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The Isotopic Signature of Fallout Plutonium in the North Pacific

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ABSTRACT

Plutonium analyses of a dated coral record from the French Frigate Shoals in the central North Pacific indicate that there are two major sources of Pu in this basin: close-in (tropospheric) fallout from nuclear weapons testing at the Pacific Proving Grounds in the Marshall Islands in the 1950s and global (stratospheric) fallout which peaked in 1962. Furthermore, the $^{240}Pu/^{239}Pu$ atom ratio of fallout from the Pacific Proving Grounds is characteristically higher (0·24) than that of global fallout Pu (0·18–0·19). Seawater and sediment samples from the North Pacific exhibit a wide range of $^{240}Pu/^{239}Pu$ values (0·19–0·34), with a trend towards higher ratios in the subsurface waters and sediment. Deep water $^{240}Pu/^{239}Pu$ ratios are higher in the vicinity of the Marshall Islands relative to stations further from this close-in fallout source. These preliminary data suggest that fallout Pu from the Pacific Proving Grounds is more rapidly removed from the surface waters than is global fallout Pu. Plutonium geochemistry appears to be related to the physical/chemical form of Pu-bearing particles generated by different fallout sources. © *1997 Elsevier Science Limited.*

INTRODUCTION

The input of fallout plutonium (Pu) from atmospheric nuclear weapons testing is the largest source of Pu to the environment, including the oceans (Harley, 1980; Perkins & Thomas, 1980). While the distribution of fallout Pu in the oceans has been well studied (Sholkovitz, 1983 and references

therein), there is little information on the different sources of fallout Pu and their relationship to Pu marine geochemistry. In this article, $^{240}Pu/^{239}Pu$ isotopic and Pu activity data for seawater, sediment, and coral samples from the North Pacific are presented. These preliminary data confirm previous studies (Bowen *et al.*, 1980) that suggest that the Pacific Proving Grounds is a major source of fallout Pu locally in the North Pacific basin. More importantly, it appears that there is a different marine geochemistry between close-in fallout originating from the Pacific Proving Grounds and stratospheric fallout Pu. The geochemical behavior of fallout Pu appears to be related to the physical/chemical form of Pu-bearing fallout particles that are generated under differing testing conditions. These findings will have major implications for geochemical studies that require a knowledge of the local fallout Pu input function. This would include studies that use Pu to estimate sediment accumulation and mixing rates (Guinasso & Schink, 1975; Krishnaswami *et al.*, 1980; Benninger & Krishnaswami, 1981; Kershaw *et al.*, 1983; Cochran, 1985), and in studies that use Pu to trace scavenging and removal processes in the water column (Bowen *et al.*, 1980; Santschi *et al.*, 1980; Fowler *et al.*, 1983; Livingston & Anderson, 1983; Bacon *et al.*, 1985).

In nuclear-weapons fallout, the dominant long-lived Pu isotopes are ^{239}Pu and ^{240}Pu, the combined activities of which have been traditionally measured by alpha-counting techniques (written as $^{239,240}Pu$). By employing mass spectrometric (m.s.) techniques, it is possible to determine separately the atomic abundances of ^{239}Pu and ^{240}Pu in marine samples (Buesseler & Halverson, 1987). Differences in weapons design and yield account for much of the variability seen in the $^{240}Pu/^{239}Pu$ ratios found in fallout samples (Perkins & Thomas, 1980; Koide *et al.*, 1985). Therefore, the isotopic signature of Pu in a given sample can be used as an indication of the different sources of fallout Pu at a given site (Beasley *et al.*, 1982; Buesseler & Sholkovitz, 1987a; Smith *et al.*, 1995).

BACKGROUND

The history of atmospheric nuclear weapons testing, and hence fallout Pu deposition, can be broadly broken down into two phases: the pre- and post-moratorium weapons testing years. During the pre-moratorium years of 1952–1958, large-scale testing programmes of the USA were conducted at the Pacific Proving Grounds, and these tests account for $> 50\%$ of the total fallout inputs at this time (calculated from announced yields in Department of Energy (1982) by Buesseler (1986)). Between November 1958 and September 1961, the testing moratorium between the United

States and the former Soviet Union (FSU) temporarily ended all new fallout inputs. Global fallout deposition peaked in 1963 during the post-moratorium period with the resumption of atmospheric testing. In 1963, the Limited Test Ban Treaty between the USA and FSU put an end to the major atmospheric nuclear testing programs. The dominant source of fallout Pu in the early 1960s was atmospheric nuclear weapons testing by the FSU. Fallout debris from the FSU testing program was largely injected into the stratosphere where it has a half-residence time in the order of one year, before being deposited globally (Joseph *et al.*, 1971; Perkins & Thomas, 1980). The FSU testing programs accounted for ≅75% of total global fallout Pu inputs (calculated from announced yields in Department of Energy (1982) by Buesseler (1986)). Limited atmospheric testing continued through the 1960s and 1970s by the French and Chinese.

The atomic ratio of $^{240}Pu/^{239}Pu$ in fallout is variable, and depends upon the specific weapons design and test yield. The global fallout average $^{240}Pu/^{239}Pu$ atom ratio is 0·18, based upon atmospheric aerosol sampling, soil samples and ice core data (HASL-273, 1973; Krey *et al.*, 1976; Koide *et al.*, 1985). However, different test series can be characterized by either higher or lower ratios. For example, fallout from the Nevada test site is characterized by generally lower $^{240}Pu/^{239}Pu$ ratios, averaging 0·035 (Hicks & Barr, 1984; Buesseler & Sholkovitz, 1987*b*). Of interest to this study are reports of elevated $^{240}Pu/^{239}Pu$ ratios (0·21–0·36) in fallout from the 1950s from the Pacific Proving Grounds. Higher ratios can be generally expected with the higher neutron fluxes associated with an increase in the yield of a nuclear detonation, and will vary with weapons design. Elevated ratios attributed to the Pacific source have been seen in ice core records (Koide *et al.*, 1985), soil samples from the Bikini atoll (Noshkin, 1978), coral samples from Bikini lagoon (Noshkin, 1978) and individual high yield tests, such as the "Mike" shot in 1952 at Enewetak (Diamond *et al.*, 1960).

In contrast to the FSU tests which were conducted primarily in the air, the majority of the tests at the Pacific Proving Grounds were conducted at or just above the earth's surface (for our purposes, we consider all announced barge, tower, surface and < 500 ft balloon tests, as 'surface' tests). Therefore, in addition to the injection of fallout debris into the stratosphere, these surface-based tests will produce significant quantities of close-in, or so-called tropospheric fallout. As the name implies, such fallout will remain within the troposphere, from which it is rapidly deposited (within minutes to a few days) (Joseph *et al.*, 1971; Holloway & Hays, 1982). Depending upon local wind conditions and wet and dry deposition patterns, tropospheric fallout will be deposited preferentially within the vicinity of the test site (Machta *et al.*, 1956).

While the relative magnitude of close-in (tropospheric) and global (stratospheric) fallout generated from the Pacific Proving Grounds tests varied considerably, as a whole, this site remains the world's largest source of close-in fallout. Based upon announced nuclear test information, the Pacific Proving Grounds tests account for >95% of all surface-based nuclear weapons tests (based upon yields calculated from Department of Energy (1982) and reported in Buesseler (1986)). By comparing marine water column and soil ^{239,240}Pu inventories, it is estimated that up to 60% of the Pu inventory in the North Pacific originated from local sources (Bowen *et al.*, 1980).

The first ocean-wide study of Pu in seawater originated with the Pacific GEOSECS sampling program in the early 1970s, some 10–15 years after the major inputs of fallout Pu. In their studies, Bowen *et al.* (1980) found two main features in the water column ^{239,240}Pu activity distributions that are characteristic of Pu in the Pacific. First, at approximately 200–500 m, they found a ^{239,240}Pu activity maximum between the surface waters that are depleted in ^{239,240}Pu, and the mid-water depths that are also low in ^{239,240}Pu. Secondly, at some of the Pacific stations, there exists a near-bottom enrichment of ^{239,240}Pu in the water column. Interestingly, this second feature is most evident in the NW Pacific stations that are more likely to be impacted by close-in fallout from the Pacific Proving Grounds. The data presented here are intended to provide insight into the link between Pu's source, and hence its isotopic signature, and these water column features.

SAMPLING AND METHODS

In this study, the mass spectrometric analyses of Pu in a coral record will be used to show the magnitude, timing and isotopic signature of fallout in the North Pacific basin, since the annual bands of corals have been shown to record the history of fallout Pu deposition quite well (Benninger & Dodge, 1986). The coral data will then be used to interpret the m.s. data from the Pacific seawater and sediment samples. All of the samples analyzed for this study were obtained from previous collections in the Pacific, and their locations are provided in Tables 1 and 2.

The coral data are from the French Frigate Shoals in the North Pacific. This coral sample had been previously sectioned into annual growth bands, and the ^{14}C record in this coral record can be found in Druffel (1987). The seawater samples were obtained from a variety of cruises between the mid-1970s and 1980 collected by the research group of V. T. Bowen *et al.* Seawater samples were collected with large volume sampling

TABLE 1
Coral Plutonium Data — French Frigate Shoals Island (24°N, 166°W)

Coral band	$^{240}Pu/^{239}Pu^{a}$ *atom ratio*	$^{239,240}Pu^{a}$ *(Bq/kg)*
1955	0·229 ± 0·036	0·367 ± 0·028
1956	0·243 ± 0·005	0·233 ± 0·008
1957–1958	0·239 ± 0·035	0·227 ± 0·047
1960	0·214 ± 0·005	0·200 ± 0·017
1961	0·203 ± 0·008	0·200 ± 0·050
1962	0·194 ± 0·002	0·188 ± 0·003
1963–1964	0·194 ± 0·002	0·150 ± 0·002

[a] $^{240}Pu/^{239}Pu$ atom ratios and $^{239,240}Pu$ activities are reported in this study with their propagated one sigma counting errors.

bottles and transferred for storage into 55-liter plastic deldrums prior to analysis. Since no filtration steps were taken upon collection, these samples represent total dissolved and particulate Pu. It has been shown that particulate Pu represents on average 6% of the total Pu activity in Pacific water samples (Krishnaswami *et al.*, 1976; Livingston *et al.*, 1987). Plutonium-239,240 water column profiles at many of these sites obtained by alpha-counting procedures have been previously reported (Noshkin & Wong, 1979; Bowen *et al.*, 1980; Nagaya & Nakamura, 1984). The seawater samples are unanalyzed duplicate samples from earlier collections that have been stored at Woods Hole since their collection. Replicate analyses ($n = 50$) of seawater samples that have been stored for up to 15 years at Woods Hole show that there are no artifacts in the Pu determinations due to the long-term storage of these samples (H. D. Livingston, personal communication). The Pacific sediment samples were collected with a box corer, and determinations of sediment $^{239,240}Pu$ activity had previously been made by alpha-counting at these sites (H. D. Livingston, personal communication; Cochran, 1985).

The analytical procedures for the Pu mass spectrometry analysis are described in detail elsewhere (Buesseler & Halverson, 1987). Briefly, a ^{242}Pu spike is added to the sample in order to follow the chemical yield of the separation and purification procedures and to determine the ^{239}Pu and ^{240}Pu concentrations by isotope dilution mass spectrometry. In addition to being able to measure the ^{239}Pu and ^{240}Pu isotopes, the m.s. techniques have a higher sensitivity for Pu than do alpha-counting methods. The thermal ionization m.s. facility used in this study is at the Savannah River Laboratory in Aiken, South Carolina, USA (Halverson, 1981). Sample sizes for the seawater averaged 4 kg, the coral samples ranged from 0·3 to 8 g, and the sediment samples were a few grams in weight (Buesseler, 1986).

RESULT AND DISCUSSION

For comparison to our data, a fallout deposition record from New York City and a coral Pu record from St. Croix in the Virgin Islands are shown in Fig. 1 along with our Pacific Coral results. The St. Croix ^{239,240}Pu activities in the annual coral bands were analyzed by Benninger and Dodge (1986). As the authors point out, there is a striking similarity between the fallout input curve and the Atlantic coral Pu record. Although the exact mechanism for the incorporation of fallout Pu into the coral matrix is not well understood, the annual bands of corals can provide us with a record of surface water Pu deposition. The French Frigate Shoals coral samples were analyzed here to examine the difference between North Pacific Pu fallout in the pre-and post-moratorium periods. The Pacific coral data contrast sharply with both of the fallout records in Fig. 1 in two major ways. First, the shape of the ^{239,240}Pu activity curve differs in the Pacific samples, in that there is no evidence of a maximum

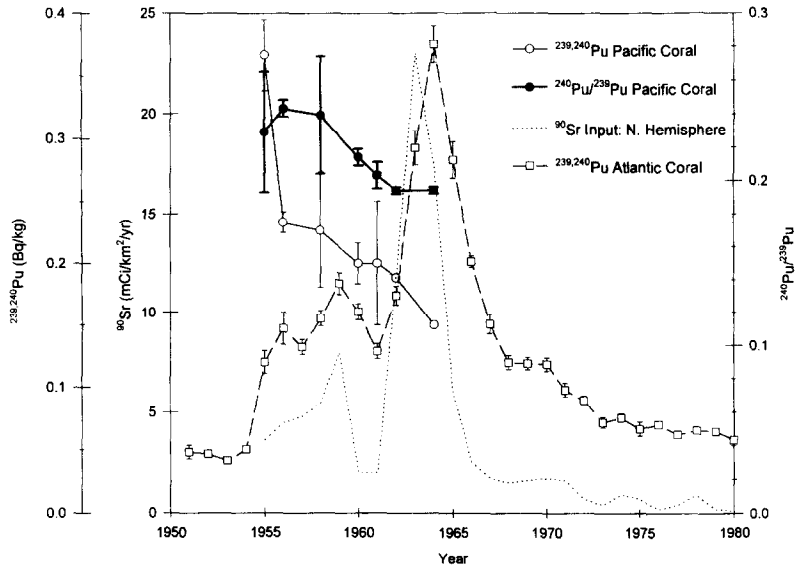

Fig. 1. A comparison of Pacific coral Pu (French Frigate Shoals—this study) and an Atlantic coral Pu record (St. Croix—Benninger and Dodge (1986)). For both coral records, total ^{239,240}Pu activities and associated errors are shown (open symbols; left x-axis). In addition, for the Pacific samples, the measured ^{240}Pu/^{239}Pu atom ratio is also shown (filled circles; right x-axis). The time-series record of ^{90}Sr fallout measured in atmospheric samples in New York is also shown (curve adapted from Larsen (1985); dotted line left x-axis). One would expect that a site receiving predominantly global fallout Pu would have a time-series record of Pu activities that was similar to the ^{90}Sr input record. Data for the Pacific Coral can be found in Table 1.

during peak global fallout input years around 1963. In fact, the 1950s Pu coral activities are highest ($>20\,dpm\,kg^{-1}$) in the earliest sample from 1955, and decrease towards the samples from 1961 and 1963. This implies that, at this central North Pacific island site, the magnitude of local fallout Pu deposition from testing in the 1950s was larger than Pu deposition from global fallout sources.

The contrasting $^{240}Pu/^{239}Pu$ ratios of the 1950s and 1960s coral samples are the second feature of interest in this data (Fig. 1 and Table 1). The North Pacific coral samples from peak global fallout years have a $^{240}Pu/^{239}Pu$ atom ratio of 0·19, which corresponds well with the global fallout average. However, the earlier samples from the mid- to late 1950s have a $^{240}Pu/^{239}Pu$ ratio of 0·24, which must reflect the elevated $^{240}Pu/^{239}Pu$ ratio of Pu that is characteristic of the Pacific Proving Grounds fallout.

The water column results have been divided into samples shallower than the Pu activity maximum (around 200–500 m in the 1970s) and those deeper. In this way, it can be seen that the $^{240}Pu/^{239}Pu$ ratios of the shallower samples are consistently lower than those in the deep waters, and are not significantly different to the global fallout average (Fig. 2A). The deep water values range from >0·20 to as high as 0·30, with some evidence of an increase towards the deeper samples (Table 2). These data are consistent with those of Bertine *et al.* (1986) (shown in Fig. 2A) which indicate higher ratios in deep samples, and ratios closer to the global fall-

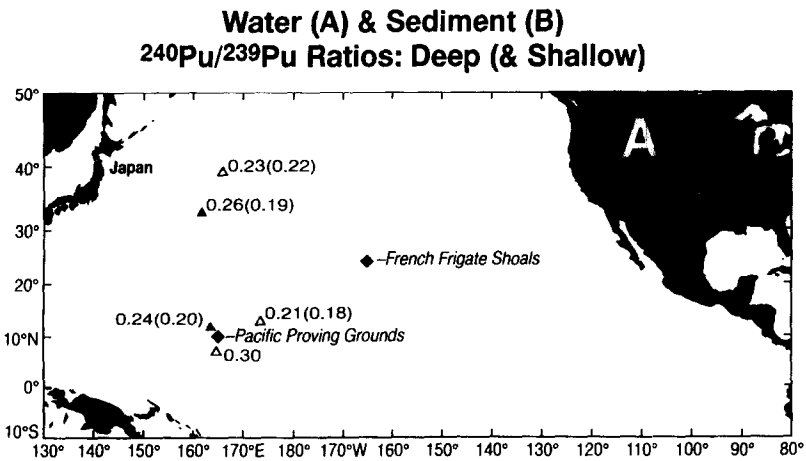

Fig. 2A. Map showing location and values of $^{240}Pu/^{239}Pu$ atom ratio in the water column. Average ratios from samples collected above the Pu maximum (water depths < 200–500 m) and those from deeper waters are indicated where available. The data are either from this study (open triangles, results in Table 2) or from Pu data from two sites reported in Bertine *et al.* (1986) (filled triangles).

TABLE 2
North Pacific $^{240}Pu/^{239}Pu$ Data

Cruise	*Station*	*Depth (cm)*	*Latitude (°N)*	*Longitude*	*Year collected*	$^{240}Pu/^{239}Pu \pm error$
Sediments						
MANOP	C	Surface	1°1·6′	138°56·1′W	1982	0·293 ± 0·038
MANOP	C	0–1	1°1·6′	138°56·1′W	1982	0·204 ± 0·004
MANOP	H	0–1·2	6°32·9′	92°53·4′W	1981	0·209 ± 0·004
MANOP	M	0–2	8°47·1′	103°59·5′W	1981	0·226 ± 0·009
MANOP	S	0–1	11°3·1′	140°4·7′W	1982	0·203 ± 0·016
MANOP	S	Surface	11°3·1′	140°4·7′W	1982	0·185 ± 0·003
Kn73/6	14	0–1	7°03·2′	164°47·3′W	1978	0·336 ± 0·012
Yaquina	59	0–1	33°10·65′	150°54·4′W	1974	0·187 ± 0·019
Hakuho Maru	8	0–1	38°00·0′	179°45·0′W	1980	0·365 ± 0·041
TT 141–2	15	1–2	39°22·5′	127°06·1′W	1979	0·175 ± 0·005
Seawater		Depth (m)				
Kn73/6	14	5925	7°03·2′	164°47·3′W	1978	0·298 ± 0·016
Kn73/6	3	400	12°52·0′	173°30′E	1978	0·178 ± 0·008
Kn73/6	3	750	12°52·0′	173°30′E	1978	0·201 ± 0·003
Kn73/6	3	1500	12°52·0′	173°30′E	1978	0·209 ± 0·011
Hakuho Maru	6	200	39°01·0′	166°00′E	1980	0·220 ± 0·009
Hakuho Maru	6	500	39°01·0′	166°00′E	1980	0·216 ± 0·029
Hakuho Maru	6	2250	39°01·0′	166°00′E	1980	0·250 ± 0·011
Hakuho Maru	6	5470	39°01·0′	166°00′E	1980	0·236 ± 0·015

out average in surface waters. All of the stations sampled for seawater Pu are within a region of the NW Pacific where total $^{239,240}Pu$ water column inventories can be as much as three to ten times greater than that expected from similar latitudes based upon estimates of global fallout inventories found in soil samples (Bowen *et al.*, 1980; Nagaya & Nakamura, 1984). The elevated water column $^{240}Pu/^{239}Pu$ ratios thus confirm earlier speculation that the enhanced inventories were related to close-in fallout from testing at the Pacific Proving Grounds.

The surface sediment samples analyzed for $^{240/239}Pu$ ratios were taken from a much wider geographical range in the Pacific (Fig. 2B). Those stations with an elevated deep water $^{240}Pu/^{239}Pu$ signature also had substantially elevated sedimentary $^{240}Pu/^{239}Pu$ ratios of up to 0·36. Slightly elevated $^{240}Pu/^{239}Pu$ ratios ($>0·20$) are seen in surface sediments from the equatorial region, and global fallout ratios of 0·18–0·19 were seen north of Hawaii and near the eastern Pacific margin. While these surface sediment ratios are not sufficient (either in depth or aerial coverage) to calculate total inventories of global fallout vs. close-in fallout in this basin, they clearly indicate an enhanced deposition of high-ratio material near the Pacific Proving Grounds.

It has been suggested that the physical/chemical form of fallout Pu is not identical from all of its sources, and this has played a major role in determining Pu's geochemical behavior in the oceans (Buesseler & Sholkovitz, 1987). In global fallout, Pu is carried by sub-micron-sized particles that are composed primarily of iron-oxides (Adams *et al.*, 1960; Joseph *et al.*, 1971; Weimer & Langford, 1978). These particles result from the vaporization and subsequent condensation of nuclear weapons materials without any interaction with the earth's surface.

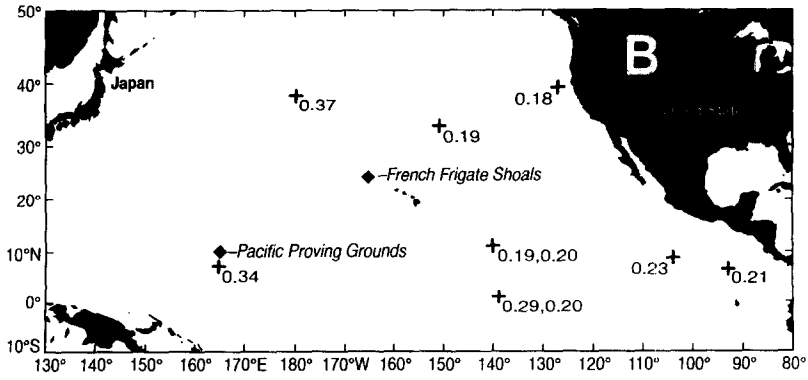

Fig. 2B. Map showing location and values of $^{240}Pu/^{239}Pu$ atom ratio in surface sediments. All data are from this study and can be found in Table 2.

Weimer and Langford (1978) found that 30–97% of the ^{55}Fe activity of a stratospheric fallout aerosol sample was released to solution upon 24-h contact with seawater.

With surface-based testing on a coral atoll such as Bikini or Enewetak, the particles carrying fallout Pu would be quite different. Such a blast will incorporate large quantities of partially and completely vaporized calcium, carbonates, calcium oxides and calcium hydroxides which are generated from the coral matrixes (Adams *et al.*, 1960). Upon hydration of the calcium oxides to calcium hydroxides, the fallout particles swell and develop into what is described as a 'crumbly, fluffy structure' (Adams *et al.*, 1960). Upon interaction with seawater, there is a partial solubilization of the calcium hydroxide-based fallout particles. This is accompanied by the release of hydroxyl ions and the interaction with magnesium ions in seawater to form a shell of insoluble magnesium hydroxide on the fallout particle.

The physical/chemical form of tropospheric fallout particles from the Pacific Proving Grounds is therefore quite different than that of global stratospheric fallout in general. The enhanced isotopic ratios in the deep waters and sediments of the NW Pacific suggest that the Pacific Proving Grounds fallout is more rapidly removed from the surface waters than is global fallout. This is consistent with sediment and water column ^{239,240}Pu inventory data that suggest that the percentage of Pu in the sediments out of the total sediment and water column inventory is largest at the stations that are closest to the Marshall Islands (Bowen *et al.*, 1980, and unpublished results; Nagaya & Nakamura, 1984).

More extensive Pu isotopic studies in the Atlantic have now demonstrated that a geochemical separation can occur in the ocean between Pu from different sources. In the North Atlantic, it has been shown (Buesseler, 1986; Buesseler & Sholkovitz, 1987*a*) that silicate-based fallout debris from surface-based testing on soils at the Nevada Test Site is more efficiently removed from the surface ocean to deep ocean sediments than is global fallout Pu. The Nevada Test Site fallout is a very small fraction of the total Pu inventory in the Atlantic ($< 1\%$), and hence it is only detected in the deep ocean sediments where it is enriched relative to global fallout. The Nevada fallout has an average ^{240}Pu/^{239}Pu ratio of 0·035 (Hicks & Barr, 1984; Buesseler, 1986), and in the North Atlantic (Buesseler, 1986; Buesseler & Sholkovitz, 1987*a*) and Gulf of Mexico (Scott *et al.*, 1983) the sediment ^{240}Pu/^{239}Pu ratios decrease with increasing water depth as the percentage of Nevada fallout in the sediment increases.

Silicate-based tropospheric fallout particles from Nevada are not identical to the calcium carbonate-based Marshall Islands fallout particles, but

both are apparently more efficiently removed from the surface waters than Pu from global fallout sources. In a more recent study, similar principles of a geochemical fractionation of fallout Pu from different sources have been applied to account for anomalously low $^{240}Pu/^{239}Pu$ ratios in a core off Novava Zemlya (Smith *et al.*, 1995). In this study, a single underwater test is identified as being the most likely source of local fallout Pu that had preferentially accumulated in a marine core.

In general, it now appears that global fallout Pu behaves as a rather soluble tracer in the ocean (< 1–6% particulate in sub-surface waters; Cochran *et al.*, 1987; Livingston *et al.*, 1987). While some fraction of fallout Pu is rapidly removed from the surface ocean and accumulates in marine sediments, the bulk of the stratospheric fallout Pu remains in solution with a much longer residence time (> 10–100 years). Pacific Proving Grounds fallout represents a large fraction of the total magnitude of Pu inputs to the North Pacific. Fallout Pu from this source is preferentially removed from the water column, and hence its influence on the Pu distribution will be considerable in this basin.

CONCLUSIONS

In summary, there are a number of features that these Pu isotopic data suggest. First, the coral data show that the Pacific Proving Grounds are a major source of fallout Pu for the North Pacific, and furthermore, these data confirm that fallout from this source is characterized by an elevated $^{240}Pu/^{239}Pu$ ratio (> 0·20) relative to global stratospheric fallout (0·18–0·19). Second, it appears that the Marshall Islands fallout is more rapidly removed from the surface waters than is global fallout, such that higher $^{240}Pu/^{239}Pu$ ratios are preferentially found at depths below the subsurface Pu maximum, in the deeper water samples, and in the underlying sediments. Finally, the data show elevated $^{240}Pu/^{239}Pu$ ratios in subsurface waters and sediments as far north as 38°N in the NW Pacific, but the influence of this source is less evident in the NE Pacific. For a more complete understanding of Pu's behavior in the North Pacific, one will need to consider the two major sources of Pu in this basin, i.e. stratospheric fallout and local tropospheric fallout from the Pacific Proving Grounds, as being separate in terms of both their input histories and subsequent scavenging and removal rates.

As a consequence of this idea of distinct fallout sources with different Pu geochemistries, the use of Pu as a geochemical tracer will need to be modified. For instance, in order to use $^{239,240}Pu$ in sediments to model

sediment mixing and accumulation rates, an input function for fallout Pu to the sediments must be chosen (Guinasso & Schink, 1975; Cochran, 1985; Buesseler, 1986; Lapicque *et al.*, 1987). If Pu from the Marshall Islands source is rapidly removed to sediments, then this will need to be incorporated into a more "pulse-like" Pu input function. Using present-day Pu removal rates may not be sufficient. In a general sense, if Pu's geochemistry is strongly influenced by its specific source, then any predictions of Pu's behaviour will need to include this consideration. Plutonium isotopic ratios will provide a unique tool with which to study questions.

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REFERENCES

Adams, C. E., Farlow, N. H. & Schell, W. R. (1960). The compositions, structures and origins of radioactive fall-out particles. *Geochim. Cosmochim. Acta*, **18,** 42–56.

Bacon, M. P., Huh, C.-A., Fleer, A. P. & Deuser, W. G. (1985). Seasonality in the flux of natural radionuclides and plutonium in the deep Sargasso Sea. *Deep-Sea Res.*, **32,** 273–86.

Beasley, T. M., Ball, L. A., Andrews, J. E. III & Halverson, J. E. (1981). Hanford-derived plutonium in Columbia River sediments. *Science*, **214,** 913–5.

Beasley, T. M., Carpenter, R. & Jennings, C. D. (1982). Plutonium, ^{241}Am and ^{137}Cs ratios, inventories and vertical profiles in Washington and Oregon continental shelf sediments. *Geochim. Cosmochim. Acta*, **46,** 1931–46.

Benninger, L. K. & Dodge, R. E. (1986). Fallout plutonium and natural radionuclides in annual bands of the coral *Montastrea annularis*, St. Croix, U.S. Virgin Islands. *Geochim. Cosmochim. Acta*, **50,** 2785–97.

Bertine, K. K., Chow, T. J., Koide, M. & Goldberg, E. D. (1986). Plutonium isotopes in the environment: Some existing problems and some new ocean results. *J. Environ. Radioactivity*, **3,** 189–201.

Bowen, V. T., Noshkin, V. E., Livingston, H. D. & Volchok, H. L. (1980). Fallout radionuclides in the Pacific Ocean: Vertical and horizontal distributions, largely from GEOSECS stations. *Earth Planet. Sci. Lett.*, **49,** 411–34.

Buesseler, K. O. (1986). Plutonium isotopes in the North Atlantic. Ph.D. Thesis, MIT/WHOI Joint Program in Oceanography, 220 pp.

Buesseler, K. O. & Halverson, J. (1987). The mass spectrometric analysis of fallout ^{239}Pu and ^{240}Pu in marine samples. *J. Environ. Radioactivity*, **5,** 425–44.

Buesseler, K. O. & Sholkovitz, E. R. (1987*a*). The geochemistry of fallout plutonium in the North Atlantic: I. A pore water study in shelf, slope and deep-sea sediments. *Geochim. Cosmochim. Acta*, **51,** 2605–22.

Buesseler, K. O. & Sholkovitz, E. R. (1987*b*). The geochemistry of fallout plutonium in the North Atlantic: II. ^{240}Pu/^{239}Pu ratios and their significance. *Geochim. Cosmochim. Acta*, **51,** 2623–37.

Cochran, J. K. (1985). Particle mixing rates in sediments of the eastern equatorial Pacific: Evidence from ^{210}Pb, ^{239,240}Pu and ^{137}Cs distributions at MANOP sites. *Geochim. Cosmochim. Acta*, **49,** 1195–210.

Cochran, J. K., Livingston, H. D., Hirschberg, D. J. & Surprenant, L. D. (1987). Natural and Anthropogenic Radionuclide distributions in the Northwest Atlantic Ocean. *Earth Planet. Sci. Lett.*, **84,** 135–52.

Department of Energy (1982). Announced United States Nuclear Tests, July 1945 through December 1981. NVO-209, Office of Public Affairs, US Dept. of Energy, Nevada Operations Office.

Diamond, H., Fields, P. R., Stevens, C. S., Studier, M. H., Fried, S. M., Inghram, M. G., Hess, D. C., Pyle, G. L., Mech, J. F., Manning W. M., Ghiorso, A., Thompson, S. G., Higgins, G. H., Seaborg, G. T., Browne, C. I., Smith, H. L. & Spence, R. W. (1960). Heavy Isotope Abundances in Mike Thermonuclear Device. *Physical Rev.*, **119,** 2000–4.

Druffel, E. R. M. (1987). Bomb radiocarbon in the Pacific: Annual and seasonal timescale variations. *J. Mar. Res.*, **45,** 667–98.

Fowler, S. W., Ballestra, S., LaRosa, J. & Fukai, R. (1983). Vertical transport of particulate-associated plutonium and americium in the upper water column of the Northeast Pacific. *Deep-Sea Res.*, **30,** 1221–33.

Guinasso, N. L. & Schink, D. R. (1975). Quantitative estimates of biological mixing rates of abyssal sediments. *J. Geophys. Res.*, **80,** 3032–43.

Halverson, J. E. (1981). A three-state mass spectrometer for isotope analysis of radionuclides in environmental samples. E.I. du Pont de Nemours & Co. Environmental Sci. Div. Rpt. DP-1611, pp. 1–13.

Harley, J. H. (1980). Plutonium in the environment—a review. *J. Radiat. Res.*, **21,** 83–104.

HASL-273 (1973). Global atmospheric plutonium-239 and plutonium isotopic ratios for 1959–1970. In: HASL-273 (Fallout Program Quarterly Summary Report, US Dept. of Energy), pp. III-2–III-28.

Hicks, H. G. & Barr, D. W. (1984). Nevada test site fallout atom ratios: ^{240}Pu/^{239}Pu and ^{241}Pu/^{230}Pu. UCRL-53499/1 (Lawrence Livermore National Laboratory), 4 pp.

Holloway, R. W. & Hayes, D. W. (1982). Mean residence time of plutonium in the troposphere. *Environ. Sci. Technol.*, **16,** 127–29.

Joseph, A. B., Gustafson, P. F., Russell, I. R., Schuert, E. A., Volchok, H. L. & Templin, A. (1971). Sources of radioactivity and their characteristics. In *Radioactivity in the Marine Environment* (National Academy of Sciences), Chapter 2, pp. 6–41.

Kerhsaw, P. J., Swift, D. J., Pantreath, R. J. & Lovett, M. B. (1983). Plutonium redistribution by biological activity in Irish Sea sediments. *Nature*, **306,** 774–5.

Koide, M., Bertine, K. K., Chow, T. J. & Goldberg, E. D. (1985). The $^{240}Pu/^{239}Pu$ ratio, a potential geochronometer. *Earth Planet. Sci. Lett.*, **72,** 1–8.

Krey, P. W., Hardy, E. P., Pachucki, C., Rourke, F., Coluzza, J. & Benson, W. K. (1976). Mass isotopic composition of global fall-out plutonium in soil. In *Transuranium Nuclides in the Environment*, IAEA-SM-199/39 (IAEA, Vienna), pp. 671–8.

Krishnaswami, S., Benninger, L. K., Aller, R. C. & von Damm, K. L. (1980). Atmospherically-derived radionuclides as tracers of sediment mixing and accumulation in near-shore marine and lake sediments: evidence from ^{7}Be, ^{210}Pb, and $^{239,240}Pu$. *Earth Planet. Sci. Lett.*, **47,** 307–18.

Lapicque, G., Livingston, H. D., Lambert, C. E., Bard, E. & Labeyrie, L. D. (1987). Interpretation of $^{239,240}Pu$ in Atlantic sediments with a non-steady state input model. *Deep-Sea Res.*, **34,** 1841–50.

Larsen, R. J. (1985). Worldwide deposition of ^{90}Sr through 1983. US Dept. of Energy Report EML-444, 159 pp.

Livingston, H. D. & Anderson, R. F. (1983). Large particle transport of plutonium and other fallout radionuclides to the deep ocean. *Nature*, **303,** 228–31.

Livingston, H. D., Mann, D. R., Casso, S. A., Schneider, D. L., Surprenant, L. D. and Bowen, V. T. (1987). Particle and Solution Phase Depth Distributions of Transuranics and ^{55}Fe in the North Pacific. *J. Environ. Radioactivity*, **5,** 1–24.

Machta, L., List, R. J. & Hubert, L. F. (1956). World-wide travel of atomic debris. *Science*, **124,** 474–7.

Nagaya, Y. & Nakamura, K. (1984). $^{239,240}Pu$, ^{137}Cs and ^{90}Sr in the central North Pacific. *J. Oceanogr. Soc. Japan*, **40,** 416–24.

Noshkin, V. E. (1978). Transuranium radionuclides in components of the benthic environment of Enewetak Atoll. Preprint UCRL-80587 (Lawrence Livermore Laboratory).

Noshkin, V. E. & Wong, K. M. (1979). Plutonium mobilization from sedimentary sources to the marine environment. In Third Nuclear Energy Agency Seminar on Marine Radioecology (Tokyo).

Perkins, R. W. & Thomas, C. W. (1980). Worldwide fallout. In *Transuranic Elements in the Environment*, ed. W. C. Hanson, pp. 53–82. Tech. Inf. Center US Department of Energy.

Santschi, P. H., Li, Y.-H., Bell, J., Trier, R. & Kawtaluk, K. (1980). Pu in coastal marine environments. *Earth Planet. Sci. Lett.*, **51,** 248–65.

Scott, M. R., Salter, P. F. & Halverson, J. E. (1983). Transport and deposition of plutonium in the ocean: Evidence from Gulf of Mexico sediments. *Earth Planet Sci. Lett.*, **63,** 202–22.

Sholkovitz, E. R. (1983). The geochemistry of plutonium in fresh and marine water environments. *Earth Sci. Rev.*, **19,** 95–161.

Smith, J. N., Ellis, K. M., Naes, K., Dahle, S. & Matishov, D. (1995). Sedimentation and Mixing Rates of Radionuclides in Barens Sea Sediments off Novaya Zemlya. *Deep-Sea Res. II*, **42,** 1471–93.

Weimer, W. C. & Langford, J. C. (1978). Iron-55 and stable iron in oceanic aerosls: Forms and availability. *Atmos. Environ.*, **12,** 1201–5.