TRANSPORT OF PLUTONIUM ($^{239/240}$ Pu) AND CAESIUM (^{137}Cs) IN THE IRISH SEA: COMPARISON BETWEEN OBSERVATIONS AND RESULTS FROM SEDIMENT AND CONTAMINANT TRANSPORT MODELLING

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Introduction

Significant quantities of artificial radionuclides reside in the sub-tidal sediments of the Irish Sea as a result of authorised discharges from the Sellafield nuclear reprocessing plant. An understanding of the processes that may determine the fate of this material is of considerable interest, both from environmental and political perspectives and the need to assess dose to the human population. The work presented here forms part of a long-term study program, funded until 1999 by the UK Ministry of Agriculture Fisheries and Food (MAFF), that has monitored radionuclide distributions and undertaken studies of the processes controlling radionuclide movement in the Irish Sea.

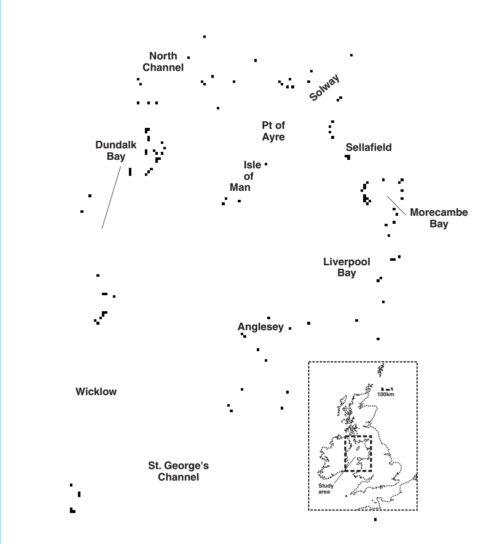


Figure 1. Model grid (mesh size; 1/20-degree longitude, 1/30-degree latitude).

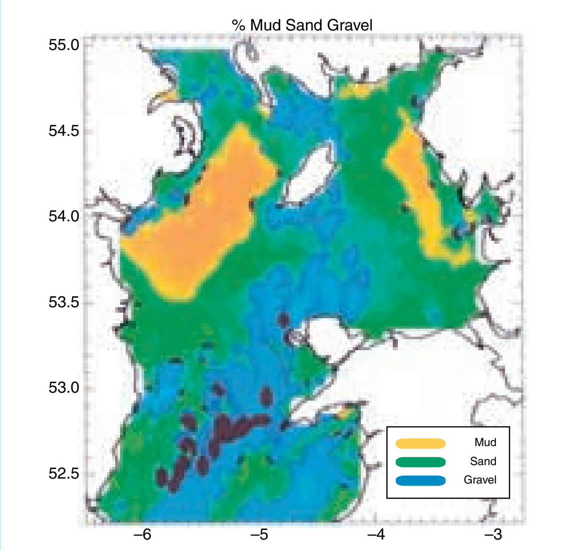


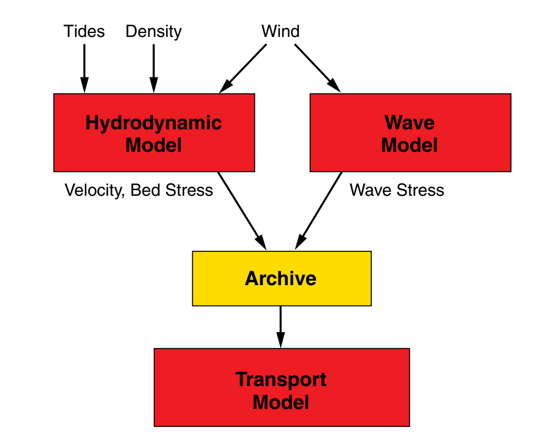
Figure 2. Bed type distribution. Source British Geological Survey.

A model for radionuclide transport in the marine environment is described and applied to the movement of ^{137}Cs and $^{239/240}$Pu discharged from Sellafield in the Irish Sea. The model resolves processes operating on tidal time scales whilst being able to run over decadal time periods. Modelled processes include transport in the water column, exchange of contaminants between dissolved and particulate phases, wave-current sediment resuspension, sediment transport and mixing of material within the seabed. Quantities are solved in depth integrated form. A layered seabed is incorporated with transfers between layers representing biological and physical mixing processes. Model simulations were carried out on the grid shown in Figure 1.

Figure 2 shows the distribution of bed types in the Irish Sea based on data supplied by the British Geological Survey (BGS) from 900 bed samples. Regions of predominantly fine muddy sediment occur to the east and west of the Isle of Man, where tidal currents are generally less than 0.3 m s^{-1}. Regions of fine sediments in particular can act as temporary sinks for many discharged radionuclides, such as plutonium, that have an affinity for particulate material.

Model

Flow fields were calculated using a three-dimensional model developed at the Proudman Oceanographic Laboratory. The model has been widely reported in the literature and extensively validated for the Irish Sea (Davies & Aldridge, 1993; Davies & Lawrence 1993).



Measurements in the shallow eastern Irish Sea indicate that wind waves play a significant role in resuspending sediment (Aldridge, 1996; Philpott, 1997). A relatively simple approach to calculating the time varying surface wave spectrum was used based on a duration and fetch limited JONSWOP spectrum (Hasselmann *et al.*, 1973). A wave current interaction model (Fredsøe, 1984) combined wind wave and current stress to yield the time varying wave-current bed stress at each point in the model domain.

Hydrodynamic and wind-wave models are used to produce a series of year-long data sets with one hour temporal resolution, archived for subsequent transport model calculations. A calculation to predict radionuclide movement is accomplished by taking a series of these year-long flow and wave fields. For longer term calculations, the sequence of year-long flow fields are repeated as necessary for the duration of the run. The assumption is made that the flow and wave conditions are a statistically representative of overall conditions.

Results for ^{137} Cs transport

The dispersion of the ^{137}Cs is used to validate the water column transport in the model. Comprehensive datasets of the distribution of ^{137}Cs exist for the Irish Sea, together with good knowledge of Sellafield discharges. Although partly sorbed onto sediment, previous studies have shown that the assumption that ^{137}Cs is conservative is well justified during the period of high discharge during the 1970's and early 1980's. Flow fields based on winds from a 2-year period closest to the 1970-1995 average wind speed are used as a basis. A comparison between the model and observations for September 1977 is shown on the right.

This shows good agreement in the eastern Irish Sea, but suggests flushing through the western Irish Sea is over estimated.

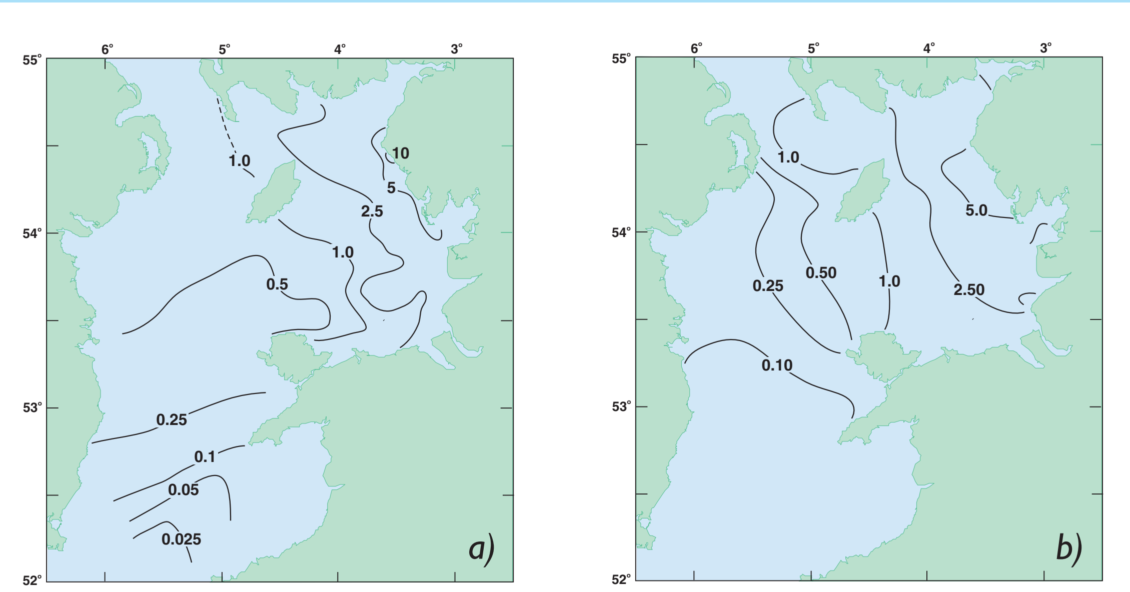


Figure 3. ^{137}Cs Distribution in September 1977 (Bq kg^{-1}). a) Observed, b) Modelled.

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Results for $^{239/240}$ Pu transport

Consideration of seabed sediments is essential in dealing with the transport of plutonium, the behaviour of this element being dominated by the interaction with particulate material. The partitioning coefficient (Kd) together with the desorption rate constant (McCubbin & Leonard, 1996) enable a relatively simple treatment of transfer between dissolved and particle phases to be developed. To complete the formulation it is necessary to predict concentrations of sediment in the water column and the rate of erosion and deposition at the seabed. Model calculations of these quantities have been validated against available observations.

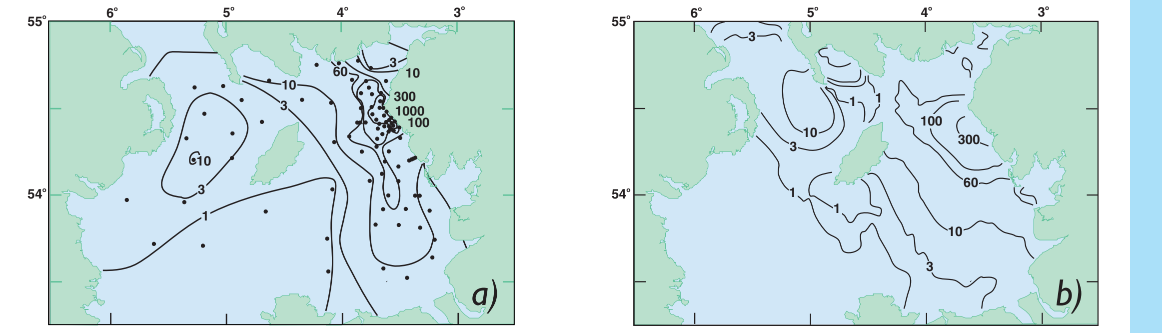

Figure 4. Total seabed inventory of $^{239/240}$Pu for 1995 (Bq m^{-2}). a) Observed, b) Modelled.

Transport calculations for $^{239/240}$Pu start from 1965; beginning with an assumed non-contaminated bed and are carried forward till 1995 with known Sellafield input. Archived two-year long flow fields are repeated as necessary to cover the simulation period, with the simulation taking about 2 days of computer time.

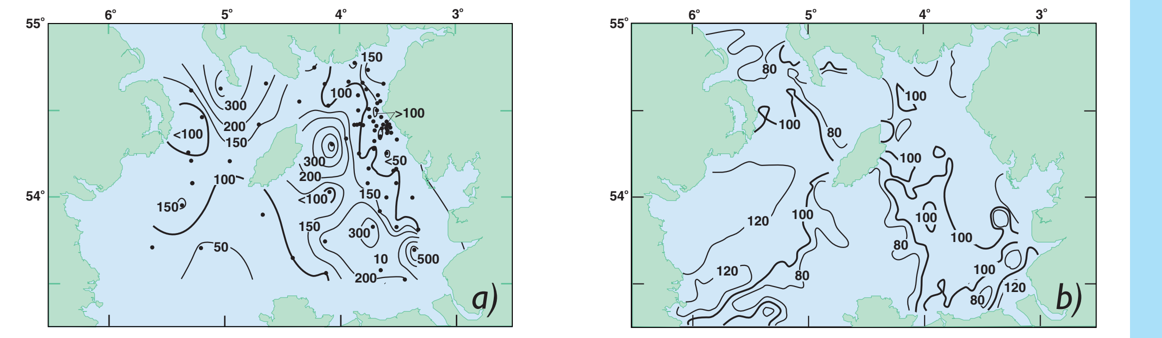

Figure 5. Seabed inventory ratio 1995/1988 for $^{239/240}$Pu (% 1995/1988). a) Observed, b) Modelled.

The mechanism in the model for accumulation of $^{239/240}$Pu in the seabed is scavenging of dissolved radionuclides by resuspended mud/silt and fine sand, or by pore water exchange with the top bed layer. Mixing of this material deeper into the seabed is then allowed simulating biological and physical exchange processes. The results shown in Figure 4 compare the observed and modelled seabed inventories (Bq m^{-2}) for 1995. In general, good agreement is found for the levels and distribution of material in the seabed. Clearly the pattern is influenced by the bed composition with high inventories associated with the finer muddy sediments.

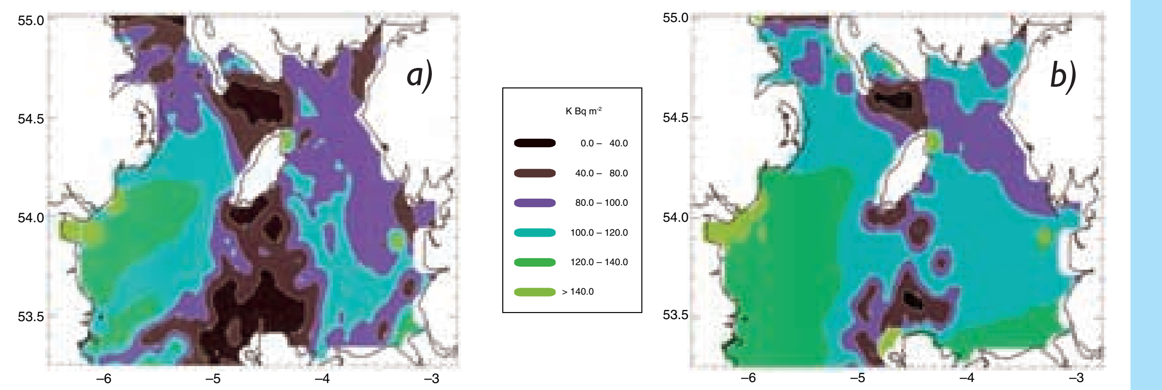

Figure 6. Modelled seabed inventory ratio 1995/1988 for $^{239/240}$Pu (% 1995/1988), a) with sediment transport and b) without sediment transport.

An earlier bed inventory survey was undertaken in 1988. The ratio of the 1995 to the 1988 bed inventories (Figure 5) is revealing. Both observations (Kershaw *et al.*, 1999) and model results present similar and consistent patterns, with net loss from along the Cumbrian coast and gains in Liverpool Bay and the western Irish Sea. The increase in Liverpool Bay is somewhat patchier, and in places significantly smaller in the model than observed. The patchiness seen in the model results is due to the patterns of sediment erosion and deposition and this aspect of the model requires further investigation. However the agreement is encouraging.

Once incorporated into the bed sediments, radionuclide re-distribution can then occur by two principal mechanisms. Directly, by the transport of contaminated sediment, or indirectly via exchange and transport in dissolved form (dissolution). The latter process operates when tidal, wind or trawling activity re-suspends bed material allowing transfer of radionuclides to the water column. This transport mode can now operate because input from Sellafield has decreased sufficiently for radionuclide concentrations on bed sediments to become on average higher than water column levels. Subsequent transport in dissolved form allows scavenging and sorption by less contaminated sediments elsewhere. Figure 6a & b shows contour plots of the full simulation with a run that has direct movement by sediment transport 'switched off'. There is a broadly similar pattern of gain and loss, although some local differences due to absence of sediment transport are apparent, particular in the eastern Irish Sea. The implication is that dissolution is the primary mechanism determining the overall changes and transport over larger scales, but that sediment transport acts to modify this locally.

Summary

Results demonstrate that the model is able to reproduce observed concentrations of radionuclides in the water column, the build-up of $^{239/240}$Pu in the seabed over a thirty year time span and the recent changes in seabed distribution. The model is used to identify and quantify the mechanisms responsible for the recent redistribution of $^{239/240}$Pu in the region. The results suggest that whilst sediment transport plays an important role in redistributing contaminated sediment in the eastern Irish Sea, desorption, followed by transport in the dissolved phase and re-absorption onto particulate material is the primary mechanism for re-distribution of $^{239/240}$Pu over longer distances.

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