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FROM THE CENTRAL OREGON COAST

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The radioisotopes plutonium-239,240 and americium-241 have been
measured in the mussel _Mytilus californianus_ from the region of
Coos Bay, OR. The flesh of this species has a plutonium concentration
of about 90 fCi/kg, and an Am-241/Pu-239,240 ratio that is high
relative to mixed fallout, ranging between two and three. Transur-
anic concentrations in sediment, unfiltered water, and filterable
particulates have also been measured; none of these materials has
an Am/Pu ratio as greatly elevated as the mussels, and there is no
apparent difference in the Am/Pu ratio of terrestrial runoff and
coastal water. Sediment core profiles do not allow accumulation
rates or depositional histories to be identified, but it does not
appear that material characterized by a high Am/Pu ratio has ever
been introduced to this estuary. Other bivalves (_Tresus capax_ and
Macoma nasuta) and a polychaete (_Abarenicola_ sp.) do not have an
elevated Am/Pu ratio, although the absolute activity of plutonium
in the infaunal bivalves is roughly four times that in the mussels.

The high Am/Pu ratio in M. californianus therefore appears to be a species-specific physiological response. Comparison of the plutonium concentration ratios for this species and other members of the genus suggests that the elevated ratio is the result of an atypically high content of americium in M. californianus.

Americium and Plutonium in Water,
Biota, and Sediment from the
Central Oregon Coast

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INTRODUCTION

The Mussel Watch program (Goldberg et al. 1978) was an ambitious
attempt to assess a variety of ocean pollutants over a wide geographic
area. The entire U.S. coastline was sampled, several species of
bivalves collected, and analyses performed for petroleum hydrocarbons,
halogenated hydrocarbons, heavy metals, and radionuclides. Bivalves
have a number of characteristics that make them particularly suitable
as indicator organisms, most important being their abundance,
accessibility, and high concentration factors for most pollutants.
However, their use must rest upon sound knowledge of the relationship
between pollutants in the environment and in the animal. This
relationship can vary with environmental conditions and between
different members of a class of pollutants (Phillips 1976, 1977).
A high concentration factor is not, itself, sufficient information
with which to interpret analytical results.

The results of Mussel Watch revealed striking differences in the
behavior of transuranic elements between east and west coasts.
Mussels from the Pacific had considerably higher activity ratios of
americium-241 to plutonium-239,240[1] than those from the Atlantic
or oysters from the Gulf of Mexico, and also generally higher
concentrations of both Am and Pu. The reason for these discrepan-
cies was not revealed by Mussel Watch data.

[1]Alpha spectrometry does not allow the two plutonium isotopes to
be distinguished.

Continual change is occurring in the Am-241/Pu-239,240 activity ratio due to the production of Am-241 by decay of Pu-241. Since the cessation of above-ground nuclear testing, very little additional Pu-241 can have been introduced to the global environment; calculation of the Am-241/Pu-239,240 ratio at any one time, therefore, is based on the amount of Pu-241 presumed to have been produced during weapons testing. Early tests (1952-58), amounting to only about 25% of the total megatonnage detonated (Carter and Moghissi 1977), apparently produced fallout with a Pu-241/Pu-239,240 ratio of about 27 (Diamond et al. 1960, Hisamatsu and Sakanoue 1978, Koide and Goldberg 1981), whereas in the later test series (1961-62) the same ratio was only about 13 (Anon. 1973). By making the assumption that Pu-239,240 was produced in proportion to megatonnage, the Am-241/ Pu-239,240 ratio in mixed fallout at any date can be calculated, as was done by Koide and Goldberg (1981). This method provides estimated values with which actual measurements can be compared. Some representative values are: 1970 - 0.22, 1976 (the date of Mussel Watch collections) - 0.31, and 1980 - 0.35. These numbers are almost certainly not exactly correct, due to inherent assumptions about the production, distribution, and mixing of radionuclides; nonetheless, they are probably the best estimates currently available. All environmental measurements must be considered in relation to this changing ratio.[2]

Mussel Watch collections from the west coast consisted principally

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[2]All isotope ratios presented here are expressed as relative activities.

of Mytilus californianus, with some M. edulis; from the east coast
only the latter species and the oyster Crassostrea virginica were
collected. Both species from the Atlantic had low Am/Pu ratios;
the averages were 0.24 ± 0.09 and 0.20 ± 0.09 for the mussel and
oyster, respectively. In the Pacific, however, the ratio was much
higher: 0.62 ± 0.20 in Mytilus edulis and a remarkable 2.4 ± 1.0
in M. californianus. The absolute amount of Pu-239,240 in M. calif-
ornianus was also twice as high, on the average, as in its congener.
East and west coast populations of M. edulis, however, showed no
significant difference in absolute activity.

There are two principal phenomena - not necessarily mutually
exclusive - by which the results of Mussel Watch may be explained.
The first is a physiological difference between bivalve species or
populations, the second is a greater availability of radionuclides
in the Pacific.

Goldberg et al. (1978) argue against a species-specific physio-
logical explanation on the basis of the different ratios in M. edulis
populations on east and west coasts, and of the habitat difference
between M. edulis and M. californianus. The former species character-
istically lives in the protected and brackish water of estuaries, the
latter upon the open coast. As a consequence of physical and chemi-
cal differences between their habitats, the amounts and forms of
radionuclides supplied to the two species may differ, thereby resulting
in different tissue concentrations.

Other marine species for which both Am and Pu have been measured
show no unusual elevation of the Am/Pu ratio. Table 1 summarizes all

Table 1. Plutonium and Americium in Selected Biota.

a. Bivalves

species	location	date	Pu-239,240 (fCi/kg, wet)	Pu CR[a]	$\dfrac{\text{Am-241}}{\text{Pu-239,240}}$	reference
Mytilus edulis	E. coast		180 ± 80	286		b
"	" "		270 ± 20	250-350		c
"	" "		150 ± 40		0.09 ± 0.01	d
"	" "	1976	95 ± 27		0.24 ± 0.09	e
"	W. coast	1976	72 ± 27		0.62 ± 0.20	e
"	Baltic Sea		0.6			f
californianus	W. coast	1964	96 ± 7	230		g
"	" "	1964	120 ± 12	290		g
"	" "	1976	180 ± 50		2.4 ± 1.0	e
gallo-provincialis	Med. Sea		0.1 - 0.2	150-500	0.4 - 0.9	h
Crassostrea virginica	E. coast	1976	77 ± 23		0.20 ± 0.09	e
"	Gulf coast	1976	77 ± 18		0.22 ± 0.10	e

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Table 1. continued

b. other marine species

species	location	date	$\dfrac{\text{Am-241}}{\text{Pu-239,240}}$	reference
Buccinum sp.	E. coast	1974		d
flesh			0.19 ± 0.05	
shell			0.29 ± 0.06	
Lunatia sp.	" "	1974	0.14 ± 0.04	d
Strongylocentrotus sp.	" "	1974		d
spines			0.15 ± 0.04	
Sargassum sp.	" "	1966	0.045 ± 0.003	i
Fucus vesiculosus	" "	1972	0.06 ± 0.02	i
Desmaidetta sp.	" "	1973	0.22 ± 0.04	i
Daisia sp.	" "	1973	0.22 ± 0.12	i
Chondrus crispus	" "	1973	0.18 ± 0.16	i

a CR - Concentration Ratio
b Wong 1971
c Noshkin et al. 1971
d Bowen et al. 1976
e Goldberg et al. 1978

f Miettinen et al. 1975
g Pillai et al. 1964
h Ballestra 1980
i Livingston and Bowen 1976

those determinations for which stratospheric fallout (rather than reactor effluent or close-in fallout, which may exist in different chemical forms) is presumed to be the source. It should be noted that few measurements exist from the Pacific coast. Covariance of plutonium and americium is the general rule, just as in the terrestrial environment. Species exposed to fuel-reprocessing waste, however, apparently can take up Am in preference to Pu. and partition the elements differently among tissues (Pentreath 1981). Such a response to fallout-derived transuranics has not yet been demonstrated, however, nor has it been shown to characterize bivalve molluscs. Among the bivalves, *Mytilus galloprovincialis* exhibits a slightly elevated Am/Pu ratio; rather than being a species-specific response, however, this is likely a consequence of the unique transuranic geochemistry of the Mediterranean.

Fractionation of Pu from Am occurs in the Mediterranean, apparently as a consequence of their different affinities for particulate matter. Only about 5% of the Pu but 15% of the Am in the water is attached to the solid phases (Holm *et al*. 1980, Ballestra 1980). This results in an Am/Pu ratio on particles that increases with depth in the water column (Ballestra 1980) and a ratio in bottom sediment that reaches 1.0 (Livingston *et al*. 1977). Sinking of particles thereby separates the elements. The reason for this pronounced fractionation has been suggested to be the large proportion of terrigenous debris in the Mediterranean, debris which may have a greater affinity for Am than does material of marine origin (Livingston *et al*. 1977, Holm *et al*. 1980).

The same process seems to be taking place, to at least a limited extent, in the Atlantic Ocean. Water column profiles show Am/Pu ratios of 0.1-0.2 in surface water, which increase to 0.2-0.3 at 3000 meters (Livingston and Bowen 1976). Although bottom sediment in the Atlantic does not show a general elevation of the Am/Pu ratio, it does so in the western sub-tropical region, an area subject to the influence of terrigenous debris from the Amazon River; the ratio there varies from 0.44 to 1.2 (Livingston and Bowen 1976). This is not only consistent with the mechanism suggested to operate in the Mediterranean, but also is evidence that significant local variations can exist.

In the Pacific, deep-water profiles do not show either changes in the Am/Pu ratio over a 6000 meter water column or an elevated ratio in bottom sediment, the value being about 0.3 in both cases (Livingston and Bowen 1976). Recent measurements of filterable material, however (Koide et al. 1981), suggest that in localized areas the Am/Pu ratio can be considerably elevated.

The Pacific may be unique in quite another way, however. Bowen et al. (1980) record significantly higher total amounts of plutonium in Pacific water than is typical of other oceans. In equatorial and sub-equatorial regions, Pu-239,240 inventories are 10-20 times higher than is to be expected from direct fallout alone; in the North Pacific, inventories are elevated by a factor of about two. Water-column profiles revealed an unusually high plutonium concentration at about 450 meters, and Bowen et al. (1980) suggest that there may be a thin layer at this depth throughout the central North Pacific in which plutonium concentrations range from 3.5 to 4.0 fCi/kg.

Only the measurements of Pillai et al. (1964) showed a similar
concentration in the central Pacific, but this was immediately follow-
ing the most intensive testing period. U.S. coastal water at the same
date - far from the area of testing - had only 0.42 fCi/kg (Pillai
et al. 1964).

Representative measurements of Pu in ocean water are shown in
Table 2. In the Atlantic, surface water has concentrations of
0.3 - 1.0 fCi/kg, and although a high value of 3.5 fCi/kg has been
measured in one subsurface sample, subsurface concentrations are
generally much lower (Bowen et al. 1971). Water-column inventories
in the Atlantic are also not elevated with respect to fallout deliv-
ery (Noshkin and Bowen 1973).

Comprehensive data on americium concentrations in ocean water do
not exist for either the Atlantic or Pacific, but the higher plutonium
activities observed in the Pacific suggest that this ocean may also
have an unusual americium content.

The measurements of Bowen et al. (1980) and Goldberg et al.
(1978) provide circumstantial evidence that west-coast bivalves may be
exposed to water that has not only high Pu and Am concentrations, but
a high Am/Pu ratio as well. If Mytilus californianus is faithfully
reflecting environmental conditions, then the fractionation of these
elements in the Pacific may also be considerable. Given the possibil-
ity of local variations, however, and the caveats that must be applied
to the use of indicator species, this conclusion is not strongly sup-
ported. The influence of habitat (i.e. the source of transuranics),
although suggested to be important by Goldberg et al. (1978),

Table 2. Plutonium and Americium in Ocean Water[a]

location	date	Pu-239,240 (fCi/l)	$\frac{\text{Am-241}}{\text{Pu-239,240}}$	reference
North Pacific				
California coast	1964	0.40 ± 0.04		Pillai et al. 1964
30°N, 140°W	1964	3.0 ± 0.2		" " " "
44°N, 151°E	1968-73	0.43		Miyake and Sugimura 1976
30°N, 151°E	" "	1.13		" " " "
45°N, 175°W	1973-74	0.2 - 0.4		Bowen et al. 1980
500 m		0.9 - 1.8		
25°N, 170°W	" "	0.2 - 0.4		" " " "
500 m		3.4 - 4.1		
31°N, 158°W				Livingston and Bowen 1976
all depths			0.3	
North Atlantic				
Massachusetts coast		0.18 - 1.2		Bowen 1975
N.Y. Bight		0.25 - 1.2		Simpson et al. 1980
36°N, 70°W	1968	0.7 ± 0.1		Wong 1971
53°N, 37°W	1972			Livingston and Bowen 1976
0-1000 m		0.77 - 1.4	0.22	
>1000 m		0.63	0.31	
Mediterranean				
42°32'N, 07°32'E	1976	1	0.06 - 0.11	Fukai et al. 1979
2000 m		0.4	0.23	

a All values for surface water unless otherwise indicated.

is also not clear.

Data are presented here which bear directly upon the relationship
between americium and plutonium in water, suspended material,
sediment, and biota. These address the relative importance of
marine and terrestrial material as sources of transuranics to Mytilus
californianus, including the effects of seasonal variations in
transuranic availability, Am and Pu fractionation upon particles,
and species-specific differences in affinity for these radionuclides.

<u>METHODS</u>

<u>Overview</u>

All samples for this investigation were taken in the region of Coos Bay, OR. In the survey of Goldberg et al. (1978) mussels from this location had high concentrations of both plutonium and americium and one of the highest recorded Am/Pu ratios. If these effects are a reflection of the physical character of the environment, then presumably the conditions giving rise to them would be most easily identified here. The Coos River provides terrestrial material to this region, and also this is approximately the latitude at which the North Pacific Current approaches the coast; anomalous isotope concentrations or ratios characteristic of the central North Pacific may therefore be most identifiable at this location.

Four types of samples were taken for analysis. Biological samples consisted principally of <u>Mytilus californianus</u>, which were analyzed to confirm the findings of Mussel Watch and to establish the existence and range of seasonal variations and the relationship between radionuclide concentrations in the mussels and in possible sources. Other biota were also sampled to determine whether elevated Am/Pu ratios are common to local biota, and whether feeding strategy influences the ratio. Three species could be collected in amounts sufficient for analysis. These were <u>Tresus capax</u>, a large filter-feeding clam living in sandy bottoms, <u>Macoma nasuta</u>, a small deposit-feeding clam which ingests the top millimeter of sediment, and <u>Abarenicola</u> sp., a burrowing polychaete which feeds by ingesting

sediment. The butter clam, Saxidomus giganteus, was also collected on one occasion.

Sediment cores were also raised from the estuary. These were intended principally to provide a means of measuring the Am/Pu ratio on particulate matter, the material on the bottom being presumed to resemble that in the water column. By taking cores, it was hoped to capture the entire history of transuranic deposition and determine whether any material of high Am/Pu ratio had, at any time, been introduced to the estuary, and particularly, whether high ratios are characteristic of terrestrial debris. A measurement of the complete inventory of Pu-239,240 in these cores would also allow an assessment of whether total deposition has exceeded the average for fallout at this latitude.

Water was collected from the Coos River and offshore in order to judge the relative amounts of radionuclides entering the estuary from terrestrial and marine sources, and the Am/Pu ratio characteristic of each source.

Suspended particulate matter was collected by filtration, from both the Coos River and the offshore region. Filtered material was intended to provide a more direct measure of the nature of available particulate matter than is provided by the cores.

Sampling

Mytilus californianus was taken from two locations (Fig. 1). The principal site was a rocky shelf at Coos Head, inside the southern breakwater and just at the mouth of the estuary. The other site was at Cape Arago, again on a rocky shelf, but in this
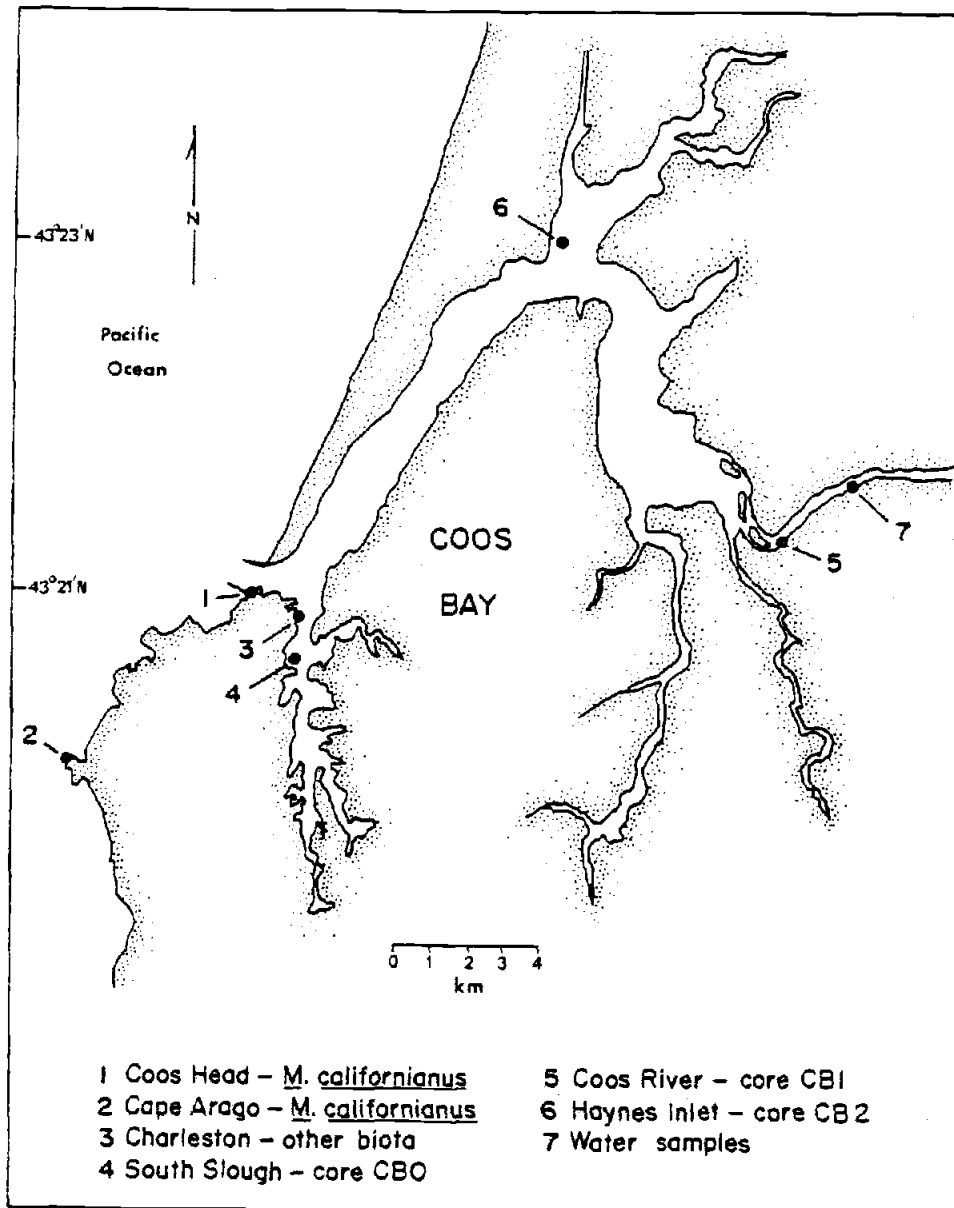
Figure 1. Sampling locations for biota, sediment, and Coos River water. Coastal water was collected in a variety of locations, as described in the text.

case exposed directly to the ocean with no adjacent sources of terrestrial material. Individuals from Cape Arago were generally smaller than those from Coos Head, their respective shell lengths being, typically, 4-5 cm and 6-8 cm. Individuals from Coos Head averaged 47 g wet weight, of which 66% was shell. The size difference is presumed to reflect exposure of the location rather than the age of the population. There is no reliable way to determine the age of individuals of M. californianus, and the effect of this variable on radionuclide burden has not been quantified. Different size classes of M. edulis have, however, been demonstrated to have similar transuranic activities (Bowen et al. 1976), and different sizes of M. galloprovincialis take up plutonium at the same rate (Fowler et al. 1975).

Collections of Mytilus generally consisted of a five-gallon bucket of live individuals, a total of 200-250 animals. Samples from Cape Arago were only two to three gallons, as the population was smaller.

Collections were made from Sept. 1979 through Nov. 1980, on roughly a quarterly schedule, with more frequent sampling during the summer of 1980 in an attempt to identify changes associated with upwelling. The dates of sampling were September 1979, January, March, June, August, September, and November 1980; Cape Arago was sampled only in March, June, and August. A large sample was taken from Coos Head in June, and half of the animals were dissected before analysis. The purpose of the dissection was to separate those tissues most likely to encounter labelled environmental

materials - and thus be involved in assimilation of transuranics - from those presumed to acquire transuranics only through metabolic pathways. As all the tissues are bathed in seawater this distinction is not easily made; separation of the tissues was performed principally on the basis of their exposure to particulate matter. Gill and gut fell in the former category, these being most involved in the handling of particles. The gill could be completely isolated, but the gut was retained with the entire visceral mass. Mantle and gonad, together, were taken to be representative of tissues most likely to receive transuranics metabolically. More exacting tissue separations were not performed because the resultant sample sizes would have been inconveniently small. For example, five gallons of mussels, live, yielded only 35 g, dry weight, of gill.

Other biota (Tresus, Macoma, Abarenicola, and Saxidomus) were collected from the tidal flats adjacent to the Charleston Boat Basin, about 2 km from the mouth of the estuary. Macoma was also taken from the vicinity of Empire, OR, about 6.4 km upriver. Tresus was collected on four occasions: October 1979 and January, April, and June of 1980, the samples being about 15-20 individuals. Half of the April sample was dissected, the fractions being neck, gill, mantle+gonad, and gut+viscera. The other three species were not collected seasonally, principally because of the difficulty of obtaining a sufficient quantity. Abarenicola sp. was sampled in October 1979 and January 1980. Macoma nasuta in January and April 1980, and Saxidomus giganteus in October 1979.

All biological samples were homogenized and stored in dry form

prior to analysis. Homogenization was performed to reduce the effect of individual variability among mussels. Bivalve shells were cleaned of epifauna and loose material before shucking, and byssal threads were removed from the mussels. Both Macoma and Abarenicola were depurated to remove sediment from the gut. Wet tissue was pooled, pureed in an electric blender, and then dried to constant weight at 60°C. This material was homogenized once again, and stored in airtight polyethylene jars.

Sediment cores were all taken on the first sampling date, in September 1979, at the locations shown in Fig. 1. The first of these (CB0) was taken in South Slough in a shallow, quiet area unfrequented by boat traffic. This location is quite close to the mouth of the estuary and the sampling locations for Mytilus and other biota. The second core (CB1) was taken from the Coos River, about 27 km upstream from the mouth. Log rafts and tugs make heavy use of the river, and although the core was taken from shallow water near shore, it contained a large amount of wood debris. The third core (CB2) was taken from Haynes Inlet; this area is about 14.5 km upriver, and also is quiet and shallow with little boat traffic.

Coring was performed with a 15.3 cm diameter gravity corer without a core liner or core catcher. The bottom of the core was capped immediately upon breaking surface. After being brought on board, the overlying water was siphoned off and the core extruded and sectioned immediately. Inspection showed no apparent disturbance of the sediment surface in any of the cores retained. Section

thickness was one centimeter, except in the upper region of the cores where the sediment was less cohesive and two or three cm sections were taken. The outer centimeter of each section was trimmed to eliminate the effect of translocation of radionuclides along the core barrel. The sections were then frozen until they could be processed.

Prior to analysis, all sections were thawed, air-dried, homogenized, and stored in airtight polyethylene jars.

Water and suspended material were collected simultaneously. The dates of collection were September 1979 and March, May, June, August, and November 1980; at the last date no particulate material could be collected offshore. From May 1980 onward, duplicate samples of each material were collected at each location. Coos River samples were taken about 30 km above the river mouth, and ocean samples from two to five km offshore, always out of the river plume. Water was collected in 60-liter acid-washed carboys and analyzed immediately upon return to the laboratory. Suspended material was collected on 0.3 μm Millipore filters in a large volume filtration apparatus (Silker et al. 1971) providing a total filtering area of about 5900 cm^2. A thousand liters were generally filtered, except in those cases when the flow rate became substantially slowed by clogging. All filtered material was frozen until the time of analysis.

Although decreasing retention of plutonium with increasing volume filtered has been observed in some circumstances (Wong et al. 1980), the apparatus used here has been demonstrated to provide a constant

filtering efficiency for volumes much greater than 1000 liters
(Holm et al. 1980).

Analysis

Isolation of transuranic elements was accomplished by solvent
extraction and precipitation techniques. Recovery is not constant
with these methods, so known quantities of yield tracers were added to
every sample before analysis. The tracers used were Pu-242 and Am-243;
about two disintegrations-per-minute of each was added. This quantity
resulted in relatively little contribution to the total error of a
measurement from the counting error of the yield determinant, yet
provided good spectral separation of the isotopes. A small part
(0.2%) of the activity of the Am-243 tracer was due to Am-241, an
effect that was accounted for.

An important consideration pertaining to the use of tracers is
that they be thoroughly equilibratied with the nuclide of interest,
so that subsequent chemical separations will return comparable propor-
tions of each. Strenuous efforts were applied to ensure that such
equilibration was attained. These centered about the total dissolu-
tion of the sample, so as to render all transuranics soluble.
Valence adjustments were performed throughout the analysis to main-
tain all plutonium isotopes in the same form. The preparation of
samples for analysis varied according to their nature, as follows.

Fifty grams of dry, homogenized biological material was ordinar-
ily prepared for analysis. Yield determinants were added to the dry
material, and the sample ashed at 500°C. Higher temperatures were

not used so as to avoid the formation of refractory oxides.
The resulting ash was then treated with nitric and perchloric acids
to oxidize any residual carbonaceous material. Six identical samples
were prepared, three of which were ashed by this procedure, and
three of which were completely wet-ashed with nitric and perchloric
acids alone. All samples yielded identical estimates of Pu and Am
activity; isotopes of neither element are rendered insoluble by the
dry-ashing, and so this technique was used exclusively owing to its
greater convenience. After ashing, the solution was diluted and all
plutonium reduced to (+3,4) with hydroxylamine hydrochloride, followed
by coprecipitation of the transuranics with calcium oxalate to
remove the majority of the iron in the samples. Re-solution of the
transuranics was then accomplished by destruction of the oxalate
with nitric and perchloric acids. An unidentified amorphous white
precipitate - possibly calcium phosphate - remained after this step.
It was eliminated - after again reducing the plutonium - by transpo-
sition to a calcium fluoride precipitate. The fluoride was then
dissolved with nitric and boric acids, resulting in complete solubil-
ization of the sample.

Sediment analyses were performed on ten-gram aliquots. Yield
determinants were added and the minerals decomposed with $8\underline{N}$ HNO_3
and concentrated HF. Further dissolution of the residue was
accomplished with $6\underline{N}$ HCl. Remaining insoluble material was separa-
ted by filtration and further digested with nitric and perchloric
acids to solubilize any calcium or barium sulfate or residual
carbonaceous material. The resulting solution was filtered, the

filtrate combined with that from the previous step, and all the plutonium reduced to (+3,4) before analysis.

Water samples were acidified with 500 ml of concentrated HNO_3 and the yield determinants added. The acidified solution was then stirred vigorously for a week to ensure equilibration of the isotopes. Stable iron carrier (30 mg) was added and the transuranics coprecipitated with $Mg(OH)_2$ by addition of NaOH. From this solution, after warming, $Fe(OH)_3$ was precipitated with NH_4OH. This precipitate was collected and then dissolved and digested with nitric and perchloric acids. Plutonium valences were then adjusted to (+3,4) before analysis.

Filters carrying the particulate material were first thawed, then dissolved in 200 ml concentrated HNO_3. Yield determinants were added and the volume reduced by evaporation until the digestion was completed with $HClO_4$. Any residue that remained was filtered from the solution and washed with hot 6N HCl. This wash was combined with the filtrate prior to analysis.

Plutonium and americium were separated from trace metals and all naturally occurring alpha emitters by a series of precipitation and solvent extraction steps. After completion of the solvent extraction, the transuranics were electrodeposited on stainless steel plates and their activities measured by alpha spectrometry. Electroplating was carried out at 1.2 amperes from an ammonium-sulfate solution, following the method of Talvitie (1972). Alpha spectrometry was performed using a silicon surface-barrier detector and multichannel analyzer calibrated at 15.6 keV/channel. Counting times were gener-

ally seven to ten days. Although the chemical procedure provided excellent decontamination from thorium, the presence of Th-228 could be inferred from the activity of its decay products Ra-224, Rn-220, and Po-216. After electroplating, the samples were allowed to sit for two weeks so that Th-228 would come into secular equilibrium with these daughters, and if present was thus accounted for. Background corrections were also made for each detector.

The accuracy of the analytical procedures performed here is confirmed by comparison of the results of analyses of a reference sediment (SLOSH III). This comparison is presented in Table 3. Overall, there are no significant differences between these sets of measurements.

Table 3. Pu-239,240 Activities Measured in Reference Sediment SLOSH III (Standard Lamont Observatory Sediment from the Hudson, supplied by Lamont-Doherty Geological Observatory).

lab	Pu-239,240 (dpm/kg)
Lamont-Doherty Geological Observatory	58.8 ± 1.1 (3)[a]
Woods Hole Oceanographic Institute	55.3 ± 3.8 (2)
Oregon State University[b]	59.2 ± 2.5 (16)
University of Washington	54.6 ± 5.1 (2)
Present	60.8 ± 1.1 (4)

a Number of replicates in parentheses. Values for the first four laboratories are taken from Carpenter and Beasley 1981.
b Previous measurements made at this laboratory.

No such suite of intercalibration data is available for Am-241, but four previous measurements made at this lab (OSU) on SLOSH III are in good accord with analyses performed for this investigation. The means are, respectively, 13.8 ± 1.0 dpm/kg (Carpenter and Beasley 1981) and 12.5 ± 0.9 dpm/kg.

These comparisons and the results of other intercalibration exercises which included biological material (Volchok and Feiner 1979) indicate that the techniques used here for transuranic analyses are both accurate and precise, and the yield determinants used to quantify Am-241 and Pu-239,240 are accurately calibrated.

Errors are expressed as one standard deviation, and are derived principally from counting error. Propagation of errors has been carried out according to standard methods (Wang et al. 1975). Empirical analysis of ratio variables (Atchley et al. 1976) shows that the ratio of two normally distributed variables can, under certain conditions, have a highly non-normal distribution. This effect occurs when the ratio of the coefficients of variation of the two variables is less than 0.7. For radioisotope measurements made over equivalent counting times, where the standard deviation is proportional to the mean, this takes place when the ratio itself is about 2.0. The implications for the present work are that calculated activity ratios of two or greater may be distributed non-normally. Although counting times were generally adjusted so that an equivalent precision was obtained for all measurements, some of the data presented here seem to exhibit this effect.

Where replicate analyses of plutonium and americium have been

made, a weighted average (Jaffey 1960) has been taken. In such cases it must be kept in mind that the error term does not directly represent the dispersion among replicates, but rather is an estimate of the precision of the measurement. All experimental data are presented in the appendices.

Throughout the following text the term 'concentration ratio' (CR) is used to denote the observed degree of enrichment of an element in an animal relative to the water, and is distinguished from the 'concentration factor', which is appropriate only when the animal is at equilibrium. With environmental measurements such as these, equilibrium cannot be assumed.

RESULTS

Sediments

Sediment profiles of Pu-239,240 and Am-241/Pu-239,240 are shown
in Fig. 2. The first two cores clearly did not penetrate to the
greatest depth of radionuclide deposition. In the third (CB2), there
is a sharp decrease in the Pu profile from 29 to 34 cm; this, and the
low final value, suggest that all the plutonium in the sediment
column has been collected.

Plutonium inventories for these cores have been calculated by
multiplying the specific activity of each section by the weight of
that section, summing over the length of the core, and dividing by
the trimmed area of the core. (For core CB1, the activity of unana-
lyzed sections was estimated by linear interpolation.) Minimum esti-
mated inventories for cores CB0 and CB1 are 0.92 and 2.3 mCi/km^2,
respectively. Core CB2, in which the entire record of deposition
was captured, has an inventory of 0.68 ± 0.14 mCi/km^2. These
differences in inventory parallel the differences in absolute activ-
ity (Fig. 2); both appear to be explicable in terms of the sediment
grain size. Porosity, the proportion of sediment volume occupied by
water, provides a rough measure of grain size; finer sediment has a
higher water content. The porosities for cores CB0, CB1, and
CB2 average 0.40, 0.65, and 0.40, respectively. The upriver core,
which has the most fine-grained material, also has the highest
specific activity and the greatest inventory. This is in accord with
observations of Hetherington et al. (1975) which show Pu uptake to
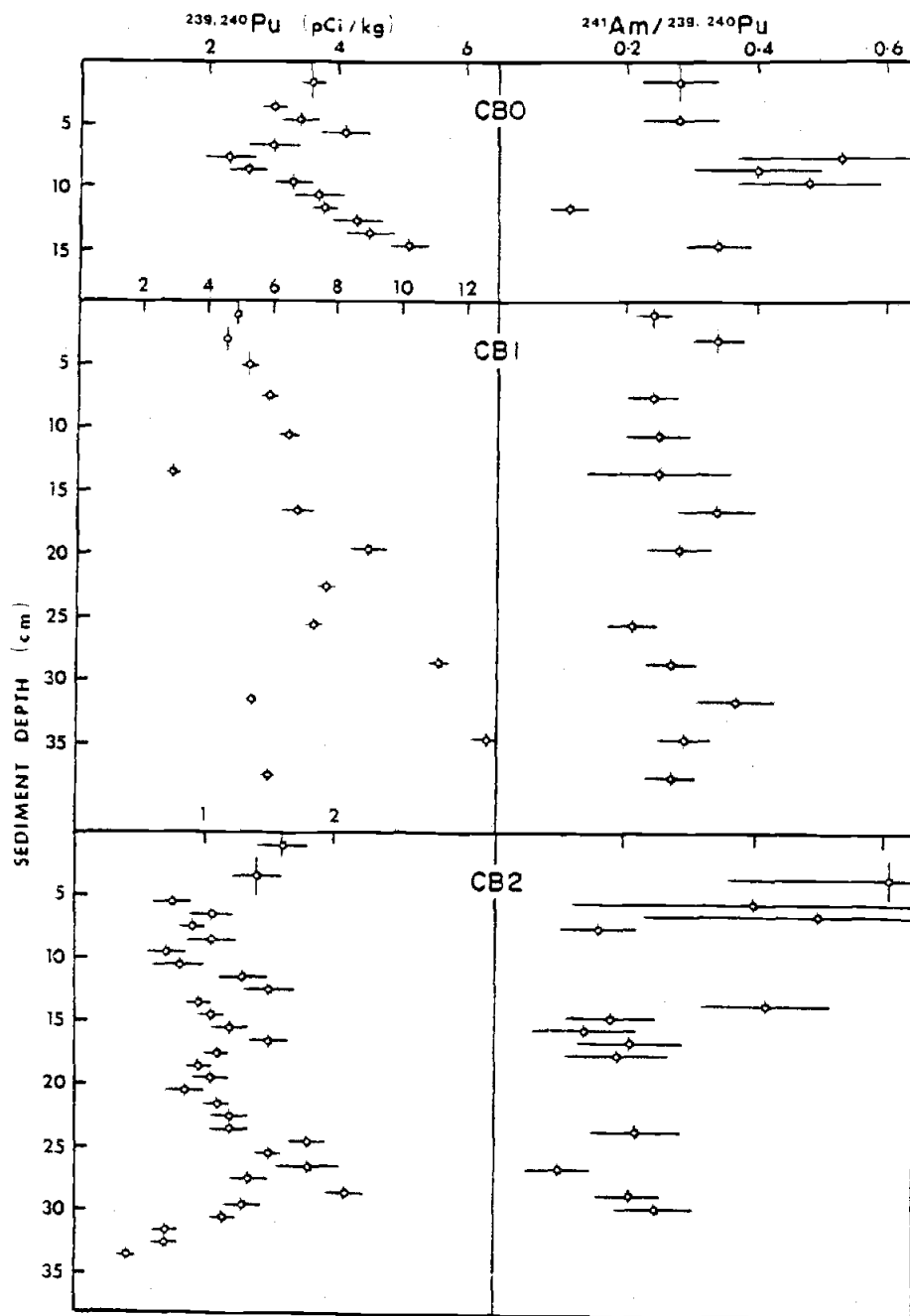
Figure 2. Sediment core profiles. Vertical bars: width of section; horizontal bars: one standard deviation, from counting error.

be related to surface area. It is surprising, perhaps, to find more fine material accumulating upriver than in embayments that are more isolated from the channel, but this may attest to the ability of tidal flushing to winnow and remove fine sediment from the lower estuary.

Attempts to establish sedimentation rates for these cores on the basis of sedimentary geochronologies (Goldberg and Bruland 1974) have all been unsuccessful. Plutonium profiles do not exhibit peaks that are clearly related to the periods of maximum weapons testing in the late fifties and early sixties. Plutonium-238/Pu-239,240 ratios show no change with depth corresponding to the SNAP- 9A burnup, which released 17 kCi of Pu-238 to the atmosphere in 1964 (Hardy et al. 1973). Within measurement error the ratio is invariant, having an average value of about 0.04 at all depths in all cores; this is similar to the ratio to be expected at this latitude following the deposition of SNAP-9A plutonium (Hardy et al. 1973).

Lead-210 and cesium-137 analyses were performed on cores CB0 and CB1, respectively (Table 4). The first of these had an erratic profile, and there was no decrease with depth. The cesium profile, like that of plutonium, showed no peaks that could be identified with periods of testing. Cesium-137 was not measureable in core CB2.

All of these methods - as well as the inventory data - indicate that some disturbance of the sediment column has taken place at each location. This may have been by mixing or by a variable rate of deposition. Only for core CB2 can a sedimentation rate be calculated, but little confidence can be placed in the result. If the

deepest plutonium in the core (at 34 cm) represents debris from 1952 tests, the sedimentation rate has been 1.3 cm/yr. However, this calculation supposes constant deposition and no mixing, a supposition that is unsupported by the profile.

Table 4. Lead-210 and Cesium-137 Profiles.

core CB0		core CB1	
depth (cm)	Pb-210 (pCi/kg)	depth (cm)	Cs-137 (pCi/kg)
0-3	220 ± 10	0-2	130 ± 30
4-5	260 ± 10	2-4	120 ± 25
7-8	210 ± 10	4-6	150 ± 30
10-11	400 ± 20	7-8	170 ± 30
13-14	350 ± 20	10-11	125 ± 35
14-15	380 ± 20	13-14	85 ± 20
		16-17	210 ± 30
		19-20	180 ± 30
		22-23	190 ± 45
		26-27	170 ± 35
		27-28	145 ± 30
		28-29	200 ± 30
		29-30	190 ± 20
		32-33	120 ± 20
		35-36	245 ± 30
		38-39	235 ± 25

Although sedimentation rates are unknown, there have apparently been no changes throughout time in the Am/Pu ratio of deposited material. In all the cores the Am-241/Pu-239,240 ratio (Fig. 2) is close to the expected value of 0.34 for mixed fallout in 1979 (calculated as per Koide and Goldberg 1981). In core CB2 there is an apparent increase in the ratio from depth to surface, although

the error terms of higher values render this an uncertain conclusion. Deeper sections have values slightly lower than is to be expected in mixed fallout, upper sections slightly higher values. This may be a reflection of temporal changes in the nature of deposited material that have not been obliterated by mixing, but as the phenomenon is not entirely clear from these data, neither is the interpretation. None of these cores, however, provide evidence of highly americium-enriched material in the estuary.

That material in the upriver core (CB2) is of terrestrial character is shown by its Pu-239,240/Cs-137 ratio. This ratio has a value of 0.04, and is constant with depth. The ratio delivered in rain at this latitude is about 0.02 (Thien et al. 1980), and most of the cesium, like plutonium, is bound to particles in fresh water (Simpson et al. 1976). Typical Pu/Cs ratios in marine sediment are much higher, around 0.3 (Noshkin and Bowen 1973, Beasley et al. 1982). Thus, fluvial material entering the estuary has an Am/Pu ratio no different from that in more marine regions.

Americium-241/Pu-239,240 ratios identical to those in Coos Bay are found in sediments in the Pacific. In deep central water (5600m) the ratio was about 0.30 ± 0.05 in 1973 (Livingston and Bowen 1976); closer to the continent, off Cape Mendocino (4000m) it ranged 0.30-0.38 in 1981 (Beasley, unpublished data); on the continental shelf off Washington it is 0.28-0.33 (Beasley et al. 1982); and in sediment from Saanich Inlet (200m) the ratio is constant with depth, averaging 0.30-0.35 in 1979 (Carpenter and Beasley 1981). Relative enrichment of americium apparently does not occur in marine sedi-

ments of the northeast Pacific; resuspension of sediments in the coastal region, whether they are of marine or terrestrial origin, will not suffice to explain elevated Am/Pu ratios in biota.

Water

The concentrations of transuranics in whole water samples and on particulate matter are shown in Tables 5 and 6.

The yearly range of Pu activity in unfiltered offshore water is 0.12 - 0.53 fCi/l; this is essentially identical to the values found by Pillai et al. (1964) in California coastal water. Similar values also characterize Atlantic water (Table 2), although these have a larger range. The high values recorded by Bowen et al. (1980) in the central North Pacific are not found in Oregon coastal water. This indicates that either the high-activity water of the submerged maximum is not carried onto the continental shelf, or that removal processes are sufficiently active in coastal water to maintain low plutonium concentrations. Two mechanisms - biological productivity and scavanging by resuspended sediment - may increase plutonium removal in coastal water; simultaneous determination of changes in plutonium concentrations and hydrographic parameters across the gyre boundary and at the shelf break might reveal whether there is any penetration of high-activity water onto the shelf.

Plutonium concentrations in Coos River water (Table 5) are slightly lower than those offshore. They are within the range of particle-free Hudson River water and of unfiltered Columbia River water, for which average values are 0.3 ± 0.2 and 0.29 ± 0.15 fCi/kg, respectively (Simpson et al. 1980, Beasley et al. 1981). The lower

Table 5. Plutonium and Americium in Unfiltered Water.
All values in fCi/kg.

Coos River

date	Pu-239,240	Am-241	Am/Pu
9/79	0.116 ± 0.008	0.11 ± 0.04	0.95 ± 0.35
3/80	0.23 ± 0.03	0.078 ± 0.018	0.34 ± 0.09
5/80	0.17 ± 0.03	0.047 ± 0.13	0.28 ± 0.77
6/80	0.23 ± 0.02[a]	0.046 ± 0.013	0.20 ± 0.06
8/80	0.084 ± 0.013	-	-
11/80	0.24 ± 0.02	0.097 ± 0.058	0.40 ± 0.24
			0.26 ± 0.05[b]

Offshore

date	Pu-239,240	Am-241	Am/Pu
9/79	0.40 ± 0.04	0.030 ± 0.016	0.08 ± 0.04
3/80	0.53 ± 0.05	-	-
5/80	0.19 ± 0.02[a]	0.029 ± 0.025	0.15 ± 0.13
6/80	0.12 ± 0.02[a]	0.04 ± 0.02[a]	0.33 ± 0.18
8/80	0.49 ± 0.03[a]	0.088 ± 0.030	0.18 ± 0.06
11/80	0.28 ± 0.06[a]	0.074 ± 0.046	0.26 ± 0.17
			0.12 ± 0.03[b]

a Weighted average of duplicate measurements.
b Weighted average of ratios at all dates.

Table 6. Plutonium and Americium on Filterable Particulates.
All values in fCi x 10^{-2} per kg of water filtered.

Coos River

date	Pu-239,240	Am-241	Am/Pu
9/79	3.8 ± 0.2	1.2 ± 0.3	0.32 ± 0.08
3/80	9.3 ± 0.9	-	-
5/80	5.4 ± 0.3[a]	1.5 ± 0.4[a]	0.28 ± 0.08
6/80	10 ± 1	-	-
8/80	1.7 ± 0.2	0.3 ± 0.4	0.18 ± 0.24
11/80	6.0 ± 0.5	3.0 ± 0.7	0.50 ± 0.12
			0.33 ± 0.05[b]

Offshore

date	Pu-239,240	Am-241	Am/Pu
9/79	4.8 ± 2.7	1.2 ± 0.3	0.25 ± 0.15
3/80	1.1 ± 0.2	1.4 ± 1.0	1.3 ± 0.9
5/80	2.3 ± 0.4	0.78 ± 0.40	0.34 ± 0.18
6/80	0.78 ± 0.16	0.42 ± 0.17	0.54 ± 0.24
8/80	3.2 ± 0.4	‹2	‹0.63
			0.35 ± 0.10[b]

a Weighted average of duplicate measurements.
b Weighted average of ratios at all dates.

average concentration in the Coos River is reflected in lower sediment activity relative to the Hudson (20 pCi/kg) and Columbia (10 pCi/kg) (Simpson et al. 1976, Beasley et al. 1981).

Neither Coos River water nor offshore water have Am-241/Pu-239,240 ratios that are elevated with respect to mixed fallout. The Coos River is comparable to the Columbia, which had an average yearly ratio of 0.24 ± 0.06 in 1978 (Beasley et al. 1981). Offshore, the ratio is generally lower; this is the same effect seen in the surface water of the Mediterranean (Fukai et al. 1979), where americium seems to be preferentially associated with particles and removed from the water column.

The Am-241/Pu-239,240 ratios on particles upriver and offshore (Table 6) are not substantially elevated. Particularly in offshore water, the ratios are higher than those of unfiltered water, but are not greater than is to be expected in mixed fallout. Indeed, weighted averages of the ratios on particles are exactly as predicted by the calculation of Koide and Goldberg (1981).

There is, however - at least in offshore water - a greater proportion of Am than Pu attached to particles (Table 7). In the Mediterranean, where the same phenomenon occurs, the proportions of particle-bound transuranics are quite similar to those off Coos Bay: 2-9% of Pu and 8-15% of Am (Holm et al. 1980). Whether the same phenomenon occurs in the Coos River is difficult to judge, owing to the few and unreliable values; these data suggest, however, that the effect may not be so pronounced as in seawater.

Table 7. Percent of Plutonium and Americium Attached to Particles.

| date | Coos River | | Offshore | |
	Pu	Am	Pu	Am
9/79	33 ± 3	11 ± 5	13 ± 8	40 ± 20
3/80	40 ± 7	-	2 ± 0.5	-
5/80	32 ± 6	(33 ± 93)	12 ± 3	28 ± 28
6/80	43 ± 6	-	7 ± 2	10 ± 7
8/80	20 ± 4	-	7 ± 1	-
11/80	25 ± 3	31 ± 20	-	-
weighted average	29 ± 2	12 ± 5	3.4 ± 0.4	14 ± 6

Suspended particulate material, both terrestrial and oceanic, has an Am/Pu ratio little different from settled particulate material, as represented by the cores. Similarly, the weight-specific activity of plutonium on suspended particles is reasonable in light of that of the core material. Although the weight (i.e. concentration) of filterable material was not measured directly, it can be estimated by using the expression of Duursma and Bosch (1970):

$$Y/\rho = (\frac{100}{P} - 1) \times \frac{10^6}{K}$$

where
       Y = sediment concentration (mg/l)
       ρ = specific weight of dry sediment
       P = percentage of radionuclide in dissolved form
and

$$K = \frac{activity/volume \ of \ sediment}{activity/volume \ of \ water} \ .$$

This is equivalent to

$$Y = (\frac{100}{P} - 1) \times \frac{10^6}{K_d}$$

where K_d, the distribution coefficient, is expressed in the more usual units of

$$\frac{activity/mass \ of \ sediment}{activity/mass \ of \ water} \ .$$

Using a K_d for Pu of 10^5 (Eyman and Trabalka 1980) and the data of Table 7 (P = 70% in the Coos River, 87-98% offshore), the concentrations of suspended material are calculated to be about 4.3 mg/l in the Coos River and 0.2-1.5 mg/l offshore. Transforming these to weight-specific Pu activities using the data in Table 6 yields values of about 14 pCi/kg for the Coos River and 17-125 pCi/kg offshore. Although the range of the offshore values is high, neither of these estimates is unreasonable. Plutonium activity at the surface of the upriver core (closest to the water sampling site) is 5-6 pCi/kg (Fig. 2); considering the correspondence between activity and surface area (Hetherington et al. 1975), suspended material must have a diameter only one-half to one-third of sediment at the upriver core in order to carry the calculated label. In other rivers (Simpson et al. 1976, Beasley et al. 1981) and in

basins on the continental shelf (Carpenter and Beasley 1981)
settled material has similar activities.

Altogether, neither the Coos River nor offshore water show
anomalously high Am/Pu ratios or Pu activities.

Biota

Transuranic activity in M. californianus and other biota (Tables
8, 9, and 10) is reported here in relation to the total weight
(wet weight) of the tissue. Wet weight/dry weight ratios were
variable, averaging 6.2 ± 1.3 in M. californianus and ranging as
high as 20 in other species. Because there is no information availa-
ble on the extent to which transuranics are partitioned among fluid
and solid components of tissue, expression of activities on a
dry-weight basis has been avoided as it neglects the possible role of
tissue fluid in determining transuranic burden. The interpretation
of results presented here is generally unaffected if, in fact,
activity is expressed in terms of dry weight. In those cases where
a difference would result, this is noted. Americium/plutonium
ratios are, of course, unchanged however activities are expressed.

Mytilus californianus has a year-round Pu-239,240 concentration
of about 90 fCi/kg (Table 8). This is essentially identical to the
values found for the same species by Pillai et al. (1964; Table 1).
Goldberg et al. (1978), however, report activity in animals from the
same location to be almost three times as great. Their reported
activity of 4.4 ± 0.4 dpm/kg, dry, and wet/dry ratio of 7.5 corre-
sponds to 260 ± 20 fCi/kg, wet. (On a dry-weight basis this differ-
ence is even greater.) Although their Coos Head sample had one of

Table 8. Plutonium and Americium in Flesh of _Mytilus californianus_.
Activities in fCi/kg of fresh tissue.

date	site[a]	Pu-239,240		Am-241		Am/Pu
9/79	CH	111 ± 4	(6)[b]	218 ± 8	(6)	2.0 ± 0.1
1/80	CH	71 ± 4	(3)	193 ± 9	(3)	2.7 ± 0.2
3/80	CH	83 ± 5	(5)	190 ± 10	(3)	2.3 ± 0.2
	CA	90 ± 5	(4)	360 ± 20	(3)	4.0 ± 0.3
6/80	CH	75 ± 10	(1)	150 ± 20	(1)	2.0 ± 0.4
	CA	178 ± 6	(3)	360 ± 10	(3)	2.0 ± 0.1
8/80	CH	96 ± 5	(3)	170 ± 20 68 ± 9		1.8 ± 0.2 0.7 ± 0.1
	CA	79 ± 6	(2)	111 ± 6	(2)	1.4 ± 0.1
9/80	CH	180 ± 10	(2)	480 ± 50 140 ± 10		2.7 ± 0.3 0.78 ± 0.07
11/80	CH	80 ± 5	(3)	470 ± 20	(2)	5.9 ± 0.4

a CH: Coos Head; CA: Cape Arago
b The number of analyses is in parentheses.

Table 9. Tissue Fractionation of Plutonium and Americium in Mytilus californianus. Sample taken from Coos Head in June 1980. Activities in fCi/kg of fresh tissue.

tissue	relative mass	Pu-239,240	Am-241	Am/Pu
gill	0.081	58 ± 6 (2)[a]	130 ± 20 (2)	2.2 ± 0.4
gut+viscera	0.552	84 ± 5 (2)	190 ± 10 (2)	2.2 ± 0.1
mantle+gonad	0.367	41 ± 4 (2)	140 ± 10 (2)	3.4 ± 0.4
"entire"[b]		66 ± 3	167 ± 7	2.5 ± 0.2
byssus[c]		1090 ± 70 (2)	200 ± 40 (2)	0.19 ± 0.04

a The number of analyses is in parentheses
b Reconstructed by summing the weight-corrected activities of each tissue.
c From September 1979 collection at Coos Head.

Table 10. Plutonium and Americium in Other Biota from Coos Bay.
Activities in fCi/kg of fresh tissue.

organism	date	tissue	Pu-239,240	Am-241	Am/Pu
Tresus capax	10/79		326 ± 14 (3)[a]	210 ± 10 (3)	0.64 ± 0.04
" "	1/80		375 ± 12 (3)	190 ± 60 (1)	0.51 ± 0.16
" "	4/80		320 ± 9 (3)	200 ± 30 (1)	0.63 ± 0.10
" "	4/80	neck[b]	990 ± 30 (3)	490 ± 60 (1)	0.49 ± 0.06
		gut+viscera	300 ± 30 (1·)	160 ± 30 (1)	0.53 ± 0.11
		"entire"[c]	>590	>300	~0.51
" "	6/80		460 ± 30 (1)	200 ± 10 (1)	0.43 ± 0.04
Macoma nasuta	1/80		500 ± 30 (1)	270 ± 30 (1)	0.54 ± 0.07
" "	4/80		250 ± 20 (2)	96 ± 13 (1)	0.38 ± 0.06
Abarenicola sp.	10/79		19 ± 7 (1)	11 ± 9 (1)	0.58 ± 0.52
Saxidomus giganteus	10/79		590 ± 160 (1)	-	-

a The number of analyses is in parentheses.
b Neck and gut+viscera made up 46% and 43%, respectively, of the total weight of tissue.
c Reconstructed from the neck and gut+viscera fractions alone, therefore minimum estimates.

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the highest activities reported (Goldberg et al. 1978), their average for all M. californianus (180 ± 50 fCi/kg, wet) is also higher than was generally found in this investigation - thus, there appears to be a systematic difference between these two sets of measurements. The reason for this is not apparent.

Despite the disparity in absolute concentrations of transuranics, the Am/Pu ratio measured in 1979-80 is essentially identical to that found by Mussel Watch in 1976. The average ratio lies between two and three, almost tenfold higher than is found in water or on particulate matter. (Replicate Am analyses of August and September samples from Coos Head had very different results, and both are presented in Table 8. On the basis of the Am/Pu ratio calculated from each of these values, it seems likely that the lower result, in each case, is erroneous. A complete tabulation of analytical results is presented in the appendices.) In two instances an unusually high Am/Pu ratio has been obtained; in neither case are the plutonium or americium concentrations outside their normal range. These large values may well be a manifestation of the skewed distribution common to ratio variables (Atchley et al. 1976).

There is no distinct difference between mussels from Cape Arago and Coos Head, in either plutonium concentrations or Am/Pu ratio. Although the high activities in June 1980 at Cape Arago are not duplicated at Coos Head, there is no similarly marked difference at other dates. Nor is the high value at Cape Arago out of the range of the Coos Head population, for they reach a comparable concentration in September.

Tissue distribution of transuranics in M. californianus is shown in Table 9. Slight differences appear to exist between the tissues. The gut+viscera fraction has the highest concentrations of both Pu and Am; this may be the consequence of food in the digestive tract. Nonetheless, the Am/Pu ratio in this tissue is ordinary. It is the mantle+gonad fraction which has an unusual ratio; this results from a plutonium content that is proportionally lower, in relation to other tissues, than is that of americium.

Byssus material is distinctly different from soft tissue, having a plutonium content that is tenfold greater. This is not a surprising result - it has been observed before, not only for transuranics (Fowler et al. 1975), but for other metals as well (Roberts 1976). Byssal threads are secreted by the foot, a small organ in Mytilus, which has been included in the gut+viscera fraction. If that organ had characteristics like the threads themselves, then the fraction containing it might be expected to also show a marked elevation of the plutonium content or depression of the Am/Pu ratio; it does not. Differences between byssus and soft tissue presumably reflect different mechanisms of uptake, or different sources, of transuranics; the distinct chemical structure of the byssus may be important.

By summing the weight-corrected activities of each tissue fraction, the plutonium and americium activities in the entire animal can be reconstructed - the result is shown in Table 9. This can be compared to another sample collected on the same date and analyzed without dissection (Table 8). Americium and plutonium activities are much alike in the two samples. The variation is

no greater than be ascribed to counting error alone. (In this case also, there is more variation when the comparison is made on a dry-weight basis, as a result of the different wet/dry weight ratios of these samples.) Although these two samples do not provide a measure of population variability, they do indicate that the number of animals collected was sufficient to integrate across any such variability. Despite the similar activities in each sample, the Am/Pu ratios are somewhat different, an illustration of the effect that small variations in measured activity can have.

Other species collected from Coos Bay (Table 10) differed from M. californianus in both plutonium content and Am/Pu ratio. Plutonium activities in Tresus, Macoma, and Saxidomus were much greater, that in the worm was much less. All of these species have Am/Pu ratios that are much lower than M. californianus, ranging from 0.4 to 0.6. Like the ratio in Mytilus californianus, however, these values appear to be systematically, if only slightly, higher than those of surrounding water and sediment.

Tissue fractions of Tresus, like those of M. californianus, have Am/Pu ratios like the whole animal. The distribution of absolute activity is not so homogenous, however. By far the most activity is localized in the neck. This distinction, more than the higher concentrations and lower Am/Pu ratio, suggests that Tresus and M. californianus have innately different responses to transuranics.

Summation of the activities in the tissues of Tresus yields a 'reconstructed' animal, just as with Mytilus, but the lack of

measurements for two tissues only allows a lower bound to be put on the total activity of each isotope. Even so, the 'reconstructed' animal has more activity than its undissected counterpart. This may be a consequence of the smaller number of individuals of _Tresus_ that were analyzed; thus, within-population variation appears to be detectable in samples of about two dozen animals - the same scale of variation may not apply to M. _californianus_, however.

The existence of individual responses to transuranics even among infaunal species is testified to by the measurements of activity in _Abarenicola_. This worm is quite unlike the bivalves. Despite the difference in absolute concentration, though, the bivalves and _Abarenicola_ have indistinguishable Am/Pu ratios.

DISCUSSION AND CONCLUSIONS

Plutonium activity in Coos River and nearby water varied considerably from date to date, but not in a way that can be related to known seasonal changes in the environment. Increased runoff during winter and spring and upwelling during the summer are the major factors which might be expected to influence transuranic activity.

In the Coos River there is no systematic difference in Pu activity between winter and summer (Figure 3). Water and particle samples show similar variations from date to date (r = 0.88), confirming than the fluctuations observed are actual effects and not simply the result of sampling and analytical error.

Because this is a relatively small watershed, short-term changes in the flow regime may be equivalent in magnitude to the largest seasonal variations. More frequent measurements and simultaneous estimates of river discharge might resolve this, but in any event it seems clear that no extraordinarily high or low levels of transuranics in river water characterize particular periods of the year.

Plutonium activity in offshore water is more variable than in the river (Fig. 3), and, as in the river, the variation of particle-bound Pu is greater than that of whole-water samples. Particulate and unfiltered samples do not covary as strongly offshore as in the river, however. In large part this is due to the March 1980 sample, in which the particulate fraction makes up an unusually small proportion of the total (Fig. 3 and Table 7). Although impossible to
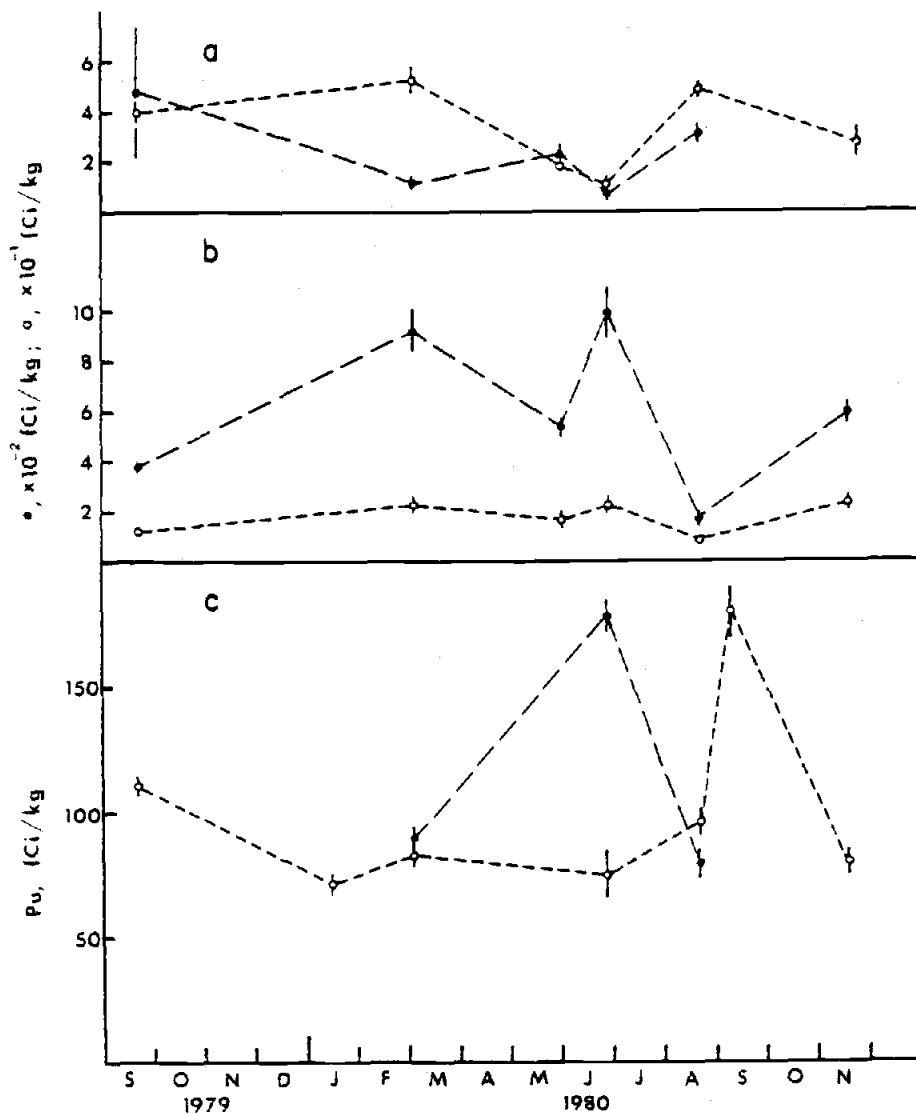
Figure 3. Seasonal variation of Pu-239.240 in water and
           mussels. a) nearshore water. b) Coos River water.
           For a and b, o - unfiltered water, • - filterable
           particulates. c) Mytilus californianus; o - Coos
           Head, • - Cape Arago.

ascertain in retrospect, a low particle population or leakage through a ruptured filter may have caused this. Correlation of the two forms is poor (r = 0.32) but rises considerably (r = 0.79) when the March data are excluded.

Upwelling seems to exert little influence: although the Pu concentration in whole water samples is lowest during May and June, at the start of upwelling, in August it is quite high. Particulate Pu concentrations offer no additional evidence for upwelling effects. It may be that water at the depth of upwelling (150-200m) is not significantly different from that at the surface. No great change in Pu activity is seen over this depth range in the central Pacific (Bowen et al. 1980).

The most marked anomaly in transuranic behavior does not occur between dates, or between locations, but between elements. Americium shows a clearly higher affinity for particulate material, at least in coastal water. The similarity between Coos Bay and the Mediterranean is striking; the same mechanism may be operating. A preponderance of the particles in the nearshore region may be of terrestrial origin and have a greater affinity for Am than Pu, as suggested by Livingston et al. (1977) and Holm et al. (1980). The consequence, however, is not an Am/Pu ratio on the particles that is elevated above that expected in mixed fallout; rather, the ratio of dissolved forms seems to be depressed. Evidence of this can be seen in the ratios of whole water samples (Table 5), which are mixtures of dissolved and particulate species, and have ratios lower than the particulate material alone (Table 6). From the data collected here it is theoretically possible to calculate Pu and Am activities in

the dissolved fraction and take their ratio. Doing so does yield

low ratios, but the propagation of errors through such a process

renders the results of little significance; therefore the calculated

ratios are not presented here.

Although these measurements are not precise enough to support a

strong conclusion, fractionation of Am and Pu upon particles may,

in this instance, produce material - dissolved species - with a

slightly lowered Am/Pu ratio. In any case, the ratio in neither

dissolved nor particulate forms is substantially elevated.

Just as suspended particulate matter has an Am/Pu ratio that is

'normal' (for 1979-80), so also does the bottom sediment in Coos Bay.

Thus, in the absence of any environmental material with a

similar characteristic, the elevated Am/Pu ratio in Mytilus califor-

nianus appears to be a consequence of the mussel's metabolism.

This conclusion is contrary to that reached by Koide et al. (1981).

On the basis of a geographically widespread series of measurements,

Koide and his colleagues suggest that the Am/Pu ratio on particles

is typically high, and that M. californianus merely reflects this.

A conclusion clearly supported by their data, but not emphasized by

the authors, is that there is considerable variability of the Am/Pu

ratio on suspended particles. The measurements of Koide et al.

(1981) are summarized in Table 11, including the propagated error

of the ratios computed from data given by the authors.

Local variations are marked. Several locations are outstanding,

but at most of these the ratio is not as elevated as that

in M. californianus. Within this tabulation there are samples from

Table 11. Am-241/Pu-239,240 Ratios of Particulate Material.
         Data from Koide et al. (1981)

location	date	depth (m)	Am/Pu
Scripps Canyon	7/70	100	1.0 ± 0.3
San Clemente Basin	7/70	500	0.3 ± 0.14
San Diego Trough	7/70	1000	1.5 ± 0.6
		1600	3.2 ± 0.9
Baja California	6/71	2000	0.5 ± 0.1
Ross Sea	2/72	125	2.2 ± 0.6
		350	2.3 ± 0.9
		550	1.6 ± 0.4
San Onofre Outfall	6/75	3	1.0 ± 0.1
	4/76	3	0.7 ± 0.2
	6/76	3	0.6 ± 0.2
		3	2.1 ± 0.5
		4	1.3 ± 0.4
		75	1.0 ± 0.2
Del Mar-Cardiff, CA	3/76	5	2.1 ± 0.9
		5	2.6 ± 0.8
		5	1.4 ± 0.5
Central N. Pacific	6/76	1-3	1.0 ± 0.3
		10	0.9 ± 0.3
		100	1.0 ± 0.3
		500	0.5 ± 0.09
		1994	0.8 ± 0.2
		4010	0.3 ± 0.1
		5682	1.0 ± 0.4

two locations which do not bear directly on the question of transuranic availability to Mytilus: the Ross Sea, which is geographically distant, and the San Onofre outfall, at which the influence of the

reactor is undetermined. Taken as a whole, however, these determinations reveal the existence of distinct and highly localized differences in the Am/Pu ratio. The cause of these differences is unknown, but may be related to the origin and history of the particles. Terrestrial material entering the ocean will already have fairly high activities and a 'normal' Am/Pu ratio, whereas authigenic material - which must start out with little or no activity - will accumulate Am and Pu in a ratio dictated by their relative affinities, presumably acquiring a high ratio. Once placed in the marine environment, terrestrial material may also accumulate more Am than Pu, but progression toward a higher ratio will be slowed by the 'inertia' of the transuranics already attached. In nearshore water, to which mussels are exposed, it would not be unreasonable to find terrestrial material predominating, and so a low ratio.

Whatever the cause of this variation, however, elevated Am/Pu ratios do not appear to be as general as Koide et al. (1981) suggest - certainly it is not the case in Coos Bay. Consequently the conclusion that filterable material controls the mussels' radionuclide content is weakened. In Coos Bay, in fact, it is unsupportable, and may be so elsewhere also.

Filterable material, however, is a term and a quantity that covers a multitude of sins. Within the population of suspended particles there are both organic and inorganic forms, and these may be different in terms of their relative affinity for transuranics. If Mytilus californianus is capable of distinguishing between these forms, it may be exposed to material that is of different character

than that collected by mechanical filtration. It is hypothetically possible that these mussels are ingesting only a small mass fraction of the total particulate material, which has a small percentage of the solid-phase transuranics but a high Am/Pu ratio.

Figure 4 shows the Am/Pu ratio of a varying mixture of inorganic sediment and plankton, in which the plankton has a CR (concentration ratio) for Pu of 5000 (as proposed by Eyman and Trabalka 1980) and an Am/Pu ratio of 2.0 (i.e. like the mussels). The sediment is presumed to have a distribution coefficient for Pu of 10^5, and an Am/Pu ratio of 0.33. For this calculation the seawater is assumed to have an Am/Pu ratio of 0.33, as is to be expected in mixed fallout. The figure illustrates that a relatively large mass fraction of the seston can be made up of such 'plankton' before the Am/Pu ratio rises so high that it clearly deviates from the expected value. If M. californianus derives all of its transuranics from this 'plankton' fraction, then it could well have the observed ratio while filterable material appears normal.

At the present time, however. there are no data which indicate that this situation actually occurs. Two of the assumptions are unsupported. One is the ability of M. californianus to derive its Pu and Am solely from the 'plankton' fraction. Mytilus edulis, in fact, does not distinguish between inorganic and organic particles when filtering (Foster-Smith 1975), and on the basis of morphological similarity it is unlikely that M. californianus does either. This species would therefore have to selectively acquire transuranics from organic material in the gut - possible, perhaps, but not the
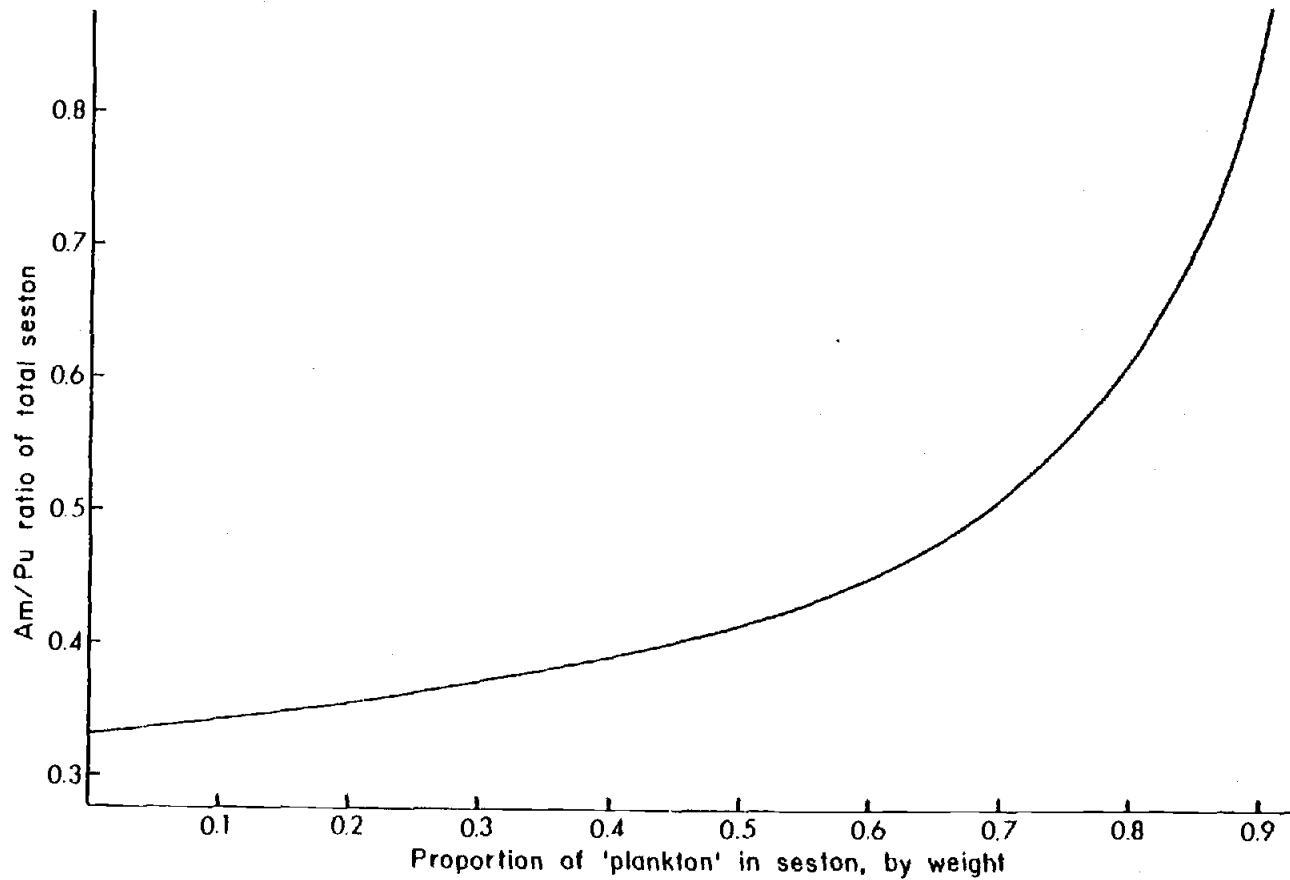
Figure 4. Am/Pu ratio of seston made up of 'plankton' with Pu CR = 5000 and Am/Pu ratio = 2 and tripton with Pu $K_d = 10^5$ and Am/Pu ratio = 0.33.

sort of feat to be taken on faith. The second unsupported assumption

is that of a high Am/Pu ratio in plankton. Despite the assertion

of Eyman and Trabalka (1980) that phytoplankton have a CR for Am

that is ten times that for Pu, there simply are no pertinent data.

To date, there have apparently been no determinations of the Am/Pu

ratio in pure phytoplankton samples. Circumstantial evidence suggests

that in fact there is no Am enrichment in the first trophic level:

anoxic Black Sea sediments, which are rich in plankton detritus and

from which no loss of transuranics is to be expected (Carpenter and

Beasley 1981) have an Am/Pu ratio (in 1973) of only 0.29 ± 0.05

(Beasley, unpublished data); the herbivorous copepod Calanus

finmarchicus had an Am/Pu ratio (in 1975) of 0.14 ± 0.01 (Ballestra

1980); and marine macrophytes show no elevation of the ratio

(Livingston and Bowen 1976). Moreover, the CR for Pu in phytoplanton

may be considerably greater than the value of 5000 used to construct

Fig. 4. Fisher et al. (1980) measured CRs of 8.5×10^4 to

2.8×10^6 in Thalassiosira pseudonana; if these values are typical

of marine plankton, and the plankton do have a high Am/Pu ratio.

then their influence would be detectable at much lower levels than

Fig. 4 suggests. Certain resolution of any differences between

organic and inorganic particles, however, waits upon the separate

collection and analysis of each fraction - a task not without

considerable practical difficulty.

Further evidence of a species-specific response in Mytilus

californianus is presented by the other Coos Bay biota. Because

the characteristics of water upriver and offshore are virtually

identical, these estuarine organisms must be exposed to material like that available to M. californianus - yet their Am/Pu ratios are different. These ratios appear to be slightly elevated with respect to available material, although measurement uncertainty weakens this conclusion. If substantiated, it would not be unreasonable to expect that this takes place by a mechanism that is biochemically distinct from that operating in M. californianus: not only is the degree of elevation much less, but the tissue distribution of transuranics in Tresus is also much different. All of the estuarine bivalves also have higher transuranic activities than the mussel. This could be a further indication of species-specific physiological differences, but may alternatively be a consequence of their benthic habitat. Evidence for the latter possibility is presented later.

Although M. californianus does not reproduce the Am/Pu ratio of suspended particles - or any other source - the radionuclides in the mussel must originate in some fraction of the environment. There are four fractions to be considered: particulate and dissolved forms, from seawater and the river. There are several means other than the Am-241/Pu-239,240 ratio by which the source might be identified.

First is the plutonium isotope ratio Pu-238/Pu-239,240. Noshkin (1972) has used this quantity to identify the source of transuranics to several marine species. In that case the possible sources had distinctly different ratios, and these were reflected in the biota. A similar situation does not occur in Coos Bay

(Table 12). No significant differences occur between source terms or between the biota and these sources. The ratio in all is about 0.04.

Table 12. Pu-238/Pu-239,240 Ratios.

source		Pu-238/Pu-239,240
sediment		
	CB0	0.032 ± 0.004
	CB1	0.040 ± 0.002
	CB2	0.038 ± 0.005
water		
	ocean	0.036 ± 0.011
	river	0.048 ± 0.013
particles		
	ocean	-
	river	0.048 ± 0.013
biota		
M. californianus Coos Head		0.036 ± 0.004
M. californianus Cape Arago		0.050 ± 0.020
Tresus capax		0.032 ± 0.004
Macoma nasuta		0.016 ± 0.006

A second possibility is seasonal covariance of isotope concentrations in Mytilus and the major source material. This comparison is complicated by a number of factors. Principal among these is the biological half-life of transuranics in Mytilus; measurements made with Mytilus galloprovincialis suggest that this may be days to many weeks (Fowler et al. 1975, Guary and Fowler 1981). Activity in the mussels is therefore unlikely to have any simple relationship to water-borne activity at any one time, but to represent a complicated integration of the water activity for some previous period . Also, the covariance of water and particle activities in the Coos

River means that if the mussels correlate well with one, they will necessarily be correlated with the other. (The possibility of such covariance in offshore water has been discussed; an error in one of the values, as suggested, may obscure any relationship with the mussels.) Lastly, there are relatively few points at which to compare activities in the water and animals; this can lead to high, but spurious, correlations. In fact, there are no unequivocal similarities between mussels and any single source term (Fig. 3). Plutonium in Coos Head mussels most closely followed particulate material in seawater, and americium most closely followed particulate material in river water. The implicit conclusion that there is a different source for each element is unsupportable, however. Mussels from locations without fluvial input - Cape Arago and elsewhere (Goldberg et al. 1978) - not only accumulate americium, but do so in a high proportion to plutonium.

The third means of identifying the origin of transuranics is by consideration of the concentration ratios (Table 13). These have been calculated, as is usually done, in relation to unfiltered water. In doing so the possible differing roles of particulate and dissolved transuranics is disregarded. A caveat must therefore be applied: if these roles differ greatly and the proportion of particle-bound isotopes varies widely from place to place, then the calculated CR will show little constancy. Bearing this in mind, the CRs of Coos Head and Cape Arago mussels can be compared, and these compared with other values for the genus.

The concentration ratios of Table 13 highlight the similarity

Table 13. Concentration Ratios of Pu-239,240 in Mytilus
californianus.

| | Coos Head mussels | |
date	relative to offshore water	relative to Coos R. water
9/79	250 ± 56	900 ± 190
3/80	170 ± 40	390 ± 100
6/80	520 ± 160	270 ± 70
8/80	180 ± 40	1040 ± 280
11/80	240 ± 75	280 ± 70
	200 ± 25[a]	350 ± 40[a]

| | Cape Arago mussels |
date	relative to offshore water
3/80	250 ± 60
6/80	1470 ± 390
8/80	170 ± 40
	200 ± 30[a]

a Weighted average of values at all dates.

between Coos Head and Cape Arago mussels relative to the difference between river and ocean water. Despite their location within the mouth of the estuary, Coos Head mussels are clearly most strongly tied to the ocean environment.

Other mussels (Table 1) have similar concentration ratios. In particular, the only previous measurement of M. californianus (Pillai et al. 1964) is not greatly different from the present, despite the sixteen year interval. Several conclusions can be drawn from these data: a) all species of the genus show a similar - perhaps identical - response to Pu in the water, b) environmentally measured concentration ratios in Mytilus are probably good estimates of actual concentration factors, and c) the caveat above does not seriously compromise the use of concentration ratios - i.e. either the partition of transuranics between dissolved and particulate forms is everywhere alike, or mussels respond similarly to both forms.

The first of these conclusions leads further to the result that the species-specific differences in Am/Pu ratio of M. californianus must be the consequence of an atypically greater uptake or retention of americium. This is seen also in a comparison of the CR for Am of M. californianus and M. galloprovincialis (the only other species for which such a value is available). These two values are, respectively, about 2500 (this work) and 500-1500 (Ballestra 1980).

The similarity of Pu CRs for all species leads to the second conclusion. It is supported by the biological half-life (days to weeks) of the majority of Pu in Mytilus galloprovincialis (Fowler et al. 1975, Guary and Fowler 1981), which indicates that the mussels

might respond with fair rapidity to changes in the Pu concentration of seawater, and thus remain at or near equilibrium. Such seems to be the case, but it should be noted that on two occasions mussels did show unusually high activities which were not consistent with levels measured in the water.

The third conclusion above presents two alternatives, either or both of which may be true. It is not easy to judge the veracity of either.

Laboratory experiments confirm that Mytilus (galloprovincialis) can take up dissolved Pu (Fowler et al. 1975, Guary and Fowler 1981) but it has not yet been shown that the uptake of dissolved forms is alone sufficient to bring about the concentrations found in natural populations. The role of particulate matter is not clear.

Measurements of the proportion of particle-bound Pu in seawater span a broad range: from only a few percent (Holm et al. 1980, this study) up to 70% (Bowen 1975). This proportion will depend on both particle concentration (Duursma and Bosch 1970) and composition (Livingston et al. 1977, Holm et al. 1980). Whether or not such differences are systematic and characteristic of particular marine regimes has not been established.

The strongest evidence that particles can be a major source of transuranics comes from comparisons of benthic and pelagic species. In both freshwater (Eyman and Trabalka 1980) and marine habitats (Guary and Frazier 1977) infauna and epifauna have higher plutonium concentrations than organisms living in the water column. The same relationship occurs in Coos Bay: infaunal bivalves (Tresus, Macoma,

and Saxidomus) have more plutonium than Mytilus. Even within the
genus Mytilus this phenomenom occurs: a population living within
the sediment of the Baltic Sea (Miettinen et al. 1975; Table 1) has
more activity than others.

Suspended and sedimented particulate matter, however, are not
necessarily comparable sources of plutonium. Although Mytilus
is a sessile species, its principal contact with particles is a
consequence of filtration and ingestion rather than physical proxi-
mity. An indication that uptake by adsorption may be overwhelmingly
greater than that by ingestion is provided by a comparison of
Macoma nasuta and Tresus capax. Although Macoma ingests sediment
and Tresus does not, the former does not show a distinctly higher
plutonium activity; if there are no great physiological differences
between these species in their treatment of transuranics, then inges-
tion appears to be an insignificant route of uptake. Thus the lower
Pu activity in mussels relative to infauna does not eliminate the
possibility that mussels acquire their transuranics from resuspended
sediment or other particulate matter. However, neither these data
nor any others directly address the relative roles of particulate
and dissolved transuranics. Experimental studies may be the only way
to distinguish their influences.

Whatever the actual source of transuranics, uptake itself may
proceed by either adsorption or assimilation. Neither need necessar-
ily reproduce the Am/Pu ratio of source material; uptake mechanisms
for the two elements may differ. Alteration of the Am/Pu ratio may
be a consequence of the animal's general physiology or an anomalous

response of a single organ. The dissections of M. californianus bear upon these alternatives.

Transuranics - particularly americium - appear to be metabolically deposited in Mytilus tissue rather than simply adsorbed to it. This follows from the nearly constant wet-weight activity in different fractions, each of which presumably differs in suface area. The gill in particular will have a proportionally greater surface area than other organs, yet its concentration of transuranics is unremarkable.

This relatively homogenous distribution of activity among tissues is in sharp contrast to the distribution of Am in Mytilus galloprovincialis following a single exposure (Guary and Fowler 1981). The two sets of data are not directly comparable owing to the fact that M. galloprovincialis was exposed only to dissolved Am and for a period of only 28 days; nevertheless the absence of any tendency toward equilibration of Am concentrations in different tissues of M. galloprovincialis over the course of a year provides a further suggestion of fundamental differences between these two species.

Organs of M. californianus may nevertheless respond differently to Pu and Am: the Pu concentration in the mantle+gonad fraction is low. This alone, however, is not sufficient to explain the elevated Am/Pu ratio of the entire animal. Other organs also show a substantial elevation; whatever metabolic action is responsible, its influence is not confined to a single tissue. The simplest explanation of americium enrichment is through the operation of a single mechanism; the only point at which a single mechanism might operate and

influence the entire animal is at the integument, at which uptake and loss occur. The principal sites for such exchange are gill and gut (uptake) and nephridia (loss). Even if these tissues could be completely separated and analyzed, however, their relative importance could not be determined from concentration data alone.

The low plutonium concentration in the mantle+gonad fraction may be a consequence of the mantle's role in shell formation. As much as 90% of the plutonium in the entire animal is deposited in the shell (Fowler et al. 1975, Guary and Fowler 1981), and it may be incorporated there rather than adsorbed, as is shown by the insignificant amounts that are removed by rinsing with 0.1 $\underline{N}$ HCl (Fowler et al. 1975). (Loss of Pu from the shell can occur following exposure to seawater, however, as has been shown by Guary and Fowler (1981). The nature of this binding, therefore, is not clear.) Plutonium presumably must be moved through the mantle if it is to be metabolically deposited in the shell, and a low concentration in the mantle may be a manifestation of a large loss pathway to the shell.

In contrast to the flesh, transuranic uptake by byssal threads appears to be principally adsorptive. The relative plutonium activities in byssus and flesh found in the Sept. 1979 mussels can be compared with those found by Fowler et al. (1975) in an experimental study. Theirs was a laboratory measurement of uptake and loss rates of Pu-237 by Mytilus galloprovincialis. Although the species concerned differ, comparison of the two sets of results can be justified on the basis of their similar CRs for plutonium.

The uptake studies of Fowler et al. (1975) ran for only 25 days, after which time the mussels had not reached equilibrium with seawater; concentration ratios of whole mussels were only 27-70. In these M. galloprovincialis, not at equilibrium, the byssus contained 30-63% of the plutonium in the entire animal (flesh + byssus + shell). In M. californianus (Sept. 1979) the byssus contained only 18% of the total plutonium in flesh + byssus; if shell had been measured as well, this proportion would presumably be lower. Thus, it appears that the byssus takes up plutonium faster than the flesh, approaching equilibrium more quickly. This supposition is in accord with the observation of Fowler et al. (1975) that loss of Pu-237 occurs from two pools, and that the faster loss occurred from a pool containing about 35% of the total Pu. Adsorption to and desorption from the byssus may take place faster than biochemically mediated exchange between flesh and water. If adsorptive processes do govern transuranic attachment to the byssus, then it is no surprise that byssal threads have an Am/Pu ratio more like the water than like mussel flesh. In fact, the ratio of the byssus (0.19 ± 0.04) is lower than the "expected" value of about 0.34, but as has been suggested, the greater affinity of Am for particulate matter may result in a lowering of the ratio in the dissolved fraction, and the byssus appears to provide an indirect confirmation of this.

Although there is no apparent seasonal variation of activity in M. californianus, both populations show one unusually high value. Because the June peak in the Cape Arago population is not duplicated at Coos Head, this is apparently not the result of a widespread

introduction of high-activity material. Water samples do not show fluctuations linked with those of the mussels, so an event supplying high activity to the mussels must have been highly localized in time as well as space. These constraints make it appear unlikely that the mussels are simply responding to a change in the supply of transuranics. It is possible that the mussels at Cape Arago in June and Coos Head in September obtained their transuranics not from local water at the time of sampling but from water which they had encountered days or weeks before. Based on current knowledge, the only source of high-activity water would be the central Pacific; that a packet of such water sufficiently small to influence mussels at one site but not the other could travel into the coastal region and retain its integrity long enough to influence the mussels does not seem plausible. The possibility cannot be disregarded, however. There are two alternatives. First is contamination of the sample after collection. Both samples of mussels, however, have a typical Am/Pu ratio; any contaminant must have had this ratio also, which would be a remarkable occurrence. The other alternative is that many of the mussels in each population have, in synchrony, undergone a physiological change resulting in greater uptake or longer retention of transuranics. This possibility cannot be evaluated directly, as no measurements of physiological condition were made. Spawning in populations of M. californianus is not confined to a single season, nor does it occur simultaneously throughout a population; little more seems certain, however, about seasonal physiological changes in this mussel (Gabbot 1976).

Thus, the cause of these changes is unclear. The rate of loss of Pu from each peak to the following date corresponds to biological half-lives of 50 and 60 days, for the Coos Head and Cape Arago populations, respectively. Goldberg et al. (1978) noted similarly rapid changes in monthly samples from Bodega Head, although there also there were no systematic seasonal changes. Biological half-lives measured in M. galloprovincialis (Guary and Fowler 1981) would allow a halving of the activity in only about 30 days, which could account for the changes seen here. That the rapidity of uptake at Coos Head is reasonable cannot so easily be established, as there are no experimentally measured rates that are suitable for comparison. The changes seen here indicate that uptake can, under certain (unknown) conditions, be quite rapid and be apparently unrelated to environmental levels. This point bears upon the utility of M. californianus as an indicator organism: if fluctuations as great as those seen here can occur in the absence of a proportionate change in environmental levels, then the animal's ability to signal such changes is compromised. Given the short turnover times of transuranics, more frequent sampling of mussels and water - monthly or even biweekly - would be necessary to judge the existence and effect of such large short-term changes in water-borne Pu and Am. If such changes do occur and are reflected in M. californianus, then the sampling interval for a monitoring program might have to be equivalently short.

SUMMARY

Mytilus californianus, unlike many other marine species, including closely related bivalves, is capable of distinguishing between plutonium and americium. Its response to plutonium is like that of its congeners, as is shown by the similarity of their concentration ratios; americium is maintained at a much higher concentration, with Am-241/Pu-239,240 ratios in the mussels ranging from two to three.

Plutonium and americium concentrations are not substantially elevated in the Coos Bay area. Plutonium concentrations in near-shore water are 0.1-0.5 fCi/kg, far less than the high values recorded in the central Pacific by Bowen et al. (1980), but typical of other oceanic measurements. River water is similar. Neither water nor sediment entering the estuary has a characteristically high Am/Pu ratio, and the high ratio found in Mytilus californianus does not exist in other intertidal biota. This mussel is therefore concluded to have a species-specific response to transuranics.

The elevated Am/Pu ratio in M. californianus is not the result of accumulation of americium in any one particular tissue, but of a heightened concentration throughout the body. Organs of assimilation and excretion are therefore likely to be the site of discrimination between these elements, and the discrimination is suggested to proceed by differential transport across membranes

rather than by the condensation of americium into some non-labile storage form. It is not possible, however, with these data, to judge whether uptake or loss mechanisms control this process.

Also unresolved is the relative importance of particulate and dissolved forms of transuranics. As is the case elsewhere, infaunal species have higher plutonium activities than others (i.e. Mytilus) not in such intimate contact with the sediment. This does not serve, however, to support the conclusion that particulate material is the most important source of transuranics to bivalves - particularly in light of species-specific differences.

The physical behavior of plutonium and americium in the Coos Bay region suggests that local conditions may greatly influence the distribution of these two elements. Americium has a greater affinity for particles, as in the Mediterranean, but the Am/Pu ratio on those particles is not elevated to the extent seen elsewhere in the Pacific (Koide et al. 1981). The presence of large amounts of terrestrial material with a previously established label may hinder the observation of the results of purely marine processes. Subdivision of the seston and separate analysis of its components - terrestrial and marine, organic and inorganic - may provide not only a better understanding of the geochemistry of these elements, but also of their availability to biota.

Although Am/Pu ratios in Mytilus californianus accorded with those measured by Goldberg et al. (1978), the absolute concentrations of transuranics were only one-half to one-third as great. This difference, as well as short-term changes in the activity of

M. californianus that appear unrelated to environmental fluctuations,
suggest that caution be applied to the interpretation of data from
this organism. In Mytilus the effects of age, temperature, salinity,
and body size - as well as the nature of the source - on transur-
anic uptake are all unknown. The need for an understanding of all
the responses of an indicator organism is highlighted by Mytilus
californianus' unusual affinity for americium.

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APPENDICES

APPENDIX A

Plutonium and Americium Activities in Sediments of Coos Bay.

Activities in pCi/kg.

depth (cm)	porosity	Pu-239,240	Pu-238 / Pu-239,240 (%)	Am-241
Core CB0				
0-3	0.426	3.6 ± 0.2	18 ± 3	1.0 ± 0.2
3-4	0.408	2.1 ± 0.7	2.5 ± 1.0	-
		3.1 ± 0.2	-	-
4-5	0.403	3.4 ± 0.3	1.6 ± 0.9	2.0 ± 0.8
		-	-	0.9 ± 0.2
5-6	0.405	4.1 ± 0.4	1.7 ± 1.2	-
6-7	0.399	3.0 ± 0.4	2.9 ± 2.0	-
7-8	0.396	2.3 ± 0.4	6.3 ± 3.7	1.2 ± 0.3
8-9	0.398	2.6 ± 0.3	6.7 ± 3.0	1.0 ± 0.2
9-10	0.407	3.3 ± 0.3	4.4 ± 2.0	1.6 ± 0.3
10-11	0.408	3.7 ± 0.4	4.2 ± 1.9	-
11-12	0.430	4.0 ± 0.4	0.7 ± 0.7	0.43 ± 0.11
		3.6 ± 0.3	4.4 ± 1.8	-
12-13	0.446	4.3 ± 0.4	3.9 ± 1.6	-
13-14	0.419	4.5 ± 0.4	-	-
14-15	0.427	5.1 ± 0.3	4.1 ± 1.0	1.7 ± 0.2
Core CB1				
0-2	0.665	4.9 ± 0.4	5 ± 2	1.2 ± 0.2
		5.1 ± 0.2	6 ± 1	1.2 ± 0.2
		4.6 ± 0.2	2.8 ± 0.9	-
2-4	0.646	4.2 ± 0.4	6 ± 2	1.5 ± 0.2
		4.6 ± 0.2	4.7 ± 0.9	1.5 ± 0.2
		4.8 ± 0.2	2.6 ± 0.7	-
4-6	0.701	5.6 ± 0.4	4 ± 1	-
		4.6 ± 0.6	3 ± 3	-
7-8	0.694	5.8 ± 0.4	2.3 ± 1.5	1.4 ± 0.2
		6.0 ± 0.4	3 ± 1	-
		5.3 ± 0.2	4.7 ± 0.8	-
10-11	0.710	5.9 ± 0.4	4.6 ± 1.0	1.5 ± 0.3
		7.7 ± 0.6	5 ± 1	-
13-14	0.612	2.9 ± 0.3	5 ± 3	-
		3.2 ± 0.3	4 ± 2	-
		2.6 ± 0.3	2 ± 2	0.65 ± 0.28
16-17	0.731	6.8 ± 0.5	3 ± 1	2.3 ± 0.4

depth (cm)	porosity	Pu-239,240	$\frac{\text{Pu-238}}{\text{Pu-239,240}}$ (%)	Am-241
19-20	0.773	9.0 ± 0.6	2.7 ± 0.9	2.5 ± 0.4
22-23	0.808	7.8 ± 0.4	4 ± 1	-
		7.5 ± 0.5	3 ± 1	-
25-26	0.818	7.9 ± 0.5	4 ± 1	-
		7.0 ± 0.4	5 ± 1	-
		7.3 ± 0.6	3.3 ± 1.3	1.5 ± 0.3
28-29	0.715	11.9 ± 0.6	4.4 ± 0.9	-
		10.8 ± 0.4	4.3 ± 0.8	-
		11.7 ± 0.7	3.6 ± 1.1	3.1 ± 0.4
31-32	0.548	6.2 ± 0.4	5 ± 1	-
		5.3 ± 0.4	5 ± 2	2.0 ± 0.3
34-35	0.673	13.1 ± 0.7	6 ± 1	-
		12.3 ± 0.7	3.5 ± 1.2	3.6 ± 0.5
37-38	0.612	5.5 ± 0.3	3 ± 2	1.7 ± 0.2
		6.7 ± 0.4	6 ± 1	1.4 ± 0.3

Core CB2

depth (cm)	porosity	Pu-239,240	$\frac{\text{Pu-238}}{\text{Pu-239,240}}$ (%)	Am-241
0-2	0.446	1.6 ± 0.2	3.7 ± 2.6	-
2-5	0.396	1.4 ± 0.2	-	0.83 ± 0.30
5-6	0.371	0.75 ± 0.14	-	0.30 ± 0.20
6-7	0.377	1.1 ± 0.2	7.1 ± 4.2	0.52 ± 0.27
7-8	0.383	0.89 ± 0.12	1.6 ± 1.6	0.14 ± 0.05
8-9	0.386	1.1 ± 0.2	-	-
9-10	0.383	0.71 ± 0.14	-	-
10-11	0.381	0.82 ± 0.18	-	-
11-12	0.390	1.3 ± 0.2	2.8 ± 2.8	-
12-13	0.398	1.5 ± 0.2	13 ± 5	-
13-14	0.408	0.94 ± 0.10	4.4 ± 2.5	0.39 ± 0.08
14-15	0.391	1.1 ± 0.1	6.6 ± 3.0	0.19 ± 0.07
15-16	0.400	1.2 ± 0.1	-	0.16 ± 0.09
16-17	0.387	1.5 ± 0.1	4.9 ± 2.2	0.32 ± 0.11
17-18	0.384	1.1 ± 0.1	7.1 ± 3.3	0.20 ± 0.08
18-19	0.384	0.97 ± 0.12	2.9 ± 2.1	-
19-20	0.391	1.0 ± 0.1	-	-
20-21	0.397	0.86 ± 0.13	4.7 ± 3.3	-
21-22	0.393	1.1 ± 0.1	3.5 ± 2.3	-
22-23	0.394	1.2 ± 0.2	-	-
23-24	0.400	1.2 ± 0.1	-	0.26 ± 0.08
24-25	0.407	1.8 ± 0.2	3.9 ± 2.1	-
25-26	0.393	1.5 ± 0.1	3.3 ± 1.5	-
26-27	0.401	1.8 ± 0.3	6.4 ± 3.7	0.18 ± 0.09
27-28	0.403	1.4 ± 0.1	3.7 ± 1.9	-
28-29	0.407	2.1 ± 0.2	6.9 ± 1.8	0.44 ± 0.10
29-30	0.403	1.3 ± 0.2	5.7 ± 2.9	0.32 ± 0.07

depth (cm)	porosity	Pu-239,240	$\dfrac{\text{Pu-238}}{\text{Pu-239,240}}$ (%)	Am-241
30-31	0.411	1.1 ± 0.1	2.0 ± 1.4	-
31-32	0.421	0.69 ± 0.09	3.4 ± 2.4	-
32-33	0.440	0.70 ± 0.09	3.5 ± 2.5	-
33-34	0.452	0.38 ± 0.07	11 ± 7	-

APPENDIX B

Plutonium and Americium Activities in Water.

Activities in aCi/kg.

sample	date	sample size (kg)	Pu-239,240	Pu-238/Pu-239,240 (%)	Am-241
Unfiltered water					
Coos River	9/79	55	120 ± 20	10 ± 7	110 ± 40
	3/80	60	230 ± 30	3.8 ± 1.6	79 ± 18
	5/80	55	170 ± 30	-	47 ± 130
	6/80	54	250 ± 30	-	28 ± 19
		54	210 ± 30	-	63 ± 18
	8/80	58	110 ± 20	-	-
		58	47 ± 19	-	-
	11/80	54	230 ± 30	6.5 ± 3.0	-
		55	240 ± 30	6.5 ± 3.9	97 ± 58
nearshore	9/79	56	400 ± 35	4.1 ± 3.1	30 ± 16
	3/80	62	530 ± 45	2.2 ± 1.9	-
	5/80	60	180 ± 24	-	29 ± 25
		55	260 ± 50	-	-
	6/80	53	170 ± 36	-	40 ± 20
		53	110 ± 20	15 ± 7	54 ± 29
	8/80	57	500 ± 34	3.8 ± 1.5	88 ± 30
		58	470 ± 50	-	-
	11/80	55	240 ± 70	-	-
		55	410 ± 120	-	74 ± 46
Filterable particulates					
Coos River	9/79	795	38 ± 2	3.5 ± 2.2	12 ± 2.8
	3/80	871	93 ± 9	-	-
	5/80	999	40 ± 3	-	11 ± 4.1
		1022	85 ± 5	5.1 ± 1.4	51 ± 12
	6/80	492	100 ± 11	3.3 ± 3.4	-
	8/80	1075	17 ± 1.6	-	3 ± 4
	11/80	1173	60 ± 5	-	30 ± 7
nearshore	9/79	1045	48 ± 27	-	12 ± 2.7
	3/80	1096	11 ± 2	-	14 ± 10
	5/80	666	23 ± 4	-	7.8 ± 4.0
	6/80	1111	7.8 ± 1.6	-	4.2 ± 1.7
	8/80	712	32 ± 4	-	1.9 ± 4.6

APPENDIX C

Plutonium and Americium Activities in Biota From Coos Bay.
All activities in fCi/kg of dry tissue.

species	date	location	wet/dry ratio	Pu-239,240	Pu-238/Pu-239,240 (%)	Am-241
M. californianus	9/22/79	Coos Head	5.56	680 ± 70	5.0 ± 2.1	1200 ± 80
				640 ± 50	3.0 ± 1.4	1250 ± 280
				630 ± 50	7.4 ± 2.2	1160 ± 100
				640 ± 80	13 ± 5	1400 ± 100
				600 ± 40	2.9 ± 1.2	1450 ± 190
				580 ± 50	2.9 ± 1.3	1100 ± 100
		(byssus)	4.05	4300 ± 400	9.8 ± 3.1	860 ± 270
				4800 ± 600	2.9 ± 2.0	800 ± 220
	1/15/80	Coos Head	7.66	440 ± 60	3.5 ± 2.5	1600 ± 100
				530 ± 60	4.3 ± 2.2	1300 ± 100
				640 ± 60	3.1 ± 1.6	1600 ± 100
	3/4/80	Coos Head	6.66	510 ± 70	-	-
				560 ± 60	2.5 ± 2.2	-
				600 ± 90	-	980 ± 100
				380 ± 70	-	1500 ± 200
				630 ± 50	6.8 ± 2.9	1350 ± 70
	3/1/80	Cape Arago	8.83	730 ± 70	5.6 ± 3.4	3200 ± 200
				650 ± 180	8.0 ± 6.0	2800 ± 300
				880 ± 130	-	-
				870 ± 70	-	3400 ± 300
	6/27/80	Coos Head	5.18	390 ± 50	-	770 ± 80

species	date	location	wet dry ratio	Pu-239,240	Pu-238 Pu-239,240 (%)	Am-241
M. californianus	6/27/80	Coos Head (gill)	9.32	490 + 90	-	1300 ± 200
				580 ± 70	2.7 ± 1.9	1000 ± 300
		(mantle+gonad)	6.30	240 ± 30	7 ± 8	970 ± 80
				270 ± 30	4.1 ± 2.1	580 ± 120
		(gut+viscera)	7.27	600 ± 50	-	1300 ± 100
				620 ± 40	1.5 ± 1.3	1400 ± 700
	6/27/80	Cape Arago	6.09	1100 ± 40	3.6 ± 2.6	2800 ± 300
				1000 ± 100	4.3 ± 2.2	1900 ± 100
				1500 ± 600	8.5 ± 9.1	2390 ± 90
	8/20/80	Coos Head	5.42	490 ± 40	3.0 ± 2.0	900 ± 90
				620 ± 50	3.6 ± 1.8	-
				520 ± 60	1.7 ± 1.7	370 ± 50
	8/21/80	Cape Arago	6.58	630 ± 60	-	690 ± 50
				440 ± 50	-	780 ± 50
	9/9/80	Coos Head	5.28	1200 ± 100	3.8 ± 2.0	2500 ± 300
				870 ± 60	2.5 ± 1.6	730 ± 60
	11/19/80	Coos Head	5.20	420 ± 30	5.6 ± 1.7	2500 ± 200
				400 ± 40	-	-
				1400 ± 300	-	2500 ± 100
Tresus capax	10/19/79	Charleston	6.88	2600 ± 200	4.5 ± 1.2	1600 ± 100
				2100 ± 200	2.8 ± 1.3	1200 ± 200

species	date	location	wet dry ratio	Pu-239,240	Pu-238 / Pu-239,240 (%)	Am-241
Tresus capax	10/19/80	Charleston	6.88	2200 ± 200	3.9 ± 1.6	1300 ± 100
	1/15/80	Charleston	7.01	2600 ± 300	-	-
				2800 ± 200	3.9 ± 1.1	-
				2600 ± 100	3.2 ± 1.0	1300 ± 400
	4/14/80	Charleston	10.17	3400 ± 400	-	-
				3500 ± 200	3.0 ± 0.8	-
				3200 ± 100	3.2 ± 0.8	2000 ± 300
		(neck)	6.31	6400 ± 300	3.0 ± 0.8	-
				6100 ± 300	3.8 ± 0.7	-
				5700 ± 900	-	3100 ± 400
		(gut+viscera)	7.45	2200 ± 200	5.5 ± 1.5	1200 ± 200
	6/27/80	Charleston	10.02	4600 ± 300	2.0 ± 0.9	2000 ± 100
Macoma nasuta	1/15/80	Charleston	6.44	3200 ± 200	1.9 ± 0.8	1700 ± 200
	4/15/80	Charleston, Empire	6.79	1700 ± 400	-	650 ± 90
				1700 ± 100	1.2 ± 0.8	-
Abarenicola sp.	10/19/79	Charleston	19.74	380 ± 140	25 ± 20	210 ± 180
Saxidomus giganteus	10/19/79	Charleston	6.79	590 ± 160	-	-

APPENDIX D

Weighted Averages

For a series of observations x_1, x_2,...x_i...x_n, each with an associated standard deviation s_1, s_2,...s_i...s_n, the weighted average is computed as follows:

$$\text{weighted } \overline{x} = \frac{\sum_{i=1}^{n} x_i w_i}{\sum_{i=1}^{n} w_i}$$

$$\text{weighted } s = \frac{1}{\sqrt{\sum_{i=1}^{n} w_i}}$$

$$\text{where } w_i = \frac{1}{s_i^2}.$$

Each value is weighted by w, the inverse of its variance. Observations with small variances are thereby given more weight than those with large variances. An example calculation is given below, using plutonium measurements from the topmost section of core CB1 (see Appendix A).

i	x	s	w	x·w
1	4.9	0.4	6.25	30.6
2	5.1	0.2	25	127.5
3	4.6	0.2	25	115
			56.25	273.1

$$\text{weighted } \overline{x} = \frac{273.1}{56.25} = 4.86$$

$$\text{weighted } s = \frac{1}{\sqrt{56.25}} = 0.13$$