Radioactive pollution

The relative importance of physical, chemical and biological processes in the Irish Sea

by David Jones

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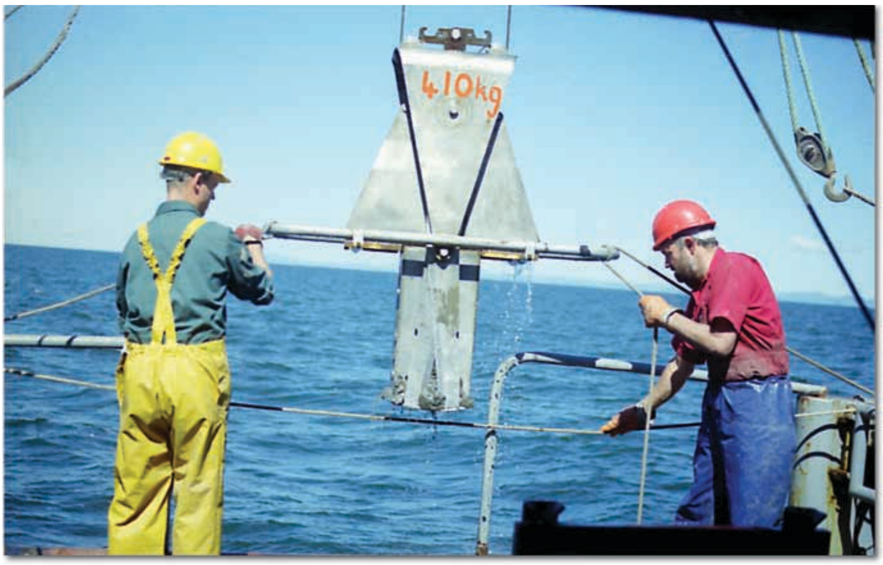
The UK has obligations to protect its marine environment under international conventions such as OSPAR (Convention for the Protection of the Marine Environment of the North-East Atlantic). In order to do this we need to carry out robust science to help us understand the processes and influences that impact on the marine environment and to underpin government policy. As part of a research consortium, we have recently examined the relative influence of physical, chemical and biological processes on the transport and speciation of pollutants in the Irish Sea. The emphasis was on radionuclides discharged from Sellafield. The report was timely because it preceded a pilot study on marine nature conservation in the Irish Sea.

The first stage of the work was a review of existing knowledge. This was followed by detailed studies of sediment cores from the area close to Sellafield. Intertidal cores were also collected, to greater depths than previously. These provided a contrast to the offshore cores, in a zone more accessible to man, and improved our knowledge of the inventory of radionuclides in this environmental compartment. The sediment studies were supplemented by modelling work that included 3D hydrodynamic modelling and chemical modelling.



Collecting a box core sample of Irish Sea sediment.

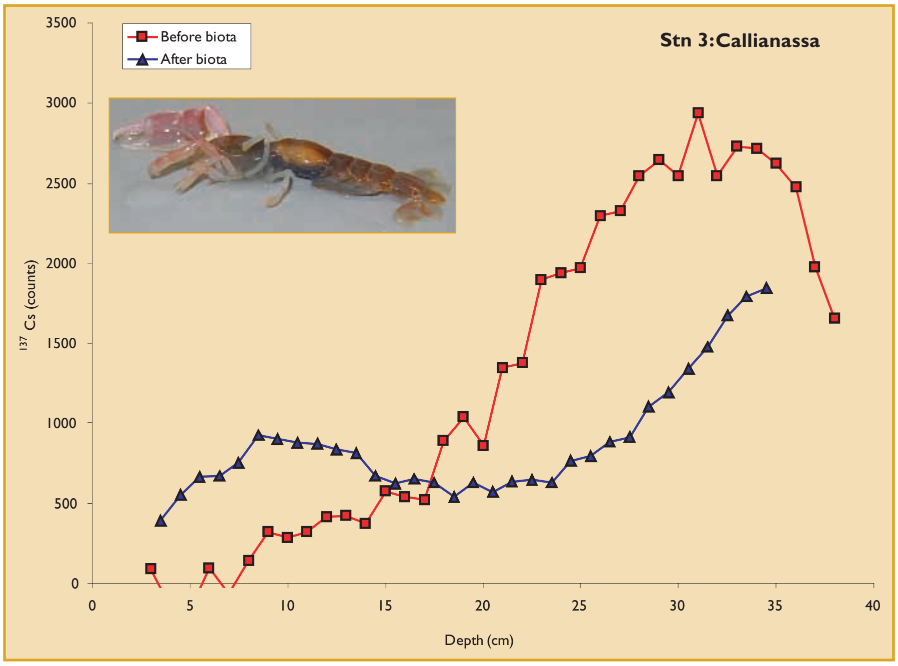
The study has shown that physical, chemical and biological factors are all important. Their relative importance varies from place to place and differs markedly between intertidal and offshore environments. Further work is needed to move from a qualitative assessment to a more quantitative approach. Some of the methodologies developed for this study offer this possibility, particularly with respect to biological processes.

The offshore sediments are, typically, burrowed to depths of 10 to 20 cm, but at greater depth physical sedimentary structures are preserved. Profiles of radionuclide concentrations with depth are consistent with the Sellafield discharge history, modified by mixing by burrowing organisms. This is supported by ratios of plutonium isotopes that reflect progressively earlier discharges with depth. Laboratory work shows that mud shrimps (*Callianassa*, *Upogebia*) effectively redistribute radionuclides in the sediments, whereas other species studied appear to have relatively little impact. Modelling techniques were used to estimate the degree of mixing produced by different species. Taken in conjunction with information on species density and geographical distribution, the impact of bioturbation could, in future, be estimated quantitatively.

Physical processes are more important offshore below the surface oxygenated zone. Sedimentary structures are apparent below the top 10 to 30 cm and include probable storm layers. The impact of storms was also examined by three-dimensional hydrodynamic modelling. Water movements in detail were controlled by the direction and duration of the storm, but showed the potential to move contaminated sediment into the estuaries of the eastern Irish Sea and more soluble phases over wider areas. Removal of oxygenated surface sediment could expose reduced sediments to aerated water and this could have implications for the release of redox sensitive contaminants. Bioturbation also has the potential to introduce local oxic conditions or recycle material from depth to the oxic near-surface environment.

Geochemical modelling has shown that plutonium is partly soluble under oxygenated conditions, with up to 10% being available for removal in the aqueous phase. This fits well with the observed loss of plutonium from the sediments over time. The much greater proportionate loss of caesium is well documented from both subtidal and intertidal sediments. Sequential extraction techniques suggest that much of the remaining caesium is relatively tightly bound in clays and therefore loss from the sediment may be expected to now be much reduced. Caesium and uranium are more prevalent in the deeper sediments, perhaps also indicating their removal from near-surface material. Pore water measurements, microbiological work and modelling all confirm the presence of a shallow (typically 10 to 15 cm deep) oxygenated layer with increasingly reducing conditions below this. Sulphate-reducing bacteria are probably controlling sulphide production at depth, seen as dark coloration in the cores and as microscopic pyrite framboids within the sediments.

Bulk mineralogy does not appear to be a controlling factor in the uptake of radionuclides by the sediments. Grain size does, however, play a part. Radionuclides are distributed in the fine matrix of the sediments but are also associated with hot particles. Contrary to previous studies, hot particles of possible effluent origin, or associated with iron minerals, have not been observed. Rather, the hot spots seen are organic gel-like particles with a high magnesium content or consist of mud pellets (possible faecal) or pellicles around shell fragments. It appears the latter may be associated with local reducing conditions, perhaps related to the breakdown of an organic coating. This is supported by the presence of pyrite. Shells lacking an organic coating (possibly older fragments) appear not to have the more radioactive rim. The



The effect of the mud shrimp Callianassa *on Caesium-137 in a sediment core. The profile of the radionuclide before introduction of this burrower (red) is altered substantially after a few months of burrowing (black).*

difference in hot particle characteristics may reflect diagenetic breakdown of effluent particles, with the high magnesium, perhaps reflecting the magnesium-alloy casing on Magnox fuel

The intertidal sediments appear to be dominated by physical sedimentary processes; cores are characteristically well laminated. It seems that migration of tidal channels causes extensive reworking of the sediments and obliterates smaller-scale mixing by burrowing. The estuaries, with their tidal dominance, are filling with sediment from seawards with little landward input from rivers. They therefore remain a sink for particle-associated contaminants, although they have clearly released significant amounts of more mobile caesium since Sellafield discharges were reduced.

Coring of the intertidal sediments has, in places, reached uncontaminated sediment at depths of less than a metre. Further from the shore, radionuclides are in many places still present down to at least 2.5 metres. Additional deeper coring is required to finally define the volume of contaminated sediment, but known inventory estimates for caesium-137, americium-241 and plutonium isotopes in the Solway Firth and Morecambe Bay can now be revised upwards by a factor of around four. ▮

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For further information, contact:

David Jones
Tel: +44(0)115 936 3576
e-mail: dgj@bgs.ac.uk