*Biological control of transuranic flux through the upper water column of the northeast Pacific Ocean

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Abstract

Analyses of sediment trap samples and freshly-produced biogenic detritus have demonstrated the overall importance of the rarer, large biogenic particulates in rapidly removing Pu and Am from the euphotic zone and transporting these radionuclides to depth. Am/Pu ratios in the particulate samples indicated that at depth Am was scavenged to a greater extent than Pu. The vertical flux of transuranics was significantly greater in a highly productive area of the California current than in oligotrophic waters either off Mexico or in the north Pacific gyre.

Résumé

Les analyses des échantillons des pièges à sédiments ainsi que des produits d'origine zooplanctonique fraîchement excrétés, ont démontré l'importance générale des particules larges et rares d'origine biogénique dans l'élimination rapide du Pu et Am de la zone euphotique vers le fond. Les rapports d'activité Am/Pu dans les échantillons de particules indiquent qu'en profondeur l'Am est entraînée par les particules de façon plus importante que le Pu. Le flux vertical des transuraniens est significativement plus important dans une zone de grande productivité influencée par le courant Californien, que dans les eaux oligotrophiques soit au large du Mexique, soit au nord des Iles Hawaii.

Sinking particulate material, both inorganic and biogenic, is probably the primary means by which surface-introduced transuranium nuclides are transported to the sediments in the marine environment. Measurements of filtered particles have verified the association of plutonium and americium with suspended particulate matter in the upper

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mixed layers of the ocean. Furthermore, recent studies have shown that large, rapidly sinking biogenic particulates such as zooplankton fecal pellets contain relatively high concentrations of plutonium and americium. Despite the growing amount of indirect evidence supporting the theory of rapid vertical transport of transuranics by particulate matter, direct measurements of transuranic flux via sinking particles are few.

Through the VERTEX programme, concentrations of plutonium (Pu) and americium (Am) were measured in sea water, suspended particulates, sediment trap samples and biogenic material from different sites in the northeast Pacific. Details on methods of collection, preparation and analyses of samples are reported elsewhere (Fowler et al., 1983).

At the VERTEX I station in the California current off Monterey, water measurements showed the presence of subsurface maxima of $^{239+240}Pu$ and ^{241}Am between 100 and 750 m and 250 and 750 m, respectively. A large fraction of the filterable $^{239+240}Pu$ (32%) and ^{241}Am (21%) in surface waters was associated with phytoplankton. Am/Pu ratios in surface water and filtered particles indicated that Pu was accumulated by cells to a greater degree than Am; however, measurements at depth showed an enrichment of Am over Pu on fine suspended particles. Both transuranic concentrations in trapped particles (largely of biogenic origin) and transuranic flux tended to increase with depth to 750 m suggesting active scavenging of Pu and Am in the upper water column by the sinking particulates. Am/Pu ratios in the trapped particles indicated that at depth Am was scavenged by particles to a greater extent than Pu. Interestingly, the trend for increasing transuranic concentration in particles and to a lesser extent, transuranic flux with depth generally was correlated with a marked, nearly linear increase in flux of alumino-silicate particles from the surface to 100 m. Reactive transuranic elements are known to be strongly sorbed by clay particles, and these are readily filtered by zooplankton and incorporated into fecal pellets. Thus, sinking biogenic aggregates such as these with their incorporated transuranic load could feasibly scavenge additional clay particles during their descent (resulting in increased alumino-silicate and transuranic flux) and rapidly reach great depth. Flux-derived mean residence times for Pu and Am of 2-13 yr and 1.5-4.5 yr respectively were computed for the upper mixed layers (<250 m) at the VERTEX I site. These values are of the same order as previous estimates based on delivery rate and mixed-layer transuranic inventory data from the Mediterranean.

During the VERTEX II and III cruises in oligotrophic waters off central Mexico, a subsurface Pu maximum was apparent near 450 m. No similar subsurface maximum was noted for Am; in fact Am concentrations were fairly constant throughout the upper 1500 m. The very high Am concentration in surface waters during VERTEX III resulted in an Am/Pu ratio (4.6) significantly higher than any of those found at other depths or during VERTEX I. Very little association of transuranics with fine suspended particles was observed ($<1\%$ in surface, $\sim 3\%$ at

450 m). This may have been due to the order of magnitude lower concentrations of suspended matter (0.27 mg l^{-1}) in surface waters off Mexico compared to that measured off California ($\sim$3.5 mg l^{-1}).

Data from the analyses of biological samples again showed a substantial enrichment of transuranics, particularly Am, in zooplankton fecal pellets compared to the organisms that form them. A layer of nearly pure diatoms (Coscinodiscus walesii) was present at $\sim$100 m during VERTEX III. These algal cells showed a marked preference for accumulation of Pu (dry concentration factor $\sim 5 \times 10^4$) over Am (CF_{dry} $\sim 5 \times 10^3$) when their transuranic concentrations were compared to those in the surrounding water. On the other hand, copepods displayed a fairly similar degree of accumulation for the two radionuclides ($\sim 2\text{-}3 \times 10^3$).

During VERTEX II, two different size fractions of copepods were collected to assess the effect of relative surface area on transuranic accumulation. Within the limits of error, transuranic concentrations were roughly the same in the two size fractions suggesting that the relative difference in animal size (i.e. surface area) was not sufficient to substantially alter the amounts of radionuclide taken up.

Sediment trap results from both cruises again indicate the general tendency for increasing transuranic concentration in the large, rapidly sinking particles with depth, at least to a depth of 450/470 m. Below this depth, the Am concentration decreased dramatically. Likewise transuranic flux, particularly that for Am, was at maximum at 450 m and decreased in a regular manner below this depth to 1470 m. There is evidence at 450 m that Am was scavenged to a greater degree than Pu. In contrast to the VERTEX I site, the alumino-silicate flux in waters off Mexico was very low and constant with depth; thus, the changes in transuranic concentration and flux at this site do not appear to be related to alumino-silicate levels in the water. In general, the vertical fluxes of Pu and Am were correspondingly lower in these waters due to the overall lower biomass present when compared to the VERTEX I site in the California current. For example, computed mean residence times for Pu and Am in the upper 100 m off Mexico were approximately 91 and 42 yr, respectively.

The same trends were observed at the VERTEX IV site in oligotrophic waters of the central gyre north of Hawaii. For example Pu and Am concentrations in large particles generally increased at least to a depth of 1000 m. Increasing Am/Pu ratios in trapped particles from 140 m to 1500 m indicated that Am was scavenged to a greater extent than Pu. Corresponding fluxes of transuranics also increased to 1000 m and then decreased by nearly a factor of two at 1500 m. These fluxes (4-20 fCi $m^{-2}d^{-1}$ for $^{239+240}Pu$ and 1-13 fCi m^{-2} d^{-1} for ^{241}Am) were of the same order as those measured off Mexico but were lower than the fluxes observed in the more productive waters of the California current.

These in situ sediment trap measurements along with analyses of zooplankton fecal pellets from different sites have clearly demonstrated the importance of the rarer, large biogenic particles in rapidly removing transuranics from the euphotic zone and transporting them to depth. Nevertheless, the exact role these particles play in controlling the observed transuranic levels in the surrounding sea water (e.g. transuranic maxima) is still unclear. Furthermore, it is not clear from these experiments the extent of depth to which these biogenic vectors operate. Measurements of Am/Pu ratios in large particles and suspended particulates at greater depths as well as in underlying surface sediments would shed some light on this question.

References

FOWLER, S.W., S. BALLESTRA, J. LA ROSA and R. FUKAI. Vertical transport of particulate-associated plutonium and americium in the upper water column of the Northeast Pacific. Deep-Sea Res. 30: 1221-1233 (1983).

Discussion

P. MIRAMAND: Avez-vous étudié les populations planctoniques, existantes le long de la colonne d'eau et en particulier les phénomènes éventuels de migration planctoniques? Existe-t'il une corrélation entre ceux-ci et les profils du plutonium et d'américium observés?

S. FOWLER: No. This information is being compiled by a student as part of his thesis. When the information becomes available, I would like to examine any possible relationship.

F. CARVALHO: Have you identified the particulate matter found in the PITS?

S. FOWLER: Yes, most of this work has been done by the Santa Cruz group. They find a preponderance of fecal pellets at all depths as well as coccoliths, diatoms, marine snow, ciliates, etc.

G. LAPICQUE: Is it possible to install an automatic sampling device at a fixed position so as to give reliable information over a year and thus reduce temporal variations?

S. FOWLER: Yes, in fact this is one of the future aims of the VERTEX programme. In the Mediterranean we are now using a sequential sampling sediment trap with which I hope to obtain data of that nature for the French ECOMARGE programme.

P. GERMAIN: With respect to the Pu peak at about 500 m, do you know if there is a large concentration of suspensivores at that depth?

S. FOWLER: There is quite a population of copepodites and other organisms at this depth. In particular, enhanced microbiological chemolithotrophic activity in this region has resulted in an increased flux of organic matter in the aphotic zone. However, it is still not clear what, if any, relationship these organisms have with the maintenance of the subsurface transuranic maxima.

A. MANGINI: How does one explain the differences in residence times for ^{241}Am and ^{239}Pu in the Pacific? These residence times were derived for transuranics on trapped particles. How large is their share in the flux in comparison to the total particulate radionuclides?

S. FOWLER: The differences are a direct function of the elements' reactivity. For example Am is scavenged more readily at depth than Pu; therefore the Am residence time is shorter. It is now well-established that the rarer, fast sinking large particles are responsible for the bulk of the downward mass flux. Therefore, the large particles are extremely important in controlling the residence, or removal time of the radionuclides in the upper layers of the sea.