von Bertalanffy equation $X(T) = X_{max} - (X_{max} - X_0)e^{-RT}$ to generate ontogenetic growth curves; $X(T)$ is shell length at age T, X_0 is the settling size, assigned a length of 400 μm [R. A. Lutz, D. Jablonski, D. C. Rhoads, R. D. Turner, *Mar. Biol.* **57**, 127 (1980)], where X_{max} is the maximum shell length, and R is a time constant. The parameters X_{max} and R were obtained from the von Bertalanffy equation, rewritten as $\Delta X_t = X(T + \Delta T) - X(T) = AX + B$, where $A = -(1 - e^{-R\Delta T})$, where $B = X_{max}(1 - e^{-RT})$ and ΔX_t is the total measured increment in shell length during the period ΔT. The regression coefficients in Fig. 1B yielded estimates of A and B. The parameters X_{max} and R were then computed from $X_{max} = -B/A$ and $R = -M(1 + A)/\Delta T$.

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13. We thank J. F. Grassle for critical review of the manuscript and the Department of Zoology, Rutgers University, for use of facilities. Contribution No. 24 from the Galápagos Rift Hot Springs Biology Expedition and a paper of the Journal Series, New Jersey Agricultural Experiment Station, Cook College, Rutgers University, New Brunswick. This work was supported by NSF grant OCE-78-08854 to D.C.R. and NSF grant OCE-80-24897 to R.A.L.

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Hanford-Derived Plutonium in Columbia River Sediments

Abstract. *Mass spectrometry data on plutonium isolated from Columbia River sediments exhibit mean ratios of plutonium-240 to plutonium-242 consistent with those observed for integrated global fallout. Ratios of plutonium-240 to plutonium-239 show marked deviations from accepted fallout values, suggesting a second source of plutonium-239. This additional plutonium-239 arises from the decay of neptunium-239 produced in reactor effluent water from the old plutonium production reactors located on the Hanford Reservation. An estimated 20 to 25 percent of the total plutonium inventory in sediments behind McNary Reservoir, the first downriver site of fine sediment accumulation below the Hanford Reservation, is ascribed to reactor operations.*

The lower Columbia River received large amounts of artificial radioactivity during the period from 1944 to 1970 as a consequence of the operation of single-pass plutonium production reactors located on the Hanford Reservation in Washington State (*1*, *2*). Although much information has accumulated concerning the absolute activity levels and geochemical behavior of elements with short and moderately long-lived radioisotopes in the river system, the same cannot be said for the very long-lived transuranic radionuclides, that is, ^{239}Pu (half-life $t_{1/2}$ = 24,131 years), ^{240}Pu ($t_{1/2}$ = 6570 years), and ^{242}Pu ($t_{1/2}$ = 376,300 years) (*3*). We report here the results of mass spectrometry and absolute activity determinations of plutonium isolated from Columbia River sediments for the period from 1944 to 1977. The data indicate that reactor-derived plutonium constitutes between 20 and 25 percent of the integrated plutonium inventories in sediments 100 km downstream of the reactors. Furthermore, the plutonium contributed by the reactors appears to have been only ^{239}Pu.

Sediment cores were raised during August 1977 from McNary Reservoir on the Columbia River (*4*) and from Ice Harbor Reservoir on the Snake River (Fig. 1). The Ice Harbor samples served as controls since the plutonium present in these sediments is derived entirely from atmospheric fallout. The absolute amounts of $^{239, 240}$Pu in our samples were determined by the addition of known amounts of ^{242}Pu to 10 to 20 g of dry sediment prior to processing and subsequent α-spectrometry measurements; no plutonium isotopes were added to portions of the same sediment (50 g) from which plutonium was isolated for mass spectrometry measurements. These latter samples were leached three times with 6*N* HCl, and the plutonium was isolated and purified by our standard laboratory procedures. Further purification at the Savannah River Laboratory entailed small-column (0.4 ml), ion-exchange chromatography, the plutonium being finally concentrated onto two or three AG 1 × 4 resin beads (50 μm in diameter). The dried beads were then loaded into the mass spectrometer V filaments for analysis. The mass spectrometer used was a surface-ionization, three-stage instrument constructed at the Savannah River Laboratory.

Table 1 lists the absolute concentrations of $^{239, 240}$Pu at the various horizons sampled and the isotopic composition of the plutonium in both our Ice Harbor Reservoir and McNary Reservoir cores as of November 1980. The mean sedimentation rate at the McNary Reservoir coring site (~ 1.4 to 1.5 cm year^{-1}) was determined from the position of the $^{239, 240}$Pu subsurface maximum observed at 19 to 20 cm, which we attribute to maximum fallout delivery in mid-1963 (*5*). We observed a marked increase in the ratio of ^{238}Pu to $^{239, 240}$Pu between 13.5 and 16.5 cm (Table 1), which we attribute to the arrival of SNAP-9A (Systems for Nuclear Auxiliary Power) in the Northern Hemisphere in mid-1966 (*6*).

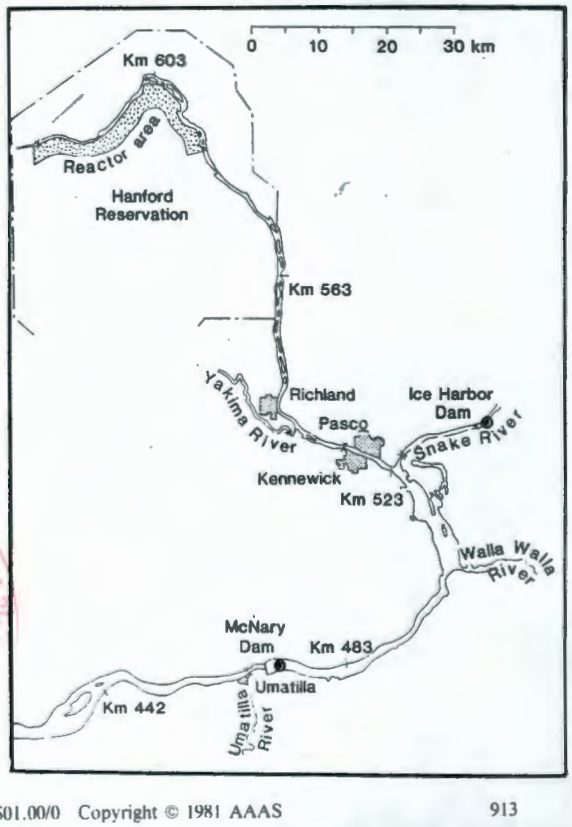

Fig. 1. Sampling sites for sediment cores raised from the Ice Harbor Reservoir and McNary Reservoir.

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Geochronologies based on the use of ^{210}Pb are not applicable to McNary Reservoir sediments (*4*). Although not given in Table 1, the plutonium profile for our Ice Harbor core evidenced the same type of distribution; sedimentation rates at that location were ~ 2 cm year^{-1}, and the total core length was 33 cm.

In interpreting our results, we have relied extensively on the data of Krey *et al.* (*7*), who reported plutonium isotopic ratios from homogenized soil cores (30 cm) collected from around the world and contaminated only by fallout debris. Their mean plutonium ratios as of 1 January 1971 were as follows: $^{240}Pu/^{239}Pu = 0.176 \pm 0.014$; $^{241}Pu/^{239}Pu = 0.0086 \pm 0.0017$; and $^{242}Pu/^{239}Pu = 0.0044 \pm 0.0011$. From these data we calculate a $^{240}Pu/^{242}Pu$ ratio of 40 ± 10. The mean $^{240}Pu/^{239}Pu$ and $^{240}Pu/^{242}Pu$ ratios for the fallout-contaminated sediments from the Snake River are 0.175 ± 0.002 and 37 ± 1, respectively (1σ propagated error), in excellent agreement with the soil data.

Our mean $^{240}Pu/^{242}Pu$ ratio for the sediments from McNary Reservoir (Table 1) is 36 ± 10, a value so consistent with the terrestrial fallout data that addition of either isotope to the sediments cannot be reasonably ascribed to reactor operations. Low-irradiation plutonium typical of uranium-fueled reactors exhibits $^{240}Pu/^{242}Pu$ ratios near 500 (*8*). We have analyzed various sediment horizons from other cores collected throughout the McNary Reservoir; the mean $^{240}Pu/^{242}Pu$ ratio in these nine samples was 41 ± 9.

The mean $^{240}Pu/^{239}Pu$ ratio in the McNary Reservoir sediments (Table 1) is 0.126 ± 0.031, a value significantly different from that derived from terrestrial soil data. Inclusion of the $^{240}Pu/^{239}Pu$ ratios from the additional cores mentioned above changes this ratio slightly to 0.121 ± 0.025. The difference in our $^{240}Pu/^{239}Pu$ ratios and those seen in global fallout must be due to additional ^{239}Pu contributed by the reactors. We believe that this ^{239}Pu originated through the decay of ^{239}Np ($t_{1/2}$ = 2.3 days), one of the isotopes of greatest abundance in Columbia River water during the years of reactor operation (*1*). This isotope is produced by slow neutron capture in uranium [$^{238}U(n,\gamma)^{239}U$] with subsequent decay of the ^{239}U ($t_{1/2}$ = 23.5 minutes) to ^{239}Np. The source of the uranium was that present in river water (~ 1 μg liter^{-1}) used to cool the reactor cores plus uranium occluded to the outside of the aluminum-clad fuel elements ("tramp" uranium).

To calculate the amount of ^{239}Pu added to river sediments by the reactors, we use the expression of Krey *et al.* (*7*):

$$\frac{(\text{Pu activity})_1}{(\text{Pu activity})_2} = \frac{(R_2 - R)(1 + 3.67 R_1)}{(R - R_1)(1 + 3.67 R_2)} \quad (1)$$

where R is the atom ratio of ^{240}Pu to ^{239}Pu measured in river sediments, R_1 is the atom ratio of ^{240}Pu to ^{239}Pu in the products of the reactors, R_2 is the atom ratio of ^{240}Pu to ^{239}Pu observed in integrated global fallout, plutonium activities 1 and 2 are the absolute activity ratios contributed by the reactors and global fallout, respectively, and the constant, 3.67, is the ratio of the half-lives of ^{239}Pu to ^{240}Pu (*9*). As the $^{240}Pu/^{239}Pu$ ratio contributed by the reactors is in this case zero, Eq. 1 simplifies to

$$\frac{(\text{Pu activity})_1}{(\text{Pu activity})_2} = \frac{R_2 - R}{R} \times \frac{1}{(1 + 3.67 R_2)} \quad (2)$$

Using a mean $^{240}Pu/^{239}Pu$ ratio of 0.126 for our McNary Reservoir core and 0.176 for global fallout, we calculate that, of the integrated inventory of 10 mCi km^{-2} of $^{239, 240}Pu$ (Table 1), approximately 2 mCi km^{-2} is reactor-derived. Using a mean $^{240}Pu/^{239}Pu$ ratio of 0.121 obtained by averaging all sediment analyses from McNary Reservoir increases this estimate to only 2.2 mCi km^{-2}. In all, a reactor plutonium contribution to these reservoir sediments of between 20 and 25 percent seems reasonable (*10*).

The mean $^{241}Pu/^{240}Pu$ ratio for the Ice Harbor Reservoir sediments, corrected to 1 January 1971, is 0.049 ± 0.008, in excellent agreement with the value of 0.049 ± 0.010 calculated from the fallout

Table 1. Absolute $^{239, 240}Pu$ activities and the plutonium isotopic composition in Snake River and Columbia River sediments. Uncertainties for sediment activities represent 1σ propagated errors in counting statistics for both photopeak and background α-spectrometry measurements. Errors for mass spectrometry measurements represent 1σ errors in the total ion counts observed at each mass number. Samples were collected in August 1977; we calculated the approximate deposition times for each sediment horizon by using sedimentation rates of 2 and 1.4 cm year^{-1} for the Ice Harbor core and McNary Reservoir core, respectively; N.M., not measured.

Depth interval (cm)	Time deposited	$^{239, 240}Pu$ [dpm kg^{-1} (dry)]	$^{238}Pu/^{239, 240}Pu$	Pu isotopic composition (atom percent)			
				239	240	241	242
			Ice Harbor				
0 to 2	Early 1977	27 ± 2	0.049 ± 0.016	84.6 ± 0.5	14.6 ± 0.4	0.44 ± 0.05	0.39 ± 0.04
6 to 7	Mid-1974	26 ± 1	0.067 ± 0.016	84.1 ± 0.3	15.0 ± 0.3	0.39 ± 0.05	0.40 ± 0.05
14 to 15	Mid-1970	59 ± 3	0.049 ± 0.006	84.4 ± 0.2	14.6 ± 0.2	0.40 ± 0.04	0.62 ± 0.08
16 to 17	Mid-1969	38 ± 1	0.049 ± 0.006	83.4 ± 0.7	15.8 ± 0.7	0.42 ± 0.17	0.42 ± 0.13
26.5 to 27.5	Early 1964	115 ± 4	0.024 ± 0.004	83.3 ± 0.1	15.8 ± 0.1	0.48 ± 0.01	0.42 ± 0.01
27.5 to 28.5	Mid-1963	120 ± 3	0.026 ± 0.005	83.5 ± 0.1	15.4 ± 0.1	0.64 ± 0.02	0.44 ± 0.03
			McNary				
0 to 2	Early 1977	32 ± 2	0.055 ± 0.010	86.6 ± 0.3	12.4 ± 0.3	0.6 ± 0.1	0.3 ± 0.1
4 to 6	Early 1974	31 ± 2	0.048 ± 0.009	86.0 ± 0.5	13.1 ± 0.5	0.44 ± 0.05	0.44 ± 0.06
7 to 8	Early 1972	30 ± 2	0.056 ± 0.013	85.3 ± 0.3	13.6 ± 0.3	0.60 ± 0.04	0.36 ± 0.04
10 to 11	Early 1970	30 ± 2	0.080 ± 0.010	85.5 ± 0.4	13.5 ± 0.4	0.65 ± 0.07	0.33 ± 0.04
13 to 14	Early 1978	41 ± 2	0.080 ± 0.011	86.5 ± 0.2	12.4 ± 0.2	0.57 ± 0.03	0.40 ± 0.04
16 to 17	Late 1965	92 ± 5	0.037 ± 0.009	88.3 ± 0.2	11.0 ± 0.2	0.44 ± 0.03	0.21 ± 0.03
19 to 20	Mid-1963	181 ± 7	0.032 ± 0.004	88.4 ± 0.2	11.0 ± 0.2	0.41 ± 0.01	0.25 ± 0.01
22 to 23	Mid-1961	137 ± 6	0.035 ± 0.005	89.5 ± 0.3	9.9 ± 0.3	0.33 ± 0.01	0.21 ± 0.03
25 to 26	Mid-1959	58 ± 3	0.035 ± 0.008	88.1 ± 0.3	11.3 ± 0.3	0.30 ± 0.03	0.29 ± 0.03
28 to 29	Early 1957	27 ± 2	0.021 ± 0.008	87.8 ± 0.3	11.4 ± 0.4	0.33 ± 0.03	0.39 ± 0.06
31 to 32	Early 1955	84 ± 3	0.018 ± 0.004	87.8 ± 0.3	11.5 ± 0.3	0.32 ± 0.02	0.36 ± 0.02
34 to 35	Early 1953	35 ± 2	0.010 ± 0.006	93.1 ± 0.2	6.6 ± 0.2	0.13 ± 0.01	0.15 ± 0.02
38 to 39	Early 1950	3 ± 0.7	N.M.	91.6 ± 0.6	7.2 ± 0.6	0.41 ± 0.12	0.59 ± 0.16
49 to 50		N.M.	N.M.	94.0 ± 0.4	5.4 ± 0.4	0.3 ± 0.1	0.3 ± 0.1

data of Krey *et al.* (7) for the same reference date (^{241}Pu $t_{1/2}$ = 14.4 years). Between 16 and 39 cm (~ 1967 to 1950) in the McNary Reservoir core the mean $^{241}Pu/^{240}Pu$ ratio, corrected to 1 January 1971, is 0.049 ± 0.010; this value indicates no significant reactor contribution of either isotope. However, between 0 and 15 cm, the mean $^{241}Pu/^{240}Pu$ ratio is significantly higher than 0.049. We believe that these surficial sediments reflect the input of ^{241}Pu, principally from aboveground tests conducted by the People's Republic of China, whose first thermonuclear device was detonated in June 1967 (*11*). That this perturbation in the $^{241}Pu/^{240}Pu$ ratio is not evidenced in the Ice Harbor core we attribute to the provenance of the material accumulating there (*12*).

Low $^{240}Pu/^{239}Pu$ ratios were observed in the deepest sections of our McNary Reservoir core, which correspond in time to the early atmospheric tests of the 1950's. We believe that these ratios reflect a high ^{239}Pu reactor contribution relative to fallout at that time.

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9. The constant in Eqs. 1 and 2 is slightly different from that used by Krey *et al.* (7), owing to the different half-lives used.
10. The deviation in the $^{240}Pu/^{239}Pu$ ratios from fallout values in the surface sediments of the McNary Reservoir core laid down after 1971 we attribute to the inclusion of some upriver sediment carried by bed-load transport between the Hanford Reservation and the reservoir and to sediment redistribution within the reservoir during freshets. We calculated the integrated plutonium inventory by summing the product of the mean $^{239, 240}Pu$ disintegrations per minute per gram (dry weight) and the mean bulk dry density (in grams per cubic centimeter) over each 2.5 cm of core length to yield the total activity per unit area.
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12. We believe that the fine particles imported by the Yakima and Walla Walla rivers to McNary Reservoir carry plutonium in which we can discern the ^{241}Pu contribution from the Chinese tests. Both rivers drain intensively managed agricultural lands, which would be expected to enhance erosion. By contrast, the Snake River, after it leaves the Boise Valley in Idaho, flows generally through desert lands and has several dam sites where fine sediment would be removed. Surface porosities confirm the different textural qualities of the sediments accumulating in the two reservoirs. For the McNary core porosities ranged from 0.885 to 0.815 between 0 and 6 cm, whereas the Ice Harbor core had porosities ranging from 0.785 to 0.681 over the same depth interval, an indication that finer particulate matter is accumulating at the McNary site. Moreover, the activity flux at both sites for the period from 1961 to 1977 for fallout plutonium is different: 1.02 dpm g^{-1} $year^{-1}$ for the McNary Reservoir and 0.73 dpm g^{-1} $year^{-1}$ at the Ice Harbor Reservoir. Thus, finer particles containing more plutonium activity are accumulating in McNary Reservoir. Finally, even though the tests of the People's Republic of China between 1967 and 1976 totaled only 16 megatons, the ^{241}Pu from such tests should be discernible owing to its high production in thermonuclear devices. The ^{241}Pu produced in the tests of 1961 to 1963 by the United States and the U.S.S.R. would have decayed by over one-half in the time between those dates and the time of our sample analyses (November 1980). By contrast, the amount of $^{239, 240}Pu$ added as fallout from the Chinese tests to the large reservoir of these atoms already existing in the environment would be negligible.
13. We thank P. W. Krey for helpful discussions and comments on the manuscript. Certain mass spectrometry measurements were carried out at the Knolls Atomic Power Laboratory by H. C. Hendrickson, C. F. Pachucki, and J. A. Leathan. This research was supported by the Office of Health and Environment, Department of Energy.

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Fossil Molluscan Larvae: A New Biostratigraphic Tool

Abstract. *Fossil molluscan larvae are less facies dependent and have a wider geographic range than their adult counterparts. They are also easily recovered from cores and small samples. With proper documentation, the study of fossil larvae can considerably enhance the biostratigraphic potential of macrofossils.*

A good biostratigraphic guide fossil should be geologically short-lived, geographically widespread, and present in a wide variety of sedimentary types. Because of their free-floating life habit, pelagic microfossils, such as foraminifera and coccolithophorids, generally meet these requirements and make good biostratigraphic indices. They are also small and easily recovered from cores and well cuttings. Most macrofossils, on the other hand, live in or on the substrate which restricts them to particular sedimentary facies. However, many macrofossils, such as molluscs, have planktic larval stages that last several weeks or even months. The larvae are in the same size range as planktic foraminifera (making them easily recoverable in small samples) and are often distributed over a considerably larger area than the adults (*1*, *2*). The larval shells of molluscs are often preserved in the fossil record (*3*–*6*), and their recognition and use may help overcome some of the drawbacks of the use of macrofossils in biostratigraphy.

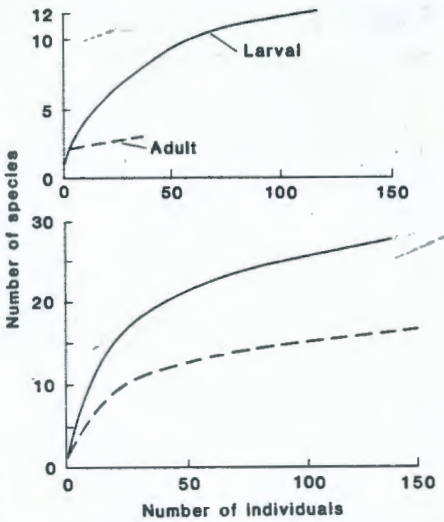

Fig. 1. Rarefaction curves for adult and larval molluscs from two samples in Eocene sediments of Texas. There are more larval than adult species for similar sample sizes.

Samples taken from Eocene sediments of the Gulf Coast, in environments ranging from open marine to brackish estuarine, were analyzed for adult and larval molluscan shells. In general, the diversities of larval shells are higher than those of adult specimens both in absolute numbers of species per volume of sediment (as might be expected because of the small size of larvae) and in numbers of species among similar numbers of individuals (Fig. 1). If larval shell distributions are considered, the known geographic range and facies independence of a species can be greatly increased. Also, because of the diversity among larvae, they provide more biostratigraphically useful species than do the adults.

There are several reasons why free,

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