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Biological Fate of Cobalt-60 Released During the Corrosion of Neutron-Activated Stainless Steel in Seawater

J. S. Young
Marine Research Laboratory,
Sequim, Washington

March 1982

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BIOLOGICAL FATE OF COBALT-60 RELEASED DURING THE CORROSION OF NEUTRON-ACTIVATED STAINLESS STEEL IN SEAWATER

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March 1982

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Biological Fate of Cobalt-60 Released During the Corrosion of Neutron-activated Stainless Steel in Seawater

ABSTRACT

Passing seawater over radioactive Type 347 stainless steel in a sediment/seawater laboratory system and exposing marine animals to this environment provided information on the bioaccumulation of ^{60}Co from radioactive structural material. Exposure of marine organisms to radioactive corrosion products and directly to radioactive stainless steel in seawater simulated some of the possible conditions which could arise from the deposition of radioactive stainless steel on the ocean floor. Detectable levels of ^{60}Co in marine animals were not observed on a short term basis (5 weeks). Longterm (13 months) exposure of marine animals in a sediment/seawater system resulted in ^{60}Co bioaccumulation. The specific activity of ^{60}Co in the organisms was as much as one million times less than that initially present in the radioactive stainless steel. This was due to the dilution of ^{60}Co by stable cobalt in the seawater, sediments and organisms. As expected the ^{60}Co specific activity of the organisms never increased above that of the radioactive source. This is because ^{60}Co is chemically indistinguishable from stable Co. Increasing ^{60}Co concentration factors with decreasing ^{60}Co concentrations in the seawater and sediment media coupled with relatively constant ^{60}Co specific activities suggests a possible homeostatic control of cobalt concentrations in certain marine organisms. The evidence indicates that the marine animals derived more of the accumulated ^{60}Co from the sediments and interstitial water than from seawater. Cobalt-60 concentration factors were generally found to be lower than published cobalt concentration factors due to the predominantly insoluble nature of the corrosion products.

Baseline information is provided on trace element concentrations in deep-sea organisms. Stable Co and twenty other elements were measured in abyssal invertebrates and a fish.

INTRODUCTION

This study was designed to assess the biological fate of corrosion products from radioactive stainless steel deposited on the deep ocean bottom. To provide information on the possibility of corrosion products from neutron activated (radioactive) stainless steel being accumulated by marine organisms, several species of marine animals were exposed to radioactive type 347 stainless steel and/or its corrosion products in a variety of laboratory experiments. The animals were then analyzed for accumulation of ^{60}Co, the major long-lived (5.27 year half-life), gamma-emitting radioisotope in radioactive type 347 stainless steel and its corrosion products. It is assumed that ^{60}Co release is a fair measure of corrosion rate. The ^{60}Co specific activity was traced from the stainless steel specimens through the various transfer media into the marine organisms.

To determine the ability of deep-sea animals to concentrate radionuclides, concentration factors were determined for cobalt and nickel by utilizing stable nuclide analogs as well as ^{60}Co. Twenty-one (21) trace element concentrations in deep-sea organisms were measured to provide baseline information.

This report is a summary of the work completed on the biological accumulation of corrosion products from radioactive stainless steel in seawater and sediments. These studies are sponsored by the Department of Energy under the auspices of the Knolls Atomic Power Laboratory. The experiments were conducted at the Battelle Marine Research Laboratory, Sequim, Washington.

MATERIALS AND METHODS

Biological Uptake of ^{60}Co from Radioactive Corrosion Products

Five representative species of marine organisms were used in a series of seven laboratory experiments to determine the biological fate of radioactive corrosion products (Table 1). All of the animals chosen were benthic organisms that simulate the types of animals expected to be in closest contact with radioactive stainless steel and corrosion products deposited on the ocean bottom. Trace element concentrations of Co, Fe, Mn and Ni in the organisms and sediments were measured using atomic absorption spectrophotometry (ERCO: Energy Resources Co., Inc., 1981). Wet weight to dry weight ratios were determined from the weights of animals blotted on paper towels then freeze-dried.

All experiments were maintained at 12°C in closed or recirculating aquarium systems to prevent the loss of any radioactive material. A basic design of one such system is pictured in Figure 1. Five of the experiments utilized radioactive stainless steel specimens while two experiments used radioactive corrosion products obtained by exposing radioactive type 347 stainless steel to clayey silt from Sequim Bay

Table 1. Characteristics of marine animals utilized for biological uptake experiments.

Species	Type Organism	(a) Trace Element Concentrations (µg/g dry weight) Co	Fe	Mn	Ni	Wet Weight / Dry Weight	Applicable Experiment(s)	Feeding Habits
Neanthes virens	Polychaete worm	0.60	290	5.8	1.7	7.19	1,2,3,4A,4B	Burrowing predator
Lytechinus pictus	Sea urchin	0.16	340	19	0.89	2.91	1,2,3,4A,4B, 5A,5B	Epibenthic
Macoma inquinata	Clam	0.08(b)	---	---	---	2.15	2,3,4A,4B	Deposit feeder
Anonyx laticoxae	Amphipod (Crustacean)	---	---	---	---	---	2	Food scavenger
Abarenicola pacifica	Polychaete worm	0.60(c)	---	---	---	---	3	Sediment ingesting

(a) Determined by ERCO 1981

(b) Smith and Carson 1981

(c) Cobalt concentration assumed to be the same as N. virens.

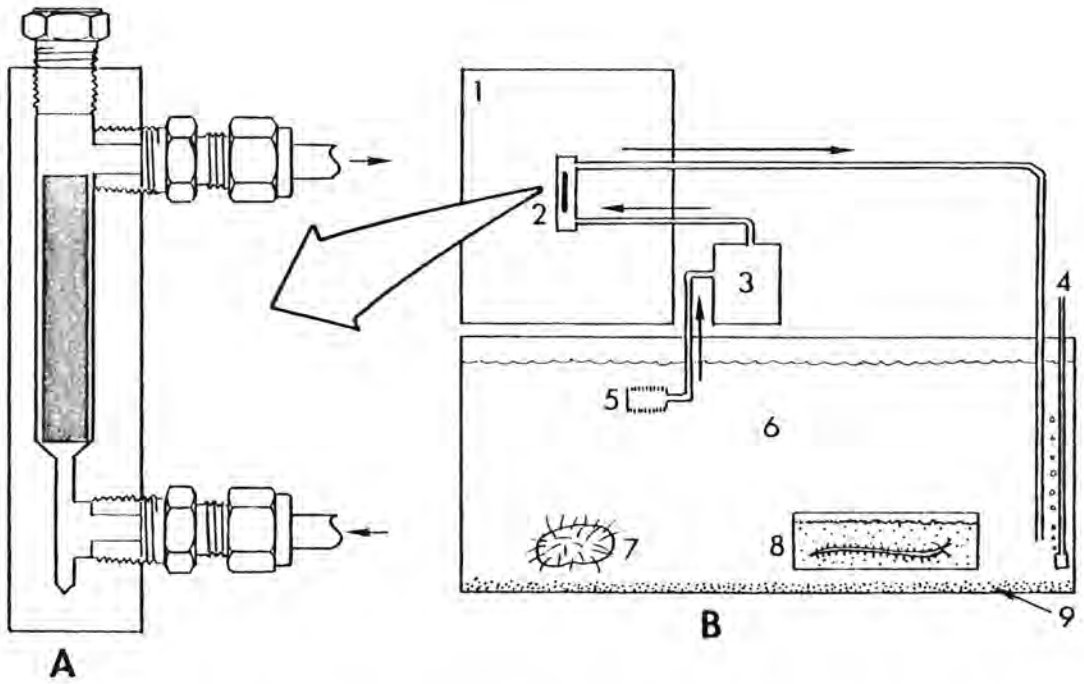

Figure 1. Irradiated stainless steel strip and its holder and closed aquarium system. A. Activated stainless steel strip (shaded) in PBC holder with nylon fittings. Arrows indicate direction of water flow. B. Closed, recirculating aquarium system: (1) aluminum canister with lead shielding, (2) stainless steel strip in holder, (3) pump, (4) aerator, (5) water intake screen, (6) seawater, (7) sea urchin, (8) tray of sediment containing infauna, (9) biological filter gravel.

under anoxic conditions (Table 2). Radioactivity and weight of the stainless steel specimens were determined on July 1, 1979, and the decay corrected to the date of initiation of each experiment. Twenty milliliter aliquots of Sequim Bay sediments were used to determine the radioactivity of the corrosion products used in Experiments 4A and 4B.

In experiments 1 to 3, the stainless steel specimens were contained in a polyvinylchloride holder with nylon fittings that was housed in an aluminum canister filled with lead shot. Intake and outlet tubes that passed through the canister allowed seawater to be pumped into the bottom of the holder, past the radioactive metal strip, and out the top. All experiments used aerated seawater. In Experiments 4A and 4B, a two-hundred milliliter slurry of Sequim Bay sediment containing radioactive corrosion products was used to spike each aquarium. In experiments 5A and 5B, sea urchins (Lytechinus pictus) were exposed directly to radioactive stainless steel strips in vessels of seawater.

Biological, sediment, filter gravel, and seawater samples were counted at least 1000 minutes for ^{60}Co using a Ge(Li) detector coupled to a computer-interfaced Canberra 8180 multichannel analyzer. All samples were background corrected. To include the effect of radioactive decay on biological accumulation, radioactivity determinations were decay corrected to the sample date except that Experiments 5A and 5B were decay corrected to the date of experiment initiation for comparison purposes. Concentrations of ^{60}Co for all sediment and biological samples are expressed in units of pico-curies (pCi) per gram (g) dry weight (dw) of material. For seawater, the ^{60}Co concentration is expressed in picocuries per milliliter (pCi/mℓ). Detection limits for ^{60}Co varied with sample size and counting time.

Total activity for the marine organisms was calculated by multiplying the ^{60}Co concentration by the total dry weight of the samples collected. To increase detection sensitivity in the smaller organisms, samples were grouped to provide additional mass. The number of organisms comprising each sample is indicated by parentheses in the applicable tables. Data for Experiments 1, 5A and 5B were calculated per organism, whereas, the other experiments were based on the number of organisms indicated.

Accumulation of ^{60}Co (%/organism) was calculated by determining the percentage of radioactivity from the stainless steel corrosion products which is incorporated into the organisms, sediment, or seawater. By using the stable cobalt and ^{60}Co concentrations, specific activities were calculated in units of microcuries ^{60}Co per gram of cobalt ($\mu Ci^{60}Co/gCo$). Maximum cobalt-60 concentration factors ($CF_{^{60}Co}$), or ratios of ^{60}Co in the animals to that in the medium in which they were living, were calculated in relation to sediment and seawater concentrations. The sediment $CF_{^{60}Co}$ was based on dry weight concentration, whereas the seawater $CF_{^{60}Co}$ was based on the wet weight

Table 2. Radioactivity of stainless steel specimens utilized for biological uptake experiments.

Experiment Number	Date Initiated	^{60}Co Stainless Steel Specimen Utilized	Activity (7/1/79)	Activity at Experiment Initiation	Weight	Specific Activity(a) ($Ci^{60}Co/g$ Co)	Exposed Surface Area (cm^2)
1	9/17/79	Specimen #1	1.86 mCi	1.81 mCi	1.73 g	1.31	9.69
2	2/5/80	Specimen #1	1.86 mCi	1.72 mCi	1.73 g	1.24	9.69
3	3/31/80	Specimen #2	0.892 mCi	0.808 mCi	1.93 g	0.523	10
4A	11/3/80	Corrosion Products	NA	7.95 nCi	26.1 g(b)	2.77×10^{-5}(b)	---
4B	4/15/81	Corrosion Products	NA	5.99 nCi	25.7 g(b)	2.12×10^{-5}(b)	---
5A	2/14/80	Sample 1 - Specimen #2	0.892 mCi	0.822 mCi	1.93 g	0.532	10
		Sample 2 - Specimen #3	0.854 mCi	0.796 mCi	0.629 g	1.58	3.67
5B	4/17/81	Specimen #1	1.86 mCi	1.48 mCi	1.73 g	1.07	9.69

(a) Specific activity calculated based on nominal 0.08% (weight) of cobalt impurities in Type 347 Stainless Steel. Value is specified as of date of experiment initiation.

(b) Weights and specific activities are based on the Sequim Bay sediment slurry added to each sample and do not include dilution by beach sand. Cobalt concentrations in this sediment were determined to be 11 μg/g (Schmidt, 1982).

concentration. Seawater concentration factors should be calculated in relation to the wet weight of an organism "to reflect the actual role of living aquatic organisms in concentrating chemical elements from aqueous solutions" (Polikarpov, 1966). Wet/dry ratios from Table 1 were used in calculating seawater $CF_{^{60}Co}$ values. Each $CF_{^{60}Co}$ represents an upper limit by assuming concentration of nuclides entirely from one medium. Cobalt-60 accumulation rates

$$\left(\frac{\text{pCi } ^{60}\text{Co uptake}}{\text{g dry weight x year x nCi corrosion products}}\right)$$

were calculated for each experiment by determining the ratio of ^{60}Co concentration in the animals to the total radioactivity of the corrosion products added to the system and normalizing to the amount of activity released for one year. Since ^{60}Co is dependent on water and sediment volume, it should be emphasized that percent accumulation and accumulation rates are useful only for comparisons and partitioning within the same experiment and not between experiments or as generalizations. The total radioactivity released to each system as corrosion products was determined by measuring each portion of the system (seawater, sediments, organisms and filter gravel) for ^{60}Co. Normalizing this data to the area of stainless steel exposed and the length of time of exposure produced estimated corrosion product release rates.

Trace Element Concentrations in Deep Sea Organisms

Four marine organisms typical of the deep-sea regions of interest were analyzed for tissue concentrations of trace elements. The metals cobalt (Co), nickel (Ni), selenium (Se), antimony (Sb), zinc (Zn), iron (Fe), scandium (Sc), chromium (Cr), europium (Eu), terbium (Tb), cesium (Cs), mercury (Hg), hafnium (Hf), thorium (Th), strontium (Sr), tantalum (Ta), manganese (Mn), potassium (K), vanadium (V), aluminum (Al) and titanium (Ti) were analyzed by neutron activatior analysis using a high sensitivity gamma-ray spectrometer that incorporates a high resolution Ge(Li) detector and a NaI(Tl) well detector. This design provided high sensitivity for the measurement of both the coincident and non-coincident gamma-ray emitting radionuclides (Cooper and Perkins, 1971).

The species analyzed include the amphipod, Eurythenese gryllus, from the mid-Pacific gyre (30°N, 158°W at a depth of 5700 meters), the fish, Coryphaenoides armatus, from the Western N. Atlantic (38°N, 70°W at a depth of 3000 meters), and the brittle stars, Amphiophiuria bullata and Ophiomusicum armigorum, from the Western N. Atlantic (38°N, 69°W at a depth of 3600 meters). All dissections were carried out under strict conditions to prevent contamination, such as working in a laminar-flow

hood and dissecting with acid-cleaned quartz knives. Analyses were conducted on samples of sixteen homogenized amphipods, several pooled brittle stars and a single fish specimen. Mean values as well as 95% confidence limits were calculated.

RESULTS

Biological Uptake of ^{60}Co from Neutron-activated Corrosion Products

In all seven experiments, the analysis of control samples yielded no detectable ^{60}Co accumulation in either the animals, sediment, seawater or filter gravel. Since minimum detectable activity varied with sample size, background and type of sample, no particular value is specified as being the lower limit of detection.

Experiment 1. The first experiment used a temperature controlled 95-liter Instant Ocean® aquarium containing two shallow-water species; the echinoid, Lytechinus pictus; and the polychaete, Neanthes virens. L. pictus was chosen because it is epibenthic, usually crawling on substrate surfaces, and echinoids are common in abyssal regions where 53 or more species exist (Heezen and Hollister, 1971).

Polychaetes, of which N. virens is a benthic errantiate representative, are a major infaunal component of deep-sea sediments (Young, 1979). It burrows, but it generally feeds by predation and does not typically ingest large volumes of sediment. Neanthes can be found as deep as 2000 meters.

Four individuals of each species were placed in an aquarium containing 79.9 liters of seawater and 2.995 Kg of sediment. N. virens were maintained in sediment and fed clams, and L. pictus were fed brown macroalgae. The animals were exposed to seawater that continually passed over a strip of radioactive material containing 1.81 mCi of ^{60}Co. Two of each species were removed after 42 days exposure; the others after 94 days. Non-exposed samples maintained by the same method served as controls.

The animals were depurated of sediments for 24 hours, then all biological samples and sediments were freeze-dried and sent with the water and control samples to Knolls Atomic Power Laboratory (KAPL) for ^{60}Co analysis.

The results of this initial three-month experiment are summarized in Table 3. The ^{60}Co accumulation is based on the average of two organisms of each species at each sample time. Although the organisms did accumulate detectable levels of ^{60}Co, each animal only assimilated an average of 0.16% of the total radioactivity released to the system by the corrosion of the radioactive stainless steel. This corresponds to a

Table 3. Accumulation of ^{60}Co by animals, sediment and seawater (Experiment 1)[a].

Species & Material	Exposure Time Days	^{60}Co Concentration (pCi ^{60}Co/g dry wt.)	Total Activity (pCi/organism)	^{60}Co Accumulation (%/organism)	Specific Activity (μCi ^{60}Co/g Co)	^{60}Co Conc. Factors Sediment	Water	^{60}Co Accumulation Rates ($\frac{pCi}{gdw \times y \times nCi}$)
Neanthes virens (2)	42	2.23	6.09	-----	3.71	-----	ND	-----
(2)	94	3.37	2.51	0.154	5.62	6.21	ND	8.06
Lytechinus pictus (2)	42	0.65	3.53	-----	4.03	-----	ND	-----
(2)	94	0.35	2.90	0.179	2.16	0.64	ND	0.837
Sediment	94	ND	1618[b]	99.7[c]	0.23[d]			
Seawater	94	ND	ND	ND	ND			

[a] Data listed as mean value, parenthesis ( ) indicates number of organisms, ND not detectable, broken lines indicate no data.

[b] pCi

[c] %

[d] Sediment (beach sand) contained 2.35 μg/g extractable Co.

final stable cobalt accumulation of 0.0003% (average) of the initial cobalt concentration. The specific activity in terms of ^{60}Co activity per gram of cobalt decreased by a factor of 3.4×10^5 from the initial ^{60}Co specific activity present in the stainless steel to the average specific activity present in the marine animals. Although the concentration of ^{60}Co in the sediment was the same order of magnitude as in the organisms, the large mass of sediment resulted in a total accumulation of 99.7% of the radioactivity released to the system in the form of corrosion products. Since the ^{60}Co concentration in the water was below the minimum detectable, it was not possible to calculate ^{60}Co concentration factors based on the water. It is apparent from the data that N. virens, the sediment dwelling polychaete, accumulated more ^{60}Co per unit weight than the sea urchin L. pictus, probably because the polychaete has a soft, absorptive body surface that was in continuous contact with the contaminated sediments.

The total radioactivity of the corrosion products released to the system during the 94-day exposure was 1.623×10^3 pCi or 8.97×10^{-5}% of the initial activity of the stainless steel specimen. This was accumulated at a rate of 8.06 pCi/gdw/y/nCi for N. virens and 0.84 pCi/gdw/y/nCi for L. pictus. The average rate at which corrosion products were released by the stainless steel was 0.62 $\mu g/y/cm^2$ (Table 13).

Experiment 2. A second more extensive biological experiment using the same ^{60}Co source but additional species examined the amount of corrosion product accumulation over a longer time course. The added species were Anonyx laticoxae, a scavenging amphipod in the same family as the deep-sea Eurythenese gryllus, and a deposit feeding clam, Macoma inquinata. Deposit feeding is common to many animals in eutrophic parts of abyssal regions. Six trays of sediment each containing 2.996 Kg were placed in the aquarium. Samples including marine organisms, seawater and sediment were obtained at 1, 3, 6, 9 and 13 months and handled as described in Experiment 1.

The results of this long-term study are summarized in Table 4. N. virens again accumulated a relatively greater concentration (weight basis) of ^{60}Co than the other organisms (Figure 2). The amphipod Anonyx laticoxae survived only the first month's sampling in the aquarium system before being eaten by the other animals. Lytechinus pictus and the deposit feeding clam, Macoma inquinata, both maintained lower concentrations of ^{60}Co than the sediments as indicated by Figure 2 and their sediment $CF_{^{60}Co}$ values. The total ^{60}Co(nCi) released as corrosion products by the stainless steel specimen is plotted in Figure 3. The bioaccumulation of the ^{60}Co from these corrosion products by the experimental organisms is expressed as a percentage of the corrosion product radioactivity released to the system. It was evident from Figure 3 that the corrosion products were released at a relatively

Table 4. Accumulation of ^{60}Co by animals, sediment and seawater (Experiment 2)[a]

Species & Material	Exposure Time Months	^{60}Co Concentration ($pCi^{60}Co$/g dry wt.)	Total Activity (pCi)	^{60}Co Accumulation (%/organism)	Specific Activity ($\mu Ci^{60}Co$/g Co)	^{60}Co Conc. Factors Sediment	Water	^{60}Co Accumulation Rates ($\frac{pCi}{gdw \times y \times nCi}$)
Neanthes virens (3)	1	ND	ND	ND	ND	ND	ND	ND
(3)	3	0.61	3.47	0.018	1.02	5.35	170	0.370
(3)	6	1.1	5.40	0.014	1.83	2.58	437	0.166
(3)	9	0.73	5.66	0.011	1.22	1.86	203	0.057
(4)	13	1.4	8.08	0.013	2.33	2.32	389	0.085
Lytechinus pictus (3)	1	ND	ND	ND	ND	ND	ND	ND
(3)	3	ND	ND	ND	ND	ND	ND	ND
(3)	6	0.21	2.65	0.007	1.31	0.49	206	0.032
(3)	9	0.23	3.38	0.007	1.44	0.59	158	0.018
(5)	13	0.28	7.87	0.010	1.75	0.46	192	0.017
Macoma inquinata (3)	1	ND	ND	ND	ND	ND	ND	ND
(3)	3	ND	ND	ND	ND	ND	ND	ND
(3)	6	0.20	1.26	0.003	2.5	0.47	266	0.030
(3)	9	0.15	1.38	0.003	1.88	0.38	140	0.012
(4)	13	0.35	3.41	0.006	4.38	0.58	326	0.021
Anonyx laticoxae (1)	1	ND	ND	ND	ND	ND	ND	ND
Sediment	1	0.041	737	26.7[c]	0.017[d]			
	3	0.115	1845	28.0[c]	0.049			
	6	0.43	5620	42.4[c]	0.183			
	9	0.39	5261	30.7[c]	0.166			
	13	0.60	6519	42.8[c]	0.255			
Seawater	3	0.0005[b]	288	3.46[c]	25.0[e]			
	6	0.00035[b]	33.9	2.56[c]	0.60[e]			
	9	0.0005[b]	48.5	2.83[c]	---			
	13	0.0005[b]	48.5	3.18[c]	---			

[a] ND = not detectable, broken lines indicate no data, parentheses ( ) indicates number of organisms.
[b] pCi/ml
[c] %
[d] Sediment (beach sand) contained 2.35 µg/g extractable Co.
[e] Seawater from 3 and 6 months contained 0.2 and 0.58 µg/ℓ total Co, respectively.

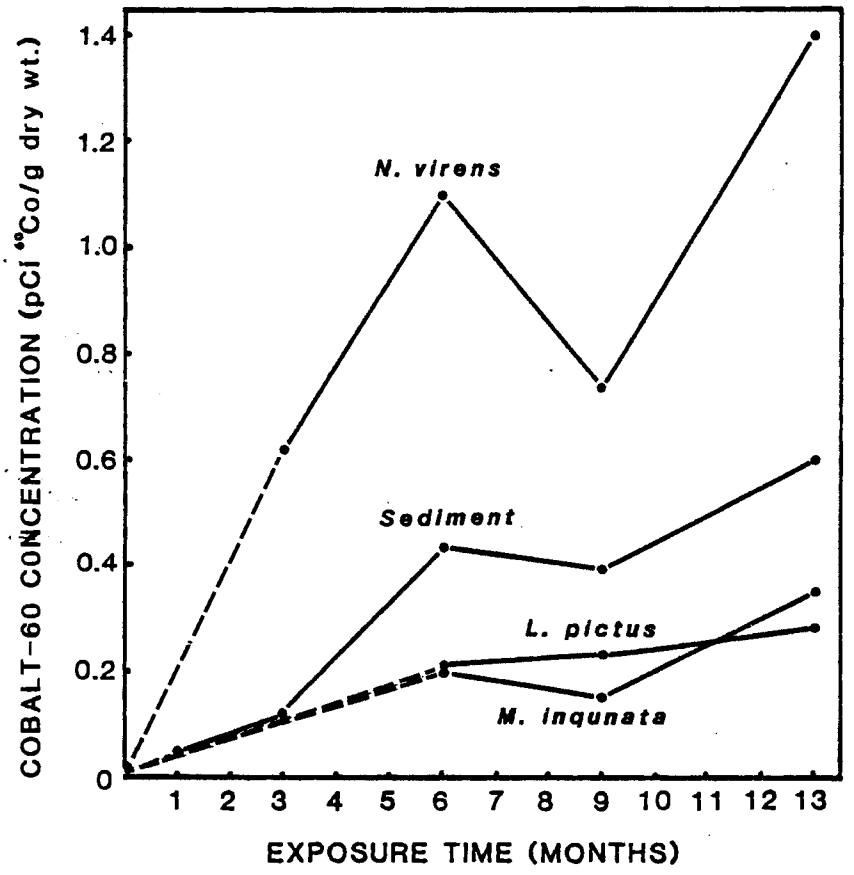

Figure 2. Cobalt-60 concentration in marine organisms and sediments (Experiment 2).

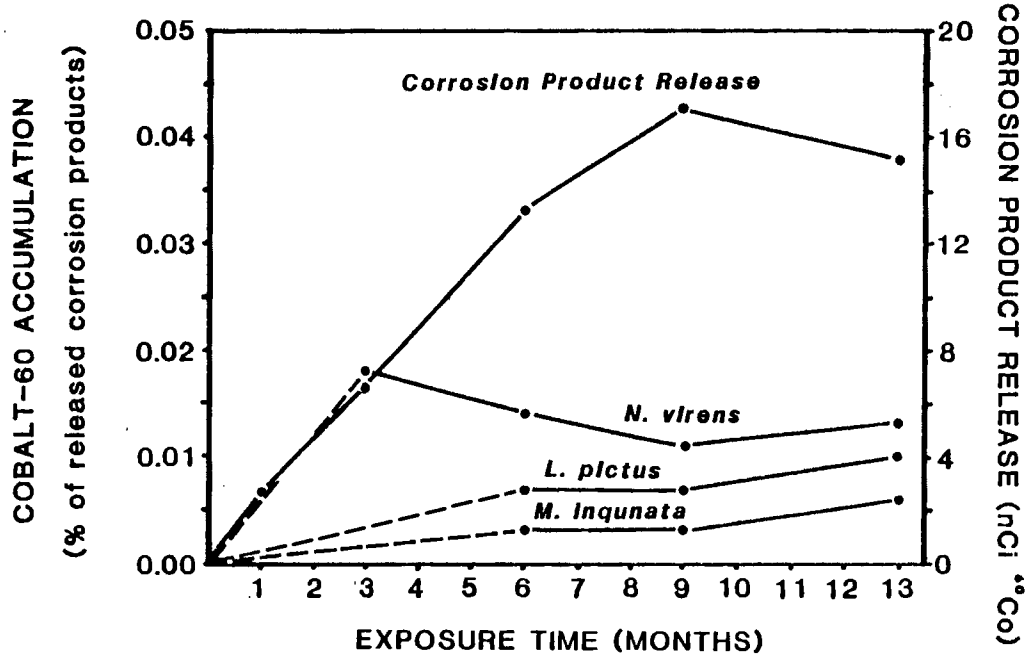

Figure 3. Cobalt-60 corrosion product release and bioaccumulation (Experiment 2).

constant rate until the nine-month sample at which time the release rate decreased to essentially zero. L. pictus and M. inquinata accumulated a higher precentage of the corrosion products after the corrosion release rate decreased. N. virens accumulated a higher percentage of ^{60}Co during the initial stages of corrosion product release.

The ^{60}Co specific activity in the animals sampled after 13 months was a factor of 4.4×10^5 less than the initial specific activity of the stainless steel. The specific activity of the seawater was about a factor of ten higher than that of the organisms. M. inquinata maintained a higher specific activity of ^{60}Co than the other organisms, possibly due to the smaller amount of stable cobalt dilution afforded by its lower cobalt concentration of 0.08 µg/g dry weight (Figure 4), a value similar to typical cobalt concentrations of 0.1 µg/g in marine clams reported by Cole and Carson (1981). The ^{60}Co specific activity in the sediment was lower than that in the organisms (Figure 5) because the ^{60}Co in the sediment was diluted by the higher initial concentrations of nonradioactive cobalt. Also, there may have been some accumulation of ^{60}Co from the seawater.

The ^{60}Co concentration factors relative to water ranged as high as 437. The highest ^{60}Co concentration factor from sediment was 5.35 for N. virens at 3 months. The concentration factors from sediment in the sediment-dwelling animals are more meaningful than those from the water since most of the ^{60}Co accumulation was derived from the sediments and their interstitial waters.

Cobalt-60 accumulation rates were about ten times higher for N. virens than for L. pictus or M. inquinata. The accumulation rates generally decreased with increasing exposure time. The slower accumulation rates for L. pictus and M. inquinata resulted in ^{60}Co concentrations being less than our detection capability until the six-month sample.

As seen in Table 13, the 15.231 nCi of ^{60}Co corrosion products released by the stainless steel resulted in an overall corrosion product release rate of 1.46 µg/y/cm^2. The corrosion products were released at a faster rate during the earlier stages of the experiment.

Experiment 3. This experiment was designed to measure short-term accumulation of radioactive corrosion products in sediment-dwelling animals. L. pictus, M. inquinata, N. virens and Abarenicola pacifica, a sediment-ingesting polychaete were placed in a 95-liter aquarium containing 79.9 ℓ of seawater and 6.85 Kg of sediment. The seawater was pumped past a stainless steel strip containing 0.808 mCi of ^{60}Co. The animals and sediment were sampled weekly for five weeks. The water was sampled at the end of five weeks. Samples were handled as described in Experiment 1.

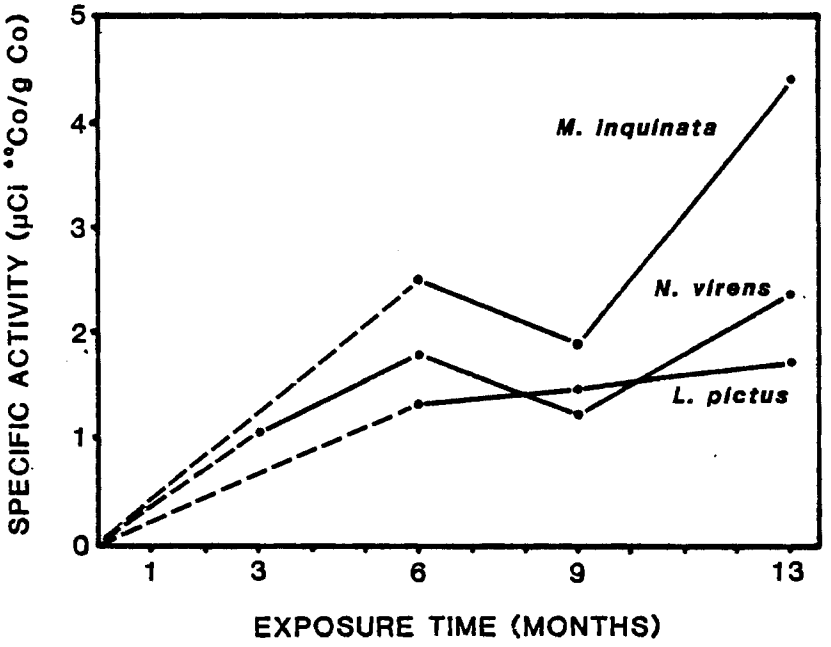

Figure 4. Cobalt-60 specific activity variation (Experiment 2).

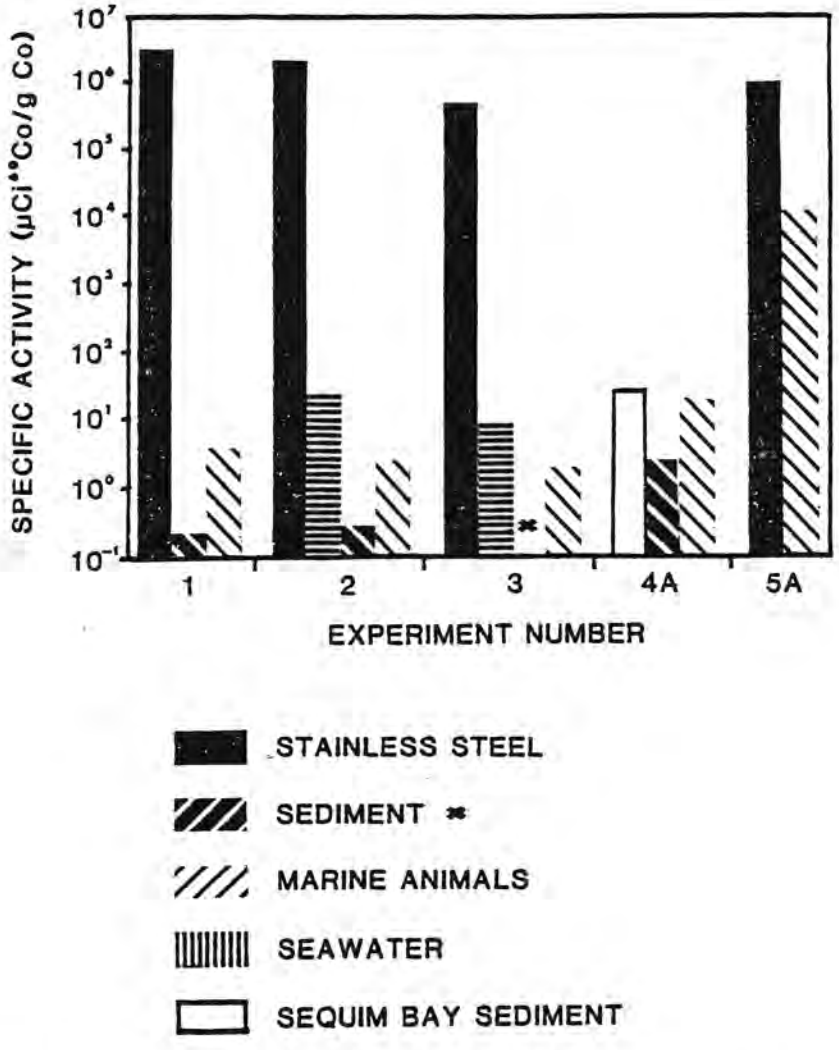

Figure 5. Cobalt-60 specific activity variation (Experiment 2).

The results are presented in Table 5. Weekly samples were obtained of the animals L. pictus, M. inquinata, N. virens and Abarenicola pacifica, a sediment ingesting polychaete. Since the samples from the fourth and fifth week yielded no positive indication of ^{60}Co accumulation, the samples from the first three weeks were not counted. The sediment samples indicated approximately a factor of ten reduction in the ^{60}Co accumulation compared to the other experiments due to the shorter exposure period. The ^{60}Co values reported for the marine organisms in Table 5 should be considered maximum possible values since they are predominantly based on the minimum detectable concentration for the size of the sample. The short-term accumulation of ^{60}Co by marine animals under the conditions of this experiment was so low that tissue levels of ^{60}Co were less than the detection capabilities of the instrumentation.

Experiment 4A. This experiment was designed to determine the ability of marine animals to accumulate ^{60}Co strictly from corrosion products derived from radioactive type 347 stainless steel and deposited in the sediments. Two hundred milliliters (200 mℓ) of a slurry of Sequim Bay clayey silt sediment containing radioactive corrosion products was used to spike two of three glass aquaria, each containing 1700 grams of washed beach sand and five liters of Sequim Bay clayey silt. In one aquarium, the slurry was thoroughly mixed with the beach sand, and then the burrowing animals M. inquinata, N. virens, the sea urchin L. pictus, and seawater were added. In the second aquarium, the slurry was spread on the surface of the beach sand in which the burrowing animals and seawater had already been introduced. The third glass aquarium held sand, animals and seawater without added ^{60}Co and served as a control. Live animals were collected after 22 days and were processed as in Experiment 1.

The resulting ^{60}Co concentrations in the animals (Table 6) were slightly higher than concentrations measured in Experiments 1-3. Over similar exposure times the ^{60}Co accumulation and accumulation rates were about tenfold higher in this experiment, possibly due in part to the higher initial specific activity of the ^{60}Co in the supplemented sediments over that in sediments where the corrosion products were adsorbed from the water.

The only organism having a higher ^{60}Co concentration than the sediment (sediment $CF_{^{60}Co} > 1.0$) was again N. virens and this time only in the mixed sediments environment. The ^{60}Co specific activity in the animals averaged eight times as high as in Experiment 2, much of this due to the relatively high ^{60}Co specific activity of M. inquinata (49.2 $\mu Ci^{60}Co/g$ Co, average). Again, as in Experiment 2, the ^{60}Co specific activity in the sediment was lower than in the organisms because the ^{60}Co in the sediment is diluted by the higher initial concentrations of nonradioactive cobalt. Even though the fraction of ^{60}Co accumulated was much greater for the supplementary radioactive corrosion products

Table 5. Accumulation of ^{60}Co in animals, sediment and seawater (Experiment 3)[a].

Species & Material	Exposure Time Weekly	^{60}Co Concentration ($pCi^{60}Co$/g dry wt.)	Total Activity (pCi)	^{60}Co Accumulation (%/organism)	Specific Activity ($\mu Ci^{60}Co$/g Co)	^{60}Co Conc. Factors Sediment	Water	^{60}Co Accumulation Rates ($\frac{pCi}{gdw \times y \times nCi}$)
Neanthes virens (3)	4	<1.2[b]	<2.4[b]	<0.243[b]	<2.0[b]	<25.0[b]	---	<47.4[b]
(3)	5	<0.41[b]	<1.12[b]	<0.074[b]	<0.68[b]	<6.61[b]	<57.0[b]	<8.44[b]
Lytechinus pictus (3)	4	<0.26[b]	<1.46[b]	<0.148[b]	<1.63[b]	<5.42[b]	---	<10.3[b]
	5	<0.1[b]	<0.62[b]	<0.62[b]	<0.63[b]	<1.62[b]	<34.4[b]	<2.06[b]
Macoma inquinata (3)	4	<0.13[b]	<2.55[b]	<0.258[b]	<1.63[b]	<2.71[b]	---	<5.14[b]
(3)	5	<0.064[b]	<1.02[b]	<0.067[b]	<0.8[b]	<1.03[b]	<29.8[b]	<1.32[b]
Abarenicola pacifica (1)	4	<3.8[b]	<1.90[b]	<0.578[b]	---	<79.2[b]	---	<150[b]
Sediment	4	0.048	329	---	0.020			
	5	0.062	425	84.2[d]	0.026			
Seawater	5	0.001[c]	79.9	15.8[d]	7.69[e]			

[a] Broken lines indicates no data, parentheses ( ) indicate number of organisms.

[b] These numbers represent maximum possible values since ^{60}Co photopeaks were barely indicated after a 17-hour sample count.

[c] pCi/ml

[d] %

[e] Seawater contained 0.13 µg/ℓ total Co.

Table 6. Accumulation of ^{60}Co in animals, sediment and seawater (Experiment 4A).

Species & Material	Type of Supplement	^{60}Co Concentration ($pCi^{60}Co$/g dry wt.)	Total Activity (pCi)	^{60}Co Accumulation (%/organism)	Specific Activity ($\mu Ci^{60}Co$/g Co)	^{60}Co Conc. Factors Sediment	Water	^{60}Co Accumulation Rates ($\frac{pCi}{gdw \times y \times nCi}$)
Neanthes[a] virens (2)	Surface	4.37	18.4	0.11	7.28	0.895	35.8	8.51
(2)	Mixed	6.47	26.2	0.15	10.8	1.31	75.0	12.4
Lytechinus pictus (1)	Surface	0.98	2.13	0.025	6.13	0.20	19.8	1.91
(1)	Mixed	1.17	7.0	0.081	7.31	0.24	33.5	2.24
Macoma inquinata (1)	Surface	3.31	75.5	0.222	41.4	0.68	90.1	6.45
(3)	Mixed	4.55	63.2	0.243	56.9	0.93	176	8.71
Sediment	Surface	4.88	8423[d]	98.9[c]	2.22			
	Mixed	4.93	8510[d]	98.2[c]	2.74			
Seawater	Surface	0.017[b]	85.0	0.998[c]				
	Mixed	0.012[b]	60.0	0.692[c]				

[a] Parentheses ( ) indicate number of organisms.

[b] pCi/ml

[c] %

[d] Total activity measured is greater than that calculated as being added to the system due to incomplete mixing of sediments.

deposited in the sediments than from corrosion products adsorbed from in seawater, the amount in each organism was still only an average of 0.14% of the added radioactivity.

Experiment 4B. Experiment 4A was verified by Experiment 4B. Two hundred milliliters (200 mℓ) of a Sequim Bay clayey silt slurry containing 5.99 nCi ^{60}Co and 25.7 g of sediment was mixed with 5600 grams of beach sand and 32.8 liters of seawater. Four animals each of M. inquinata, L. pictus and N. virens were added to an experimental and control aquarium. The exposure lasted 44 days at which time the animals were removed and processed as in Experiment 1.

The results are presented in Table 7. N. virens again had the highest concentration of ^{60}Co and L. pictus the smallest. Cobalt-60 accumulations of individual organisms averaged about 0.06% of the initial radioactivity added to the system. This is about half that measured in Experiment 4A probably due to the larger aquarium volume in Experiment 4B. The ^{60}Co specific activities were comparable except that N. virens was about twice as high in Experiment 4B whereas M. inquinata was half as much in Experiment 4B as in Experiment 4A.

The concentration factors related to sediment increased by a factor of 6 for N. virens, 3 for L. pictus and 1.3 for M. inquinata. This may be a function of the lower ^{60}Co concentration in the larger sediment volume of Experiment 4B combined with the amount of sediment association by the three species involved.

Accumulation rates of ^{60}Co were lower than in Experiment 4A but were still much higher than for values determined for accumulation of ^{60}Co directly from corrosion of the stainless steel specimens.

Experiment 5A. To assess the capability of marine organisms to accumulate ^{60}Co directly from radioactive corrosion products in seawater, an experiment was conducted utilizing two sea urchins. One L. pictus was placed in each of two 1-liter vessels of seawater containing a radioactive stainless steel specimen. The sea urchins were fed brown algae and removed after 39 days. Specimens and water samples were processed as in Experiment 1 with the exception that the seawater was filtered through 0.45 micron membrane filters which were subsequently analyzed for ^{60}Co.

The results of this experiment in which L. pictus was exposed directly to radioactive stainless steel in seawater are summarized in Table 8. The ^{60}Co concentrations and accumulation rates were more than one thousand times as high as in any of the previous experiments for L. pictus. Likewise, the seawater concentrations were several hundred times as high as in Experiments 1-4. The average ^{60}Co accumulation by the sea urchins was 27% of the activity released by the stainless steel. The ^{60}Co specific activities were about one thousand times as high as in

Table 7. Accumulation of ^{60}Co in animals, sediment and seawater (Experiment 4B).

Species & Material	^{60}Co Concentration ($pCi^{60}Co$/g dry wt.)	Total Activity (pCi)	^{60}Co Accumulation (%/organism)	Specific Activity ($\mu Ci^{60}Co$/g Co)	^{60}Co Conc. Factors Sediment	Water	^{60}Co Accumulation Rates ($\frac{pCi}{gdw \times y \times nCi}$)
Neanthes[a] virens (4)	12.3	42.3	0.086	20.5	8.37	14.3	8.29
Lytechinus pictus (4)	1.11	20.1	0.041	6.94	0.76	3.18	0.748
Macoma inquinata (4)	1.79	34.7	0.071	22.4	1.22	6.94	1.21
Sediment	1.47	8270[d]	67.2[c]	0.615			
Seawater	0.12[b]	3936[d]	32.0[c]	----			

[a] Parentheses ( ) indicates number of organisms.

[b] p Ci/ml

[c] %

[d] Total activity measured is greater than that calculated as being added to the system due to incomplete mixing of sediments.

Table 8. Accumulation of ^{60}Co *L. pictus*, seawater and filtered material (Experiment 5A).

Sample	Type	^{60}Co Concentration ($pCi^{60}Co/g$ dry wt.)	Total Activity (pCi/organism)	^{60}Co Accumulation (%/organism)	Specific Activity ($\mu Ci^{60}Co/g$ Co)	^{60}Co Conc. Factor	^{60}Co Accumulation Rates ($\frac{pCi}{gdw \times y \times nCi}$)
1	*L. pictus*	1972	1.18×10^4	26.6	12,300	21.2	416
	Filtered seawater	32[a]	3.2×10^4	72.1[b]	----	----	----
	Filtered material	----	649	1.46[b]	----	----	----
2	*L. pictus*	1813	1.01×10^4	28.1	11,300	24.9	473
	Filtered seawater	25[a]	2.5×10^4	69.6[b]	----	----	----
	Filtered material	----	847	236[b]	----	----	----

[a] pCi Co^{60}/ml

[b] %

the previous experiments. The average ^{60}Co concentration factor in the sea urchins from seawater was determined to be 23.1.

These results are somewhat higher than would be expected and may be a result of a combination of several factors. First, the absence of sediments and filter material to adsorb the radioactive corrosion products from the seawater allowed much higher seawater concentrations of ^{60}Co to develop. Second, the stainless steel strips were not encased as in Experiments 1-3 thereby removing a barrier to the release of heavy particulate corrosion products. Third and most important, the sea urchins were capable of direct contact with the stainless steel strip thus allowing abrasion of the corrosion layer on the stainless steel by the movement of the sea urchins' calcium carbonate (calcite) spines with possible surface contamination and ingestion of abraded particles.

The average corrosion product release rate of 62 $\mu g/y/cm^2$ was more than one order of magnitude greater than in the previous experiments (Table 13).

Experiment 5B. The final experiment involving bioaccumulation of radioactive corrosion products was designed to determine if the volume of seawater used in the experiments had an effect on ^{60}Co uptake. Four sea urchins (L. pictus) were placed in each of three aquaria containing 4, 16 and 40 liters of seawater, respectively. Each aquarium utilized the same 1.48 mCi radioactive stainless steel specimen (Specimen #1) and were run successively for 25-days each. The sea urchins were not fed in order to eliminate contaminated food (algae) as a source of ^{60}Co. As in Experiment 5A, the radioactive specimens were freely suspended and accessible to the sea urchins to simulate the conditions of radioactive stainless steel immersed in the deep sea. Specimens and water samples were removed at the end of each exposure period and processed as in Experiment 5A.

The data again show a high ^{60}Co concentration in L. pictus as well as a volume effect on ^{60}Co accumulation (Table 9). The ^{60}Co concentration in the organisms decreased by 40% when the volume was increased by a factor of ten, but this relationship was not linear. An exponential relationship between volume and ^{60}Co concentration factor was observed as shown in Figure 6. This may be a result of the organism attempting to achieve a state of saturation with respect to cobalt concentration since the ^{60}Co specific activities were similar for the 4, 16 and 40 liter volumes. The concentration factors from seawater were as high as 775 for the 40 liter aquarium.

The organisms accumulated much less ^{60}Co than in Experiment 5A due to the decreased amount of direct contact with the radioactive stainless steel in the larger aquaria. The average corrosion product release rate decreased with volume for the same reason (Table 13).

Table 9. Accumulation of ^{60}Co L. pictus, seawater and filtered material (Experiment 5B).

Experiment Volume	Type	^{60}Co Concentration ($pCi^{60}Co$/g dry wt.)	Total Activity (pCi organism)	^{60}Co Accumulation (%/organism)	Specific Activity ($\mu Ci^{60}Co$/g Co)	^{60}Co Conc. Factor	^{60}Co Accumulation Rates
4 liters	L. pictus	289	4017	12.0	1,810	18.4	127
	Seawater	5.41[a]	21640	65.1	----	----	----
	Filter	----	7620	22.9	----	----	----
16 liters	L. pictus	169	1817	19.2[c]	1,060	124	261[c]
	Seawater	0.47[a]	7520	79.6[c]	----	----	----
	Filter	----	115[c]	1.2[c]	----	----	----
40 liters	L. pictus	176	2834	47.3[c]	1,100	775	429[c]
	Seawater	0.078[a]	3120	52.1[c]	----	----	----
	Filter	----	35[c]	0.6[c]	----	----	----

[a] pCi/ml

[b] Broken lines indicate no data.

[c] Does not include bottom material contribution.

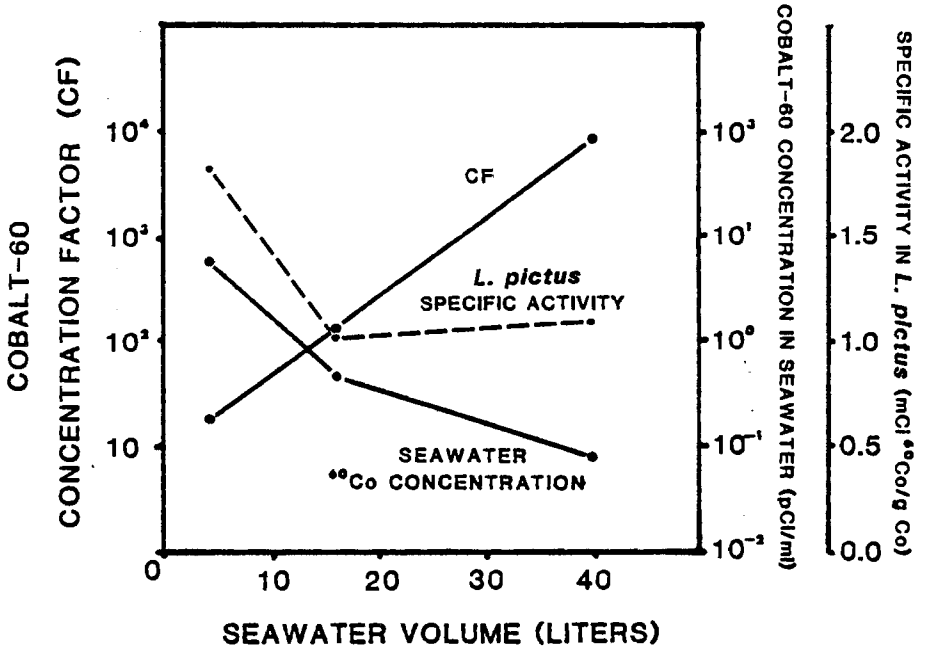

Figure 6. Effect of Volume on Cobalt-60 Concentration Factor $CF_{^{60}Co}$ in L. pictus - (Experiment 5B).

Trace Elements in Deep-Sea Organisms

Table 10 contains the mean trace element concentrations in four marine organisms determined by neutron activation analysis. The mean cobalt concentrations (in μg/g dry weight) for each species are Eurythenese gryllus 0.24, Coryphaenoides armatus 0.28, Amphiophiura bullata 0.13 and Ophiomusium armigerum 0.14. The mean nickel concentrations (in μg/g dry weight) are E. gryllus 1.3, C. armatus 0.78, A. bullata 1.2 and O. armigerum 2.0. Using the cobalt and nickel concentrations in the organisms as well as values for seawater (Table 11) and suitable wet/dry ratios, concentration factors (CF_{Co}) and (CF_{Ni}) may be obtained as shown in Table 12. The cobalt concentration factors are much higher than those determined for ^{60}Co from radioactive stainless steel and/or its corrosion products due to the reduced capability of the marine organisms to utilize the relatively insoluble, adsorbed or refractory oxide forms of cobalt corrosion products (Schmidt 1981).

The cobalt concentration factor are more than six times as great as the nickel concentration factors, reflecting the greater need for cobalt as a trace element essential to biological metabolism. As expected, both cobalt and nickel are concentrated to a much lesser extent in muscle tissue of C. armatus than in the other organs examined.

The cobalt and nickel concentrations in the organisms analyzed in this study are almost a decade lower than the levels determined by Bowen (1966) in various groups of marine animals (Young, 1979). They are significantly closer to the more recent cobalt determinations published by Cole and Carson (1981) and to analyses conducted on the animals utilized in this study by ERCO (1981).

DISCUSSION

Cobalt-60 Concentration

The concentrations of ^{60}Co in sediments and animals from Experiment 1 were higher than those in Experiment 2. Though the corrosion product release rate in Experiment 1 was lower than that in Experiment 2 (Table 13), the fact that it contained no biological filter gravel accounts for a higher fraction of ^{60}Co being concentrated in the sediments and organisms. The large amount of filter gravel in Experiment 2 competed with both the sediment and animals for available ^{60}Co, making it less available to these components of the aquarium system. The seawater of both experiments contained little measurable ^{60}Co. Solubility-absorption reactions for ^{60}Co between the overlying seawater, the interstitial waters, the sediments, and the organisms could not approach equilibrium because of the continuous input of ^{60}Co from the source.

Table 10. Mean trace element concentrations in deep sea organisms[a] (μg/g dry weight).

Organism	Co	Ni	Se	Sb	Zn	Fe	Sc[b]
Eurythenese gryllus (lysianassid amphipod) (16)	0.24 ± 0.02	1.3 ± 0.2	1.62 ± 0.13	0.42 ± 0.006	102 ± 11	79 ± 8	3.29 ± 0.26
Coryphaenoides armatus (rat-tailed fish)							
skin (3)	0.20 ± 0.02	0.54 ± 0.07	2.41 ± 0.25	0.045 ± 0.006	54 ± 6	101 ± 10	11.3 ± 0.9
muscle (3)	0.03 ± 0.00	0.12 ± 0.02	1.24 ± 0.10	0.053 ± 0.007	14 ± 2	10 ± 1	1.27 ± 0.10
liver (2)	0.25 ± 0.03	0.85 ± 0.10	1.44 ± 0.12	0.034 ± 0.004	38 ± 4	45 ± 5	2.05 ± 0.15
gill (1)	0.63 ± 0.08	1.94 ± 0.08	1.6 ± 0.16	0.075 ± 0.009	66 ± 7	895 ± 86	108 ± 8
Amphiophiura bullata (brittle star)							
arms (3)	0.10 ± 0.01	0.95 ± 0.13	0.59 ± 0.05	0.027 ± 0.003	30 ± 3	146 ± 14	24.8 ± 1.7
disks (2)	0.16 ± 0.02	1.4 ± 0.2	0.96 ± 0.08	0.019 ± 0.003	56 ± 6	207 ± 20	36.6 ± 2.6
Ophiomusium armigerum (brittle star)							
arms (2)	0.14 ± 0.02	2.0 ± 0.2	1.37 ± 0.11	0.023 ± 0.003	35 ± 4	131 ± 13	22.8 ± 1.6

[a] Parentheses ( ) indicates number of samples used to determine mean, values are means ± 95% confidence limits.

[b] ng/g dry weight.

Table 10. Mean trace element concentrations in deep sea organisms[a] (μg/g dry weight) (continued).

Organism	Cr	Eu[b]	Tb[b]	Cs[b]	Hg	Hf[b]	Th[b]
Eurythenese gryllus (lysianassid amphipod) (16)	0.37 ± 0.13	1.60 ± 0.35	<6.4	15 ± 3	1.22 ± 0.17	<17.1	<22.5
Coryphaenoides armatus (rat-tailed fish)							
skin (3)	0.24 ± 0.06	1.9 ± 0.2	3.0 ± 0.7	85 ± 13	1.9 ± 0.3	13.3	17 ± 4
muscle (3)	0.32 ± 0.06	0.18 ± 0.05	<0.72	67 ± 11	0.87 ± 0.12	<11.7	<3.3
liver (2)	0.40 ± 0.03	0.37 ± 0.08	1.43 ± 0.51	3.7 ± 0.7	1.47 ± 0.21	<3	33 ± 14
gill (1)	1.2 ± 0.2	17.6 ± 1.8	20.1 ± 2.6	88 ± 14	3.9 ± 0.5	74 ± 10	189 ± 18
Amphiophiura bullata (brittle star)							
arms (3)	0.36 ± 0.08	10.4 ± 1.0	11.2 ± 1.4	18 ± 3	0.16 ± 0.02	16 ± 4	35 ± 5
disks (2)	0.46 ± 0.09	12.1 ± 1.3	13.7 ± 1.7	19 ± 3	0.27 ± 0.04	24 ± 5	49 ± 6
Ophiomusium armigerum (brittle star)							
arms (2)	0.26 ± 0.06	7.9 ± 0.8	10.6 ± 1.4	14 ± 0.8	0.31 ± 0.04	13 ± 4	295 ± 28

[a] Parentheses ( ) indicates number of samples used to determine mean, values are means ± 95% confidence limits.

[b] ng/g dry weight.

Table 10. Mean trace element concentrations in deep sea organisms[a] (μg/g dry weight) (continued).

Organism	Sr	Ta[b]	Mn	K	V	Al	Ti
Eurythenese gryllus (lysianassid amphipod) (16)	605 ± 58	<3.5	4.9 ± 15	0.49 ± 0.15	<94.6	<0.41	<0.3
Coryphaenoides armatus (rat-tailed fish)							
skin (3)	327 ± 33	5.2 ± 1.7	36 ± 1	0.68 ± 0.07	<3.7	0.027 ± 0.03	<0.03
muscle (3)	4.7 ± 0.5	4.1 ± 1.2	2.0 ± 0.5	1.13 ± 0.182	<7.7	<0.26	<0.049
liver (2)	9.5 ± 1.	<0.75	25 ± 4	0.31 ± 0.1	<2.7	<0.61	<0.07
gill (1)	200 ± 20	19 ± 6	207 ± 13	0.48 ± 0.12	<5	0.16	<0.06
Amphiophiura bullata (brittle star)							
arms (3)	250 ± 30	5.1 ± 1.6	19 ± 2	0.23 ± 0.06	<6	0.022 ± 0.008	0.035 ± 0.020
disks (2)	375 ± 35	6.3 ± 2.0	15 ± 2	0.27 ± 0.07	2.7 ± 1.7	0.043 ± 0.006	<0.039
Ophiomusium armigerum (brittle star)							
arms (2)	295 ± 28	3.7 ± 1.2	18 ± 4	0.27 ± 0.07	4.4 ± 2.3	0.019 ± 0.008	0.055 ± 0.023

[a] Parentheses ( ) indicates number of samples used to determine mean, values are means ± 95% confidence limits.

[b] ng/g dry weight.

Table 11. Cobalt and nickel concentrations in seawater summarized from current literature.

Cobalt (µg/l)	Nickel		Summarized from
0.05	1.7		Brewer, 1975
0.021	2.4		Spencer et al., 1970
	0.35		Sclater et al., 1976
	0.47		
	0.58		
	0.58		
	0.32		
	1.8		Chester and Stoner, 1974
	1.2		
0.013	0.25		Brewer et al., 1976
0.014	0.22		
0.019	0.17		
0.023	0.23		
0.023	0.79	Mean Values	

Table 12. Cobalt and nickel concentration factors in deep-sea organisms.

Species	Cobalt Concentration Factor	Nickel Concentration Factor
Eurythenese gryllus	2520	390
Coryphaenoides armatus		
skin	1900	150
muscle	240	28
liver	6500	660
gill	5200	370
Amphiophiura bullata		
arms	1500	415
disks	2400	610
Ophiomusium armigerum	2100	870
Mean	2795	435

[a] Concentration factors were calculated using experimentally determined wet/dry ratios of 4.14 for E. gryllus, 4.5 for C. armatus skin, 5.4 for C. armatus muscle, 16. for C. armatus liver, 5.4 for C. armatus gill and an estimated value of 2.9 for the echinoderms A. bullata and O. armigerum.

Table 13. Corrosion product release rates[a].

Experiment	Exposure Time	Total Corrosion Products Released (nCi)	Corrosion Product Release Rate (μg/y/cm^2)
1	94 days	1.623	0.622
2	1 month	2.756	3.43
	3 months	6.596	2.74
	6 months	13.242	2.75
	9 months	17.145	2.37
	13 months	15.231	1.46
3	4 weeks	0.329	1.02
	5 weeks	0.505	1.25
5A - Sample 1	39 days	44.4	97.6
- Sample 2	39 days	35.9	72.2
5B - 4 liter	25 days	33.2	58.5
- 16 liter	25 days	9.45[b]	16.6
- 40 liter	25 days	5.99[b]	10.5

[a] Measured by detecting ^{60}Co released from stainless steel specimens.

[b] Does not include bottom material.

Experiment 3, the five week exposure, had a larger mass of sediment in which the corrosion products were distributed, probably affording a greater dilution of ^{60}Co than found in Experiment 1. The resultant ^{60}Co concentrations in the marine animals were below the detection capabilities of the instrumentation although some ^{60}Co was detectable in the sediment and seawater.

The sediment and seawater ^{60}Co concentrations were about an order of magnitude higher in Experiments 4A and 4B, in which corrosion products that were formed under anoxic conditions in Sequim Bay sediments were added to the experimental sediments. The resultant ^{60}Co concentrations in the marine animals were more than ten times as high as the concentrations measured in Experiments 1-3 due to the increased ^{60}Co concentrations in the sediments. However, the concentration factors were not much different.

Much higher ^{60}Co concentrations were detected in the sea urchin L. pictus when exposed directly to the radioactive stainless steel in a very small volume of seawater (Experiments 5A and 5B). Some of this ^{60}Co may have been derived from the fractions that Schmidt (1982) had shown could dissolve though the bioavailable form is not known. Most of the ^{60}Co concentrated by L. pictus in Experiment 5A was probably due to the small volume of seawater contained in the vessels and abrasion of the low temperature oxide film by the sea urchins calcareous spines as well as the lack of sediments and filter material to adsorb and remove the corrosion products. Some of the apparent ^{60}Co bioaccumulation may have been contamination on the surface of the sea urchins. Increasing the volume of seawater decreased the ^{60}Co concentrations in the sea urchins in Experiment 5B. This was the result of dilution and reduced contact of the urchins with the stainless steel.

Cobalt-60 Accumulation

The ^{60}Co accumulation is expressed in terms of percent per organism (%/organism) and is a measure of the percentage of radioactivity from the stainless steel corrosion products which was incorporated into the organism. For sediments and seawater it is calculated in terms of the total percentage in that medium.

It should be emphasized that ^{60}Co accumulation as calculated in this study is a function of an organism's mass and ^{60}Co concentration. The highest ^{60}Co bioaccumulation from the stainless steel specimens in the sediment/seawater experiments (Experiments 1-3) was by L. pictus (0.179% see Table 3). This is less than two thousandths of the radioactivity added to the system by the corrosion of stainless steel and less than two billionths of the radioactivity in the stainless steel specimen itself. The highest ^{60}Co accumulation by an organism in the supplemented sediments experiments was M. inquinata (0.222%). In the

small volume seawater experiment (5A), L. pictus accumulated as much as 28.1% of the released radioactivity much of this was due to the experimental conditions previously discussed.

The higher concentrations of ^{60}Co accumulated by N. virens in the sediment/seawater experiments were probably due to interactions with the sediments or interstitial waters. Since this non-calcareous sediment was predominantly aerated almost half of the corrosion products should be refractory, with much of the remaining partitioned as reductant soluble oxides, adsorbed to particulates, or biologically available (Schmidt, 1982). Non-oxygenated sediments result in a soluble fraction of ^{60}Co that can be released to interstitial and overlying waters and may be biologically available, and an increased fraction adsorbed to sediment oxides (Schmidt, 1982). The large, adsorptive epidermal surface of N. virens was probably a route of uptake of soluble corrosion products in the interstitial waters. Another route may have been through the gut by the ingestion of sediment microflora contaminated with ^{60}Co. The greater accumulation of ^{60}Co by N. virens in the sediment/seawater experiments indicates that soft-bodied infaunal animals may potentially accumulate more ^{60}Co than their shelled counterparts, such as clams, presumably because they have a higher ratio of sorptive outer surface to body mass. The early demise of the amphipod Anonyx laticoxae makes it difficult to speculate on uptake rates of burrowing crustaceans, however, since they do possess exoskeletons, the sorptive surfaces are diminished and they should have a lower uptake rate than soft-bodied animals. The sea urchins, which are epifaunal, accumulated the least concentration of ^{60}Co, probably because fewer sorptive surfaces were in contact with sediment and the contact was not continuous because the urchins were frequently on the walls of the aquarium.

Cobalt-60 Accumulation Rates

To normalize the ^{60}Co accumulation by marine animals in terms of size (grams dry weight) and time of exposure (years), ^{60}Co accumulation rates were determined and expressed as picocuries (pCi) ^{60}Co uptake per gram dry weight (gdw) per year (y) exposure per nanocurie (nCi) of added radioactive stainless steel corrosion products.

Neanthes virens accumulated ^{60}Co much faster than any other organism. In Experiments 1-3, its ^{60}Co accumulation rate was about ten times that of the other organisms. The fact that N. virens is in contact with the sediments to a much greater extent than the other organisms supports the hypothesis of greater ^{60}Co uptake from the sediments or interstitial water.

In Experiment 2, the rate of ^{60}Co accumulation in N. virens decreased after three months whereas L. pictus and M. inquinata had much more constant but lower accumulation rates (Figure 7). The overall ^{60}Co

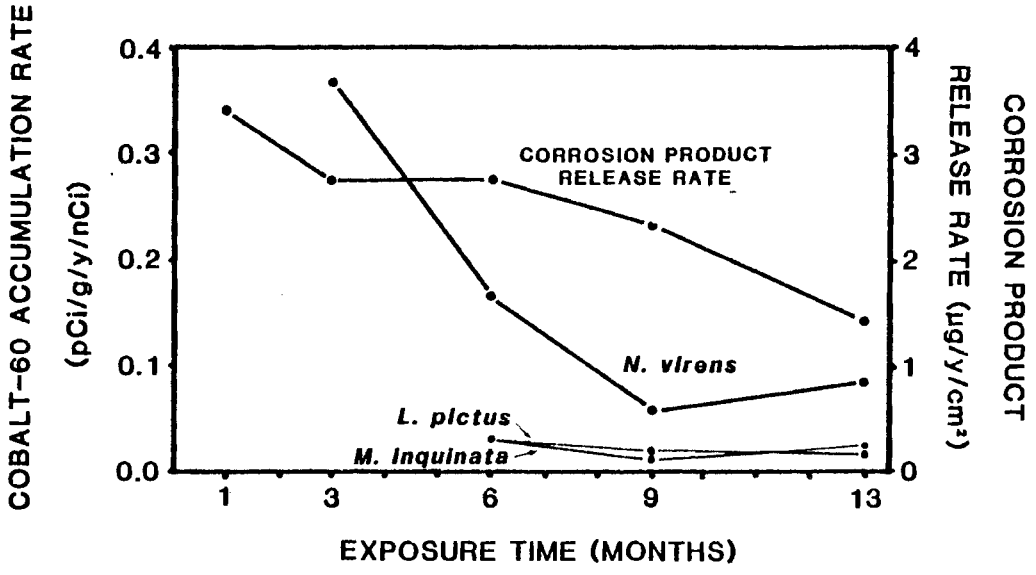

Figure 7. Rate of Cobalt-60 Accumulation and Corrosion Product Release (Experiment 2).

accumulation rates were so low as to preclude the detection of any ^{60}Co that may have accumulated on a short term basis as shown by the five week exposure of Experiment 3.

The ^{60}Co accumulation rates for L. pictus in Experiments 5A and 5B were high. This may have been due to the small vessel sizes and direct contact and abrasion of the stainless steel by the sea urchins, and may reflect contamination as much as an active biological concentration mechanism.

Corrosion Product Release Rates

The corrosion product release rates (Table 13) calculated in these experiments ranged from 0.622 to 97.6 $\mu g/y/cm^2$ (0.0622 to 9.765 $mg/y/dm^2$) with the average corrosion product release rate in the sediment/seawater experiments (1-3) being 1.96 $\mu g/y/cm^2$ (0.196 $\mu g/y/dm^2$). The release rates in Experiments 5A and 5B were much higher due to abrasion by the calcarious spines. Schmidt (1982) calculated corrosion product release rates ranging from 1.38 to 10.58 $\mu g/y/cm^2$ for similar specimens in various sediment/seawater systems. These extremely low corrosion products release rates combined with the physical and chemical forms of the corrosion products reduce the accumulation of ^{60}Co by the marine organisms.

Cobalt-60 Specific Activity

In a comparison of ^{60}Co specific activities of the stainless steel and the various experimental systems the ^{60}Co specific activity in the sediment/ seawater systems (Experiments 1-3) decreased by a factor of almost one million because the ^{60}Co from the stainless steel was isotopically diluted by stable cobalt in the seawater, sediment and marine organisms (Figure 5). Even with direct contact between the organisms and radioactive specimens (Experiments 5A and 5B) the ^{60}Co specific activity decreased more than 100 times. The fact that the organisms achieved a higher ^{60}Co specific activity than the sediments in Experiments 1-4B was not due to a selective concentration of ^{60}Co over stable cobalt but was due to a larger amount of stable cobalt found initially in the sediment.

In the long term exposure of Experiment 2, N. virens maintained a lower ^{60}Co specific activity in spite of its higher ^{60}Co concentration due to its higher stable cobalt concentration. In the seawater Experiment 5B the specific activity was relatively constant over a wide range of seawater ^{60}Co concentrations indicating a possible homeostatic control of cobalt concentrations (Figure 6).

Cobalt-60 Concentration Factors

The capacity of an aquatic organism to accumulate an element (or radionuclide) is expressed by the concentration factor (CF) where

$$CF = \frac{c}{c^1}$$

and c and c^1 are, respectively, the concentration of the element in the organism and the concentration of the element in the aquatic medium (Polikarpov, 1966). The sediment dwellers in this study assimilated ^{60}Co not only from the overlying water but from the sediments, their interstitial waters, detritus, and food. Because nuclides were taken up from so many different sources, "one is faced with the problem of selecting the environ-mental basis for comparing the amounts of radionuclides in the organisms," (Loman, Rice, and Richards, 1971). It can be argued that "comparisons of the amounts of trace elements in an infaunal organism with the amounts in seawater are not representative, since, for many elements even the concentrations of dissolved elements in the interstitial water of sediment differ significantly from those in 'normal' seawater," (Loman, Rice and Richards, 1971), or it can be argued that no matter what environment contributed the assimilated elements, water was the ultimate source, and that comparisons of elements in any aquatic organism with those in the water is valid. In this study, to avoid dispute, concentration factors were calculated in relation to the concentration of ^{60}Co in the sediments and in the seawater. The concentration factor of a radionuclide should correspond to that of its stable element analog providing both isotopes are in the same physical/chemical form. Neanthes virens had a higher sediment $CF_{^{60}Co}$ and generally a higher seawater $CF_{^{60}Co}$ than the other animals.

It is not clear whether the burrowing animals got all their ^{60}Co from the sediments and their interstitial waters, or whether a portion came from the overlying water. Macoma inquinata, for example, may have absorbed some ^{60}Co through the gills from the overlying water during respiration, and the remainder through the mantle and foot tissues from the sediment/interstitial water. Some of the ^{60}Co may have been accumulated from simple contamination. Therefore, both seawater and sediment concentration factors are included for all animals.

Neanthes virens had generally more ^{60}Co although the data was quite variable (Figure 8). This would tend to indicate some type of homeostatic control of cobalt assimilation in which the tissue concentration is independent of ambient levels. Jinks and Eisenbud (1972) reported this occurrence in water for the elements potassium, calcium and manganese. L. pictus and M. inquinata had ^{60}Co concentration factors from sediment that were relatively independent of sediment ^{60}Co concentration. Cole and Carson (1981), in a review of

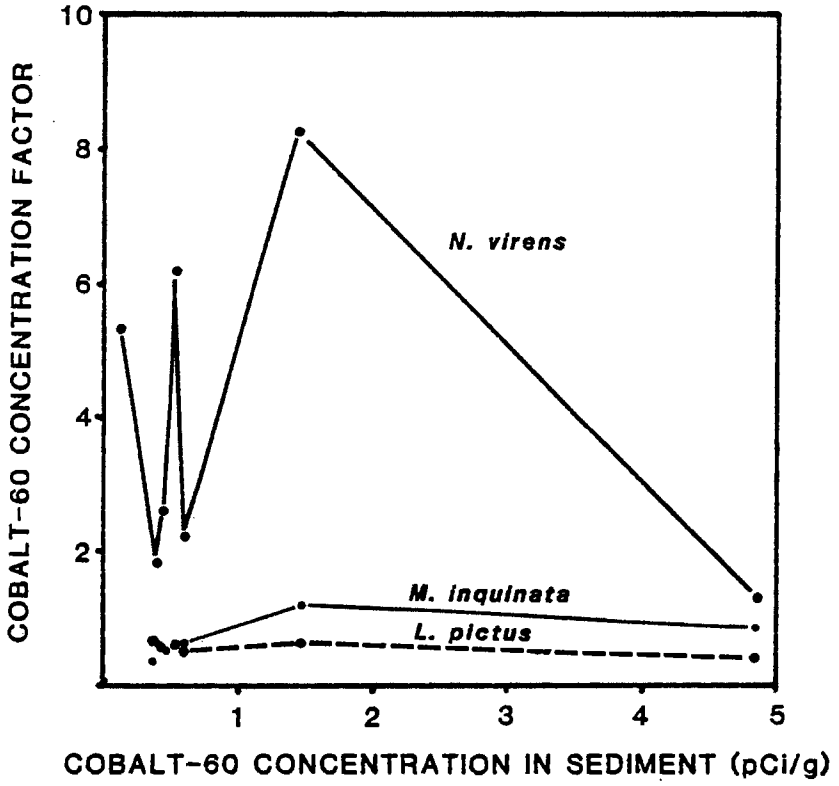

Figure 8. Concentration factors of Cobalt-60 from sediment.

cobalt in the environment, state that homeostatic control of cobalt in higher plants, invertebrates and fish is a major unanswered question at this time. However, Harrison (1973) concluded from studies on the marine clam, Mya arenaria, that cobalt regulation may take place at high concentrations, and that ^{60}Co turnover was fastest in organs with high concentration factors. Also, Feldt and Melzer (1978) found evidence of homeostatic control of cobalt by fish.

Cobalt is a micronutrient used in the formation of cobalamin (Vitamin B-12). Most essential trace elements are regulated, and it is not surprising that that cobalt would be likewise. It is probable that at high water or sediment concentrations of ^{60}Co the tissue concentration will be homeostatically controlled, albeit at higher than normal Co levels.

The increasing amounts of ^{60}Co concentrated by the animals with decreasing seawater ^{60}Co concentrations coupled with the relatively constant specific activities of Experiment 5B again support the homeostatic concentration hypothesis discussed earlier (Figure 6). Similar results in water were obtained by Lentsch, et al. (1971) in which they showed that tissue concentrations of manganese in Hudson River fish remained constant in spite of the large variations in the manganese content of water.

Cobalt concentration factors for marine organisms have been reported by Eisenbud (1973) and are summarized in Table 14. Pritchard (1959) estimated ^{60}Co concentration factors of 10^4 for marine invertebrates and 10^3 for fish. The stable cobalt concentration factors for C. armatus (Table 12) determined in this study are slightly higher but of the same order of magnitude as published values. The ^{60}Co concentration factors determined in this study were generally much lower than published values due to the chemical and physical forms of the corrosion products. Schmidt (1982) showed that exposure of the irradiated stainless steel to aerated seawater for 108 days resulted in corrosion products that were ~30% soluble (< 0.4 μm) and ~70% associated with insoluble suspended particulates. The soluble fraction could be chelated, most of which (~70%) was than associated with organic molecules of $<10^3$ nominal molecular weight. Only a small fraction was ionic. Much (46%) of the particulate fraction was inorganic or weakly complexed. The highest ^{60}Co concentration factor in any of the experiments was 775 relative to water and 8.4 relative to sediment. For the organisms involved in this study, the sediment CF ^{60}Co is more meaningful since the animals were more closely associated with the sediment than the seawater. The maximum sediment ^{60}Co concentration factor for assimilation of the predominantly insoluble corrosion products is a factor of 1000 less than the value for soluble ^{60}Co reported by Prichard (1959).

Table 14. Reported values of cobalt concentration factors for various classes of marine organisms[a].

Group	Concentration Factors Range	Mean
Plants	60-1400	553
Molluscs	1-210	166
Crustacea	300-4000	1700
Fish	20-5000	650

[a] Adapted from Eisenbud (1973).

These experiments have examined the biological fate of ^{60}Co from radioactive stainless steel and its corrosion products under laboratory simulation of ocean conditions. Similar investigations on the physicochemical speciation of corrosion products released from stainless steel upon exposure to seawater and sediments under selected environmental conditions have been accomplished at Pacific Northwest Laboratory by Schmidt (1982). Together these investigations have resulted in a more complete understanding of the fate of radioactive stainless steel deposited in the benthic boundary layer.

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REFERENCES

Brewer, P.G. 1975. Minor elements in seawater. In: J.P. Riley and G. Skirrow (eds.), Chemical Oceanography, Vol. 1. Academic Press, New York.

Brewers, J.M., B. Sundby and P. A. Yeats. 1976. The distribution of trace metals in the western North Atlantic off Nova Scotia. Geochem. Acta 40: 676-696.

Chester, R. and J.H. Stoner. 1974. The distribution of zinc, nickel, manganese, cadmium, copper, and iron in some surface waters from the world ocean. Mar. Chem. 2:17-32.

Cooper, J.A. and R.W. Perkins. 1971. A versatile Ge(Li)NaI(TL) coincidence-anticoincidence gamma-ray spectrometer for environmental and biological problems. BNWL-SA-3972. Pacific Northwest Laboratory, Richland, Washington.

Heezen, B.C. and C.D. Hollister. 1971. In the Face of the Deep. Oxford University Press, New York, 659 p.

Lowman, F. G., T. R. Rice, and F. A. Richards. 1971. Accumulation and redistribution of radionuclides by marine organisms. pp. 161-199. In: Radioactivity in the Marine Environment. National Academy of Sciences.

Polikarpov, G.G. 1966. Radioecology of Aquatic Organisms. North-Holland Publishing Co., Amsterdam, 314 p.

Sclater, F.R., E. Boyle and J.M. Edmond. 1976. On the marine geochemistry of nickel. Earth Planet. Sci. Letters. 31:119-128.

Schmidt, R.L. 1982. Fate of corrosion products released from stainless steel in marine sediments and seawater. PNL-3466 Pts. 1-5., Pacific Northwest Laboratory, Richland, Washington.

Spencer, D.W., D.E. Robertson, K.K. Turekian and T.R. Folsom. 1970. Trace element calibrations and profiles at the Geosecs test station in the Northeast Pacific Ocean. J. Geophys. Res. 75:7688-7696.

Young, J.S. 1979. Food web transport of trace metals and radionuclides from the deep sea: A review. PNL-2960, Pacific Northwest Laboratory, Richland, Washington.

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