CHEMICAL PARTIONING OF PLUTONIUM AND AMERICIUM IN SEDIMENTS FROM THE PALOMARES MARINE ECOSYSTEM

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The marine environment adjacent to the coastal village of Palomares (Southeastern Spain) became a suitable area to investigate the behavior of the transuranics that reached the Mediterranean Sea after the partial land-to-sea transfer of the contamination dispersed as a consequence of the non-nuclear explosion of two thermonuclear bombs accidentally released during a plane crash in 1966 (GASCÓ *et al.*, 1992 and ROMERO *et al.*, 1992). To determine the potential post-depositional remineralization of these transuranics, their bioavailability to bottom feeding biota, along with the effect of their source term on their distribution within the major sedimentary phases, the geochemical association of these longlived radionuclides has been evaluated.

In this study, two sections from Station 13 (50 m depth, 37°11.21 N 1°47.1 W) were selected: PASD13(01) corresponds to the first centimeter of the core; PASD13(09) is a deeper layer, and it corresponds to the 8-9 cm section. Station 13 is located south of the Almanzora river mouth in an area of the continental shelf where enhanced concentrations of transuranics have been previously found (GASCÓ *et al.*, 1992).

Chemical partioning of Pu and Am was performed by applying the following sequential leaching procedure: 12 g subsamples were stirred for 18 h with the appropriate amount of extractant, as shown in Table 1. The supernatant was filtered through a Whatman GF/C filter paper. Spikes of ^{242}Pu and ^{243} Am were added to determine the radiochemical yield of the procedure (COOK *et al.*, 1984).

Fraction	Reagant	Volume(ml/g)
Readily available	$CaCl_2$ 0.05 M	20
Exchangeable	CH_3-COOH 0.05 M	20
Organically bound	$Na_4P_2O_7$ 0.1 M	100
Oxide bound	$(NH_4CO_2)_2$0.175 M/$C_2O_4H_2$ 0.1 M	75
Residual	HNO_3/HF/HCl conc.	75

Table 1. Scheme of the leaching procedure for marine sediments

The results of chemical partitioning for Station 13 are summarized in Tables 2 and 3. The order of association of Pu in PASD13(01) is (Table 2): organic > oxide > residual > exchangeable > readily available. The fractions considered most mobile (readily available/exchangeable) contain less than 3% of the plutonium. The majority is associated with insoluble organic chelated complexes (66%).

The order of association of Am in PASD13(01) is (Table 2): exchangeable > organic > residual > oxide > readily available. Almost 50% of the Am is linked to the exchangeable phase, known as a "soluble" phase. Am also appears to be less associated with the sesquioxides (Al,Fe,Mn) than plutonium.

The isotopic ratios ^{238}Pu/^{239}Pu=0.04±0.01 and Am/Pu=0.3±0.1 indicate global fallout as the source term of these transuranics.

Fraction	^{239}Pu activ. Bq/kg d.w.	% ^{239}Pu content	^{241}Am activ. Bq/kg d.w.	% ^{241}Am content
Read. avail.	BDL	-----	BDL	-----
Exchangeable	0.06 ± 0.01	2.5 ± 0.4	0.24 ± 0.02	40. ± 6.1
Organ. bound	1.53 ± 0.10	66. ± 2.3	0.2 ± 0.01	33.3 ± 3.7
Oxide bound	0.51 ± 0.04	22. ± 1.7	0.05 ± 0.02	8.3 ± 4.1
Residual	0.22 ± 0.05	9.4 ± 2.0	0.10 ± 0.04	16.6 ± 7.9
Σ activity	2.3 ± 0.12		0.6 ± 0.05	

Table 2. Sequential leaching of PASD13(01). Uncertainties are given in ± 1σ.

Fraction	^{239}Pu activ. Bq/kg d.w.	% ^{239}Pu content	^{241}Am activ. Bq/kg d.w.	% ^{241}Am content
Read. avail.	0.01 ± 0.005	0.09 ± 0.05	BDL	-----
Exchangeable	0.15 ± 0.02	1.36 ± 0.21	0.51 ± 0.06	17.3 ± 0.04
Organ. bound	0.91 ± 0.06	8.23 ± 0.85	0.46 ± 0.10	15.6 ± 0.03
Oxide bound	0.80 ± 0.05	7.23 ± 0.71	0.05 ± 0.03	1.7 ± 0.003
Residual	9.19 ± 0.92	83.1 ± 1.52	1.93 ± 0.18	65.4 ± 0.14
Σ activity	11.06 ± 0.92		2.95 ± 0.22	

Table 3. Sequential leaching of PASD13(09). Uncertainties are given in ± 1σ.

The order of association for Pu is (Table 3): residual > organic > oxide > exchangeable > readily available. Most of the Pu appears in the residual fraction (88%), suggesting that the Pu is very refractory, like Pu in the aerosol dispersed during the accident in 1966. The order of association for Am is (Table 3): residual > exchangeable > organic > oxide > readily available. Most of the Am also appears in the residual fraction, however, almost 20% is in the exchangeable phase.

The isotopic ratios ^{238}Pu/^{239}Pu=0.02±0.005 and 241Am/239Pu=0.24±0.03 suggest weapon grade Pu ratios, indicating that the transuranics detected at this depth originate from the Palomares accident.

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