MINERALOGICAL MAGAZINE, APRIL 1985, VOL. 49, PP. 177-194

The disposal of radioactive wastes into the marine environment: the presence of hot particles containing Pu and Am in the source term

E. I. HAMILTON

Institute for Marine Environmental Research, Prospect Place, The Hoe, Plymouth PL1 3DH, Devon

ABSTRACT. Radioactive particulate matter (identified as hot particles) is present in the effluent discharged by the British Nuclear Fuels Ltd (BNFL) uranium reprocessing plant at Sellafield, Cumbria, UK. There is very little information on the abundance or chemical and physical forms of solid matter in the effluent; even less is known of the significance of particulate debris in relation to the uptake of radionuclides for non-occupationally exposed people as a result of transfer along marine foodchains. Some observations on the occurrence and abundance of hot particles in the vicinity of Sellafield are reported, with special reference to those that contain transuranic radionuclides (Pu,Am,Cm). Some of the uncertainties are discussed in an evaluation of the significance of hot particles, albeit aggregates of colloids for the smallest particles, and exposure to man from ionizing radiation. There is no evidence that hot particles derived from BNFL and subsequently dispersed into the marine environment represent a hazard to man. However, further studies are required in order to determine whether or not the pathways followed by the particles are significant, or different to those of other radionuclides through which the radiation exposure of man within this region of Cumbria is assessed.

KEYWORDS: radioactive waste, marine environment, Sellafield, Cumbria, Irish Sea.

A PRIME objective in the disposal of radioactive wastes into near-shore waters of the marine environment is to ensure that the procedures adopted provide for an adequate dispersion of the effluent, such that now, and in the future, the practice should not constitute a hazard to man or to parts of marine ecosystems. In practical terms this statement requires some clarification:

(i) Any hazard should be evaluated in relation to other accepted risks which are associated with man's cultural and technological advancement.

(ii) The important components of the total cost-benefit equation should be identified, in particular the accuracy of the data which are used, and an identification of those components of the equation which are most sensitive to change, or for which quantitative data are not available.

(iii) In the disposal of radioactive wastes the average concentration of a radionuclide over several decades, the short-term maximum in a small region, or the occurrence of extreme transfer events should not lead to a potentially significant hazard.

Man appears to be the form of life which is most sensitive to the effects of ionizing radiation: in the marine environment, no effects on biota are seen which can be unequivocably attributed to the presence of ionizing radiation (above that which occurs naturally) as a consequence of the current practices for the disposal of radioactive wastes into the sea. This does not mean that such effects do not occur, but rather that because of the very high natural mortality in the seas, any small contribution arising from the disposal of radioactive wastes cannot be identified. The complexity of the problems associated with the disposal of radioactive wastes, and the limited availability of data have resulted in the adoption of various types of predictive models. It is essential that these models are evaluated for sensitivity to all processes likely to be operating. One small aspect of the problem is considered here, namely the high levels of radioactivity associated with small (hot) particles, which is a neglected field of study. They occur in the natural environment and are derived from the nuclear industry: similar particles form in the sea as a result of natural processes.

The health of man is assumed to be safeguarded by the practice of monitoring seafoods available for consumption and identification of critical pathways for transfer of radionuclides, together with more detailed studies of individuals who are subject to unusual exposures. With the exception of occupationally exposed individuals, most of the data in current use for the purposes of radiological protection does not distinguish between conservative and non-conservative forms of radionuclides; very little

is known of the abundance and composition of so-called hot particles.

The study area is in the NE Irish Sea, which receives high-level radioactive wastes* from the British Nuclear Fuels (BNFL) reprocessing plant at Sellafield, Cumbria, UK. According to the Ministry of Agriculture, Fisheries and Food Certificate of Authorization for the Disposal of Radioactive Waste (Great Britain, 1960), 'liquid radioactive waste means waste consisting of liquid with or without solid matter in suspension therein'.

Reports (e.g. Atherton, 1979, 1980; Hetherington, 1976; Howells, 1977; Hunt, 1979–83; Mitchell, 1967–77) on radioactive discharges into the Irish Sea do not identify the amount, particle size, or composition of solids which are discharged. The data presented here are based mainly on preliminary observations which are considered relevant for non-occupationally exposed individuals; a more detailed account of the procedures adopted will be presented elsewhere. The data cannot be extrapolated to other sites and are only applicable to the study area; attention is mainly focused on the alpha-emitting heavy radionuclides (Pu,Am,Cm).

The quality of data used in radiological protection of the general population

Two extreme approaches are often taken when considering the subject of environmental radioactivity: first, the use of experimental systems to understand the processes controlling the dispersion, uptake, loss, and retention of radionuclides in marine ecosystems (to be of practical value they need to be validated on natural systems); secondly, an evaluation of the extent to which man-produced radionuclides behave as analogues of stable elements. In the disposal of radioactive wastes to the oceans, the nature and composition of the source term could initially influence or even control subsequent distributions (source term effects are most likely to occur near the site of discharge). Most radionuclides which enter the seas, in diverse chemical and physical forms, will gradually adopt characteristics which are identical or similar to those of stable element analogues through processes of geochemical and biological recycling. An exception would be a highly resistant and non-degradable type of waste which, in the water column or in bottom sediment, would probably act in a non-conservative manner. Therefore in order to understand better the processes affecting the cycle of radionuclides in the sea, it is important to consider the short- and long-term influence of the source term. Unfortunately, for most industries, and the nuclear industry in particular, information which adequately defines the source term is rarely available.

One of the primary aims of radiological protection is to ensure that radiation exposures are kept 'as low as reasonably achievable' (ALARA). The radiation exposure of the population (called the collective dose) is used as a measure of the health detriment in the irradiated population to ensure that the ALARA criterion is met. Prediction of the collective dose in marine systems involves an assessment of the dispersion of radioactivity throughout the oceans and its subsequent return via marine pathways to man. A large number of factors needs to be identified, which may be often restricted to defined geographical or climatic regions of the world, or to particular situations such as proximity to a source. The National Radiological Protection Board and the Commissariat à l'Energie Atomique (NRPB/CEA, 1979) have created a compartmental model which, in the UK, has been further developed by the MAFF Fisheries Radiological Laboratory [FRL] (Camplin *et al.*, 1982) to obtain estimates of collective dose. In the UK the various compartments of the model and transfer factors are considered in more detail for special areas, such as those affected by the operations at BNFL; similar models are being considered for the open ocean dump sites (GESAMP, 1983).

It is important to establish the relationship between field data and the effects of radiation. The accuracy and uncertainties of the data which are related to effects from ionizing radiation should be considered in relation to those concerned with the nature and rates of processes in models for the disposal of radioactivity into the marine environment. Although many of the models appear acceptable they still contain terms based on inadequate data.

The extent of further research required on the exposure of general members of the population to low levels of ionizing radiation can only be determined when more data become available for the concentration of radionuclides in human tissues at critical sites.

If radioactive (hot) particles enter the body and are retained as discrete particles they may act as loci of high radiation doses; high concentrations could lead to death or to a shortening of life by destroying functional tissue; lower concentrations may be detrimental by causing progressive fibrosis. The respiratory and lymphatic tissues are of particular concern in the inhalation of particles, and the possible development of radiation-induced neoplasia in these systems requires further consideration. The problem is so complex that data of direct

*Arising principally from the reprocessing of spent Magnox uranium fuel rods.

relevance to effects are not presented here; indeed no effects have been observed beyond the occasional accidents among occupationally exposed individuals, or individuals in close proximity to nuclear detonations. Nevertheless, areas of uncertainty for normal members of the population, which are evident from a study of the principles adopted by the International Commission on Radiological Protection (ICRP), are as follows:

(i) The ICRP (1972, 1981) describes three classes of solubility for retention of radionuclides in the body which partly reflect their chemical and physical forms. These classes may be appropriate for exposed workers but there is little information concerning the abundances or chemical form of radioactive particles which are present in the natural environment.

(ii) Considerable uncertainties exist in the influence or significance of chemical and physical form of radionuclides in transfer of materials from lung to lymph nodes. The relative roles of particle form or macrographic processes on translocation are not discussed.

(iii) No human beings (apart from those exposed in the industry or accidents) have shown health effects that can be associated with inhalation of radionuclides. It is therefore necessary to obtain data from animal experiments, where almost all the data are derived from the inhalation of simple chemical and physical forms of radionuclides which may not be present, or even stable, in the natural environment.

(iv) The ICRP does not recommend the use of its data and models for exposed individuals to estimate committed dose equivalents to members of the general population by adjusting solely on the basis of differences in mass of organ and magnitude of intake; the data were not collected with this purpose in mind.

(v) The ICRP Annual Limit on Intake (ALI) does not consider chemical toxicity. By definition a hot particle contains a high concentration of an element in a small volume, hence the chemical nature of the material may be 'recognized' by a biological process and retention and translocation within the cell, organ or whole body may be influenced by such a process.

(vi) For the purposes of environmental exposure the ICRP does not consider chelated forms of radionuclides (e.g. Pu,Am,Cm), although they are considered to be more mobile in the body than other compounds (Durbin, 1973) and therefore may be more rapidly excreted than other forms.

(vii) For non-occupationally exposed individuals, an evaluation of lung burdens of the heavy radionuclides by *in vivo* counting is of no practical value. Even for those occupationally exposed, the limits of detection are poor and measurements pose many difficulties, so that only cases of very severe exposure can be diagnosed. Information concerning the distribution of radiation dose or the influence of translocation processes is not available even in these circumstances.

There are also uncertainties associated with the efficiency of field sampling of natural aerosols and estimating their transfer from the natural environment to the human lung. It is contended by some that lung deposition of radioactive hot particles containing a certain amount of alpha-activity represents a risk of lung cancer substantially greater than if the same amount of radionuclide were distributed among small sources, or present as a true diffuse distribution (Tamplin and Cochran, 1974). The National Resources Defence Council (NRDC) in the United States considers that the critical particle contains a minimum of between 0.14 and 4.3 pCi; in this work an average value of 0.6 pCi is used (Tamplin and Cochran, 1974; ICRP, 1980). In this study the ICRP (ICRP, 1980) description of hot particles is adopted. They are recognized in autoradiographs (for alpha sources) as 'alpha stars', generated by alpha radiation emanating from larger particles of high specific gravity or smaller particles inhaled, the solubility and the propensity to aggregate, determine the characteristics of alpha radiation, e.g. the formation of single tracks or stars. Recognition of a hot particle by a star depends on the duration of autoradiographic exposure. If long exposure times are used most diffuse distributions will eventually be represented by 'alpha stars'. It is difficult to determine the specific activity of a hot particle whose density has to be assumed and which can range from about 1–12 g cc^{-1}. In autoradiography hot particles are seen as 'fixed phenomena'; *in vivo* the particle would be expected to move over tissue surfaces, and hence the dose delivered per unit volume of tissue with time would be much less. From a consideration of radiation effects some cells adjacent to a hot particle would be killed, which is a common biological process. However, the probability that a cancer will be induced by a given particle will depend upon whether or not the radioactivity of the particle is sufficient to impart just enough energy to cause changes in adjacent cells leading to cell transformations and eventually cancer (ICRP, 1980). Evidence that a single radioactive particle can lead to histological changes and eventually neoplasia has not been demonstrated.

The foregoing statement thus outlines the need for research requiring evaluation in relation to the physical and chemical properties of radioactive particulate material, and is here applied to the study of hot particles, as a component of the source

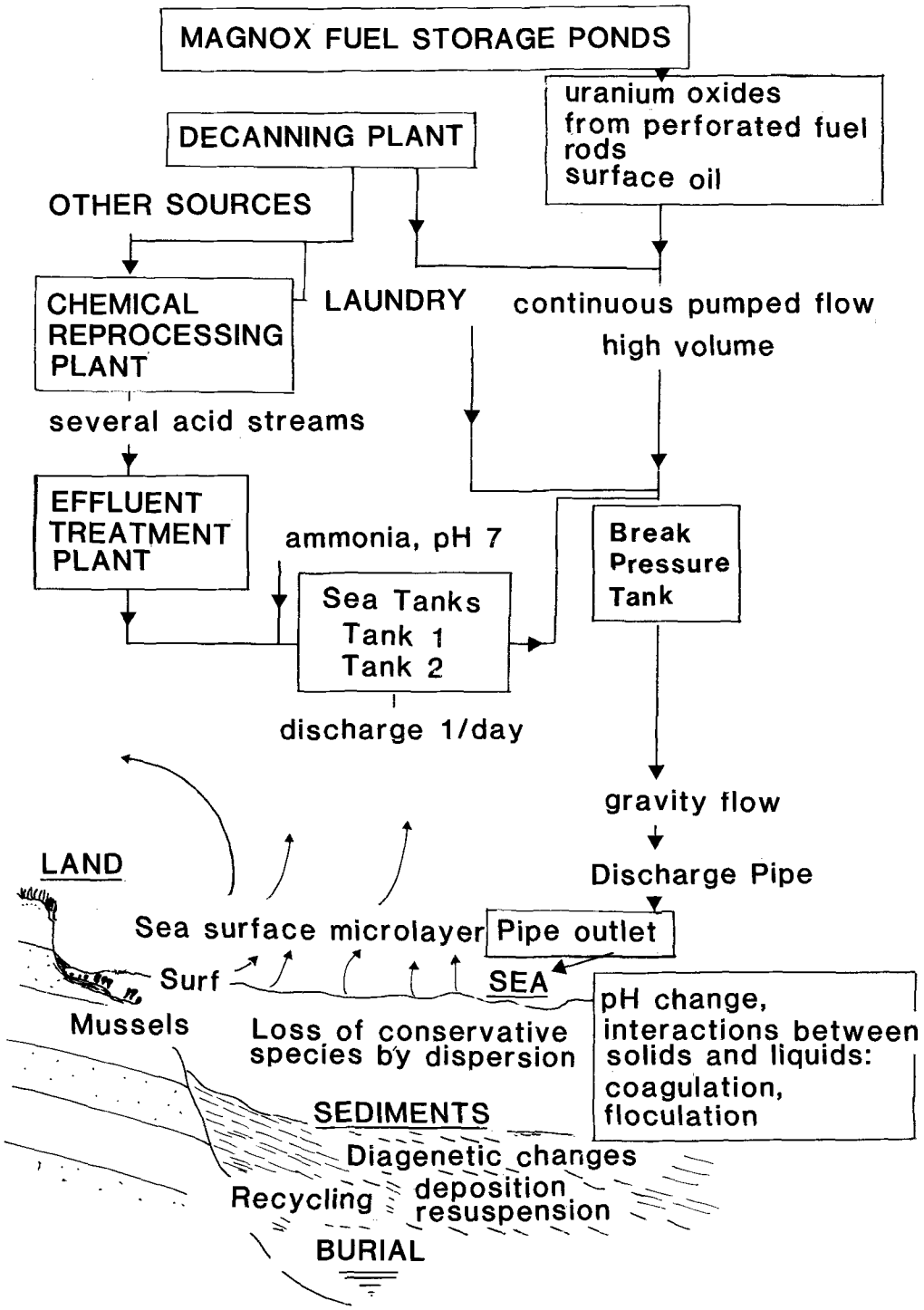


FIG. 1. Possible constituents of the BNFL effluent source term in relation to the production of hot particles.

term, released by BNFL at Sellafield into the NE Irish Sea.

An examination of the BNFL source term

No data are available on the chemical and physical form of the radionuclides in BNFL effluent; some possibilities considered in this paper are illustrated in fig. 1. If conservative or non-conservative forms are present in the source term they may require separate treatment when considering protection of the general public from sources of ionising radiation. Differences in the composition of the source term may also require consideration in modelling the behaviour of the radionuclides. Discussion will be mainly restricted to the transuranic radionuclides ^{241}Am, $^{239+240}Pu$, and ^{238}Pu, but some consideration will be given to *gross* beta-particle emissions.

The following assumptions are made in this investigation:

(i) The concentration of Pu and Am in various materials is considered in terms of their relative abundance (%), i.e.

$$\%\ ^{239+240}Pu \text{ in } \Sigma Am + Pu(\alpha) = \frac{^{239+240}Pu}{^{241}Am + ^{239+240}Pu + ^{238}Pu} \times 100$$

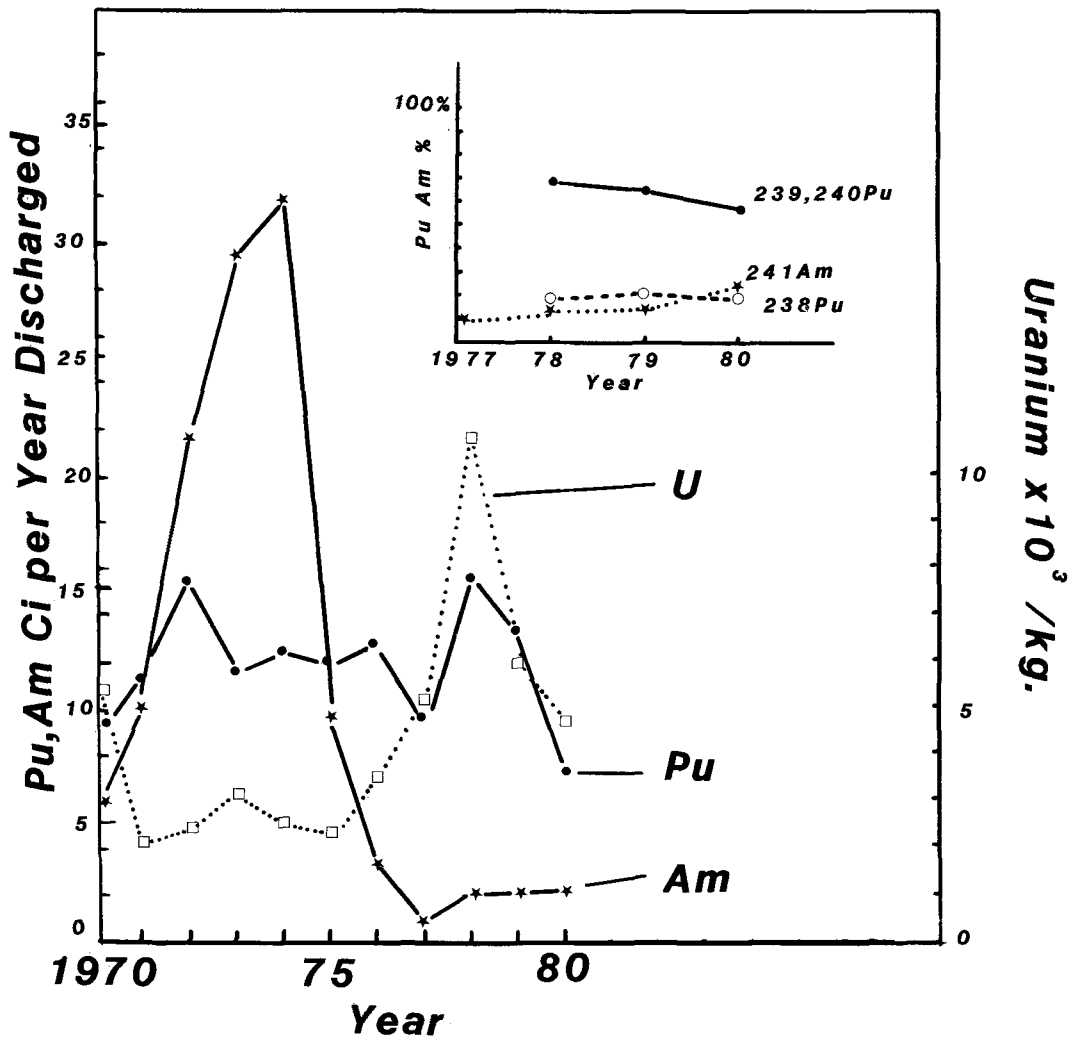


FIG. 2. Annual discharges of U (kg), $^{238,239,240}Pu$ and ^{241}Am(Ci) by BNFL Sellafield to the NE Irish Sea, 1970–80. (Insert—Data presented in terms of the relative abundances (%) for Pu and Am radionuclides.)

(ii) If the relative abundance (%) of Pu(α) and ^{241}Am in a sample is the same as that in spent Magnox fuel, then this is taken to indicate the presence of unaltered solid particles of Magnox fuel in the sample.

(iii) There are no natural processes whereby dissolved forms of either Am and Pu can recombine to give relative abundances identical to those characteristic of Magnox fuel or its products (i.e. fresh spent fuel, aged fuel, or products after reprocessing).

The period of study is 1977–80 for which published data for releases of U, Pu, and Am from BNFL are available (Atherton, 1979, 1980, 1981; Pentreath *et al.*, 1979) as illustrated in fig. 2 and the distribution of sampling sites is given in fig. 3.

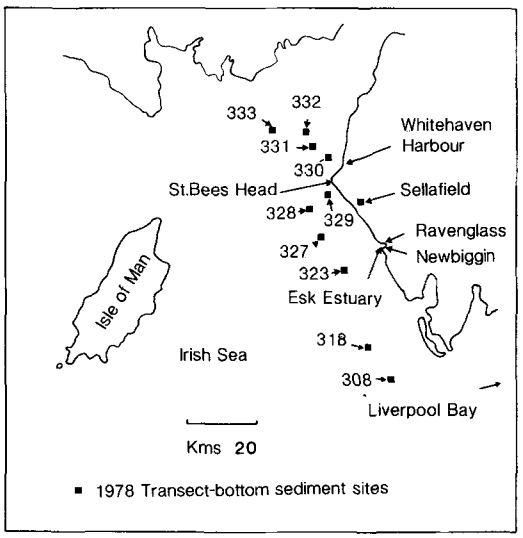


FIG. 3. Sample locations.

The relative abundance of the radionuclides studied in spent fuel and BNFL effluent are given in Table I. The main item to note is the relatively low abundance of ^{241}Am compared with radionuclides of Pu.

Relative abundance and concentration of ^{238}Pu, $^{239+240}$Pu near the BNFL marine outfall

Hetherington (1978) notes that *c.* 95% of all Pu released into the sea by BNFL is rapidly deposited in sediments; after the effluent is discharged it remains stratified for 3–4 days because of lower density as a tidal plug which is carried in a southerly direction by residual currents to Liverpool Bay, and then northerly by Atlantic water passing up the Irish Sea. Hetherington (1978) did not find any significant enhancement in the concentration factors for Pu in superficial sediment near the outfall which indicates an efficient dispersal, at least for Pu. The concentration and relative abundance of ^{238}Pu, $^{239+240}$Pu and ^{241}Am in superficial (5 cm) sediments of the NE Irish Sea, collected along an offshore transect (see fig. 3) passing Sellafield in November 1978, are given in Table II; these data are compared with those for sediments of the Esk estuary some 14 km to the south of Sellafield which contain the highest levels of radioactivity found within the region (Hamilton and Clifton, 1980; Hamilton and Clarke, 1984). All data have been normalized to values for sample 329 (Table II) which is nearest to the BNFL outfall.

The normalized data have also been considered in relation to their distance, in a northerly or southerly direction, from the outfall; fig. 4*a* illustrates the data for $\Sigma(\mathrm{Am}+\mathrm{Pu}[\alpha])$ pCi g^{-1} for each sample and shows that (in 1978) the total concentration of Pu and Am in surface sediments of the NE Irish Sea decreased with distance from the

Table I. Total Ci and relative abundance of ^{241}Am, $^{239+240}$Pu, and ^{238}Pu discharged into the NE Irish Sea, 1979–1980 compared to the relative abundance of Am, Pu in spent Magnox cooled fuel.

Year	Σ Total Ci discharged	Relative abundance (%)		
		^{241}Am	$^{239+240}$Pu	^{238}Pu
BNFL effluent[1]				
1978	1781	12.0	69.2	18.8
1979	1547	13.7	65.4	20.9
1980	959	23.3	57.4	19.4
Spent Magnox fuel[2] 2 x 10^{3}d cooled	–	8.3	70.4	21.4

1 – Cambray (1982) 2 – Mairs and Nair (1979).

Table II. The concentration (pC g^{-1}dry wt) and relative abundance of normalised values (%Norm.) for ^{241}Am, $^{239}+^{240}Pu$ and ^{238}Pu in sediments of the Irish Sea and Esk estuary

Sample No. Type Irish Sea (1978)	Distance from source, Kms. N = North S = South	$^{241}Am(\gamma)$[1] pCi g^{-1}dry wt.	$^{241}Am(\alpha)$[2] pCi g^{-1}dry wt.	%Norm[3]	$^{239}+^{240}Pu$ pCi g^{-1}dry wt.	%Norm	^{238}Pu pCi g^{-1}dry wt.	%Norm
308 Grey silt	52.0/S	5.2 ± 0.4	5.4 ± 1.3	0.8	6.7 ± 0.5	0.9	3.4 ± 0.3	2.0
318 Coarse sandy silt	41.2/S	7.9 ± 1.9	9.2 ± 2.0	1.0	8.6 ± 2.7	0.9	2.9 ± 0.2	1.3
323 Grey sandy silt	18.0/S	15.6 ± 2.6	17.4 ± 3.5	1.3	10.5 ± 0.7	0.8	3.7 ± 0.4	1.2
328 Grey sandy coarse silt	13.0/N	22.0 ± 3.9	24.7 ± 1.10	1.2	17.9 ± 0.9	0.8	5.9 ± 0.4	1.1
327 Grey sandy coarse silt	10.9/S	17.6 ± 1.2	18.6 ± 1.3	1.1	17.8 ± 1.2	1.0	3.9 ± 0.8	0.9
329 Grey silty fine sand	9.1/N	106.4 ± 3.5	108.8 ± 12.1	1.0	118.0 ± 22.7	1.0	27.7 ± 1.7	1.0
330 Grey silty mud	17.2/N	46.6 ± 2.3	45.0 ± 6.2	0.9	61.0 ± 9.8	1.1	16.9 ± 0.4	1.3
331 Grey fine silty sand	21.6/N	31.4 ± 0.5	31.0 ± 5.2	1.1	28.4 ± 0.7	0.9	9.2 ± 0.6	1.2
332 Brown grey fine sand	26.4/N	11.1 ± 0.5	10.7 ± 2.6	0.8	15.1 ± 0.1	1.1	4.4 ± 0.6	1.4
333 Brown grey fine sand	33.2/N	5.6 ± 0.6	5.2 ± 1.4	0.7	7.7 ± 2.3	1.0	3.5 ± 0.6	2.0
Esk Estuary (1980)	14/S							
Waberthwaite mud		-	62.9 ± 8.3	0.8	95.2 ± 5.0	1.1	26.1 ± 1.9	1.3
Newbiggin Viaduct mud		-	176.1 ± 11.1	1.0	210.5 ± 10.4	1.0	47.9 ± 3.8	1.0
Mussel Bed mud, Ravenglass		-	36.3 ± 4.0	0.9	45.2 ± 2.4	1.0	13.7 ± 0.9	1.3
Particulate debris suspended in water column, Newbiggin.		-	69.1 ± 8.0	0.9	85.5 ± 5.2	1.0	28.1 ± 3.9	1.4

1. Assay by γ spectrometry.

2. Assay by α spectrometry using surface barrier detectors.

3. eg. % Abundance for ^{241}Am for each sample normalised against the value for sample 329.

outfall with the sediment passing to the north having the highest activity. In fig. 4*b* the abundance (%) data for ^{241}Am shows a similar decrease in activity away from the outfall, but with slightly more ^{241}Am passing to the south. In fig. 4*c*, *d*, data are illustrated for the plutonium radionuclides; the trend is the reverse to that found for ^{241}Am and there is an apparent relative enhancement of Pu in a southerly direction, the most significant being for ^{238}Pu. Therefore Hetherington's observation (1978) that there is no build up of either Pu or Am near (*c.* 9 km) the outfall is confirmed, with a gradual decrease in concentration with distance from the outfall; however, examination of the relative abundance of the radionuclides shows an enhancement with distance for Pu, and a relative depletion in Am compared with the discharge site. This is not as expected since the decay system $^{241}Pu \xrightarrow{\beta} {}^{241}Am$, with a radioactive half life of 13.2 yrs., should enhance the pool of ^{241}Am, hence an apparent depletion in ^{241}Am relative to Pu includes any effect which is attributable to the decay of ^{241}Pu. It is assumed that the atoms of ^{241}Pu are homogenously mixed with those of other Pu radionuclides. Nevertheless this effect may be attributable to past differences in the amounts of Pu and Am in BNFL effluent (see fig. 2).

Although there is no evidence for the gradual build up of radionuclides in bottom sediments near (*c.* 9 km) the BNFL outfall, if significant amounts of particulate debris were present in the effluent, then some may be transported to intertidal zones by tides and accumulate with time in intertidal sediments. There are very few historical records through which such a process can be investigated. The gamma dose rate (μ rad h^{-1}) of intertidal sediments, between 1965 and 1980, reported in various FRL reports, has been selected as a measure of concentration processes. Sediments along the shore line at Sellafield consist of coarse-grained reworked sands; approximately equidistant to the north and south intertidal fine-grained silts are deposited at Whitehaven Harbour and Newbiggin respectively. Although sedimentation may differ at each site it is assumed to have remained constant

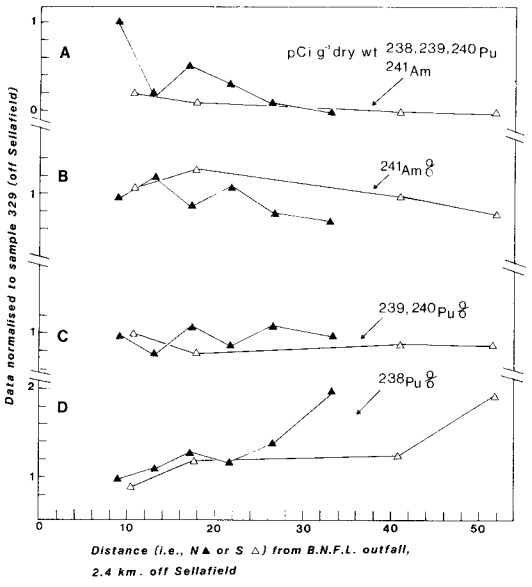


FIG. 4. The relative abundance (%) of Pu and Am normalized against sample No. 329 (see fig. 3) of the Irish Sea transect (see Table II), in relation to distance of sampling site from Sellafield. A Total $^{238+239+240}$Pu + ^{241}Am in bottom sediments; B ^{241}Am; C $^{239+240}$Pu; D ^{238}Pu. (N) northern direction. (S) southern direction.

with time; all three sites are within the high radioactivity zone offshore at Sellafield and therefore the dominant source of radioactivity will be from fresh BNFL discharges. The dose rates for Whitehaven Harbour and Newbiggin have been normalized to the value for Sellafield as illustrated in fig. 5. Overall the curves show a gradual decrease in levels of activity which can be related to discharges by BNFL. Relative to Sellafield, the Whitehaven data have remained fairly constant with time, but at Newbiggin there has been a gradual decrease; the high values of 1969 were caused by an unusual release of short-lived radionuclides which were transported preferentially to the south. Today both sites, relative to Sellafield, have similar dose rates which are about 4 times higher than those at Sellafield. The data illustrated in fig. 5 show that there has been no gradual build-up of radionuclides in intertidal sediments 10 km to the north or south of Sellafield, and hence they reflect the fairly rapid loss of radionuclides deposited on intertidal sediments as a result of either transport or burial.

Comments on the relationship between Pu and Am content of samples and grain size of sediment

It is generally accepted that Pu and Am is concentrated on the surface of sediment debris; hence the concentration of Pu and Am in a sample and grain size and surface area should be related. This topic will be discussed elsewhere; however, it should be noted that:

(i) There is an increase in Pu and Am content of a sample as the proportion of fine grained (< 60 μm) debris increases in this region.

(ii) For many sediments there is a more pronounced relationship between the total number of hot particles per unit mass of sediment and the total concentration of Pu + Am. Using 4 × 4 cm tablets of compressed sediment powder, exposed against the dielectric detector CR-39, the total number of hot particles recorded from a thick source can be determined. In fig. 6 the numbers of hot particles (normalized relative to sample 329, which is nearest the BNFL outfall) are given for various distances from the outfall, together with total Pu and Am content of the samples. The number of particles decreases fairly rapidly for the northerly flow direction, but is not so pronounced for the southerly flow. Correlation between the relative abundance of hot particles and Pu + Am content is good. Of the ten samples examined, six contained a greater abundance of < 60 μm sediment debris than was present in sample 329.

Recent studies indicate that a correlation exists between Pu and Am content and grain size for sands and other types of sediment which lack hot particles; this is not always the case for silts and muds, especially where organic-rich.

Relationships between the Am and Pu content of mussels in relation to those in sediment and BNFL effluent

Evidence derived from a study of the concentration of Am and Pu in tissues of the mussel is

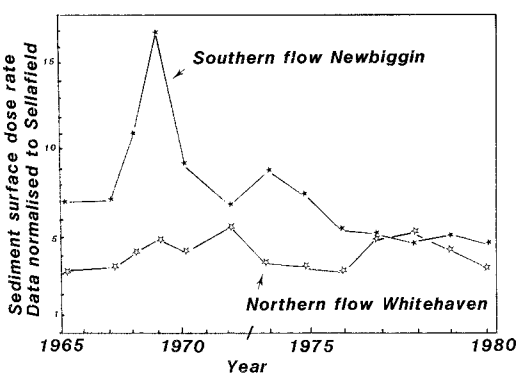


FIG. 5. Gamma dose μR (1965–80) for muds at Newbiggin and Whitehaven Harbour normalized against sands at Sellafield (see fig. 3).

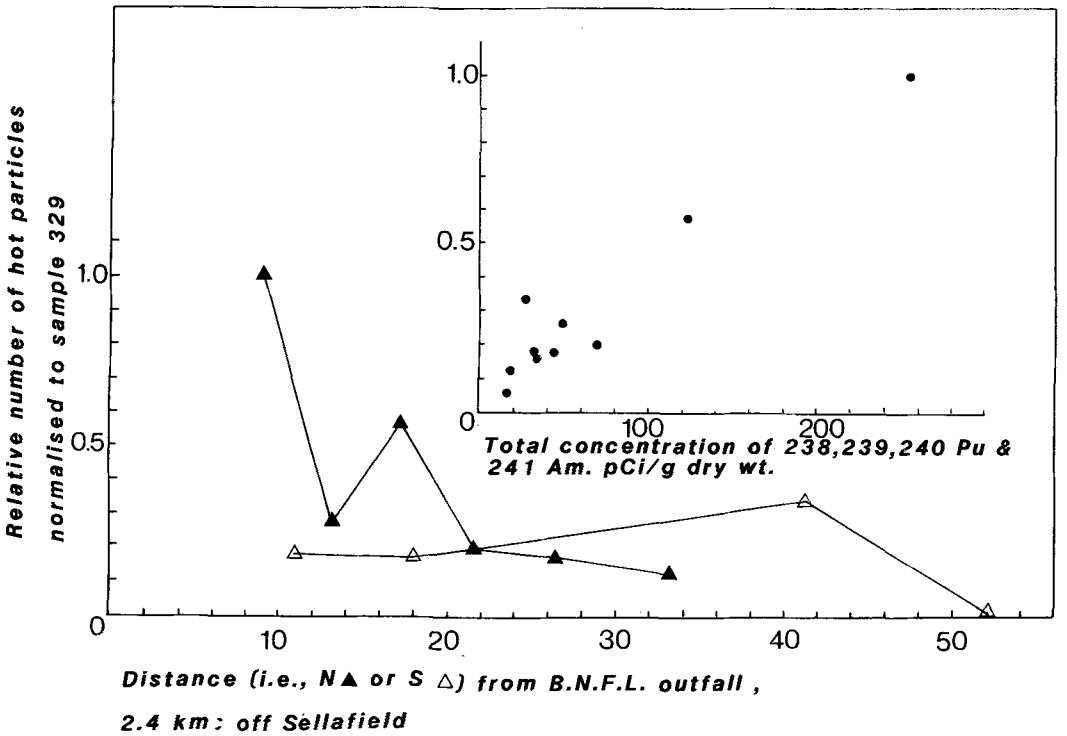


FIG. 6. The relative number of hot particles normalized to sample 329, nearest the BNFL outfall, in relation to (*a*) distance from the outfall in a northerly and southerly direction and (*b*) total concentration of $^{238,239,240}Pu + ^{241}Am$ in samples for the off-shore seabed transect.

considered next, to establish whether the mussel reflects Am and Pu levels in sediment or from a different source. Data are given in Table III and fig. 7 for the relative abundance of ^{241}Am, $^{239+240}Pu$, and ^{238}Pu in mussels calculated from data presented by Hamilton and Clifton (1980). The mussels were collected from the Ravenglass site (see fig. 3) between 1977 and 1979 and the data are compared with comparable values for mussels (total soft tissue) collected near the BNFL outfall (Hunt, 1979–81) and Newbiggin sediments, which have a composition very similar to those found at Ravenglass. The mussel only accepts particles into the digestive tract of a limited size range (Bayne 1976), although the range is not known precisely. Furthermore, Hamilton and Clifton (1980) indicated that the mussel derives some Am and Pu directly from seawater, together with small amounts which are absorbed in the digestive tract as a result of digestive processes. The highest concentrations of Am and Pu are found in the digestive gland and intestine and reflect the presence of ingested sediment. Hot particles are found (Hamilton 1981) in the digestive tract (see fig. 9*i*) and are also associated with granules present in the pericardial gland and kidney (Hamilton and Clifton 1980), which reflect ultrafiltration and excretory processes respectively. Am and Pu are retained in the mussel for a long time (Clifton *et al.*, 1983), although the pericardial gland, kidney and byssus have a more rapid turnover. The byssal threads, and to some extent the periostracum (organic covering to the external surface of the shell), may obtain a significant portion of their Am and Pu from seawater or adhering sediment (Hamilton, 1980).

Between 1973 and 1980 (fig. 2) the amount of Pu discharged into the NE Irish Sea has gradually decreased. Large amounts of ^{241}Am were discharged between 1972 and 1975, followed by a sharp decline. There was a gradual increase in the amount of uranium discharged from 1971 until 1978 and then a decline; the releases of U, ^{238}Pu, and $^{239+240}Pu$ at the peak are coincident, but were not noted in previous releases—indeed they generally vary inversely. The association of high U and Pu with low Am would be compatible with the release of unprocessed Magnox fuel debris by BNFL in 1978 or alternatively is related to the

Table III. A comparison between the relative abundances (%) of ^{241}Am, $^{239+240}Pu$, ^{238}Pu in mussels from the Esk (Ravenglass), Windscale shoreline near BNFL and Esk muds (Newbiggin).

Organ/Tissue	^{241}Am	$239 + 240Pu$	^{238}Pu	^{241}Am	$239 + 240Pu$	^{238}Pu	^{241}Am	$239 + 240Pu$	^{238}Pu
	1977			1978			1979		
Ravenglass samples (1)									
Total soft tissues	73.1	21.7	6.0	38.8	46.3	15.0	14.9	70.2	14.9
Digestive gland	65.4	24.7	9.8	43.1	45.2	11.7	6.4	79.0	14.6
Kidney	53.6	35.9	10.5	62.0	13.1	17.2	7.2	84.8	8.0
Mantle	78.8	16.7	3.9	48.1	39.0	13.0	23.6	64.6	11.8
Mantle edge	61.0	30.5	8.5	49.0	37.3	13.7	25.6	64.7	9.8
Gill	65.8	29.2	4.9	55.2	27.1	17.7	21.8	84.4	13.5
Muscle	74.2	18.2	7.6	35.9	38.9	22.1	34.8	53.6	11.6
Foot	-	-	-	3.4	68.4	28.2	15.0	67.5	17.5
Periostracum	50.0	39.0	11.2	55.5	31.9	12.6	-	-	-
Byssal threads	27.5	62.1	10.4	24.6	54.5	20.9	17.3	68.1	14.6
Windscale shoreline (2)									
Total soft tissues	40.1	47.6	11.7	39.8	48.0	12.1	35.1	51.9	13.0
Newbiggin muds (3)	42.0	45.8	12.3	36.6	50.9	12.5	31.7	54.9	13.4

(1) Clifton, Stevens Hamilton (1983) (2) Hunt 1979, 1980, 1981 (3) Hamilton and Clifton 1980.

composition of the source term without consideration of the solid content. In comparing the relative mass abundance of Am and Pu, there was a small decline in Pu between 1978 and 1980 and a slight increase in Am, albeit small. Fig. 7 shows that there is only a small difference between the abundance of Am and Pu in Windscale mussels (total soft tissue) and Newbiggin mud; hence it can be concluded that the source material has the same composition which has not changed during transport from Sellafield to Newbiggin and which would be compatible with the movement of Pu and Am in, or on, particles. The Pu/Am ratio is *c.* 1, which is similar to that found for regional sediments, providing that allowance is made for any significant changes in the amounts of these radionuclides which are released by BNFL at a particular time. Apart from the ^{241}Am content (pCi g^{-1} dry wt.) of byssal threads for Ravenglass, which appear to be similar to those observed for Windscale mussels (total soft tissue) and Newbiggin sediment, the Pu burdens indicate a more readily available direct source of Pu than for Am. Fresh BNFL effluent, which has not been mixed with the regional reservoir of Am and Pu present in sediments, is one possible source, especially Magnox fuel debris suspended in the water column, although the amounts present are small. In 1977, Hamilton (1981) observed an enrichment of Pu relative to Am in particulate debris collected from the sea surface which coincided with increased releases of uranium by BNFL, in the period 1971-6, of 3000 kgU $yr.^{-1}$ (1977—5268 kg U, 1978—10936 kg U, and 1979—6000 kg U). Hamilton and Clarke (1984) determined a lag-time for transport of Pu and Am from Sellafield to Ravenglass/Newbiggin of *c.* 1 yr., hence an increase in Pu in mussels from Ravenglass in 1979 may reflect the 1978 releases of uranium by BNFL.

In sampling mussels at Newbiggin, time and spatial variability has been reduced to a minimum by selection of animals: at the same time of the year; at similar stages of the Condition Index; of sizes between 5 and 7.5 cm. Animals removed from the same area were randomly divided into two groups, one being used for total soft tissue analysis and the other for the dissection of individual organs. Total soft tissue of mussels collected at Newbiggin and Windscale appear to reflect the composition of local sediment, but this is not observed for individual tissues and organs from mussels sampled at Newbiggin. If allowance is made for biological variability (*c.* 30%) between mussels, the trend of relative enrichment in Pu and/or relative depletion in Am, for most tissues, suggests that the mussel does not differentiate between Pu and Am, either because it cannot, or because both Pu and Am are not recognized by biological processes and are treated in the same manner; although numerous reports indicate that Am is more mobile in biological systems than Pu. An alternative hypothesis suggests that the animal does not react to Pu or Am because both are present or associated with particulate debris which would be distributed throughout the body tissues of the mussel in a random

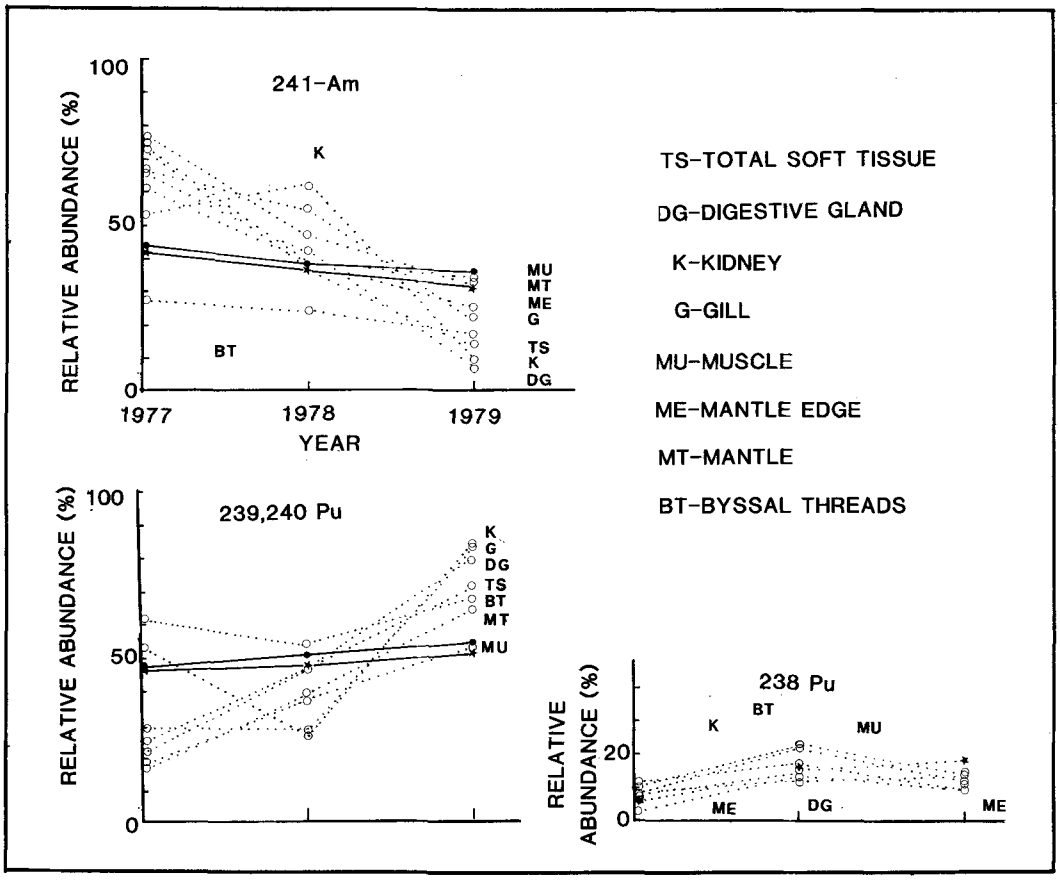


FIG. 7. The relative abundances (%) of Pu(α) and Am in various organs of *Mytilus edulis* from Ravenglass, compared to total soft tissue for animals sampled near Sellafield and muds from Newbiggin for 1977–9. Solid circles: Newbiggin muds. Stars: Windscale shoreline mussels (TS).

manner and be controlled by the animal's macrophage-lyosome system.

A significant source of Pu resulting from oxidation of reduced sediments (Pu^{4+}–Pu^{6+}) during tidal scouring is not likely as field evidence indicates that Pu is retained in the anaerobic muds and the associated flocs, which are formed during tidal disturbances. Sand could constitute a source of Pu^{6+}, since a large volume of sandy sediment with only trace quantities of Pu and Am is involved in tidal disturbance.

Composition of aerial dispersed debris from BNFL

Data are not available for the physical and chemical composition of radionuclides which are released into the atmosphere from the BNFL stacks; neither is there any information on the contribution to ground deposition which is derived from general wind blown debris off the BNFL site, for example from the uncovered Magnox fuel storage ponds. In the vicinity of the site there will be an additional source of radionuclides derived from marine aerosols, especially from the off-shore region where the BNFL effluent is discharged into the sea. In several transects Cambray and Eakins (1982) report the concentrations of Pu and Am present in soil profiles taken to a depth of 15 cm. Near the sea-shore the relative abundances of Pu and Am radionuclides are similar to those for local marine sediments, but passing inland, and up to a distance of 10–20 km, the abundance of ^{241}Am relative to $^{238+239+240}Pu$ decreases until the deposited aerosol reflects a composition which is similar to that for general global fallout debris of the UK, which is characterized by higher levels of Pu than Am. Similarly an examination of data for the composition of sea spray, described by Eakins

et al. (1982), shows a small depletion in the abundance of ^{241}Am relative to Pu at the coast near Sellafield, and with increasing distance from BNFL the composition also becomes similar to that for average marine sediments of the region. The number of atoms or particles of Am or Pu m^{-3} of air decreases with distance from the source, if particulate debris is present, although isolated samples of soil can contain much higher concentrations of Pu and Am presumably resulting from the presence of particulate debris. Gorham (1958) concluded that almost all the sodium and chloride ions in Cumbrian tarns (altitude 15–275 m) is derived from sea spray; therefore radionuclides derived from BNFL would be expected to be present at least up to 50 km inland and possibly across the whole of northern England. Therefore while almost all of the Pu and Am deposited in Cumbria is derived from global fallout debris there is a need to determine whether particulate debris does penetrate further inland than current estimates indicate. Research on this topic is currently being undertaken using media which concentrate radionuclides, such as bottom deposits of lakes and selective biological accumulators.

Hot Particles

The discussion has centred so far on identifying features which illustrate the coherent or separate behaviour of Am from Pu, in geochemical and biological systems which involves studies of conservative or non-conservative species of Am or Pu. The methods in common use are often arbitrary, for example the process of filtration is essentially concerned with the separation of solids from liquids, which is determined by the pore size of the material used for filtration, however, the properties of the residue controls what is passed through the filter paper and hence the accumulating residue can influence the separation. The commonly used 0.45 μm filter (operating efficiently) allows a considerable number of particles through.

In order to overcome some of the artefacts of separating conservative from non-conservative species in the laboratory, measurements are made directly on natural samples as described by Hamilton and Clifton (1981). There is, however, no clear definition of a hot particle, especially in relation to hazards from ionizing radiation; as the autoradiographic exposure time increases, the number of hot particles also increases. Exposure times of between 1 hr. and 600 days are used, which identify a hot particle as associated with a level of radioactivity having a small volume, which is considerably higher than that from surrounding areas. It is usually difficult to determine the composition of an individual hot particle, or the true size of particles with a MPD of < 5 μm; however, it is assumed that for smaller particles, solids are less important than solution processes. In the Cumbrian region, alpha-active hot particles usually account for at least 10% of the total radioactivity of a sample, but this may not be true for gamma-active particles, especially those found in sediments.

Beta-gamma hot particles are illustrated in fig. 8, from a sediment profile of the Esk estuary, Cumbria, where the following features are observed:

(*a*) The radioactivity is present in well-defined layers which reflect the annual releases of radionuclides by BNFL (Hamilton and Clarke, 1984).

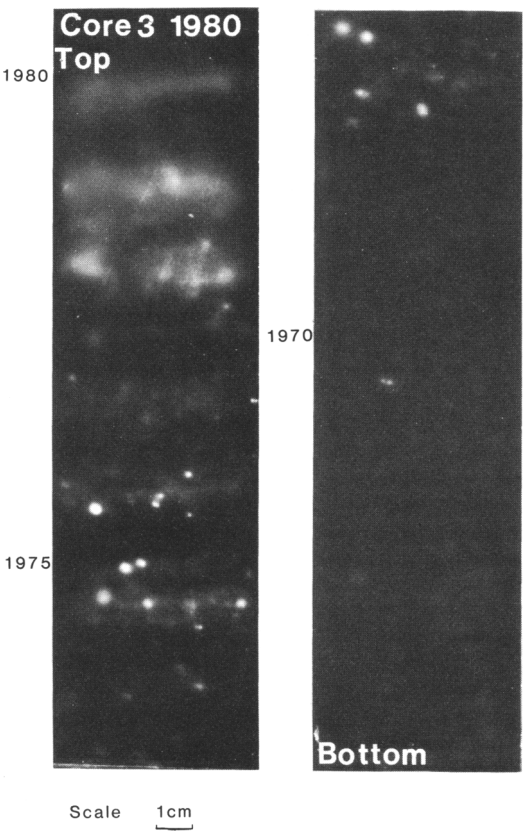


FIG. 8. Beta/gamma autoradiograph of a silt core taken from the Esk Estuary (sedimentation rate of 3.5 cm^{-1} yrs.) using Kodak Industrex C X-ray film. (White areas represent regions of high radioactivity.) Note: The zones of high radioactivity can be related to deposition of sediments between May and October; in the winter months relatively little radioactivity is retained in the sediments. Pre-1970 sediments do not appear to contain many beta/gamma hot particles.

The zones of lower activity represent deposition of sediment during the winter when there is less retention of radioactive debris because of the dominance of high energy conditions. The zones of high radioactivity, towards the top of the sediment core reflect the presence of higher concentrations of short-lived radionuclides (^{95}Zr, ^{95}Nb, ^{106}Ru) which obscures the hot particles.

(*b*) Most of the ^{137}Cs activity in the core is associated with discrete particles; 40–60% of the ^{137}Cs can be removed by acid leaching which reflects the partial dissolution of particles. Hence as the ^{137}Cs activity is also associated with other gamma emitters characteristic of BNFL effluent, the particles either represent original BNFL effluent as released, the formation of particulate matter when the effluent enters the sea, or the presence of Magnox fuel debris. The distribution of radioactivity throughout the core is not compatible with the adsorption of ^{137}Cs, which is present in the water of the river Esk, as non-conservative species on clay minerals. It is therefore concluded that the retention of BNFL effluent in the Esk is influenced by the nature of the BNFL source term, in particular the presence of solid phases. However, most of the ^{137}Cs released by BNFL is transported as conservative species in seawater and can be traced as far afield as Greenland and the Baltic.

(*c*) It is observed that the number and shape of hot particles tend to change with depth in the sediment (Hamilton, 1981; Hamilton and Clarke, 1984). Angular shapes are present at the sea/water interface and at the surface of the sediment, but disappear with increasing depth, where they are replaced by rounded forms. This probably reflects a general solution process which occurs principally near the sediment surface, which is subject to many periods of disturbance and redox recycling and weathering, throughout the year.

Alpha particle autoradiography, illustrating various types* of alpha hot particles are shown in fig. 9. *In situ* exposures for 2 hrs. at Newbiggin, using the dielectric detector CR39, have identified the presence of a few hot particles; some of the more common types recorded are as follows:

(*a*) 'Golf balls', defined as very hot particles where the detector has been totally burned out.

(*b*) Hot particles with well-defined forms generally tabular in nature, and which are believed to represent fresh BNFL debris.

(*c*) Very small hot particles, (1–5 μm MPD) which are only just visible above the diffuse background levels.

(*d*) Particles with a well-defined form, indicating that they are either fragments of larger original debris or composite particles, part of which is inactive.

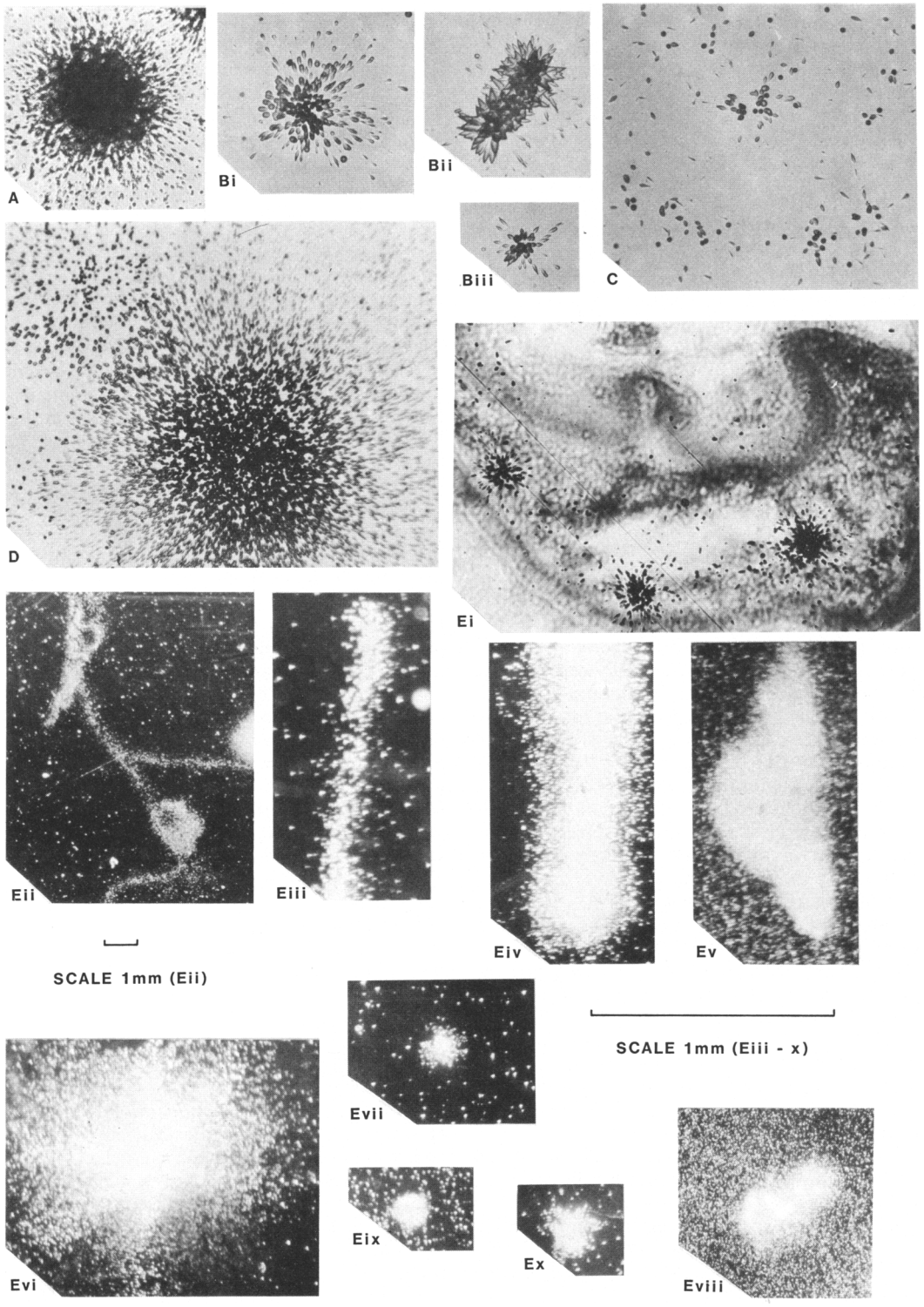
(*e*) Biological debris represents some of the most radioactive types of macro hot particle found in the Esk. Although still subject to study, it is clear that fragments of byssal thread, and especially the terminal caps, at all stages of degradation do give rise to many of the macro-hot particles found on the surface of Esk sediment. Other evidence indicates that the alpha-activity associated with the byssal threads is only superficial, therefore the specific alpha-activity of the tanned surface protein of the threads will be very high.

This paper does not consider the question of whether or not a hot particle, if inhaled, constitutes a risk of lung cancer. For members of the general public within the Cumbrian region, no information is available for intake and retention of such particles. For the sake of completeness their concentrations and particle sizes should be determined together. Considerable gaps exist in our knowledge concerning inhalation of particulate debris; at present it is not possible to specify rates of transfer of airborne particulate debris from the environment to the human lung. Much of the highly radioactive debris present in the region, especially biological debris, is unlikely to become airborne unless surfaces of sediment are dry. The size of the hot particles range from sub-micron to at least a few mm MPD. The most likely sites for the generation of hot particles (into the air) would be from mud flats and vegetation rather than high-energy regions such as those associated with the surf-zone of sandy foreshores. Of particular interest is the extent to which large hot particles are trapped in the naso-pharyngal cavity, a site known to be sensitive for the induction of cancers (i.e. woodworkers). At present the ICRP Lung Model does not consider the nasal-pharyngal areas for the site of induction of cancers as a result of exposure to ionizing radiation, but it is recognized that the tissues are sensitive to damage.

Radioactivity of hot particles

At their present stage of development the techniques used in this work do not permit an accurate estimation of the size of submicron hot particles. However, if a sample contains discrete sources of highly radioactive submicron particles emitting alpha particles, their presence would easily be identified. In the CR-39 etching technique (Hamilton and Clifton 1981) a *c.* 5 MeV alpha will give rise to tracks *c.* 10 μm in total length, hence a < 1 μm particle will give rise to a hot particle with a maximum diameter (for the extreme ends of the

* This study only concerns hot particles with a MPD of > 1 μm.


A
Bi
Bii
Biii
C
D
Ei
Eii
Eiii
Eiv
Ev
SCALE 1mm (Eii)
SCALE 1mm (Eiii - x)
Evi
Evii
Eviii
Eix
Ex

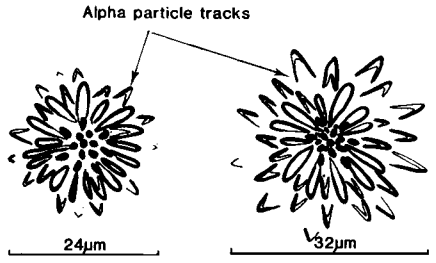


FIG. 10. Diagrams illustrating features of α-track length produced in CR.39 for: (*a*) BNFL particle (Pu,Am,Cm); (*b*) Natural particle (U + Th decay series).

tracks) of *c.* 20 μm which would clearly be identified as illustrated in fig. 10.

An illustration of one class of hot particle ('golf balls') present in suspended particulate debris in 51 aliquots of seawater collected throughout one tidal cycle in the Esk is given in fig. 11, and shows that the radioactivity of a few particles approaches the *c.* 0.6 pCi level identified by NRDC. Data given in Table IV compare the alpha-activity of hot particles removed from the surface of sediment as determined by ZnS scintillation counting and surface barrier alpha-particle spectrometry; except for the particle with the highest activity, the agreement between both techniques is good.

The routine methods deployed so far only serve to identify one class of hot particle, namely those which have levels of radioactivity far in excess of the background levels (diffuse activity). However, by using autoradiographic exposure times of up to 600 days the general picture (hot particle: diffuse activity) does not change, hence it is concluded that isolated particles containing > 1 pCi are probably rare. Using the data of Mairs and Nair (1979) a *c.* 10 pCi particle of fresh unprocessed Magnox fuel which has cooled for between one and ten years will have a MPD of *c.* 8 μm, whereas reprocessed fuel material (99.9% Pu and U removed) will have an activity of *c.* 0.1 pCi. Hence it can be concluded that while acknowledging uncertainties in the measurements, in the Esk region at least, particles of fresh Magnox fuel or fresh BNFL effluent are present. In maintaining a correct perspective which links hot particles and possible effects arising from ionizing radiation it should be noted that Fews and Henshaw (1981) found small clusters of alpha-active particles, containing between 10^{-4} and 10^{-1} pCi of activity, in the human lung of non-occupationally exposed people. The alpha-particle activity arises because of the presumed presence of common accessory minerals of igneous rocks, such as zircon, which are ubiquitous in most soils. Data are given in Table V for the U, Th, and Zr content of some

Table IV. Total ∝activity, $^{239}+^{240}Pu$, $^{238}Pu + ^{241}Am$, ^{242}Cm and $^{243}+^{244}Cm$ activities (pCi g^{-1}dry wt) for hot particles analysed in situ.

Sample No.	Total Activity ZnS pCi g^{-1}	Surface barrier measurements pCi g^{-1}				
		Total Activity	$^{239}+^{240}Pu$	$^{238}Pu + ^{241}Am$(1)	^{242}Cm	$^{243}+^{244}Cm$
121	1.90	4.82	3.3	3.3	0.2	0.02
141	0.32	0.38	0.14	0.11	0.1	0.03
131	0.43	0.63	0.31	<0.32	<0.1	<0.01
128	0.82	0.89	0.59	0.30	ND	ND
109	0.068	0.06	0.04	0.02	ND	ND
94	0.59	0.53	0.38	0.15	ND	ND

(ND - Not detected)

(1) Using a surface barrier detector and without chemical separation; energies from ^{238}Pu and ^{241}Am cannot be resolved.

FIG. 9 (*opposite*). Track lengths *c.* 10 μm. A 'Golf balls' where the detector is saturated. B Highly radioactive particles, often tabular in shape, which are considered to represent fragments of Magnox fuel debris. C < 1 μm highly radioactive particles. D Large particles with well-defined form; often found adhering to sand grains. E (i) Cross section of the digestive tract of the mussel (Ravenglass) showing the presence of discrete hot particles in the lumen. Exposure time 166 days. Activity of particles *c.* 0.0006 pCi. (ii) *Macro Particles* (Dark field illumination). (iii) Byssus of *Mytilus edulis* illustrating threads and caps. (iv) Higher magnification of a portion of byssal thread. (v) Very high activity byssal thread. (vi) Degraded byssal cap. (vii) Fresh byssal cap illustrating stellate structure. (viii-x) Byssal caps illustrating various stages of degradation.

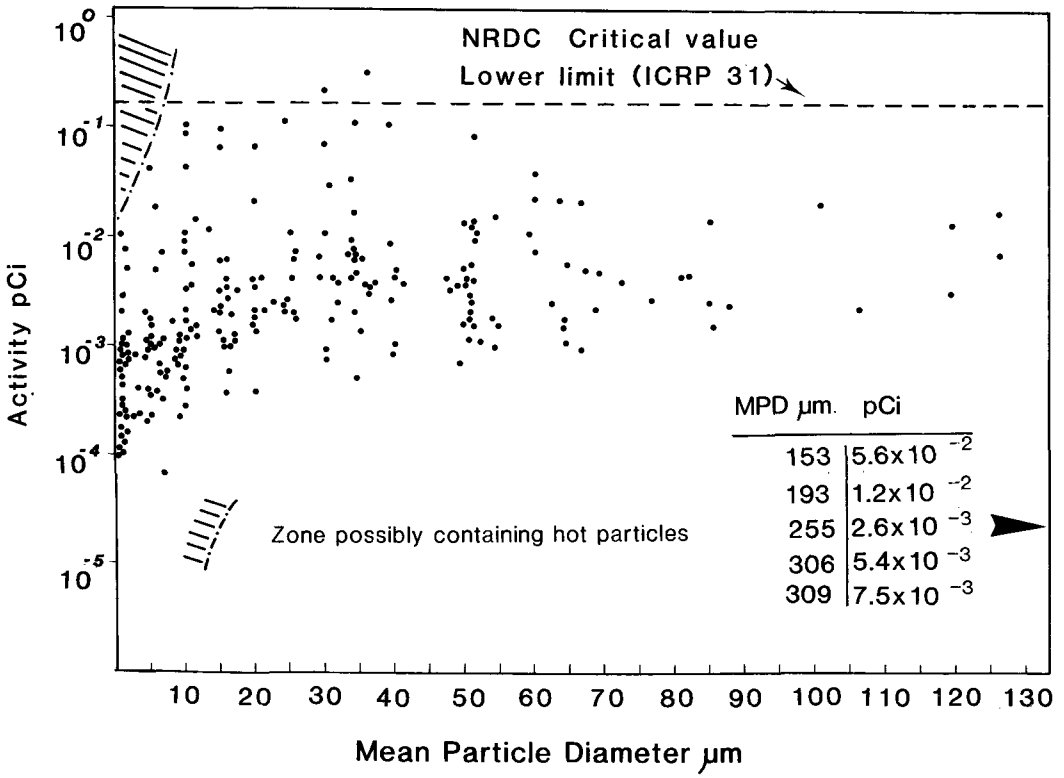


FIG. 11. Activity and particle size of hot particles, detected in CR.39, for a series of samples filtered (0.22 μm) from seawater throughout a tidal cycle, Esk Estuary, Cumbria.

human tissues (Hamilton, 1979) which show an enhancement in U and Zr in lung and lymph nodes which may support the observations of Fews and Henshaw (1981), although it should be noted that in recent years Zr has been widely used in various deodorant aerosols.

Conclusions

1. An examination of the relative abundance of Pu and Am in sediments and tissues of the mussel along the Cumbrian coast and nearshore sediments indicates changes in composition with distance from the source. The observed trends are considered more likely to reflect those related to geochemical and biological processes rather than variability in composition of Pu and Am in the fresh effluent.

2. The gamma-dose rate from coarse grained sands of the Sellafield foreshore has been used as an index of radionuclide emissions to the marine environment. However, the direction of movement of the sediment is not constant, although movement to the north and south of Sellafield has been similar in recent years.

3. The BNFL effluent discharged into the sea contains solid debris, some of which has characteristics compatible with relatively unaltered Magnox fuel.

4. In the Esk, some hot particles have activities which are similar to those identified by NRDC as being of concern; more recent measurements, using different techniques, have shown that high activity

Table V. The concentration of U, Th, Zr (μg g^{-1} wet wt) in some human tissues (from Hamilton, 1979)

Organ	U	Th	Zr
Blood (whole)	0.0008 ± 0.0001	0.002 ± 0.0004	0.02
Liver	0.0003	-	0.03
Lung	0.001	0.01	0.06
Lymph Nodes	0.01 ± 0.002	0.2 ± 0.1	0.3
Muscle	0.0002 ± 0.001	-	0.02
Bone	0.007	<0.04	<0.1

particles with a MPD of < 1 μm are also present. The activity of the particles found in the Esk are similar to those found in the lung of non-occupationally exposed individuals.

5. Hot particles have also been observed which consist of debris from the byssus of the mussel and contain Pu and Am which is believed to be derived from seawater. With erosion of byssal debris in the marine environment, the activity of these particles is likely to decrease rapidly as the surface coatings are worn away; if Magnox particles are present, solution of the uranium is likely to result in the production of particles with a much higher specific radioactivity. A single (*c.* 1×3 μm) particle, if coated with a single layer of ^{241}Am (close packing) would contain *c.* 8×10^6 hydrated Am nuclei and would give rise to a *c.* 0.01 pCi particle whose activity is a factor of about 60 below the NRDC value.

6. In the Esk sediments various gamma and beta particle (^{137}Cs) emitters are present in hot particles; hence it is not acceptable to consider the radioactivity of these sediments simply in terms of adsorption by conservative species of radionuclides on to mineral debris.

7. An examination of the composition of Pu(α) + Am in mussels collected near Windscale indicates that while total soft tissues contain Pu and Am in amounts which are similar to those found in fine-grained silts, those sampled some 14 km from the BNFL outfall at Newbiggin show a relative enrichment in Pu, and a relative depletion in Am, which possibly indicates a different source for these two radionuclides. This may reflect indiscriminate uptake by the mussel of fine-grained particulate debris which is present in BNFL effluent and has been rapidly transported in the sea to the Esk from Sellafield (Windscale).

8. The composition of the source term does exert an influence on the distribution of radionuclides which are released into the Irish Sea by BNFL. The effluent does contain solid debris, some of which is transported to the Esk estuary and is found in sediments. A radionuclide such as ^{137}Cs is dispersed over very large distances simply because in the open sea most of it is present in a conservative form. However some of the ^{137}Cs is also associated with particulate forms and is present as a constituent of the sediment. When using models to evaluate the transport of radionuclides released by BNFL into the marine environment there may be a need to define more precisely their distribution between conservative and non-conservative phases with respect to relative abundances in the source term. Such features may be important when using general open-ocean models to predict transfers for the near-shore environment, especially following accidental or unusual releases of radionuclides. From the evidence available, for the Esk, it appears that, once deposited in sediments, there is little subsequent migration of radionuclides associated with hot particles. Clearly, until samples of BNFL effluent can be examined, there are some areas of doubt concerning any evaluation of the transport of radionuclides off the Cumbrian coast. There is little evidence to indicate that such studies will identify an unusual hazard from ionizing radiation but they are likely to improve our understanding of geochemical and biological processes which control the observed distribution of radionuclides in coastal waters which receive effluent from the reprocessing of uranium fuel.

Future research requirements

The nature of hot particles released by BNFL, under normal working conditions has not been adequately characterized so far. Their relevance as a significant component in the transfer of radionuclides to man has not been evaluated for Cumbria, and deserves further study. Changes in the criteria used in dose assessment, for example a change in the gut transfer factor for Pu, or the selection of different critical groups for an evaluation of non-occupation exposure (see Hunt, 1983), may indicate a need to determine the pathways whereby hot particles can enter man. In predictive evaluation, and in modelling the pathways of radionuclide to man, more attention should be directed towards the presence of hot particles and their availability to man, by taking into account changes in their composition and forms from the point of discharge to eventual sites of deposition which should include data on the direction of movement for nearshore sediments off the Cumbrian coast.

Acknowledgements. This work forms part of the environmental radioactivity programme of the Institute for Marine Environmental Research, a component body of the Natural Environment Research Council, and was supported in part by funds provided by the Commission of the European Communities, Contract BIO-B-438-81-UK. I express my sincere thanks to R. J. Clifton, R. S. Cambray, P. J. Moore, R. J. Pentreath, D. F. Jefferies, G. J. Hunt, R. S. Atherton, H. Longley, and J. Vennart for useful discussions. The opinions expressed in this paper are those of the author only. J. M. Miller of the NERC, Institute of Geological Sciences provided samples of bottom sediments from the Irish Sea and H. E. Stevens provided technical assistance.

REFERENCES

Atherton, R. S. (1979) British Nuclear Fuels Ltd., Health and Safety Directorate. *Annual Report on Radioactive*

Discharges and Monitoring of the Environment, 1978, p. 11. Risley, Warrington, UK.
——(1980) Ibid. 1979, p. 11.
——(1981) Ibid. 1980, p. 11.
Bayne, B. L. (1976) *Marine Mussels: Their Ecology and Physiology*. Int. Bio. Prog. 10. New York, Cambridge University Press, pp. 506.
Cambray, R. S., and Eakins, J. D. (1982) *Nature*, **300**, 46–8.
Camplin, W. C., Durance, J. A., and Jefferies, D. F. (1982) *A marine compartment model for collective close assessment of liquid radioactive effluents*. Sizewell Inquiry Series No 4. MAFF/S17 Lowestoft, UK.
Clifton, R. J., Stevens, H. E., and Hamilton, E. I. (1983) *Concentration and depuration of some radionuclides present in a chronically exposed population of mussels (Mytilus edulis)*. Mar. Ecol. Prog. Ser. 11, 245–56.
Durbin, P. W. (1973) In *Metabolism and biological effects of the transplutonium elements* (H. C. Hodge, J. N. Stannard, and J. B. Hursh, eds.) 739–896. Springer Verlag, Berlin.
Eakins, J. D., Lally, A. E., Burton, P. J., Kilworth, D. R., and Pratley, F. A. (1982) *Studies of Environmental Radioactivity in Cumbria*. Pt 5. (AERE-R10127), HMSO, London.
Fews, A. P., and Henshaw, D. L. (1981) *Analysis of uranium fragments found in the human lung*. In *Int. Conf. on Solid State Nuclear Track Detectors*. Bristol, UK.
GESAMP Dent No. 19 (1983) *An oceanographic model for the dispersion of wastes disposed of in the deep sea*. IAEA, Vienna.
Gorham, E. (1958) *Phil. Trans. R. Soc. Lond. Ser. B*, **241**, 147–78.
Great Britain-Parliament (1960) *Radioactive Substances Act 1960*. HMSO, London, 28 pp.
Hamilton, E. I. (1979) *The Chemical Elements and Man*. C. C. Thomas Pub. Springfield. Illinois USA, 386–94.
——(1980) *Mar. Ecol. Prog. Ser.* **2**, 61–73.
——(1981) *Nature* **290**, 690–3.
—— and Clarke, K. R. (1985) Sedimentation history of the Esk estuary, Cumbria UK: the application of radiochronological methods. *Sci. Total Environ.* **35**, 325–86.
—— and Clifton, R. J. (1980) *Mar. Ecol. Prog. Ser.* **3**, 267–77.
——————(1981) *Int. J. Appl. Rad. Isotopes.* **32**, 313–24.
Hetherington, J. A. (1976) *Radioactivity in surface and coastal waters of the British Isles, 1974*. Tech. Rept. FRL 11, pp. 35.
——(1978) In *Environmental Toxicity of Aquatic Radionuclides: Models and Mechanisms* (M. W. Miller and J. N. Stannard, eds.) Ann Arbor Science Pub. Inc. Mich., 81–106.
Howells, H. (1977) *British Nuclear Fuels Ltd., Windscale and Calder Works, Health and Safety Department. Radioactive waste disposal and associated monitoring data 1971–1976*.
Hunt, G. J. (1979) *Radioactivity in surface and coastal waters of the British Isles, 1977*. Aquat. Environ. Monit. Rep., MAFF Direct. Fish. Res. Lowestoft (3), 36 pp.
——(1980) Ibid. 1978 (4), 37 pp.
——(1981) Ibid. 1979 (6), 32 pp.
——(1982) Ibid. 1982 (8), 35 pp.
——(1983) Ibid. 1981 (9), 36 pp.
International Commission on Radiological Protection (1972) *The metabolism of compounds of plutonium and other actinides*. Pergamon Press, Oxford (ICRP, Pub. 19).
——————————(1980) *Biological effects of inhaled radionuclides* (ICRP, Pub. 31).
——————————(1981) *Limits for intakes of radionuclides by workers. Part 1* (ICRP, Pub. 30 Part 1).
Mairs, J. H., and Nair, S. (1979) *The inventories of actinide and fission product arisings in spent nuclear fuel: results from the Rice Code*. Central. Elect. Gen. Board. Res. Div. Berkeley Nuclear Labs. RD/B/N4579, UK.
Mitchell, N. T. (1967) *Radioactivity in surface and coastal waters of the British Isles*. Tech. Rept. FRL 1, pp. 36.
——(1968) Ibid. 1967. Tech. Rept. FRL 2, pp. 41.
——(1969) Ibid. 1968. Tech. Rept. FRL 5, pp. 39.
——(1971*a*) Ibid. 1969. Tech. Rept. FRL 7, pp. 33.
——(1971*b*) Ibid. 1970. Tech. Rept. FRL 8, pp. 34.
——(1973) Ibid. 1971. Tech. Rept. FRL 9, pp. 35.
——(1975) Ibid. 1972–3. Tech. Rept. FRL 10, pp. 40.
——(1977*a*) Ibid. 1976, Part 1. The Irish Sea and its environs. Tech. Rept. FRL 13, 15 pp.
——(1977*b*) Ibid. 1975. Tech. Rept. FRL 12, 32 pp.
National Radiological Protection Board and the Commissariat à l'Energie Atomique (1979) *Methodology for evaluating the radiological consequences of radioactive effluents released in normal operations*. Joint report for Commission of the European Communities. CEC, Luxembourg. Doc. V/3865/79-EN, 292 pp.
Pentreath, R. J., Lovett, M. B., Harvey, B. R., and Ibbett, R. D. (1979) In *Biological Implications of Radionuclides Released from Nuclear Industries* Vol. II IAEA. SM-237/1, 227–45.
Tamplin, A. R., and Cochran, T. B. (1974) *Radiation Standards for Hot Particles*. Washington DC: Natural Resources Defense Council.