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Flux of Transuranium Nuclides in the Northwestern Mediterranean following the Chernobyl Accident

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Introduction

Biogeochemical flux studies have furnished data on the vertical flux of natural and artificial radionuclides through the water column in the North Pacific and North Atlantic, however, until recently similar information for the Mediterranean has been lacking (Fowler et al., 1987; in press). As part of the French DYFAMED Programme, sediment traps were moored in the Ligurian Sea at 200 m depth approximately 15 nautical miles off the coast of Corsica. The total depth of the water column at this station was 2200 m. The automated sediment traps were set to collect six consecutive samples at intervals of every 6.25 days during April-May 1986. On 26 April 1986, the accident at Chernobyl occurred and subsequent sampling of air, sea water, plankton and sedimenting particles allowed assessing the behaviour and transport of Chernobyl-derived radionuclides in the northwestern Mediterranean Sea (Fowler et al., 1987; Whitehead et al., 1988; Holm et al., 1988). Here we report the concentrations of $^{239+240}Pu$ and ^{241}Am in various samples collected before and after the Chernobyl fallout was detected in this region.

Results and Discussion

The Chernobyl fallout was first detected in Monaco on 30 April and subsequent wet and dry fallout analyses indicated that peak Chernobyl fallout delivery to the sea surface near Monaco occurred during 4-5 May (Whitehead et al., 1988). The total integrated deposition of $^{239+240}Pu$ at Monaco following the accident was 10±1 mBq m^{-2}. The $^{241}Am/^{239+240}Pu$ activity ratio was 0.13±0.03 corrected to 26 April. This ratio is lower than the integrated ratio today from nuclear test fallout (0.36) but will increase rapidly due to the decay of ^{241}Pu since the $^{241}Pu/^{239+240}Pu$ ratio was unusually high (≈86) in the Chernobyl fallout. The deposition of $^{239+240}Pu$ was only about 0.02% of the previous integrated deposition from nuclear test fallout, which means that post-Chernobyl samples contain activity from both source terms. The two sediment trap samples collected before 26 April are considered to contain only background levels of $^{239+240}Pu$ and ^{241}Am arising from nuclear testing fallout (Table 1). Thus, an average ^{241}Am background concentration in sedimenting particles of 0.78 Bq kg^{-1} dry can be computed. Plutonium concentrations in the first two samples varied to a greater degree and an average concentration of 3.7 Bq kg^{-1} may be representative. Fission product analyses indicated that the maximum concentrations of Chernobyl-derived radionuclides were found in particles at 200 m during 8-15 May (Fowler et al., 1987). Comparison of the average pre-Chernobyl levels with the transuranic concentrations measured in particles collected between 8-15 May indicate an increase in $^{239+240}Pu$ and ^{241}Am by a factor of 2.6 and 4.7, respectively (Table 1). Transuranic concentrations decreased thereafter similar to those of the fission products (Fowler et al., 1987) indicating the pulsed nature of the vertical flux of Chernobyl-derived radionuclides associated with sinking particles.

Microscopic examination of the sediment trap samples showed that a large proportion of the particulates was zooplankton fecal material. Zooplankton netted over the traps and particularly their freshly excreted fecal pellets also contained relatively high concentrations of transuranics (Table 1). In the case of plutonium, concentrations were quite similar in fresh fecal pellets and in the particulates from 200 m. If we assume that most of the particles in the traps were fecal pellets (Fowler et al., 1987), the increased Am/Pu ratio in the sinking particles from 8-15 May compared to that in pellets produced over the traps on 6 May suggests that sinking fecal pellets scavenged ^{241}Am to a greater extent than $^{239+240}Pu$ as they sank through the water column. A similar observation has been made for these transuranics in north Pacific waters (Fowler et al., 1983).

Table 1. Concentrations, activity ratios and vertical fluxes of transuranics in the northwestern Mediterranean before and after the Chernobyl accident. Concentrations in zooplankton and their fecal pellets are also given for comparison.

Sample/Date	Concentration (Bq kg^{-1} dry) $^{239+240}Pu$	Concentration (Bq kg^{-1} dry) ^{241}Am*	Ratio Am/Pu	Mass flux (mg $m^{-2}d^{-1}$)	Flux (mBq $m^{-2}d^{-1}$) $^{239+240}Pu$	Flux (mBq $m^{-2}d^{-1}$) ^{241}Am
Sediment trap (200 m)						
13-20 April	5.43	0.87	0.16	213.7	1.16	0.19
20-26 April	2.00	0.68	0.34	111.5	0.22	0.076
26 April-2 May	3.00	1.51	0.50	63.9	0.19	0.096
2-8 May	3.22	1.05	0.33	65.5	0.21	0.069
8-15 May	9.70	3.63	0.37	53.6	0.52	0.19
15-21 May	4.71	2.83	0.60	57.6	0.27	0.16
Zooplankton (0-100 m) 6 May	0.016	0.004	0.25			
Fecal pellets (0-100 m) 6 May	7.4	0.63	0.09			

*^{241}Am values corrected for ingrowth from ^{241}Pu

From Table 1, the integrated vertical flux through 21 May of post-Chernobyl $^{239+240}Pu$ associated with sinking particles is calculated to be approximately 7.5 mBq m^{-2}. Comparison of this value with the total integrated wet and dry deposition at Monaco cited above suggests that 75% of the plutonium deposited in this region had fluxed through 200 m depth within one month following the accident. To our knowledge there are no other comparable transuranic data from sediment trap studies which were underway in European waters following the accident. Nevertheless, our results have demonstrated that small, but significant inputs of $^{239+240}Pu$ and ^{241}Am resulting from the Chernobyl accident were measureable in the northwestern Mediterranean. Our data also suggest that both radionuclides were rapidly scavenged from the surface layers and transported to depth by sinking biogenic debris.

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