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Uptake of americium-241 from two experimentally labelled deep-sea sediments by three benthic species: a bivalve mollusc, a polychaete and an isopod

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ABSTRACT: Laboratory experiments were conducted to determine the bioavailability of ^{241}Am in contaminated deep-sea sediments to 3 benthic organisms: the bivalve mollusc *Venerupis decussata*, the polychaete *Hermione hystrix* and the isopod *Cirolana borealis*. The biological availability was determined in 2 different sediments: the 'Atlantic sediment', originating from the NEA dumpsite in the NE Atlantic and the 'Pacific sediment', originating from a seabed disposal feasibility investigation site in the Pacific Ocean. The affinity of ^{241}Am for the sediments was very high: K_d values amounted to $1.5 \times 10^5 \pm 3.0 \times 10^4$ for the Atlantic and $1.8 \times 10^5 \pm 2.0 \times 10^4$ for the Pacific sediment. Accumulation of the radionuclide by the 3 species, measured over a period of 40 to 50 d, yielded transfer factors (TF = radioactivity g^{-1} animal wet weight/radioactivity g^{-1} wet sediment) lower than unity. Uptake of ^{241}Am was highest in the polychaete (TF Pacific = 0.12; TF Atlantic = 0.05), whereas radionuclide accumulation in the isopod (TF Pacific = 0.032; TF Atlantic = 0.006) was comparable with uptake by the bivalve (TF Pacific = 0.02; TF Atlantic = 0.004). Most of the radionuclide in the clam (56 to 75 %) was fixed to the shell; 78 % to 96 % of the ^{241}Am was fixed to the body wall of the polychaete. The relative distribution of the radionuclide among the animals' tissues did not vary with the sediment type, whereas transfer factors in shell and viscera of clams and body wall of worms were dependent upon sediment type. In all cases transfer factors were up to 2 to 5 times higher for animals in Pacific sediment, although K_d's in both sediments were not statistically different. ^{241}Am K_d values in these sediments were found to be insensitive indicators of relative bioavailability of the radionuclide; however, the different geochemical associations of ^{241}Am in both sediments (in particular a highly resistant fraction not removed by hot acid leaching) can be used to explain the observed differences in bioavailability.

INTRODUCTION

Most of the transuranium elements now present in the environment result from nuclear weapon tests. Also, the continuing growth of the nuclear industry will lead to an unavoidable increase in the production of high-level radioactive waste. One option under study to alleviate this problem is permanent disposal of such wastes in deep-sea sediments (Anderson, 1979; Gomez et al., 1980). This consideration has emphasized the need to study the potential for transfer of long-lived α-emitting transuranium elements to sediment dwelling benthic biota. Among the radionuclides currently of concern, the quantitatively more important are ^{241}Am and $^{239+240}$Pu, with minor amounts of neptunium and curium isotopes as well as the β-emitter ^{241}Pu which decays *in situ* to the α-emitter ^{241}Am (Grimwood and Webb, 1976).

Recently, studies have begun to address the bioavailability of ^{241}Am from sea water and food to different marine organisms. Various aspects of the behaviour of transuranics in marine organisms have been examined both in laboratory experiments (Fowler and Heyraud, 1974; Murray et al., 1978; Guary, 1980; Hoppenheit et al., 1980; Grillo et al., 1981; Guary and Fowler, 1981) and field surveys (Bowen et al., 1976; Hetherington et al., 1976; Livingston and Bowen, 1976; Pentreath and Lovett, 1976). We report here results on

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the transfer of Am-241 from artificially contaminated deep-sea sediments to 3 different types of invertebrates which inhabit the sediment-water interface: the clam *Venerupis decussata* (Linneaus, 1758) (= *Tapes decussatus*), the isopod *Cirolana borealis* (Lilljeborg, 1851) and the polychaete worm *Hermione hystrix* (Savigny, 1820). The clam and worm are typical infaunal organisms, while the isopod can burrow in sediment or may swim above it. The 2 sediments used in these experiments came from the OECD/NEA North-East Atlantic low-level dumpsite and from a location in the North Pacific which is currently under study for use as a sub-seabed high-level radioactive waste disposal site.

MATERIALS AND METHODS

Different sediments from 2 locations were used in these experiments. The Atlantic sediment was sampled from a depth of 4640 m at the OECD/NEA dumpsite at 46°02' N, 16°55' W. The Pacific sediment was obtained from a depth of 5700 m at 31°N 159°W. Details of the general sedimentological properties of the 2 sediments are shown in Table 1.

For the bioaccumulation experiments 750 g wet of Atlantic and Pacific sediment were each slurried into 100 ml of sea water spiked with ^{241}Am (125 Bq g^{-1} wet weight in Atlantic and 100 Bq g^{-1} wet Pacific sediment), and left to equilibrate for 1 d.

To determine pore water concentrations of ^{241}Am and to calculate K_d values, defined as radioactivity g^{-1} dry sediment/radioactivity g^{-1} pore water (Duursma and Bosch, 1970), 100 g wet of each sediment was labelled with 10-fold the ^{241}Am activity used in the bioaccumulation experiments and left to equilibrate for 1 d. These more active sediments were centrifuged at 2700 g for 20 min and the water filtered through 0.45 µm membrane filters to separate pore water and sediment grains. The 2 fractions were subsequently radioanalysed, and data from these experiments allowed computation of pore water ^{241}Am levels in the sediments used to label the animals.

Spiked sediments for the K_d and bioaccumulation experiments were poured into plastic basins and allowed to settle. The settled sediments were then placed under running unfiltered sea water which ranged in temperature from about 12 to 14 °C during the experiments.

Clams *(Venerupis decussata)* were purchased from a local dealer who collects the animals near Sete (France). The isopods *(Cirolana borealis)* were caught in baited traps moored off the coast of Monaco at a depth of 250 m. In the same location polychaete worms *(Hermione hystrix)* were captured using bottom trawls. All animals were individually numbered and acclimated in running sea water for 1 wk prior to the experiment. Three isopods were placed in each of 5 perforated plastic tubes. In the case of the larger polychaetes only a single individual was introduced into each tube. All tubes were then partially buried in the spiked sediments. This technique ensured that the same individual(s) living within the sediment could be identified each time they were removed for radioanalysis. Individually numbered clams were placed directly in the sediment. During the course of the experiments, animals were periodically removed from the sediments and counted. The clams were first

Table 1. Sampling locations and general sedimentological properties of the deep-sea sediments

Sediment	Location	Water depth (m)	% Carbonate	% Organic C	% Grainsize distribution (µm)		Clay mineralogy*
North East Atlantic	46° 02′ N 16° 55′ W	4640	83	0.3	>63	3.1	Illite
					31–63	3.2	Kaolinite
					16–31	2.1	Chlorite
					8–16	4.2	
					4–8	16.8	
					2–4	5.3	
					<2	65	
Central Pacific	31° 00′ N 159° 00′ W	5700	7.6	0.4	>63	8.7	Illite
					31–63	0.3	Kaolinite
					16–31	0.6	Chlorite
					8–16	4.5	
					4–8	9.4	
					2–4	18	
					<2	59	

* Dominant clays in descending order of abundance

washed with a fine jet of sea water from a wash bottle, followed with further cleaning by carefully scrubbing with filter paper. Afterwards, clams were placed for 20 min in clean flowing sea water and then counted for ^{241}Am in a NaI(Tl) well detector connected to a pulse height analyser. The ^{241}Am was measured using its 59.5 Kev photopeak. A preliminary experiment was designed to test if there was a difference in ^{241}Am loss from animals held in flowing sea water for 20 min or for periods up to 3 h. No difference in loss was observed whether the clams were rinsed for 20 min or 3 h, therefore it was assumed that 20 min was sufficient to remove adherent contaminated sediment grains.

In a separate experiment, clams exposed to contaminated sediment and rinsed for 20 min, were then counted for ^{241}Am content and subsequently fed with phytoplankton for 2 h. Following feeding, the animals were recounted and the particulate matter at the bottom of the beakers was collected by filtration and counted to determine the ^{241}Am content of the excreted faeces. This purging experiment was carried out to estimate the fraction of ingested sediment that was not excreted during the 20 min rinsing period.

Before radioanalysing the isopods, the animals were allowed to swim freely in clean sea water for about 20 min. If necessary, they were further cleaned with filter paper. Groups of 2 or 3 animals were then radioanalysed together in one counting tube filled with 10 ml of clean sea water. As with clams, worms were also washed with a fine jet of clean sea water and then placed in running sea water for 20 min. Occasionally sediment aggregations were trapped in the mucous layer on the body surface. In this case, animals were individually checked for adhered sediment grains and the sediment was removed with a pair of tweezers. All worms were then individually radioanalysed in a tube filled with 15 ml clean sea water.

None of the animals received extra food over and above the natural particulate matter in the sediments and that entering with the running sea water. At the termination of the experiments, the clams and worms were dissected to determine ^{241}Am distribution in their tissues.

RESULTS

^{241}Am radioactivity in Atlantic and Pacific sediment

The ^{241}Am radioactivity in the Atlantic and Pacific sediments used in the uptake experiments amounted to 125 Bq g^{-1} wet and 100 Bq g^{-1} wet, respectively. From 2 test experiments in which a higher ^{241}Am concentration was used, the mean K_d was calculated to be $1.5 \times 10^5 \pm 3.0 \times 10^4$ (1σ propagated counting error) in the Atlantic sediment and $1.8 \times 10^5 \pm 2.0 \times 10^4$ in the Pacific sediment. These very high K_d values illustrate the strong association of this transuranium nuclide with the sediment particles. Furthermore, within experimental errors, K_d values as well as total radioactivity in the sediment did not change during the course of the experiments. Considering the errors associated with the K_d values, there was no significant difference in K_d between the 2 sediments.

Uptake from Atlantic and Pacific sediments

Increase of the transfer factor with time for *Venerupis decussata* and *Hermione hystrix* are illustrated in Fig. 1. The transfer factors (TF) were calculated as ^{241}Am g^{-1} animal wet weight divided by ^{241}Am g^{-1}

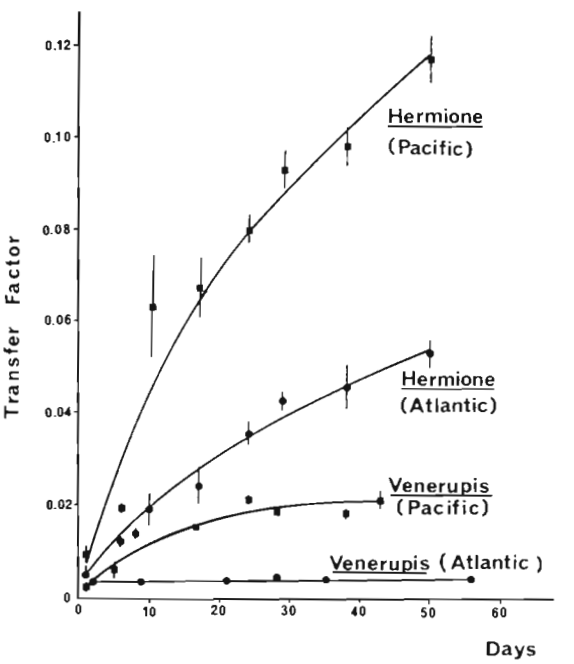

Fig. 1. *Hermione hystrix* and *Venerupis decussata*. Americium-241 transfer factors as a function of time for individuals exposed to contaminated Atlantic and Pacific sediments. Error bars represent ± standard error on the mean for 4 individuals. Curves fitted by eye

total wet sediment. Transfer factors for *V. decussata* in Atlantic sediment reached steady state after 2 d of uptake. The transfer factor from the Atlantic sediment averaged $4 \times 10^{-3} \pm 0.2 \times 10^{-3}$ (mean for 4 animals in 6 successive measurements ± S.E. of the mean). Uptake from the Pacific sediment resulted in a mean transfer factor of $20 \times 10^{-3} \pm 0.7 \times 10^{-3}$ (mean of 4 animals for at least 4 measurements). The accumulation curve clearly indicated that maximum values for ^{241}Am transfer from sediment to clam were achieved much more slowly in the Pacific sediment than in the

Atlantic sediment. At the termination of the experiment uptake was about 5 times higher from the Pacific sediment than from the Atlantic sediment.

In *Hermione hystrix*, ^{241}Am accumulation from both sediments had not reached a steady state after 50 d (Fig. 1). The transfer factor increased more rapidly with the Pacific sediment. At the end of the experiment the maximum transfer factor was $53 \times 10^{-3} \pm 3 \times 10^{-3}$ (mean value for 4 animals ± S.E. of the mean) in the Atlantic sediment and $117 \times 10^{-3} \pm 5 \times 10^{-3}$ in the Pacific sediment. Furthermore, the difference between the 2 sediment types may increase with time since uptake from the Pacific sediment appeared to be more rapid than that from Atlantic sediment. If we assume that uptake continued at similar rates measured over the first 50 d, near equilibrium transfer factors of approximately 0.1 and 1.8 can be estimated for worms in Atlantic and Pacific sediments, respectively.

Fig. 2 illustrates the change in ^{241}Am transfer factors in benthic isopods with time. As in the case of the

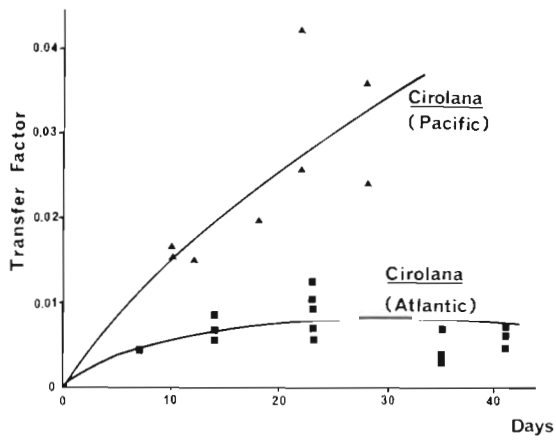

Fig. 2. *Cirolana borealis*. Americium-241 transfer factors as a function of time for individuals exposed to contaminated Atlantic and Pacific sediments. Each point represents a pooled measurement for 2 or 3 individuals. Curves fitted by eye

worms, the isopods did not reach equilibrium by the end of the experiment in Pacific sediment, and the scatter in the data makes it difficult to estimate the equilibrium TF value. As ^{241}Am accumulation by the isopods was very low it was not possible to monitor the ^{241}Am radioactivity in a single individual within the time-span that it could remain out of the experimental solution. Therefore, samples were composed of all the remaining animals from each tube and each point in Fig. 2 represents the pooled measurement of 2 to 3 individuals. Even then, the propagated counting error at 1σ was greater than 50 % in some samples. If the total net counts were not higher than twice the counting error, the radioactivity was regarded as 'not measurable' and therefore not included in Fig. 2. The relative counting error (1σ) of each measurement in the Atlantic experiment was within 20 to 40 % of the total net counts. No significant increase in the transfer factor was noted in the Atlantic sediment group after 7 d; therefore an average transfer factor for these isopods was estimated to be $6 \pm 1 \times 10^{-3}$. In the Pacific sediment experiment, the mean transfer factor was calculated for the 6 individuals (2 groups each containing 3 ind.) which survived longer than 20 d. These TF values computed between 22 and 28 d averaged $32 \pm 4 \times 10^{-3}$. Thus, the mean transfer factor from Pacific sediment was roughly 5 times higher than that from Atlantic sediment.

^{241}Am uptake relative to pore water radioactivity

Assuming that bioaccumulation of ^{241}Am results mainly from uptake from pore water radioactivity, a concentration factor (CF) can be estimated as ^{241}Am

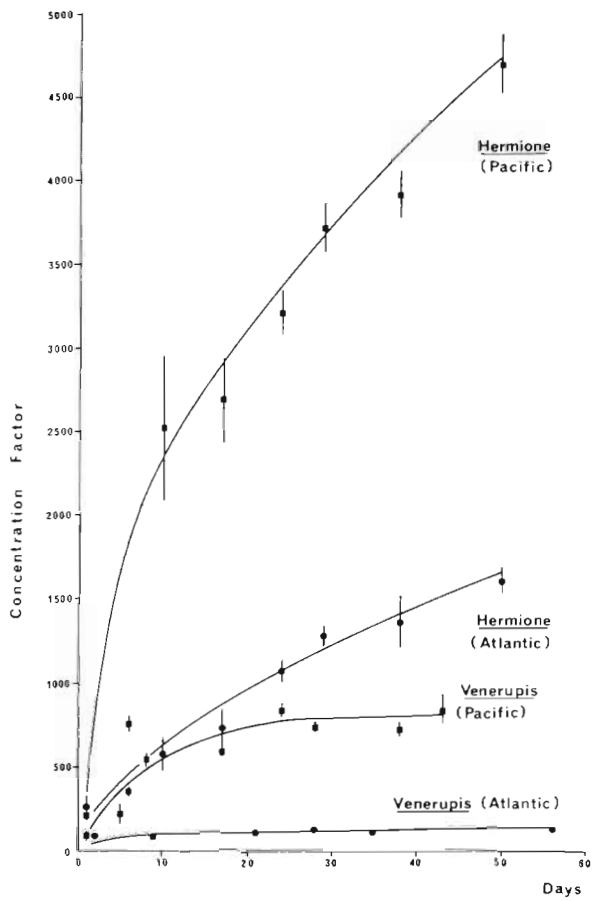

Fig. 3. *Hermione hystrix* and *Venerupis decussata*. Americium-241 concentration factors based on pore water as a function of time for contaminated Atlantic and Pacific sediments. Error bars represent ± standard error on the mean for 4 individuals. Curves fitted by eye

radioactivity g^{-1} animal/^{241}Am radioactivity ml^{-1} pore water. Pore water ^{241}Am activities were derived from the previously determined K_d values. The computed concentration factors are illustrated for the clam and the polychaete worm in Fig. 3. Whereas CF values for *Venerupis decussata* in the Atlantic sediment experiment were constant after 2 d of uptake $\bar{x} = 106 \pm 5$ ($n = 24$ measurements), CF in the Pacific sediment experiment increased, reaching a maximum value at steady state of 780 ± 30 ($n = 16$ measurements).

Pore water CF values in *Hermione hystrix* were significantly higher than in clams; in the Atlantic sediment a CF of 1600 ± 80 was reached after 50 d. In the Pacific sediment experiment uptake was more rapid resulting in an average CF of 4690 ± 180 after the same period of time. It was evident from both uptake curves that ^{241}Am accumulation had not reached equilibrium after 50 d. Likewise, isopods (not shown) attained much lower pore water CF values in Atlantic sediment (180 ± 16 after 1 to 6 wk) than in the Pacific sediment (1290 ± 170 after 3 to 4 wk).

^{241}Am organ distribution

Clam

In an attempt to discern the fraction of accumulated ^{241}Am represented by residual contaminated sediment in the digestive tract of clams, radioactivity in 4 individuals taken from the spiked Atlantic sediment was measured before and after a 2 h feeding period on non-radioactive phytoplankton. In 3 of 4 individuals, total body radioactivity decreased between 13 to 24 % after feeding suggesting excretion of some residual sediment (Table 2). Virtually all the ^{241}Am lost from the clams was recovered with the faeces on filter paper. In 1 individual loss did not occur and furthermore, no faeces were observed although the animal had extruded both siphons and appeared to be in good health.

Following 56 d and 43 d of ^{241}Am uptake by the clams from Atlantic and Pacific sediment, respectively, all remaining individuals were dissected in order to examine distribution of ^{241}Am in the tissues (Table 3). The relative distribution of radionuclide was roughly comparable in the individuals from both sediment types. The preponderence of ^{241}Am was found in the shell (56 to 70 % in Atlantic sediment; 60 to 75 % in Pacific sediment), whereas most of the remaining radionuclide (20 to 30 %) was located in the viscera. Only small amounts of ^{241}Am were measured in pallial fluid, muscles (adductor-muscles and foot-muscle), mantle plus siphons and gill. The relative ^{241}Am bioavailability from Atlantic and Pacific sediment is also reflected in the radionuclide TF values in some of the tissues. For example, the shells and viscera of individuals in the Pacific sediment had transfer factors

Table 2. *Venerupis decussata*. Loss of radioactivity from individuals after 2 h feeding on unlabelled phytoplankton. The 4 clams used in this experiment had accumulated ^{241}Am for 28 d from Atlantic sediment. Data are counts per minute ± 1σ counting error and % of prefeeding radioactivity

Clam no.	cpm ± 1σ before feeding	cpm ± 1σ after feeding	cpm ± 1σ lost by feeding (% loss)	cpm ± 1σ recovered on filter (% recovered)
1	4016 ± 183	4214 ± 183	−198 ± 91 (0 %)	not measurable
2	4918 ± 176	3751 ± 173	1167 ± 93 (24 %)	806 ± 164 (16 %)
3	8422 ± 195	7175 ± 191	1307 ± 125 (15 %)	1530 ± 176 (18 %)
4	5175 ± 177	4489 ± 175	686 ± 98 (13 %)	1045 ± 164 (20 %)

Table 3. *Venerupis decussata*. Transfer factors (TF) and tissue distribution (% of total animal radioactivity) of ^{241}Am after 56 and 43 d exposure to ^{241}Am in contaminated Atlantic and Pacific sediments, respectively. Values are means ± standard error for 9 clams radioanalyzed in 3 groups each with 3 individuals

Organ	Atlantic sediment		Pacific sediment	
	TF ($\times 10^{-3}$)	%	TF ($\times 10^{-3}$)	%
Shell	5.1 ± 0.3	61 ± 4	19 ± 1	66 ± 5
Pallial fluid	0.8 ± 0.1	4 ± 1	3.5 ± 1.5	3 ± 1
Muscle (foot + adductor)	1.1 ± 1.1	1 ± 1	3 ± 2	0.5 ± 0.3
Mantle + siphons	4.3 ± 1.4	5 ± 1	11 ± 4	5 ± 2
Viscera	27 ± 2	25 ± 2	80 ± 14	25 ± 4
Gill	8 ± 2	4 ± 1	2.5 ± 2.5	0.3 ± 0.3

about 3 times higher than values for individuals exposed to Atlantic sediment. However, in all other organs, transfer factors were not significantly different at the 2σ level for the 2 sediment types.

Polychaete

The ^{241}Am tissue distribution and transfer factor values in *Hermione hystrix* after a 50 d exposure in both sediment types is depicted in Table 4. As with clams, no major difference in the percentage tissue distribution could be found between the 2 sediment types. Furthermore, the enhanced uptake in Pacific sediment compared to Atlantic sediment (Fig. 1) is also evident from the roughly similar relative difference between transfer factors in the body wall of the worms.

Table 4. *Hermione hystrix*. Transfer factors (TF) and tissue distribution (% of total animal radioactivity) of ^{241}Am after 50 d exposure to ^{241}Am in contaminated Atlantic and Pacific sediment. Mean value ± standard error given for 4 individuals

Organ	Atlantic sediment		Pacific sediment	
	TF ($\times 10^{-3}$)	%	TF ($\times 10^{-3}$)	%
Body wall	54 ± 15	86 ± 5	183 ± 55	95 ± 1
Gut	2 ± 1	5 ± 2	1 ± 1	0.4 ± 0.4
Coelomic fluid	5 ± 1	9 ± 2	10 ± 4	4.8 ± 0.4

DISCUSSION

Uptake of ^{241}Am by the animals

Bioaccumulation of ^{241}Am from the contaminated deep-sea sediments is low as evidenced by transfer factors lower than unity for all animals tested in these experiments (Figs. 1 and 2). Low bioavailability of ^{241}Am from a variety of sediments seems to be the case for several marine organisms (Table 5). Miramand et al. (1982) observed higher ^{241}Am uptake from coastal sediments by the amphipod *Corophium volutator* (TF = 0.1) than by either the bivalve mollusc *Scrobicularia plana* (TF = 0.01) or the worm *Arenicola marina* (TF = 0.003). Our data indicate a much higher ^{241}Am accumulation by *Hermione hystrix* (TF Atlantic = 0.05; TF Pacific = 0.12) compared to *A. marina*, whereas uptake by *Cirolana borealis* (TF Atlantic = 0.006; TF Pacific = 0.032) is lower than *C. volutator* exposed to ^{241}Am in coastal sediments. *Venerupis decussata* attained transfer factors (TF Atlantic = 0.004; TF Pacific = 0.02) that were similar to those reported for *S. plana*.

Miramand et al. (1982) reported that all animals tested in their experiments ingested contaminated sediment, but all were allowed to void their gut for 16 h under running sea water before measurement. While some faeces were excreted from clams during our 20 min rinsing period, we observed that about 13 to 24 % of the total radioactivity of the clam could be excreted during a 2 h period of intense feeding (Table 2). This suggests that some residual contaminated sediment remained in the gut after the 20 min rinse. However, the excretion of the ^{241}Am during the 2 h feeding cannot be unequivocally attributed to removal of sediment grains since recent autoradiographic studies of clam viscera (Miramand, 1983) have shown that ^{241}Am taken up from water is heavily concentrated on surfaces of the gut wall and in the tubules of the digestive gland. Hence, food in the form of phytoplankton cells moving along the gut lumen and through the digestive gland tubules during digestive processes could feasibly remove ^{241}Am located in those tissues. Regardless of the origin of the residual ^{241}Am that can be purged through active feeding, our transfer factors reported for clams (Fig. 1), if considered in terms of transfer into tissue, may be as much as 20 % high. Nevertheless, the use of these factors is not an unrealistic approach in the ecological sense since animals with full guts are regularly eaten by organisms from higher trophic levels.

Some individual isopods exhibited a dark staining of the alimentary tract at the start of the experiment suggesting that food was present prior to exposure. During the course of the experiments the dark area in the gut disappeared indicating that digestion or regurgitation of 'food' particles (as was often observed) had occurred. However, a restaining of the alimentary tract was never observed during the experimental period. This indicated that these animals did not take up any sedimentary particles during the experiments. Moreover, isopods swam freely in the continuously renewed sea water in the upper portion of the partially buried plastic tubes and only occasionally burrowed into the sediment. These observations may explain the low TF for ^{241}Am in these animals. In addition a low transfer factor was observed for the gut of *Hermione hystrix* (Table 3) which also suggests that relatively little residual sediment was retained by the animals at time of dissection.

Sediment type-induced differences in bioaccumulation of ^{241}Am

The present results indicate that the sediment type induces appreciable differences in total ^{241}Am uptake by the animals. A different metal (silver, cobalt and zinc) availability from various sediments has been

Table 5. Americium-241 transfer factors for uptake from sediment by benthic organisms expressed as the ratio: radioactivity g^{-1} wet weight animal/radioactivity g^{-1} wet weight sediment. The mode of contamination of the sediment is indicated

Animal	^{241}Am transfer factor	Sediment origin	Mode of contamination	Reference
Amphipoda				
Corophium volutator	0.12	Cotentin coast (France)	Labelled	Miramand et al. (1982)
Isopoda				
Cirolana borealis	0.007	Atlantic deep-sea	Labelled	This study
	0.032	Pacific deep-sea	Labelled	This study
Polychaeta				
Nereis diversicolor	0.0003	Bikini Atoll	*In Situ* weapon test	Beasley & Fowler (1976)
Nereis diversicolor	0.0006	Windscale	*In Situ* reprocessing effluent	Beasley & Fowler (1976)
Arenicola marina	0.003	Cotentin coast (France)	Labelled	Miramand et al. (1982)
Hermione hystrix	0.053	Atlantic deep-sea	Labelled	This study
	0.117	Pacific deep-sea	Labelled	This study
Mollusca				
Scrobicularia plana	0.009	Cotentin coast (France)	Labelled	Miramand et al. (1982)
Venerupis decussata	0.004	Atlantic deep-sea	Labelled	This study
	0.02	Pacific deep-sea	Labelled	This study

observed by Luoma and Jenne (1975). Their results show that the highest metal availability to clam *(Macoma balthica)* soft parts was achieved in sediment types with the lowest K_d values, i.e. for those sediments from which most metal was desorbed back into solution. In our experiments, however, the K_d values for ^{241}Am are very high (1.5 to 1.8 $\times$ 10^5), and taking into account the observed propagated counting error ($1\sigma = 2–3 \times 10^4$), it is evident that the difference between K_d for these 2 sediments is not significant at the 1σ level.

tion. We conclude that the 5-fold difference in the resistant fraction of sediment-bound ^{241}Am provides a ready explanation for the up to 5-fold difference in transfer factors between the 2 sediments. This underscores the fact that biological availability of radionuclides is, at least for americium, highly dependent on site-specific geochemical associations. Beasley and Fowler (1976) attributed the observed difference in ^{241}Am availability to *Nereis diversicolor* (Table 5) to different ^{241}Am origins in the sediments used. In the Bravo crater sediment the americium principally

Table 6. Geochemical partitioning of ^{241}Am in Atlantic and Pacific sediments. (After Aston et al., in prep.)

Partition phase	Extraction technique	Atlantic sediment (%)	Pacific sediment (%)
Exchangeable	1 M NH_4CH_3COO	≤ 1	≤ 1
Carbonates	1 M CH_3COOH	18	12
Fe/Mn hydrated oxides	0.1 M NH_2OH HCl + 25 % v/v CH_3COOH	≤ 1	42
Organic matter	40 vol H_2O_2 + 1 M CH_3COOH	18	33
Resistant	hot conc. HNO_3 + HF + H_3ClO_4	62	12

Sequential leaching experiments performed on these 2 sediments labelled with ^{241}Am have been used to determine the geochemical partitioning of this radionuclide (Aston et al., in prep.). The leaching techniques used in this study and the differences in partitioning of ^{241}Am in the Atlantic and Pacific sediments are given in Table 6. The results reveal that for the Atlantic sediment 62 % of the ^{241}Am was present in a highly resistant form which required hot concentrated acid attack for removal, while for the Pacific sediment only 12 % was present in this resistant fraction. We conclude that the 5-fold difference in the resistant fraction of sediment-bound ^{241}Am provides a ready explanation for the up to 5-fold difference in transfer factors between the 2 sediments. This underscores the fact that biological availability of radionuclides is, at least for americium, highly dependent on site-specific geochemical associations. Beasley and Fowler (1976) attributed the observed difference in ^{241}Am availability to *Nereis diversicolor* (Table 5) to different ^{241}Am origins in the sediments used. In the Bravo crater sediment the americium principally comes from the decay of ^{241}Pu, while the ^{241}Am in the Windscale sediment comes partially from ingrowth of ^{241}Pu but mainly from ^{241}Am already present in the processed waste. The observed differences in bioavailability in our experiments cannot be attributed to a different ^{241}Am origin, as all sediments used in these experiments have been identically labelled in the laboratory.

It is commonly assumed that the bioavailability of ^{241}Am from sediment can be controlled by two processes: (1) indirect transfer of the radionuclide from the

interstitial water to the animal, i.e. a simple water/animal transfer, and (2) direct transfer from the sediment to the animals. The role of the indirect transfer route (water/animal) was estimated to be high for Pu uptake in worms. Murray and Renfro (1976) followed plutonium uptake from spiked Mediterranean sediment and reported that the worm *Nereis diversicolor* obtained more than 98% of its body radioactivity directly from the pore water. Beasley and Fowler (1976) calculated that roughly 83% of the plutonium taken up by the same species of worm exposed to environmentally contaminated Bikini Atoll and Windscale sediment would be derived from the pore water. Miramand et al. (1982) compared americium CF values from pore water uptake, as calculated in a sediment-uptake experiment, with the CF values from sea water uptake as obtained from a parallel sea water-uptake experiment. They found that in the 3 benthic species tested *(Arenicola marina, Corophium volutator, Scrobicularia plana)*, the pore water CF values were higher than the sea water CF values for all animals except for the shell of the clam. From these results they concluded that the uptake could not be explained exclusively by a simple interstitial water/organism exchange but that part of the ^{241}Am in the animals was directly transferred from the sediments to the animals.

To determine the relative importance of indirect transfer from interstitial water to animals in the present experiments, we calculated the CF values for ^{241}Am relative to the interstitial water (Fig. 3) and compared these data with published ^{241}Am CFs for the same species determined by uptake from sea water. Grillo et al. (1981) measured a CF of about 350 in the clam *Venerupis decussata* following 22 d of ^{241}Am uptake from sea water. Within this period no equilibrium was reached and the CF was still increasing by the end of the experiment. In our experiments (Fig. 3), the same species reached a pore water CF of 840 for animals in Pacific sediment and 170 for those in Atlantic sediment. In the latter sediment an apparent equilibrium was reached after 2 d and in the former there was an indication of approach to steady state after the second week. It is evident that the total amount taken up, as well as the uptake rate in our experiments, are somewhat different from the data obtained by Grillo et al. (1981) and are sediment-type dependent; hence, it is difficult to draw any valid conclusions from a comparison of the data. Nevertheless, in the case of the Pacific sediment, the nearly 2-fold enhanced pore water CF compared to CF values based on uptake from water over a similar period of time (Fig. 3; 22 d) suggests that indirect transfer of ^{241}Am from interstitial water only can account for about 50% of the total radionuclide accumulated by the clams. The same transfer process is apparently not operating with clams exposed to Atlantic sediment and, perhaps due to the particular chemical nature of the sediment, uptake may better reflect slow leaching of ^{241}Am from ingested sediment grains than indirect transfer from pore water. Clearly, the differences in geochemistry of these 2 sediment types are of overriding importance in determining the transfer processes involved.

For *Hermione hystrix* the pore water CF value of 1060 (after 24 d) in Atlantic sediment is roughly comparable with the sea water CF (CF = 1000) measured after 22 d by Grillo et al. (1981). The pore water CF in the Pacific (pore water CF = 3210) is, however, substantially higher. From these data we estimate that about 30% of all ^{241}Am taken up by the worm in the Pacific sediment could have come from the pore water whereas in the Atlantic sediment almost all ^{241}Am in the worm can be accounted for by direct uptake from pore water. A comparison of these results for worms with those for clams also underscores the fact that the ^{241}Am transfer pathway is highly dependent upon animal species.

From the above discussion it may be concluded that simple interstitial water/animal transfer cannot explain all the results from our experiments. It seems that direct transfer from the sediment to the animal can play an important role, and this is also indicated by the fact that ^{241}Am uptake from both sediments cannot readily be related to the measured K_d, but depends upon the americium geochemical fractionation in the sediments.

^{241}Am organ distribution in the clam

Although accumulation of ^{241}Am by the clam from the sediment was followed for a considerably long time in these experiments (56 d for Atlantic and 43 d Pacific sediments), only relatively low concentrations of the radionuclide were detected in the soft tissues. By far the largest proportion of ^{241}Am in soft tissues (between 20 and 30%) was measured in the viscera (Table 3), but the feeding experiment (Table 2) indicated an excretion of about 13 to 24%, so it may be assumed that the bulk of the ^{241}Am concentration measured in viscera is actually a readily excretable ^{241}Am fraction such as ingested sediment or that loosely bound to external tissues in the digestive tract.

Small percentages of total body ^{241}Am have been found in gill, foot and adductor muscle and in the pallial fluid (Table 3). Similarly small, but measurable fractions have been measured in these tissues when ^{241}Am is taken up from water by clams (Grillo et al., 1981). The relative tissue distribution of ^{241}Am in the clam is comparable for both sediment types. However, in terms of relative bioavailability to tissues (Table 3),

more ^{241}Am was taken up by the shell and the viscera of animals living in the Pacific sediment.

^{241}Am organ distribution in the worm

The ^{241}Am tissue distribution in *Hermione hystrix* showed a high affinity for the external surface (Table 4). Also, Grillo et al. (1981) measuring uptake from water in the same animal, reported a very high ^{241}Am concentration factor for body wall (CF = 1900–2200) and setae (CF = 8000–13 000). These authors found about 11 to 30 % total body radioactivity on the mucus-covered setae alone. In the present experiments the setae were not measured separately and they are included in the 'body wall' (Table 4). It was unexpected to find that the coelomic fluid contained relatively high percentages of total body activity (4 to 13 % in Atlantic sediment and 4 to 6 % in Pacific sediment) when compared to tissues in worms exposed to ^{241}Am in sea water (Grillo et al. (1981): 2 % of total ^{241}Am in coelomic fluid). The reason for this difference in relative tissue distribution is not clear but is probably pathway induced.

Our experimental results show that benthic species generally take up ^{241}Am from artificially labelled deep-sea sediments to a very low extent. This finding appears to be a common occurrence for a variety of ^{241}Am-labelled sediment types (Table 5). Maximum uptake as well as the uptake rate varies with the species; however, even within the same species, sediment type can induce large differences. A geochemical characterization of the sediments by sequential leaching techniques is an important prerequisite for predicting the bioavailability of sediment-bound ^{241}Am at specific sites in the deep-sea.

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