NASA TECHNICAL NOTE

NASA TN D-8196

NASA TN D-8196

RADIONUCLIDES IN CHESAPEAKE BAY SEDIMENTS

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NATIONAL AERONAUTICS AND SPACE ADMINISTRATION • WASHINGTON, D. C. • APRIL 1976

1. Report No. NASA TN D-8196	2. Government Accession No.	3. Recipient's Catalog No.
4. Title and Subtitle Radionuclides in Chesapeake Bay Sediments		5. Report Date April 1976
		6. Performing Organization Code 923
7. Author(s) Philip J. Cressy, Jr.		8. Performing Organization Report No. G-7661
9. Performing Organization Name and Address Goddard Space Flight Center Greenbelt, Maryland 20771		10. Work Unit No. 176-52-41-04
		11. Contract or Grant No.
12. Sponsoring Agency Name and Address National Aeronautics and Space Administration Washington, D.C. 20546		13. Type of Report and Period Covered Technical Note
		14. Sponsoring Agency Code
15. Supplementary Notes		

16. Abstract

Natural and manmade gamma-ray emitting radionuclides were measured in Chesapeake Bay sediments taken near the Calvert Cliffs Nuclear Power Plant site. Samples represented several water depths, at six locations, for five dates encompassing a complete seasonal cycle. Radionuclide contents of dry sediments ranged as follows: ^{208}Tl, 40 to 400 pCi/kg; ^{214}Bi, 200 to 800 pCi/kg; K, 0.04 to 2.1 percent; ^{137}Cs, $<$ 5 to 1900 pCi/kg; ^{106}Ru, $<$ 40 to 1000 pCi/kg; ^{60}Co, $<$ 1 to 27 pCi/kg. In general, radionuclide contents were positively correlated with each other and negatively correlated with sediment grain size.

17. Key Words (Selected by Author(s)) Chesapeake Bay, Sediments, Natural radioactivity (Th, U, K), Nuclear fallout (^{137}Cs, ^{60}Co, ^{106}Ru)	18. Distribution Statement Unclassified—Unlimited Cat 45

19. Security Classif. (of this report)	20. Security Classif. (of this page)	21. No. of Pages	22. Price*
Unclassified	Unclassified	20	$3.25

*For sale by the National Technical Information Service, Springfield, Virginia 22161

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BACKGROUND

This document describes a study conducted in cooperation with the Hallowing Point Field Station, Natural Resources Institute, University of Maryland. The purpose was to establish base levels of natural and manmade gamma-ray emitting radionuclides in surface sediments near the Calvert Cliffs Nuclear Power Plant site on the Chesapeake Bay.

Before one can hope to measure the contribution of the power plant to the radioactivity in the Chesapeake Bay, it is clearly necessary to know the nature, amount, and distribution of radioactivity from other sources. Most of the radioactivity in the Chesapeake Bay sediments is due to naturally occurring potassium, uranium, and thorium found in clays and trace minerals. A second component is due to nuclear fallout from weapons testing. A general assessment of radioactivity level can be obtained from conventional gross beta and gross gamma measurements. The low-background system used for the present study permitted precise and specific measurements of the following gamma-ray emitting radionuclides: ^{40}K, ^{60}Co, ^{106}Ru, ^{137}Cs, ^{208}Tl, and ^{214}Bi, the latter two being decay products of ^{232}Th and ^{238}U, respectively.

EXPERIMENTAL PROCEDURE

Samples were taken quarterly, from the sites indicated in figure 1, by personnel of the Hallowing Point Field Station. The abbreviations are Scientists' Cliffs (SC), Long Beach (LB), Flag Pond (FP), Calvert Cliffs Outfall (CCO), Calvert Cliffs South (CCS), and Rocky Point (RP). The Roman numerals I, II, III, and IV in the site designations refer to water depths of about 3, 6, 9 to 12, and 27 m, respectively. Two samples were acquired successively at each site by using an anchor dredge which removes surface sediment to depths on the order of 15 cm. The dredge, while quite satisfactory for the primarily biological studies for which it was intended, is not ideal for chemical or radiochemical sampling. Samples from known depths, such as from cores, are much more informative. However, the extremely close agreement between the duplicate CCO-II samples of May 1972 argues for thorough homogenization of bottom sediments, at least in that area, down to the sampling depth. Five sets of these samples, covering a complete seasonal cycle, were studied in this work.

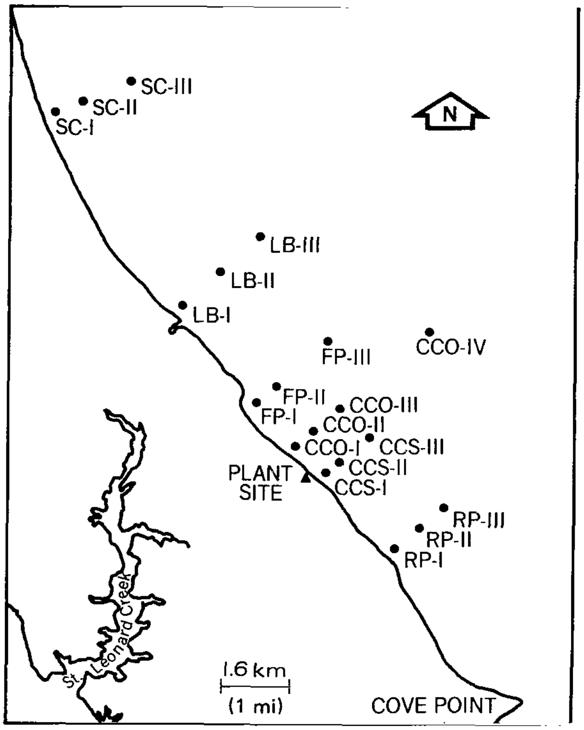

Figure 1. Site locations of the Calvert Cliffs sampling area for the quarterly sampling program of the Hallowing Point Field Station. CCO-IV is no longer visited.

The gamma radioactivities of the samples were measured by nondestructive gamma-gamma coincidence spectrometry. Sediments were dried at 368 K (95°C) at the Hallowing Point Station and ground to coarse powder with a mortar and pestle. Samples ranging from ~ 10 to 300 g were placed in lucite holders, which were mounted in reproducible geometry between two 10- by 10-cm (4- by 4-in.) NaI (Tl) detectors. These sample detectors were placed inside a 50-cm long, 40-cm outer diameter, plastic scintillator, anticoincidence detector. The entire assembly was shielded by 15 cm of steel on all sides and an extra 5 cm of lead on the top. Pulses from the two sample detectors were stored in a 4096-channel, dual parameter, pulse height analyzer as a function of gamma-ray energy, either as single events from either detector or as coincident events resulting from simultaneous detection of gamma-ray emissions in both detectors (time resolution is about 1 μs). Backgrounds were measured by counting dunite powder in the same geometries as the samples. Five radionuclides, ^{208}Tl, ^{214}Bi, ^{60}Co, ^{106}Ru, and ^{134}Cs, were determined from the coincidence spectra: the first two from several coincident gamma-ray peaks and the others after correcting peak counting rates for ^{208}Tl and ^{214}Bi interference. Potassium and ^{137}Cs were determined from the noncoincident spectra after appropriate interference corrections.

Standardized sources of ^{40}K, ^{60}Co, ^{137}Cs, U, and Th (the last two in equilibrium with their daughters) in dunite powder were counted in the various sample geometries for detector calibration. Uncalibrated sources of ^{106}Ru and ^{134}Cs were used for energy calibrations. The identification of ^{106}Ru in samples is based on: (1) excess counting rate in the 0.5- by 0.6-MeV coincidence region, after subtraction of Th, U, and K contributions; (2) the resemblance of the residual spectral pattern to that of the energy calibration source; and (3) recounting CCO-IIA (August 1972) after a 10-month interval and observing decay in this counting region consistent with the 368-day half-life of ^{106}Ru. The identification of ^{134}Cs is based only on its interference in the 0.6- by 0.8-MeV coincidence peak of ^{214}Bi and comparison with the energy calibration source; net ^{134}Cs (half-life = 2.1y) activities were too low for decay measurements. The individual gamma-ray counting yields for these nuclides were calculated from plots of gamma-ray energy versus photopeak counting yield for the sample geometries of interest, were combined for coincidence counting yield assuming no directional correlation of the gamma-rays, and were corrected for gamma-branching fractions to obtain nuclide counting yields.

RESULTS

The results of the analyses of Calvert Cliffs bulk sediments are given in tables 1 through 5. The values for ^{137}Cs ($T_{1/2}$ = 30y), ^{106}Ru, and ^{60}Co ($T_{1/2}$ = 5.26y) have been corrected for radioactive decay from time of sampling to time of measurement. The potassium content (%K) was calculated from ^{40}K assuming normal potassium isotopic composition. Except for K, concentrations are given in picoCuries* per kilogram of dry sediment (pCi/kg). Typical standard deviations for ^{208}Tl, ^{214}Bi, ^{137}Cs, and K were about 5 percent or less.

The Φ values in the first column of tables 1 through 4 were provided by the Hallowing Point Field Station; values for November 1972 samples were not available for this work. The Φ value is not an entirely satisfactory characterization of median grain size distribution. A value of 4 corresponds to fine silt, while coarse sands typically have $\Phi \sim 2$. Even casual inspection of tables 1 through 4 indicates a definite trend of increasing radioactivity with decreasing grain size, that is, with increasing total surface area per unit mass. Unfortunately, this general correspondence cannot be translated into a quantitative relationship sufficient for predictive purposes.

Phi is merely the median grain size; 50 percent of the sample is coarser and 50 percent is finer. The median says nothing concerning the actual distribution about that size. The three sets of data in table 6, at Φ = 2, 2.5, and 3.4, illustrate this. The pair of samples with Φ = 2.5 provides the most dramatic example. LB-I is most probably comprised of essentially one component. CCO-II, however, has at least two components and a great deal more surface area per unit mass. This is reflected in the respective radionuclide contents of the two samples (tables 1 and 3).

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*One picoCurie (pCi) = 2.22 disintegrations per minute (dpm).

Table 1
Radioactivities in Calvert Cliffs Sediments—November 1971

Site	Φ	^{208}Tl pCi/kg	^{214}Bi pCi/kg	%K	^{137}Cs pCi/kg	^{106}Ru pCi/kg	^{60}Co pCi/kg
SC- II*	1.95	354	648	1.65	1211	815 ± 344	10.6 ± 5.4
III	3.55	346	646	1.81	1358	1004 ± 282	24.0 ± 6.3
LB- II	1.98	55	290	0.061	20	< 232	< 2
III	3.18	382	690	1.83	540	< 323	< 13
FP- II	3.39	244	748	1.13	778	571 ± 285	10.0 ± 5.7
III	3.20	369	628	1.96	976	594 ± 314	26.8 ± 7.3
CCO-II	2.53	123	570	0.31	137	265 ±154	3.7 ± 2.1
III	3.35	378	629	1.90	1827	396 ± 288	16.8 ± 6.5
IV	3.40	374	617	1.99	1332	< 780	13.1 ± 5.4
CCS- II	2.05	65	332	0.18	84	151 ± 109	< 2.8
III	3.85	381	613	1.93	1589	< 780	24.0 ± 6.6
RP- II	1.92	40	223	0.082	19	< 230	3.2 ±1.6
III	3.44	358	683	1.83	1212	532 ± 329	13.3 ± 6.5

*Radioactivity not consistent with coarse grain size distribution.

Table 2
Radioactivities in Calvert Cliffs Sediments—February 1972

Site	Φ	^{208}Tl pCi/kg	^{214}Bi pCi/kg	%K	^{137}Cs pCi/kg	^{106}Ru pCi/kg	^{60}Co pCi/kg
SC- II	2.35	99	454	0.31	102	189 ± 129	< 3.1
III	4.01	332	808	1.46	633	< 500	12.2 ± 5.6
LB- II	2.14	60	296	0.96	25	< 157	< 1.9
III	3.55	372	661	1.85	1500	1037 ± 228	22.7 ± 5.6
FP- II	3.72	233	831	1.03	643	589 ± 225	6.1 ± 4.4
III	3.60	382	664	1.92	1490	499 ±246	18.9 ± 5.7
CCO-II	2.17	114	493	0.31	117	410 ± 205	< 5.5
III	4.00	354	731	1.89	1641	329 ± 260	9.5 ± 5.8
IV	3.75	360	559	2.01	1074	< 500	11.8 ± 5.7
CCS- II	1.88	54	344	0.10	81	< 520	< 10.5
III	3.59	403	632	1.81	1321	< 630	21.5 ± 4.9
RP- II	1.60	43	262	0.036	< 6	101 ± 84	< 3.3
III	2.15	49	216	0.084	39	< 154	< 2.3

Table 3
Radioactivities in Calvert Cliffs
Sediments—May 1972

Site	Φ	^{208}Tl pCi/kg	^{214}Bi pCi/kg	%K	^{137}Cs pCi/kg	^{106}Ru pCi/kg	^{60}Co pCi/kg	^{134}Cs pCi/kg
SC- I	2.15	43	294	0.085	12	–	< 1.7	–
II	2.00	94	450	0.196	63	157 ± 65	3.0 ± 1.6	–
III	4.00	339	802	1.56	991	133 ± 64	8.7 ± 3.5	16 ± 7
LB- I	2.51	39	280	0.101	11	–	3.1 ± 1.5	–
II	2.00	70	321	0.044	8	< 45	< 1.2	–
III	4.00	391	706	1.96	849	< 470	9.3 ± 5.0	–
FP- I	2.45	54	379	0.087	16	–	< 1.4	–
II	3.30	221	853	0.93	501	327 ± 55	4.1 ±2.1	–
III	4.00	396	706	1.97	873	< 240	16.5 ± 6.0	–
CCO- I	2.45	40	280	0.095	9	74 ± 23	< 3.3	–
II A1	2.01	129	556	0.342	158	143 ± 16	3.8 ± 1.0	–
II A2	–	131	586	0.351	168	173 ± 33	2.2 ± 1.2	–
II B	–	119	589	0.338	162	115 ± 27	3.3 ± 1.1	–
III	4.00	374	684	1.98	1903	198 ±81	24.0 ± 5.1	24 ± 8
IV	3.90	363	641	2.07	1391	178 ± 65	19.8 ± 4.1	7 ± 6
CCS- I	2.24	41	311	0.084	2	193 ± 67	< 2.3	–
II	2.00	65	355	0.159	55	97 ± 34	< 2.3	–
III	1.98	41	193	0.116	77	55 ± 30	2.2 ± 1.4	5 ± 3
RP- I	2.55	55	336	0.110	20	–	1.8 ± 1.5	–
II	1.95	37	233	0.052	13	–	< 2.5	–
III	3.78	404	743	1.83	1405	163 ± 75	12.0 ± 3.7	15 ± 7

Table 4
Radioactivities in Calvert Cliffs
Sediments—August 1972

Site	Φ	^{208}Tl pCi/kg	^{214}Bi pCi/kg	%K	^{137}Cs pCi/kg	^{106}Ru pCi/kg	^{60}Co pCi/kg
SC- I	2.40	46	294	0.105	10	–	< 1.6
II	3.15	100	536	0.27	72	75 ± 77	< 3.6
III	4.00	358	801	1.64	1328	208 ± 91	5.5 ± 4.3
LB- I	2.46	58	351	0.107	14	–	< 0.3
II	2.31	61	325	0.111	31	–	< 2.6
III	3.39	378	649	1.90	1423	635 ± 178	18.5 ± 5.9
FP- I	2.00	24	180	0.042	6	–	< 3.4
II	3.30	212	779	0.90	459	342 ± 72	< 1.4
III	4.00	404	691	1.98	1343	533 ± 201	20.4 ± 6.1

Table 4 (Continued)

Site	Φ	^{208}Tl pCi/kg	^{214}Bi pCi/kg	%K	^{137}Cs pCi/kg	^{106}Ru pCi/kg	^{60}Co pCi/kg
CCO- I	2.63	55	363	0.132	19	32 ± 24	2.2 ± 1.2
II	2.81	145	712	0.544	109	66 ± 39	1.5 ± 1.5
III	3.56	402	776	1.94	1532	767 ± 97	19.5 ± 5.5
IV	4.00	383	638	2.08	1414	109 ± 58	20.5 ± 3.7
CCS- I	2.55	60	385	0.139	22	–	< 3
II	2.01	60	346	0.140	59	39 ± 27	< 3
III	4.00	402	700	1.87	1446	266 ± 87	12.4 ± 4.8
RP- I	2.50	81	446	0.179	32	79 ± 66	2.3 ± 1.5
II	1.95	45	294	0.040	3	–	< 1
III	4.01	394	787	1.83	1505	88 ± 68	10.7 ± 3.7

Table 5
Radioactivities in Calvert Cliffs
Sediments–November 1972

Site	^{208}Tl pCi/kg	^{214}Bi pCi/kg	%K	^{137}Cs pCi/kg	^{106}Ru pCi/kg	^{60}Co pCi/kg
SC- II	57	246	0.12	44	< 137	< 3.2
III	362	695	1.83	1666	584 ± 194	25.8 ± 7.0
LB- II	48	193	0.050	13	< 46	< 3.3
III	378	657	1.83	1390	285 ± 153	6.6 ± 4.5
FP- II	233	803	1.02	558	222 ± 133	3.1 ± 2.3
III	389	733	1.92	1172	443 ± 179	13.3 ± 4.7
CCO- II	141	713	0.38	98	116 ± 88	< 3.0
III*	358	739	1.92	1721	644 ± 175	14.5 ± 5.7
CCS- II	73	362	0.183	55	< 179	< 3.6
III	387	664	1.89	1297	488 ± 197	19.3 ± 6.9
RP- II	45	254	0.110	16	60 ± 38	1.0 ± 0.9
III	354	671	1.82	1391	< 420	21.6 ± 5.7

*^{134}Cs: 18 ± 13 pCi/kg.

The other two sets in table 6, while less extreme, are still typical of the grain size characterization problem. An additional aspect suggested by these samples is the possibility that the sieving process did not completely segregate the grain sizes. Fine fractions especially tend to resist sieving efforts.

Table 6
Grain Size Distributions and Φ Values

Sample	Date	Φ	> 1 mm	> 0.5	> 0.25	> 0.125	> 0.0625	< 0.0625
CCO-III	11/71	3.35	0	0.8	16.2	14	50	19
CCO-IV	11/71	3.40	0	1.9	16.1	25	17	40
RP-III	11/71	3.44	0	0.7	9.3	24	36	30
LB-1	5/72	2.51	0.2	0.2	5	86.6	7.5	0.5
CCO-II	11/71	2.53	5.0	6	19	37	21	12
LB-II	11/71	1.98	1.2	6.8	48	40	3.4	0.6
CCO-II	5/72	2.01	8.5	11	30.5	30	16	4
SC-I	5/72	2.15	0.3	0.5	37.2	60.2	1.5	0.3

BULK SEDIMENT SAMPLES

Inspection of the data in tables 1 through 5 suggests several possible correlations. The accompanying figures may help to clarify these relationships.

In figure 2, ^{60}Co and ^{137}Cs, both nuclear fallout products, are plotted against K for all samples. Since both ^{60}Co and ^{137}Cs are adsorbed onto sediment grains, a relationship here implies either (1) K is also adsorbed, or (2), which is more likely, the primary site for adsorption is a K-rich mineral. However, the tendency for ^{137}Cs and especially ^{60}Co to vary somewhat randomly at K = 1.8 to 2.1 percent implies that concentration of this hypothetical K-rich species is not the only parameter of importance. Grain size, specifically of the adsorbing species, is a logical candidate for a second parameter. As mentioned above, the Φ value is too crude a measure of grain size to substantiate this possibility.

As a first approximation, ^{137}Cs content in these sediments can be estimated (±10 to 20 percent) from K content by the least squares regression curve shown in figure 2. The open circles in the figure represent data from CCO-IV, LB-III, and FP-III on different dates. These data do not appear to conform to the same trends as data from other sites and were not included in the correlation calculations. These three sites may be comprised of different sediment/silt materials than the others.

In figure 3, ^{214}Bi and ^{208}Tl are plotted against K for all sediment samples. In the Chesapeake Bay sediments, ^{208}Tl is positively correlated with K content in a fairly smooth relationship. Like ^{60}Co and ^{137}Cs, ^{208}Tl also varies appreciably and randomly at K > 1.8 percent. The ^{214}Bi variation with K is a much more complex situation. It is likely that several sources of ^{214}Bi are contributing to the overall pattern.

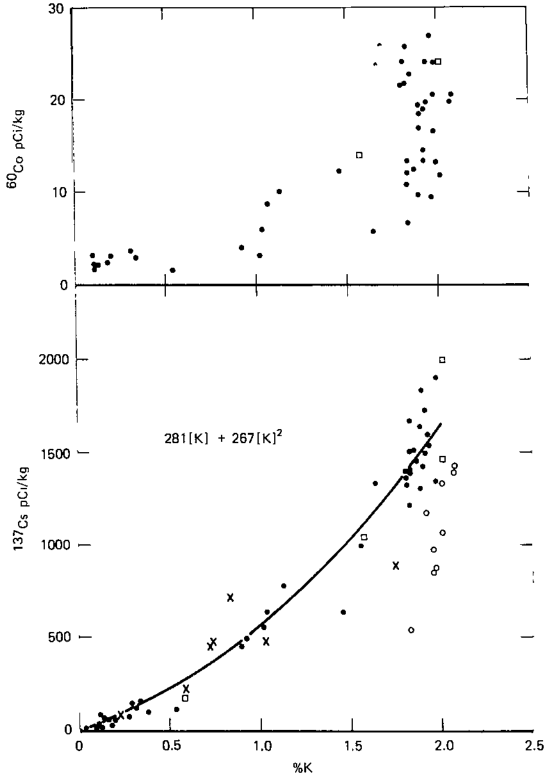

Figure 2. ^{60}Co and ^{137}Cs as a function of K-content in sediment samples. Solid and open circles are Calvert Cliffs bulk samples from tables 1 through 5 (see text). The separated fractions of CCO-IIA (table 7) are denoted by X, and Potomac River sediment data (table 8) are plotted as boxes (□).

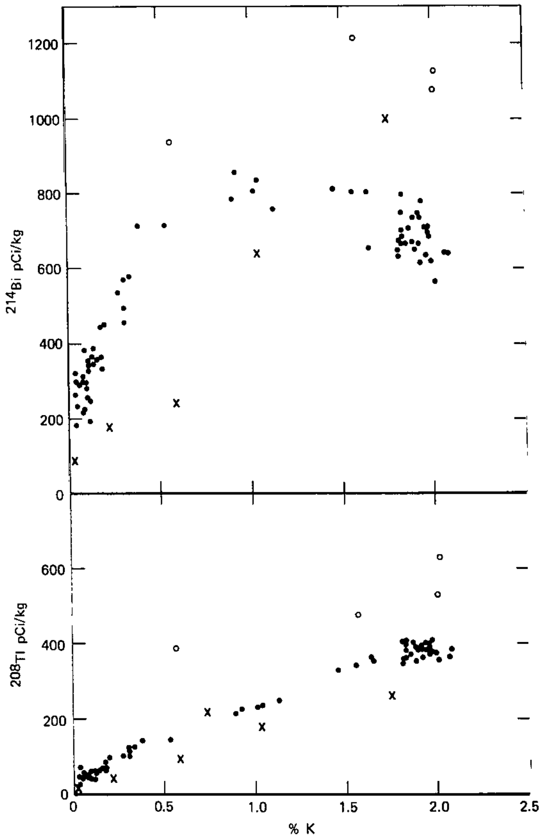

Figure 3. ^{214}Bi and ^{208}Tl as a function of K content. Solid points are from bulk Chesapeake Bay sediments (tables 1 through 5); X's indicate CCO-IIA separates (table 7); open circles (○) indicate Potomac sediment data (table 8).

In figure 2, fallout radionuclide concentrations were compared with a natural radionuclide indicative of an adsorbing species. In figure 4, two fallout radionuclides, ^{60}Co and ^{137}Cs, are intercompared. The samples are designated as "before" and "after" tropical storm Agnes (July 1972). The pre-Agnes data are consistent with a linear relationship between ^{60}Co and ^{137}Cs, although the statistical uncertainties in ^{60}Co measurements could easily conceal a more complex pattern. In the lower part of figure 4, post-Agnes data are plotted. The dashed line is the pre-Agnes trend. Although the ^{137}Cs activities have clearly changed (only one point is between 200 and 1100 pCi/kg), the line is still a reasonable representation of the ^{60}Co variation. Such a general correlation between two fallout nuclides of different half-lives implies that they are fairly uniformly mixed throughout the sampling depth.

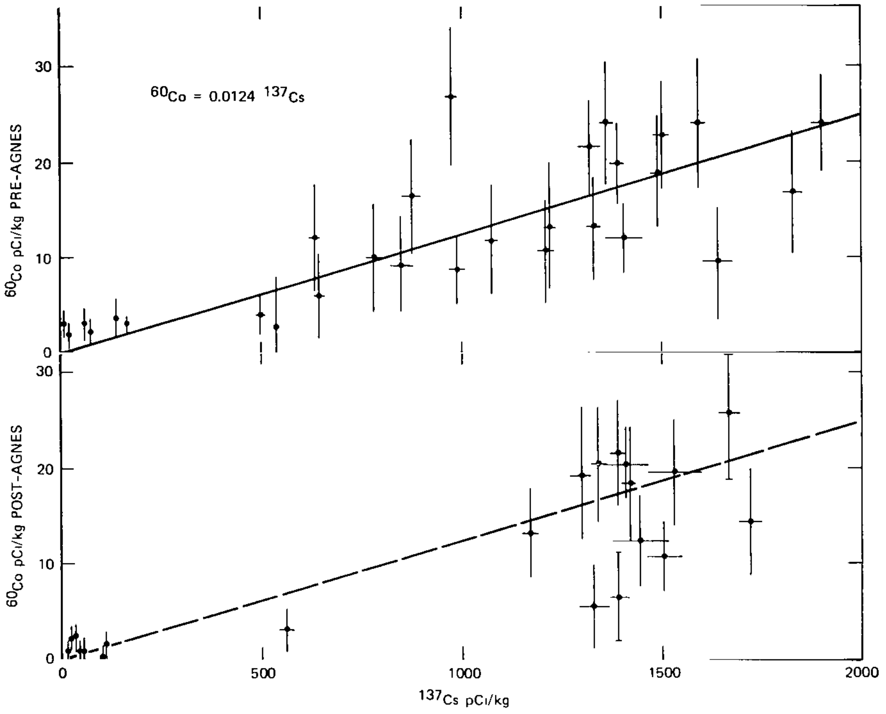

Figure 4. ^{60}Co as a function of ^{137}Cs in Chesapeake Bay sediments, before and after tropical storm Agnes (July 1972). Error bars are one σ standard deviations from counting statistics.

In figure 5, ^{106}Ru is plotted against ^{137}Cs. In the samples taken before Agnes, ^{106}Ru is poorly or not at all correlated with ^{137}Cs. The May data, immediately before Agnes, have a noticeably flat distribution, indicating ^{106}Ru was primarily a surficial deposit at this time, relatively uniform at all water depths. The February 1972 and November 1971 samples include some higher ^{106}Ru results, but with no apparent pattern and with large statistical uncertainties. With some obvious exceptions, the post-Agnes data describe a positive

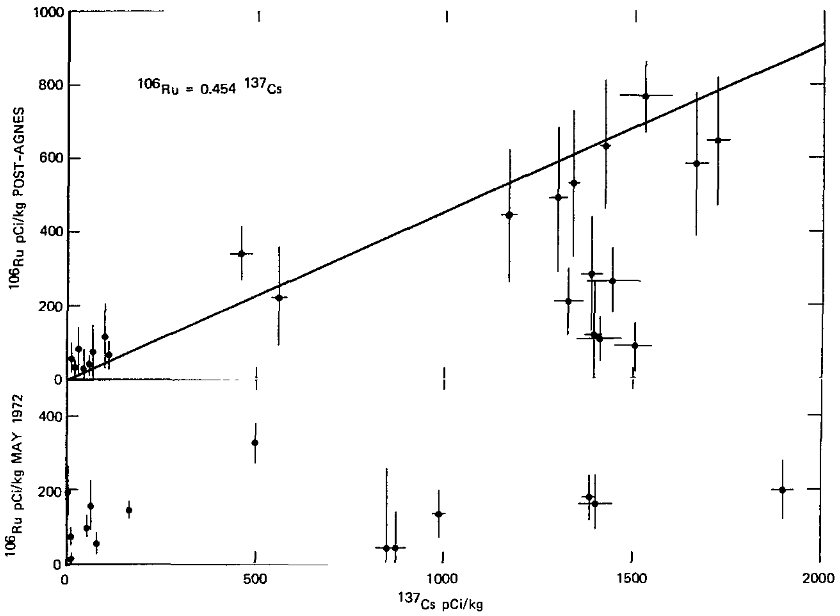

Figure 5. ^{106}Ru as a function of ^{137}Cs, in May 1972 and after Agnes. Error bars are one σ standard deviations.

correlation between ^{106}Ru ($T_{1/2}$ = 368 d) and ^{137}Cs ($T_{1/2}$ = 30 y). Redistribution of ^{106}Ru throughout the sampling depth has apparently occurred, presumably as a result of tropical storm Agnes.

SEASONAL TRENDS AT EACH SITE

Figures 6 through 18 illustrate the trends in radionuclide concentrations at each site over five dates covering a full season. In general, the patterns of change of the different nuclides reflect one another at each site. Many apparent inconsistencies in ^{106}Ru trends may be due to large (⩾ 40 percent) statistical uncertainties. Several specific observations may also be drawn.

- The FP-II site consistently has an unusually large Φ, that is, a large proportion of fine material, for a II site at 6 m. Radionuclide contents are consistent with this.
- The radioactivity data for SC-II, November 1971, are inconsistent with a Φ of 1.95. Either the reported Φ is wrong, or the sample was mislabeled. It is also surprising to observe virtually no change in radioactivity from February (Φ = 2.35) and May (Φ = 2.0) to August (Φ = 3.15) 1972.
- Phi for RP-II is uniformly low. The relative K variation is considerable, but the absolute amounts of implied K-rich material are too small to seriously affect radionuclide contents.

- The SC-III site experienced a dramatic drop in K, ^{137}Cs, and ^{106}Ru without any corresponding change in Φ, prior to February 1972. This could be explained by a deposition of low ^{137}Cs - ^{106}Ru "silt."
- At LB-III, ^{137}Cs, ^{60}Co, and ^{106}Ru co-vary, but seem independent of K. Again, this suggests a mixture of "clean" and "hot" sediments.
- At FP-III, ^{137}Cs and ^{106}Ru show behavior similar to that at LB-III. At both sites, one major change occurred after tropical storm Agnes. In this respect, Agnes' effect appears to have not been more serious than the effect of an earlier, unidentified event.
- At CCO-III, ^{106}Ru increased substantially after Agnes, while ^{137}Cs actually decreased from a level unusually high for this part of the Chesapeake Bay. Sediment redistribution seems the most likely cause.
- A complete change in the nature of the sediment occurred at CCS-III between February and May 1972. This is obviously not associated with Agnes and was probably caused by dredging operations in the area at that time. A similar event appears to have occurred at RP-III between November 1971 and February 1972. In both instances, sediments returned to their customary state before the next sampling period.

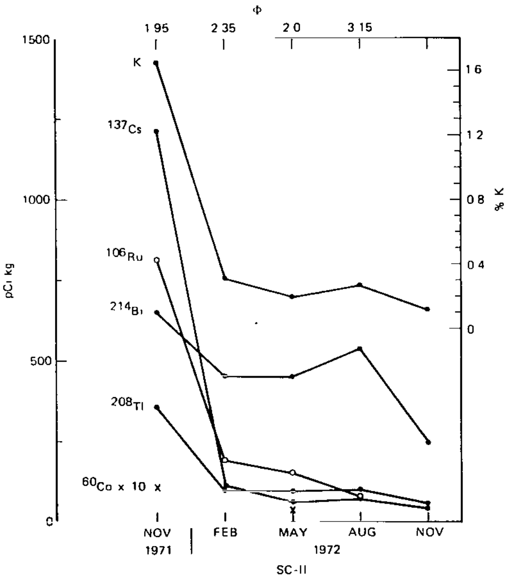

Figure 6. Radionuclide concentrations at SC-II in five successive quarterly samples. The Φ value reported for each sample is given along the top of the figure. Open circles here and in later figures denote ^{106}Ru; ^{60}Co, multiplied by 10 for easier plotting, is denoted by X.

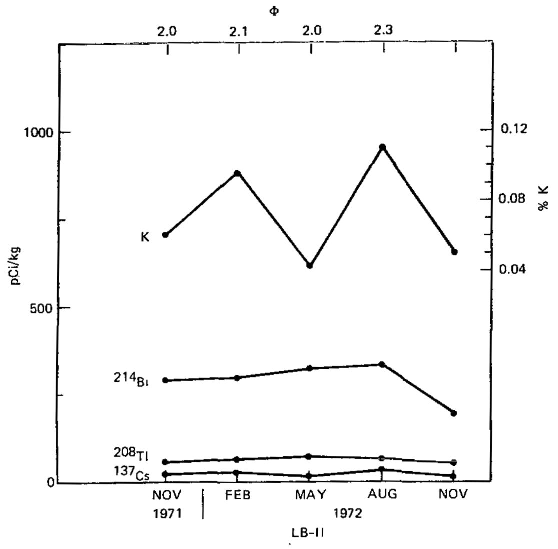

Figure 7. Radionuclide contents at LB-II in five samples.

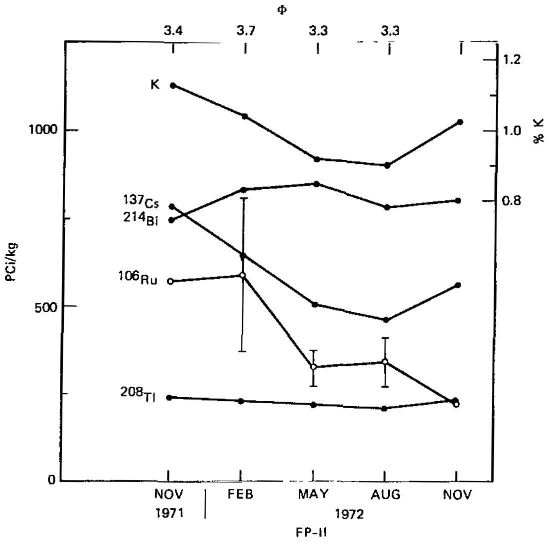

Figure 8. Radionuclide contents at FP-II in five samples. Error bars are attached to ^{106}Ru points here and in the other figures when the standard deviations are less than about 30 percent.

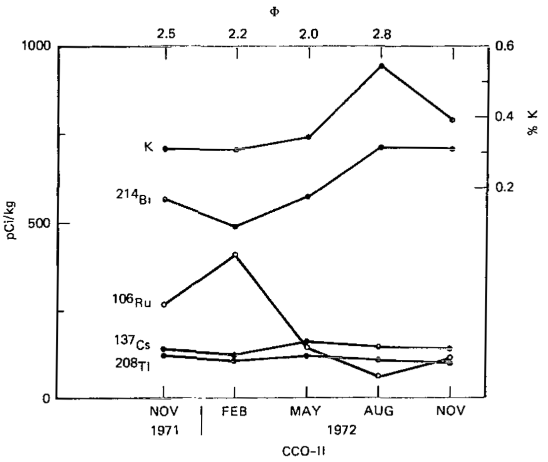

Figure 9. Radionuclide contents at CCO-II in five samples. Clustering of the data did not permit drawing the ± 21 pCi/kg uncertainty in the May ^{106}Ru point.

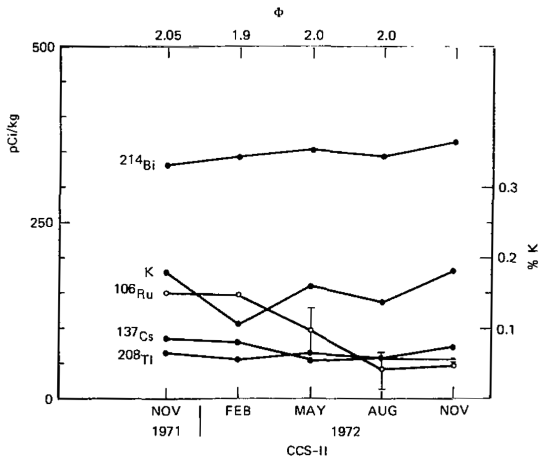

Figure 10. Radionuclide contents at CCS-II in five samples.

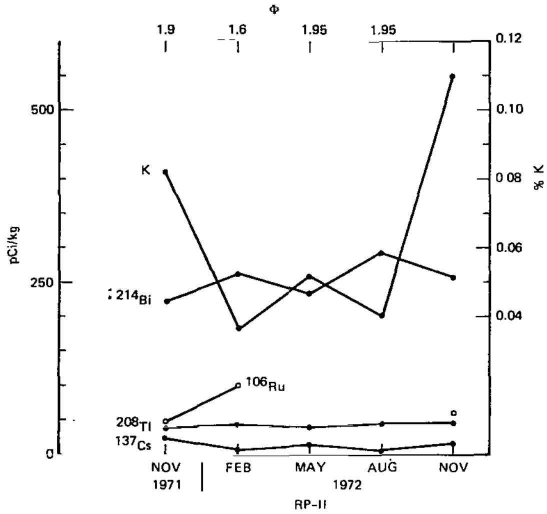

Figure 11. Radionuclide contents at RP-II in five samples.

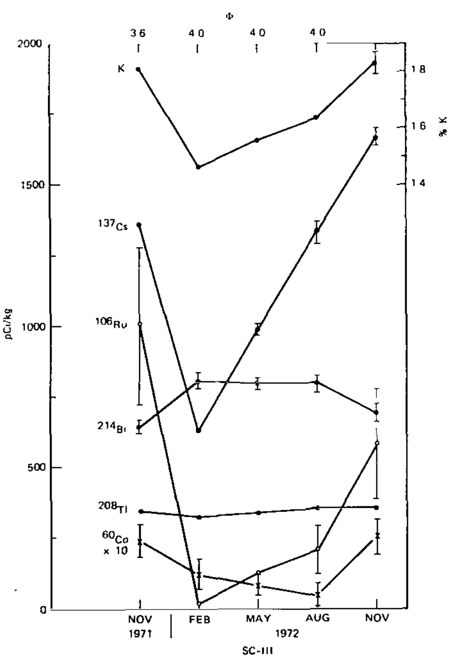

Figure 12. Radionuclide contents at SC-III in five samples. One σ uncertainties in ^{60}Co data are indicated by error bars in this and later figures, except where the uncertainties are so large as to clutter the figure. Uncertainties in K, ^{214}Bi, and ^{208}Tl are also indicated whenever they are appreciably greater than the points themselves.

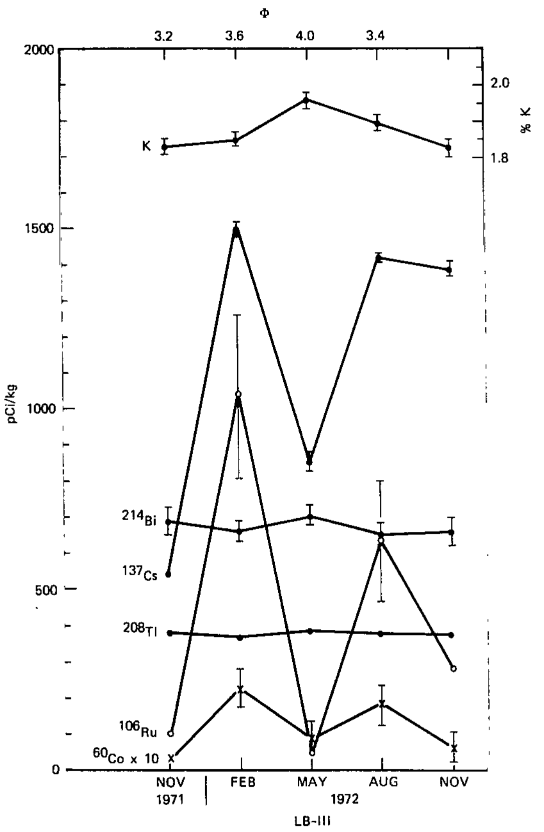

Figure 13. Radionuclide contents at LB-III in five samples.

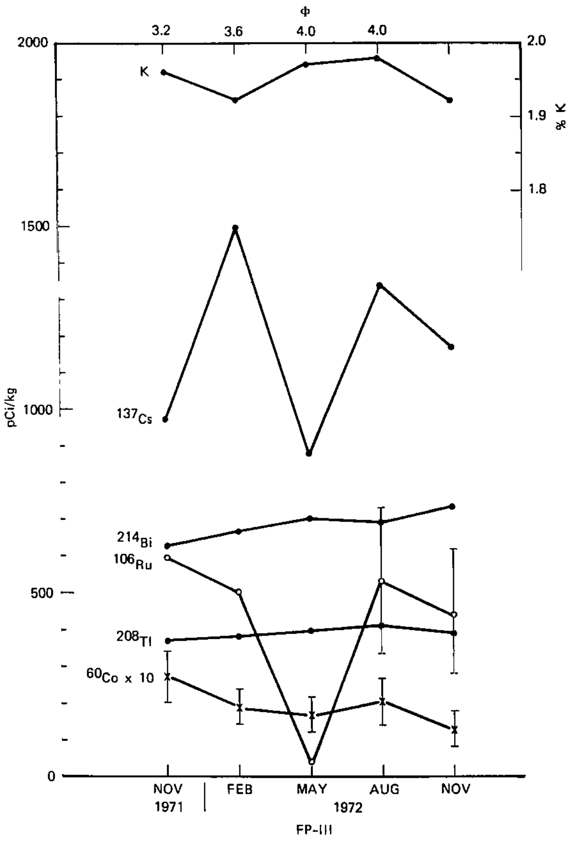

Figure 14. Radionuclide contents at FP-III in five samples.

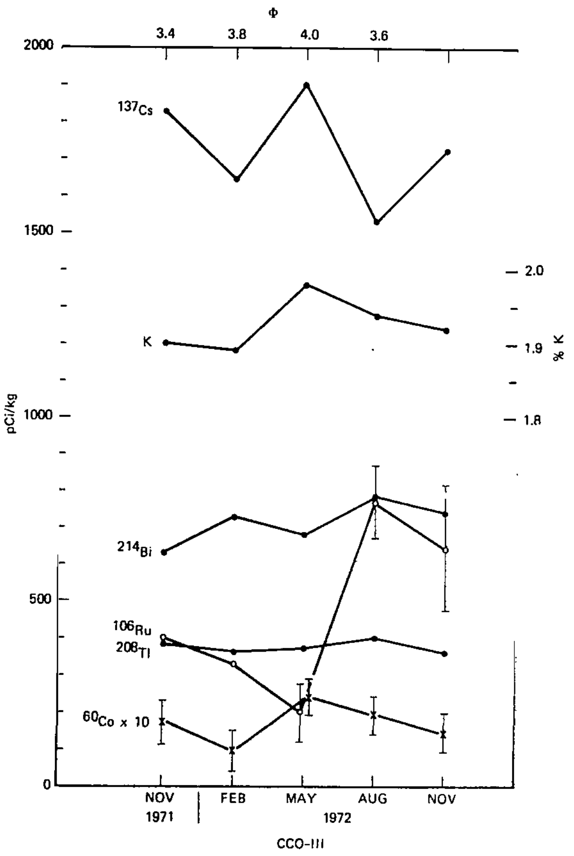

Figure 15. Radionuclide contents at CCO-III in five samples.

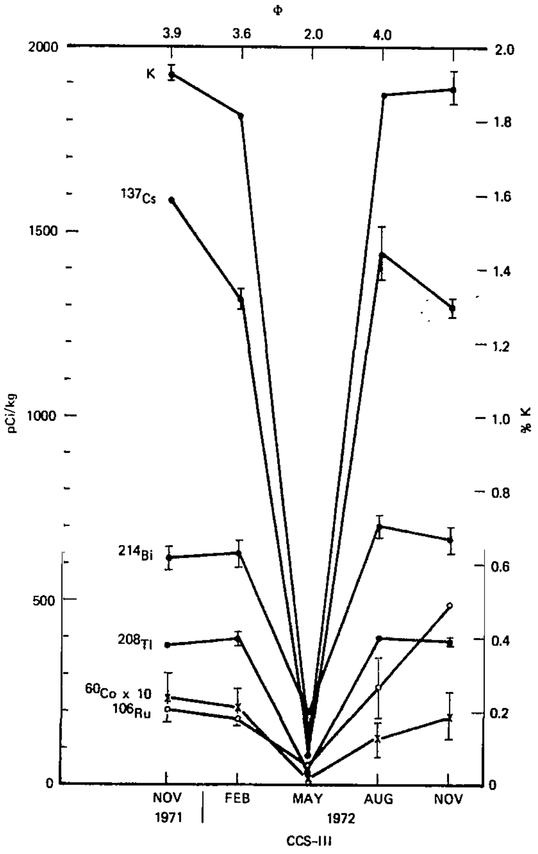

Figure 16. Radionuclide contents at CCS-III in five samples.

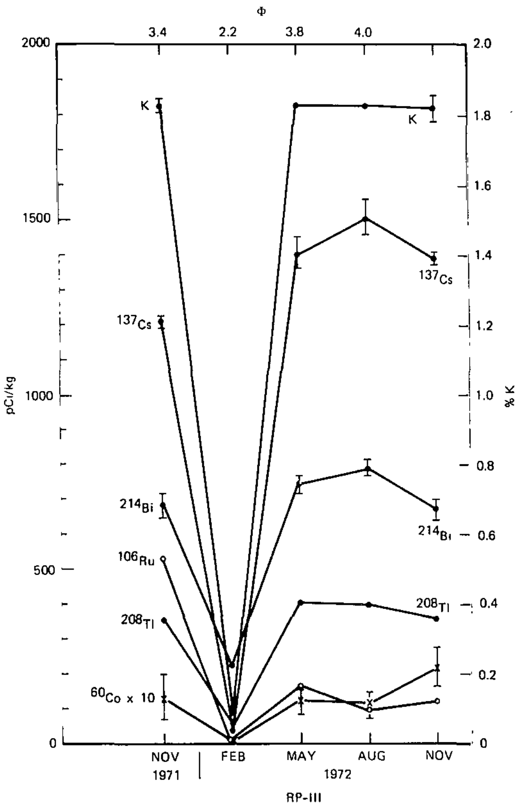

Figure 17. Radionuclide contents at RP-III in five samples.

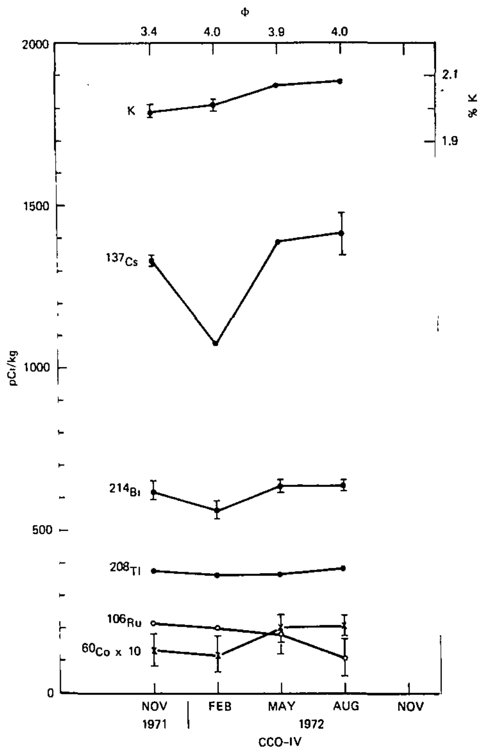

Figure 18. Radionuclide contents at CCO-IV in four samples. Sampling at this site was discontinued after August 1972.

CCO-II—MAY 1972

This site was selected for more detailed analysis because the visual appearance of the sample and the grain size distribution (table 6) suggested a wide variety of components. The agreement between samples CCO-IIA and B (table 3) has already been mentioned. One portion of CCO-IIA was subjected to mineral separation techniques in an attempt to gather more information about the distribution of radioactivity among the various components of Chesapeake Bay sediments. The 300-g portion was first divided into four size fractions, ~500, 150, 75, and < 75 μm. An appreciable number of shell fragments were handpicked from the coarsest fraction. The four size fractions were then treated with aqueous Clerici (thallium malonate-formate) solution at successive densities of 2.8, 3.17, 3.45, and 4.25 g/cm^3. It was expected that the density separation in this aqueous medium would remove a significant part of the adsorbed radionuclides, but that the mineralic K, U, and Th would remain untouched. Fractions were rinsed with deionized water to remove traces of Clerici, and the radionuclide contents were determined in the coincidence counting system. The results of these analyses are given in table 7.

Table 7
Radionuclides in Size/Density Fractions of
CCO-IIA—May 1972

Sample	Mass, g	^{208}Tl pCi/kg	^{214}Bi pCi/kg	%K	^{137}Cs pCi/kg	^{106}Ru pCi/kg
F@2.8/on 500 μm	66.5	16 ± 5	84 ± 12	0.03 + 0.01	< 14	< 103
F@2.8/on 150 μm	128.1	46 ± 5	177 ± 12	0.22 ± 0.01	77 ± 5	< 67
F@2.8/on 75 μm	50.2	90 ± 7	239 ± 14	0.59 ± 0.01	220 ± 14	208 ± 43
F@2.8/on 75 μm	11.9	182 ± 19	642 ± 45	1.04 ± 0.03	486 ± 27	350 ± 106
F@3.17/on 75 μm	1.6	358 ± 82	2616 ± 193	0.73 ± 0.18	468 ± 165	–
F@3.17/< 75 μm	0.9	263 ± 158	997 ± 420	1.75 ± 0.35	892 ± 368	–
F@3.17, 3.45/ on 150 μm	14.3	221 ± 16	1890 ± 47	0.74 ± 0.02	477 ± 25	161 ± 98
F@3.45/on 30, on & < 75 μm	2.4	715 ± 94	4442 ± 264	0.84 ± 0.21	715 ± 188	–
F@4.25/on 500 μm	0.2	< 680	< 2490	4.02 ±1.51	–	–
F@4.25/on 150 μm	0.4	1515 ± 303	8989 ± 808	–	–	–
F@4.25/on 75 μm	0.2	5110 ± 757	38232 ± 2650	–	–	–
F@4.25/< 75 μm	0.1	7344 ± 979	47004 ± 2938	–	–	–
S@4.25	1.6	4404 ± 201	17529 ± 662	< 0.8	403 ± 317	–
Shells, white	5.9	49 ± 19	367 ± 55	–	–	–
Shells, all	17.3	63 ± 10	815 ± 36	< 0.08	< 42	–
Total Mass Integrated Activity	295.7	101 ± 3	505 ± 9	0.30 ± 0.01	127 ± 6	73 ± 19
CCO-IIA (table 3)	–	130	571	0.35	163	158

Samples are identified by sieve size and float (F) or sink (S) at various Clerici densities. Some fractions were recombined on the basis of X-ray diffraction measurements. As only about 1 percent of the sample was lost during the procedure, it is believed that the results are representative. It is apparent that about 20 percent of the original ^{208}Tl, ^{214}Bi, and ^{137}Cs were lost, presumably due to contact with the aqueous solution. It is remarkable that so much material remained persistently adsorbed despite the treatment. The difference in K could almost be due to loss of the brackish water residue dried onto the sample. Although about half of the ^{106}Ru seems to have been lost, small but appreciable amounts may have gone undetected in the various fractions.

The K and ^{137}Cs data for these fractions are plotted in figure 2 with the bulk sediment data. Their general correspondence to the calculated relationship indicates that desorption in the separated fractions did not destroy the general pattern in the original material. Figure 3 also contains ^{214}Bi and ^{208}Tl data from these fractions. Desorption seems to have had a more pronounced effect in these cases.

The ^{208}Tl and ^{214}Bi data can also be compared in terms of the corresponding Th and U contents, assuming secular equilibrium in the two decay chains. Since ^{228}Ac was actually measured as part of the Th determination, this assumption is more justifiable for Th than for U, for which only ^{214}Bi was measured. Figure 19 is a plot of Th versus U for bulk and separated samples. The line in this figure was drawn from the CCO-II fraction data for F at 3.45 and F at 4.25 g/cm^3, with an average Th/U ratio of 1.28. The data for coarse sediments, having low Th and U, agree fairly well with this line and suggest that the ^{208}Tl and ^{214}Bi in these samples are due primarily to mineralic Th and U. Deviations from the line at higher concentrations can be attributed to increasing adsorption of Th in fine-grained sediments. Several of the light/fine fractions of CCO-IIA appear to follow this trend. The heaviest fraction (S at 4.25 g/cm^3) showed ilmenite and zircon in its X-ray diffraction pattern and had Th/U = 2.11, which is intermediate between the 1.28 value for other heavy fractions and the 2.83 ratio for the Long Beach Shore sand (table 8).

The ^{208}Tl and ^{214}Bi in the shells present an interesting situation. The handpicked white shell fragments had radioactivities equivalent to 1.3 ppm Th and 1.1 ppm U, very near the correlation line in figure 19. These fragments were then combined with other fragments stained purple. The combined sample ("all" in table 7) had 2.5 ppm U and 1.6 ppm Th. Mathematically removing the "white" component from the mixture results in the "stained" shells having 3.2 ppm U and 1.7 ppm Th. The Th contents of the two kinds of shells are statistically the same, but the U difference is real—almost a factor of three enrichment.

LONG BEACH SHORE SAMPLE

The Long Beach sample reported in table 8 was collected in April 1972 by R. McLain of the Benedict Estuarine Laboratory, Philadelphia Academy of Natural Science. It was a mixture of rather coarse sand and conspicuous black grains later identified (by X-ray diffraction) as primarily ilmenite containing monazite. The measured ^{208}Tl and ^{214}Bi correspond to 47.5 ppm Th and 16.8 ppm U, with a Th/U ratio of 2.83.

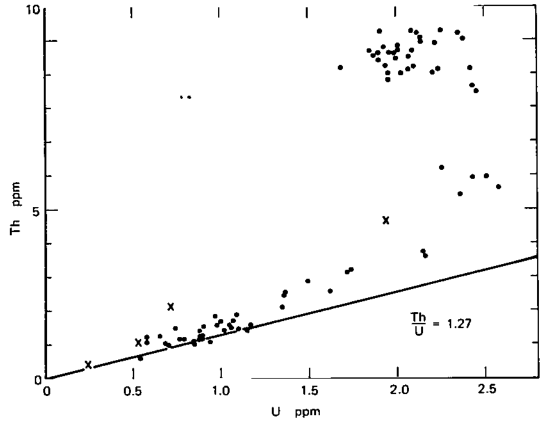

Figure 19. Th versus U for Chesapeake Bay sediment samples. Solid points are for bulk samples (tables 1 through 5); X's are CCO-IIA separate data (table 7). The Th/U correlation line was calculated from mineralic Th/U in dense fractions of CCO-IIA.

Table 8
Radioactivities in Other Sediment Samples

Site		^{208}Tl pCi/kg	^{214}Bi pCi/kg	%K	^{137}Cs pCi/kg	^{106}Ru pCi/kg	^{60}Co pCi/kg
Long Beach (Shore)		1869	5563	0.01	365	788 ± 225	< 11
Potomac River	A	631	1124	2.02	1993	1219 ± 170	24 ± 8
	B	532	1076	2.01	1453	–	< 27
	C	478	1214	1.58	1041	321 ± 248	14 ± 11
	D	392	938	0.59	189	–	< 25

The black grains were easily separated from the sand by heavy liquids (S at 4.25 g/cm^3). An electromagnetic separator was used to obtain a fraction enriched in monazite. The purified monazite sample was too small ($\ll$1 g) for accurate calibration in the present counting system. However, the thorium radioactivity was easily detectable and corresponds to a Th concentration in the percent range.

POTOMAC RIVER SEDIMENTS

The Potomac River sediment samples, taken June 5, 1972, were obtained from the Benedict Estuarine Laboratory. Samples A and B in table 8 are from the Pope's Creek area; C and D were taken near the Morgantown Steam Electric Power Station. The variations of

^{60}Co and ^{137}Cs with K content are consistent with the trends in the Chesapeake Bay sediments (see figure 2). In figure 3, the Potomac River sediment ^{208}Tl and ^{214}Bi data lie consistently and conspicuously above the Chesapeake Bay data, due presumably to either different Th and U influx rates or different sedimentation rates in the two areas. Although only four Potomac River sediment samples were analyzed, the ^{214}Bi and ^{208}Tl data suggest patterns similar to those for Chesapeake Bay sediments. The calculated Th/U ratios of the Potomac River samples lie between 3.3 and 4.7, consistent with the high values from figure 19 for the Chesapeake Bay samples; the amounts are different but the ratios are not. The ^{106}Ru activity in A is also high in comparison with Chesapeake Bay sediments and thus also consistent with a different influx or sedimentation rate.

Goddard Space Flight Center
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