* EFFECTS OF A LONG-TERM RELEASE OF PLUTONIUM AND AMERICIUM INTO AN ESTUARINE AND COASTAL SEA ECOSYSTEM

II CHEMICAL SPECIATION AND ENVIRONMENTAL FACTORS

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Summary

Continuing the development of a methodology for the assessment of the distribution and fate of actinide radioactivity in the case of its introduction into surface waters of a coastal marine ecosystem, the consequences of different chemical and environmental factors are discussed in relation to actinide transport and bioavailability.

Resumé

En poursuivant le développment d'une méthodologie pour l'évaluation de la distribution et de la destinée des actinides dans l'hypothèse de leur introduction dans les eaux de surface d'une ecosystème littoral, on discute des conséquences des différentes espèces chimiques des actinides et des paramètres liés au milieu sur la mobilité des actinides et leur biodisponibilité.

A methodology for the assessment of the distribution, fate and associated hazard due to a release of long-lived alpha emitting radionuclides into surface waters of an estuarine-coastal sea ecosystem is being developed ; MURRAY and AVOGADRO (1978). The study is a part of a project on the assessment of risk associated with the long-term storage of solidified high-level radioactive wastes in geological formations; GIRARDI et al. (1977).

The present analysis has been developed to consider new data especially in relation to differences in chemical reactivity of plutonium and americium. The following environmental regions have been considered : river, estuary and coastal sea.

The occurrence of various chemical species means that it is not possible to use single values for distribution and transfer coefficients within the different compartments considered in Figure 1. These differences will thus probably play an important role in the following environmental processes a) sorption and transport, b) bottom sediment migration and c) biological accumulation.

The formation of different species of plutonium has already been demonstrated thermodynamically. The hexavalent form and its complex ions (mainly carbonate) are much more mobile than other species, and may play the most important role in regulating the long-term transport and bioavailability in aquatic systems.

It has been demonstrated that differences in K_d for Pu (IV) and (VI) species lead to changes in the distribution of "soluble" forms of plutonium during their transport from freshwater (pH 6.5) to seawater (pH 8.0). An increase of 0.3 to 10% in the Pu (VI) "soluble" form has been calculated to occur in an estuarine environment.

Taking the results reported by HETHERINGTON et al. (1975) for plutonium concentrations in seawater and sediments of the Irish Sea, a possible interpretation is proposed for the apparent decrease of sediment K_d with distance from the input source, based on the co-existance of two species of plutonium (IV) and (VI) respectively.

Fig 1 Model-compartments

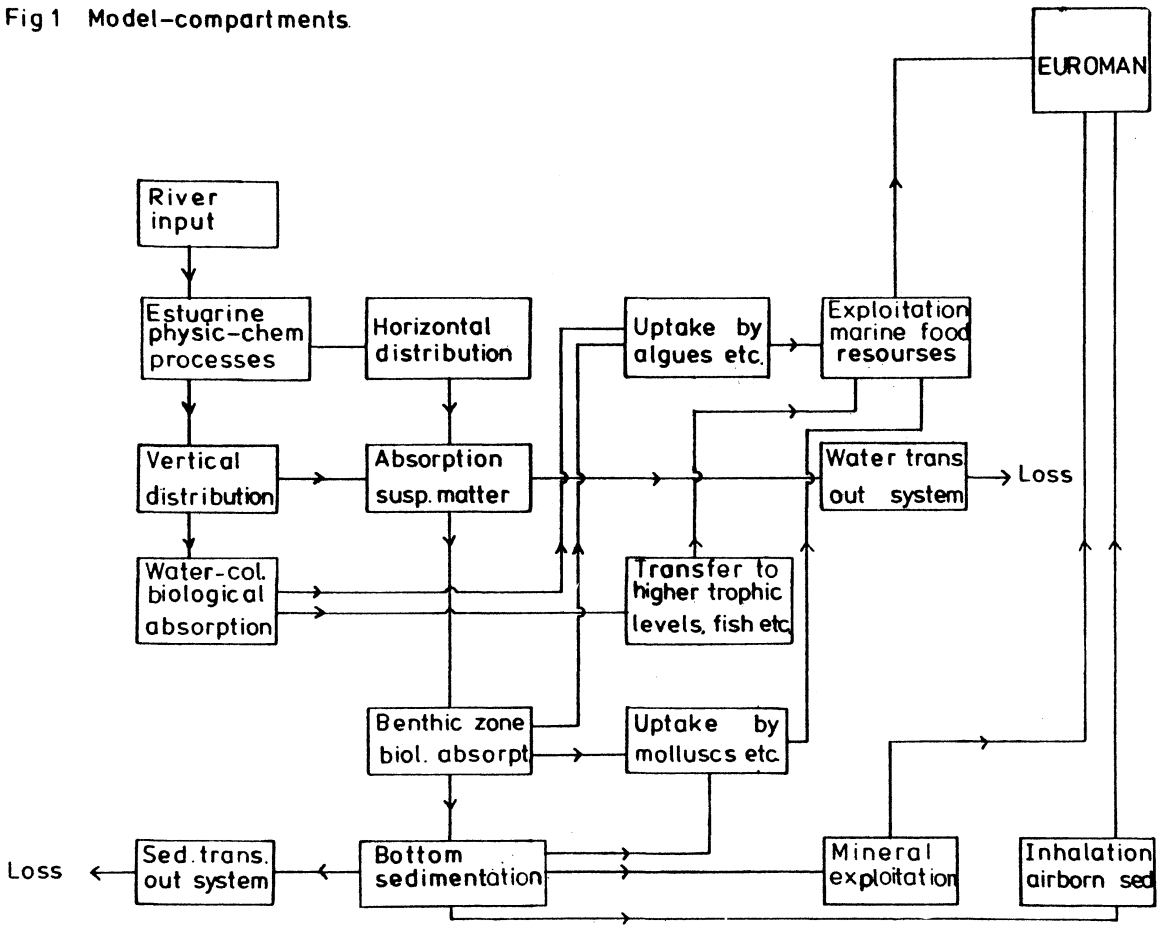
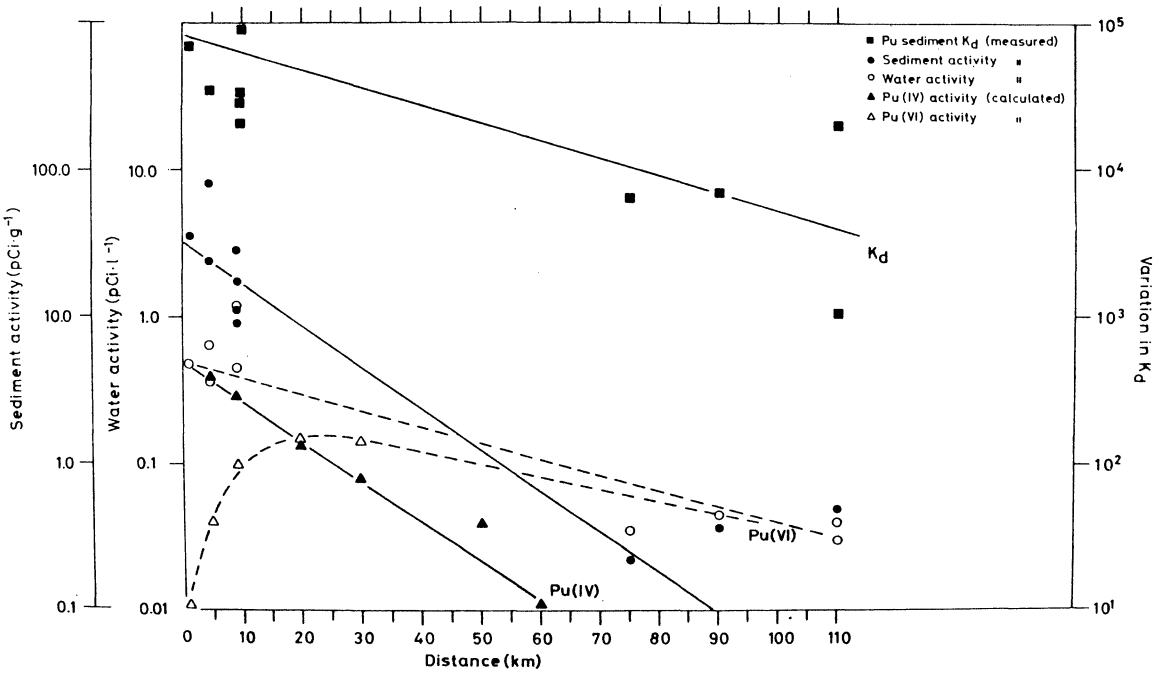


Figure 2 Distribution of Pu (IV) and (VI) activities with distance in Irish Sea water and sediment



These forms have been shown to exist by NELSON and LOVETT (1978). Table 1 shows the variations of the ratio in water of the two valence states. As can be seen in Fig. 2 the decrease of K_d may be explained by the percentage increase of the more mobile plutonium (VI) species with distance.

References

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Some investigations into the behaviour of plutonium in the marine environment
In Int. Symp. Impact of Nuclear Releases into the Aquatic Environment. IAEA Vienna 193-212

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Effect of a long-term Release of Plutonium and Americium into an Estuarine and Coastal Sea Ecosystem
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The Oxidation states of plutonium in the Irish Sea
(submitted for publication)

Table 1 Distribution of plutonium (IV) and (VI) activ

Irish Sea water and sediment

Seawater		1 Km	4.5 Km	9 Km
Pu (IV)	pCi Kg^{-1}	0.49	0.39	0.30
Pu (VI)		0.01	0.04	0.09
total		0.50	0.43	0.39
Pu $\frac{(VI)}{(IV)}$	%	2	9	23
sediment	pCi g^{-1}	34	24	18

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"Effects of a long-term release of plutonium and americium into an estuarine and coastal sea ecosystem. II - Chemical speciation and environmental factors"

Paper presented by C.N. Murray (CCR Euratom)

Discussion

G.G. POLIKARPOV: Did you try to estimate the dose-rates of Am in animals and the distribution of the absorbed doses?

C.N. MURRAY: Unfortunately we are not in a position to calculate the organ dose ad the present time, but as I pointed out at the start of my paper, we recognize the importance of carrying out these types of calculations in code to discuss the effect of radiation on aquatic organisms.