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Uptake of Americium and Plutonium from Contaminated Sediments by Three Benthic Species: *Arenicola marina, Corophium volutator* and *Scrobicularia plana*

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ABSTRACT: The biological availability of americium (^{241}Am) and plutonium (^{238}Pu) contained in a muddy sediment was investigated in 3 burrowing coastal benthic species: the polychaete annelid *Arenicola marina* (L.), the bivalve mollusc *Scrobicularia plana* (da Costa) and the amphipod crustacean *Corophium volutator* (Pallas). The biological availability was expressed in terms of a transfer factor defined as the activity in the organisms (cpm g^{-1} wet weight) relative to the activity in the sediment (cpm g^{-1} wet weight). For the 3 species and both radionuclides, the transfer factors observed were low (< 1) after 14 d in contact with the contaminated sediment. Uptake by *C. volutator* (TF = 0.1) was 10 times (TF ≃ 0.01) and 50 times (TF ≃ 0.002) greater than uptake by *S. plana* and *A. marina* respectively. A comparative investigation of the biological availability of americium and plutonium contained in seawater showed, for the same exposure period, that the concentration in the organism relative to the concentration in the water (C.F.) was about 1000 for americium and 780 for plutonium in *C. volutator*, but much lower for the other 2 species. The data obtained demonstrate that part of the americium transferred to the organisms comes from the sediment's interstitial water. Part of the americium retained by the animals could therefore result from direct transfer from sedimentary particles to organisms.

INTRODUCTION

Numerous studies on the biogeochemistry of transuranic elements in marine environments indicate that ultimately most of the plutonium and americium deposited in the ocean from atmospheric fallout or wastes from nuclear industries is transferred to the sediments (Noshkin and Bowen, 1973; Hetherington et al., 1975; 1976; Livingston and Bowen, 1976a, b; Ballestra, 1980; and references therein). However, it appears that the transuranic elements contained in the solid wastes that might be emplaced within deep-sea sediments, would also be concentrated in the sediments over a long period of time (Sousselier, 1977; Talbert, 1977). Recent investigations show that, once they are adsorbed, these transuranic elements are not irreversibly retained on the sedimentary particles but obey the fundamental distribution law. Thus they can be released on contact with interstitial and surface waters which contain lower concentrations than required by the mass action constant for the reaction (Noshkin and Wong, 1980; Pentreath et al., 1980). The problem is to understand the biological availability of these radionuclides for benthic species living in contact with the sediment. To our knowledge, all experimental studies on the uptake of plutonium and americium from sediments by benthic species have used only the polychaete worm *Nereis diversicolor* (Beasley and Fowler, 1976; Murray and Renfro, 1976). Thus studies should be developed to include other species representative of benthic indofauna and epifauna.

This paper deals with the results of experiments on the uptake of plutonium and americium from contaminated sediments by 3 coastal benthic species: the polychaete annelid *Arenicola marina* (L.), the bivalve mollusc *Scrobicularia plana* (da Costa), and the amphipod crustacean *Corophium volutator* (Pallas). These species live in sediments, or at the sediment-sea water interface, and belong to zoological groups which occur in deep water (Hessler and Sanders, 1967; Hessler and Jumars, 1977; Rice, 1978; Sibuet, 1979).

A first experiment (Experiment A) quantifies the

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transfer of these radionuclides from labelled sediment to organisms (T.F.), while a second experiment (Experiment B) studies the uptake from labelled seawater by the same organisms (C.F.).

MATERIALS AND METHODS

Corophium volutator were trapped on the shore near the port of Saint Vaast-la-Hougue (eastern coast of Cotentin). *Scrobicularia plana* and *Arenicola marina* were collected from mud near the port of Ouistreham (Calvados).

The ^{241}Am (half-life: 434 y), ^{239}Pu (half-life: 24,386 y), and ^{238}Pu (half-life: 86 y), conditioned in HNO_3 1 N were obtained from the C.E.A. (France). Before use, ^{239}Pu and ^{238}Pu, were put into a (VI) oxidation state according to Duursma and Parsi's method (1974). ^{241}Am is assumed to be in a (III) state.

The ^{241}Am gamma ray (60 keV) was measured with 25 % efficiency with a gamma spectrometer coupled to a NaI(Tl) well crystal.

^{239}Pu and ^{238}Pu alpha emissions were counted by liquid scintillation spectrometry using Picofluor 15 as the scintillator. Before counting, the samples were pretreated as follows:

(1) After collection the individuals were rinsed, weighed, dissected *(S. plana)* and then oven-dried for 48 h (T = 95 °C). The tissues were mineralised using hot HNO_3 directly in the counting vials. After mineralisation, the HNO_3 was removed by evaporation, the dry residue redissolved in 3 ml of HCl 0.1 N, and 15 ml of scintillator was added.

(2) The radioactivity of 2 ml of sea water from the experimental aquarium was directly counted in 15 ml of scintillator.

(3) 5 g of dried sediment were calcined for 24 h at 600 °C, then shaken for 24 h, in a 50:50 mixture of HCl – HNO_3 (200 ml 80 °C). The acid extraction was repeated. Numerous tests, we have realized, have demonstrated that about 96 % of the plutonium in the sediments can be desorbed in this way. After centrifugation, aliquots of 50 and 100 µl of supernatants were directly placed in 15 ml of scintillator and counted.

An external standard ratio calibration curve was plotted; the count yield was in all cases greater than 95 %.

Accumulation from Sediment (Experiment A)

Muddy sediment was collected at the same time as the *Corophium volutator* Table I shows how the sediment was prepared (sifting) and labelled with ^{241}Am and ^{238}Pu. The sediment was labelled in such a way that the radionuclide concentration in sea water was always less than 10^{-8} M in order to avoid any polymerization of these isotopes.

The animals were introduced into the sediment: 100 individuals weighing on average 0.015 g wet weight for the *Corophium volutator*, 15 individuals of around 3 g wet weight for the *Arenicola marina* and 8 individuals averaging 9 g wet weight for the *Scrobicularia plana*. The 14 d experiment was carried out under clean running sea water. After sampling, the individuals were placed in clean running sea water for 16 h to void their gut before preparation for analysis (vide supra), and were then carefully rinsed. Measuring the radioactivity in the organism as well as in the sediment makes it possible to calculate the transfer factor (T.F.): cpm g^{-1} animal wet weight/cpm g^{-1} sediment wet weight.

After the biological uptake experiments the sediment was analysed to determine the binding strength of ^{241}Am to the sediment. Aliquot portions were extracted with acetic acid at pH2, and then with a 50:50 mixture of concentrate HCl · HNO_3 at 80 °C. Interstitial water was extracted from two 600 g samples of sediment using a press similar to that described by Presley et al. (1967). The concentration of ^{241}Am in the interstitial water was then measured. As both the concentrations of radioactivity in the sediment *(AS)* and the interstitial water *(A isw)* are known, the distribution coefficient of ^{241}Am between the sediment and its interstitial water *(Kd = AS/A isw)* can be calculated.

Table 1. Scheme of contamination processes of sediments used in our experiments (dry to wet ratio = 0.41)

Sediment sifting 6.25 mm / sifting 0.25 mm results in 12.8 kg wet weight, of which 92.2 % has a Ø < 50 µm and 250 µm > 7.8 % > 50 µm	
3 × 21 bottles (2.1 kg bottle^{-1}) + 1 l filtered sea water (0.45 µm) bottle^{-1} + ^{241}Am 259.10^6 mBq bottle^{-1} (8.50.10^{-9} M) (7 µCi bottle^{-1})	3 × 21 bottles (2.1 kg bottle^{-1}) + 1 l filtered sea water (0.45 µm) bottle^{-1} + ^{238}Pu 259.10^6 mBq bottle^{-1} (7 µCi bottle^{-1}) (1.7 10^{-9} M)
Shaking for 48 h (T = 4 °C)	Shaking for 48 h
Results in contaminated sediment at 123.10^6 mBq kg^{-1} (3.3 µCi kg^{-1})	Identical process as for ^{241}Am
Placed into basin at the rate 10 cm basin^{-1} of wet sediment	
Decanting	
Placed under running sea water 100 ml min^{-1} T = 14° ± 2 °C	

Uptake from Sea Water (Experiment B)

For each isotope, 100 *Corophium volutator*,, 30 *Arenicola marina*, and 30 *Scrobicularia plana*, all weighing the same as those used previously, were placed individually in plastic aquaria containing 20 ml (*C. volutator* and 200 ml *A. marina* and *S. plana*) of sea water filtered on a millipore filter (0.45μm), at 14 °C. subsequently, 6 475 mBq ml^{-1} (175 pCi ml^{-1}) for ^{241}Am, and 1 776 mBq ml^{-1}(48 pCi ml^{-1}) for ^{239}Pu were added. The sea water was changed every 48 h using water of the same quality containing the same concentrations of radioisotopes. Each time the water was changed 10 individuals (*C. volutator*) and 4 individuals (*A. marina* and *S. plana*) were collected as well as aliquots from the new and old solutions for analysis. Counting their radioactivity made it possible to calculate the concentration factor defined as cpm g^{-1} animal wet weight/ cpm ml^{-1} sea water.

Table 2. Percentage of ^{241}Am desorbed by 2 successive acid attacks on the contaminated sediment and percentage, before acid extraction, of ^{241}Am retained on the contaminated sediment fractions smaller or larger than 50 μm

Extract (%)		Activity (%)	
Acetic acid pH2	HCl-HNO_3 80 °C	Particles of Ø > 50 μm	Particles of Ø < 50 μm
66	31	8.6	91.4

RESULTS

For the 2 extractions, the ^{241}Am activity of the contaminated sediment-interstitial water was around 7.4 mBq ml^{-1} (0.2 pCi ml^{-1}). The sediment-interstitial water Kd obtained from this value was 5.10^4. It is interesting to note as well that 91 % of the element was attached to sedimentary particles of less than 50 μm in diameter and that 66 % of the ^{241}Am could easily be desorbed from the sediment by acetic acid at pH2 (Table 2).

The transfer factors of ^{241}Am to the *Corophium volutator*, *Scrobicularia plana*, and *Arenicola marina* obtained at the end of Experiment A were low, less than 1 (Table 3). This was also true for plutonium whose transfer factors were the same as for americium

Table 3. Transfer factor of ^{241}Am and ^{238}Pu from contaminated sediment to *Corophium volutator*, *Arenicola marina* and *Scrobicularia plana*. T = 14 °C. Radionuclide concentrations in sediment (cpm g^{-1} wet weight): ^{238}Pu = 8828; ^{241}Am = 2276 / dry to wet ratio: sediment = 0.41; *Corophium volutator* = 0.19; *Arenicola marina* = 0.14; *Scrobicularia plana*, entire = 0.41, soft part = 0.07, shell = 0.86

Species	Radioisotope	Duration of experiments (d)	Number of individuals	cpm g^{-1} wet weight	Transfer factor
Corophium volutator					
	^{241}Am	4	60	228	0.10
	^{241}Am	14	14	273	0.12
	^{238}Pu	4	14	883	0.10
	^{238}Pu	14	21	883	0.10
Arenicola marina					
	^{241}Am	6	8	4.55	0.002
	^{241}Am	14	4	6.83	0.003
	^{238}Pu	6	8	17.66	0.002
	^{238}Pu	14	4	17.66	0.002
Scrobicularia plana					
Entire	^{241}Am	7	4	15.93	0.007
	^{241}Am	14	4	20.48	0.009
	^{238}Pu	7	4	88.28	0.01
	^{238}Pu	14	4	88.28	0.01
Soft part	^{241}Am	7	4	13.66	0.006
	^{241}Am	14	4	22.76	0.01
	^{238}Pu	7	4	79.45	0.009
	^{238}Pu	14	4	44.14	0.005
Shell	^{241}Am	7	4	15.93	0.007
	^{241}Am	14	4	22.76	0.01
	^{238}Pu	7	4	88.28	0.01
	^{238}Pu	14	4	88.28	0.01
Pallial fluid	^{241}Am	7	4	11.38	0.005
	^{241}Am	14	4	4.55	0.002
	^{238}Pu	7	4	17.66	0.002
	^{238}Pu	14	4	26.48	0.003

(Table 3). However, differences in uptake of the 2 transuranic elements by the 3 benthic species tested can be observed: *C. volutator* (T.F. = 0.1) accumulated about 10 times more of the transuranic elements from the contaminated sediment than did *S. plana* (T.F.≃0.01) and 50 times more than *A. marina* (T.F.≃0.002). These differences were also found for the concentration factors from labelled sea water in Experiment B (Fig. 1, 2, 3). For the same time period in the labelled water, viz. 12 d, *C. volutator* accumulated 10 times more americium (C.F.≃1000) and 14 times more plutonium (C.F.≃780) than *S. plana* (whole) and 78 times more americium and 180 times more plutonium than *A. marina*.

For uptake from water (Experiment B), the 3 tested species preferentially accumulated americium rather than plutonium (Fig. 1, 2, 3). *Scrobicularia plana* shells accumulate a greater concentration of the 2 transuranic elements than do the soft part or the pallial fluid (Fig. 2); the soft part seems to be in isotopic equilibrium with the elements in the water from the 14th day for ^{239}Pu and the 20th day for ^{241}Am (Fig. 2). However, for the other *S. plana* tissues, as well as for *Corophium volutator*, and *Arenicola marina*, the plutonium and americium accumulation remains linear at the end of the different periods of experimentation (Fig. 1, 2, 3).

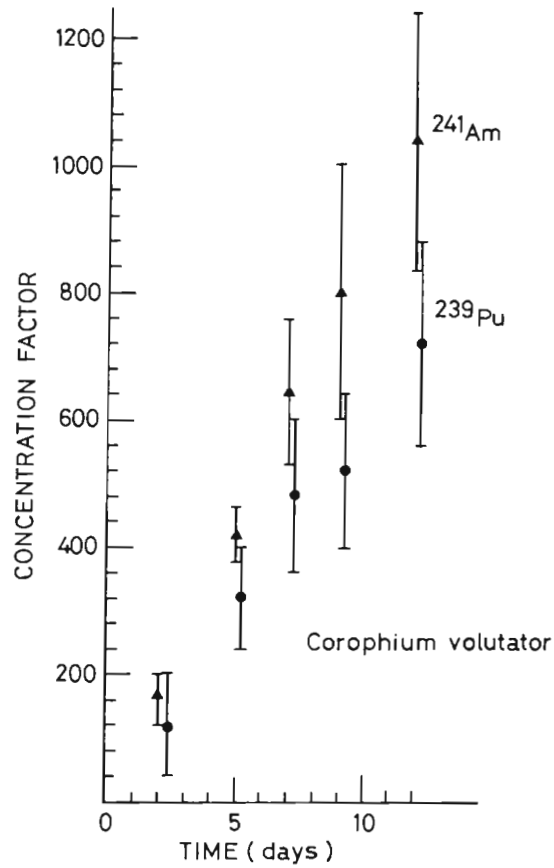

Fig. 1. *Corophium volutator*. Accumulation of ^{241}Am and ^{239}Pu from labelled sea water. T = 14 °C (n = 10 per group, mean wet weight of 100 individuals ≃ 0.015 ± 0.005 g)

Table 4 shows americium and plutonium distributions in various parts of *Scrobicularia plana*, after accumulation of these elements from labelled sea water (Experiment B) or contaminated sediment (Experiment A). In both cases, the greatest percentage of americium and plutonium is found in the shells.

DISCUSSION

After 2 weeks of contact with the highly contaminated sediments, the radioactivity found in the 3 tested species, although measurable, is low. The animals voided their gut for 16 h under running sea water. However, in spite of this procedure, several residual grains of sediment could remain in the individuals, and because of their high activities, would falsify the measurement. In fact, the problem does not occur. For *Corophium volutator* the residual sediment weight should correspond to around 40 % of the sample, and around 1.5 % for *Scrobicularia plana* and *Arenicola marina*, i.e. from 30 to 140 mg wet weight of sediment; however, when the animals are mineralized for plutonium measurement, once the organic matter is destroyed no trace of sedimentary particles is found in the samples. Our experiments demonstrate that americium and plutonium transfer from contaminated sediments does occur in *C. volutator*, *A. marina*, and *S. plana*. This transfer is low and varies with the species (from 0.1 for *C. volutator* to 0.002 for *A. marina*).

Our results for *Arenicola marina* confirm the transfers observed by Beasley and Fowler (1976) and by Murray and Renfro (1976), between *Nereis diversicolor* and various contaminated sediments (T.F. between 0.0015 (Pu) and 0.0003 (Am)). Our data also show that other burrowing benthic species such as *Scrobicularia plana* and *Corophium volutator* can accumulate plutonium and americium from contaminated sediment from 5 to 50 times more, respectively, than the annelids.

It would seem that the transuranic elements retained on the sediment are only slightly accumulated by the benthic species, as already observed for certain heavy metals and fission products (Renfro, 1973: Ueda et al., 1977; 1978 Jennings and Fowler, 1980 and references therein).

The transfer of radionuclides from sediment to burrowing benthic species can take place in 2 ways: (1) direct uptake by desorption of the transuranic elements retained in the ingested sediment when passing through the digestive tract; (2) indirect uptake via interstitial water, i.e., simple water-organism

Table 4. *Scrobicularia plana*. Tissue distribution of americium and plutonium following exposure to labelled sea water or contaminated sediment. T = 14 °C (± = standard deviation for 4 individuals)

Tissue	% Total Americium content		% Total Plutonium content		% Total Wet wt
	After 32 d exposure to labelled sea water	After 14 d exposure to contaminated sediment	After 42 d exposure to labelled sea water	After 14 d exposure to contaminated sediment	
Shell	87 ± 2	58 ± 5	89 ± 4	66 ± 18	35 ± 2
Soft part	12 ± 3	36 ± 4	9 ± 3	30 ± 12	34 ± 3
Pallial fluid	1 ± 1	5 ± 2	1 ± 1	4 ± 1	34 ± 2

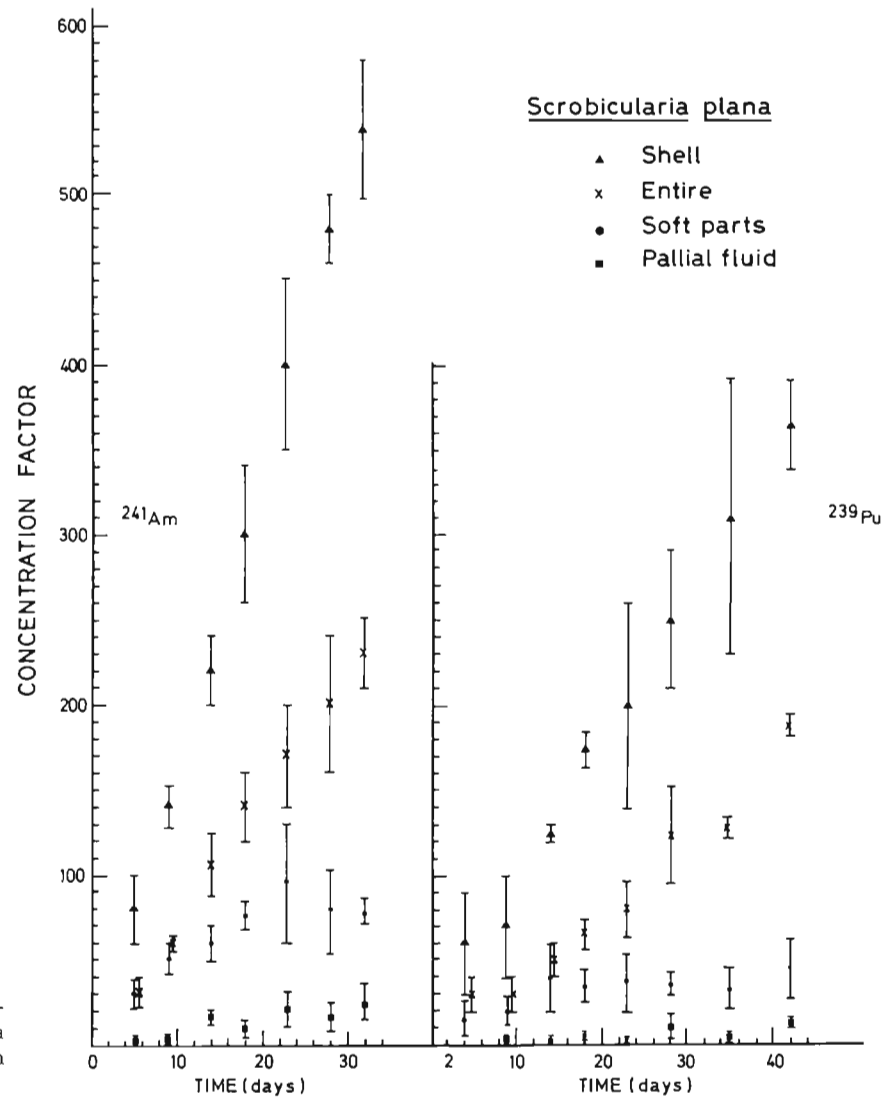

Fig. 2. *Scrobicularia plana*. Accumulation of ^{241}Am and ^{239}Pu from labelled sea water. T = 14 °C (n = 4 per group, mean wet weight of 60 individuals = 9 ± 2 g)

exchange. In our experiments with americium, we found low quantities of this element in the interstitial water of the sediment (≈0.2 pCi ml^{-1}). This allows the determination in Experiment A (transfer sediment-species) of the C.F. reached by the animals from the sediment's interstitial water, i.e. *CF isw*. The ratio CF_{isw}/CF^{*}_{sw} calculated for the same experimental

* *CF sw*: Concentration factor found in Experiment B (uptake from labelled sea water) and reported in Figs. 1, 2 and 3

Table 5. Comparison of *CF sw* for americium found in Experiment B (uptake from labelled sea water) and *CF isw* calculated from the value of interstitial sea water in Experiment A (transfer from labelled sediment)

Species		*CF* from interstitial water *(CF isw)*	*CF* from labelled sea water *(CF sw)*	*CF isw/CF sw*
Corophium volutator		2700	1200	2.3
Arenicola marina		68	16	4.3
Scrobicularia plana	Soft part	137	60	2.3
	Shell	228	220	1.0

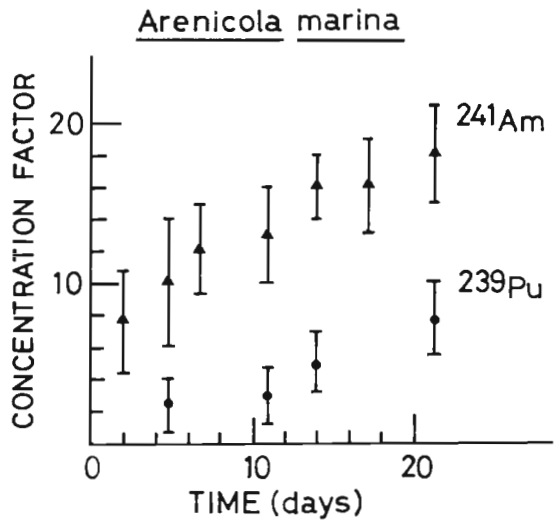

Fig. 3. *Arenicola marina*. Accumulation of ^{241}Am and ^{239}Pu from labelled sea water. T = 14 °C (n = 4 per group, mean wet weight of 44 individuals = 3 ± 1 g)

period (14 d) is greater than 1 (Table 5). Thus it would seem that the transfer of ^{241}Am (T.F.) observed in Experiment A cannot be exclusively explained by a simple interstitial water/organism exchange. Consequently, part of the americium taken up by the animals may be directly transferred from the sediment, probably by desorption of the americium in the ingested sediment.

All animals tested continuously ingested contaminated sediment. *Corophium volutator* can only ingest sedimentary particles with diameters between 4 and 63 µm (Fenchel et al., 1975), and 92 % of the experimental sedimentary particles had a diameter of less than 50 µm and fixed more than about 91 % of the radioactivity. Throughout the test, *Arenicola marina* burrowed into the sediment, and *Scrobicularia plana* inhaled surface sediment through the siphon. In any event, in spite of the continuous ingestion of highly contaminated sediments, the observed uptake was very low. The extractions of americium from the sediment by pH2 acetic acid, which indicate a 66 % desorbtion of this element, do not seem to prove the bio-availability of this fraction.

Our experimental results show that benthic species can mobilize the transuranic elements within the biodisturbed layer. They demonstrate that the transfer of ^{241}Am sorbed on sediment to benthic species could occur through two different processes: (1) indirect transfer via interstitial water and (2) direct transfer from sediment to species. The transfer factors we measured in a short period seem to be low, but one cannot predict the effect of this pathway on the contamination of sea products reaching man. Such an effect could only be evaluated by means of specific transfer models including the data obtained in the present work. In both cases, within the limit of extrapolation of the experimental results obtained with the 3 coastal species belonging to zoological groups present in deep water, small amphipods would seem to represent a more significant dissemination vector for transuranic elements in the marine environment than bivalves or polychaetes.

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