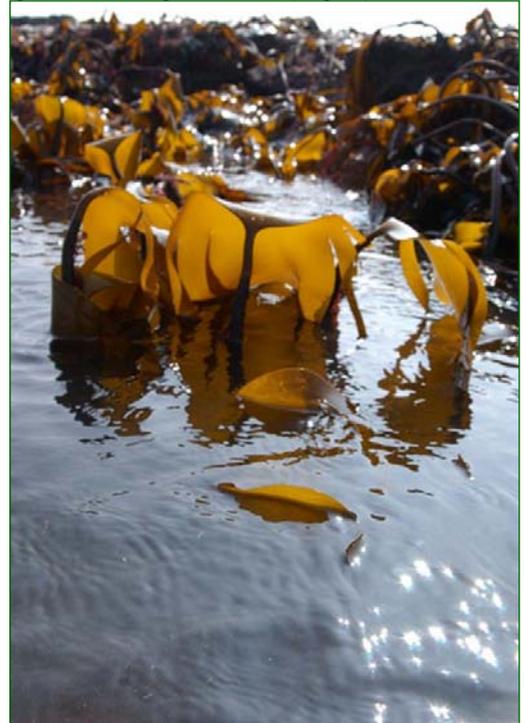
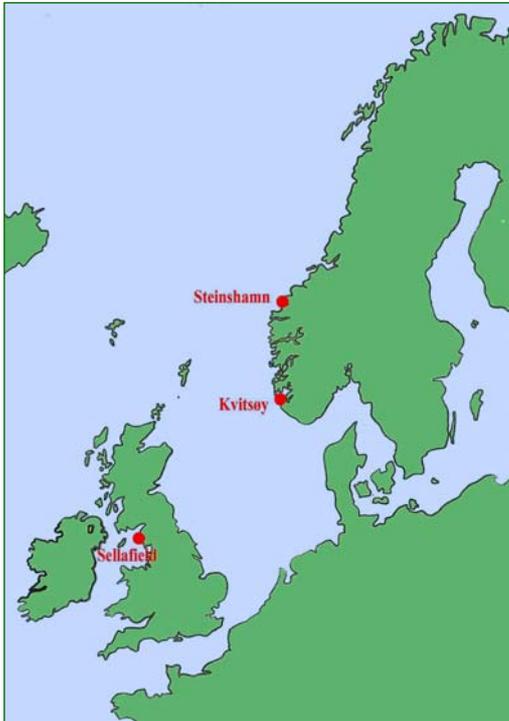


Figure 5: *Laminaria hyperborea*

Figure 6: Algae sampling sites on the Norwegian coastline. The algae are contaminated with ⁹⁹Tc from the effluents from

BNFL Sellafield nuclear facility in Cumbria, UK.

The presence of ⁹⁹Tc in *Laminaria hyperborea* is also of commercial interest because the algae is routinely harvested from the Norwegian coastline for alginate production. Samples of *Laminaria hyperborea* were collected each month from sites along the Norwegian coastline (Fig.3) and the blades and stipes were separated, dried, grounded and homogenized before analysis. ⁹⁹Tc was extracted using microwave acid digestion followed by separation on a TEVA ion-exchange column. ^{99m}Tc was used as a yield monitor. The concentration of ⁹⁹Tc in the extracts was determined using ICP-MS (Skipperud et al. in prep).

Table 2 Sampling site and information

Area	Medium	Nuclides	Time	Number	Modelling	Contact
Norwegian coast					NRPA box model	Lindis Skipperud
Kvitsoy	Seaweed	⁹⁹ Tc	1996-2003	2x12/yr	(M. Iosjpe, NRPA)	and Brit Salbu,
Steinshamn	Seaweed	⁹⁹ Tc	1996-2003	2x12/yr		AUN

The concentration of ^{99}Tc is higher in the stipes than in the blades. There are apparent seasonal fluctuations, with activity higher in winter/spring and lower in summer/autumn. Factors involved may include plant growth patterns, seasonal changes in ocean circulation and short-term variations in actual ^{99}Tc discharges from Sellafield.

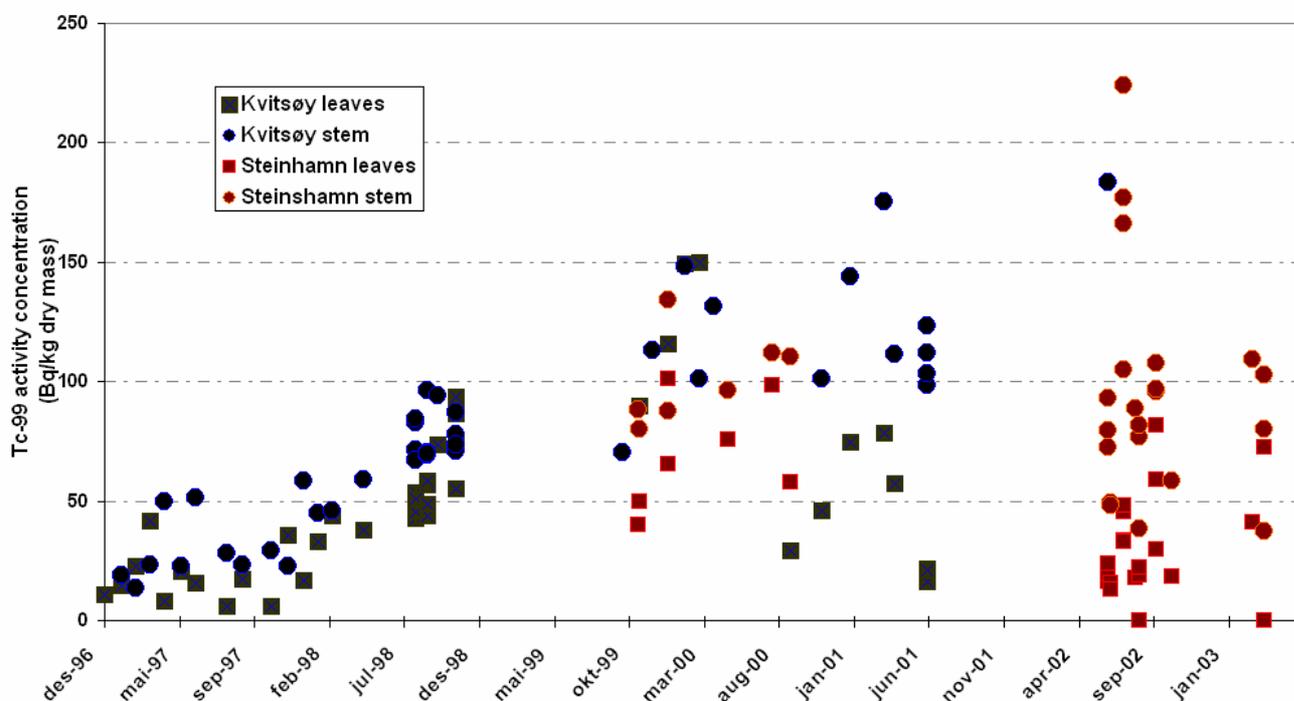


Figure 7: Activity of ^{99}Tc in the stipes and blades of *Laminaria hyperborea* collected from Kvitsøy and Steinshamn since 1997.

Results from Kvitsøy (Fig. 7) indicate an increase in ^{99}Tc activity in both stipes and blades between 1997 and late 2000. ^{99}Tc activity in the blades, which are replaced annually, appeared to peak in 2000, but activity in the stipes, which last the entire life of the plant and constitute approximately 60 % of the algal biomass, remained at elevated levels until July 2002. Results from Steinshamn, further down-stream from the discharge, show a similar pattern, with a continuing increase in stipe ^{99}Tc activity up to late 2002.

The time trend for activity concentration of ^{99}Tc in the blades suggests that the discharge plume from Sellafield reached these sites in 1996, and the accumulation continued in blades until 2000. For the stipes, however, the ^{99}Tc levels continues to accumulate (Mobbs and Salbu 2002) until late 2000. Measurements of ^{99}Tc in *Laminaria hyperborea* will be continued.

Temporal variations in uptake of ^{99}Tc and Stable Analogues (Re, Mo, W) in Seaweed

Figure 8 and 9 show ICP-MS measurements of ^{99}Tc and stable analogues (Re, Mo, W) in a series of seaweed collected from Northern Norway. Analysis was carried out on 0.5 g microwave digest of the seaweed. The results show that Mo, Tc and partially Re follow the same pattern.

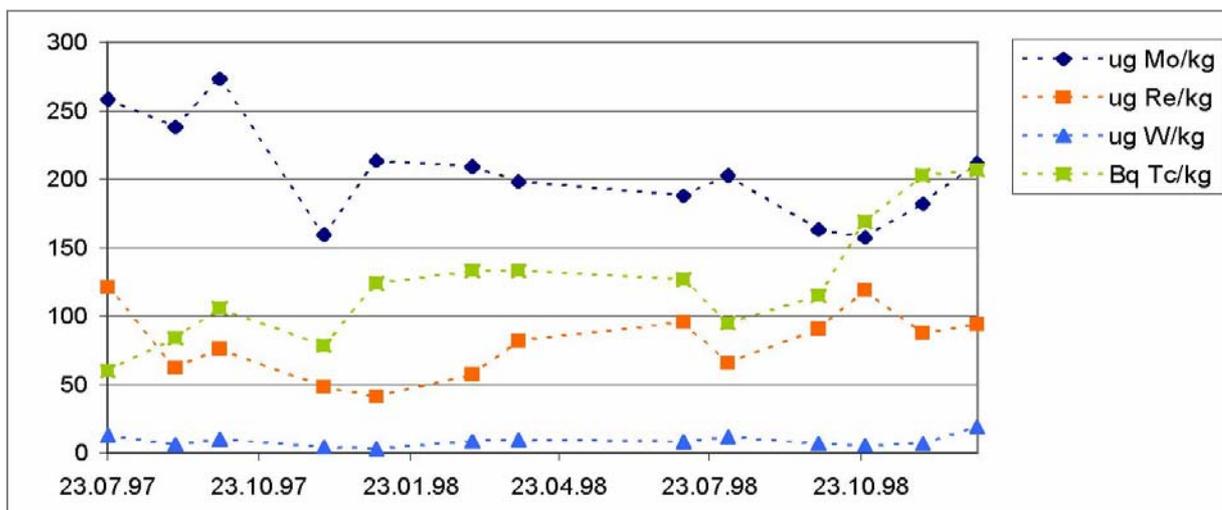


Figure 8. ICP-MS determination of Tc, Re, W and Mo in a series of seaweed collected from Northern Norway

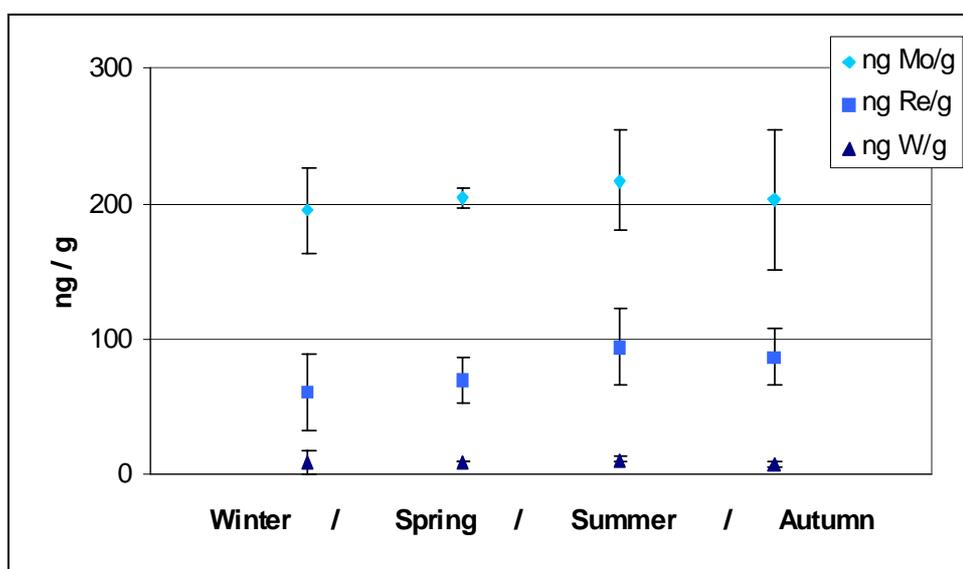


Figure 9: Mean activity concentration of Re, W and Mo in a series of seaweed collected during different seasons from Northern Norway

Rhone mouth, Mediterranean

The Rhône River is one of the major rivers flowing to the Mediterranean Sea. This area has been selected as a study area for the REMOTRANS project because i) it has been demonstrated that the coastal deposits of fine-grained sediments (i.e. prodeltas) at the mouths of Mediterranean rivers are efficient traps for contaminants transported from the catchment basins, ii) several nuclear installations are authorized to discharge liquid effluents into the Rhone river, main releases arising from the reprocessing plant in Marcoule.

Table 3 : Available data off The Rhône mouth for the sediment compartment

Area	Medium	Nuclides	Time	Number
Mediterranean Sea (Rhône mouth)	Sediment (box-corers or multicorers)	Gamma emitters	1990-1991 2001-2002	12 cores sampled at the same locations at both periods (see map)
Mediterranean Sea (Rhône mouth)	Sediment (box-corers or multicorers)	Gamma emitters	1984 1990 1994 1997 2001	5 cores at the same location very close to the Rhône mouth (ROUS)
Mediterranean Sea (Rhône mouth)	Sediment (piston corers)	Gamma emitters Pu	1991 1996 2001	3 long cores at ROUS

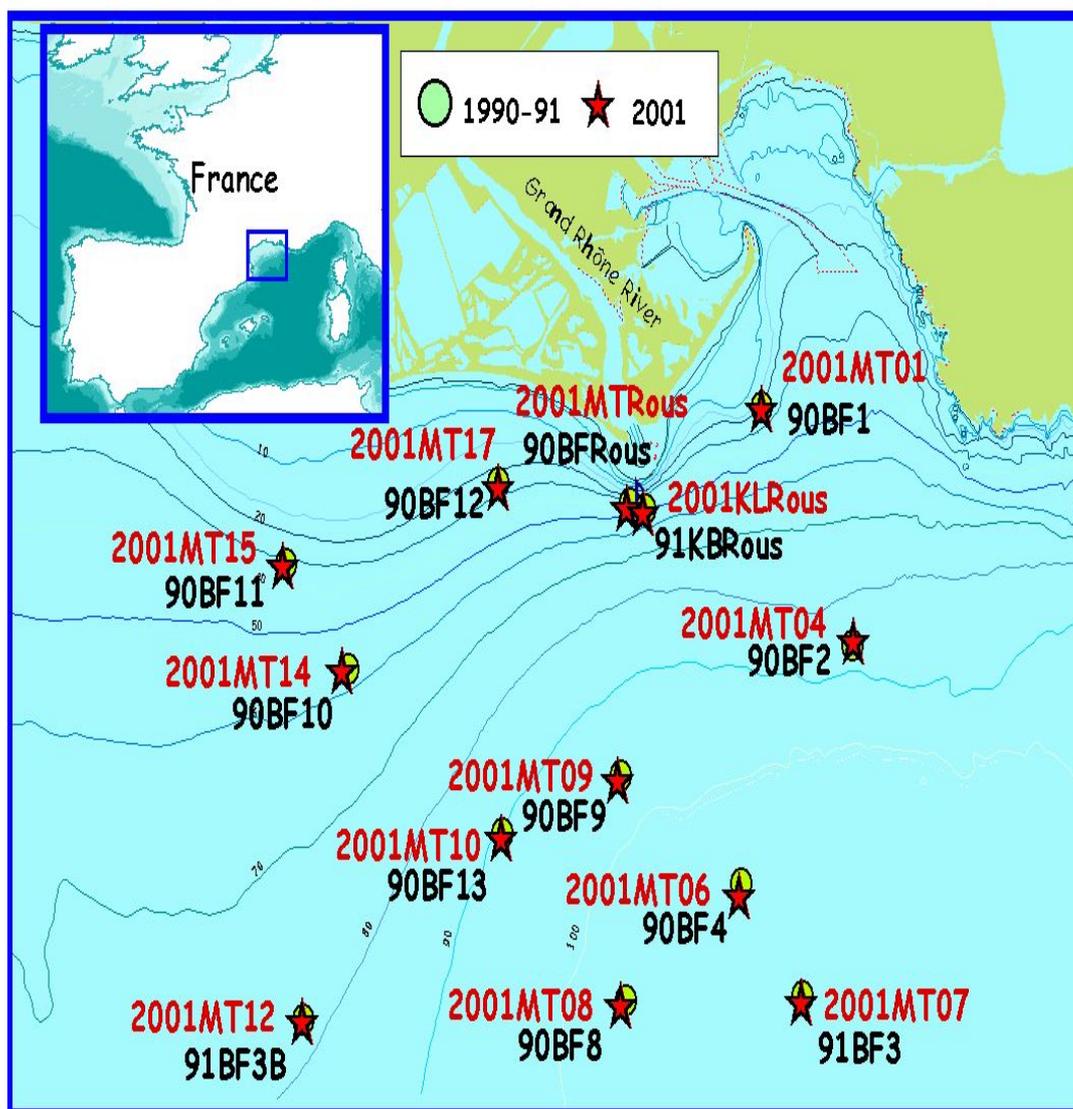


Fig. 10. Vertical profile sites close to the Rhône mouth.

In the Rhône prodelta, sediments were sampled at various periods with box-corers or multicorers (table 1) with a maximum penetration depth of 45cm. But the very high sedimentation rates in this area (up to 50 cm y-1), prevented us from superposing the different vertical

profiles in order to obtain almost continuous time series record. However, the various vertical profiles obtained over time (1984, 1990, 1994, 1997, 2001) close to the Rhône mouth (figure 10) clearly underline the decrease in the Marcoule discharges with two main events : in 1991 when the new treatment station for liquid effluents was brought into use and in 1997 when the reprocessing operations were stopped. In 2001, ^{137}Cs is still detected through gamma spectrometry on the whole core but the levels are lower than 15 Bq kg^{-1} dry weight and ^{60}Co is episodically found in the upper part of the core only.

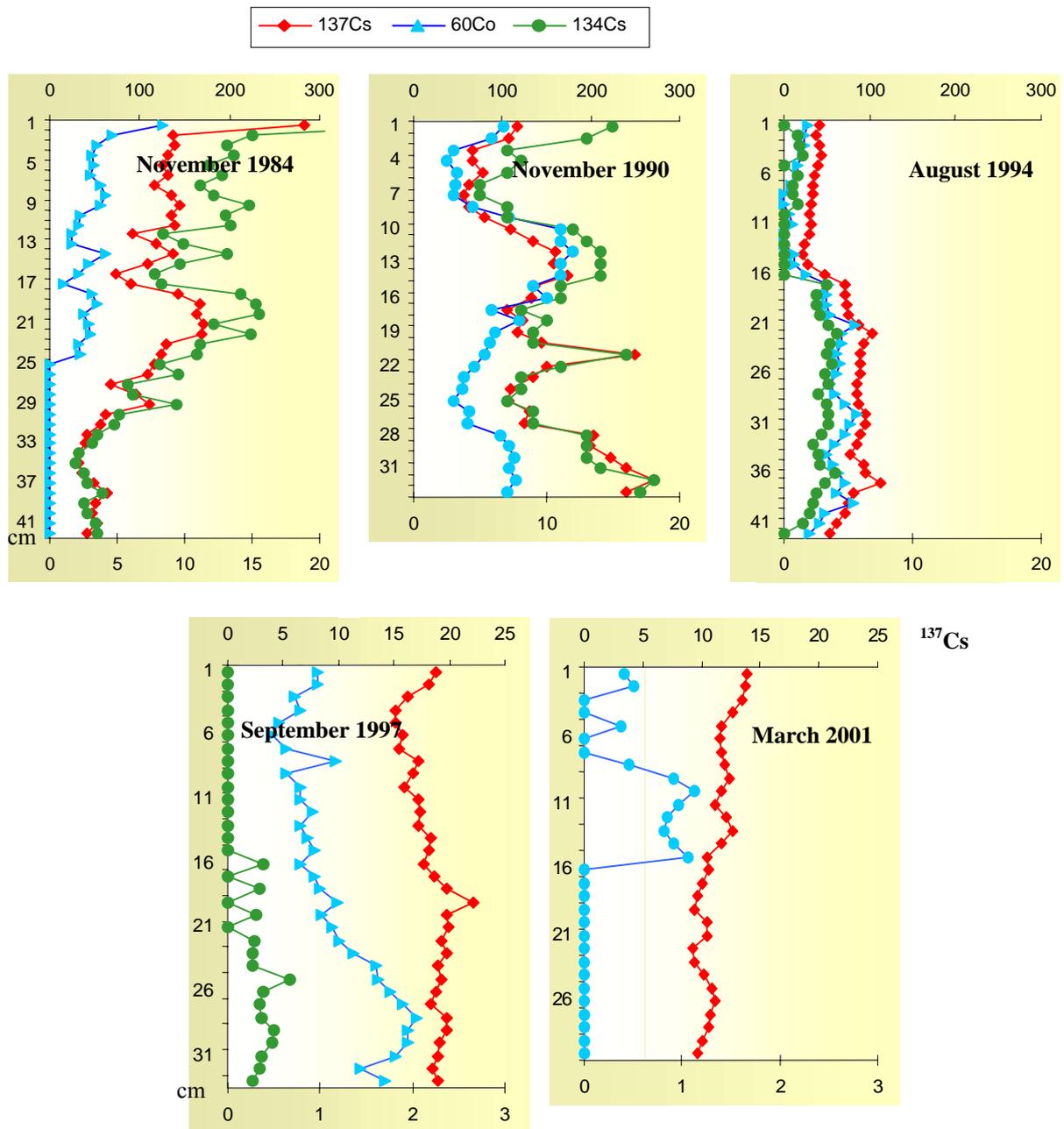


Figure 11 : Vertical profiles of ^{137}Cs , ^{134}Cs and ^{60}Co (Bq kg^{-1} dry weight) in cores sampled at the Roustan station close to the Grand Rhône mouth in 1984, 1990, 1994, 1997 and 2001. (Vertical scale in cm, notice the change of horizontal scale from 1997 onwards)

Even for long cores (several meters) sampled with piston corer close to the mouth (Table 3, figure 2), superposition of the profiles is difficult due to strong disturbances in sediment record in this shallow area especially during meteo-climatic events (see WP3). Indeed,

$^{137}\text{Cs}/^{134}\text{Cs}$ ratio allowed us to determine that during the major flooding which occurred in 1994 more than the upper 40cm of the sediment record in this area were flushed away.

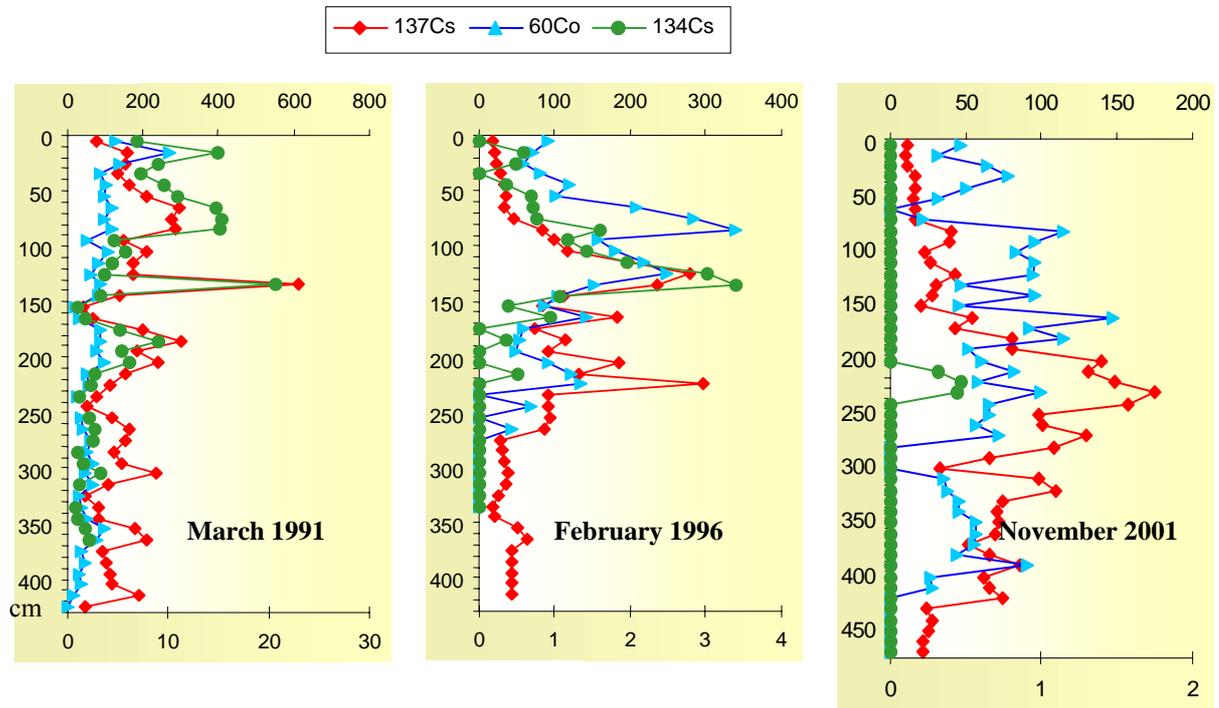


Figure 11: Vertical profiles of ^{137}Cs , ^{134}Cs and ^{60}Co (Bq kg⁻¹ dry weight) in long cores (>400cm) sampled at the Roustan station close to the Grand Rhône mouth in 1991, 1996 and 2001

Regarding inventories, data in table 4 clearly underline the decrease with time in ^{137}Cs inventories. Indeed, a decrease by a factor of 2 is observed over a 10 years period at the Roustan station close to the Rhone mouth for long cores (430cm and 470cm) which is far more than the decrease due to the decay. $^{239+240}\text{Pu}$ inventories within these cores appear quite constant i.e. 1991 (>10,2 kBq m⁻²) and 2001 (>10,0 kBq m⁻²) underlying the stronger binding of Pu isotopes on particles compared to ^{137}Cs .

Table 4: ^{137}Cs inventories in kBq m⁻²

Cores	Depth	1990-1991	1996	2001-2002
91/96/2001KLRous	30-40m	>603	>316	>292
90BF1/2001MT01	20 m	1.64		0.41
90BF2/2001MT4	95 m	2.55		1.83
91BF3/2001MT7	110 m	0.84		1.07
91BF3b/2001MT12	80 m	1.91		2.37
90BF4/2001MT6	104 m	2.69		2.86
90BF8/2001MT8	102 m	3.79		1.78
90BF9/2001MT09	95 m	9.17		>5.58
90BF10/2001MT14	56 m	7.71		>4.62
90BF11/2001MT15	30 m	3.96		0.93
90BF12/ 2002MT01Bis	23 m	8.85		>3.66
90BF13/2001MT10	90 m	8.92		>6.28

➤ ^{137}Cs is still detected in the bottom of the core, the inventory is thus incomplete.

In summary, it can be underlined that the Rhone prodeltaic area is characterized by high apparent accumulation rates but the stability of the deposits seems to be low. This shallow depth area (20-50m) is subject to resuspension phenomena during intense meteorological events and can thus be considered as temporary sinks/storages for particle-bound contaminants.

Odiel and Tinto estuary

The task of the University of Seville in WP2 was the measurement of natural radionuclides in the Odiel and Tinto estuary, in order to obtain information about the possible remobilization of radionuclides deposited in the sediments of these rivers in former years.

The especial characteristic of this system is that was being continuously contaminated by the releases of phosphate fertiliser plants that stopped direct discharges in 1998. These plants used to release part of their residues directly to the Odiel River (a map of the zone is shown in WP1 report). Apart from this, solid wet phosphogypsum rests were deposited near the Tinto River in piles. These piles were leached by rain and other weather factors and the radionuclides passed to the River water and to the sediments. In the last years, these piles have been substituted by a new especially protected pile that prevents the river contamination to a high extent.

Radionuclides as ^{226}Ra , ^{238}U , ^{210}Po and ^{210}Pb have been analyzed both in water and sediments in most cases and the results are listed in the tables included in the deliverable.

The analyses have been carried out using different techniques: alpha spectrometry, liquid scintillation counting and proportional gas counting.

Table 5 (see deliverables) shows the activities measured for ^{226}Ra in river water. A similar behaviour can be detected in the three zones studied. In concrete, activities seem to decrease clearly between 1999 and 2002. This effect could be expected after the stop of the discharges to the rivers in 1998 and the installation of the protected new pile in that year. Possibly, the successive tides tend to dilute the river waters producing a reduction in the activity.

However, this reduction is not as fast as it would be expected if the presence of radionuclides was only in the river water. Most of the analyzed locations present also a reduction in the ^{226}Ra activity in sediments between 1999 and 2001. This fact can be more clearly observed when the recent results are compared to the ones obtained by the University of Seville in 1993 and 1996, much higher (figure 12). This effect suggests that the radionuclides accumulated for years in the sediment floor are acting as source to the rivers water. The radionuclides are then slowly remobilizing from the sediment to the water. Then, the whole system seems to be experiencing a washing process after the stop of the discharges.

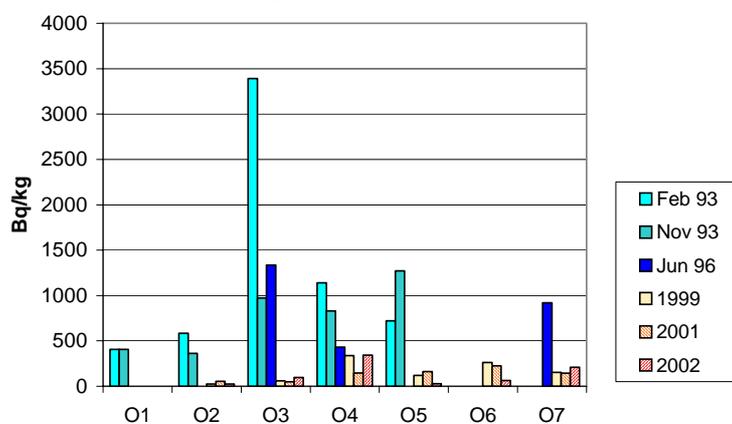


Figure 12. ^{226}Ra in Odiel river sediments.

The same conclusions refer to uranium isotopes in river water and sediments. As before, a global decrease tendency exists in the activities in water and sediments. Although the effect is not completely clear from 1999 to 2001, it is really evident when these results are compared to the ones measured in 1993 and 1996, much higher. Then, a self-cleaning of the estuary can be deducted also for U isotopes. The differences between U and Ra could be due to their chemical properties and also to the influence of the old phosphogypsum piles still present at the Tinto River side.

Figure 13 shows the ratio $^{234}\text{U}/^{238}\text{U}$ as a function of the ^{238}U activity in water and sediments at the studied zone. It can be appreciated that high ratios (signal of natural uranium) correspond to low ^{238}U activities. These are the samples taken at the locations less affected by the factories discharges. The most contaminated samples present $^{234}\text{U}/^{238}\text{U}$ ratios close to the unity, lower than the natural equilibrium ratio.

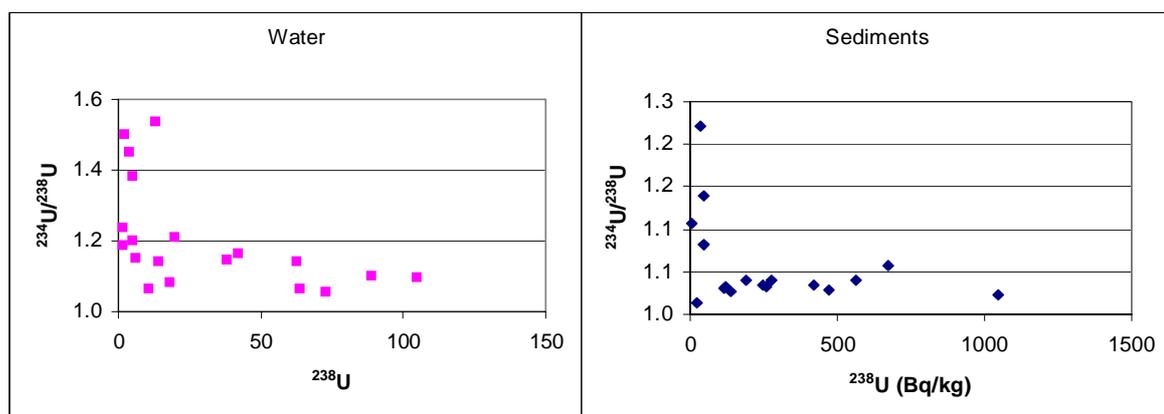


Figure 13. $^{234}\text{U}/^{238}\text{U}$ as a function of the ^{238}U activity in water and sediments.

An important event that affected the presence of radionuclides at the zone was the pile accident that took place in 1999 at the Tinto River side. This strongly affected the self-cleaning process of the estuary increasing the radionuclides activities in the water (figure 14). However, the measurements carried out for samples collected after 1999 show that these activities have clearly decreased in the years following the accident.

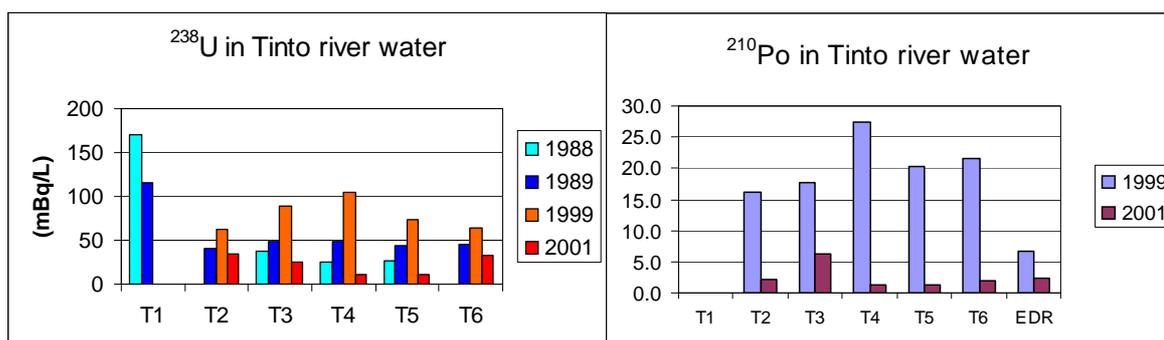


Figure 14. Effect of the piles accident in 1999.

Skagerrak and Kattegatt

The activity concentration of ^{99}Tc and the activity ratio $^{99}\text{Tc}/^{137}\text{Cs}$ in brown seaweed (*Fucus vesiculosus* and *Fucus serratus*) and seawater from the Swedish west coast in 2001 were determined. The ^{99}Tc activity concentrations vary from approximately 11 Bq kg^{-1} to 135 Bq kg^{-1} from the south to the north, respectively. The ^{99}Tc activity concentrations data in seaweed appears to be depending on latitude and salinity. The results from the one site differs from the

rest due to the mixing of seawater with low salinity freshwater from a nearby river. The ^{99}Tc concentrations in seawater are relatively constant along the coast with an average concentration of $1.05 \pm 0.17 \text{ Bq m}^{-3}$. This corresponds to a dilution factor of approximately 3000 for the Sellafield discharge to the Swedish coastal waters. It has been estimated previously by others that approximately 2% of the Sellafield and 10% of the La Hague ^{99}Tc discharge is transported by the sea current in the North Sea to the coastal waters of Sweden. The measured $^{99}\text{Tc}/^{137}\text{Cs}$ activity ratio in *Fucus vesiculosus* and seawater vary between 0.4 ± 0.1 to 35.1 ± 2.5 and 0.018 ± 0.002 to 0.085 ± 0.021 , respectively. The ratio is strongly affected by the outflow of ^{137}Cs contaminated water from the Baltic Sea. For comparison, the reported $^{99}\text{Tc}/^{137}\text{Cs}$ value from the Sellafield EARP pulses in 1995-1996 was approximately 15.

The uptake of Tc in *Fucus vesiculosus* appears to have a strong dependence on the salinity concentration in seawater with higher uptake for higher salinity. However, the opposite relation is observed for Cs. The concentration factors for Tc and Cs in *Fucus vesiculosus* to coastal seawater at the west coast are presented in Figures 3 and 4 with salinity concentrations for comparison. The CF for Tc and Cs varies between $0.31 \pm 0.03 \times 10^5$ to $1.32 \pm 0.19 \times 10^5 \text{ dm}^3 \text{ kg}^{-1}$ and 136 ± 16 to $549 \pm 63 \text{ dm}^3 \text{ kg}^{-1}$, respectively, with an average of $0.79 \pm 0.30 \times 10^5 \text{ dm}^3 \text{ kg}^{-1}$ and $308 \pm 112 \text{ dm}^3 \text{ kg}^{-1}$, respectively.

The uptake of ^{99}Tc differs between the two species of seaweed, *Fucus vesiculosus* and *Fucus serratus*, sampled at the same time and location. A comparison between the two species showed an average ratio of 0.61 ± 0.16 , which is in good agreement with earlier studies in the same environment. For ^{137}Cs there is no significant differences in uptake between the two species with an average ratio of 1.08 ± 0.23 .

The ^{99}Tc activity has altered during the years influenced by the Sellafield and La Hague reprocessing plants. Since the uptake in seaweed of Cs is competing with the potassium content in seawater, a better understanding of the coastal gradient is obtained with the $^{99}\text{Tc}/(^{137}\text{Cs}/^{40}\text{K})$ ratio (Fig. 6) instead of the $^{99}\text{Tc}/^{137}\text{Cs}$ activity ratio. The average ^{40}K activity concentration for 1991, 1995 and 2001 were $808 \pm 125 \text{ Bq kg}^{-1}$, $1123 \pm 140 \text{ Bq kg}^{-1}$ and $783 \pm 136 \text{ Bq kg}^{-1}$, respectively. The gradients along the coast are more pronounced for the 1991 and 2001 samples than for the 1995 sample. The 2001 samples is strongly affected by the EARP pulses in 1995-1996 and 1991 samples are presumed being influenced by the La Hague discharge a few years earlier.

The ^{99}Tc activity concentrations in *Fucus serratus* at the Särödal site, collected 1967-2000, with the annual ^{99}Tc discharges from Sellafield and La Hague are presented in Figure 15. The early Sellafield data until 1977 were incomplete and were estimated by environmental measurements of brown seaweed, using data from Särödal as well as from sampling spots from Arctic waters, in the period 1963-1982.

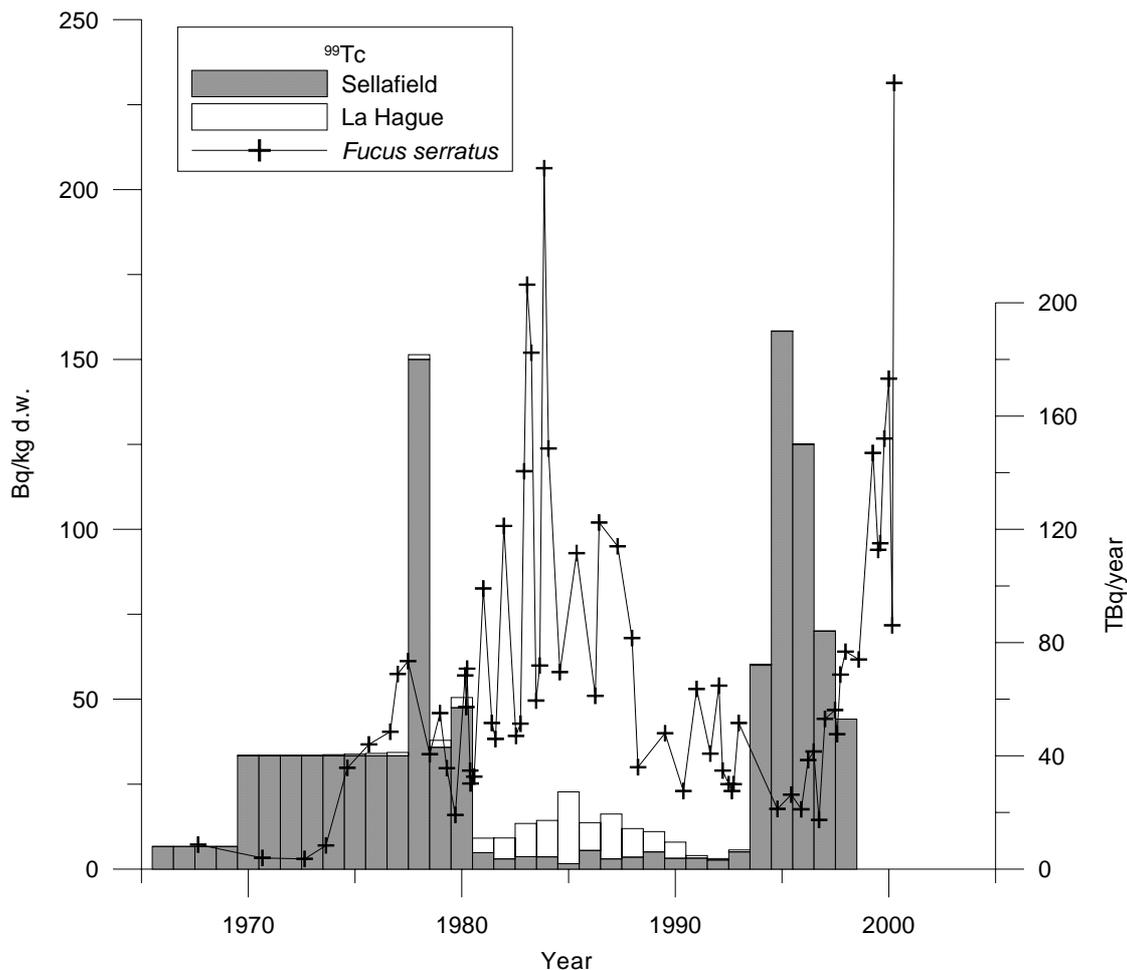


Figure 15. Technetium-99 activity concentration in *Fucus serratus* (primary axis) from Särödal (56.76 °N, 12.63 °E), 1967-2000, and annual ^{99}Tc activity discharge from Sellafield (U.K.) and La Hague (France) (secondary axis).

The ^{99}Tc concentration in *Fucus serratus* has increased approximately with a factor 20 in the year 2000 to approximately 200 Bq kg^{-1} due to the EARP pulses in 1995-1996. The transfer time to the Swedish west coast of Sellafield derived ^{99}Tc is estimated to 5-6 years. The response from the ^{99}Tc discharge at the Särödal site is not a distinct peak but is smoothening in time due to the dispersion of seawater and natural variability between different samples.

The Tc-pulses from 1995-1996 have not yet stagnated at the Swedish west coast. The seasonal variation of the ^{99}Tc concentrations in the 1994-2000 samples is not as pronounced as in previously studies. One explanation can be that the seawater movement has changed over the years and altered the salinity and the transport routes in the coastal area.

Most of the ^{237}Np on the Swedish west coast are believed to originate from the Sellafield nuclear reprocessing plant. In Table 5, the ^{237}Np activity concentrations in *Fucus vesiculosus* from the Swedish west coast during the summers of 1991, 1995, and 2001 are listed. The concentrations ranged from 0.16 ± 0.02 to $1.02 \pm 0.09 \text{ mBq kg}^{-1}$ depending on the locations and sampling year. The concentrations show a slight tendency to decrease along the coast from north to south with small deviations, but not as pronounced as in earlier studies of ^{99}Tc (Lindahl et al., 2002).

Table 5. Neptunium-237 activity concentration (mBq kg^{-1} , dry weight) in *Fucus vesiculosus* from the Swedish west coast collected 1991, 1995, and 2001 (Uncertainty expressed as one standard deviation).

No.	Sampling sites	Locations	1991	1995	2001
1	Saltö	58.87 °N, 11.12 °E	0.96 ± 0.08	0.88 ± 0.08	0.17 ± 0.02
2	Smögen	58.35 °N, 11.22 °E	0.47 ± 0.04	0.43 ± 0.04	0.66 ± 0.06
3	Gullholmen	58.18 °N, 11.42 °E	0.49 ± 0.04	0.66 ± 0.06	0.38 ± 0.03
4	Marstrand	57.90 °N, 11.63 °E	0.31 ± 0.03	0.37 ± 0.03	0.41 ± 0.04
5	Sandö	57.48 °N, 11.88 °E	0.33 ± 0.03	0.51 ± 0.05	1.02 ± 0.09
6	Getterön	57.12 °N, 12.20 °E	0.57 ± 0.05	0.53 ± 0.05	0.55 ± 0.05
7	Glommen	56.93 °N, 12.35 °E	0.31 ± 0.03	0.37 ± 0.03	0.34 ± 0.03
8	Görvik	56.65 °N, 12.77 °E	0.47 ± 0.04	0.67 ± 0.06	0.23 ± 0.02
9	Kattvik	56.47 °N, 12.78 °E	-	-	0.26 ± 0.03
10	Torekov	56.42 °N, 12.63 °E	0.43 ± 0.04	0.39 ± 0.03	-
11	Arild	56.27 °N, 12.58 °E	-	-	0.16 ± 0.02

In Figure 16, the $^{239+240}\text{Pu}$ activity concentrations in *Fucus vesiculosus* from the Swedish west coast collected in 2001 are displayed. The $^{239+240}\text{Pu}$ concentrations showed a more pronounced gradient along the coast. One explanation could be the mixing of the low-salinity water from the Baltic Sea with the high-salinity water from the North Sea.

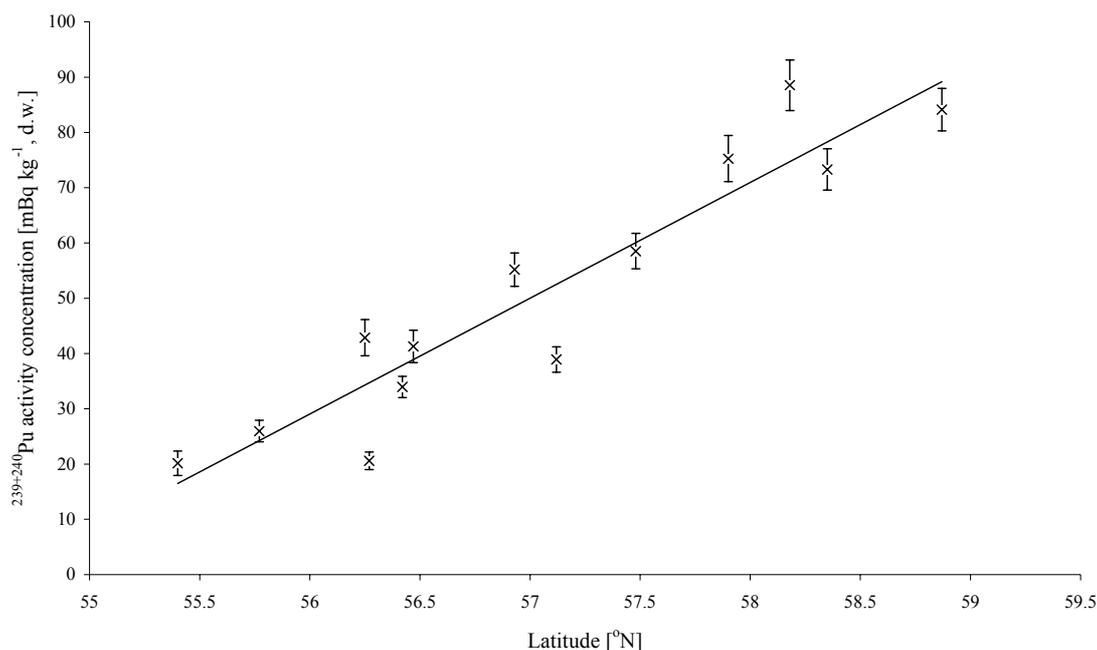


Fig. 16. $^{239+240}\text{Pu}$ activity concentration in *Fucus vesiculosus* from the Swedish west coast collected in 2001.

Danish waters

Time series of ^{137}Cs in Danish waters has indicated, that there may be no net sedimentation of ^{137}Cs in the Baltic Sea as seen previously. As earlier results indicated a fast sedimentation of ^{137}Cs in the Baltic Sea due to a high general sedimentation rate and a high K_d in the brackish waters, the explanation could be that the sedimentation is now counterbalanced by remobilisation from the large amounts of Chernobyl ^{137}Cs deposited in the Baltic sediments and to a smaller extent by runoff from land.

The Northern Adriatic Sea has been selected as a study area for the REMOTRANS project because it is the Italian area most affected by direct Chernobyl fallout and by the delayed input of Chernobyl radionuclides through two main rivers: Isonzo and Tagliamento. The study area is shallow (maximum depth: 20 m) and the fine sediments exported by the rivers are accumulated in two elongated lenses near the river mouths and in the inner Gulf of Trieste. The results of the campaigns conducted in this area in June and November 1987 indicated that most Chernobyl radionuclides were confined in the fine grained sediments close to the shore, while very low levels characterised the offshore residual sand. The prodelta acted as reservoirs for radionuclides transported by the rivers (both concentrations and inventories of ^{137}Cs and ^{134}Cs were about one order of magnitude higher than in the mud shelf sediments).

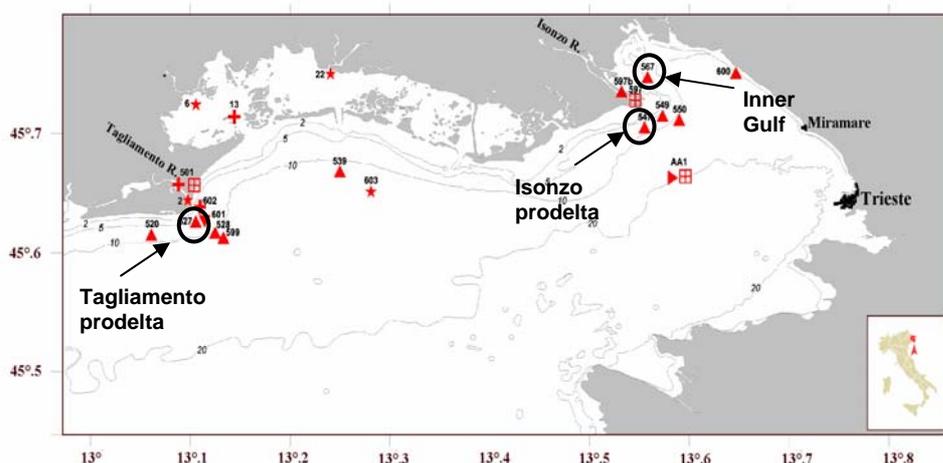


Fig. 17 – Study area and sampling points, 1987 and 2001.

A sampling campaign was conducted in June 2001, re-visiting the stations already examined in 1987-89: the objective was to assess the stability of the sedimentary deposits of radionuclides (^{137}Cs , $^{210}\text{Pb}_{\text{ex}}$, $^{234}\text{Th}_{\text{ex}}$ and ^7Be) with time.

In Spring 2001, the short-lived $^{234}\text{Th}_{\text{ex}}$ and ^7Be were present only in the upper 1-2 cm of sediment, indicating a limited sediment accumulation.

The time trend of radionuclide vertical profiles is examined at three stations, representative of areas characterised by different sedimentation regimes: St. 527 (Tagliamento prodelta) St. 567 (inner Gulf of Trieste) and St. 547 (Isonzo prodelta).

In 1987 – 1989 all vertical profiles near the Tagliamento river mouth were characterised by the presence of Chernobyl-derived ^{137}Cs and ^{134}Cs only and an excess ^{210}Pb in a sediment layer 10 – 15 cm thick, overlying old sediment layers, that did not show evidence of global fallout radionuclides (Fig. 18). This indicated that all sediment deposited from 1963 to 1986 had been re-suspended and transported away. ^{137}Cs concentrations ranged between 200 and 300 Bq kg^{-1} and the inventories between 2 and 12 kBq m^{-2} .

The 2001 profiles show significant differences with respect to the past. Again, ^{137}Cs is only present in the upper 20 cm, but its concentration is significantly lower than expected from the old profiles, decay corrected. The analysis of the vertical profile of excess ^{210}Pb (Fig. 19)

gives an explanation for this trend: again, there is no excess ^{210}Pb below 22 cm, and all sediment between the sediment surface and 20 cm depth shows constant concentration, and has probably been deposited at the same time. The big flood of November 2000 could be responsible for the deposition of such a thick layer of sediment. The new data confirm the hypothesis made in 1987–89: the sediments transported by the river are temporarily stored in the marine area facing the river mouth, but the recent sediment can be completely re-suspended by intense meteo-marine events. The same holds for all vertical profiles near the Tagliamento river mouth.

Completely different is the situation in the Gulf of Trieste. Here sedimentation processes are regular and the radionuclide vertical profiles have maintained the same characteristics in the period 1987 – 2001. Figure 20 shows the vertical profiles of cesium isotopes at St. 567, in the inner part of the Gulf.

The vertical profile of excess ^{210}Pb (Fig. 21a) indicates regular sediment accumulation rate ($0.53 \text{ g cm}^{-2} \text{ y}^{-1}$). Based on ^{210}Pb dating, it is possible to re-construct the vertical profile of the 2001 core as it was in 1987 (Fig. 21 b). The vertical profile obtained in this way almost exactly reproduces that found in 1987. We can conclude that the inner Gulf of Trieste is an area of regular sediment accumulation and that the radionuclides are here permanently stored. The present inventory of ^{137}Cs is around 6 kBq m^{-2} , equal to that obtained in 1987, indicating that the new input from the river has till now compensated radioactive decay.

The third type of vertical profile is that characterising the prodelta of the Isonzo river, where in 1987 concentrations and inventories of ^{137}Cs and ^{134}Cs were highest and sediment accumulation rate was fast. The Chernobyl peak was recorded at a depth of 16 cm (Fig. 22a). In the 2001 vertical profile (Fig. 22b), ^{137}Cs concentrations are one third those measured in 1987 and slowly increase downcore. There is no evidence of a Chernobyl peak, that instead in 1987 was very pronounced. The excess ^{210}Pb shows the same trend, with increasing values downcore, indicating a very fast sediment accumulation rate. In any case, despite the limited water depth (8 m) at this station, meteo-marine events seem not able to re-suspend the large amount of sediment transported by the river.

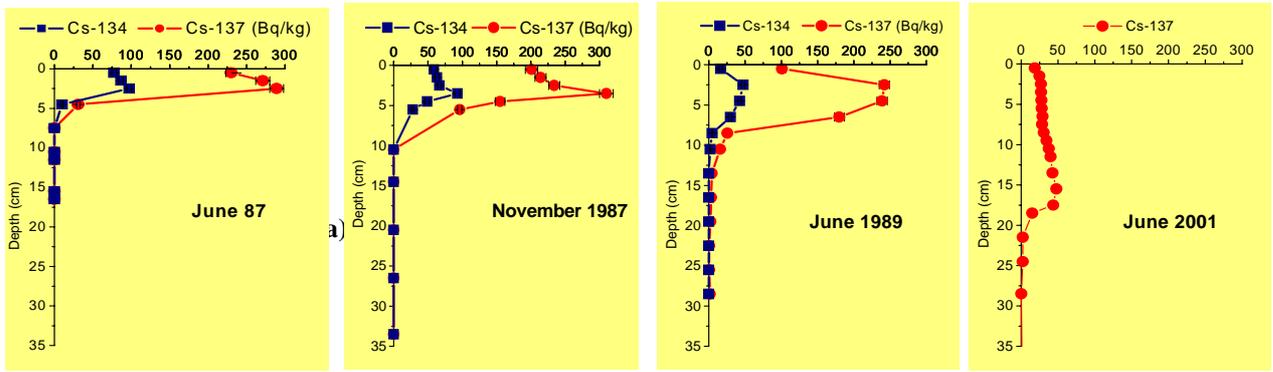


Fig. 18 - Vertical profiles of Cs isotopes at station 527, 1987 – 2001.

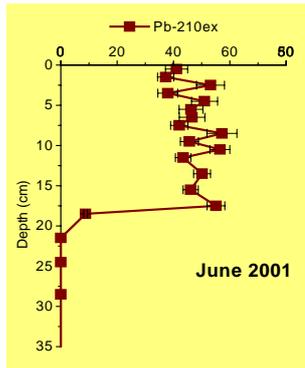


Fig. 19 Vertical profile of ^{210}Pb at St. 527, 2001.

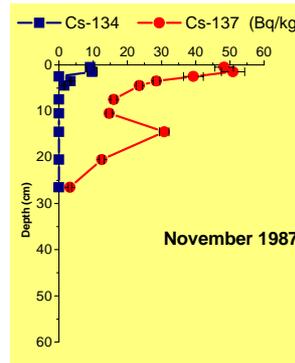


Fig. 20 - Vertical profiles of ^{137}Cs and ^{134}Cs at St. 567, in the inner Gulf of Trieste, 1987 (a), and 2001 (b).

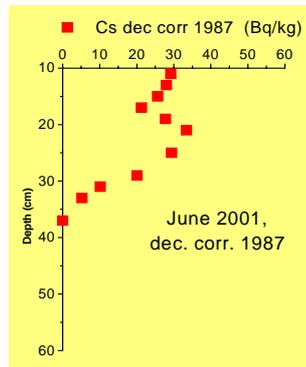
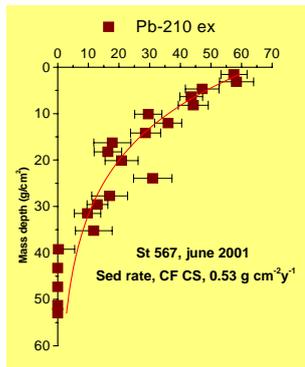
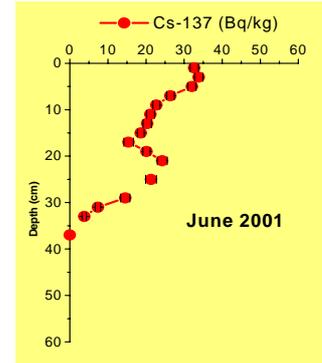


Fig. 21 – Vertical profile of the excess ^{210}Pb and of the ^{137}Cs decay corrected to 1987, based on ^{210}Pb dating of the core collected in 2001 at St. 567, in the inner Gulf of Trieste.

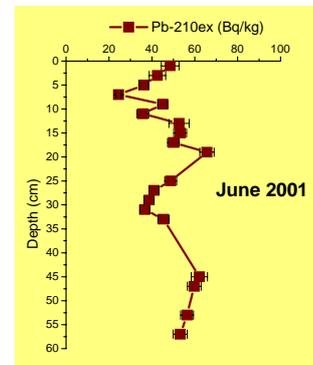
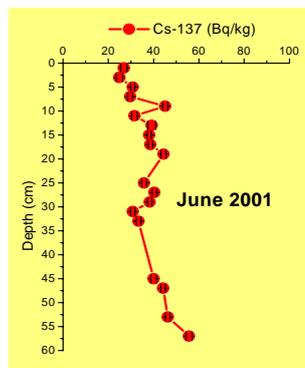
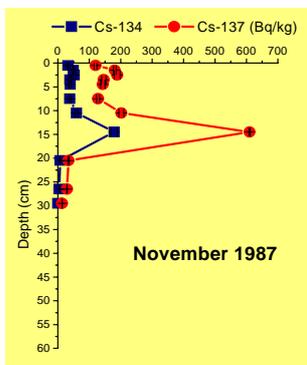


Fig 22 – Core 547, Isonzo river prodelta: vertical profiles of ^{137}Cs in 1987 and 2001 and of the excess ^{210}Pb in 2001. Core length: 30 cm in 1987 and 80 cm in 2001.

Work Package 3:

Objectives

To carry out a comprehensive study on the processes controlling radionuclide remobilisation from sediments.

Participants: ULUND, IRSN, NRPA, AUN, CEFAS, UCD, ENEA, CIEMAT

The overall objectives of WP3 were to (i) carry out a comprehensive study of the processes controlling radionuclide remobilisation from contaminated oxic and anoxic sediments, and (ii) establish the influence of these processes on the long-range transport of remobilised radionuclides. To achieve these objectives, new field and laboratory experiments were carried out with a view to investigating the role of environmental parameters, radionuclide speciation and solid partitioning in sorption/desorption processes at the sediment-water interface. Studies were conducted in a number of diverse environments, including the Irish Sea, the Rhône Estuary, the English Channel, the Kongsfjord-Krossfjord system (Svalbard), Rørholtjord (Norway), the Skagerrak area in the North Sea, the Baltic and the NW Mediterranean. The work package was divided in four main tasks:

- Solid partitioning – identification of carrier phases;
- Physical processes and remobilisation;
- Remobilisation processes in fjords;
- Remobilisation processes in freshwater.

Results

Solid partitioning – identification of radionuclide carrier phases

Sequential extraction analysis

Traditionally, the distribution of radionuclides between dissolved and solid phases has been described using equilibrium K_d values, in which the solid phase (suspended particulate, sediment) is assumed to be a homogeneous medium. In reality, however, suspended particulate and sediment are complex heterogeneous mixtures of various components, including residues of weathering and erosion such as clays and other aluminosilicates, substances produced by biological activity both organic (living organisms and humic substances), and inorganic (carbonates, phosphates and silica), and diagenetic products including iron and manganese oxyhydroxides and sulphides. The diverse components that constitute the solid matrix are usually present not as separate particles, but rather as aggregates, and act as reservoirs for radionuclides in the environment. Further, changes in environmental conditions (e.g. salinity, redox potential, organic ligand concentration) can lead to the remobilisation of radionuclides from the sediment to the liquid phase. Hence, identification of the main binding sites and phase associations of radionuclides in sediment can provide insight into the geochemical processes taking place and facilitate a realistic assessment of the potential for remobilisation.

Although significant advances have been made in the direct determination of the solid phase speciation of radionuclides, the task is often hampered by the low concentrations found in most contaminated sediments. An alternative (indirect) approach is sequential extraction analysis, which can provide important information on the chemical and physical conditions that govern the remobilisation of radionuclides in natural water systems.

The method uses a succession of chemical reagents (extractants) chosen to selectively dissolve the various geochemical phases likely to be affected by changes in environmental conditions within the sediment and/or the surrounding water. Sequential extraction schemes have often been criticised because the analytical results are affected by a number of pitfalls, the most serious of which relate to the poor selectivity of the reagents used and the post-extraction resorption of released radionuclides. In addition to these analytical problems, there are also potential artefacts associated with the preservation of sample integrity between the

time of sample collection and the extraction and, in the case of anoxic sediment, the presence/absence of oxygen during the extraction steps. Despite these limitations, satisfactory results can be obtained by a careful choice of the extractants and a stringent validation of the protocol employed.

As part of the REMOTRANS project, a five-day workshop on the solid partitioning of plutonium in anoxic sediments was held in April 2001 at the Laboratoire d'Études Radioécologiques de la Façade Atlantique (IRSN-LERFA) in Cherbourg. The main objective of this workshop was to share experience and expertise in the field of sequential extraction analysis between the participating laboratories, and to arrive to a consensus protocol for the study of the solid partitioning of plutonium in anoxic sediments, to be applied in tidal and subtidal sediments from the Irish Sea in the course of the project. The consensus protocol adopted is based on the procedure proposed by Tessier *et al.* (1979) and partitions radionuclides into five operationally-defined geochemical fractions: exchangeable; acid-soluble; reducible; oxidisable and residual. The protocol was specifically modified to allow controlled dissolution of oxygen sensitive sulphide species in the first extraction step while preserving the reductive nature of the sediment during subsequent extractions (prior to the oxidisable extraction step). In addition, a complexing agent (sodium citrate) was employed to inhibit post extraction resorption, and stringent precautions were taken throughout to preserve the natural geochemical conditions of the sediment column.

A rigorous evaluation of the selectivity of the proposed scheme was undertaken to validate the efficacy of the protocol. The results of these experiments indicated that post-extraction resorption of plutonium in the course of sequential extraction analysis is greatly reduced by the addition of sodium citrate to each extractant. While no significant dissolution of any non-targeted geochemical phases was observed, it was clear, however, that plutonium bound to carbonate minerals can be solubilised during the exchangeable extraction due to ligand competition with sodium citrate. Nevertheless, although the data showed that the addition of sodium citrate does modify the selectivity of the protocol, on balance its use is to be recommended if resorption and redistribution are to be minimised.

The decision as to whether a chelating agent is used in a sequential extraction protocol should be governed by the objectives of the study. If the primary question concerns the determination of precise geochemical associations, rather than the simulation of actual environmental processes, the inclusion of such an agent is indicated as it facilitates the identification of those sediment phases with which plutonium is predominantly associated.

Nevertheless, a number of other sequential extraction protocols widely used in the literature were also applied in the course of the project in order to determine the solid partitioning of plutonium in oxic sediments from different marine environments.

Plutonium carrier phases in Irish Sea sediments

A collaborative expedition (DIAPLU) to the eastern Irish Sea and to intertidal areas of the Cumbrian coast took place in July 2002. The objectives of this sampling campaign were to (i) determine the solid partitioning of plutonium originating from the discharges to the north-eastern Irish Sea by the Sellafield reprocessing plant in tidal and subtidal sediments from the Cumbrian area and (ii) investigate the influence of pore-water chemistry and the development of anoxic conditions on the remobilisation of this element from the sediment to the overlying water column. The main challenges faced by the participating laboratories were the recovery of high-quality sediment and pore-water samples, and the maintenance of anoxic conditions throughout the analytical processes employed. Sub-tidal sediment cores were retrieved using a modified FLUCHA box-corer from aboard the French research vessel *Côtes de la Manche* (INSU/CNRS). Sediment description and pH-eH profiling were performed onboard immediately after retrieval, and sub-cores taken for sedimentological and diagenetic studies. In the intertidal zones, cores were retrieved by driving PVC pipes into the sediment and excavating the surrounding sediment. Back in the (field) laboratory, sediment core extrusion and sectioning was carried out in an oxygen-free atmosphere under nitrogen, within a glove

box specially designed for the purpose. The cores were sectioned at 2 cm intervals, an outer rind of sediment was removed to eliminate smear contamination, and the sediment placed in pore water squeezing pots. The pots were placed into a pneumatic squeezing rig, where porewater from each section was extruded and collected in 50 ml disposable plastic syringes. The pore water was then transferred to a separate nitrogen-filled glove box, where it was filtered (0.45 μm) in an inert (N_2) atmosphere and subdivided into aliquots prior to the determination of various pore water parameters referred to above. The squeezing pots were disassembled and the sediment immediately frozen inside heavy-duty plastic bags purged with nitrogen, for storage until further analysis by sequential extraction.

A number of parameters were determined on water samples (major and trace elements, sulphides, sulphates and dissolved organic carbon) as well as on sediments (some major and trace elements, particulate organic carbon, carbonate content, grain-size distribution, gamma-emitting radionuclides). Special attention was paid to the determination of acid-volatile sulphides (AVS) and chromium reducible sulphides (CRS) and to the solid partitioning of plutonium. A summary of the results is given below.

Intertidal sites – the Esk Estuary and the Solway Firth

A number of sediment cores were taken from a site (Stn. INT-010; 54° 20.35'N, 03° 24.09'W) in the intertidal reaches of the Esk Estuary, some 10 km south of Sellafield. The sampling site was a non-vegetated area, located in the mid-region of the saltmarsh, away from any major edge features or creeks. Previous studies have shown that this site is geomorphologically stable, at least over periods of decades.

Grain size analysis indicated that the sediment was predominantly composed of silt (64–83%), with smaller quantities of sand (16–41%) and clay (8–11%). Although little variation in particle size was observed in the top 20 cm of the core, a layer containing an increased proportion of sand was evident between 20 and 34 cm. Loss on ignition data showed that the organic carbon content of these sediments was very low (<1.4% in all strata), though the uppermost layers exhibited the highest levels. On the other hand, the calcimetry data show little variation in carbonate content (~ 6%) down the profile.

Field measurements of the redox potential in the interstitial pore water showed a rapid decrease in Eh with depth, with values changing from +45 mV at 1 cm to –350 mV at 24 cm and below, clearly reflecting the anoxic character of the subsurface sediment. A similar trend was observed for the pH profile, which relaxed smoothly from 7.3 at 1.0 cm to about 6.7 at 15 cm. Both Eh and pH measurements were carried out using well-calibrated microelectrodes.

A characteristic vertical and time-related redox reaction sequence was evident in the concentration profiles of dissolved Fe, Mn, SO_4^{2-} and alkalinity. Clear evidence of diagenetic redox cycling was apparent, with sulphate reduction accompanied by the production of large pore water concentration gradients of alkalinity and the formation of subsurface maxima in Mn and Fe. Moreover, the drop in sulphate concentration by at least a factor of 3 in the first few centimetres of the profile clearly reflects the anoxic character of the sub-surface sediment.

The concentration profiles of ^{137}Cs , ^{241}Am , ^{99}Tc and $^{239,240}\text{Pu}$ for the core are shown in Figure 1. In the case of ^{137}Cs and ^{241}Am , pronounced sub-surface maxima were observed at depths of approximately 26 and 36 cm, respectively. The $^{239,240}\text{Pu}$ profile also peaked at about 30 cm. Similar sub-surface maxima had previously been observed at shallower depths (~10 cm) in cores taken during the late 1970s in the same general. The change in the depth of these peaks reflects the progressive burial of the Sellafield signal in a zone of active sediment accretion. Clearly, these sub-surface maxima in ^{137}Cs , $^{239,240}\text{Pu}$ and ^{241}Am concentrations correspond to the periods of maximum discharges of these nuclides from nearby Sellafield, and imply a sedimentation rate of approximately 1.2 cm yr^{-1} , which is consistent with sedimentation rates previously reported for this zone. It is also consistent with the sedimentation rate determined from the ^{99}Tc concentration profile, which displays a clear sub-surface maximum at around 6 cm. The discharge history of technetium is very different to those of ^{137}Cs , $^{239,240}\text{Pu}$ and ^{241}Am , and is characterised by relatively low discharges during the 1980s and early 1990s, followed

by a pronounced peak in 1994–98. These trends are clearly reflected in our core. Furthermore, the presence of well-defined maxima in these profiles, decades after input, indicates that relatively little post-depositional mixing of sediment has occurred during this period.

Data on the solid partitioning of plutonium at progressively increasing depths are presented in graphical form in Figure 2. At all depths, a substantial proportion of the plutonium was found to be associated with the R0 exchangeable/readily-oxidisable (mean: $29 \pm 8\%$; range: 13–45%) and R1 acido-soluble (mean: $45 \pm 6\%$; range: 30–54%) phases. Smaller amounts were associated with the R2 reducible (mean: $5 \pm 2\%$; range: 3–10%), R3 oxidisable (mean: $10 \pm 3\%$; range: 6–15%), R4 strongly bound (mean: $8 \pm 4\%$; range: 3–14%) and residual (mean: $4 \pm 2\%$; range: 2–11%) phases.

The small amount of plutonium associated with the exchangeable/readily-oxidisable phase in the oxygenated surface layer compared to that associated with this phase in the deeper anoxic layers, would suggest that some, at least, of the latter is associated with readily-oxidisable geochemical species developed under reducing conditions (e.g. acid volatile sulphides), a result confirmed by measurement of AVS concentrations in the sediment core (Figure 3).

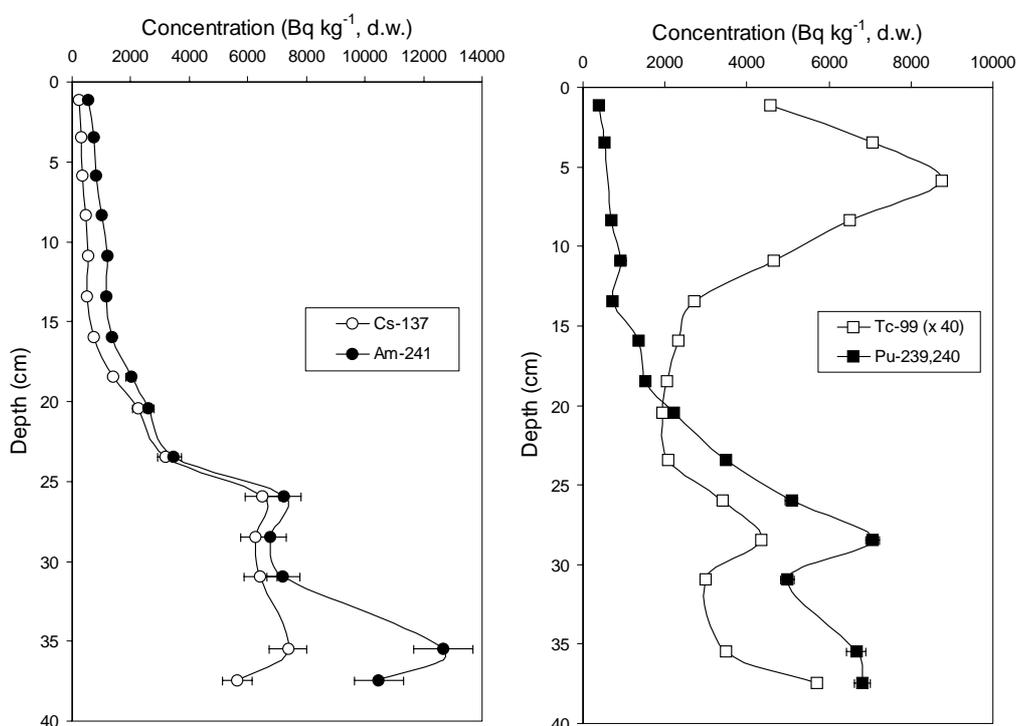


Figure 1. ^{137}Cs , ^{241}Am , ^{99}Tc and $^{239,240}\text{Pu}$ concentration profiles (Bq kg^{-1} , dry weight) in sediment (core INT-010). In some cases, measured uncertainties ($\pm 2 \sigma$) are masked by the data points

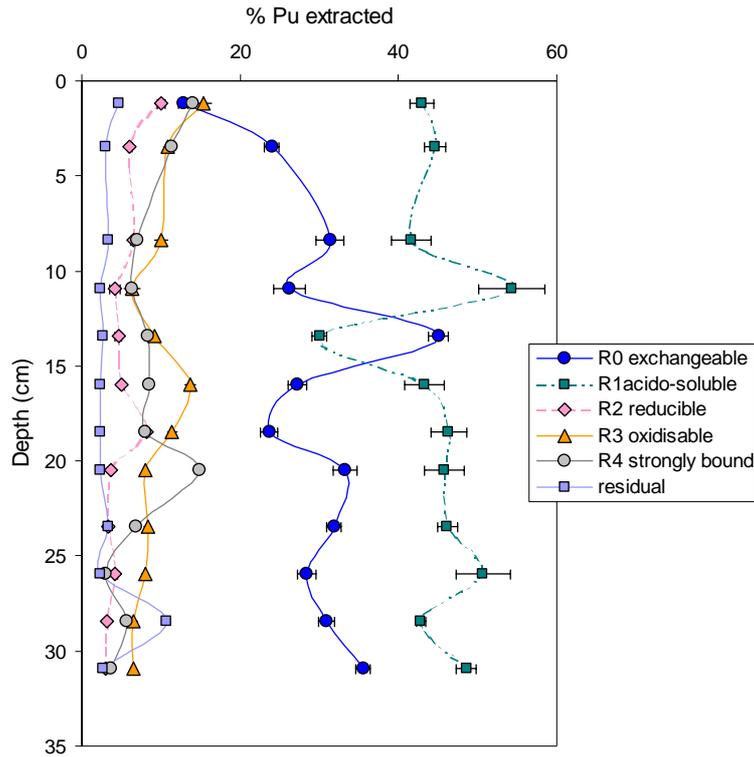


Figure 2. Sequential extraction of $^{239,240}\text{Pu}$ from successive sediment horizons (core INT-010)
Uncertainties are given to $\pm 2\sigma$

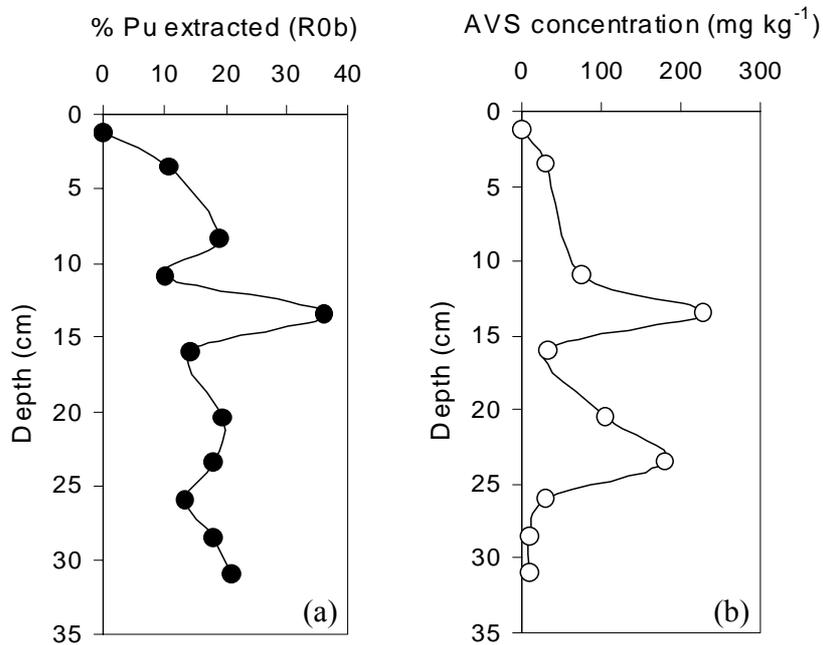


Figure 3. Profiles of (a) fraction of Pu in an easily-exchangeable / readily-oxidisable form and
(b) the concentrations of AVS (mg kg^{-1}) in successive sediment horizons of the Ravensglass
Estuary core

More plutonium was found to be associated with the reducible phase in the surface layer compared to deeper sediments, reflecting the geochemical behaviour of iron and manganese. A similar pattern was evident in the case of the oxidisable phase, more than likely reflecting the somewhat higher concentrations of organic matter in the surface layer. It is interesting that the portion of plutonium associated with the oxidisable (R3) extraction, which is designed to target organic matter among other phases, closely agreed with the organic content of the sediment.

Overall, the results indicate that plutonium at this site is primarily associated with acido-soluble minerals, and that association to acid volatile sulphides and other readily-oxidisable geochemical phases is also of importance in the case of anoxic sediments. Moreover, it is clear that diagenetic reactions taking place during the development of anoxic conditions influence the partitioning of plutonium in the solid phase.

Similar results were obtained for a core taken in an intertidal site in the Solway Firth (Scotland), with the bulk of the plutonium associated with the acido-soluble (R1) and exchangeable (R0) phases.

Subtidal site – the eastern Irish Sea mud-patch

Measurements carried out in a subtidal (Stn. SUB079; 54° 20.026'N, -03° 40.069'W) sediment core retrieved from the muddy sediments just off the Sellafield discharge point showed no dissolved oxygen in the pore water below the top 5 mm. The constant sulphate and alkalinity profiles suggest that if any sulphate reduction is taking place at this site, the sulphate pool is actively being renewed by the overlying water, and is not acting as a limiting factor for the reduction reaction. Nevertheless, evidence that the sediments have undergone some suboxic diagenesis was provided by the presence of subsurface peak concentrations of Fe and Mn at depths of 1 and 5 cm, respectively, as well as by the negative Eh values (~200 mV) measured at depths of ~20 cm down the core. The fact that pore-water Fe concentrations never reached zero indicates that dissolved sulphide production is not very active or that it occurs in anoxic 'micro-niches'. In addition, peak values of both AVS and particulate organic carbon were found at 11 and 13 cm, suggesting that AVS could be produced by the anaerobic decay of autochthonous organisms.

The non-reactive sediment parameters such as carbonate content or ⁴⁰K concentrations were essentially constant, while the ¹³⁷Cs and ²⁴¹Am profiles showed only a very slight gradient of decreasing concentrations with depth, suggesting extensive down-core mixing. Total concentrations and elemental solid partition between acido-soluble fraction (1M HCl) and residual fraction showed that most of the Ca and Sr was acido-soluble. This is not surprising, as these two elements are known to be associated to the carbonate fraction. In the case of Mg, the acido-soluble component was smaller (69%), with the remainder most likely bound to the clay minerals, as is most (92%) of the Al. Significant amounts of Mn (71%) and, to a lesser extent, Fe (26%) were in acido-soluble forms. The latter is known to be associated with carbonate phases in anoxic sediments.

Sequential extraction analyses carried out on six samples of sediment from this core gave a similar solid partitioning for plutonium to that observed in the Esk estuary, with most of the plutonium in the acido-soluble (R1) and exchangeable (R0) phases. In this case, however, no clear correlation with reactive solid phases was found, a result that is consistent with the higher bioturbation activity observed at this subtidal site. Mean ²⁴⁰Pu/²³⁹Pu and ²⁴¹Pu/^{239,240}Pu activity ratios in these samples showed little variation between the different extracted fractions or with depth down the core, showing the mixed (integrated) nature of the signal.

On the whole, the DIAPLU study has shown that plutonium bound to sediment in the Irish Sea is potentially more bioavailable than hitherto believed. Sequential extraction analyses showed that a large proportion of the plutonium in Irish Sea sediments is loosely bound to sites that readily exchange with seawater, readily oxidised upon air contact or easily dissolve upon changes in pH. Moreover, the data clearly indicate that diagenetic reactions taking place during the development of anoxic conditions influence the partitioning of plutonium in the solid phase. Plutonium is known to behave non-conservatively in the Ravenglass Estuary,

with low salinity water and changes in pH causing the rapid desorption of a labile form of plutonium. The present work supports this observation, and indicates that this labile fraction is rather large. Although the results of this work contradict many previous sequential extraction studies, which have reported that little of the plutonium in Irish Sea sediments is in a readily available form, with most publications identifying primary associations with oxide and organic sediment phases, some, at least, of the disparity is almost certainly attributable to failure to take account of the resorption of plutonium in the course of sequential extraction. In addition, many researchers have not taken the necessary precautions to preserve the anoxic integrity of the sediment during sampling, storing and extraction, an omission which undoubtedly leads to unwanted, non-quantifiable phase changes in the course of analysis.

Plutonium carrier phases in other environments

The consensus sequential extraction protocol for the study of the solid partitioning of plutonium in anoxic sediments was also applied to a number of samples from a sediment core taken in the Roads of Cherbourg (English Channel), affected by the discharges from the La Hague reprocessing plant. Once more, the results revealed a significant extraction of plutonium in the exchangeable and acido-soluble fractions.

An alternative protocol widely used in the literature, not using sodium citrate to inhibit post-extraction resorption, was also employed for the determination of the solid partitioning of plutonium in oxic sediments from different marine environments, including the Baltic Sea, the NW Mediterranean, Svalbard, the Irish Sea and the Rhône Estuary. The protocol used also partitions radionuclides into five operationally-defined geochemical fractions, although the sequence of the extractions is slightly different: exchangeable (using calcium chloride as extractant); acido-soluble (using 0.5M ammonium acetate as extractant); oxidisable (using sodium pyrophosphate as extractant); reducible (using a buffer of oxalic/oxalate as extractant) and residual. In all cases, the bulk of the plutonium was found to be associated to the oxidisable and reducible phases, with little of the plutonium in a exchangeable or acido-soluble form. These data contrast with those obtained in the Irish Sea and the English Channel using the consensus modified protocol, and clearly highlight the caution required to interpret results arising from sequential extraction analysis. It is recommended that a number of parameters on pore-waters and sediments be measured in order to help interpretation.

Physical processes and remobilisation

Physical processes in the Rhône prodelta

The possible impact of physical processes on the remobilisation of radionuclides were studied in the course of the REMOTRANS project using the Rhône prodelta as an example of dynamic, non-stable system. Prodeltaic areas are well known for their ability in trapping fine-grained sediment and associated contaminants. These areas are characterized by high sedimentation rates and a low stability in the deposits. In the Rhône area, the prodelta is located between 20–50m depth and sedimentation rates can reach values as high as 50 cm y⁻¹. The shallow depths favour physical resuspension during storms or flood events. Indeed, analysis of archive data gathered in 1984, 1990 and 1994 allowed us to demonstrate that during a major flood in 1994, more than 40 cm of sediment were flushed away from regions with depths around 20-30 m in the prodelta. This result was obtained by studying the ¹³⁷Cs/¹³⁴Cs activity ratio in cores sampled at the same location at these three periods. In order to study the effects of flow velocity on sedimentation and remobilisation processes, of clear importance for the remobilisation and traslocation of radionuclides, a series of experiments were conducted within the frame of REMOTRANS project in order to determine the value of the critical shear stress of erosion in this area.

Experiments were carried out in a flume tank with a view to determining this parameter on sediment cores sampled close to the Rhône mouth. Critical values ranged between 0.08 and 0.12 N m⁻². Since, to our knowledge, no data has been published regarding values for this parameter in the area of study, an ADCP (AWAC - Acoustic Waves And Current) was moored for 35 days from 29 April to 4 June 2002 off the Rhône mouth at 16m depth in order to obtain field values to compare with the flume tank experiments. The mean speed of the bottom current was 7 cm s⁻¹ while instantaneous reached 30 cm s⁻¹. The maximum shear stress due to the current only was 0.11 N m⁻² while the maximum value related to the waves was near four times higher, at 0.4 N m⁻². Shear stress was more than 15% of the time above the critical value determined experimentally. It therefore appears that waves are the main factor in resuspension phenomena in this area.

The backscattered signal showed various events where resuspension processes could be involved. One is linked to an increase in the Rhone flow (from 700 m³ s⁻¹ to 2700 m³ s⁻¹), leading to an increase in the turbidity signal in the whole water column. This increase is due both to export of particulate matter from the river and to resuspension due to an increase in the shear stress on the bottom. Several other events concern only the bottom layer and are related to strong periods of wind especially south-southeast winds. Indeed, this area is characterized by two main types of wind, namely north-northwest winds (i.e., Mistral and Tramontane) and south-southeast winds. The former can reach very high speeds but their interactions with the bottom are quite low due to the protection from the coastline; in contrast, winds from the south-south west lead to the formation of large waves, resulting in strong resuspension events. In these latter cases, resuspended particulate matter is then transported by bottom current towards the south west i.e. the gulf of Lion.

Long-range transport of remobilised radionuclides

In order to study the transport of remobilised radionuclides from the Irish Sea into the North Sea and, in particular, the Skagerrak, a sediment core collected in the north-eastern part of this region was retrieved during an expedition in 2000 from a water depth of 100 m using a hydraulically-damped multiple corer. Analyses of ²¹⁰Pb, ¹³⁷Cs, ²³⁸Pu, ^{239,240}Pu and ²³⁷Np were carried out using a combination of gamma, alpha and mass spectrometry.

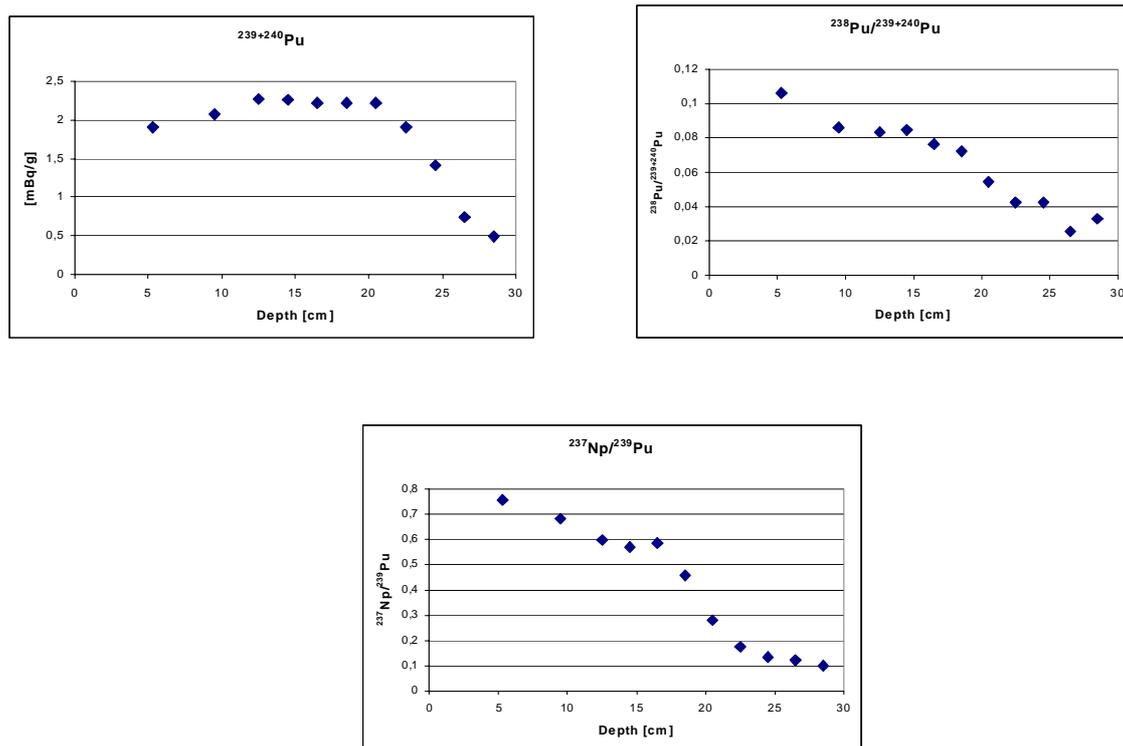


Figure 4. $^{239,240}\text{Pu}$ profile and transuranium isotopic ratios in a sediment core from Skagerrak, 2000

From the ^{210}Pb data, an average sediment accumulation rate of approximately 0.5 cm y^{-1} was estimated. It is well known that this region is subject to high accumulation rates due to the slow down of the anti-clockwise moving currents bringing in large amounts of suspended matter from the North Sea. This is reflected in the high ^{210}Pb excess inventory, at about 13500 Bq m^{-2} , and that of $^{239,240}\text{Pu}$, at 350 Bq m^{-2} .

As seen in Figure 4, the $^{239,240}\text{Pu}$ concentrations are starting to decline but, at the same time, the $^{238}\text{Pu}/^{239,240}\text{Pu}$ activity ratios have steadily increased, reflecting the transport of remobilised plutonium from the Irish Sea or from deposition areas in the western North Sea. Even though ^{237}Np fluxes to sediments are believed to be small in well-oxygenated waters, the concentrations measured in these sediments are relatively high. The $^{237}\text{Np}/^{239}\text{Pu}$ atom ratio down the core shows a similar increase with time to that observed for the $^{238}\text{Pu}/^{239,240}\text{Pu}$ ratio. The integrated weapons fallout ratio is about 0.45, while that from the integrated Sellafield discharges is about 1.7. The more recent inputs of suspended matter to the Skagerrak region clearly show the transport of material with a Irish Sea origin.

Additional surface sediments and sediment cores were collected from the North Sea, Skagerrak and Kattegatt areas in 2001 (Figure 5). Surface sediments (upper 2 cm) and two cores from the Skagerrak and Kattegatt areas were analysed for ^{137}Cs (Stns. 1427 and 1430). In the surface sediments, activity concentrations ranged from 6.4 Bq kg^{-1} (dry wt.) to 13.2 Bq kg^{-1} (dry wt.) in the Skagerrak area. In the sediment core from Stn. 1427, the ^{137}Cs activity signal was detectable down to approximately 12 cm, giving an estimated sediment accumulation of 0.3 cm y^{-1} . In the core from Stn. 1430, ^{137}Cs activities were measurable below 16 cm depth, suggesting a sedimentation rate in excess of 0.4 cm y^{-1} or more. However, dating with the $^{210}\text{Pb}_{\text{excess}}$ method measurement give lower sedimentation rates, suggesting that some vertical mixing of ^{137}Cs has taken place in these cores. In the core from station 1427, the highest ^{137}Cs concentration is seen in the surface layer (upper 1 cm), and the concentration is gradually reduced with depth. In the core from station 1430, maximum concentrations are seen in the layers between 1-6 cm. This could be due to the ^{137}Cs concentrations in the outflowing Baltic contaminated with Chernobyl fallout after 1986.

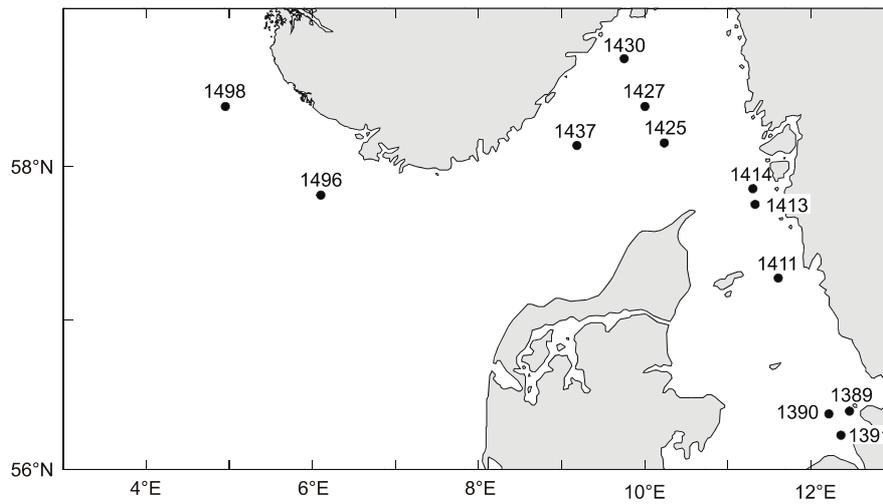
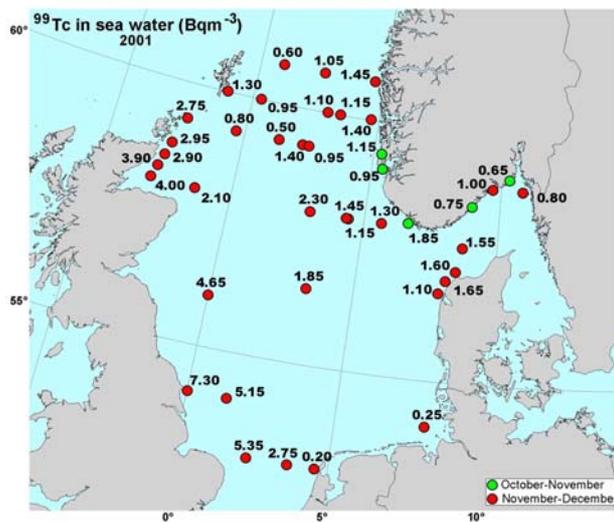
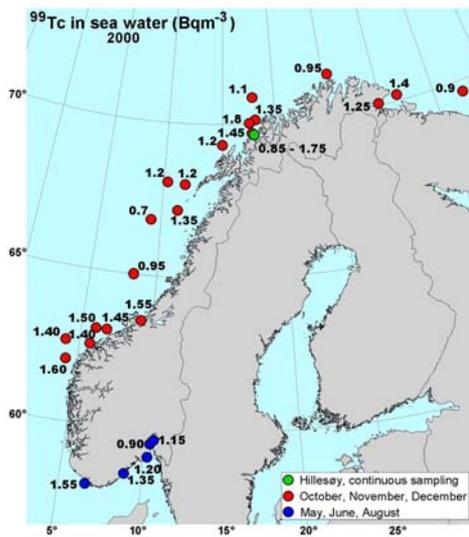
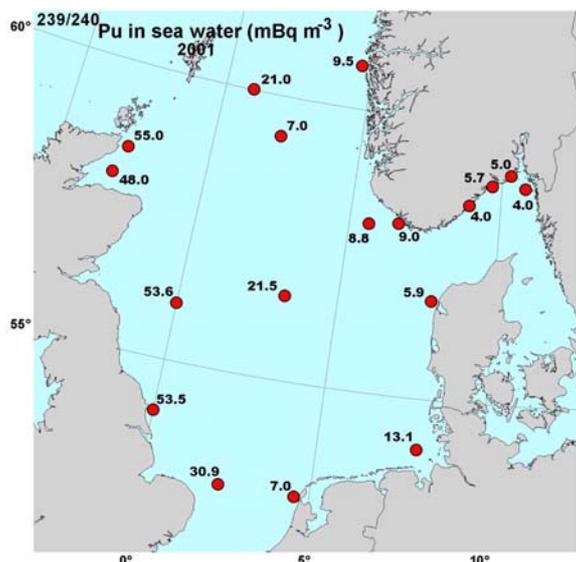


Figure 5. Sediment sampling stations in the Skagerrak/Kategat area, 2001



Results on plutonium 239, 240 in the North Sea in 2001 showed clearly elevated levels along the east coast of Scotland and England, which can only be explained by remobilisation from Irish Sea sediments. The activity ratio Pu-238 /Pu-239, 240 also suggested historical Sellafield releases as the main source.



All results on Tc-99 and Pu-239, 240 in sea water will be used for development of the NRPA box model. In addition, Tc-99 data will be used for the NOASIM model of the Alfred Wegner Institute (M. Kärcher).

Remobilisation processes in fjords

Scavenging, sedimentation and resuspension processes at the glacier-sea interface - the Kongsfjord (Svalbard)

In the period 2001-2003, three campaigns were carried out in the Kongsfjord, an arctic fjord on the western coast of the island Spitsbergen, Svalbard archipelago, in order to investigate the sedimentation/resuspension processes at the glacier-sea interface, the origin and pathways of radionuclides associated with sedimentary particles, the role of colloid in the scavenging of particles from the water column, and the stability of sedimentary deposits. The disequilibrium between ²³⁴Th and ²³⁸U in the water column was used as tracer for scavenging processes of particle-associated radionuclides, while the vertical distribution of ¹³⁷Cs, ²¹⁰Pb_{ex} and ²³⁴Th_{ex} was used to trace sediment accumulation on different time-scales and derive information on the stability of sedimentary deposits at the glacier-sea interface.

The vertical profiles of ²³⁴Th in soluble and particulate form were determined in the outer fjord and at the glacier-sea interface both in summer and in winter (iced) situation. The distribution of suspended particles shows a maximum in the southern part of the inner fjord, close to Kongswegen, the most productive glacier. The release of particles from the glacier is huge, both in summer and winter, under the ice cover: consequently, a marked disequilibrium between ²³⁴Th and ²³⁸U is observed in both seasons, indicating that scavenging processes are active all over the year. Typical profiles of ²³⁴Th and ²³⁸U (calculated from salinity) are shown in Figure 6.

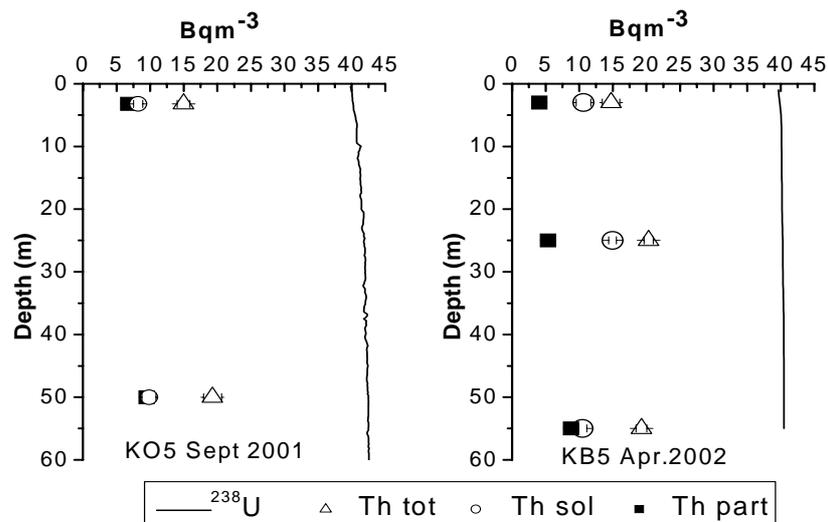


Figure 6. Vertical profiles of soluble and particulate ^{234}Th in the water column of the inner Kongsfjord in summer (September 2001) and winter (April 2002).

^{234}Th concentration on particles increases with distance from the glacier and particulate ^{234}Th fluxes are high and of the same order of magnitude in the whole fjord (around $30 \text{ Bq m}^{-2} \text{ d}^{-1}$). Particle associated radionuclides are efficiently scavenged both in the inner fjord (high particle population, low ^{234}Th concentration) and in the outer part (lower particle population, higher concentrations). Little or no colloidal intermediates are involved in the process. In late summer, ^{234}Th derived particle fluxes are in the range $5 - 9 \text{ g m}^{-2} \text{ d}^{-1}$ in the outer fjord and $2 - 4 \text{ g m}^{-2} \text{ d}^{-1}$ in the central/outer fjord. Also in late winter, particle fluxes in the water column of the inner fjord are relatively high: $7 - 8 \text{ g m}^{-2} \text{ d}^{-1}$.

Sedimentation processes were studied at seven stations: four in the inner and three in the outer fjord. Sediment accumulation rates decrease from $0.02 - 0.09 \text{ g cm}^{-2} \text{ y}^{-1}$ in the outer fjord, to $0.2 - 0.4 \text{ g cm}^{-2} \text{ y}^{-1}$ in the central fjord, to $2 - 5 \text{ g cm}^{-2} \text{ y}^{-1}$ in the inner fjord (Figure 7).

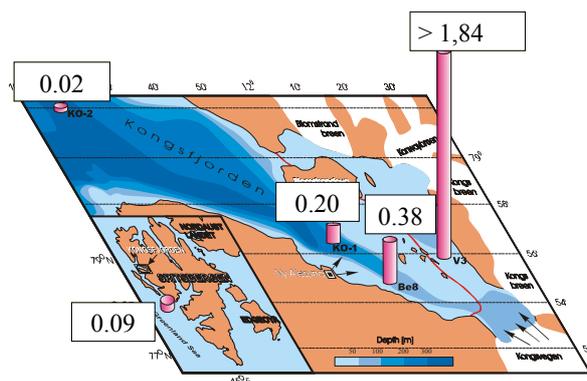


Figure 7. Sedimentation rates in Kongsfjord. ($\text{g cm}^{-2} \text{ y}^{-1}$)

Near Kongsvegen, the concentrations of $^{210}\text{Pb}_{\text{ex}}$ and ^{137}Cs were almost constant along the cores, and an estimate of the sediment accumulation rate could only be done using the short-lived ^{234}Th . In this area, the downcore penetration of $^{234}\text{Th}_{\text{ex}}$ is not due to bioturbation, as the high particle flux at the glacier-sea interface does not allow the establishment of a stable benthic community. The presence of an excess ^{234}Th at depth in the core is then related either to sedimentation or to lateral sediment transport. The vertical profile of excess ^{234}Th at the station close to the glacier indicates that during the summer season, some 15 g cm^{-2} of sediment is accumulated. In winter, the sampling was repeated twice: in the first case (2002,

April) an excess of ^{234}Th was detected in the sediment down to 15 cm, in the second case (2003, March) it was only limited to the first cm. The integrated deficit of ^{234}Th in the water column was then compared to the excess inventory in the sediment (Figure 8): results (2002: +2900 Bq m^{-2} ; 2003: - 100 Bq m^{-2}) show evidence of a) lateral advection, and b) re-suspension processes.

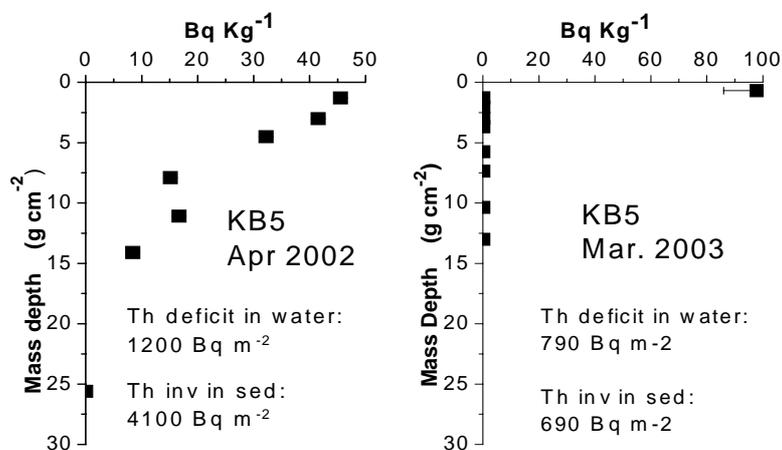


Figure 8. Vertical profiles of excess ^{234}Th in sediments near Kongsvegen.

In summary:

- The scavenging processes are efficient both in the summer (highest) and in winter (iced sea);
- Sediment accumulation rates increase of two orders of magnitude from the inner to the outer fiord;
- The particles discharged from the land (mainly by glacier melting) into the fiord and the associated radionuclides are trapped in the inner part: post-depositional re-mobilisation of sediments takes place, but there is no evidence of their transport to the outer fjord.

Remobilisation processes in Lake Rørholtfjord

Rørholtfjord is a branch of Lake Tokke, in southeast Norway, and is situated ca 60 meter above sea level. The drainage of Lake Tokke is about 900 km^2 with no tributary drainage directly into Rørholtfjord. The maximum depth of the fjord is 147.5 meter. The water masses are stratified with an upper fresh-water layer of 135 m and a bottom layer of 12.5 m where the water is anaerobic and saline with a maximum salinity of 17%. Following the withdrawal of the inland ice, the area was gradually lifted above sea level 8500 – 8000 years ago.

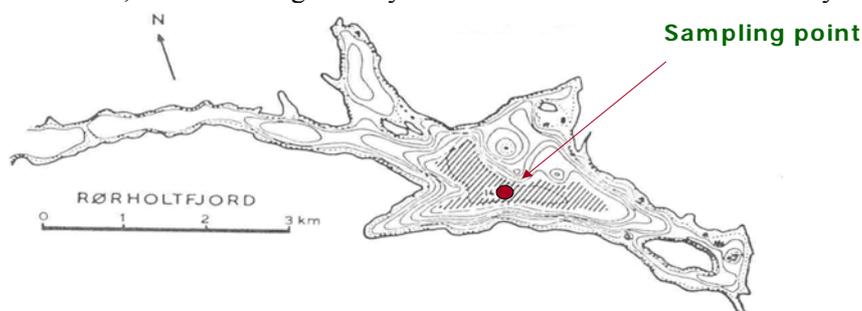


Figure 9. Depth contours (20 m apart, additional 10 m contour (dashed line)) of Lake Rørholtfjord. The sampling point is indicated.

The Rørholtfjord bottom saline water is alleged to be trapped seawater from the time the lake was part of the sea, with additional contribution from underlying marine clays due to upward flow of artesian groundwater.

The fieldwork in the Lake Rørholtfjord (Figure 9) included sampling of fresh water, fossil seawater and sediment cores. For the first time *in-situ* fractionation was carried out in deep (135–147 m) anaerobic fossil seawater. Using 150 m tubes, echo sound and peristaltic pumps, waters at defined depths were transported through the fresh water oxic layer and fractionated by 0.45 µm Millipore filters and 10 kDa Amicon hollow fibers onboard ship, with all fractionation equipment contained in N₂ tent.

Vertical distribution in water

The vertical distribution of most elements (e.g. Cs, Fe and U, Table 1), radionuclides (e.g. ¹³⁷Cs and ^{239,240}Pu (Figure 10) and total organic carbon (TOC) in the water profile were significantly enriched in the saline anoxic bottom water. It is clear from these measurements that due to the anoxic conditions and the high concentration of divalent Fe and low molecular mass total organic carbon (TOC) in the saline layer, most of the elements and radionuclides originally deposited on the lake surface, have been kept in solution and have not deposited in the underlying anoxic sediments. A major fraction of ^{239,240}Pu in the fresh water was found to be associated with colloids (Figure 10d). In the saline anoxic water, however, no association with colloidal material could be identified.

Table 1. Salinity and concentration of stable Cs, Fe and U in the vertical water profile of Rørholtfjord

Depth (m)	Salinity (ms/cm)	Stable Cs (µg/L)	Fe (ng/L)	U (µg/L)
5	0	0.20 ± 0.03	0.011 ± 0.002	0.53 ± 0.06
130	0	< 0.1	0.131 ± 0.019	0.45 ± 0.05
136.5	8.2	0.19 ± 0.03	89 ± 9	1.5 ± 0.1
140.5	15.1	0.30 ± 0.05	391 ± 39	2.6 ± 0.3
143.5	15.1	< 0.3	242 ± 29	2.3 ± 0.2

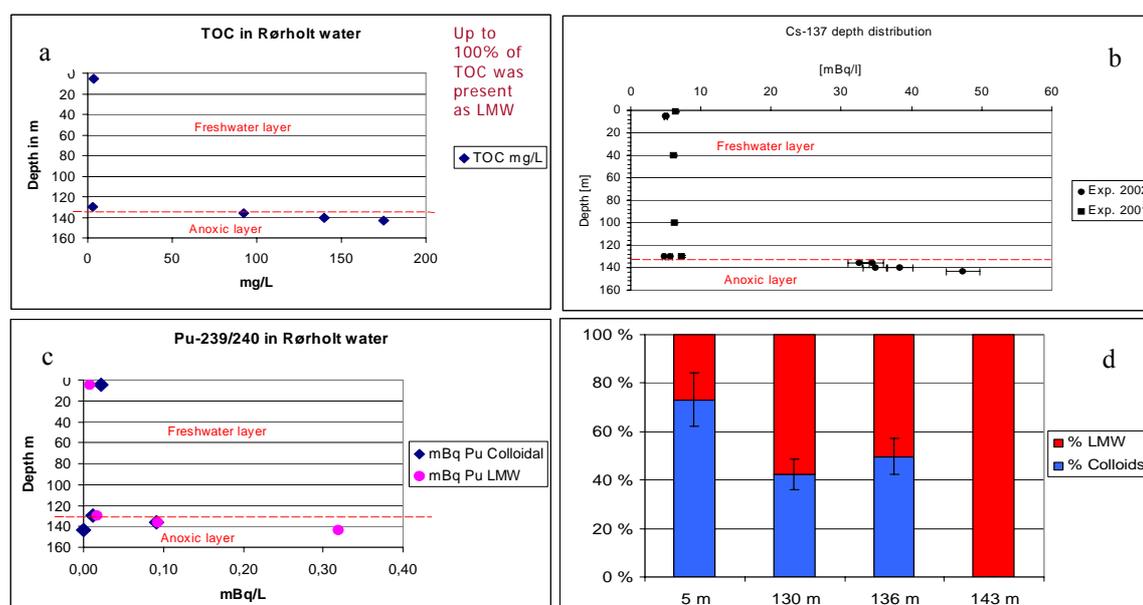


Figure 10. Vertical distribution of the concentration of a) total organic carbon, b) ¹³⁷Cs, c) ^{239,240}Pu in the water profile, and d) the speciation of ^{239,240}Pu in the waters.

Vertical distribution in sediments

The sediment is characterized by a gyttja and blackish silt layer (upper core) covering a grayish silt layer (Figure 11), with high clay content (18 – 30 %) and TOC concentration (50 %) in the uppermost part of the core, and sandy silt and lower TOC concentration (30 %) in the lower depths. The sedimentation rate, as determined by the $^{210}\text{Pb}_{\text{excess}}$ method, was found to be constant over the last 100 yrs.

The activity concentrations of ^{137}Cs , ^{241}Am and $^{239,240}\text{Pu}$ (Figure 12) were significantly enriched in the upper sediment (0-5 cm). ^{137}Cs concentration varied from about 3 Bq kg^{-1} in the lower layers to almost 400 Bq kg^{-1} in the top layers. Based on the $^{210}\text{Pb}_{\text{excess}}$ chronology, the deposition of ^{137}Cs took place during 1950 to 1990, reflecting weapon fallout and fallout from the Chernobyl accident in 1986. The activity concentrations of ^{241}Am and $^{239,240}\text{Pu}$ were low, varying from 0.3 to 2.2 Bq kg^{-1} for ^{241}Am and from 0.2 to 17 Bq kg^{-1} $^{239,240}\text{Pu}$. Both radionuclides were slightly enriched in the upper sediment layers.

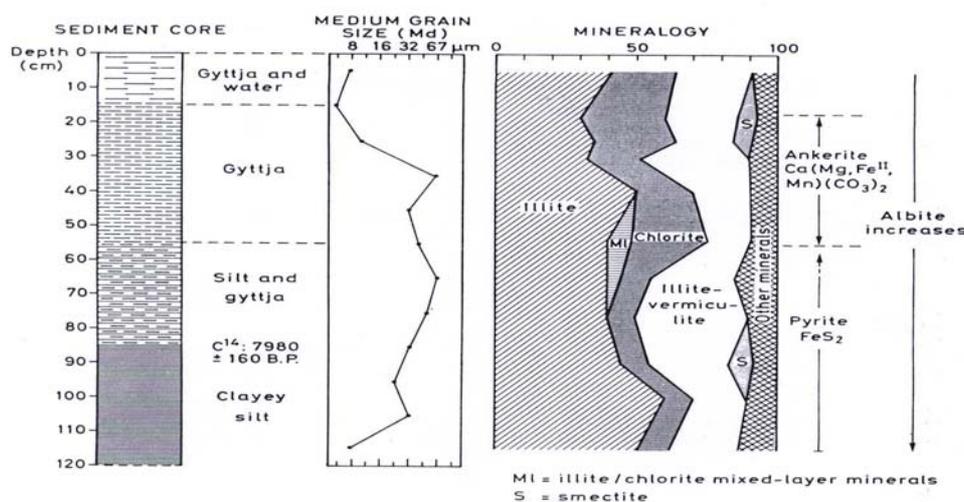


Figure 11. Vertical distribution of texture and mineralogical composition of sediments in Lake Rørholtfjord

Apparent K_d in anoxic saline systems

The apparent K_d values (l kg^{-1}) derived from the bottom water and surface sediment concentrations for trace metals, ^{137}Cs and $^{239,240}\text{Pu}$ (Table 2) were found to be significantly lower than those observed in oxic marine systems. This means that the mobility of the trace metals and radionuclide species is high and the retention by sediments low. The apparent K_d obtained for Pu in the Rørholtfjord is of the same order as that observed in the Yenisey and Ob Estuaries ($1.0 \times 10^4 \text{ l kg}^{-1}$ and $1.6 \times 10^4 \text{ l kg}^{-1}$ respectively) and a factor of two lower than in the marine system of the Kara Sea ($2.3 \times 10^4 \text{ l kg}^{-1}$), but is two orders of magnitude lower than that measured for plutonium in the Irish Sea and the North Atlantic.

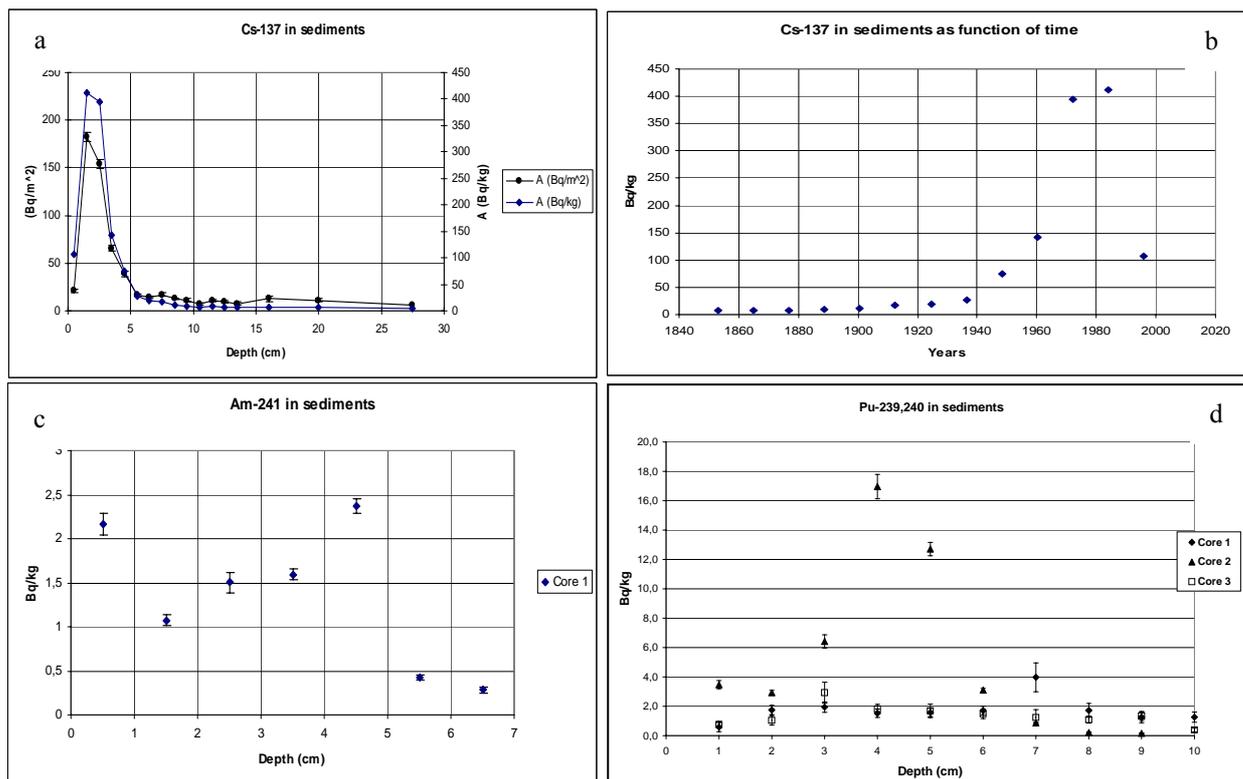


Figure 12. Vertical distribution of (a) ^{137}Cs concentrations in sediments, (b) ^{137}Cs as function of time, (c) ^{241}Am and (d) $^{239,240}\text{Pu}$ concentrations in the sediment cores

Table 2. K_d (1 kg^{-1}) for stable and radioactive elements in lake Rørholtfjord (anoxic sediments/anoxic sea water)

Element	K_d , L/kg	Element	K_d , L/kg
Na	7	Cr	$> 10^5$
Rb	200	U	3×10^3
Cs	2×10^3	Th	$> 5 \times 10^5$
^{137}Cs	2.2×10^3	Fe	2×10^5
Ca	60	Co	2×10^4
Ba	$> 10^3$	Au	200
Sb	10^3	Zn	> 100
Br	0.2	Cd	200
Sc	3×10^4	Hg	200
La	1.5×10^4	$^{239,240}\text{Pu}$	1.1×10^4

Laboratory derived K_d values for anoxic saline systems.

The distribution of radionuclides between sediments and water (i.e. the distribution coefficient K_d) depends on the radionuclide species in question, sediment surface composition and is a function of contact time with the sediments. Furthermore, K_d is a conditional constant, i.e. it is susceptible to physical and chemical changes such as pH, temperature, salinity as well as redox conditions. If the kinetics of sediment-tracer interactions in contaminated sediments is slow, then the apparent K_d may not reflect equilibrium conditions. Unless equilibrium conditions are confirmed, information about the kinetics of these interactions could be more relevant for predictive models than the apparent distribution of

radionuclides between water and sediment. Thus, tracer experiments under controlled pH, redox, salinity and temperature conditions were performed in the course of the project. Sediment samples from the Irish Sea mud patch (collected during the DIAPLU expedition, July 2002) and sediments from the Lake Rørholtfjord (June 2002), varying in TOC concentrations (LOI 3.2 % in Irish Sea sediment, LOI 39.2 % Rørholt sediment) were included in the tracer experiments.

To perform the experiments, aliquots of homogenised sediment (Irish Sea sediments: 10-30 cm, 5 g dry weight in contact with artificial sea water (28‰), Rørholtfjord: 0-30 cm, 0.5 g dry weight in contact with anoxic Rørholtfjord bottom water) were introduced in centrifugation tubes. Tracers prepared in artificial seawater (i.e. $^{236}\text{Pu(V, VI)}$, $^{239}\text{Pu(III, IV)}$, ^{241}Am , ^{60}Co and ^{134}Cs) were added (Irish Sea sediment ratios: 1:2; Rørholtfjord sediments: 1:10 to 1:20, pH 8) under anoxic conditions (N_2 flask system, N_2 flush, $\approx 20^\circ\text{C}$). Redox potentials and pH were continuously monitored using sensors (Hamilton Flushtrode 325, HI8615L ORP transmitters) during the duration of the experiment (1-month). Samples were taken after a contact time of 1 hour (n=3), 1 day (n=3), 1 week (n=3) and 1 month (n=3), and the aqueous phase for each sample was assayed for radionuclide content following high speed centrifugation (Compact centrifuge, Avanti 30, Beckman). The sorption of tracers to tube walls was shown to be less than 5%. During the experiments, the redox conditions were kept anoxic.

The results of these anoxic tracer experiment show that pseudo-equilibrium conditions were reached for ^{134}Cs in Irish Sea sediments after 1 month contact time, when the K_d was $22 \pm 12 \text{ l kg}^{-1}$, a factor of 10 lower than that measured in Irish Sea sediments under oxic conditions. For ^{60}Co , pseudo-equilibrium conditions were not reached in Irish Sea sediments after 1 month contact time. The measured K_d reached $311 \pm 218 \text{ l kg}^{-1}$ (n=3) after one month contact time, and was also significantly lower than previously observed in oxic experiments, which range from 20000 – 100000 l kg^{-1} .

Pseudo-equilibrium conditions were also reached for ^{134}Cs in the Rørholtfjord sediments after one month contact time, with a final K_d of $296 \pm 231 \text{ l kg}^{-1}$, a factor of 10 higher than the K_d for the Irish Sea sediments. For ^{60}Co , pseudo-equilibrium conditions were not reached in Rørholtfjord sediments after one month contact time. The K_d reached a value of $6000 \pm 1800 \text{ l kg}^{-1}$ after 1 month, and was a factor of 20 higher than the K_d for the Irish Sea sediment.

WP3.4. Remobilisation processes in freshwater systems

Extraction experiments were carried out in the course of the project to investigate the remobilisation of ^{137}Cs , ^{60}Co , ^{99}Tc and ^{90}Sr from contaminated freshwater sediments when exposed to seawater. The purpose of these experiments was to simulate the remobilisation processes taking place during the transport of sediments contaminated from Mayak PA (Urals) via the Ob river system (freshwater) to the Kara Sea (high ionic strength, high pH, estuarine environment). For all the radionuclides studied, transfer from a freshwater to an estuarine environment lead to a decrease in the apparent distribution coefficient, resulting in an increased mobility of the radionuclides. Total remobilisation of radionuclides from sediments when exposed to seawater for 80 days was 5% for ^{137}Cs , 15% for ^{60}Co , 1% for ^{99}Tc and 23% for ^{90}Sr . It is clear from these experiments that large amounts of radionuclides could be remobilised in the Ob estuary in the event of a major release of sediments from the Mayak reservoirs.

Sequential extraction experiments carried out on the sediments showed that, as expected, ^{90}Sr is quite mobile, while ^{137}Cs is more strongly bound, with only 20% extracted in the exchangeable, acido-soluble, reducible and oxidisable fractions. For ^{99}Tc , up to 85% of the activity was extracted in the oxidisable fraction, showing lower mobility than expected, while ^{60}Co was more mobile than anticipated, perhaps as a result of the slightly reducing character of these sediments.

Pu isotope ratios in the Ob and Yenisey Estuaries and radionuclides associated with colloids in aquatic systems

The Ob and Yenisey influence is particularly interesting as both rivers have weapons-grade plutonium sources in their catchment areas, including the Russian plutonium production and reprocessing plants at Mayak, Tomsk-7 and Krasnoyarsk, and the Semipalantinsk nuclear weapons testing site in Kazakhstan (Figure 13), and together represent the largest input of freshwater to the Arctic area. Hence, measurement of the plutonium-isotope ratios offers both a means of identifying the origin of radionuclide contamination and the influence of the various nuclear installations on inputs to the Arctic, as well as a potential method for following the movement of water and sediment loads in the rivers. The river systems represent a source of a variety of pollutants to the Arctic and information on their environmental status and behaviour is of interest to various scientific disciplines.

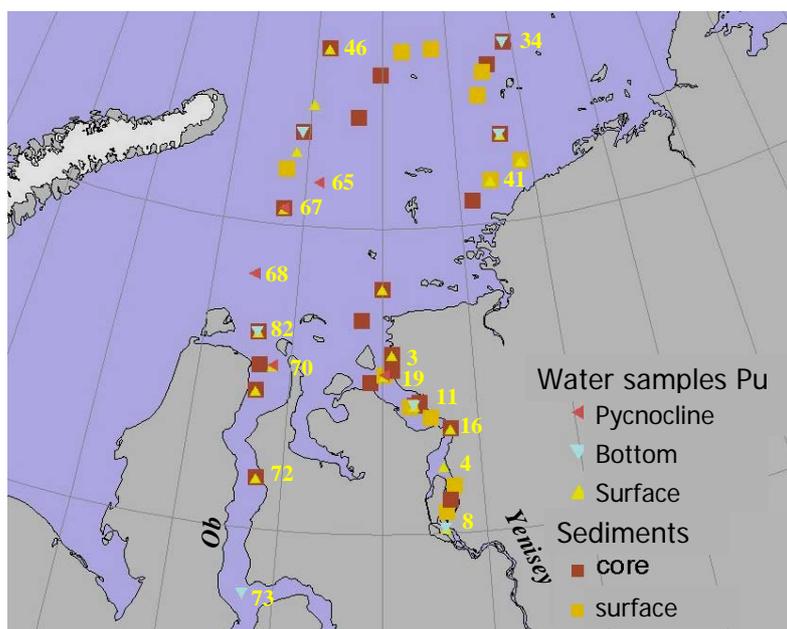


Figure 13. Sampling sites in the Kara Sea and the Yenisey and Ob Rivers during the Kara Sea expedition 2001 with RV "Akademik Boris Petrov". Water samples included in the present work are indicated with the station number.

A total of 50 surface sediments have been measured from the various sites in Yenisey River, Ob and Yenisey Estuaries and Kara Sea from the "SIRRO 2001" expedition. The data indicated a clear influence from a low $^{240}\text{Pu}:^{239}\text{Pu}$ source in surface sediments collected from the Yenisey Estuary. Previous studies have indicated similar results, although based on much fewer samples than the present study (Beasley et. al 1995, Oughton et. al. 1999a). The results also show an increase in plutonium concentration and a decrease in isotope ratio going upstream from the estuary. The sample collected from the site closest to the Krasnoyarsk site having the highest concentration (11.02 ± 0.09 mBq/g $^{239}, ^{240}\text{Pu}$) and lowest observed $^{240}\text{Pu}:^{239}\text{Pu}$ isotope ratio (0.049 ± 0.001). Measurements of plutonium isotope ratios in Kara Sea and Ob sediments gave no evidence for significant non-global fallout sources, which is in agreement with published data (Sayles etc), again on a much lower number of samples. Finally it should be stressed that the concentration level of plutonium in both estuaries is low, and many orders of magnitude below that reported in the river catchment sites close to the nuclear installations.

Tangential Flow ultrafiltration combined with AMS measurements have been used to study the size distribution, in particular the colloidal fraction, of ^{239}Pu and ^{240}Pu in the Arctic rivers, Ob and Yenisey, and their estuaries. For size fractionated samples with ultra low Pu activity concentrations, AMS has proved to be most useful. The results should improve the

understanding of plutonium speciation and processes in estuaries, and be of relevance for impact assessments in case of accidental releases from the nuclear installations in the Urals and Siberia.

The activity concentration of $^{239,240}\text{Pu}$ in the Ob and Yenisey river waters varied within 6-43 mBqm^{-3} , somewhat higher than in the open Kara Sea (4-11 mBqm^{-3}). Results indicate that the major fraction of Pu in the estuarine waters was present in dissolved form ($<0.45 \mu\text{m}$), while the particulate fraction of Pu was high in Ob and Yenisey river waters probably reflecting the high load of suspended particulate material (Fig. 14).

The colloidal fraction in the rivers ranged from 29 – 48% (River Yenisey) and 29 – 73% (River Ob) whereas about 50% of the Pu in Kara Sea water influenced by the river Ob was associated with colloids.

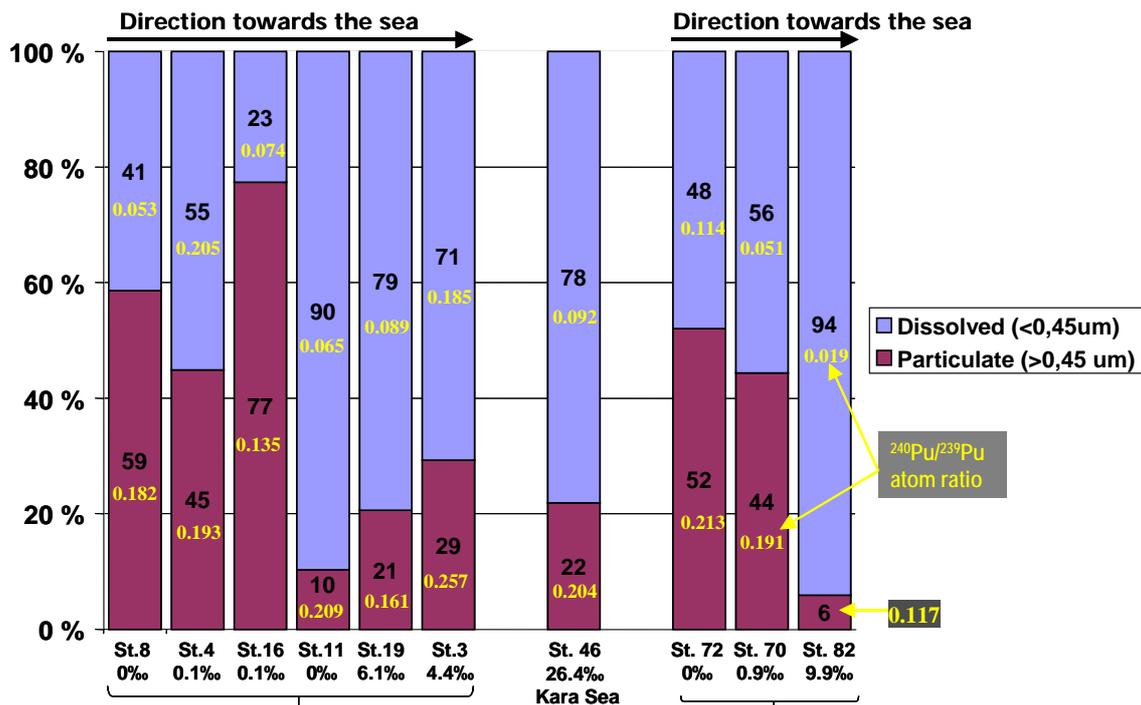


Figure 14. Distribution (%) of $^{239+240}\text{Pu}$ between particulate and dissolved forms in surface water collected in the Kara Sea and the Yenisey and Ob Rivers. $^{240}\text{Pu}/^{239}\text{Pu}$ atom ratio (yellow) indicated for each fraction.

Furthermore, the AMS results demonstrate that the $^{240}\text{Pu}/^{239}\text{Pu}$ atom ratios varied within the samples, being rather low for the dissolved phases (colloids and LMM species) and significantly higher for the particulate fraction. Thus, particulate Pu is apparently derived from global fallout, while colloidal and LMM Pu has another origin.

For both rivers the various nuclear installations are more than 2000 km from the estuaries potential weapon's grade sources. The Ob River has more sources than the Yenisey, and higher concentrations at source (i.e., Mayak), thus it is interesting that it should be the Yenisey that shows the non-fallout influence in sediments. Some possible explanations can be; stronger turbulence caused by a reduction of flow speed as the Ob widens into the Ob Bay and the Yenisey into the southern Kara Sea probably accounts for most of the sedimentation in these two areas, flocculation and coagulation due to fresh and salt water mixing adding to the deposition (Gebhardt *et al.*, 2003). Very little sedimentation of suspended matter from the Yenisey seem to occur until it reaches North of Sibiryakova Island (Dittmers *et al.*, 2003). AMS measurements of sediment cores from the main deposition area North of Sibiryakova Island show that the Pu deposited there is mainly derived from global fallout with $^{240}\text{Pu}/^{239}\text{Pu}$ atom ratios of 0.16 ± 0.01 (Skipperud *et al.*, 2003). Outside the deposition area South of

Sibiryakova Island, however, the $^{240}\text{Pu}/^{239}\text{Pu}$ atom ratios of surface sediments were significantly lower ranging between 0.09 – 0.11 in the estuary and 0.05 – 0.13 in the river (freshwaters). This seems consistent with the observed isotope ratios for particulate Pu expected to constitute most of the sedimented Pu. In the Ob River, however, the sedimentation takes place all the way along the Ob Bay, through the river mouth and north to 76°N (Gebhardt *et al.*, 2003). All the AMS measurements of the Ob Bay and adjacent Kara Sea station sediment cores reported by Skipperud *et al.* (2003) showed Pu to be global fallout derived.

Work Package 4:

Objectives

To assess bioavailability of artificial radionuclides which have been remobilised from sediments.

Participants: RPII, NRPA, AUN, RISOE, IFE, CEFAS

The overall objective of WP4 was to assess the impact of remobilisation on bioavailability, by means of laboratory experimentation and by investigating the degree of intra-species variation on both regional and time-dependent scales. The focus was to use concentration factors (CF: Bq kg⁻¹ biota/Bq l⁻¹ seawater) as a common measure of bioavailability. The problem to be addressed was whether uptake by biota of artificial radionuclides remobilised from aquatic sediments may be significantly different from uptake in the past when the radionuclides were originally introduced. In many regions of Europe artificial radionuclides present in the sediments of the seabed, lakes, deltas and fjords - as a result of past inputs from a variety of sources (e.g. fallout, Chernobyl accident, reprocessing wastes) - represent a significant actual or potential source to aquatic ecosystems. The extent to which remobilised radionuclides behave in a similar manner, with respect to uptake by biota, to the original inputs is poorly known. For this reason the potential dose consequences to the human population cannot be predicted with certainty.

Progress and Results

Full details of the main progress and results in WP 4 are provided in Deliverable 4.1 (Concentration Factors), and a number of publications (Kershaw *et al.*, in prep; Lindahl *et al.*, in press; Nawakowski *et al.*, in press; Strålberg *et al.*, in prep).

The WP was divided into 3 main tasks:

- 4.1 Estimation of concentration factors (CFs) based on field measurements.
- 4.2 Comparison of recent CFs with those from data archives, based on field measurements.
- 4.3 Mesocosm experiments to establish radionuclide uptake from contaminated sediments.

WP4.1 & 4.2 – Field-based observations

The aim of WP4.1 was to determine equilibrium CFs for a number of key radionuclide and biota combinations (e.g. Pu, Am, Cs, Tc; lobster, nephrops, crab, *Mytilus sp.*) from the Celtic Seas, North Sea, Norwegian Sea, Skagerrak and the Baltic, and examine the data for evidence of regional differences. In addition, an experimental cannulation procedure was developed to provide information of the internal distribution of radionuclides in fish. Despite the large data set amassed (see Annex in Deliverable 4.1), no statistically-significant regional differences in CFs, for identical radionuclide-biota pairs, were found, other than for the special case of the Kattegat where significant salinity gradients do effect uptake of ^{99}Tc and ^{137}Cs by *F. vesiculosus* (Lindahl *et al.*, 2003).

The aim of WP4.2 was to establish whether CFs for certain biota-radionuclide pairs have changed over time, particularly in areas like the eastern Irish Sea where remobilisation has been demonstrated. In addition, the data were examined for evidence of inter-annual variability due to seasonal effects and other environmental factors. In most cases no

statistically significant trends were observed, despite datasets covering several decades. In only 2 cases could a trend be shown: ^{137}Cs in winkles and crabs from the eastern Irish Sea (Fig 4.1), over a period of 40 years. Some of the variability of ^{99}Tc in *F. vesiculosus* appeared to be due to seasonal patterns of growth, but it was difficult to quantify this effect (Nawakowski et al., in press).

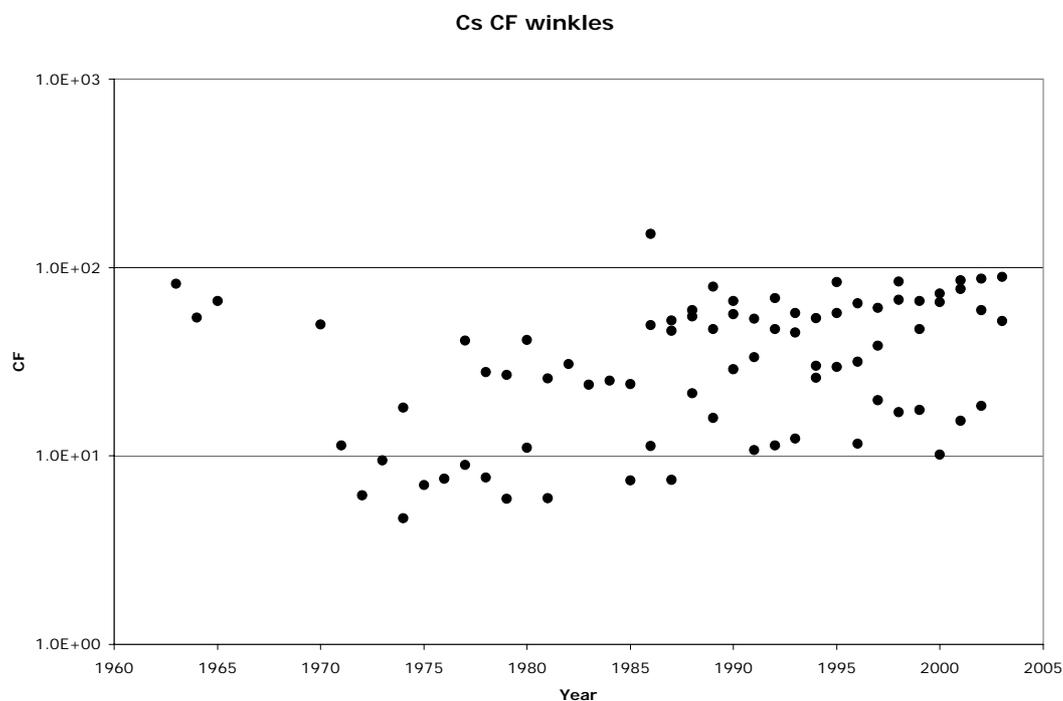


Figure 4.1 CF for ^{137}Cs in winkles from the eastern Irish Sea, 1962-2003

4.3 Mesocosm-based experiments

The aim was to use sediment cores from areas of significant contamination (e.g. Rhone delta, Irish Sea, Norwegian fjords) to conduct remobilisation and uptake experiments, under controlled conditions, in specially designed mesocosms. Experiments were designed to assess the influence of changes to the redox conditions and physical disturbance on the release of radionuclides from the sediment, using a set of primary chambers. The remobilised radionuclides then passed to a second set of chambers containing *Littorina littorea*, and *Fucus vesiculosus* for the degree of uptake to be monitored, and compared with the field-based observations. The sediment was obtained from the Irish Sea, contaminated by discharges from BNFL Sellafield. All experiments were performed using unfiltered deep seawater samples (salinity 34-35 ‰) from near Drøbak in the Oslo Fjord. The experimental and analytical methods used and results of the biota uptake experiments are described in more detail in Strålberg et al. (in prep.).

Sediment diffusion and disturbance experiments were performed with the aim of exploring how much plutonium and caesium were made available for transport over a longer distance under different conditions, using unfiltered water samples to derive K_t (transport coefficient). Passive diffusion experiments were carried out using both homogenised sediment (5cm deep) and sediment cores (diameter 6 cm, 10 cm deep) with contact times of 4 and 10-26 weeks respectively. All uncertainties are given as one standard deviation.

Table 4.1. Passive diffusion using undisturbed sediment cores.

	Sediment concentration (Bq kg ⁻¹ d.w.)	Water concentration (Bq litre ⁻¹)	K _t (litre kg ⁻¹)
¹³⁷ Cs – location #75	114 ± 11	-	-
¹³⁷ Cs – location #78/79	282 ± 20	0.45 ± 0.11	6.3 · 10 ²
^{239,240} Pu – location #75	57 ± 5	0.0019 ± 0.0006	3.0 · 10 ⁴
^{239,240} Pu – location #78/79	253 ± 18	0.0036 ± 0.0007	7.1 · 10 ⁴

A third set of experiments simulated “storm” disturbance using a pump to generate water circulation near the bottom. The experiments were running for 4 days, and the water samples were collected 2 hours after the “storm” had ended so that the largest particles had resettled and only the finer particles that are likely to be transported over larger distances were kept in the water column.

Table 4.2. Simulated storm disturbance using homogenised sediment.

	Water concentration (Bq litre ⁻¹)	K _t (litre kg ⁻¹)
¹³⁷ Cs – location #75	-	-
¹³⁷ Cs – location #78/79	1.6 ± 0.4	3.9 · 10 ²
^{239,240} Pu – location #75	0.083 ± 0.009	9.2 · 10 ²
^{239,240} Pu – location #78/79	0.125 ± 0.017	2.8 · 10 ³

In a fourth set of experiments “trawler” disturbance was simulated by shaking the sediment with seawater, and after a contact time of 24 hours the sediment was allowed to settle for 2 hours before the seawater was removed.

Fresh sediments from the Esk estuary in the Irish Sea were collected for the biota experiments. The activity concentrations in these sediments were higher than in those used in the diffusion experiments and are shown in Table 4.3. Samples of *F. vesiculosus* and *L. littorea* were collected from a site north-west of Fredrikstad in southern Norway. The sediment and biota samples were kept in different chambers connected through a closed loop for water circulation. The set-up consisted of 3 parallel units; all located in a climate-controlled room at 10-12 °C (Fig. 4.2). Water samples were collected from the chambers after an exposure time of 16 days. The activity concentrations are shown in Table 4.3. Portions of the biota species were removed from the chambers after different exposure times. All results are shown in Table 4.4 and represent average and one standard deviation of the three parallel units. Reference values (background concentrations) are given for comparison. Only the soft parts of the winkles were used in the analyses. Average ratio between dry and fresh weight of the soft parts of the winkles was 0.21 ± 0.01 (one standard deviation).

Of the 4 radionuclide-biota pairs, only ¹³⁷Cs in *L. littorea* had reached apparent equilibrium within 46 days (Fig 4.3). The CF obtained was within the IAEA recommended range. It is not possible to directly compare the other pairs, due to lack of equilibrium.

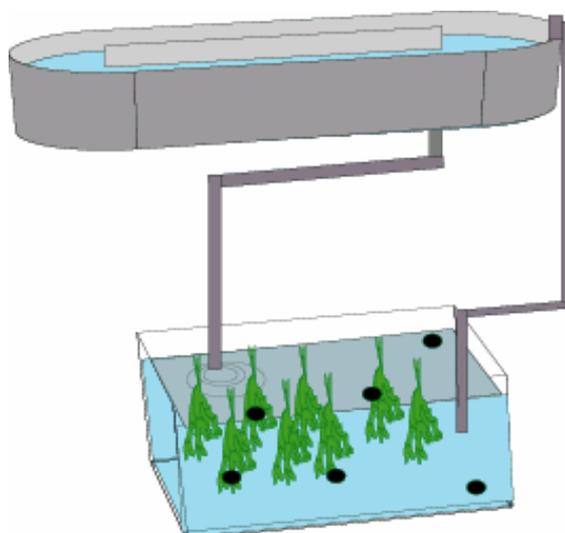
Table 4.3. Activity concentrations in homogenised sediment and water (collected after 16 days) used in the biota experiments. All uncertainties are given as one standard deviation.

Nuclide	Sediment concentration (kBq kg ⁻¹ d.w.)	Water concentration (Bq litre ⁻¹)
¹³⁷ Cs	4.21 ± 0.24	3.71 ± 0.05
^{239,240} Pu	3.23 ± 0.23	0.016 ± 0.004

Table 4.4. Biota experiments. All uncertainties are given as one standard deviation.

Species	Days of exposure	¹³⁷ Cs (Bq kg ⁻¹ d.w.)	^{239,240} Pu (Bq kg ⁻¹ d.w.)
<i>Fucus vesiculosus</i>	Reference	< 3	0.21 ± 0.03
	2	22.3 ± 2.7	12.0 ± 1.5
	7	36 ± 5	22 ± 5
	14	55 ± 10	28 ± 6
	23	71 ± 4	37 ± 4
	46	93 ± 11	68 ± 13
<i>Littorina littorea</i>	Reference	< 3	1.1 ± 0.9
	2	26.4 ± 2.2	6.8 ± 0.5
	7	58 ± 7	5.8 ± 1.0
	14	79 ± 6	6.6 ± 0.4
	23	74 ± 5	10.8 ± 1.7
	46	76 ± 10	17.5 ± 1.9

Figure 4.2. Experimental set-up.



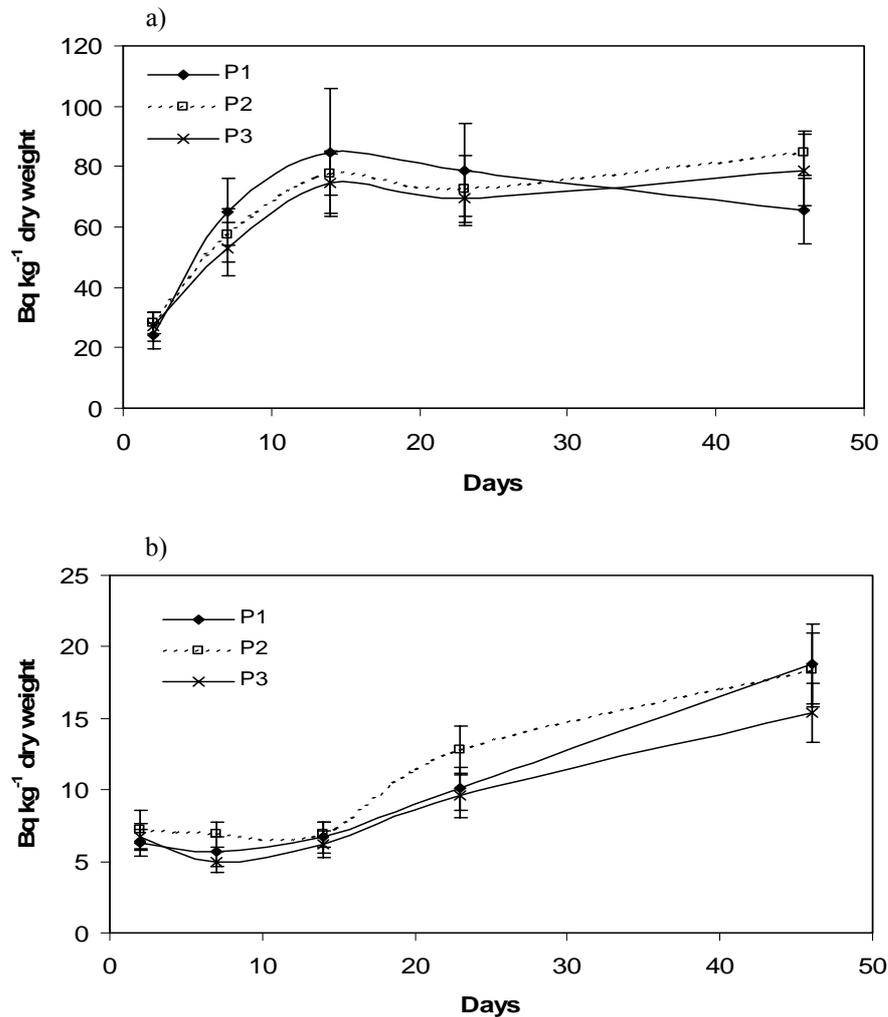


Figure 4.3 Uptake of ¹³⁷Cs (a) and ^{239,240}Pu (b) in *Littorina littorea*. Zero level values are given in Table 4.3.

Additional experimental studies – cannulation technique

Uptake studies in fish show that the uptake of radionuclides depends on the speciation of radionuclides and that the interaction with organic materials in waters changes the bioavailability of radionuclides. Thus, well-defined radioactive tracers were injected into rainbow trout using a cannulation technique (Fig. 4.4), to study the internal distribution in fish. Exposure time was 27 hours. All the injected ^{99m}TcO₄⁻ was found in spleen (52%) and kidneys (40%), while traces were found in the brain and the eyes (Fig. 2) (Heier et. al. 2002).

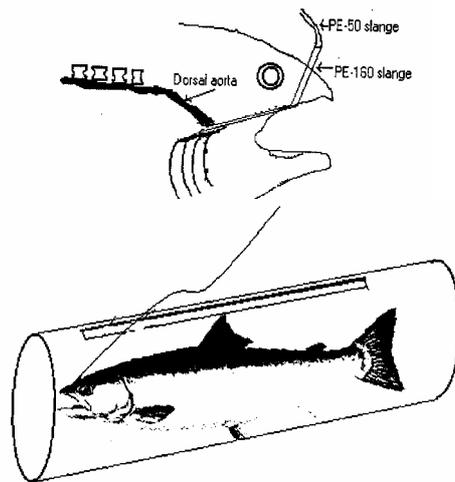


Figure 4.4: Cannulation of fish, with minimal stress to the fish.

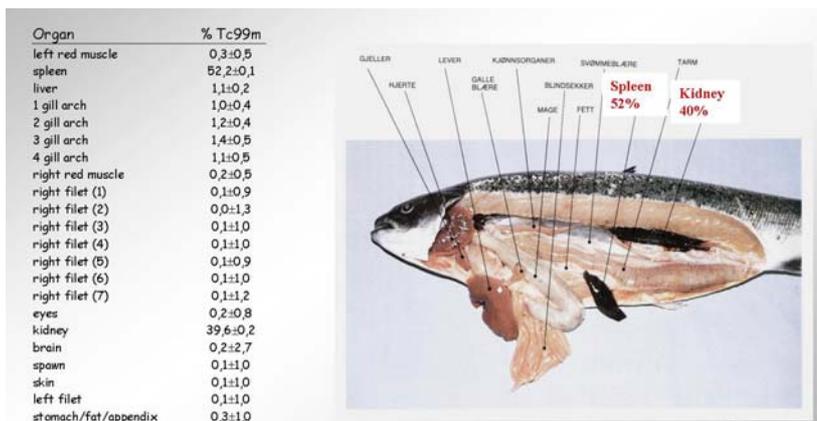


Figure 4.5: Distribution of Tc-99 in fish 27 hours after cannulation.

$\text{Pu}^{\text{V,VI}}$ have also been injected into rainbow trout using the same cannulation technique, to determine the distribution of Pu in selected organs. The results (Table 4.5) show that $\text{Pu}^{\text{V,VI}}$ species are predominantly retained in liver, gills and kidney, when looking at soft tissue. It is interesting that a certain fraction can be observed in the filet.

Table 45. Distribution of Pu in fish, soft tissue, 9 days after cannulation.

Organs	% distribution
Blood	0.70 ± 0.03
Gills	0.8 ± 0.1
Liver	1.0 ± 0.4
Bile	0.07 ± 0.02
Spleen	0.10 ± 0.02
Rowan	0.4 ± 0.2
Stomach	0.4 ± 0.2
Kidney	0.6 ± 0.2
Muscle/Filet	0.6 ± 0.4

Work Package 5:

Objectives

To improve the methodology for describing sediment-water interactions in numerical hydrodynamic compartment models.

Participants: RPII, NRPA, AUN, RISOE, CEFAS, UCD, USEV

The main objectives of Work-package 5 were to improve the methodology for describing sediment-water interactions, with special regards to studying radionuclide remobilisation mechanisms and to estimate how an improved description of dispersion processes would influence the predicted contamination of sea areas. To attain these ends, it was necessary to improve model modules and update information for the key parameters on the basis of information for parameterisation, validation and corroboration of the models as well as to compare the predictions using different approaches for modelling and model tools.

3. Progress and Results

Detailed information about the main progress and results in WP 5 are provided in Deliverables 5.1 (Radionuclide dispersion models for Arctic, Atlantic and Mediterranean seas. Estimation of radiological sensitivity of marine areas), and in a number of publications, which are listed below.

The WP was divided into 3 main tasks:

- 5.1 Development of model tools based on improved information provided during the project.
- 5.2 Development and improvement of key modules of the models.
- 5.3 Sensitivity evaluation of radionuclides transport, uptake and doses to man for improvement of key modules.

WP5.1 & 5.2 – Development and improvement of model tools and key modules

The radionuclide interaction processes occurring between the dissolved phase and sediments have been studied by means of numerical modelling. A set of models with different time and space resolution have been developed and improved under the course of the REMOTRANS project for the following regions: the Irish Sea (a high resolution 3D hydrodynamic model that can explicitly solve tidal mixing, and the COSE model which resolves processes operating on tidal time scales), the English Channel (a long-term dispersion model based upon averaged circulation in the Channel), the Odiel-Tinto Estuary (a high resolution 2D hydrodynamic model), the Rhone River mouth (a high-resolution 3D baroclinic model that

solves water circulation, salinity changes and dispersion of suspended matter and radionuclides in the river plume), and the North Atlantic and Arctic Oceans including the North-West European coastal areas (the model is based on the modified approach for box modelling, which includes time-dependent dispersion of radionuclides i.e. non-instantaneous mixing in oceanic space).

In parallel with the use of different models, different approaches for the treatment of water-sediment interactions have been under consideration. Hydrodynamic models have been used to carry out testing and evaluation of the 1- and 2-step kinetic models (Fig. 5.1) for water-sediment interactions (two reversible models and one irreversible was additionally used for the English Channel). It was found that the transfer of radionuclides from the liquid to the solid phase depended on the amount of particle surfaces per water volume unit, which can be written as a function of particle size and density, among other parameters.

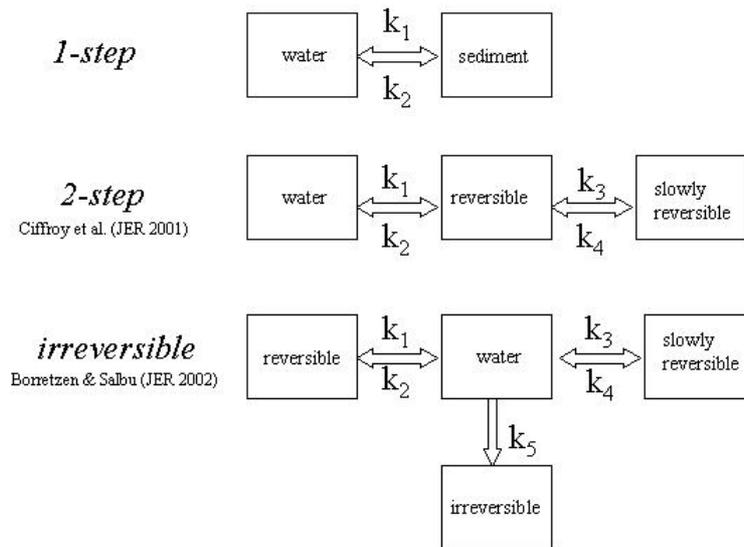


Figure 5.1. Kinetic models included in the hydrodynamic dispersion models

When using the box model, it is assumed that, at any given time, the activity in the water column is partitioned between the water phase and the suspended sediment material. Activity associated with suspended sediments is lost to the underlying boxes when particles settle out. The model also includes the diffusivity processes of radioactivity through the pore water, resuspension, mixing due to bioturbation and burial of activity into a deep sediment layer (regarded as a sink) based on Fick's law. Under the course of the project, two approaches for treating of water-sediment interactions have been under consideration with the following differences: (i) the resuspension rate describing radionuclide remobilisation is defined as an depth-dependent empirical expression based on an equilibrium assumption between deposition and remobilisation for the whole oceanic space and (ii) the description of radionuclide remobilisation from the seabed is linked to the sediment reworking and porewater turnover rates. In addition, the influence of using a time-dependent laboratory derived sediment-seawater distribution coefficient (K_d) for dispersion of radionuclides has been assessed when modelling radionuclide discharges into the Ob Bay (the Kara Sea).

A detailed description of models may be found in Deliverables 5.1.

Examples of simulation results are shown on Figures 5.2 – 5.6.

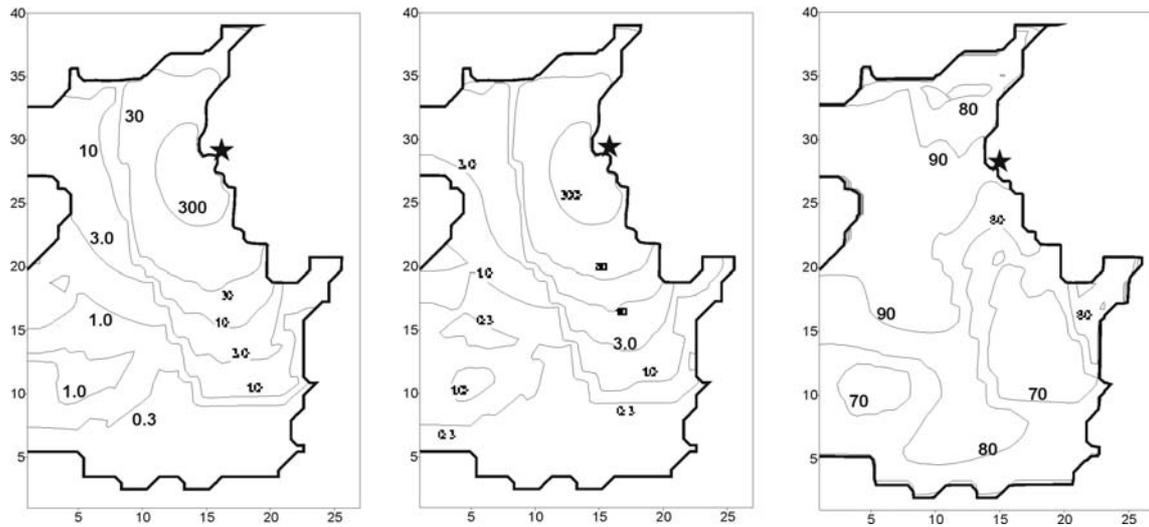


Figure 5.2. Examples of the computed distributions of $^{239,240}\text{Pu}$ in bed sediments (kBq/m^2) for year 1977 with the 1-step (left) and 2-step (centre) models. The star is Sellafield reprocessing plant. The map on the right is the computed fraction of reduced Pu in bed sediments (the same result with both models)

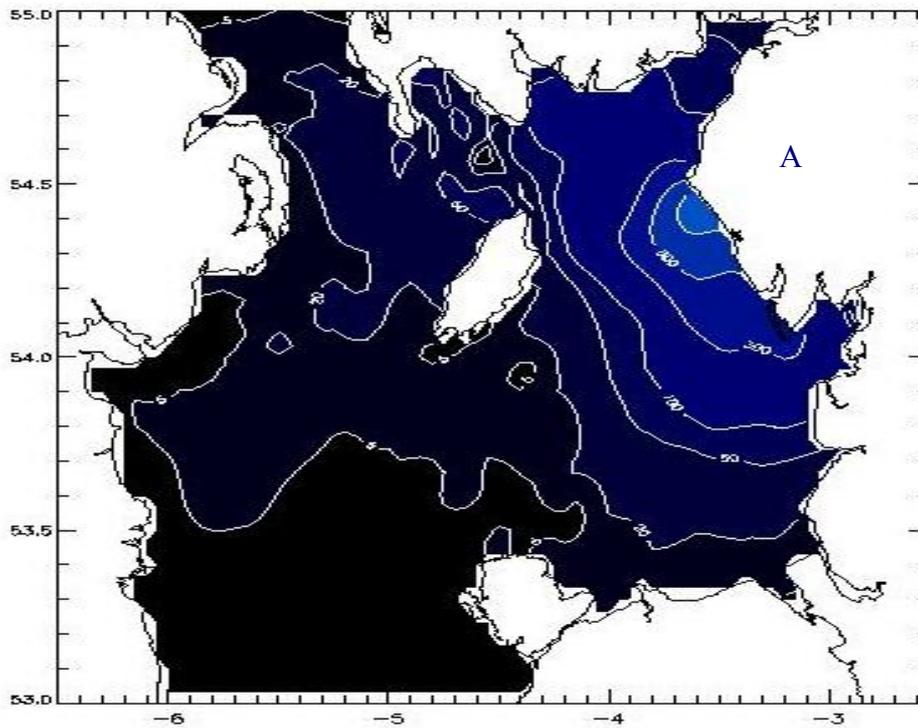


Figure 5.3. $^{239/240}\text{Pu}$ sediment concentrations top 5 cm (Bq/Kg). A) Model

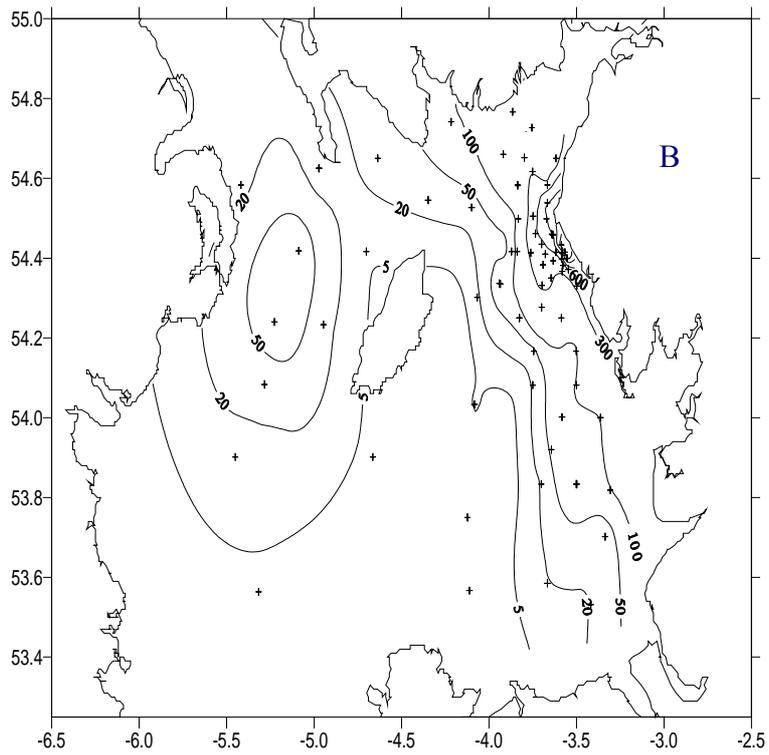


Figure 5.3. $^{239/240}\text{Pu}$ sediment concentrations top 5 cm (Bq/Kg). B) Observed

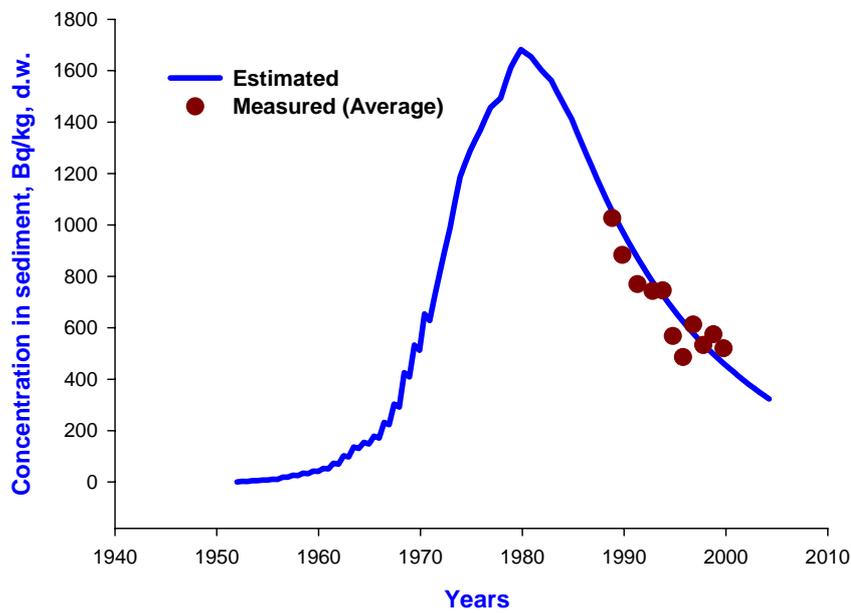


Figure 5.4. Comparison between model simulation and measurements for the Cumbrian waters for $^{239,240}\text{Pu}$ discharge into the Irish Sea

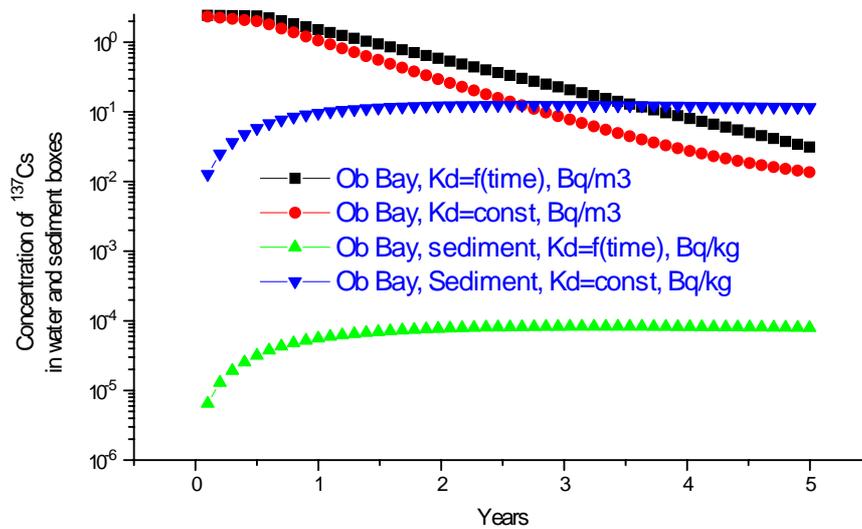


Figure 5.5. Influence of a time-dependent sediment distribution factor on dispersion of ^{137}Cs in the Ob Bay (the Kara Sea)

Simulation results for the models used in the project are generally in sufficient agreement with observations. Thus, it seems that the main processes included in the models have been adequately described. From this point of view, it is interesting to compare calculation results for the 3D hydrodynamic and box models for the Cumbrian Waters (the East Irish Sea), which are shown in Table 5.1.

Table 5.1. Comparison of the simulation results for 3D hydrodynamic and box models.

Time, years	^{137}Cs concentration in sediment, Bq kg ⁻¹ d.w.		$^{239,240}\text{Pu}$ concentration in sediment, Bq kg ⁻¹ d.w.	
	3D model	Box model	3D model	Box model
0.5	4.2	1.6	2.5	7.1
1.0	3.6	1.6	2.5	6.8
1.5	3.0	1.5	2.5	6.6
2.0	2.6	1.4	2.5	6.4

Simulations correspond to a 1 TBq discharge of radionuclides into the water column. The results for the 3D model in Table 5.1 are obtained by averaging the spatial activity concentration over an area corresponding to that of the actual box in the box model. Table 5.1 shows that in spite of different modelling approaches and description of water-sediment interaction processes, differences for this simulation are not more than a factor of three.

Figure 5.6 demonstrates some of the complexities encountered when modelling water-sediment interactions. Using the box model, the figure shows a sensitivity analysis examining how the dispersion of radionuclides in the Cumbrian Waters (CW) and the North part of the Irish Sea (ISN) depends on the sediment reworking rate (R_w). Calculations correspond to the discharge of 1 TBq ^{239}Pu into the water column in the Cumbrian Waters. The results show clearly that the radionuclide concentration in solid and liquid phases can decrease as well as increase with an increasing reworking rate, depending on time and distance from the contamination source. Due to this complicated behaviour, the parameterisation process of the models is not quite trivial.

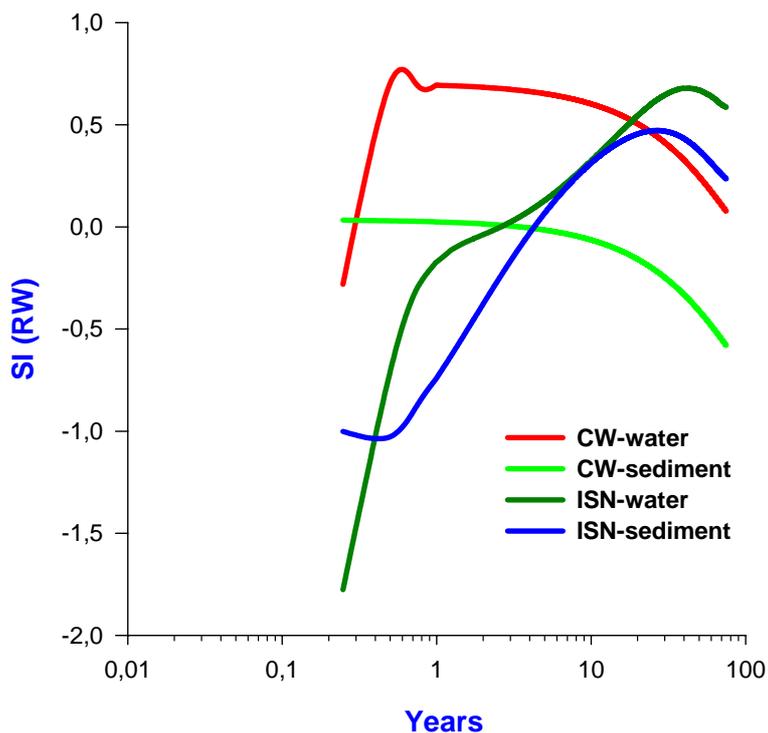


Figure 5.6. Local sensitivity analysis: relative dispersion of ^{239}Pu in the Irish Sea regions versus relative sediment reworking rate

WP5.3 - Evaluation of the radiological sensitivity

During the course of the REMOTRANS project, the box model was improved and updated with the main goal to provide improved radiological assessments for the marine environment, to be able to better evaluate the sensitivity of different sea areas regarding radionuclide discharges. In particular, the key modules, as well as the box structure, for the Arctic Ocean, the Nordic Seas and the North Atlantic were improved on the basis of new knowledge about Polar, Atlantic and Deep waters.

Calculations results presented in Figure 5.7 show clearly that coastal marine waters are most sensitive for the present scenario of radionuclide releases. More detailed information about evaluation of the radiological sensitivity of the marine environment can be found in Deliverable 5.1.

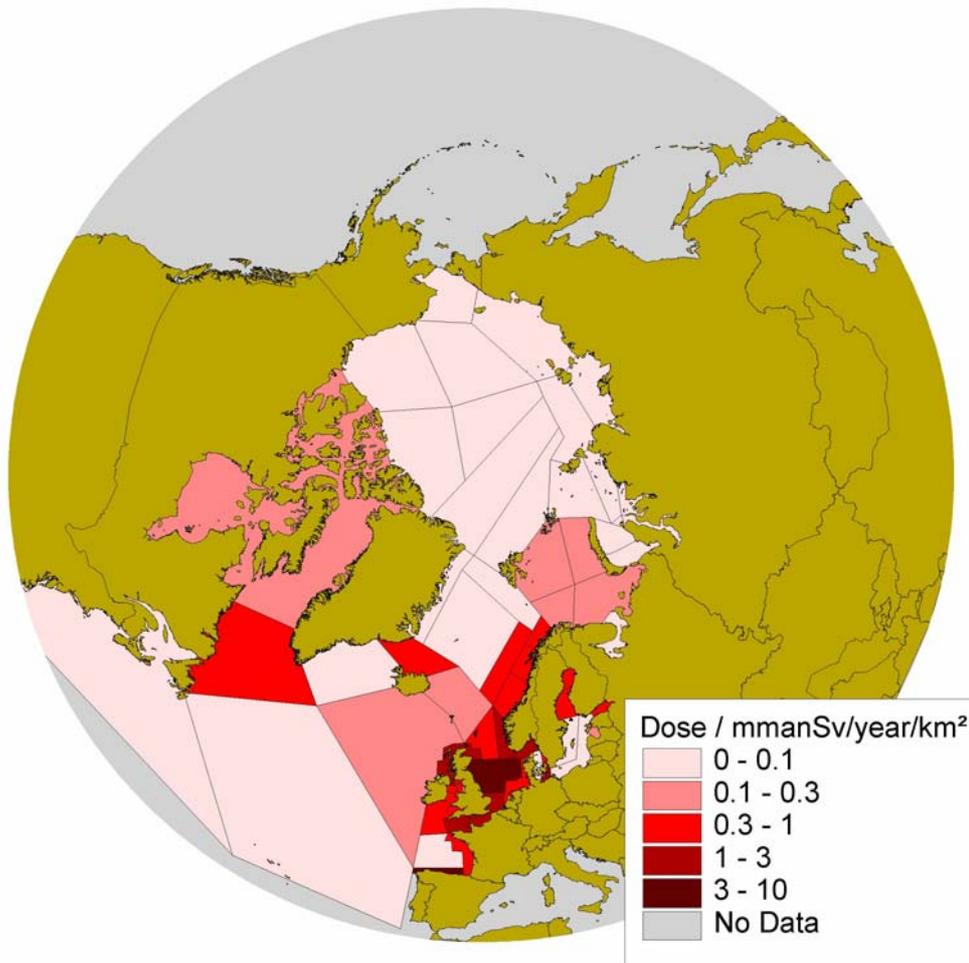


Figure 5.7. Collective dose rate after 10 years from different marine regions per 1 km² after a uniform deposition of ¹³⁷Cs at 1 kBq m⁻²

3. Deliverables

Work Package 1: Inventories

D1.1 Inventory Database. Contributors: C. McMahon (Pn 3, RPII, WP leader); P. Kershaw (Pn 8, CEFAS); D. Boust, S. Charmasson and M. Arnaud (Pn 2, IRSN); A.-L. Rudjord (Pn 4, NRPA); E. Strålberg and G. C. Christensen (Pn 7, IFE); L. León Vintró and P. Mitchell (Pn 9, NUI-UCD); P. Roos and H. Dahlgaard (Pn 6, RISØ); C. Papucci and R. Delfanti (Pn 10, ENEA); C. Gascó (Pn 11, CIEMAT).

Work Package 2. Time series

D2.1. Report of time series results on radionuclide concentrations in water, indicator species and sediment cores. Contributors H. Dahlgaard (Pn 6, RISOE, WP leader), E. Holm (Pn 1, ULUND), D. Boust S. Charmasson, (Pn 2, IRSN), C. McMahon (Pn 3, RPII), A.-L. Rudjord (Pn 4, NRPA), E. Strålberg and G. C. Christensen (Pn 7, IFE), P. Kershaw (Pn 8, CEFAS), L. L. Vintró and P. Mitchell (Pn 9, NUI-UCD), Carlo Papucci and Roberta Delfanti (Pn 10, ENEA), C. Gascó (Pn 12, CIEMAT).

Work Package 3: Processes, chemical forms, speciation and remobilisation

D3.1. Report on the physico-chemical speciation of radionuclides in the water column and sediments and implications for remobilisation, prepared by L.León Vintró, P.I. Mitchell, J.A. Lucey, D. Boust and A. Gouzy (2003).

Work package 4. Bioavailability

D4.1 Concentration Factors. Contributors: P. J. Kershaw (Pn 8, WP leader); E. Holm & P. Lindahl (Pn 1, Lund); M. Masson (Pn 2, IRSN); C. McMahon (Pn 3, RPII); A.-L. Rudjord (Pn 4, NRPA); B. Salbu and L. Skipperud (Pn 5, AUN); E. Strålberg and Gordon C. Christensen (Pn 7, IFE) ; Torleif Bækken (Pn 7, NIVA); C. Nawakowski, M. D. Nicholson, C. Smedley and J. Barry (Pn 8, CEFAS). 51pp.

Work Package 5: Modelling

D5.1 Radionuclide dispersion models for Arctic, Atlantic and Mediterranean seas. Estimation of radiological sensitivity of marine areas. Contributors: M. Iosjpe (Pn 4, WP leader), C. McMahon (Pn 3, RPII), B. Salbu & L. Skipperud (Pn 5, AUN), P. Roos and H. Dahlgard (Pn 6, RISOE), P. Kershaw (Pn 8, CEFAS), L. Vintró and P. Mitchell (Pn 9, NUI-UCD), M. Garcia-Leon, R. Perriñez (Pn11, USEV).

5. Discussion

Work Package 1: Inventories

A considerable body of data has been generated and compiled in the course of the Remotrans project. Areas such as the northern Adriatic Sea, the Rhone Delta, the western Irish Sea and the Gulf of Cadiz, where only limited work had been done to take stock of the radionuclide inventories in the underlying sediments, have now been comprehensively examined. The size of the secondary sources potentially available for remobilisation in the five marine zones studied has been better quantified through a number of collaborative studies within the REMOTRANS project.

As examples of the data generated, in the western Irish Sea mud patch $^{239,240}\text{Pu}$ inventories in the range 350 – 36000 Bq m⁻² were measured. It was calculated that 20 TBq of $^{239,240}\text{Pu}$ and 120 TBq of ^{137}Cs are currently deposited in the western Irish Sea mud patch. These data were obtained on the basis of measurements on 23 sediment cores in conjunction with the sedimentological information. In all but one of the sediment cores, the bottom five centimetres of the core corresponded to less than 10% of the total inventory activity, indeed, less than 1% in the majority of the cores. In the Rhone Delta, the decrease in ^{137}Cs inventory with time was evident, while for $^{239,240}\text{Pu}$ the inventories have remained relatively constant over the ten-year time period.

The database generated (Deliverable 1.1) brings together the results of a number of sediment contamination studies, including REMOTRANS, in one easily accessible interactive package. This database is also being shared with the IAEA and its member states through inclusion in the GLOMARD database.

Work package 2. Time series

During 1970ies and to 1986 (the Chernobyl accident) the major source for ^{137}Cs and ^{99}Tc in Nordic waters was the Sellafield reprocessing plant. The process change in the plant resulted in much lower releases of radiocaesium but much higher levels of ^{99}Tc . The major source for radiocaesium became outflow from the Baltic Sea (about 60 TBq/year). The Baltic Sea has always contained relatively high levels of radiocaesium and radiostrontium due to that it is a marginal sea with long residence time for dissolved substances. Transit times from Sellafield of ^{99}Tc , which is supposed to behave conservatively, to for example the Swedish west coast

has been calculated. This shows that there is no significant difference if one compares the pulse of ^{99}Tc in 1978 with the releases 1994 -2001.

To explain the balance of radiocaesium in the Baltic Sea a remobilisation from the sediments must take place.

The seasonal variations in *Fucus sp.* with maxima in winter and minimum in summer for ^{99}Tc are difficult to explain. This is the opposite behaviour to radiocaesium. The concentration factor related to salinity is also the opposite for Tc compared to Cs. This can be part of the explanation since during summer, water with lower salinity flows out from the Baltic Sea and during autumn winter weather with low pressure dominate and this is the period when the Baltic Sea is ventilated with more saline water from the North Sea. However similar, if not so strong, seasonal effects are also shown along the Norwegian coast which can not be explained in the same way.

The results for the Rhone prodeltaic area is that the remobilisation is subject to resuspension phenomena during intense meteorological events. The remobilisation in shallow areas of the Baltic Sea is probably of the same nature. The relative part which is shallow is also much larger for the Baltic Sea than the Mediterranean. The run off from rivers, carrying sedimentary materials and radionuclides associated with colloids is also much greater for the Baltic Sea

The analysis of the time trend of the vertical profiles of ^{137}Cs in sediment cores of the Northern Adriatic Sea, together with sediment dating on different time-scales, allowed us to elucidate the mechanisms of re-distribution of the radionuclides exported by rivers in the Northern Adriatic Sea. Three different areas were identified:

The prodelta of the Tagliamento, where sediments transported by the river are rapidly accumulated and only temporarily stored. Intense meteorological events can totally re-suspend recent sediment and associated radionuclides and transport them to other marine areas.

The inner gulf of Trieste, where sediments transported by the Isonzo river are regularly accumulated and ^{137}Cs is stably stored.

The prodelta of the Isonzo, where sediments and associated radionuclides rapidly accumulate. Although this area is as shallow as the Tagliamento prodelta, the high sediment load transported by the river and the geographical location, more sheltered from the prevailing winds, allow a relevant net sediment accumulation and the permanent storage of the associated radionuclides.

The results obtained by the for the Tinto and Odiel Rivers estuary show that the presence of natural radionuclides released by the fertiliser plants is decreasing slowly both in sediments and river water after the stop of the discharges. It seems clear that, nowadays, the radionuclides deposited into the sediments are being remobilized to the river water. The whole process deals to a self-cleaning of the estuary, as can be deduced from the lower radionuclide concentrations and the activity ratios. However, there are still some radionuclide sources to the rivers: the former phosphogypsum piles, not protected against the meteorological phenomena, and the possible accidents, as the one that took place in 1999 at the new pile.

This work presents future perspectives. In concrete, it would be interesting to analyse the activity concentration in suspended matter in order to obtain more information about the remobilization processes. Apart from this, mathematical modelling about migrations of radionuclides today could be done, considering the high amount of data accumulated in our group.

Work Package 3. Processes, chemical forms, speciation and remobilisation

The overall objective of this work-package was to carry out a comprehensive study of the processes controlling radionuclide remobilisation from contaminated oxic and anoxic sediments, and to establish the influence of these processes on the long-range transport of remobilised radionuclides. This was achieved by carrying out by combining field and laboratory studies in a number of different environments under a widely varying range of environmental conditions (oxic/anoxic, temperate/arctic, depositional/unstable, etc.).

A substantial effort was focused on the development and testing of a sequential extraction analysis protocol specifically designed to study the solid partitioning of plutonium in anoxic sediments. An existing protocol was modified in order to (i) allow controlled dissolution of oxygen sensitive sulphide species in the first extraction step while preserving the reductive nature of the sediment during subsequent extractions (prior to the oxidisable extraction step), and (ii) avoid some of the pitfalls associated with this type of analysis. After a thorough testing and validation process, the method was applied to anoxic sediments collected in the course of a major collaborative campaign to the Cumbrian region of the eastern Irish Sea in July 2002. The results of these analyses indicate that plutonium bound to sediment in the Irish Sea is potentially more bioavailable than hitherto believed. Sequential extraction analyses showed that a large proportion of the plutonium in Irish Sea sediments is loosely bound to sites that readily exchange with seawater, readily oxidised upon air contact or easily dissolve upon changes in pH. Moreover, the data clearly indicate that diagenetic reactions taking place during the development of anoxic conditions influence the partitioning of plutonium in the solid phase.

Physical process (e.g., flood events, wind and wave action, storms) can be an important mechanism controlling remobilisation of sediment-bound radionuclides in certain non-stable environments, such as prodeltaic areas. Studies carried out in the Rhône prodelta in the course of this project suggest that these mechanisms are particularly important for this area, and that the main mechanism controlling resuspension events is wave action driven by winds blowing in a particular direction.

The well-known long-range transport of remobilised radionuclides from sediments in the Irish Sea and the North Sea was confirmed by the analysis of radionuclide concentrations and isotopic ratios in sediment cores collected in the Skagerrak area. The results of these analyses clearly show that, despite the reduction in direct discharges from the Sellafield reprocessing plant since the mid-1970s, highly-particle reactive radionuclides such as plutonium are still being transported from sediment deposits in the Irish Sea or the western North Sea following remobilisation.

Studies carried out in the Kongsfjord system (Svalbard) to study scavenging, sedimentation and resuspension processes at the glacier-sea interface, showed that scavenging processes are active and efficient both under summer and winter conditions. They also demonstrated that sediment accumulation rates decrease by up to two orders of magnitude from the inner to the outer fjord, with high sediment accumulation rates taking place close to the glacier-sea interface. Particles (and associated radionuclides) discharged from land (mainly by glacier melting) into the fjord are trapped in the inner part of the fjord. Although post-depositional remobilisation of sediments takes place within this area, there is no evidence of sediment transport to the outer fjord.

Studies were also carried out in Rørholtfjord (south-eastern Norway), an stratified lake containing an upper freshwater layer overlying a thin bottom layer of anoxic, saline water, trapped from the time the lake ceased to be connected to the sea. These studies have allowed an study to be made of differences in radionuclide mobility between oxic and anoxic environments. The vertical distribution of most trace elements and radionuclides in the water profile showed a significant enrichment in concentrations in the saline, anoxic layer. It is clear from these measurements that the anoxic conditions of the water, together with high concentrations of divalent iron and low molecular mass total organic carbon present in this layer, keeps scavenged radionuclides into solution, preventing them to reach the bottom sediment. As a result, lower sediment-water distribution coefficients are measured for both trace metals and radionuclides. Fractionation experiments of $^{239,240}\text{Pu}$ carried out under strictly controlled (anoxic) conditions showed that while most of the plutonium in the fresh water was associated to colloids, the fraction of plutonium in a colloiddally-bound form was practically zero.

Kinetic studies carried out using sediments of different composition under anoxic conditions showed that the achievement of pseudo-equilibrium in the distribution of a particular radionuclide between sediment and seawater is dependent both on the radionuclide in question and the type of sediment. These studies also confirmed that distribution coefficients

for anoxic marine sediments are lower, and in some cases much lower, than those measured for the same sediments under oxic conditions.

Finally, experiments carried out to investigate the remobilisation of radionuclides from contaminated freshwater sediments upon contact with seawater showed that a significant fraction of the total radionuclide content associated with the sediments can be remobilised within days following exposure to seawater.

Deviation from global fallout $^{240}\text{Pu}/^{239}\text{Pu}$ ratios can be identified in sediments from the Yenisey River and Estuary and also in water samples. An influence from low burn-up or non-civil sources is apparent in the Yenisey Estuary, whereas plutonium in the Ob Estuary sediments are dominated by global fallout. The data suggest that the plutonium-isotope “fingerprint” would be a useful method of following the migration and behaviour of both plutonium and sediments and particle loads in the two rivers, and further investigation will be carried out to obtain more information on the distribution of and mobility of plutonium associated with sediment samples.

Results show that the activity concentration of $^{239}, ^{240}\text{Pu}$ in the Yenisey and Ob rivers waters varied within 6 - 43 mBq m⁻³, somewhat higher than in the open Kara Sea (4 - 11 mBq m⁻³), where most of the Pu in the surface water samples collected in the freshwater of the Rivers Yenisey and Ob appears to be associated with particulate material while a majority of the Pu in both estuaries (mixing zones) is in a dissolved form. The colloidal fraction in the rivers ranged from 29 - 48% (River Yenisey) and 29 - 73% (River Ob) whereas about 50% of the Pu in Kara Sea water influenced by the river Ob was associated with colloids. AMS results indicate that the $^{240}\text{Pu}/^{239}\text{Pu}$ atom ratios are low for the dissolved phases (colloids and low molecular mass (LMM) species) and significantly higher for the particulate fraction. $^{240}\text{Pu}/^{239}\text{Pu}$ atom ratios show that particulate Pu is fallout related and that Pu in dissolved form originates from other low atom ratio sources (e.g. nuclear weapon production facilities).

Work Package 4: Bioavailability

The overall objective was to assess the bioavailability of artificial radionuclides which have been remobilised from contaminated sediments in the aquatic environment. This was achieved through a combination of controlled laboratory experiments and the analysis of environmental data. The focus was to use concentration factors (CF: Bq kg⁻¹ biota/Bq l⁻¹ seawater) as a common measure of bioavailability and compare CFs measured in situations where remobilisation was dominating with those obtained when the major source was direct releases to the water column from nuclear installations. Secondary aims were to establish whether regional differences in CFs occurred and to assess the extent of temporal variability.

In practise, the availability of environmental data is influenced both by the need to make radiological assessments in seafood and the usefulness of radionuclides as tracers of environmental processes. For the former, biota measurements are often made on the edible fraction. In some cases there may be significant differences in concentration, and hence CF, depending on whether particular organs are included (e.g. the lobster, *Homarus gammarus*, green gland) or whether the animal is allowed to depurate prior to analysis (e.g. winkle, *Littorina littorea*). In addition, there may be significant differences in CF between relatively closely-related organisms (e.g. Order Decapoda: *H. gammarus* and the edible crab *Cancer pagurus*), which may not be reflected in CF compilations based on broader classifications (e.g. IAEA, 1985). We have attempted to take account of these factors in the acquisition and selection of data in the present study.

CFs estimated from available monitoring data around the coasts of Ireland, the UK, Norway and Sweden showed considerable variability. Only in the case of ^{137}Cs in crabs and winkles was a statistically-significant increasing trend of CF over time observed. This could be taken to indicate that ^{137}Cs in the eastern Irish Sea has become more bioavailable since remobilisation from seabed sediments has become the dominant source, from the mid-1980s onwards. However, we have no complementary data to support this hypothesis. It was not possible to detect regional differences from the data available, with the exception of ^{99}Tc and

^{137}Cs in *F. vesiculosus* along the coast of southwest Sweden. In this case the spatial pattern is due to the impact of the salinity gradient resulting from the Baltic Sea outflow (Lindahl et al., 2003). Part of the variation in ^{99}Tc concentration in *F. vesiculosus* may be due to seasonal changes in apparent bioavailability, with relatively higher concentrations occurring in the winter months. It is suggested that this is due to seasonal variations in growth rates (Nawakowski et al., in press).

The measured transport coefficients (K_t) for plutonium were one order of magnitude lower for the storm and “trawler” experiments than for the passive diffusion experiments, showing that much more plutonium is made available for transport when the sediment is being disturbed. This remobilised plutonium can be transported over longer distances and made available for uptake by plant and animal species. The K_t 's for ^{137}Cs were in the same order of magnitude for all experiments. The ^{137}Cs remobilised from sediments seems to be less bioavailable than is generally observed in natural environments. The suggested explanation for this is that the ^{137}Cs is almost irreversibly bound to tiny suspended particles included in the water samples analyses. However, this particle bound ^{137}Cs will not be readily available to the biota, consequently resulting in a low CF. The results for plutonium are associated with large uncertainties so that no reliable conclusions can be drawn.

Work Package 5: Modelling

Generally speaking, work within WP5 has been focused on studying the description of the uptake/release processes of radionuclides between water and sediments. Several models using different modelling approaches with different spatial and temporal scales adapted to particular sites with different physical characteristics have been used to treat the problem under a perspective as wide as possible. The water-sediment interactions modules can fit concrete/limited experimental data relatively well, but have some difficulties with real dispersion of a wide set of radionuclides in natural marine environments. For example, several kinetic models have been proposed by different authors to fit laboratory sorption/desorption experiments, and all of them have been successful in each particular experiment. However, it has been found that differences in model outputs are drastically amplified when the different kinetic models are used in a real marine environment to simulate the same event. Computed activity levels in the water column and in the sediment are, in general, different for each kinetic model. A similar situation takes place in box modelling with different approaches to describe water-sediment interactions where the intensity of radionuclide exchange between solid and liquid phases can differ radically.

The logical question now is, obviously, which are the most suitable approaches. On the basis of the present knowledge, it is possible to make some conclusions for different model approaches:

- For hydrodynamical models, particularly for models it seems clear that a 1-step kinetic model produces a redissolution of radionuclides from sediments that is too fast when compared with experimental evidence. The use of an irreversible model introduces other problems that have been widely discussed in Deliverable 5.1. Thus, it seems that a 2-step model represents the best option, at this moment, to properly simulate adsorption/desorption processes. Nevertheless, any of the models studied can probably be used to reproduce observations through an adequate calibration of the kinetic rates. However, time scale of the processes would still remain rather different. Thus, a definitive decision on the most suitable approach cannot be given at this moment and, clearly, further work is required.
- For box model, the approach based on describing of radionuclide remobilisation from sea bed through the sediment reworking and pore water turn over rates seems more suitable for evaluation of water-sediment interactions in comparison with describing of remobilisation processes through a depth-depended empirical expression of the resuspension rate, because it provides a more complete description of radionuclide removal from the sea bed and has more adaptable possibilities for model construction.

However, this approach needs more site-specific information and very careful evaluation of parameters on the basis of simulation of a wide set of radionuclides.

It is important to note that the work provided under the course of the REMOTRANS project has demonstrated the need to test models against a wide variety of data. From this point of view, the Sellafield dataset is an excellent resource in this respect because of its unique coverage in time, space, and the radionuclides included.

6. Publications

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