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Processes Regulating Remobilisation, Bioavailability, and Translocation of Radionuclides in Marine Sediments

(REMOTRANS)

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Introduction

Marine sediments in European waters have become contaminated by artificial radionuclides as a result of a variety of planned and unplanned human activities. For example, 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 $^{239+240}$Pu and 41 PBq of ^{137}Cs have been discharged. 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 grained, muddy sediments of the west coast of Britain. Radiocaesium is more conservative in seawater but it can be scavenged by particulates, especially in coastal waters in association with clay particles. In addition, there have been significant releases of ^{99}Tc during different periods and also radionuclides such as ^{237}Np and ^{129}I. These more conservatively behaving radionuclides have been used as tracers for the water movements and mixing from the source. Releases from the French reprocessing facility, La Hague, have also contributed to increased concentrations of artificial radioactivity in European waters, in particular ^{129}I and ^{125}Sb, but the overall contribution has been relatively small.

The Baltic Sea was the sea which was most contaminated following the Chernobyl accident. The inventory of ^{137}Cs is estimated to be 5 PBq. The Baltic Sea is a marginal sea and is especially vulnerable due to its lower salinity, which causes higher Concentration Factors (CFs) in biota. While caesium is substantially more conservative than plutonium, much of the activity in the shallow Baltic sea has also been deposited in the sediments.

However, such contaminated 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 seabed sediments of the Irish Sea have been a net source of ^{137}Cs since the mid-1980s, and of plutonium throughout the 1990s, as a result of the decrease in direct discharges. It has been estimated that a significant amount of radiocaesium, about 60 TBq per year, is leaving the Baltic Sea to the North Sea associated with the net outflow, which corresponds to the inflow from rivers.

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 receiving 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 restricted to 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 1000 m, the inventories are a few percent of the cumulative fallout deposition, while on the shelf they can reach values higher than twice the 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 whole basin.

Certain marine areas, such as particular fjords, have natural boundaries such as anoxic-oxic interfaces, ice-water interfaces (Arctic) and/or saline-freshwater interfaces phases. These may cause enhanced natural accumulation and remobilisation processes, for example with higher levels in the water of especially actinides, close to the bottom.

Objectives

The overall aim of the project was to study remobilisation of radiologically important radionuclides from sediments of different characteristics in different European environments. This included the evaluation of the radiological consequences and trans-boundary exposure including uptake in biota and radioecological modelling. It was expected that the results would create more extensive knowledge of the long-term effects on the environment and man from past and present sources, and encourage a more informed debate on waste management of controlled releases of radioactivity in the marine environment. Important additional 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.

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

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 remobilisation and of bio-availability of removed radionuclide species required an innovative approach leading to a more robust understanding of marine environmental radioactivity. The multinational collaboration makes it possible to reach and perform sampling in a variety of European marine environments.

The overall objective of the project was achieved through a programme of work as follows:

- A sediment sampling and analysis programme leading to improved knowledge of inventories and quantifying potential sediment source terms
- Time-series measurements of radionuclides at key locations to quantify transport rates and transfer factors
- Development of a protocol for, and performance of, the determination of physico-chemical forms of the radionuclides in order to assess their potential remobilisation
- Laboratory experiments to assess the impact of remobilisation on bio-availability
- Development of radioecological models taking remobilisation processes into account.

Results

A sediment sampling and analysis programme was completed leading to improved radionuclide inventory estimates, and an overview of the contamination and comparison with earlier studies, in the areas of interest (the Irish Sea; the Skagerrak and Kattegat; the estuaries of the Odiel and Tinto Rivers; the Rhône Delta and the northern Adriatic Sea). To explain the balance of radiocaesium in the Baltic Sea, a remobilisation from the sediments must take place.

A number of new time-series were compiled. For example, the transit times from Sellafield of ^{99}Tc, released 1994-2001 (as a 'pulse' associated with the operation of the Enhanced Actinide Removal Plant, EARP), to the Swedish west coast was 4-5 years. This showed no apparent difference from the pulse of ^{99}Tc in released in 1978. However, more detailed examination of time-series seawater data from the Irish, North and Nordic Seas revealled apparent intra- and inter-annual variations in transport times. These appeared to be related to fluctuations in basin- and regional-scale processes (e.g. North Atlantic Oscillation, NAO).

There is a seasonal variation in ^{99}Tc concentrations in *Fucus sp.* (brown seaweed), with maxima in winter and minimum in summer. This was observed along the coasts of the UK, France, Sweden, and Norway, and appears to be related to seasonal growth patterns. The observations were more complex in the Kattegat, due to the strong gradient in salinity and seasonal variations. Radio-caesium concentrations are higher in the summer. The concentration factors are related to salinity. During summer, water with lower salinity flows out from the Baltic Sea. During autumn and winter weather, when low pressure dominates, the Baltic Sea is ventilated with more saline water from the North 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:

(i) at the pro-delta of the Tagliamento, where sediments transported by the river are rapidly accumulated and only temporarily stored, intense meteo-marine events can totally re-supend recent sediment and associated radionuclides and transport them to other marine areas

(ii) in the inner gulf of Trieste, were sediments transported by the Isonzo river are regularly accumulated and ^{137}Cs is stably stored

(iii) at the pro-delta of the Isonzo, where sediments and associated radionuclides rapidly accumulate. Although this area is as shallow as the Tagliamento pro-delta, the high sediment load transported by the river and the geographical location, more sheltered from the prevailing winds, allow net sediment accumulation and the permanent storage of the associated radionuclides.

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.

The change of CFs over time was investigated, especially in areas where remobilisation was demonstrated, such as the Irish Sea. In most cases no statistical trends were observed. In only two cases could a trend be shown: increasing ^{137}Cs CFs in winkles and crabs from the eastern Irish Sea over a period of 40 years.

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 pro-deltaic areas. Studies carried out in the Rhône pro-delta 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.

Mesocosm experiments were used to establish radionuclide uptake from contaminated sediments. Passive diffusion, and simulated 'storm' and 'trawler' disturbance were investigated. The measured transport coefficients for plutonium were one order of magnitude lower for 'storm' and 'trawler' experiments than for passive diffusion, showing that much more plutonium is available for transport when the sediments are disturbed. This plutonium can be transported over long distances and made available to plant and animal species. The transport coefficients for ^{137}Cs were in the same order for all experiments. The ^{137}Cs remobilised from sediments seems to be less bio-available than is generally observed in natural environments. Uptake in fish was studied by a cannulation technique in a separate set of experiments. This showed that the uptake depends on the speciation of radionuclides and that the interaction with organic material in waters changes the bioavailability.

A number of modelling approaches were investigated. For one of the hydrodynamical models, a 1-step kinetic model produce a redissolution of radionuclides from sediments that was too fast when compared with experimental evidence. The use of an irreversible model introduced other problems that have been widely discussed in Deliverable 5.1. In this case, it seems that a 2-step model represents the best option, at this moment, to properly simulate adsorption/desorption processes. However, there are other models which can be used to reproduce observations but including key environmental processes (e.g. bioturbation) and through an adequate calibration of the kinetic rates. However, the representation of appropriate time scales of the processes remains 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 modelling, the approach based on describing radionuclide remobilisation from the 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 remobilisation processes through a depth-dependent 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 the parameters, on the basis of simulation of a wide set of radionuclides.

It is important to note that the work provided in 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.

Implications

It is shown that there is a trans-boundary exposure from remobilised radionuclides. The CFs and Transfer Factors are similar to directly discharged radionuclides and the radiological implications can be calculated on the same basis. The consideration of long-term consequences has resulted in improvements to models and can now better be estimated as a result of the REMOTRANS project.

The REMOTRANS project has helped to maintain end also enhance European competence in the field of environmental radioactivity. It has provided training and increased mobility for younger scientists. The project has resulted in a number of PhD examinees.