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RADIONUCLIDE DISTRIBUTIONS AND SORPTION BEHAVIOR IN THE SUSQUEHANNA-CHESAPEAKE BAY SYSTEM

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FOREWORD

This Report entitled "Radionuclide Distributions and Sorption Behavior in the Susquehanna-Chesapeake Bay System" was produced for the Maryland Power Plant and Environmental Review Division of the Department of Natural Resources under Contract No. P4-84-07. Research was sponsored jointly by this agency under Interagency Agreement ERD-83-302 and the Office of Health and Environmental Research, U.S. Department of Energy, under contract DE-AC05-84OR21400 with Martin Marietta Energy Systems, Inc. Publication No. 2907, Environmental Sciences Division, Oak Ridge National Laboratory, and Publication No. PPER-R-12, Maryland Power Plant and Environmental Review Division.

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ABSTRACT

Radionuclides released into the Susquehanna--Chesapeake System from the Three Mile Island, Peach Bottom, and Calvert Cliffs nuclear power plants are partitioned among dissolved, particulate, and biological phases and may thus exist in a number of physical and chemical forms. In this project, we have measured the dissolved and particulate distributions of fallout ^{137}Cs; reactor-released ^{137}Cs, ^{134}Cs, ^{65}Zn, ^{60}Co, and ^{58}Co; and naturally occurring ^{7}Be and ^{210}Pb in the lower Susquehanna River and Upper Chesapeake Bay. In addition, we chemically leached suspended particles and bottom sediments in the laboratory to determine radionuclide partitioning among different particulate-sorbing phases to complement the site-specific field data. This information has been used to document the important geochemical processes that affect the transport, sorption, distribution, and fate of reactor-released radionuclides (and by analogy, other trace contaminants) in this river-estuarine system.

Knowledge of the mechanisms, kinetic factors, and processes that affect radionuclide distributions is crucial for predicting their biological availability, toxicity, chemical behavior, physical transport, and accumulation in aquatic systems. The results from this project provide the information necessary for developing accurate radionuclide-transport and biological-uptake models.

EXECUTIVE SUMMARY

Radioactivity is introduced into the Susquehanna River-Chesapeake Bay System from three sources: nature, atmospheric tests of nuclear weapons, and controlled low-level releases from the Three Mile Island, Peach Bottom, and Calvert Cliffs nuclear power plants. Samples of water, suspended matter, and sediments were collected over a three-year time period at four sites: Conowingo Pond, Susquehanna Flats, Annapolis, and Calvert Cliffs. These sites were chosen because they cover a significant range in estuarine salinity, span the major zones of fine-particle deposition, and are within the influence of reactor releases. Dissolved and particulate distributions of naturally occurring ^{7}Be and ^{210}Pb, fallout ^{137}Cs, and reactor-released ^{137}Cs, ^{134}Cs, ^{65}Zn, ^{60}Co, and ^{58}Co were quantified at each of these four sites. The distribution data have been used to

(1) describe sorption kinetics and estimate radionuclide removal rates from the water column,

(2) determine radionuclide partitioning among different particulate sorbing phases (clay surfaces, organics, Fe-Mn coatings, and mineral lattices), and

(3) document the important geochemical processes that affect the fate of radionuclides (and by analogy, other trace contaminants) in the Susquehanna--Chesapeake Bay System.

The concentration, transport, and fate of reactor-released radionuclides in freshwater areas is primarily governed by variations in radionuclide input and fine-particle dynamics. Because variations in input and particle dynamics are short term; chemical equilibrium is continually adjusting and is therefore rarely attained at the time of sample collection. As a result, measured radionuclide concentrations and their particle-to-water distributions vary over one order of magnitude, depending on variations in freshwater flow, sediment resuspension, and other environmental conditions. The short-term nature of these variations limits the suitability of using laboratory-derived equilibrium distribution coefficients in radionuclide transport and fate models.

In the Susquehanna River, our results indicate that ^{137}Cs, ^{134}Cs, ^{65}Zn, ^{60}Co, and ^{7}Be are rapidly removed from the dissolved phase by sorption onto suspended matter and are rapidly removed from the water column by fine-particle deposition. Variations in <u>dissolved</u> radionuclide concentrations are primarily governed by variations in radionuclide input. Following input, radionuclide sorption rates onto suspended matter from the dissolved phase range from 10-20% d^{-1}. Although fine-particle deposition in Conowingo Pond may trap as much as 80% of the radiocesium, radiocobalt and radiozinc released from the Peach Bottom Atomic Power Station during low-to-normal Susquehanna flow conditions, much of this deposited material (and associated radionuclides) may be resuspended during storms or seasonal high-flow events, and thus escape Conowingo Pond to be ultimately deposited in upper Chesapeake Bay.

In the saline waters of Chesapeake Bay, our results indicate that radiocobalt and beryllium-7 are still very particle reactive but that radiocesium tends to remain in solution because it must compete with salt-water cations (such as K^+, Na^+, and Ca^{++}) for sorption sites on particles. Consequently, the transport and fate of radiocobalt released from the Calvert Cliffs facility will be governed by sorption processes and particle dynamics, whereas the transport and fate of released radiocesium will be more strongly influenced by estuarine circulation and water dynamics.

Chemical-leaching treatments of particulate material collected from the Susquehanna River-Chesapeake Bay System indicate that all of the above radionuclides, except radiocesium, are associated with particulate Fe-Mn oxide phases. The geochemical cycling of these phases, and associated radionuclides, may be affected by changes in oxygen levels and pH, but are unaffected by changes in estuarine salinity. In contrast, radiocesium is associated with clay minerals and its desorption may be affected by estuarine salinity, depending on its contact time with the particle. Long contact times allow radiocesium to become irreversibly fixed within clay minerals by substitution for K^+ or collapse of the hydrated lattice. Short contact times leave radiocesium on the particle surface and susceptible to desorption by exchange with seawater cations. Consequently, recently introduced radiocesium (^{134}Cs and ^{137}Cs) from reactors may behave differently than fallout ^{137}Cs which has been in the Susquehanna drainage system for over two decades. This finding dictates that the transport and fate of radiocesium from power plants and fallout must be modeled differently.

The information obtained from this project is crucial for understanding the radiological effects of operating nuclear power plants and predicting the effects of future discharges of radioactivity. In addition to providing site-specific data on the fate of reactor-released radionculides in the Susquehanna--Chesapeake Bay System, the results from this project are necessary for developing accurate contaminant-transport and biological-uptake models and thus, for eliminating some of the uncertainty associated with extrapolating numerical simulations and laboratory data to natural environments.

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1. INTRODUCTION

Nuclear energy production at the Three Mile Island (TMI), Peach Bottom (PB), and Calvert Cliffs (CC) reactor sites (Fig. 1) has introduced several anthropogenic radionuclides into the Susquehanna-Chesapeake System via controlled releases of coolant-water effluent. Some of these radionuclides, such as tritium (^{3}H), iodine-131 (^{131}I), and strontium-90 (^{90}Sr), are relatively soluble in riverine-estuarine environments. Consequently, their transport and biogeochemical fates are conceptually straightforward and are mediated by water mass movements and biological uptake from the water phase. A much greater number of the released radionuclides, however, are chemically and biologically reactive, such as zinc-65 (^{65}Zn), cesium-137 (^{137}Cs), cesium-134 (^{134}Cs), cobalt-60 (^{60}Co), cobalt-58 (^{58}Co), and manganese-54 (^{54}Mn). These reactive nuclides show a strong affinity for association with fine-grained organic and inorganic particles as a result of (1) ion-exchange interactions with particle surfaces, (2) coprecipitation with amorphous Fe-Mn-Al hydrous-oxide coatings, (3) complexation with organic substances, (4) incorporation into planktonic organisms, fecal material, or mineral lattices, or (5) flocculation with colloidal organic and inorganic substances during river and seawater mixing (Jenne and Wahlberg 1968; Troup and Bricker 1975; Sholkovitz 1976; Turekian 1977; Sigleo and Helz 1981; Sigleo et al. 1981; Olsen et al. 1982; Helz et al. 1985).

The extent to which a specific radionuclide is affected by the above processes depends upon the chemical forms of the radionuclide and the chemical and physical characteristics of the particulate material and transporting medium. Therefore, quantitative estimates for the net transport and biogeochemical fate of particle-reactive radionuclides are difficult to make, especially in river-estuarine zones, where freshwater mixes with seawater. In these zones large variations occur in salinity, freshwater flow, tidal currents, pH, oxidation potentials, <u>in situ</u> productivity, biological communities, suspended matter concentrations and compositions, channel-bay morphology, and fine-particle deposition rates.

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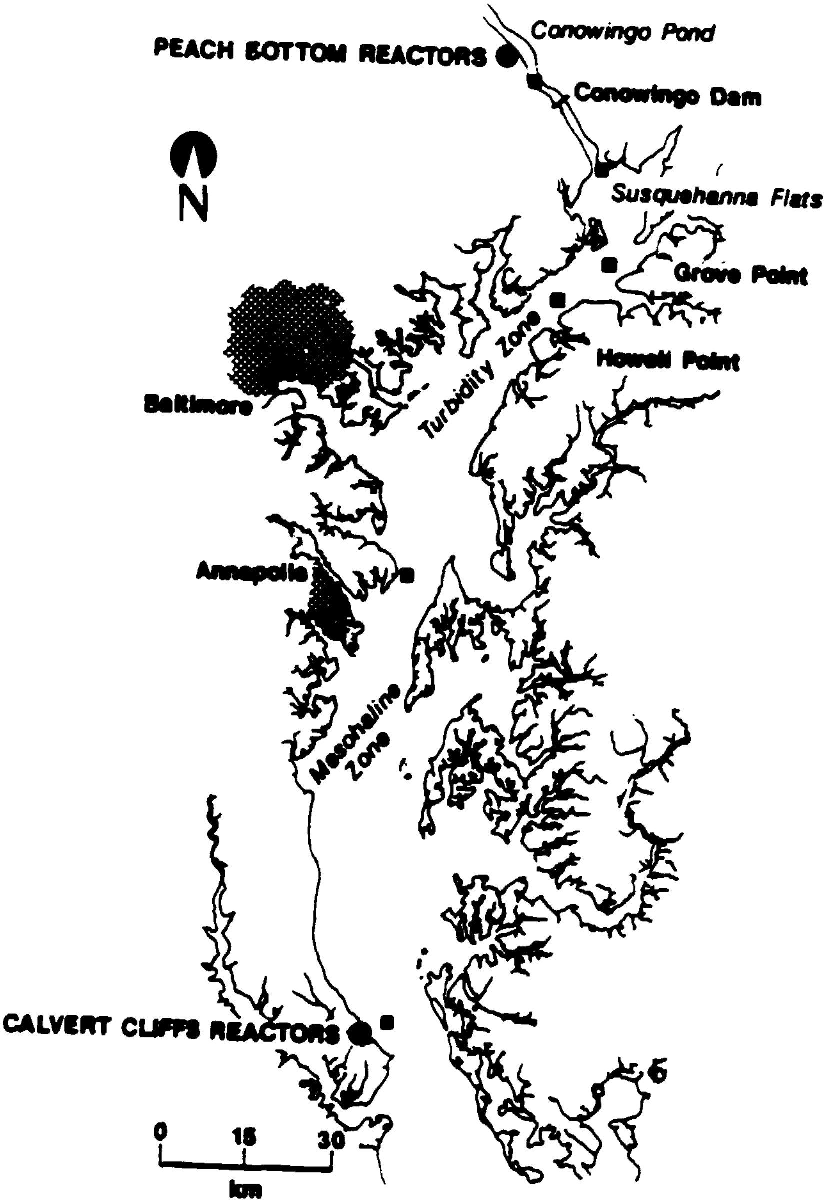

	Volume (10^6 m^3)	Surface Area (10^6 m^2)	Average Depth (m)
Conowingo Reservoir	336	37	9.0
Upper Chesapeake Bay	12,780	1,566	8.2

Fig. 1. A map of the Lower Susquehanna River and Upper Chesapeake Bay illustrating the locations of the Peach Bottom and Calvert Cliffs nuclear power plants and the locations of our large-volume water and suspended-matter samples. The Three Mile Island nuclear power plant (not shown) is located on the Susquehanna near Harrisburg, about 70 km upstream from Conowingo Dam.

In this project we have measured the particle-to-water distribution of several natural and anthropogenic radionuclides that have known sources and histories of input into the Susquehanna-Chesapeake Bay System. The field samples were collected during the fall and spring (1983-1985) to document seasonal variations in radionuclide distributions in association with low and high Susquehanna River flows, respectively. These measured distributions have been used with laboratory data to (1) identify transport mechanisms, (2) quantify sorption kinetics and removal rates from the water column, (3) determine chemical partitioning among different particulate-sorbing phases, and (4) identify the important biogeochemical processes that affect the fate of radionuclides (and by analogy, other trace substances or contaminants) in this river-estuarine system. This information is crucial for predicting the chemical behavior and biological availability of reactor-released radionuclides in the Susquehanna-Chesapeake Bay System. In addition, the site-specific results from this project allow for the development of accurate radionuclide-transport/biological-uptake/human-dose models.

1.1 ENVIRONMENTAL SETTING, HYDROLOGICAL CHARACTERISTICS AND SAMPLING LOCATIONS

The Susquehanna-Chesapeake Bay System forms the largest estuarine system on the eastern coastline of the United States and drains an area of about 165,000 km^2 (Wolman 1968). Although this hydrologic system encompasses the drainage basins of the Potomac, Patuxent, Rappahannock, James, and several lesser rivers, the Susquehanna River is by far the largest single freshwater contributor. With a mean annual discharge of about 3.46×10^{10} m^3/yr or 1095 m^3/sec (Helz et al. 1985), the Susquehanna comprises more than 50% of the total freshwater input to Chesapeake Bay and more than 85% of that introduced north of the mouth of the Potomac (Schubel and Carter 1976).

Mean monthly freshwater flows for the Susquehanna are illustrated in Fig. 2 for the years 1983, 1984, and 1985. Average monthly flows range from about 200 m^3/sec to 3500 m^3/sec, with the highest values generally occurring in March or April. Past work has shown that the

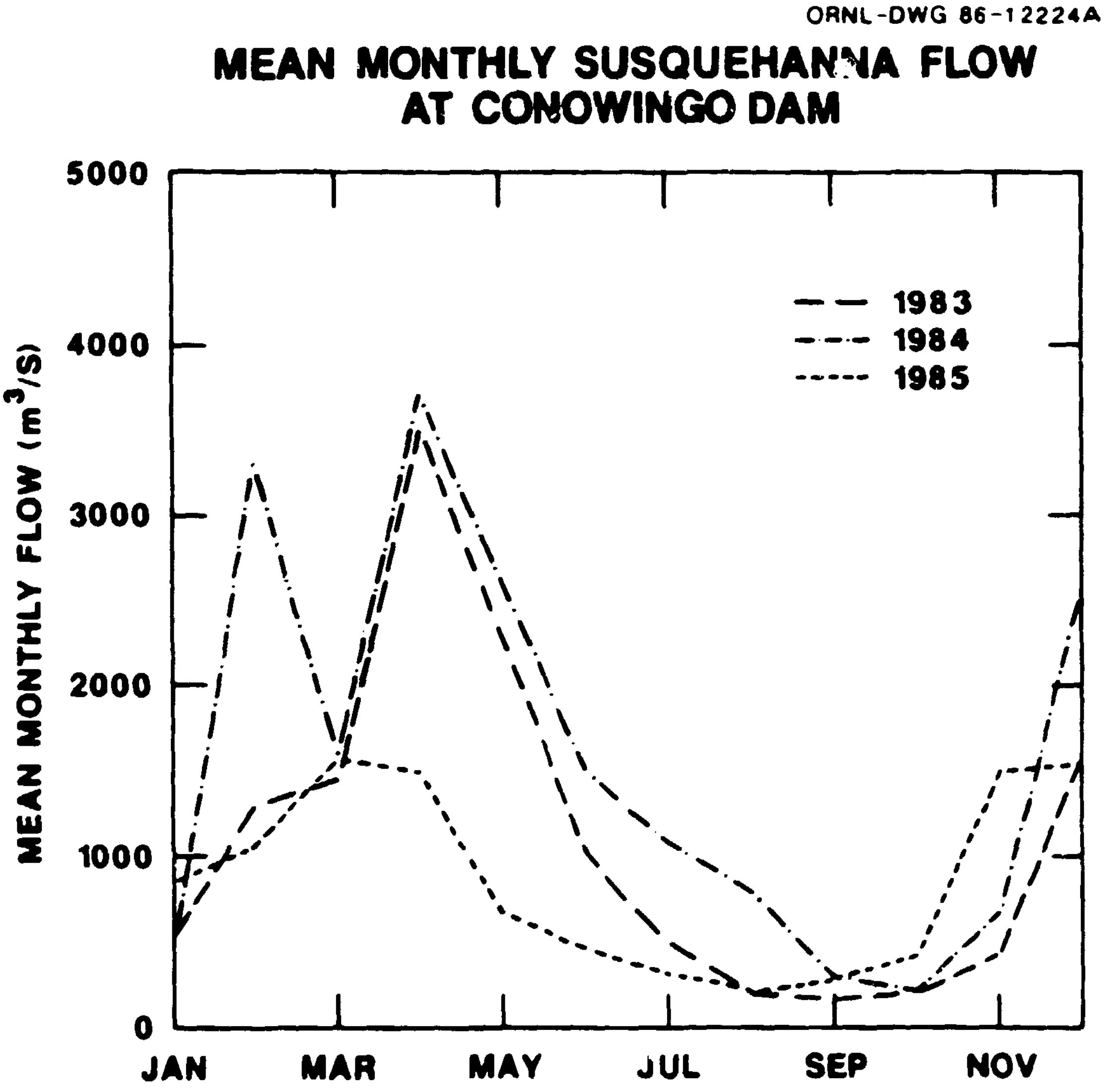

Fig. 2. Mean monthly freshwater flow in the Susquehanna River at Conowingo Dam. The data are from U.S. Geological Survey Water-Data Reports for 1983, 1984 and 1985.

amount of spring runoff entering Chesapeake Bay from the Susquehanna River can significantly affect salinity distributions and water stratification within the Bay (Pritchard 1955). Biggs (1981) has suggested that peak flows may have increased as much as 30% during the past two hundred years as a result of drainage basin deforestation. Deforestation, industrialization, and urbanization have caused an increase in nutrient and contaminant fluxes to Chesapeake Bay in addition to the increased flows (Carpenter et al. 1975; Nichols et al. 1981; Sinex and Helz 1982; Brush 1984; Helz et al. 1986).

Estimates of the amount of suspended sediment discharged from the Susquehanna have been made by several investigators (Biggs 1970; Schubel 1972; Williams and Reed 1972; Carpenter et al. 1975; Schubel and Carter 1976). After reviewing these data, Gross et al. (1978) have estimated that the long-term average sediment discharge from the Susquehanna is 0.9×10^6 tons/year (in the absence of major floods) and that 50-60% of this sediment load is transported during the spring freshet, which is caused by snowmelt. Most of this sedimentary material is deposited within the upper portions of Chesapeake Bay (Zabawa and Schubel 1974; Donoghue 1981; Helz et al. 1986). This is also a major zone of accumulation for particle-associated radionuclides that escape from behind Conowingo Dam (Olsen et al. 1981a).

The areas sampled in this study are illustrated in Fig. 1, and include Conowingo Pond, Susquehanna Flats, the estuarine turbidity zone (Howell Point and Grove Point), and the mesohaline portion of Chesapeake Bay (Annapolis and Calvert Cliffs). These sampling sites were chosen because they cover a significant range in salinity, span the major zones of fine-particle deposition, and are within the influence of reactor releases.

Conowingo Pond is a freshwater reservoir located on the Susquehanna River about 14 km upstream from its mouth in Chesapeake Bay. The reservoir is formed by Conowingo Dam, one of four hydroelectric dams on the Susquehanna River below Harrisburg, Pennsylvania. The Peach Bottom Atomic Power Station discharges directly into this reservoir on the west side of the river about 11 km upstream from the dam (Fig. 1). Previous work has shown that reservoirs formed by dams serve as efficient fine-particle and radionuclide traps (Olsen et al. 1981a; McLean et al. 1988).

Susquehanna Flats is a wide, shallow area (surface area 107 km^2, average depth about 1.6 m) at the north end of Chesapeake Bay (Fig. 1). The sediments within this area are sandy, reflecting scour during periods of high freshwater discharge and resuspension by wave and current activity. Although fresh water predominates throughout most of the year, saline water can occasionally penetrate into this area during the low-flow conditions of late summer and early fall, when fine-grained material may also be temporarily deposited.

The turbidity zone is an area of pronounced fine-particle deposition in response to estuarine circulation patterns and several biogeochemical processes that occur when fresh water mixes with saline water. Estuarine circulation in Chesapeake Bay is characterized by a lower-salinity surface layer with a net seaward flow and a denser, more saline bottom layer having a net landward flow (Pritchard 1955). The two layers are separated by a no-flow regime. Near the landward limit of the salinity intrusion, this no-flow regime intersects the estuarine floor, causing a zone of rapid deposition and fluid mud (Schubel 1968, Nichols 1985). In addition, riverborne suspended colloids (clays and organics) coagulate or flocculate into larger particles as fresh water mixes with saline water near the landward limit of the salinity intrusion. Both flocculation within the water column and resuspension from the fluid mud bottom create a turbidity maximum within this zone that causes phytoplankton production to become light limited. Water salinities generally range from about 1 to 5 ppt within the turbidity zone. Since the position of the turbidity zone is dependent on freshwater discharge, it is seasonally variable. In Chesapeake Bay, this zone of turbidity and pronounced fine-particle deposition generally occurs between Grove Point and Baltimore (Fig. 1).

The mesohaline zone is characterized by a salinity range between 5 and 18 ppt and in Fig. 1, occurs between Baltimore and Calvert Cliffs. Two sampling sites were located within the mesohaline zone, one at Annapolis near the Bay Bridge and the other about 200 m offshore from Calvert Cliffs, adjacent to the reactor discharge site. Mesohaline habitats support large benthic populations of crabs, clams, and oysters and produce most of the estuarine forage-fish biomass. In addition, these areas serve as important feeding grounds for predatory fishes (e.g., striped bass and bluefish). The bottom sediments in this area are generally a mixture of fine sands, silts, and clay. Pockets of highly liquefied or fluid mud occur in areas where tidal and estuarine flow is hindered or dissipated, such as in geomorphologic depressions greater than 10 m in depth or around obstructions such as piers and bridges. In Fig. 1, the mesohaline zone encompasses a surface area of about 1285 km^2 and is about 9.3 m deep on an average. During the summer, the water downstream of Annapolis and deeper than 10 m usually experiences oxygen depletion. On occasion, this depletion may result in anoxia and in hydrogen sulfide production, a condition that is toxic to fish and benthic organisms. In addition, this depletion may influence the partitioning of radionuclides by enhancing the redox cycling and release of several metals, including iron (Fe), manganese (Mn), and cobalt (Co) from anoxic sediments. Below Calvert Cliffs, the Patuxent and Potomac tributary estuaries enter the main Bay. Throughout this report we will refer to the Upper Bay as that portion of Chesapeake Bay from Calvert Cliffs to Susquehanna Flats.

1.2 RADIONUCLIDE INPUTS

Radionuclides are introduced into the Susquehanna-Chesapeake System from natural sources, global fallout, medical facilities, and nuclear power plants. Although this project is primarily concerned with identifying the distribution and fate of radionuclides released from nuclear power plants, we have also measured the distribution of several natural and fallout radionuclides to provide additional information for comparing relative amounts of radioactivity and for evaluating the processes that control their sorption behaviors. The natural radionuclides discussed in this report include Beryllium-7 (^{7}Be), Lead-210 (^{210}Pb), and Potassium-40 (^{40}K).

1.2.1 Natural Radionuclides

Beryllium-7 (53.3-d half-life) is produced by cosmic-ray spallation of nitrogen and oxygen within the earth's atmosphere. Like other chemically reactive and atmospherically derived radionuclides, ^{7}Be rapidly becomes associated with aerosols, and its deposition to the earth's surface is primarily a result of washout by precipitation (Olsen et al. 1985). Upon contact with rainwater, ^{7}Be is solubilized and is deposited as a dissolved constituent of precipitation. The atmospheric flux of ^{7}Be to the Susquehanna-Chesapeake Bay System has been measured at Norfolk, Virginia, and the data are listed in Appendix A. Total (wet and dry) atmospheric fluxes of ^{7}Be range from 0.15 to 1.09 pCi/cm^{2} on a monthly basis; when extrapolated over the surface area of Upper Chesapeake Bay and Conowingo Pond (Fig. 1), the amount corresponds to a monthly input ranging from 2.4 to 17.5 Ci. The particle-to-water distributions of ^{7}Be have been used to quantify radionuclide sorption rates to suspended matter and radionuclide removal rates from the water column (See Section 4.1).

Lead-210 (22-year half-life) is a daughter nuclide in the ^{238}U-^{234}U-^{230}Th-^{226}Ra-^{222}Rn-^{218}Po-^{214}Pb-^{210}Pb-^{206}Pb radioactive decay series that occurs naturally in the earth's rocks, soils, and waters. As an inert gas, some radon-222 (3.8-day half-life) emanates into the atmosphere from the earth's surface and rapidly decays, through a series of short-lived daughters, to ^{210}Pb. This atmospherically produced ^{210}Pb, like ^{7}Be, is primarily removed by rainfall forming a flux of unsupported ^{210}Pb (U-^{210}Pb) back to the earth's surface. The total monthly atmospheric flux of U-^{210}Pb to Chesapeake Bay (presented with the ^{7}Be data in Appendix A) ranges from 0.006 to 0.071 pCi/cm^{2}. The distribution of U-^{210}Pb in sediments has been used to document rates of sediment and associated radionuclide accumulation.

Potassium-40 is a naturally occurring radionuclide comprising about 0.012% of the potassium in K-bearing minerals such as muscovite, illite and K-feldspar. Consequently, the ^{40}K activity of the sediments and suspended particles primarily reflects grain size and composition. For example, quartz-rich sandy sediments are characterized by low ^{40}K activities (5,000-10,000 pCi/kg) relative to fine-grained silts and clays (15,000-20,000 pCi/kg). The activity of ^{40}K has been used to help characterize the composition of particulate samples and in some cases, to normalize the anthropogenic radionuclide data.

1.2.2 Fallout Radionuclides

Global fallout from atmospheric weapons testing has introduced fission nuclides such as ^{137}Cs and ^{90}Sr into the Susquehanna-Chesapeake Bay System over a longer time span than have nuclear power plants. The major influx of fallout ^{137}Cs and ^{90}Sr occurred between 1962 and 1964. The decay-corrected (to 1985) input of fallout ^{137}Cs and ^{90}Sr is about 95 mCi/km^2 and 60 mCi/km^2, respectively. When extrapolated over the drainage basin of Chesapeake Bay, this input corresponds to 15,700 Ci of ^{137}Cs and 9,900 Ci of ^{90}Sr. Extrapolation over the surface area of Conowingo Pond and Upper Chesapeake Bay (Fig. 1) indicates that about 150 Ci of ^{137}Cs and 95 Ci of ^{90}Sr have been directly input to the water surface by global fallout. Because ^{134}Cs, ^{65}Zn, ^{60}Co, and ^{58}Co have relatively low bomb-production yields and are primarily produced by neutron activation during reactor operations, controlled releases of reactor liquid effluent were the only significant source of these nuclides in the Susquehanna-Chesapeake Bay system during our October 1983-October 1985 sampling period.

More recently, however, detectable concentrations of ^{134}Cs as well as ^{137}Cs, ^{141}Ce, ^{140}Ba-^{140}La, ^{131}I, ^{106}Ru, and ^{103}Ru have been introduced into the Susquehanna-Chesapeake Bay system as a result of the reactor accident at Chernobyl on April 26, 1986. Total (wet and dry) atmospheric fluxes of ^{134}Cs, ^{131}I, ^{106}Ru, and ^{103}Ru (measured at Oak Ridge National Laboratory) were 0.36 mCi/km^2, 0.85 mCi/km^2, 0.34 mCi/km^2, and 0.96 mCi/km^2 respectively from April 26 to June 9, 1986, with about 50% of the total ^{134}Cs flux occurring between May 19 and 23, 1986. Although the input of Chernobyl radionuclides to

Chesapeake Bay has not substantially altered present radionuclide inventories nor posed any adverse environmental or health risks, it does provide a spiked input and a unique opportunity for quantifying the biogeochemical processes affecting the fate of radiocesium in coastal areas, where reactor-released radiocesium is undetectable (McLean, 1986).

1.2.3 Reactor-Released Radionuclides

Liquid effluents released from the TMI, PB, and CC nuclear power plants have introduced a wide variety of radionuclides into the Susquehanna-Chesapeake System including:

^{137}Cs (30-year half-life),
^{134}Cs (2.1-year half-life),
^{110m}Ag (253-d half-life),
^{90}Sr (29-year half-life),
^{65}Zn (245-d half-life),
^{60}Co (5.3-year half-life),
^{58}Co (71-d half-life), and
^{54}Mn (280-d half-life).

The TMI Nuclear Station is situated on an island in the Susquehanna River about 13 km southeast of Harrisburg, Pennsylvania, and about 70 river km upstream from Conowingo Dam (Fig. 1). Liquid releases of ^{137}Cs, ^{134}Cs, ^{60}Co, ^{58}Co and ^{65}Zn from the two pressurized-water reactor units at TMI totaled 0.15, 0.23, 0.05, 0.77, and 0.005 Ci respectively from 1975 to 1979 (Olsen et al. 1981a). Semi-annual effluent release reports submitted by the utility to the U.S. Nuclear Regulatory Commission indicate that there have been no appreciable radionuclide releases from the TMI site since 1979 (McLean et al. 1986).

Annual radionuclide release histories for ^{137}Cs, ^{134}Cs, ^{60}Co, ^{58}Co, and ^{65}Zn from the PB and CC sites are presented in Figs. 3 and 4, respectively. The annual liquid release data are listed in Appendix B with additional quarterly, monthly and daily release data relevant to our time period of sample collection (Tables B-1 to B-5). Liquid releases from the two boiling-water reactors at the PB site (Fig. 1)

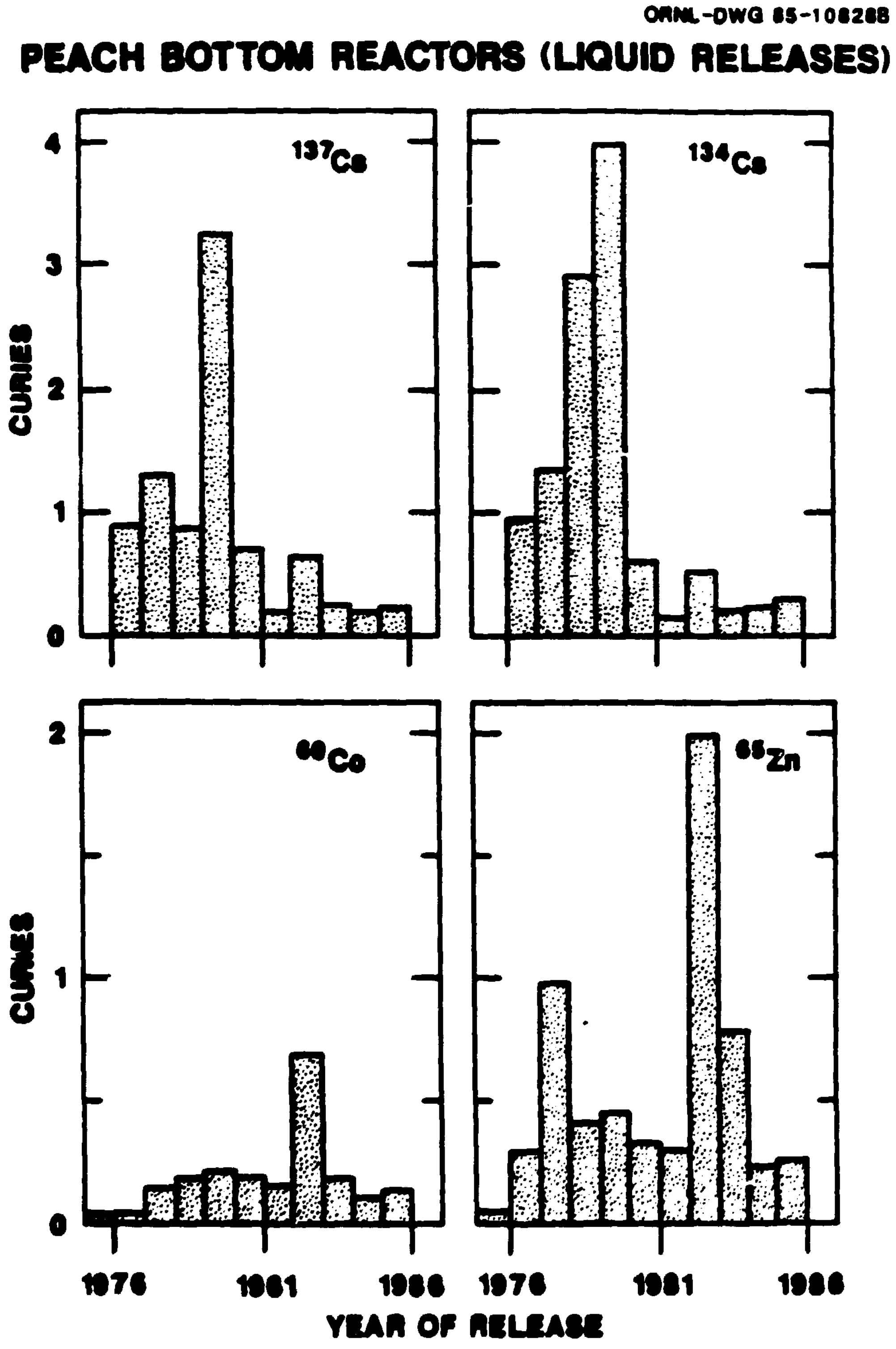

Fig. 3. Release history for ^{137}Cs, ^{134}Cs, ^{60}Co, and ^{65}Zn in liquid effluent from the Peach Bottom Atomic Power Station.

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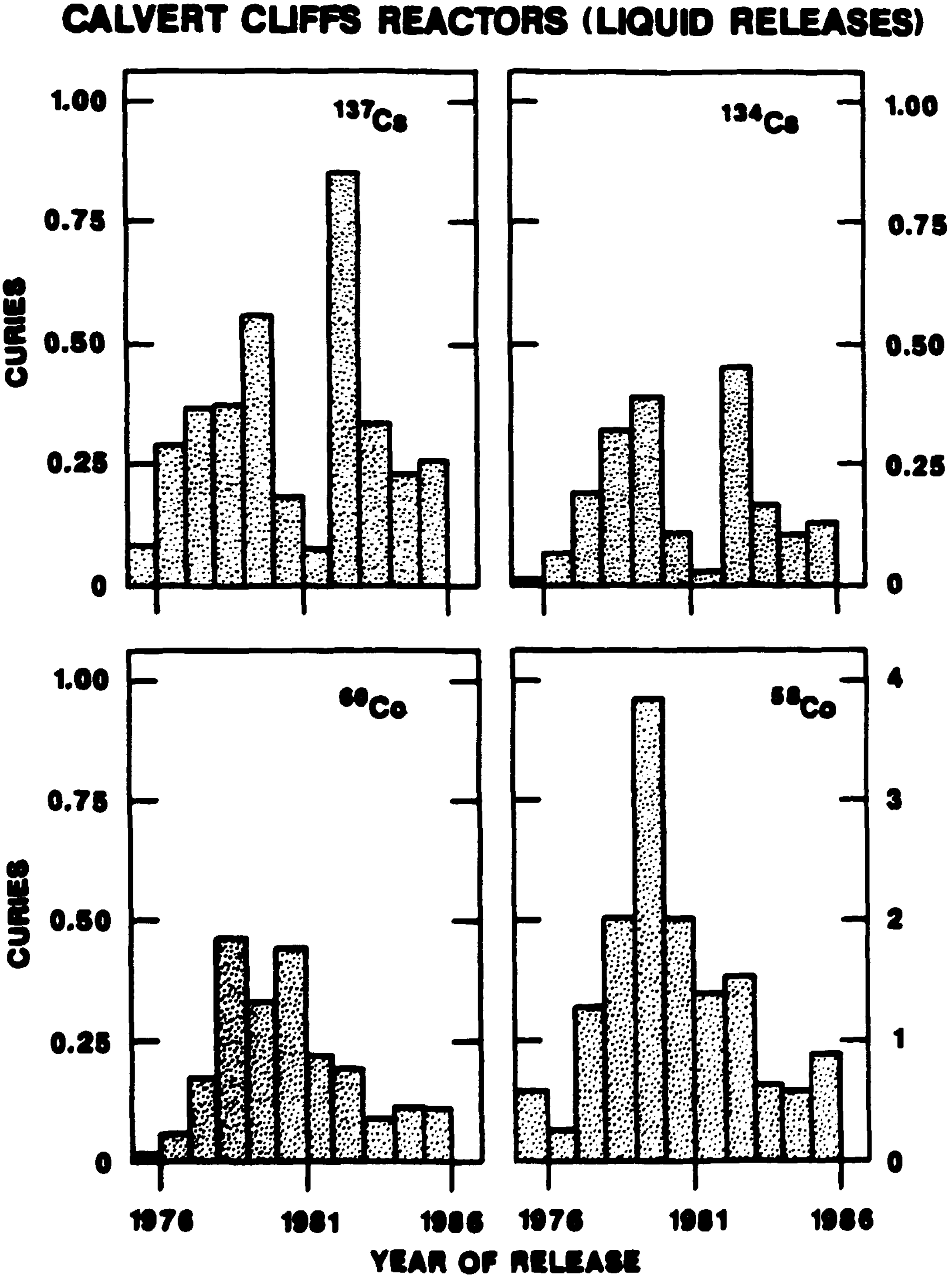

Fig. 4. Release history for ^{137}Cs, ^{134}Cs, ^{60}Co, and ^{58}Co in liquid effluent from the Calvert Cliffs Nuclear Power Plant.

totaled 8.34 Ci of ^{137}Cs, 10.6 Ci of ^{134}Cs, 1.81 Ci of ^{60}Co, 0.24 Ci of ^{58}Co, and 5.91 Ci of ^{65}Zn, as of October 31, 1985 (Appendix B-1). Despite the accident at TMI, more than 95% of the reactor-released ^{134}Cs, ^{60}Co, and ^{65}Zn introduced into the Susquehanna River is a result of routine liquid effluent disposal from the PB facility. Total radionuclide liquid releases into the mesohaline waters of Chesapeake Bay from the two pressurized-water reactors at the CC site are 3.78 Ci of ^{137}Cs, 2.00 Ci of ^{134}Cs, 2.17 Ci of ^{60}Co, 14.6 Ci of ^{58}Co, and 0.024 Ci of ^{65}Zn (Appendix B-3).

2. METHODOLOGY

2.1 SAMPLE COLLECTION

To quantify radionuclide particle-to-water distributions, we collected a total of twenty-one large-volume water samples (about 800-L each) at six different sites in the Susquehanna - Upper Chesapeake Bay System (Fig. 1). Each sample was collected about 2 m below the water surface. The samples were collected on four separate occasions during October 1983, April 1984, May 1985, and October 1985. The samples were collected in the fall and spring when freshwater flow is near its minimum and maximum, respectively (Fig. 2 and Appendix C), in an attempt to sample annual extremes in salinity distributions, dissolved-oxygen concentrations, and suspended-particle concentrations and compositions. Sample information concerning the location, date of collection, freshwater flow characteristics, suspended-matter concentrations, salinity, and meteorologic conditions during the collection period is presented in Appendix D. Two of the sites, Grove Point and Howell Point (Fig. 1), were only sampled in October 1985 in an attempt to document the sorption-desorption fates of reactor-released ^{134}Cs, ^{65}Zn, and ^{60}Co upon first contact with saline water in the turbidity zone.

Surface sediment samples and sediment cores were also collected at each of the large-volume water sampling sites. The surface grab samples were packed in 1000 cm^3 marinelli beakers, sealed with a silastic glue, and stored in an ice box or refrigerator until analyzed. The sediment cores were collected with a vibrocorer using 3-in. (7.7-cm) diameter aluminum pipe. The cores were extruded within a few hours after collection and sectioned into 2- or 4-cm increments, depending upon the depth in the core.

2.2 SAMPLE PREPARATION AND SPECTROMETRIC TECHNIQUES

The water, suspended matter, and sediment samples were analyzed for reactor-released ^{137}Cs, ^{134}Cs, ^{110m}Ag, ^{65}Zn, ^{60}Co, and ^{58}Co and for naturally occurring ^{40}K and ^{7}Be by gamma spectrometry, using low-background, high-resolution, lithium-drifted germanium detectors equipped with a Nuclear Data Model 6700 microprocessor system programmed to record gamma spectra in 4096 channels. Naturally occurring ^{226}Ra-supported ^{214}Pb and ^{210}Pb were quantified with a planar, intrinsic-germanium detector, using the same microprocessor system.

All samples were packed in 1000-cm^3 marinelli beakers, 90-cm^3 aluminum cans, or 15-cm^3 plastic petri dishes, depending on the amount of material analyzed. The detectors were calibrated for the respective geometries using a certified mixed standard (Amersham Radiochemical Centre; QCY 44/46); the calibration procedures are described elsewhere (Larsen and Cutshall 1981). Reported radionuclide concentrations are per gram dry weight and counting errors are ± one standard deviation.

Suspended matter (> 0.45 µm) was removed from the large-volume water samples by continuous-flow centrifugation within 4 h after sample collection (Fig. 5). The chemical (major and trace element) characteristics of the suspended particles and sediments were determined by inductively coupled plasma (ICP) spectroscopy.

2.3 RADIOCHEMICAL TECHNIQUES

After centrifugation, each large-volume water sample was acidified with HCl to a pH of approximately 2, and stable Cs, Zn, Co, Be, Pb, and Fe were added as carriers and yield tracers. The yield tracers were allowed to equilibrate for 6 to 8 h and were recovered from the acidified water with cation resins or with $Fe(OH)_3$ precipitates. Quantification of the yield tracers by atomic absorption spectrometry indicated that 60-90% of the Co and 70-100% of the Zn, Be, Pb, and Cs could be recovered by these procedures. The radionuclide activities for each sample were yield corrected according to the actual recovery.

After acidification, dissolved radiocesium was removed from the large-volume water sample by sorption on a cation-exchange resin (ammonium molybdophosphate). The resin was added on the same day of sample collection and was allowed to settle out of the sample overnight (Fig. 5). The water was then transferred to another 1000-L tank, and the pH was adjusted to about 10 with NaOH to allow the iron to precipitate. Dissolved ^{65}Zn, ^{60}Cc, ^{58}Co, ^{7}Be, and ^{210}Pb were removed by coprecipitation with, or sorption on, $Fe(OH)_3$.

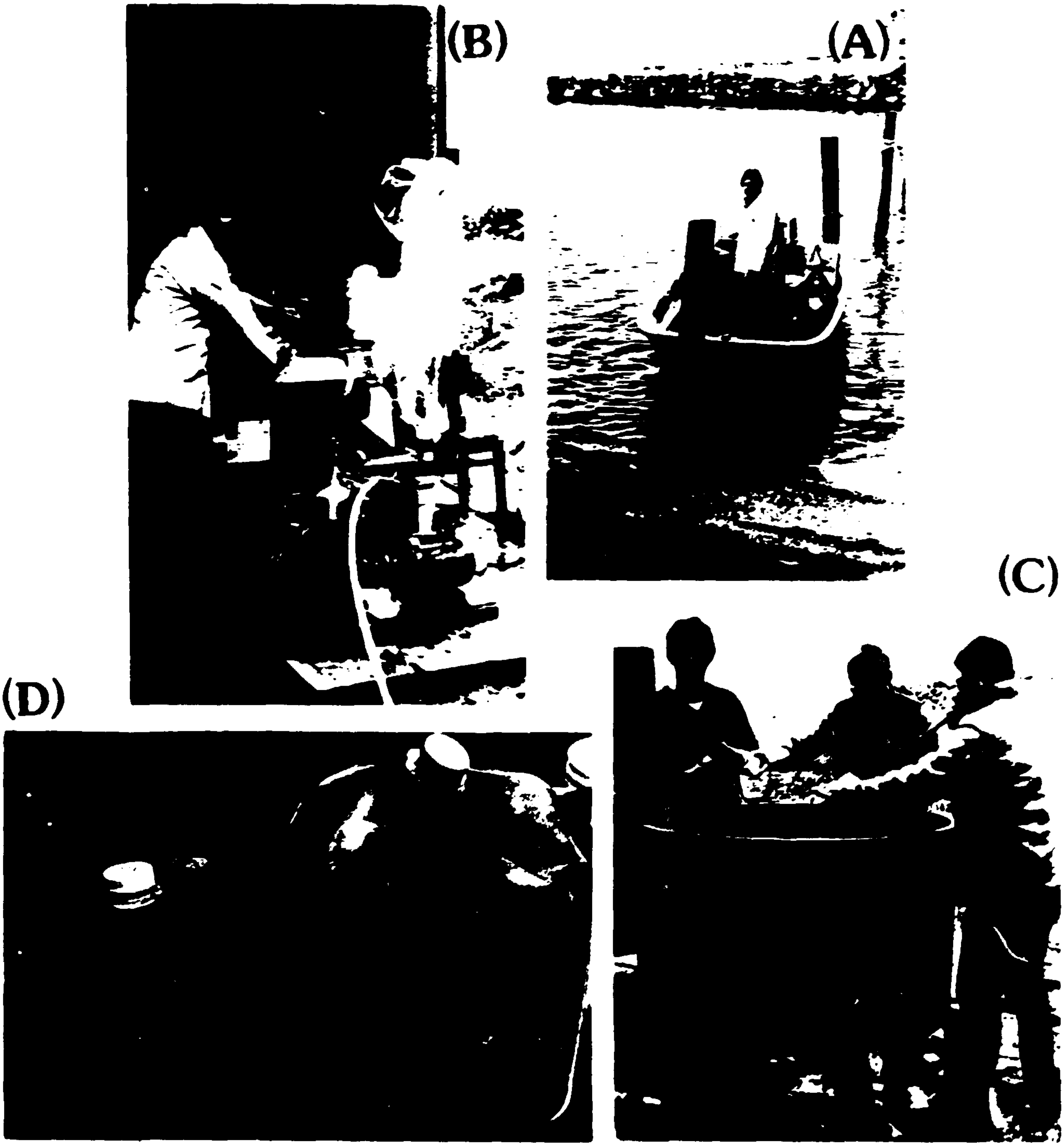

Fig. 5. A series of photos illustrating sample collection and processing procedures. (A) About 800 L of water was collected at each site and transported to the shoreline. (B) The suspended matter was removed from the large-volume water sample by continuous-flow centrifugation. (C) Radionuclides in the dissolved phase were removed using cation resins or coprecipitation with Fe(OH$_3$). (D) The suspended matter, resin and precipitate samples were placed in an ice box and brought back to ORNL for radionuclide analysis.

2.4 LABORATORY LEACHING EXPERIMENTS FOR RADIONUCLIDE PARTITIONING AMONG DIFFERENT SORBING PHASES

To examine the affects of salinity, organic decomposition, and diagenesis on the mobility and bioavailability of particle-associated radionuclides we determined the extent of radionuclide partitioning among different particulate sorbing phases (i.e., particle surfaces, organic detritus, Fe-Mn coatings, organic coatings, and mineral lattices) using an operationally defined selective extraction scheme similar to that described in Gibbs (1977), Trefry (1977) and Tessier et al. (1979). The modified extraction scheme used in this project is diagrammed in Fig. 6. From 5 to 50 g of wet particulate material (suspended matter or sediment) was placed into a 1 L bottle with 900 mL of leaching solution. Following extraction, both the particulate sample and the leach solution were gamma-counted for 24 to 48 h. The particulate sample was returned to the bottle, and the next 900 mL sequential leach solution was added. The amount of radioactivity removed by each extraction was expressed as a percent of the total amount measured initially.

The first extraction involved leaching the freshwater suspended matter and sediments from Conowingo Pond with saline Chesapeake Bay water (salinity about 10 ppt) for about 3 h to determine the extent that sorbed radionuclides could be removed by transport into estuarine areas. The second extraction involved leaching with NH_4Cl to remove the radionuclides sorbed to particle surfaces that were not removed by exchange with seawater cations. Previous work has shown that NH_4^+ (which is produced during the decomposition of organic matter in sediments) is extremely effective in replacing radiocesium on mineral surfaces (Evans et al. 1983).

After the exchangeable nuclides were removed, the sediment samples were sieved to remove organic detritus greater than 850 µm in size. Previous work by Olsen (1979) has shown organic detritus can concentrate radionuclides and that this detrital component can contain a significant fraction of the total radioactivity associated with coarse-grained sediments. Because organic detritus (>850 µm) comprised a small

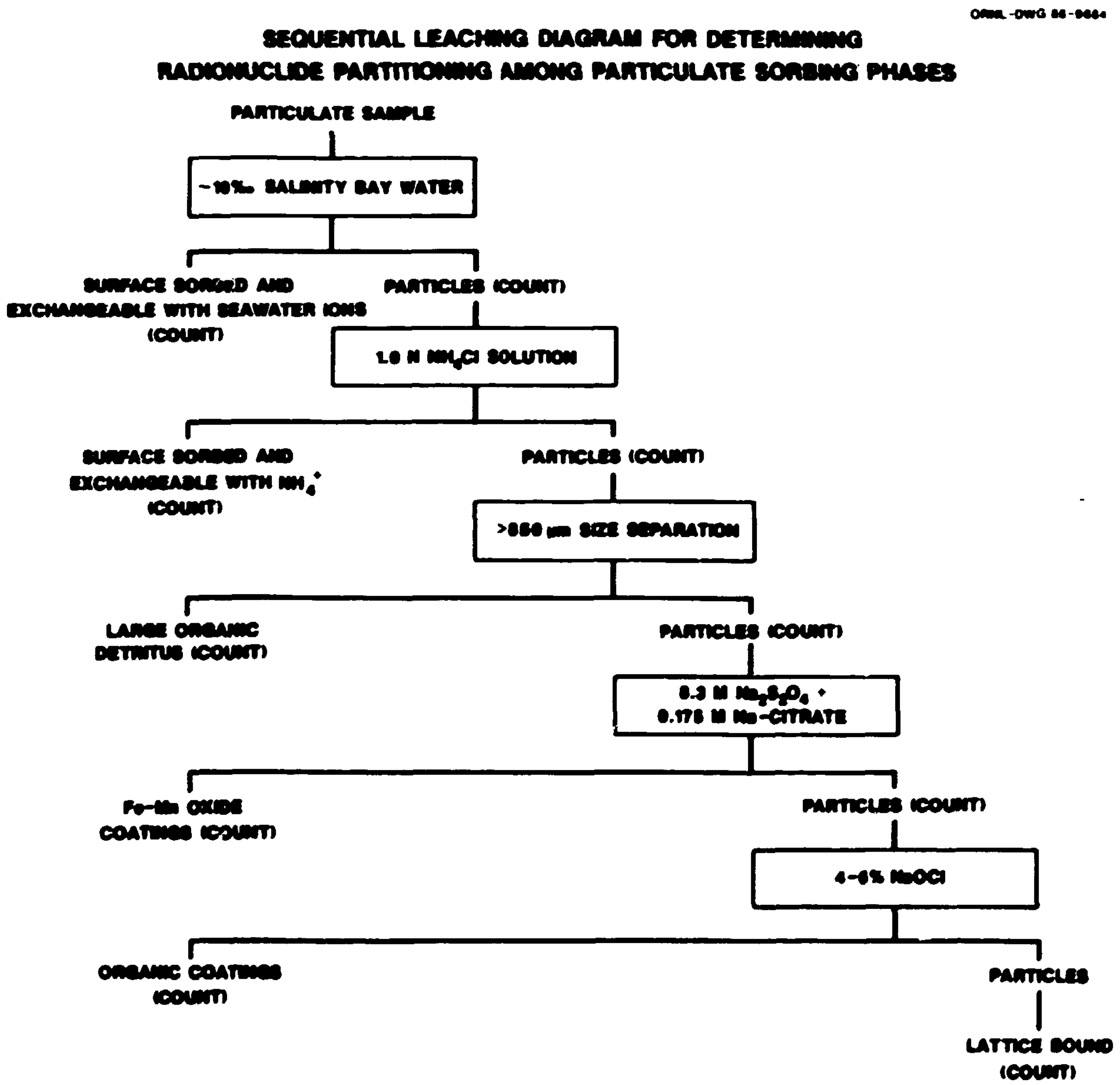

Fig. 6. The sequential leaching procedures used to identify radionuclide partitioning among the different particulate sorbing phases.

fraction of the sediment samples collected in Conowingo Pond and Upper Chesapeake Bay, the total amount of reactor-released radioactivity associated with this fraction was undetectable. The third extraction used a citrate-buffered sodium dithionite solution, which removes reducible iron, manganese oxides, and hydroxides but does not detectably alter clay minerals (Trefry 1977). This extraction was used to identify radionuclides sorbed to Fe-Mn phases which could possibly be remobilized under anoxic conditions when Fe and Mn are solubilized. The final extraction used a 4-6% purified grade of sodium hypochlorite to oxidize organics, sulfides, and radionuclides associated with these phases. After all of the above fractions had been removed, radionuclides associated with the residual material were assumed to be locked into particulate mineral lattices.

3. RESULTS AND IMPLICATIONS

3.1 RADIONUCLIDE PARTICLE-TO-WATER DISTRIBUTIONS

Many field and laboratory studies concerned with radionuclide transport and fate measure radionuclide partitioning between solid and dissolved (or complexed) phases. This partitioning is expressed quantitatively by assigning it a value or distribution coefficient (K_d), defined as

$$K_d = \frac{C_p}{C_w} ,$$

where C_p is the concentration of a specific radionuclide associated with a given weight of particles and C_w is the concentration of the radionuclide in an equal weight of water. Ideally, this ratio is a measure of the reversible equilibrium partitioning of a radionuclide between dissolved and particulate phases. The distinction between "dissolved" and "particulate," however, is generally operational (such as passage through a 0.45 µm pore-size filter or through a continuous-flow centrifuge). Consequently, particle-to-water distribution data calculated from field and laboratory data cannot often be considered true equilibrium K_ds, especially because many laboratory experiments and most natural environments are not reversible systems at equilibrium.

To avoid the equilibrium implications of a distribution coefficient, we have used a sorption ratio (R_s) which is defined as the amount of radioactivity per kilogram of particulate material divided by the amount of radioactivity per liter of water:

$$R_s = \frac{\text{(Activity/kg of sediment)}}{\text{(Activity/L of water)}} = \frac{\text{L}}{\text{kg}}$$

The particle-to-water sorption ratio (R_s) is numerically equal to K_d but is a measured empirical value that does not imply equilibrium or reversibility. R_s values are important for quantifying radionuclide sorption/solubility in models used to assess radionuclide transport and fate. Although R_s values measured in the laboratory are quite variable and are highly dependent upon experimental procedures (Higgo and Rees 1986), R_s values measured in the field provide site-specific information critical for understanding the geochemical factors that govern radionuclide fate, especially when R_s variations can be correlated with changes in water or particulate chemistry within the same field system.

Radionuclide concentration data for surface sediments, suspended matter, and water collected in the Susquehanna-Chesapeake Bay System during October 1983, April 1984, May 1985, and October 1985 are listed in Tables 1-4. Also listed in Tables 1-4 are the field-determined particle-to-water sorption ratios (R_s) for ^{137}Cs, ^{134}Cs, ^{60}Co, ^{58}Co, ^{65}Zn, and naturally occurring ^{7}Be. Field particle-to-water distributions were also determined for ^{90}Sr at two locations in May 1985 (Table 3).

3.1.1 Radionuclide Concentrations and Particle-to-Water Distributions in the Susquehanna River

Particle-to-water sorption ratios for radiocesium, radiocobalt, radiozinc, and naturally occurring ^{7}Be range from 10^4 to 10^5 in Conowingo Pond and in the Susquehanna River and Flats area downstream of Conowingo Dam. Such a high sorption ratio implies that the tendency for these radionuclides to become associated with suspended particles in freshwater environments is 10^4 to 10^5 times greater than their tendency to remain in solution. As a result, particulate phases play an important role in the removal of ^{137}Cs, ^{134}Cs, ^{60}Co, ^{58}Co, ^{65}Zn, and ^{7}Be from the water column, and sediments serve as their ultimate sink (see Section 4.1).

The removal of these radionuclides, however, may be limited by the amount of suspended particulate matter in the water column. At suspended-matter concentrations of 10 mg/L (typical during normal flow conditions), there is 10^5 times more water than suspended matter in a liter. Consequently, the strong tendency for ^{137}Cs, ^{134}Cs, ^{60}Co, ^{58}Co,

Table 1. Radionuclide concentrations in surface sediments, suspended matter, and water of the Susquehanna-Chesapeake Bay System[a], October 1983

Sample (Date)	Nuclide	Surface Sediment (pCi/kg)	Susp. matter (pCi/kg)	Water (fCi/L)	Site-specific field R_s
Conowingo	Cs-137	600±6	1,880±70	23.0±0.6	8.2×10^4
Pond	Cs-134	165±3	910±30	13.6±0.5	6.7×10^4
(3 Oct-83)	Co-60	285±6	—	—	—
	Zn-65	285±7	3,250±130	13.6±1.9	2.3×10^5
S ppt ≤ 0.1[b]	Be-7	215±29	11,800±600	55.5±5.6	2.1×10^5
mg/L = 6[c]	Pb-214	1,180±48	1,110±205	ND	—
C % = 10.6[d]	Pb-210U	1,340±180	3,510±450	ND	—
	K -40	18,350±110	15,300±1200	—	—
Susquehanna	Cs-137	510±6	1,750±70	20.0±1.0	8.8×10^4
Mouth	Cs-134	210±4	670±30	11.2±1.1	6.0×10^4
(3 Oct-83)	Co-60	61±8	—	—	—
	Zn-65	180±7	350±70	1.5±0.6	1.8×10^5
S ppt ≤ 0.1	Be-7	580±32	11,700±500	57.3±6.0	2.0×10^5
mg/L = 6	Pb-214	1,000±70	1,150±190	ND	—
C % = 10.0	Pb-210U	850±120	2,310±650	ND	—
	K -40	17,040±114	15,400±1100	—	—
Annapolis	Cs-137	435±9	330±110	60.9±0.7	5.4×10^3
Bridge	Cs-134	ND	ND	1.9±0.1	—
(4 Oct-83)	Co-60	ND	—	—	—
	Zn-65	ND	ND	ND	—
S ‰ = 12	Be-7	1,550±60	4,600±630	24.4±4.1	1.9×10^5
mg/L = 4	Pb-214	610±40	950±180	ND	—
C % = 22.5	Pb-210U	2,900±160	2,700±400	ND	—
	K -40	16,670±170	11,900±1800	—	—
Calvert	Cs-137	575±5	170±103	72.1±0.8	2.4×10^3
Cliffs	Cs-134	ND	ND	2.2±0.3	—
(5-Oct-83)	Co-60	82±7	—	—	—
	Co-58	ND	2,930±100	6.2±1.0	4.9×10^5
S ppt = 14	Zn-65	9±5	ND	ND	—
mg/L = 4	Ag-110M	12±3	ND	ND	—
C % = 15.7	Be-7	28±19	4,500±580	73.0±8.3	6.2×10^4
	Pb-214	725±36	380±160	ND	—
	Pb-210U	2,820±160	3,770±480	ND	—
	K -40	14,190±100	10,800±1700	—	—

[a] Radionuclide activities have been decay-corrected to the date of sample collection. Statistical counting errors are expressed as one-sigma. ND indicates that the radionuclide activity was below detection limits, and dashes (-) indicate that the sample was not analyzed or not calculated.

[b] S ppt = water salinity in parts per thousand.

[c] mg/L = suspended matter concentration in the collected sample.

[d] C % = percent total carbon concentration of the suspended matter.

Table 2. Radionuclide concentrations in surface sediments, suspended matter, and water of the Susquehanna-Chesapeake Bay System[a], April 1984

Sample (Date)	Nuclide	Surface Sediment (pCi/kg)	Susp. Matter (pCi/kg)	Water (fCi/L)	Site-specific field R_s
Conowingo	Cs-137	505±7	555±26	2.3±0.4	2.4×10^5
Pond	Cs-134	51±5	39±13	0.5±0.2	7.8×10^4
(4 Apr-84)	Co-60	67±5	71±26	1.1±0.3	6.5×10^4
	Zn-65	89±9	350±77	0.1±0.3	—
S ppt ≤ 0.1[b]	Be-7	740±35	8,700±320	380.0±5.0	2.3×10^4
mg/L = 15[c]	Pb-214	1,200±30	1,200±40	ND	—
C % = 2.3[d]	Pb-210U	1,130±120	1,985±150	ND	—
	K -40	19,600±130	23,800±520	—	—
Susquehanna	Cs-137	60±1	570±20	2.5±0.4	2.3×10^5
Mouth	Cs-134	10±1	ND	ND	—
(3 Apr-84)	Co-60	10±1	37±16	1.0±0.3	3.7×10^4
	Zn-65	27±1	73±20	0.4±0.3	—
S ppt ≤ 0.1	Be-7	680±14	13,700±200	215.0±5.0	6.5×10^4
mg/L = 19	Pb-214	250±20	2,110±160	ND	—
C % = 3.2	Pb-210U	160±50	2,280±180	ND	—
	K -40	5,040±25	25,000±400	—	—
Annapolis	Cs-137	535±7	660±30	29.4±0.6	2.2×10^4
Bridge	Cs-134	7±3	ND	0.7±0.3	—
(4 Apr-84)	Co-60	15±5	ND	ND	—
	Co-58	ND	ND	ND	—
S ppt = 9	Zn-65	ND	ND	ND	—
mg/L = 24	Be-7	3,350±84	12,800±300	545.0±15.0	2.3×10^4
C % = 10.6	Pb-214	630±30	1,020±70	ND	—
	Pb-210U	2,110±160	3,380±220	ND	—
	K -40	23,300±160	20,600±500	—	—
Calvert	Cs-137	24±4	240±30	41.3±0.7	5.8×10^3
Cliffs	Cs-134	5±2	ND	3.1±0.5	—
(5 Apr-84)	Co-60	78±3	44±25	0.9±0.6	4.9×10^4
	Co-58	39±3	32±15	ND	—
S ppt = 8	Zn-65	ND	ND	ND	—
mg/L = 11	Ag-110M	15±3	26±21	ND	—
C % = 33.7	Be-7	1,050±25	11,100±300	340.0±10.7	3.3×10^4
	Pb-214	720±20	280±50	ND	—
	Pb-210U	1,430±120	2,640±150	ND	—
	K -40	0200±100	5,500±500	—	—

[a] Radionuclide activities have been decay-corrected to the date of sample collection. Statistical counting errors are expressed as one-sigma. ND indicates that the radionuclide activity was below detection limits, and dashes (-) indicate that the sample was not analyzed or not calculated.

[b] S ppt = water salinity in parts per thousand.

[c] mg/L = suspended matter concentration in the collected sample.

[d] C % = percent total carbon concentration of the suspended matter.

Table 3. Radionuclide concentrations in surface sediments, suspended matter, and water of the Susquehanna-Chesapeake Bay System[a], May 1985

Sample (Date)	Nuclide	Surface Sediment (pCi/kg)	Susp. matter (pCi/kg)	Water (fCi/L)	Site-specific field R_s
Conowingo	Cs-137	405±2	2,040±30	160 ±2	1.3×10^4
Pond I	Cs-134	135±2	1,400±22	125 ±2	1.1×10^4
(16 May-85)	Co-60	345±3	265±40	1.2±0.3	2.2×10^5
	Zn-65	215±6	695±30	6.5±0.3	1.1×10^5
S ppt $\leq$ 0.1	Be-7	340±9	6,010±175	80.6±2.5	7.5×10^4
mg/L = 12	Pb-214	830±20	980±70	ND	—
	Pb-210U	510±90	1,500±210	ND	—
	K -40	11,800±40	17,300±350	—	—
Conowingo	Cs-137	—	1,930±40	135 ±2	1.4×10^4
Pond II	Cs-134	—	1,320±30	105 ±2	1.3×10^4
(17 May-85)	Co-60	—	410±50	1.3±0.4	3.1×10^5
	Zn-65	—	695±50	6.1±0.7	1.1×10^5
S ppt $\leq$ 0.1[b]	Be-7	—	8,230±340	130 ±5	6.4×10^4
mg/L = 17[c]	Pb-214	—	1,270±70	ND	—
C % = 8.9[d]	Pb-210U	—	2,050±275	ND	—
	K -40	—	20,100±500	—	—
	Sr-90	—	51±50	150 ±20	3×10^2
Susquehanna	Cs-137	125±2	680±55	6 ±0.4	1.1×10^5
Mouth	Cs-134	23±1	150±30	2 ±0.1	7.5×10^4
(15 May-85)	Co-60	15±1	265±75	1.3±0.5	2.0×10^5
	Zn-65	14±3	220±100	1.5±0.5	1.5×10^5
S ppt $\leq$ 0.1	Be-7	345±11	5,280±385	43.5±3.2	1.2×10^5
mg/L = 7	Pb-214	620±20	1,070±70	ND	—
C % =12.4	Pb-210U	520±90	1,900±190	ND	—
	K -40	7,050±40	21,500±950	—	—
	Sr-90	—	—	220 ±40	—
Annapolis	Cs-137	425±7	365±45	35 ±0.5	1.0×10^4
Bridge	Cs-134	9±1	ND	2 ±0.1	—
(20 May-85)	Co-60	10±4	ND	0.9±0.4	—
	Zn-65	ND	ND	ND	—
S ppt = 10.8	Be-7	350±30	12,500±420	105 ±4.2	1.2×10^5
mg/L = 5	Pb-214	920±30	770±120	ND	—
C % = 16.9	Pb-210U	2,000±140	2,890±260	ND	—
	K -40	18,500±120	14,800±900	—	—
	Sr-90	—	7±170	100 ±30	7×10^1
Calvert	Cs-137	345±3	350±150	50 ±0.6	7.0×10^3
Cliffs	Cs-134	5±1	ND	1 ±0.2	—
(21 May-85)	Co-60	130±3	605±140	0.7±0.4	8.6×10^5
	Co-58	14±2	1,130±100	2.5±0.3	4.5×10^5
S ppt = 13.5	Ag-110M	ND	ND	—	—
mg/L = 2	Be-7	105±14	16,100±1060	170 ±6	9.5×10^4
C % = 32.6	Pb-214	730±60	1,760±210	ND	—
	Pb-210U	3,100±250	1,800±640	ND	—
	K -40	12,200±670	7,130±1910	—	—

[a] Radionuclide activities have been decay-corrected to the date of sample collection. Statistical counting errors are expressed as one-sigma. ND indicates that the radionuclide activity was below detection limits, and dashes (-) indicate that the sample was not analyzed or not calculated.

[b] S ppt = water salinity in parts per thousand.

[c] mg/L = suspended matter concentration in the collected sample.

[d] C % = percent total carbon concentration of the suspended matter.

Table 4. Radionuclide concentrations in surface sediments, suspended matter, and water of the Susquehanna-Chesapeake Bay System[a], October 1985

Sample (Date)	Nuclide	Surface Sediment (pCi/kg)	Susp. matter (pCi/kg)	Water (fCi/L)	Site-specific field R_s
Conowingo	Cs-137	640±5	1,290±50	13.0± 0.6	9.9×10^4
Pond I	Cs-134	175±3	510±40	7.8± 0.2	6.5×10^4
(2 Oct-85)	Co-60	180±4	2,630±80	4.2± 0.4	6.3×10^5
	Zn-65	120±6	1,390±110	5.2± 0.4	2.7×10^5
S ppt ≤ 0.1[b]	Be-7	470±27	5,660±460	154 ± 6.0	3.7×10^4
mg/L = 5.3[c]	Pb-214	1,290±150	1,790±160	8.8± 0.6	—
	Pb-210U	910±160	1,650±340	26.2± 2.3	6.3×10^4
	K -40	25,000±100	23,200±900	—	—
Conowingo	Cs-137	—	670±70	3.8± 0.6	1.8×10^5
Pond II	Cs-134	—	100±30	1.4± 0.3	7.1×10^4
(3 Oct-85)	Co-60	—	310±70	1.8± 0.5	1.7×10^5
	Zn-65	—	720±460	0.7± 0.4	—
S ppt ≤ 0.1	Be-7	—	12,050±610	202 ± 6.0	6.0×10^4
mg/L = 4.6	Pb-214	—	1,520±110	13.4± 0.6	—
	Pb-210U	—	2,570±310	10.0± 2.4	2.6×10^5
	K -40		24,400±1100	—	—
Conowingo	Cs-137	—	700±60	10.8± 0.8	6.5×10^4
Pond III	Cs-134	—	240±30	5.0± 0.5	4.8×10^4
(4 Oct-85)	Co-60	—	330±60	2.0± 0.4	1.7×10^5
	Zn-65	—	390±120	1.4± 0.4	2.8×10^5
S ppt ≤ 0.1	Be-7	—	11,140±430	159 ±15	7.0×10^4
mg/L = 5.2	Pb-214	—	1,560±130	5.3± 0.6	—
	Pb-210U	—	1,970±360	4.7± 2.4	4.2×10^5
	K -40		22,400±900	—	—
Susquehanna	Cs-137	220±3	650±40	11.2± 0.9	5.8×10^4
Mouth	Cs-134	50±1	190±20	4.0± 0.7	4.8×10^4
(4 Oct-85)	Co-60	26±1	620±40	1.1± 0.4	5.6×10^5
	Zn-65	16±4	400±60	1.8± 0.4	2.2×10^5
S ppt ≤ 0.1	Be-7	510±25	7,470±240	86.9± 4.6	8.6×10^4
mg/L = 15.8	Pb-214	1,130±20	1,320±220	3.7± 0.7	—
	Pb-210U	650±90	2,270±240	6.3± 2.0	3.6×10^5
	K -40	11,300±400	20,400±600	—	—
Grove	Cs-137		540±40	14.9±0.6	3.6×10^4
Point	Cs-134		19±12	0.8±0.3	2.4×10^4
(8 Oct-85)	Co-60		90±40	1.2±0.3	7.5×10^4
	Zn-65		120±40	0.5±0.4	2.4×10^5
S ppt = 3.0	Be-7		1,720±230	46.3±5.4	3.7×10^4
mg/L = 14.0	Pb-214		1,070±80	ND	—
	Pb-210U		1,270±220	ND	—
	K -40		25,300±700	—	—

Table 4. (continued)

Sample (Date)	Nuclide	Surface Sediment (pCi/kg)	Susp. matter (pCi/kg)	Water (fCi/L)	Site-specific field R_s
Howell	Cs-137	—	560±30	23.4±0.8	2.4×10^4
Point	Cs-134	—	ND	2.9±0.7	—
(9 Oct-85)	Co-60	—	ND	ND	—
	Zn-65	—	ND	ND	—
S ppt = 5.5	Be-7	—	1,100±200	20.5±4.7	5.4×10^4
mg/L = 9.8	Pb-214	—	1,060±90	ND	—
	Pb-210U	—	1,240±240	ND	—
	K -40	—	22,100±600	—	—
Annapolis	Cs-137	160±2	210±40	63.1±0.8	3.3×10^3
Bridge	Cs-134	ND	ND	2.0±0.3	—
(8 Oct-85)	Co-60	2±1	ND	ND	—
	Zn-65	ND	ND	ND	—
S ppt = 15.2	Be-7	450±25	4,210±250	33.3±3.9	1.3×10^5
mg/L = 3.4	Pb-214	840±30	610±90	ND	—
	Pb-210U	1,890±130	1,560±230	ND	—
	K -40	7,700±40	13,700±800	—	—
Calvert	Cs-137	240±3	400±60	87.9±1.4	4.6×10^3
Cliffs	Cs-134	ND	ND	ND	—
(7 Oct-85)	Co-60	50±2	360±70	0.9±0.2	4.0×10^5
	Co-58	8±3	390±60	1.9±0.7	2.1×10^5
S ppt = 19.9	Ag-110M	ND	ND	ND	—
mg/L = 4.7	Be-7	69±27	7,190±420	42.2±6.3	1.7×10^5
	Pb-214	530±20	490±90	ND	—
	Pb-210U	2,080±100	4,050±310	ND	—
	K -40	7,500±50	16,000±1200	—	—

[a] Radionuclide activities have been decay-corrected to the date of sample collection. Statistical counting errors are expressed as one-sigma. ND indicates that the radionuclide activity was below detection limits, and dashes (-) indicate that the sample was not analyzed or not calculated.

[b] S ppt = water salinity in parts per thousand.

[c] mg/L = suspended matter concentration in the collected sample.

^{65}Zn, and ^{7}Be to become associated with particles is offset by the large amount of water relative to suspended matter. As a result, about 50% of the total radioactivity will be in the dissolved phase during low to normal flow conditions, and the remaining 50% will be associated with the suspended particles. For example, during the low-flow conditions of October 1983 (Table 1) the concentration of ^{137}Cs on suspended matter in Conowingo Pond was 1800 pCi/kg and its concentration in the water phase was 0.023 pCi/L. With a Pond suspended-matter concentration of 6 mg/L (Table 1), the total amount of ^{137}Cs associated with particles in a liter of water is calculated to be 0.011 pCi/L:

$$(1800 \text{ pCi/kg}) (10^{-6} \text{ kg/mg}) (6 \text{ mg/L}) = 0.011 \text{ pCi/L}$$

Consequently, at a suspended matter concentration of 6 mg/L about 65% of the total ^{137}Cs radioactivity in a liter of water (0.034 pCi/L) is in the water phase (0.023 pCi/L) and the remaining 35% (0.011 pCi/L) is associated with suspended particles. During high-flow conditions or during floods, however, when suspended-matter concentrations are approximately 100 mg/L (Appendix C), more than 90% of the total radioactivity will be transported with particles and only 10% will be transported in the water. Because particle and radionuclide transport in the Susquehanna is dominated by floods with high suspended-matter loads (Helz et al. 1985), reactor-released ^{137}Cs, ^{134}Cs, ^{60}Co, ^{58}Co, and ^{65}Zn will be primarily transported with fine-particles during high discharge events.

The measured sorption ratio for ^{90}Sr in Conowingo Pond is about 3×10^{2}, or about three orders of magnitude less than those measured for radiocesium, radiocobalt, or radiozinc (Table 3). This implies that much of the reactor-released ^{90}Sr will remain in solution and will be transported downstream in the water phase, even during periods of high discharge when suspended-matter concentrations greatly increase.

3.1.2 Radionuclide Concentrations and Particle-to-Water Distributions in Chesapeake Bay

The extent to which the sorption behaviors and particle-to-water distribution of radiocesium, radiocobalt, and ^{7}Be are affected by estuarine salinity when released or transported into Chesapeake Bay is illustrated in Fig. 7. The average particle-to-water sorption ratios for ^{137}Cs are 1.1×10^{5} in the fresh waters of Conowingo Pond and 5.0×10^{3} in the more saline (10-20 ppt) bay waters near Calvert Cliffs. This two-order-of-magnitude decrease primarily reflects the fact that in saline bay water, radiocesium has to compete with saltwater cations (Na^{+}, K^{+}, Ca^{++}, Mg^{++}) for sorption sites on particles. As a result, most of the radiocesium introduced into saline waters (via global fallout or reactor releases) remains in solution and dissolved ^{137}Cs concentrations in Chesapeake Bay are proportionally higher at higher salinities.

Whereas radiocesium sorption is clearly diminished with increasing salinity, the effects of estuarine salinity on the sorption behavior of ^{60}Co, ^{7}Be and ^{65}Zn are less clear. Although the sorption ratios of both ^{60}Co and ^{7}Be appear lowered within the estuarine turbidity zone relative to the Susquehanna River (Fig. 7), they increase in the saline waters near Calvert Cliffs to values which are similar to or exceed their R_s values in freshwater. A likely explanation for the lower R_s values in the turbidity zone is that older (unlabeled) particles dilute the concentration of ^{60}Co and ^{7}Be associated with the suspended matter in this zone. Such a dilution may result from the resuspension of bottom sediments (depleted in ^{60}Co and ^{7}Be) by radioactive decay.

No data are presented for ^{65}Zn in Fig. 7 because, except for one sample, it could not be detected in the saline waters of the Upper Bay. The average particle-to-water sorption ratio for ^{65}Zn in Conowingo Pond was 2.1×10^{5}. In the one instance in which ^{65}Zn was detected on the suspended particles in saline water (3 ppt salinity at Grove Point, Table 4), its sorption ratio was 2.4×10^{5} and thus appears to be unaffected by salinity.

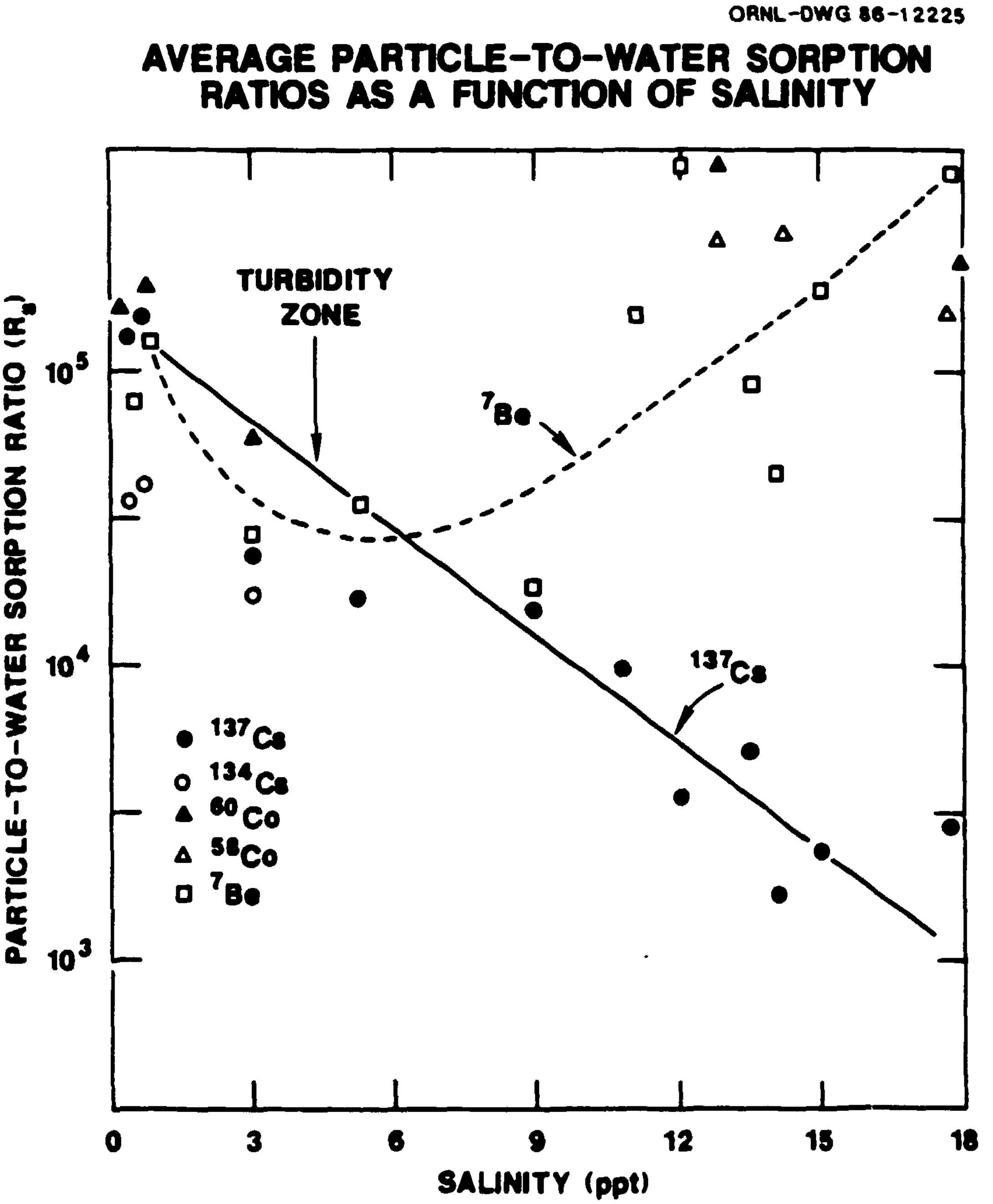

Fig. 7. The effect of salinity on the measured particle-to-water distribution for ^{137}Cs (●), ^{134}Cs (o), ^{60}Co (▲), ^{58}Co (Δ), and ^{7}Be (□). The two freshwater points are an average of all the samples collected in Conowingo Pond and Susquehanna Flats, respectively. It is apparent that the sorption of radiocobalt and ^{7}Be is unaffected by estuarine salinity, but that the sorption of radiocesium is strongly dependent on salinity.

The field R_s for ^{90}Sr at Annapolis is slightly lower than the sorption ratio measured in Conowingo Pond (Table 3). The lower sorption ratio in saline areas might be expected as a result of cation competition for sorption sites on the suspended matter, but the major uncertainties associated with the quantification of ^{90}Sr on suspended particles make the comparison tenuous, at best.

3.2 TEMPORAL VARIATIONS IN RADIONUCLIDE CONCENTRATIONS AND PARTICLE-TO-WATER DISTRIBUTIONS

In addition to the spatial or salinity effects, radionuclide concentrations and particle-to-water distributions are also affected by temporal variations in radionuclide input via effluent discharges (Appendix B) and by seasonal changes in environmental conditions such as river flow, biological productivity, and sediment resuspension (Appendix D). Increases in river flow or in biological particle production can respectively dilute dissolved and particulate radionuclide concentrations, and can thus significantly affect particle-to-water distributions. Sediment resuspension near reactor sites can increase the concentration of particle-bound reactor-released radionuclides and thereby raise particle-to-water sorption ratios. On the other hand, sediment resuspension in relatively uncontaminated areas can also dilute the concentration of particle-bound reactor-released radionuclides and thereby lower measured particle-to-water sorption ratios. For example, sediment resuspension in the Susquehanna River (upstream of the PB discharge site) can dilute reactor-released radionuclide concentrations on the suspended matter in Conowingo Pond when transported into the area. In addition, the resuspension of older sediments (i.e., sediments that have been in the system long enough for radioactive decay to have depleted the concentration of short-lived radionuclides) can also dilute radionuclide concentrations on the suspended matter. As discussed previously, the low particle-to-water sorption ratios that occur in the estuarine turbidity zone (Fig. 7), probably reflect the dilution of radionuclide concentrations on suspended matter by the resuspension of older bottom sediments, and by the input of "unlabeled" particles from other sources.

The effects of sediment resuspension on the concentration, transport, and biological fate of reactor-released radionuclides in the Susquehanna-Chesapeake Bay System greatly depend on the half-life of the radionuclide. For relatively long-lived radionuclides such as ^{137}Cs (30-year half-life), the effect of sediment resuspension on the particulate ^{137}Cs concentration and particle-to-water sorption ratios is relatively minor; the increase is on the order of a factor of two (Conowingo Pond in Table 4). For ^{60}Co (5-year half-life) and ^{134}Cs (2-year half-life), sediment resuspension is relatively more important and can affect measured concentrations and sorption ratios by a factor of about 5 in the Susquehanna River and Upper Chesapeake Bay. For short-lived radionuclides such as ^{65}Zn, ^{58}Co, and ^{7}Be (with half-lives of 245, 71, and 53 days, respectively) sediment resuspension may affect measured concentrations and particle-to-water distributions by more than one order of magnitude. Consequently, the quantification of sediment resuspension is as important as the quantification of freshwater flow in radionuclide-transport/biological-uptake/human-dose models and therefore must be explicitly considered.

3.2.1 Temporal Variations in Reactor Radionuclides

The temporal effects of reactor effluent discharges are exemplified by the much higher concentrations of dissolved ^{137}Cs and ^{134}Cs in the water of Conowingo Pond and Susquehanna Flats during May 1985 (Table 3) relative to the concentrations measured previously in October 1983 or April 1984. As indicated in Table B-2, Conowingo Pond received relatively large radiocesium releases from the PB Atomic Power Station during May 1985. The high concentrations of dissolved ^{137}Cs and ^{134}Cs resulting from these releases caused the calculated particle-to-water distribution ratios to drop to about 10^4 relative the field R_s's measured in October 1983, April 1985, or October 1985, which were about 10^5. Because the measured concentrations and particle-to-water distributions of reactor-released and natural radionuclides in Conowingo Pond, made on successive days during May 1985 (May 16 and May 17 in Table 3) were almost duplicates; we suggest that the variations associated with analyzing two different samples collected under relatively constant conditions are minor relative to variations that may occur as a result of changes in reactor input, biological productivity, or meteorological (storm, seasonal) conditions.

The effect of changing environmental conditions on measured radionuclide concentrations and distributions is best illustrated in the Conowingo Pond samples collected for three consecutive days during the course of a rainstorm in October 1985 (Table 4). The dissolved and particulate concentrations of reactor-released radionuclides were much greater in the October 2 sample (Conowingo Pond I in Table 4) collected prior to the rainstorm than in the October 3 sample (Conowingo Pond II) collected during the rainstorm or in the October 4 sample (Conowingo Pond III) collected a day after the storm event. The decrease in dissolved ^{137}Cs, ^{134}Cs, ^{60}Co, and ^{65}Zn probably reflects dilution by the increased river flow and freshwater input associated with this prolonged rainfall event (Appendix D). The decrease in reactor-radionuclide concentrations on the suspended matter probably reflects (1) the dilution of the particulate material by sediment erosion and resuspension in upstream areas and (2) the flushing reactor-tagged suspended particles from the reservoir during the storm event. The flushing of particulate material with high ^{137}Cs, ^{134}Cs, ^{60}Co, and ^{65}Zn concentrations is supported by the relatively high concentrations of these nuclides on the suspended matter collected below the dam, at the mouth of the Susquehanna, on October 4, 1985 (Susquehanna Flats in Table 4).

Resuspension of particles containing relatively high concentrations of fallout ^{137}Cs and reactor-released radionuclides during periods of high flow is also exemplified by comparing the bottom sediment data for Susquehanna Flats during the low-flow conditions of October 1983 (Table 1; Appendix D) and during the high-flow conditions of April 1984 (Table 2; Appendix D). During the low-flow conditions of October 1983, sediments near the mouth of the Susquehanna River were fine grained and contained appreciable anthropogenic radionuclide concentrations

(Table 1). In April 1984, sediment samples collected at the same site were much coarser grained and showed a sharp decrease in anthropogenic radionuclide concentrations (Table 2). This seasonal increase in grain size and the associated decrease in radionuclide concentrations probably reflects the erosion, resuspension, and down-bay transport in the spring of the fine-grained particles deposited in this area during the low-flow conditions of the previous summer and fall. Increased sediment erosion and resuspension during spring runoff also accounts for the large increase in suspended-matter concentration that was observed throughout the Susquehanna-Chesapeake Bay System in April 1984 relative to concentrations measured during the other periods of sample collection. Sediment resuspension and downstream transport may also account for the detectable concentrations of ^{134}Cs and ^{60}Co in the sediments at Annapolis during April 1984 and May 1985.

The importance of particle dynamics and dilution is exemplified in comparing the concentration of long-lived fallout ^{137}Cs on the sediments and suspended matter at sites removed from the influence of reactor releases, such as at Annapolis (Tables 1-4). In October 1983 (Table 1) and May 1985 (Table 2) when freshwater flows were low (Appendix D) and the production of in situ biological particles within the water column was high, ^{137}Cs concentrations on the bottom sediments at Annapolis actually exceeded concentrations on the suspended matter. During periods of high freshwater flow, however, such as during April 1984 (Appendix D), concentrations of ^{137}Cs on the suspended matter at Annapolis exceeded October and May concentrations by about a factor of two and were very similar to concentrations on the bottom sediments. This seasonal variation in particulate ^{137}Cs concentrations is explained by the interplay of three processes, one sedimentological, one geochemical, and one biological: (1) most of the sediment accumulating in the Annapolis area originates from upstream (freshwater) areas and is resuspended, transported, and deposited during storms or seasonal periods of high flow, (2) particles transported downstream during periods of high flow contain relatively high concentrations of ^{137}Cs because particle-to-water sorption ratios are much higher in upstream (freshwater) areas than in mesohaline areas, and (3) during periods of low flow, biological particle production in the water column dilutes the concentration of ^{137}Cs on the suspended matter.

3.2.2 Temporal Variations in Natural Radionuclides

As with the reactor-released radionuclides, the dissolved concentrations and particle-to-water distributions of naturally occurring and atmospherically derived ^{7}Be in Conowingo Pond and in Upper Chesapeake Bay are governed to a great extent by radionuclide input and by environmental conditions. Dissolved ^{7}Be concentrations were greatest in April 1984 (Table 2) when its input via atmospheric washout by precipitation was greatest (Appendix A). The average dissolved ^{7}Be concentration in the four large-volume water samples collected in October 1983 was 0.06 pCi/L (Table 1), whereas the average dissolved concentration at the same four sites in April 1984 was 0.32 pCi/L (Table 2). The highest dissolved ^{7}Be concentration was measured in a water sample collected on April 4, 1984, at the Annapolis site during a spring rain that delivered 1.8 cm of precipitation to the Bay surface (Appendix D). Within a day after this rain event, the dissolved ^{7}Be concentrations decreased by a factor of two in a nearby sample collected at Calvert Cliffs and approached dissolved values measured in the system prior to the rain event (Table 2).

The effects of input variations on dissolved and particulate concentrations of ^{7}Be are also exemplified in the three consecutive daily samples collected in Conowingo Pond prior to, during, and after a period of prolonged rain fall from October 2 to 4, 1985 (Table 4). [As an aside, this rainstorm delivered 0.15 pCi/cm^{2} (Appendix A) or a total of 0.06 Ci of ^{7}Be to the surface of Conowingo Pond. This natural event introduced more natural gamma radioactivity to Conowingo Pond than the combined releases of ^{137}Cs, ^{134}Cs, ^{60}Co, and ^{65}Zn during the entire month of October 1985 (Appendix B)]. While storm events reduce dissolved and particulate concentrations of ^{137}Cs, ^{134}Cs, ^{60}Co, and

^{65}Zn by dilution and flushing from Conowingo Pond, dissolved and particulate concentrations of ^{7}Be sharply increased with the precipitation event.

On the day after this rainstorm (Conowingo Pond III in Table 4), dissolved ^{7}Be concentrations decreased by about 20%, reflecting rapid sorption onto suspended-particulate material. Because the particle-to-water sorption ratio for ^{7}Be is similar to sorption ratios measured for ^{137}Cs, ^{134}Cs, ^{60}Co, and ^{65}Zn in Conowingo Pond (Tables 1-4), the ^{7}Be data imply that the above reactor-released radionuclides may also be removed from the dissolved phase at a rate of about 20% per day by rapid sorption onto suspended particles in Conowingo Pond. A more quantitative discussion of radionuclide sorption and water column removal rates is presented in Section 4.1 of this report.

3.3 PARTICULATE COMPOSITIONS AND RADIONUCLIDE PARTITIONING AMONG VARIOUS SORBING PHASES

Variations in the particle-to-water distribution of specific radionuclides may also reflect changes in the physicochemical properties of the sorbing particle. For example, fine-grained particles generally have a greater affinity for radionuclides than do coarser particles because they have greater surface areas per unit weight and are enriched in layered aluminosilicate minerals, which contain more binding sites per unit area. In addition, coarser particles are generally enriched in quartz (silica), which is relatively nonreactive compared to organic or clay (aluminosilicate) particles in natural waters. Different aluminosilicate minerals also have different affinities for radionuclides. Illite, for example, exhibits a very high affinity for ^{137}Cs and ^{134}Cs as a result of cesium substitution for potassium in the clay mineral lattice (Tamura and Jacobs 1960).

In this project, we have documented variations in the elemental and mineralogical composition of the suspended matter and bottom sediments in the Susquehanna River and Upper Chesapeake Bay (Appendix E). We have also conducted laboratory extraction or leaching experiments to identify the extent of radionuclide partitioning among the different particulate sorbing phases (i.e., particulate surfaces, organic detritus or coatings, Fe-Mn coatings, and mineral lattices). The results from these leaching experiments are summarized in Table 5 for samples collected in October 1983 and April 1984, and in Table 6 for samples collected in October 1985 and May 1985. The average results for all the suspended matter and bottom sediment samples collected in Conowingo Pond are also illustrated in Fig. 8.

It is apparent from Fig. 8 that there are differences in the nuclide chemical associations on suspended matter relative to bottom sediments and that radiocesium, for example, is much more readily desorbed from suspended matter than from bottom sediments. Previous work has shown that fallout ^{137}Cs in fresh water environments is irreversibly sorbed by replacing K^+ in clay mineral lattices (Lomenick and Tamura 1965; Eyman and Kevern 1975; Olsen et al. 1981b; Zucker et al. 1984). This is consistent with our data indicating that about 90% of the ^{137}Cs associated with bottom sediments in Conowingo Pond is locked within the mineral lattice (Fig. 8). Cesium-137 associated with the suspended matter, however, appears to be somewhat more exchangeable, with about 70% irreversibly sorbed in the mineral lattice, despite the fact that the sediments and suspended matter are mineralogically very similar (Appendix E). One possible explanation for this difference is that <u>in situ</u>-produced biogenic particles, which expurgate when placed in saline water, form a major component of the suspended matter. Another possible explanation is that recently introduced reactor radiocesium is rapidly sorbed to particle surfaces and is thus exchangeable unless enough time has elapsed for the radiocesium to become nonexchangeable by diffusion into the mineral lattice.

The latter explanation is supported by the ^{134}Cs data. Only about 50% of the recently introduced reactor ^{134}Cs appears to be locked into particulate mineral lattices (Fig. 8), and as much as 100% of the ^{134}Cs on the suspended matter could, on occasion (April 1984), be removed by exchange with seawater cations or NH_4^+ (Table 5). In other words, it appears that <u>the irreversible sorption of radiocesium</u>

Table 5. Radionuclide associations with chemical phases on sediments and suspended matter

Nuclide	Nuclide activity (pCi/g)	Cation exch. (leached)[a]	Mn,Fe exch. (leached)[b]	Organic exch. (leached)[c]	Non-exch. (in lattice)[d]
Conowingo Pond suspended matter (April 1984)					
Cs-137	0.55±0.03	30%	N.D.	N.D.	70%
Cs-134	0.04±0.01	100%	-	-	0%
Co-60	0.07±0.03	N.D.	100%	-	0%
Zn-65	0.17±0.04	100%	-	-	0%
Be-7	8.7 ±0.3	N.D.	100%	-	0%
K-40	23.8 ±0.5	N.D.	N.D.	N.D.	100%
Ra-226	1.2 ±0.04	55%	N.D.	5%	40%
Pb-210	3.2 ±.14	50%	10%	10%	30%
Conowingo Pond sediments (April 1984)[e]					
Cs-137	0.50±0.02	N.D.	N.D.	5%	95%
Cs-134	0.06±0.01	25%	N.D.	30%	45%
Co-60	0.07±0.01	5%	80%	10%	0%
Zn-65	0.17±0.02	45%	40%	15%	0%
Be-7	0.76±0.09	N.D.	100%	-	0%
K-40	20.4 ±0.3	N.D.	N.D.	N.D.	100%
Ra-226	1.33±0.05	5%	5%	5%	85%
Pb-210	2.54±0.16	N.D.	10%	N.D.	90%
Conowingo Pond sediments (October 1983)[f]					
Cs-137	0.62±0.03	5%	N.D.	5%	90%
Cs-134	0.15±0.02	35%	5%	5%	55%
Co-60	0.18±0.03	10%	80%	10%	0%
Zn-65	0.28±0.05	5%	95%	-	0%
Be-7	-	-	-	-	-
K-40	20.2 ±0.5	N.D.	N.D.	N.D.	100%
Ra-226	1.29±0.06	15%	5%	N.D.	80%
Pb-210	2.95±0.20	5%	35%	N.D.	60%
Calvert Cliffs sediments (April 1984)[g]					
Cs-137	0.24±0.01	5%	N.D.	15%	80%
Cs-134	-	-	-	-	-
Co-60	0.08±0.01	N.D.	15%	5%	80%
Co-58	0.02±0.004	15%	70%	35%	40%
Ag-110M	0.01±0.006	N.D.	20%	65%	15%
Be-7	0.90±0.06	N.D.	65%	15%	30%
K-40	10.4 ±0.2	N.D.	N.D.	N.D.	100%
Ra-226	0.69±0.04	N.D.	5%	N.D.	95%
UPb-210	2.30±0.14	N.D.	10%	15%	75%

[a] Percent of initial particulate radionuclide concentration first leached with saline bay water and then leached with a 1.0 M NH_4Cl solution which removes exchangeable cations on particle surfaces.

[b] Percent of the initial particulate radionuclide concentration leached with a 0.3 M $Na_2S_2O_4$ solution and a 0.175 M sodium citrate solution, which removes Mn and Fe from particle surfaces.

[c] Percent of initial particulate radionuclide concentration leached with a 4–6% NaOCl solution which digests organic materials.

[d] Percent of the initial particulate radionuclide concentration which remained on particles following all the leach treatments.

[e] The results are an average of two samples.

[f] The results are an average of three samples.

[g] The results are an average or two samples. The order of the $Na_2S_2O_4$ and NaOCl treatments was reversed for one of the samples.

Table 6. Radionuclide associations with chemical phases on Conowingo particles

Nuclide	Total Activity (pCi)	Bay Water[a] Exchange (%)	Cation[b] Exchange (%)	Fe-Mn[c] Leach (%)	Organic[d] Leach (%)	Fe-Mn -II Leach (%)	Cation-II Leach (%)	Lattice[e] (%)
Suspended Matter 15cc (October 1985)								
Cs-137	5.0±0.3	6.0%	28.0%	0.0%	4.0%	14.0%	0.0%	48.0%
Cs-134	2.0±0.2	25.0%	0.0%	5.0%	0.0%	30.0%	15.0%	25.0%
Co-60	10.3±0.4	14.6%	21.4%	55.3%	5.8%	1.0%	1.9%	0.0%
Zn-65	4.3±0.5	16.3%	0.0%	65.1%	0.0%	14.0%	0.0%	4.6%
Be-7	19.5±2.7	0.0%	12.8%	72.3%	4.1%	10.8%	0.0%	0.0%
K-40	79.1±4.6	0.0%	0.3%	12.3%	2.3%	7.2%	0.0%	77.9%
Suspended Matter 50cc (October 1985)								
Cs-137	13.5±0.5	0.0%	10.4%	2.2%	0.0%	0.0%	0.0%	87.4%
Cs-134	4.5±0.5	6.7%	0.0%	11.1%	8.9%	0.0%	28.9%	44.4%
Co-60	11.5±0.6	0.0%	3.5%	51.3%	8.7%	28.7%	0.0%	7.8%
Zn-65	8.8±0.8	9.1%	8.0%	46.6%	0.0%	26.3%	2.1%	7.9%
Be-7	190 ±5.3	0.0%	1.6%	68.3%	4.7%	15.9%	1.1%	8.4%
K-40	381 ±9.9	0.0%	0.0%	0.0%	0.0%	0.0%	0.0%	100.0%
Sediments 50cc (October 1985)								
Cs-137	24.5±0.7	0.0%	1.2%	17.1%	0.0%	0.0%	0.0%	81.7%
Cs-134	6.0±0.8	0.0%	0.0%	40.0%	0.0%	3.3%	0.0%	56.7%
Co-60	7.1±0.7	5.6%	0.0%	62.0%	5.6%	12.5%	4.3%	10.0%
Zn-65	6.1±1.0	8.2%	0.0%	44.3%	4.9%	33.2%	1.6%	8.0%
Be-7	18.2±4.7	13.7%	8.2%	72.5%	0.0%	5.6%	0.0%	0.0%
K-40	1029 ±9.9	0.0%	0.0%	0.0%	0.0%	0.0%	0.0%	100.0%
Suspended Matter 15cc (May 1985)								
Cs-137	46.5±0.7	10.0%	8.0%	0.0%	14.0%	0.0%	3.0%	65.0%
Cs-134	31.8±0.5	13.0%	15.0%	0.0%	12.0%	0.0%	3.0%	57.0%
Co-60	6.0±0.9	10.0%	8.0%	52.0%	12.0%	18.0%	0.0%	0.0%
Zn-65	15.8±0.7	0.0%	0.0%	70.0%	0.0%	30.0%	0.0%	0.0%
Be-7	137 ±4.0	0.0%	0.0%	65.0%	12.0%	14.0%	9.0%	0.0%
K-40	395 ±8.0	0.0%	0.0%	0.0%	0.0%	0.0%	0.0%	100.0%
Sediments 50cc (May 1985)								
Cs-137	33.4±0.7	0.0%	0.0%	0.0%	10.0%	0.0%	12.0%	78.0%
Cs-134	12.1±0.5	3.0%	2.0%	7.0%	25.0%	0.0%	9.0%	54.0%
Co-60	21.2±0.8	5.0%	0.0%	75.0%	8.0%	12.0%	0.0%	0.0%
Zn-65	15.7±1.0	3.0%	0.0%	56.0%	10.0%	13.0%	18.0%	0.0%
Be-7	49.5±4.4	13.0%	12.0%	60.0%	15.0%	0.0%	0.0%	0.0%
K-40	062 ±13	0.0%	0.0%	0.0%	0.0%	0.0%	0.0%	100.0%

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[a] Percent of initial particulate radionuclide concentration leached with saline bay water.

[b] Percent of initial particulate radionuclide concentration leached with a 1.0 M NH_4Cl solution which removes exchangeable cations on particle surfaces.

[c] Percent of the initial particulate radionuclide concentration leached with a 0.3 M $Na_2S_2O_4$ solution and a 0.175 M sodium citrate solution, which removes Mn and Fe from particle surfaces.

[d] Percent of initial particulate radionuclide concentration leached with a 4-6% NaOCl solution which digests organic materials.

[e] Percent of the initial particulate radionuclide concentration which remained on particles following all the leach treatments.

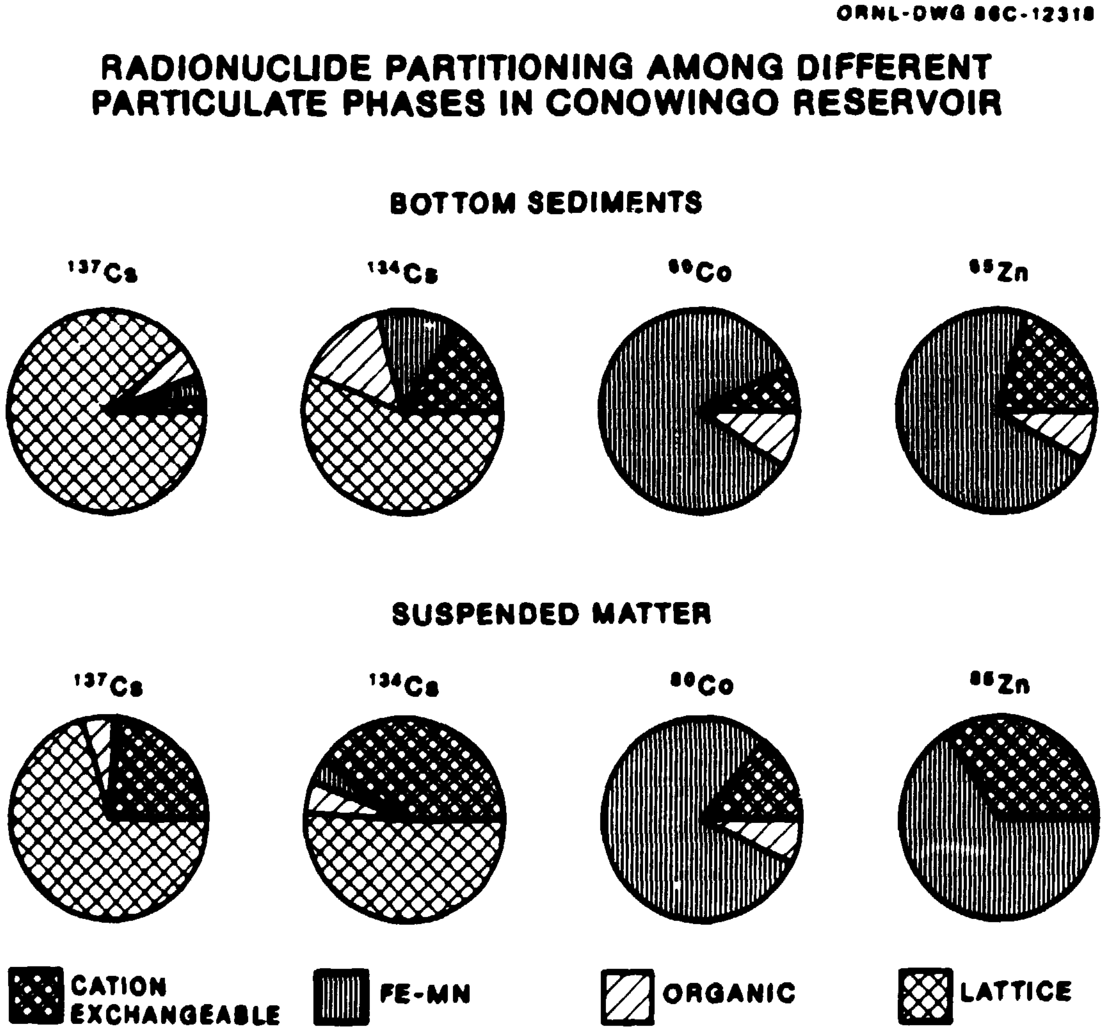

Fig. 8. Radionuclide partitioning among different particulate phases in suspended matter and sediment samples collected in Conowingo Pond.

is kinetically controlled; and, as a consequence, recently introduced reactor radiocesium (^{134}Cs and ^{137}Cs) that rapidly attaches to particle surfaces will be more vulnerable to cation exchange and desorption when transported from freshwater areas into estuarine areas than fallout radiocesium (^{137}Cs) that has been in the system long enough to diffuse into mineral lattices. This assumption implies that recently introduced radiocesium (from reactors) may behave differently than fallout radiocesium in estuarine environments. This is supported by the somewhat lower average sorption ratio of ^{134}Cs in Conowingo Pond (5.5×10^4) relative to the average R_s for ^{137}Cs (1.1×10^5). Consequently, transport and fate models based on fallout radiocesium may not be appropriate for recently introduced reactor radiocesium.

Cobalt-60, ^{65}Zn, and ^{7}Be all appear to be primarily associated with the Fe-Mn phases on both the suspended matter and bottom sediments (Tables 5 and 6). Between 10 and 20% of the these nuclides may also be associated with organic substances. Less than 10% of the sorbed ^{60}Co appears to be exchangeable with seawater cations, with the exception of one suspended-matter sample collected in May 1985 at a time when relatively large amounts of ^{60}Co were being released from the PB facility (see daily releases for May 1985, Appendix B). At present, we are still uncertain of the effects that salinity and suspended-matter chemistry may have on ^{65}Zn sorption-desorption. The leaching results indicate that a significant amount of the ^{65}Zn appears to be exchangeable with salt-water cations (Tables 5-6 and Fig. 8). The field samples collected at 3 ppt salinity in October 1985 (Table 4) indicated no change in the particle-to-water distribution for ^{65}Zn, which implies that very little (if any) of the ^{65}Zn associated with riverborne particles was desorbed when transported into this estuarine area. As in the case for radiocesium, there may be a kinetic factor involved in converting cation-exchangeable zinc into Fe-Mn associated zinc.

A comparison of the chemical associations of radionuclides on Conowingo Pond sediments with chemical associations on Calvert Cliffs sediments (Tables 5-6) indicates that organics may play a much greater role as a sorbing phase in the Bay near Calvert Cliffs than in Conowingo Pond, where Fe-Mn coatings are the most important sorbing phase. This may reflect the fact that organic particles form a greater percentage of the suspended matter in the lower Bay, as indicated by the greater total carbon concentrations relative to carbon concentrations in the suspended matter of Conowingo Pond (Table E-1).

3.4 RADIONUCLIDES IN SURFACE SEDIMENTS AND SEDIMENT CORES

Radionuclide concentrations measured in the surface (0-2 cm) sediment samples are presented with the water column data in Tables 1-4, and their vertical distribution in two of these cores, one from Conowingo Pond and one from the Upper Bay near Calvert Cliffs, is presented in Tables 7 and 8, respectively. Although measurable activities of ^{137}Cs were observed over the entire 2-meter length of the Conowingo Pond core, the activity of reactor-produced ^{134}Cs and ^{60}Co were confined to the top one-half meter (Table 7). The deepest appearance of ^{137}Cs in the Bay core near Calvert Cliffs was 75 cm, whereas reactor-produced ^{60}Co was confined to the top 40 cm. The deepest appearance of ^{137}Cs indicates the beginning of significant fallout in 1954, and the deepest appearance of reactor-released radionuclides indicates the date of initial reactor startup at PB and CC, respectively. In addition, peaks in the radionuclide profiles may be correlated with the history of fallout or reactor releases to provide additional time-stratigraphic reference levels. Sediment and radionuclide accumulation rates thus determined can be independently checked using the natural distribution of atmospherically derived ^{210}Pb (labeled U-^{210}Pb), to separate it from ^{210}Pb produced in the sediment by ^{226}Ra decay. Any rigorous attempt to obtain accumulation rates using these time-stratigraphic markers requires information concerning variations in sediment properties (such as grain size) and the extent of sediment mixing.

Table 7. Conowingo Pond Sediment Core (October 3, 1985)

Depth (cm)	Dry Wt. (g)	^{7}Be (pCi/g)	^{137}Cs (pCi/g)	^{134}Cs (pCi/g)	^{60}Co (pCi/g)	^{65}Zn (pCi/g)	^{40}K (pCi/g)	^{214}Pb (pCi/g)	^{210}Pb-U (pCi/g)
0-2	51.30	1.00±0.16	0.66±0.03	0.24±0.02	0.27±0.03	0.26±0.04	25.5±0.7	1.37±0.07	2.28±0.25
2-4	39.18	0.70±0.18	0.55±0.03	0.20±0.02	0.18±0.03	0.14±0.05	26.1±0.6	1.21±0.05	1.37±0.16
4-6	41.42	0.53±0.11	0.56±0.02	0.14±0.01	0.19±0.02	0.06±0.02	24.2±0.4	1.27±0.05	1.05±0.17
6-8	49.50	0.22±0.09	0.43±0.02	0.09±0.01	0.07±0.02	0.07±0.03	23.3±0.4	1.32±0.05	0.51±0.17
8-10	53.62	ND	0.30±0.02	ND	ND	ND	22.6±0.6	1.48±0.04	0.53±0.13
10-12	65.14	ND	0.28±0.02	ND	ND	ND	20.6±0.5	1.20±0.04	0.43±0.14
12-14	56.65	ND	0.31±0.01	ND	ND	ND	27.2±0.4	1.48±0.04	0.61±0.13
14-16	62.51	ND	0.35±0.02	ND	0.04±0.02	ND	23.4±0.5	1.30±0.03	0.54±0.11
16-18	57.58	ND	0.40±0.02	0.05±01	0.07±0.02	ND	16.4±0.4	1.11±0.04	1.09±0.14
18-20	67.04	ND	0.35±0.02	0.05±0.01	0.07±0.02	0.03±0.03	16.3±0.4	1.00±0.03	0.87±0.13
20-22	64.07	ND	0.34±0.02	ND	0.06±0.02	ND	15.8±0.4	0.95±0.03	1.37±0.15
22-24	61.93	ND	0.40±0.02	ND	0.03±0.02	0.06±0.03	15.8±0.4	1.12±0.04	1.10±0.15
24-26	53.61	ND	0.41±0.02	0.01±0.01	0.03±0.02	ND	18.2±0.4	1.17±0.04	0.86±0.12
26-28	52.81	ND	0.36±0.02	0.06±0.01	0.03±0.02	ND	20.0±0.5	1.20±0.04	0.94±0.14
28-30	50.90	ND	0.39±0.02	0.02±0.01	0.03±0.02	ND	16.5±0.4	1.09±0.04	0.88±0.14
30-32	53.29	ND	0.48±0.02	0.07±0.01	0.36±0.02	ND	16.4±0.4	1.10±0.03	1.12±0.13
32-34	51.22	ND	0.51±0.02	0.06±0.01	0.05±0.02	ND	17.3±0.4	1.15±0.04	1.42±0.17
34-36	52.50	ND	0.65±0.02	0.13±0.02	0.13±0.02	0.02±0.03	19.2±0.4	1.23±0.04	1.02±0.16
36-38	49.86	ND	1.02±0.03	0.29±0.02	0.23±0.02	0.06±0.04	20.1±0.5	1.14±0.04	1.27±0.14
38-40	48.56	ND	0.66±0.03	0.06±0.01	0.12±0.02	ND	19.9±0.5	1.33±0.04	1.01±0.14
40-42	50.50	ND	0.73±0.03	0.09±0.01	0.24±0.02	ND	18.9±0.4	1.12±0.04	1.44±0.16
42-44	43.80	ND	0.81±0.03	0.09±0.02	0.29±0.03	ND	19.4±0.5	1.21±0.04	1.23±0.15
44-46	45.54	ND	0.72±0.03	0.08±0.01	0.09±0.02	ND	18.4±0.4	1.27±0.05	1.09±0.15
46-48	49.33	ND	0.69±0.02	0.05±0.02	0.05±0.02	ND	18.4±0.4	1.13±0.03	1.02±0.15
48-50	60.41	ND	0.97±0.03	0.11±0.01	0.05±0.02	ND	18.4±0.4	1.12±0.03	1.18±0.15
50-52	60.87	ND	1.15±0.03	0.08±0.01	0.07±0.02	ND	18.3±0.4	1.04±0.04	0.93±0.14
52-54	71.63	ND	0.60±0.02	0.05±0.01	ND	ND	16.0±0.3	1.07±0.03	0.59±0.11
54-56	66.90	ND	0.46±0.02	0.02±0.01	ND	ND	15.9±0.3	1.02±0.03	0.74±0.02
56-58	68.50	ND	0.38±0.02	ND	ND	ND	16.1±0.3	1.00±0.05	1.02±0.21
58-60	63.73	ND	0.39±0.02	ND	ND	ND	16.6±0.4	1.10±0.03	1.05±0.13
60-64	71.21	ND	0.37±0.02	ND	ND	ND	16.3±0.3	0.97±0.03	0.80±0.12
64-68	58.54	ND	0.78±0.02	ND	0.03±0.02	ND	19.3±0.4	1.13±0.03	0.87±0.14
68-72	58.37	ND	0.74±0.02	ND	0.01±0.01	ND	18.5±0.4	1.10±0.04	0.93±0.15
72-76	55.70	ND	0.52±0.02	ND	ND	ND	15.5±0.4	1.01±0.04	0.58±0.11
76-80	66.64	ND	0.33±0.02	ND	ND	ND	13.3±0.3	0.83±0.03	0.65±0.11
80-84	67.57	ND	0.49±0.02	ND	ND	ND	16.8±0.4	1.09±0.03	0.60±0.13
84-88	65.40	ND	0.43±0.02	ND	ND	ND	21.4±0.5	1.18±0.03	0.80±0.13
88-92	53.53	ND	0.77±0.02	ND	ND	ND	22.3±0.4	1.33±0.04	0.69±0.14
92-96	66.63	ND	0.84±0.02	0.02±0.02	0.05±0.02	ND	19.1±0.4	1.15±0.03	0.82±0.14
96-100	65.07	ND	0.68±0.03	ND	ND	ND	19.6±0.5	1.09±0.03	0.94±0.15
100-104	66.35	ND	0.42±0.02	ND	0.02±0.01	ND	15.5±0.4	1.02±0.04	0.76±0.13
104-108	69.79	ND	0.41±0.03	ND	ND	ND	15.1±0.6	1.03±0.03	0.64±0.11
108-112	69.46	ND	0.34±0.02	ND	ND	ND	14.9±0.4	1.02±0.03	0.58±0.12
112-116	78.06	ND	0.35±0.02	ND	ND	ND	16.1±0.4	0.93±0.03	1.03±0.13
116-120	60.89	ND	0.42±0.02	ND	ND	ND	17.2±0.4	1.16±0.03	0.55±0.11
120-124	66.58	ND	0.45±0.03	ND	ND	ND	17.0±0.7	1.05±0.03	1.02±0.13
124-128	66.57	ND	0.43±0.03	ND	ND	ND	16.9±0.7	1.09±0.03	0.85±0.12
128-132	66.83	ND	0.46±0.02	ND	ND	ND	18.3±0.5	1.18±0.03	0.88±0.14
132-136	69.44	ND	0.38±0.03	ND	ND	ND	20.9±0.8	1.19±0.03	0.57±0.14
136-140	68.03	ND	0.44±0.02	ND	ND	ND	18.4±0.3	1.25±0.03	0.65±0.13
140-144	77.28	ND	0.48±0.02	ND	ND	ND	18.8±0.3	1.13±0.03	0.97±0.15
144-148	67.33	ND	0.43±0.02	ND	ND	ND	17.4±0.3	1.18±0.03	1.07±0.14
148-152	68.01	ND	0.39±0.03	ND	ND	ND	16.5±0.5	1.04±0.03	0.74±0.12
152-156	75.10	ND	0.30±0.03	ND	ND	ND	18.1±0.6	1.05±0.03	0.71±0.12
156-160	85.71	ND	0.19±0.01	ND	ND	ND	10.6±0.2	0.79±0.02	0.33±0.09
160-164	74.61	ND	0.14±0.02	ND	ND	ND	10.1±0.4	0.80±0.02	0.13±0.06
164-168	77.65	ND	0.12±0.01	ND	ND	ND	10.6±0.3	0.79±0.03	0.33±0.10
168-172	76.43	ND	0.17±0.01	ND	ND	ND	10.7±0.3	0.82±0.03	0.32±0.10
172-176	80.73	ND	0.14±0.02	ND	ND	ND	9.0±0.3	0.80±0.02	0.27±0.07
176-180	83.56	ND	0.13±0.02	ND	ND	ND	7.9±0.5	0.68±0.02	0.24±0.08
180-184	75.55	ND	0.25±0.02	ND	ND	ND	11.9±0.3	0.88±0.03	0.28±0.10
184-188	75.55	ND	0.18±0.02	ND	ND	ND	9.5±0.4	0.80±0.02	0.28±0.07
188-192	75.68	ND	0.17±0.02	ND	ND	ND	9.3±0.3	0.78±0.03	0.27±0.10
192-196	80.09	ND	0.19±0.01	ND	ND	ND	9.6±0.2	—	—
196-200	74.01	ND	0.24±0.02	ND	0.02±0.01	ND	11.2±0.3	0.89±0.02	0.21±0.08
INTEGRATED TOTAL (pCi/cm^{2})		Σ1.9±0.2	Σ58±0.4	Σ2.9±0.1	Σ3.2±0.2	Σ0.6±0.1			Σ91±1

Table 8. Calvert Cliffs sediment core (October 7, 1985)

Depth (cm)	Dry Wt. (g)	^{7}Be (pCi/g)	^{137}Cs (pCi/g)	^{134}Cs (pCi/g)	^{60}Co (pCi/g)	^{65}Zn (pCi/g)	^{40}K (pCi/g)	^{214}Pb (pCi/g)	^{210}Pb-U (pCi/g)
0 - 2	28.45	1.07±0.11	0.13±0.02	ND	0.05±0.01	ND	8.8±0.3	0.99±0.05	0.51±0.14
2 - 4	83.43	0.36±0.06	0.06±0.01	ND	0.03±0.01	ND	5.4±0.7	0.65±0.02	0.46±0.09
4 - 6	97.93	0.24±0.04	0.06±0.01	ND	0.04±0.01	ND	5.2±0.1	0.60±0.02	0.64±0.09
6 - 8	100.23	0.11±0.06	0.06±0.01	ND	0.04±0.01	ND	4.7±0.1	0.55±0.02	0.38±0.07
8 -10	93.22	ND	0.06±0.01	ND	0.04±0.01	ND	4.9±0.1	0.55±0.02	0.36±0.07
10–12	110.46	ND	0.06±0.01	ND	0.02±0.01	ND	4.1±0.1	0.53±0.01	0.20±0.05
12–14	98.27	ND	0.04±0.01	ND	ND	ND	4.3±0.2	0.50±0.02	0.35±0.06
14–16	95.28	ND	0.06±0.01	ND	0.03±0.01	ND	4.5±0.2	0.50±0.02	0.44±0.06
16–18	96.59	ND	0.10±0.01	ND	ND	ND	5.5±0.2	0.56±0.02	0.64±0.06
18–20	120.67	ND	0.05±0.01	ND	0.01±0.04	ND	4.3±0.1	0.50±0.01	0.22±0.06
20–22	113.55	ND	0.03±0.01	ND	ND	ND	3.6±0.1	0.57±0.02	0.27±0.07
22–24	111.44	ND	0.05±0.01	ND	ND	ND	5.1±0.1	0.52±0.02	0.26±0.07
24–26	119.70	ND	0.07±0.01	ND	ND	ND	4.7±0.2	0.55±0.02	0.40±0.06
26–28	114.04	ND	0.08±0.01	ND	ND	ND	5.1±0.3	0.53±0.02	0.23±0.06
28–30	113.63	ND	0.05±0.01	ND	ND	ND	4.5±0.2	0.54±0.02	0.26±0.07
30–32	114.47	ND	0.05±0.91	ND	ND	ND	4.0±0.2	0.52±0.02	0.25±0.06
32–34	108.56	ND	0.07±0.01	ND	0.01±0.01	ND	4.7±0.2	0.58±0.02	0.34±0.07
34–36	120.80	ND	0.05±0.01	ND	0.01±0.01	ND	4.1±0.2	0.54±0.01	0.29±0.06
36–38	116.01	ND	0.05±0.01	ND	ND	ND	4.1±0.1	0.54±0.02	0.26±0.00
38–40	111.52	ND	0.06±0.01	ND	0.01±0.01	ND	4.6±0.2	0.57±0.02	0.35±0.07
40–42	120.86	ND	0.08±0.01	ND	ND	ND	4.4±0.2	0.53±0.02	0.37±0.06
42–44	119.01	ND	0.06±0.01	ND	0.02±0.01	ND	4.0±0.3	0.54±0.02	0.18±0.05
44–46	102.26	ND	0.11±0.01	ND	ND	ND	3.5±0.2	0.49±0.02	0.19±0.07
46–48	95.57	ND	0.16±0.02	ND	ND	ND	4.9±0.3	0.56±0.02	0.20±0.07
48–50	119.65	ND	0.18±0.01	ND	ND	ND	4.5±0.2	0.53±0.02	0.29±0.07
50–52	94.32	ND	0.27±0.01	ND	ND	ND	5.0±0.1	0.63±0.02	0.21±0.08
52–54	106.23	ND	0.27±0.02	ND	ND	ND	4.9±0.3	0.53±0.02	0.39±0.07
54–56	111.65	ND	0.27±0.01	ND	ND	ND	4.9±0.2	0.60±0.02	0.38±0.07
56–58	104.39	ND	0.26±0.02	ND	ND	ND	5.0±0.3	0.60±0.02	0.30±0.07
58–60	92.97	ND	0.24±0.01	ND	ND	ND	5.0±0.2	0.60±0.02	0.33±0.06
60–64	100.95	ND	0.19±0.01	ND	ND	ND	5.0±0.2	0.62±0.02	0.35±0.06
64–68	120.13	ND	0.10±0.01	ND	ND	ND	4.9±0.2	0.57±0.02	0.23±0.07
68–72	96.51	ND	0.09±0.01	ND	ND	ND	5.0±0.2	0.68±0.02	0.17±0.08
72–76	111.30	ND	0.06±0.01	ND	ND	ND	5.5±0.2	0.63±0.02	0.19±0.07
76–80	118.65	ND	0.02±0.01	ND	ND	ND	4.6±0.1	0.50±0.02	0.20±0.07
80–84	97.52	ND	ND	ND	ND	ND	4.4±0.2	0.66±0.02	0.13±0.00
84–88	93.47	ND	ND	ND	ND	ND	5.6±0.2	0.66±0.02	0.14±0.00
88–92	91.11	ND	ND	ND	ND	ND	6.6±0.1	0.70±0.02	0.22±0.08
92–96	80.44	ND	ND	ND	ND	ND	7.3±0.2	0.68±0.02	0.19±0.08
96–100	63.83	ND	ND	ND	ND	ND	8.7±0.3	0.79±0.03	0.42±0.10
100–104	82.36	ND	ND	ND	ND	ND	7.6±0.2	0.79±0.03	0.32±0.11
104–108	82.07	ND	ND	ND	ND	ND	8.1±0.3	0.77±0.02	0.21±0.08
108–112	93.98	ND	ND	ND	ND	ND	7.2±0.2	0.74±0.02	0.18±0.10
112–116	89.27	ND	ND	ND	ND	ND	6.6±0.2	0.72±0.02	0.27±0.07
116–120	88.49	ND	NA	ND	ND	ND	ND	0.72±0.02	0.14±0.08
120–124	88.54	ND	NA	ND	ND	ND	ND	0.66±0.02	0.12±0.08
124–128	66.57	ND	NA	ND	ND	ND	ND	0.80±0.02	0.26±0.07
128–132	59.23	ND	NA	ND	ND	ND	ND	0.78±0.03	0.28±0.08
132–136	59.79	ND	NA	ND	ND	ND	ND	0.86±0.03	0.25±0.09
136–140	57.22	ND	NA	ND	ND	ND	ND	0.87±0.03	0.28±0.10
140–143	64.54	ND	ND	ND	ND	ND	ND	0.81±0.03	0.17±0.10
INTEGRATED TOTAL pCi/cm^2		Σ2.1±0.2	Σ9.1±0.2		Σ0.6±0.1				Σ39±2

The core profile for Conowingo Pond indicates that this reservoir serves as a very efficient fine-particle and radionuclide trap. A comparison of the total integrated activity of ^{7}Be (1.9 pCi/cm^{2}) measured in this core (Table 7) with the total integrated standing crop of ^{7}Be expected from its atmospheric flux to the pond surface (1.95 pCi/cm^{2}; Appendix A) indicates that this site is an efficient collector for particles and radionuclides during the 77-d time period, or the mean life of ^{7}Be. The mean life of a radionuclide is the average length of time that the radionuclide will be around before decay; it is numerically related to its half-life by the expression

$$\text{mean life} = \frac{\text{half-life}}{\text{Ln } 2} \quad .$$

Over longer time periods, such as over the 32-year mean life of ^{210}Pb, radionuclide accumulation at this site appears to be enhanced by particle focusing during deposition. The expected U-^{210}Pb inventory supported by its atmospheric flux during the past 32 years is about 15 pCi/cm^{2} (Appendix A) whereas the measured inventory of U-^{210}Pb in the Conowingo Pond core (Table 7) is about 7 times greater than this expected value. These results suggest that this area in Conowingo Pond is a major accumulation site for materials released into or transported downstream in the Susquehanna River. Particle and associated-radionuclide focusing may also occur at the Calvert Cliffs core site, as indicated by the ^{7}Be and ^{210}Pb core inventories of 2.1 pCi/cm^{2} and 39 pCi/cm^{2}, respectively (Table 8).

A rough maximum estimate for the trapping efficiency of Conowingo Pond for reactor-released radionuclides can also be obtained by extrapolating the respective ^{134}Cs, ^{60}Co, and ^{65}Zn inventories in the Conowingo sediment core over the surface area of the Pond (37 Km^{2}) and comparing this value with the decay-corrected reactor inputs listed in Appendix A. Such a calculation indicates that about 50% of the ^{134}Cs, 70% of the ^{65}Zn, and 100% of the ^{60}Co released from the Peach Bottom Power Atomic Station has been trapped with the reservoir sediments.

Because this core was collected at a major accumulation site, and extrapolated over the entire pond, these estimates are unrealistically high. A more refined moss balance estimate for the extent of radionuclide trapping in Conowingo Pond has been derived from a 5-year time series of surface sediment collections and from additional core data (McLean et al. 1988). These results indicate that about 28% of the ^{60}Co, 13% of the ^{134}Cs, and 31% of the ^{65}Zn are retained within the sediments of the reservoir (McLean et al. 1988).

4. DISCUSSION OF MECHANISMS AND RATES AFFECTING THE FATE OF RADIONUCLIDES IN THE SUSQUEHANNA-CHESAPEAKE BAY SYSTEM

4.1 RATES FOR RADIONUCLIDE SORPTION AND REMOVAL FROM THE WATER COLUMN USING ^{7}Be AS A NATURAL TRACER

Because the input function of naturally occurring ^{7}Be can be quantified and documented at the time of sample collection and because the sorption ratios for ^{7}Be have been shown to be similar to those measured for ^{137}Cs, ^{134}Cs, ^{60}Co, and ^{65}Zn in Conowingo Pond (Tables 1-4), we have used its particle-to-water distribution to quantify radionuclide sorption rates to suspended matter and radionuclide removal rates from the water column by settling particles. During 1983-1985, the input function of ^{7}Be has been measured from precipitation samples collected at Norfolk, Virginia (located on Lower Chesapeake Bay), and at Oak Ridge, Tennessee. Total (wet and dry) ^{7}Be atmospheric flux data for Norfolk and integrated ^{7}Be inventories calculated from these flux data are presented on a monthly basis in Appendix A. The atmospheric flux of ^{7}Be commonly supports inventories ranging from 1 to 2 pCi/cm^{2} at both Norfolk and Oak Ridge.

To investigate how long a dissolved radionuclide will remain in solution before biological uptake or sorption onto suspended matter, we determined the sorption kinetics of ^{7}Be by collecting a time series of large-volume water samples after major precipitation (input) events. Because collection in a time series requires daily samples at the same location over the course of a week or two, this work was conducted in Melton Hill Reservoir near Oak Ridge National Laboratory. The results for one such event in March 1986, illustrated in Fig. 9, indicate that the apparent sorption rate is about 15% d^{-1}. Similar time-series field experiments during different environmental conditions indicated that the apparent sorption rates are dependent on suspended matter concentration, and range from 10% d^{-1} in winter to about 20% d^{-1} in early spring, when biological productivity and runoff increases suspended matter concentrations from about 5 to 20 mg/L

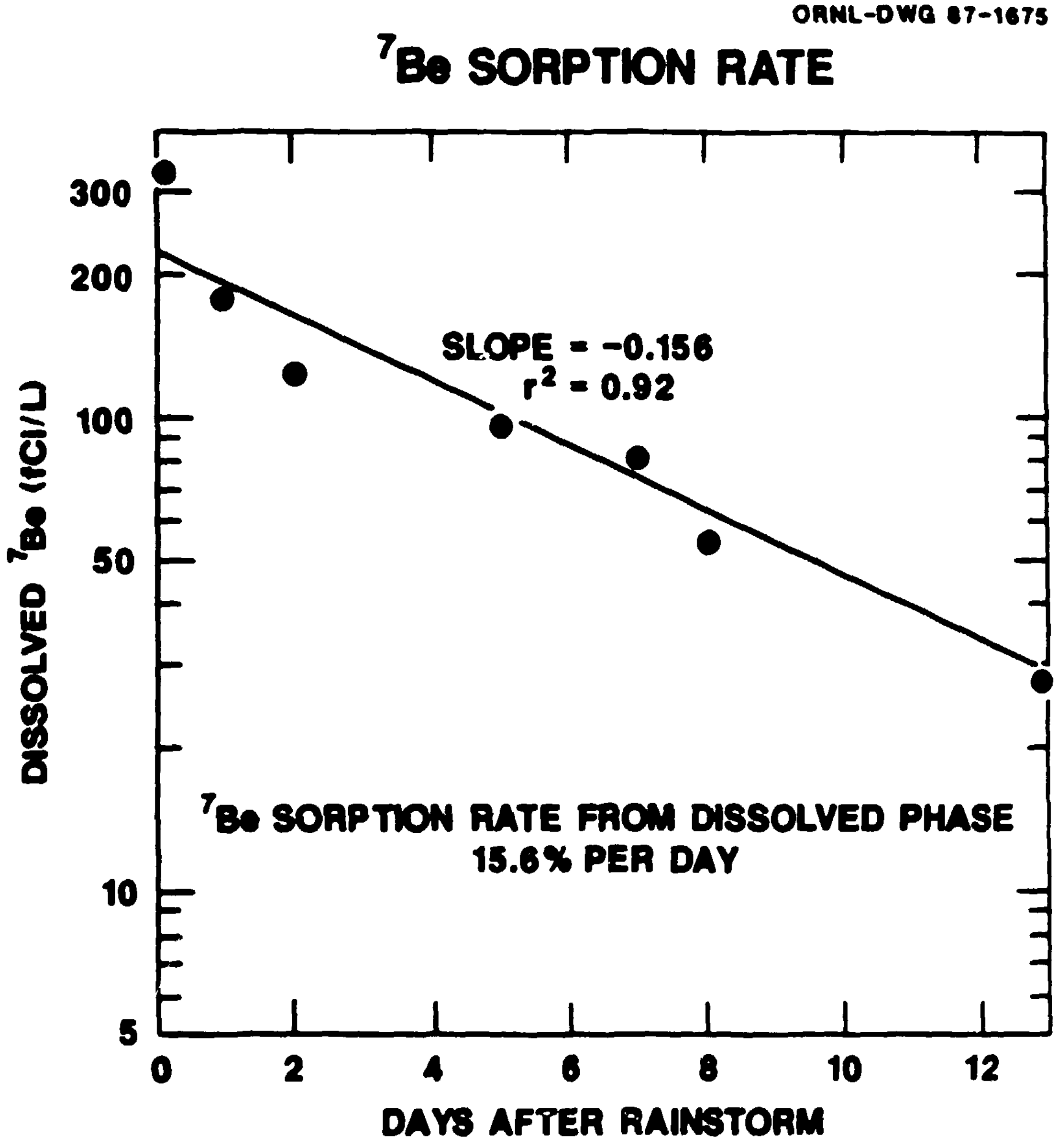

Fig. 9. Dissolved ^{7}Be concentrations measured in Melton Hill Reservoir for samples collected in a time series after a major rainstorm (^{7}Be-input) event. These results indicate that the sorption rate is about 15% d^{-1} at an average suspended matter concentration of 15 mg/L.

(C. R. Olsen, unpublished data, 1986). These suspended matter concentrations are also typical for Concwingo Pond. The drop in dissolved radiocesium concentrations following a large release into Conowingo Pond (measured on successive days during May 1985) also indicated a sorption rate onto suspended matter of about 15% to 20% d^{-1} for both ^{137}Cs and ^{134}Cs (Table 3).

To investigate the length of time a particle-reactive radionuclide will remain in the water column (i.e., its residence time) before removal to the sediments via settling particles, we have used the box model illustrated in Fig. 10 to calculate ranges in ^{7}Be removal rates. The rate of ^{7}Be removal from the water column (λ_s) can be estimated by integrating the ^{7}Be standing crop throughout the water column (A_{wc}) and relating this value to its input function (I) and rate of radioactive decay (λ). Beryllium-7 removal rates from the water column calculated from this box model are listed in Table 9.

The total inventory of ^{7}Be delivered to the surface of Conowingo Pond and Upper Chesapeake Bay was 1.10 pCi/cm^2 at the beginning of October 1983, 1.66 pCi/cm^2 at the beginning of April 1984, 1.35 pCi/cm^2 at the beginning of May 1985, and 1.96 pCi/cm^2 at the beginning of October 1985 (Appendix A). Assuming that the mean depth of Conowingo Pond is 9 m (McLean et al. 1983) and a total ^{7}Be concentration (on the suspended matter and in the dissolved phase) is 0.13 pCi/L during October 1983, 0.51 pCi/L in April 1984, 0.23 pCi/L in May 1985, and 0.22 pCi/L in October (Tables 1-4), then the total amount of ^{7}Be in the water column per cm^2 area ranges from 0.12 pCi/cm^2 (or about 10% of the total input) in October 1983 to 0.46 pCi/cm^2 (or about 25% of the total input) in April 1984 (Table 9). Using the steady-state box model in Fig. 10, the calculated removal rates for ^{7}Be from the Conowingo Pond water column range from as high as 11% d^{-1} in October to as low as 4% d^{-1} in April (Table 9). This corresponds to a ^{7}Be residence time in Conowingo Pond of about 9 days during the low-flow conditions of October to as much as 25 days during the high-flow conditions of April (Table 9). The residence time of water in Conowingo Pond ranges from about 2 days during high flow to about 19 days during low flow, as calculated by dividing the Pond volume (330×10^6 m^3) by the average

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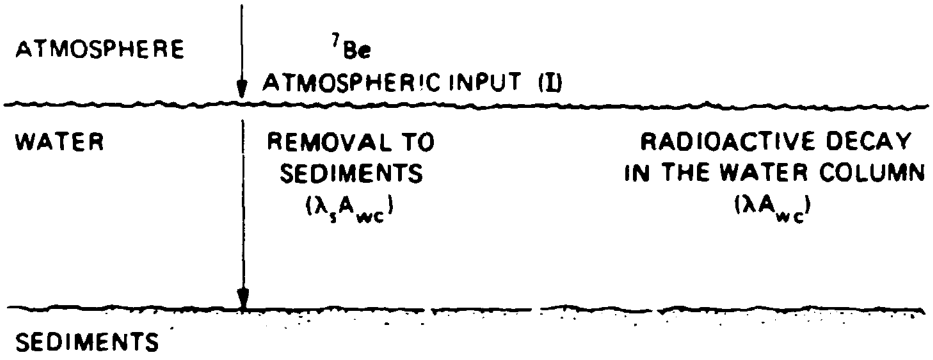
BOX MODEL FOR ^{7}Be REMOVAL FROM COASTAL WATERS BY SETTLING PARTICLES



ASSUME:

(1) STEADY-STATE FLUX CONDITIONS RANGING FROM 0.013 pCi $cm^{-2}d^{-1}$ TO 0.026 pCi $cm^{-2}d^{-1}$

(2) NO EXCHANGE OF WATER WITH SIGNIFICANTLY DIFFERENT ^{7}Be CONCENTRATIONS OVER THE 77 DAY MEAN-LIFE $(1/\lambda)$ OF ^{7}Be

THEN:

$$I = \lambda A_{wc} + \lambda_s A_{wc}$$

OR:

$$\lambda_s = \frac{I - \lambda A_{wc}}{A_{wc}}$$ FIRST-ORDER REMOVAL RATE CONSTANT FOR ^{7}Be

AND:

$$\tau = \frac{1}{\lambda_s}$$ MEAN RESIDENCE TIME OF ^{7}Be IN COASTAL WATERS

CONOWINGO RESERVOIR

DATE	INPUT (pCi/cm^2)	WATER COLUMN (pCi/cm^2)	REMOVAL RATE (λ_s)	WATER COLUMN RESIDENCE TIME (τ)
OCT 83	1.1	0.12	0.11 d^{-1}	9 days
APR 84	1.7	0.46	0.04 d^{-1}	25 days

Fig. 10. Illustration of the box model used to calculate the ^{7}Be removal rates and residence times listed in Table 9. The rate of ^{7}Be removal from the water column (λ_s) can be estimated by intergrating the ^{7}Be standing crop throughout the water column (A_{WC}) and relating this value to its steady-state atmospheric input (I) and its rate of radioactive decay (λ).

Table 9. ^{7}Be removal rates and residence times in the water column

Sample (date)	^{7}Be Input[a] (pCi/cm^{2})	Total ^{7}Be Water column[b] (pCi/cm^{2})	Removal rate (d^{-1})	Residence Time (d)	Effective Mean Residence Time[c] (d)
Conowingo Pond					
October 1983	1.1	0.12	0.11	9	8
April 1984	1.7	0.46	0.04	25	19
May 1985	(1.4)	0.21	0.07	(14)	(12)
October 1985	2.0	0.20	0.11	9	8
Upper Chesapeake Bay					
October 1983	1.1	0.07	0.19	5	5
April 1984	1.7	0.49	0.03	33	23
May 1985	(1.4)	0.13	0.13	8	(7)
October 1985	2.0	0.09	0.27	4	4
Turbidity Zone					
October 1985	2.0	0.02	1.25	0.8	0.8

[a] Be-7 input is based on the inventories listed in Appendix A for Norfolk, Virginia. Because of a collector malfunction, the input data for May 1985 was lost at Norfolk. The May 1985 input data are for Oak Ridge, Tennessee.

[b] The total amount of ^{7}Be in the water column represents both the amount in the dissolved phase and the amount associated with the suspended matter. Upper Chesapeake Bay represents an average in the water column at Susquehanna Flats, Annapolis and Calvert Cliffs. Turbidity zone represents an average for Grove Point and Howell Point (Table 4).

[c] Effective mean residence time takes into account removal by radioactive decay as well as by deposition with settling particles. For relatively short-lived radionuclides such as ^{7}Be, ^{58}Co, and ^{65}Zn, the effective mean residence time may be significantly shorter than that calculated for removal by sorption onto particles and particle deposition.

monthly high (3500 m^3/s) and low (200 m^3/s) flows for the Susquehanna River. These results imply that a significant portion of the ^7Be (and by analogy, other particle-reactive or reactor-released radionuclides) introduced into Conowingo Pond during high flow conditions will be transported past the dam into the upper portions of Chesapeake Bay.

Within the turbidity zone, our data indicate that the rate of sorption (removal from the dissolved phase) and the residence time (rate of removal from the water column) are extremely rapid. The residence time of ^7Be within the turbidity zone of Chesapeake Bay appears to be less than one day (Table 9). Sediment cores collected within this zone indicate that this area of Chesapeake Bay serves as an efficient fine-particle trap and as a site of rapid burial for particle-associated radionuclides (Olsen et al. 1981).

Similar removal rate calculations can be made for Upper Chesapeake Bay assuming a mean depth of 8.2 meters (Fig. 1, and Cronin 1971), and using an average ^7Be water column concentration for Susquehanna Flats, Annapolis, and Calvert Cliffs (Table 9). The removal rates of ^7Be from the water column during May and October ranged from 13 to 27% d^{-1} and are about the same as the rate of ^7Be sorption onto suspended particles (10 to 20% d^{-1}). This implies that during normal to low river-flow conditions, the length of time that ^7Be (and by analogy, other particle-reactive or reactor-released radionuclides) remains in the water column is primarily controlled by its sorption rate onto suspended matter. During the high-flow conditions of April, however, the removal rate of ^7Be from the water column is reduced by a factor of 3 in both the Upper Bay and in Conowingo Pond. This corresponds to an increase in the water residence time of about 25 to 33 days (Table 9). One possible explanation for this increase in residence time is that sediment resuspension (in association with storms or high seasonal flows) can reintroduce deposited ^7Be (or other particle associated contaminants) back into the water column, and thus prolong the length of time required for ultimate removal by deposition with settling particles. This implies that during high river-flow conditions, the residence time of ^{7}Be (and by analogy, other particle-reactive or reactor-released radionuclides) in the water column is primarily controlled by fine particle dynamics rather than by the rates of sorption onto suspended matter as was the case during low-flow conditions. The importance of high-flow conditions and sediment resuspension on the transport of reactor-released radionuclides has been previously discussed in Section 3.2.

4.2 TRANSPORT AND GEOCHEMICAL FATE OF RADIOCESIUM

Dissolved radiocesium rarely forms solution complexes in natural aquatic environments and therefore occurs predominantly as a monovalent cation (Cs^{+}) in the Susquehanna-Chesapeake Bay System. Consequently, radiocesium sorption appears to be relatively unaffected by organics, pH, or Fe-Mn hydroxides, but it is affected by the concentration of other cations. Average particle-to-water distribution ratios range from about 1×10^{5} in the fresh waters of Conowingo Pond to about 5×10^{3} in the more saline waters near Calvert Cliffs, where radiocesium has to compete with seawater cations (especially K^{+}) for binding sites on particles.

The relatively high particle-to-water distribution for ^{137}Cs in fresh water arises from its strong affinity for sorption onto clay minerals. Cesium sorption on clay minerals can occur at several sites, including (1) easily exchangeable sites on the clay particle surface, (2) fixed sites at the edges of the clay-mineral lattice, and (3) nonexchangeable, interlayer sites of 1.0-nm (illitic) micaceous minerals where cesium is entrapped during lattice collapse (Tamura and Jacobs 1960). This is consistent with our radionuclide partitioning data (Fig. 8, and Tables 5 and 6) that indicate that a significantly greater proportion of the recently introduced reactor-released radiocesium (^{134}Cs and ^{137}Cs) is surface sorbed and exchangeable, whereas most of the fallout radiocesium (^{137}Cs) has had enough time to become irreversibly sorbed into interlayer lattice sites.

Little is known about the kinetics of cesium fixation into clay interlayer sites after rapid initial sorption in fresh waters (Evans et al. 1983). Our laboratory nuclide-partitioning results (Tables 5 and 6, however, indicate that about 50% of the reactor-released ^{134}Cs associated with both suspended matter and bottom sediments is already irreversibly sorbed into interlayer lattice sites, implying that fixation kinetics may occur relatively rapidly, on the order of weeks to months. A more accurate estimate for the kinetics of radiocesium fixation could possibly be obtained by examining the relative amount of ^{134}Cs fixation with depth in dated sediment cores.

Previous work on the biogeochemical fate of ^{137}Cs in the water, sediments, macrophytes, algae, zooplankton, and several species of fish in a eutrophic lake indicated that 97% of the ^{137}Cs was in the sediments (Eyman and Kevern 1975). Likewise, in Conowingo Pond, particles rapidly remove radiocesium from the water column, and sediments are the ultimate repository for radiocesium in this freshwater ecosystem.

In the estuarine waters near Calvert Cliffs, the particle-to-water distribution of radiocesium is almost two orders of magnitude lower than respective distributions measured in the fresh water of Conowingo Pond. This decrease may reflect (1) the tendency of radiocesium to remain in solution because of its competition with seawater cations (especially K^+) for binding sites on particles, or (2) the desorption of recently introduced radiocesium from particulate surfaces by exchange with seawater cations. Previous work has indicated that radiocesium desorption from particles may occur in Chesapeake Bay (Reil 1970) and in other estuarine systems (Jinks and Wrenn 1976; Patel et al. 1978; Zucker et al. 1985). Much of the field evidence for desorption has been based on a sharp decrease in suspended-particulate and sediment ^{137}Cs concentrations in estuarine areas relative to freshwater areas. Such a decrease in particulate ^{137}Cs concentrations with salinity is clearly evident in the distributional data listed in Tables 1-4. As noted previously, however, this decrease may also reflect the dilution of riverborne particles with biological particles produced in situ or with particles transported into the estuary from marine or shoreline sources. One way to help differentiate desorption from dilution is to document an increase in dissolved ^{134}Cs or ^{137}Cs within the turbidity zone or mesohaline waters relative to values expected from the mixing of Susquehanna River water, adjacent coastal water, and reactor discharges.

The results from such a three end-member box model are presented in Table 10 and indicate no appreciable radiocesium desorption from riverborne particles. Dissolved fallout ^{137}Cs concentrations in the surface mixed layer of near-shore ocean water (27-32 ppt salinity) are about 100 fCi/L along the southeastern coastline of the United States, and dissolved river ^{134}Cs was undetectable prior to the recent nuclear accident at Chernobyl (Olsen, unpublished data, 1986). With salinity as a tracer for marine water in Upper Chesapeake Bay, the marine input of dissolved ^{137}Cs is estimated to range from 10 fCi/L at 3 ppt salinity to 67 fCi/L at 20 ppt salinity (Table 10). An estimate for the dissolved input of both fallout ^{137}Cs and reactor-released ^{137}Cs and ^{134}Cs to the Upper Bay is obtained for each of the specific sampling time periods from the radiocesium data for Susquehanna Flats (Tables 1-4). As discussed in Section 3.2.1, the concentration of dissolved radiocesium in this area is quite variable and depends primarily on temporal variations in reactor discharges and river flow. Therefore, the quantification of actual radiocesium fluxes into Chesapeake Bay over longer time periods requires the utilization of a coupled hydrological, geochemical, and reactor-release model. Such a model is currently being developed by the Power Plant and Environmental Review Division and Versar Corp. Nevertheless, the synergistic data set collected during October 1985 (Table 4), provides a snapshot of radiocesium transport and geochemical fate within the turbidity zone of Upper Chesapeake Bay.

Because reactor releases from the PB facility are the only significant source for ^{134}Cs measured in the waters from Susquehanna Flats to Annapolis, and because a greater portion of the sorbed ^{134}Cs may be attached to <u>exchangeable</u> sites on particle surfaces relative to sorbed ^{137}Cs, the ^{134}Cs particle-to-water distribution data (Table 4) for Grove Point (3 ppt salinity), Howell Point (5.5 ppt salinity), and

Table 10. Dissolved radiocesium in Chesapeake Bay

Location and date	Salinity (ppt)	Seawater input[a] (fCi/L)	Freshwater input[b] (fCi/L)	CC-Reactor input[c] (fCi/L)	Estimated total (fCi/L)	Measured total (fCi/L)
Grove Point						
October 1985	3.0	10±1	10±5		20±5	15±0.6
(Cs-134)[d]		(0)	(3.6±2)		(3.6±2)	(0.8±0.3)
Howell Point						
October 1985	5.5	18±2	9±5		27±5	23±1
(Cs-134)		(0)	(3.3±2)		(3.3±2)	(2.9±0.7)
Annapolis						
October 1983	12.0	40±4	12±6		52±7	61±0.7
April 1984	9.0	30±3	0±1		30±3	29±0.6
May 1985	10.8	37±4	4±2		41±4	35±0.5
October 1985	15.2	50±5	6±2		56±5	63±0.8
(Cs-134)		(0)	(2±1)		(2±1)	(2±0.3)
Calvert Cliffs						
October 1983	14.0	47±5	11±5	3±1	61±7	72±0.8
April 1984	8.0	27±3	1±1	12±1	40±3	41±0.7
May 1985	13.5	45±5	3±2	3±1	51±5	50±0.6
October 1985	19.9	67±7	3±2	0±1	70±7	88±1.4

[a] Seawater input was estimated by assuming that the dissolved radiocesium at 30 ppt salinity was 100 fCi/L.

[b] Freshwater inputs were estimated from the dissolved radiocesium concentrations respectively measured at Susquehanna Flats.

[c] Reactor inputs from Calvert Cliffs were estimated from the ^{134}Cs/^{137}Cs ratio (see text).

[d] Parentheses indicate data for dissolved ^{134}Cs, all other data are for ^{137}Cs.

Annapolis (15.2 ppt salinity) provide an ideal tracer for the sorption-desorption fate of radiocesium. Although the concentration of ^{134}Cs on suspended particles decreases rapidly from 190 pCi/kg to not detectable (<10 pCi/kg) levels within the turbidity zone, dissolved ^{134}Cs concentrations actually decreased rather than increased as would be expected if desorption were occurring (Table 4). This may imply that (1) Susquehanna River water is not well mixed and moves as a plume through the turbidity zone and higher salinity waters of Chesapeake Bay, (2) dissolved radiocesium is actually removed from the water column by sorption onto particles in areas where the turbidity increases but that the concentration of ^{134}Cs on suspended matter is diluted by this increased turbidity, or (3) a combination of both. This sorption/desorption behavior with transport is also confirmed by the ^{137}Cs data. There are no significant differences between the dissolved ^{137}Cs concentrations estimated for marine, freshwater (fallout), and reactor sources and the concentrations of dissolved ^{137}Cs measured throughout the Upper Bay (last two columns in Table 10). This implies that ^{137}Cs is not measurably desorbed from sediments and suspended matter in Chesapeake Bay and that desorption is not significantly contributing to dissolved ^{137}Cs concentrations.

The lack of ^{134}Cs exchange with seawater cations is contradictory to our leaching results (Fig. 8), which indicate that 30-40% of the ^{134}Cs on suspended particles in Conowingo Pond is exchangeable. Two possible explanations for this contradiction are that (1) the "exchangeable" ^{134}Cs sorbed to particle surfaces in Conowingo Pond is fixed into nonexchangeable lattice sites during the flocculation of clays and dissolved organic substances with increasing salinity in the estuarine turbidity zone, and (2) the sorption rate and water-column removal rate of radiocesium in Conowingo Pond is fast enough that most of the reactor-released ^{134}Cs is deposited to the reservoir sediments, where it remains long enough to allow for fixation in nonexchangeable sites before it is resuspended and transported into the saline portions of Upper Chesapeake Bay during high flow events.

In the mesohaline waters of Chesapeake Bay, ^{134}Cs was only detected in the dissolved phase (Tables 1-4). This implies that most of the radiocesium (both ^{134}Cs and ^{137}Cs) introduced into this portion of the Bay via effluent discharges from the Calvert Cliffs Nuclear Power Plant remains in solution. The effect that these releases will have on dissolved ^{134}Cs and ^{137}Cs concentrations can be calculated by dividing the total respective quarterly release (Appendix B-4) by the total volume of mesohaline water (9.4×10^{12} L), assuming that the residence time of mesohaline water is on the order of three months:

$$\text{Residence Time (months)} = \frac{\text{Mesohaline Volume}}{\text{Susquehanna Discharge}} = \frac{0.94 \times 10^{13}\ \text{L}}{0.29 \times 10^{13}\ \text{L/month}} = 3.2 \text{ months}$$

Such calculations for quarterly releases during 1983 to 1985 indicate that the reactor effluent from Calvert Cliffs will increase dissolved ^{134}Cs concentrations by about 1.5 to 3.5 fCi/L, which are typical of the values measured in the mesohaline waters near the Calvert Cliffs site (Tables 1-4). Similar calculations for ^{137}Cs indicate that quarterly releases will increase dissolved ^{137}Cs concentrations by about 3 to 12 fCi/L (Table 10). These dissolved ^{137}Cs values are consistent with estimates determined by assuming that all of the dissolved ^{134}Cs in the waters near Calvert Cliffs comes from the CC reactors, and calculating the ^{137}Cs input from the measured ^{134}Cs/^{137}Cs ratios in the water and the decay-corrected ^{134}Cs/^{137}Cs ratios in the reactor effluent. The quarterly ^{134}Cs/^{137}Cs release ratios (computed from the release data in Appendix B-3) range from 0.773 to 0.270. The dissolved ^{137}Cs concentrations calculated from these ratios are presented in Table 10 for mesohaline water near Calvert Cliffs. Since most, if not all, of the dissolved ^{134}Cs in the mesohaline waters near Annapolis was probably transported downstream from the PB facility rather than derived from the CC facility, inputs of dissolved ^{137}Cs from Calvert Cliffs was considered to be insignificant at the Annapolis site (Table 10).

Because there does not appear to be any appreciable desorption of ^{134}Cs or ^{137}Cs within Chesapeake Bay (Table 10), the two-order-magnitude decrease in the particle-to-water distribution observed for radiocesium in the mesohaline waters of Chesapeake Bay (Tables 1-4) must reflect (1) the tendency for radiocesium to remain in solution in saline water and (2) the dilution of Susquehanna-derived particles with particles from other Upper Bay tributaries, <u>in situ</u> productivity, or 'ine sources. In fact, dissolved radiocesium may actually be incorporated into particles within the estuarine turbidity zone as a result of the increased suspended-matter concentration or the flocculation of dissolved organic and inorganic substances that occurs in this area (Bond and Meade 1966; Sholkovitz 1976; Schubel and Meade 1977; Sigleo and Helz 1981; Smith and Ellis 1982). This suggests that the flocculation of clay minerals may in some way lock radiocesium into nonexchangeable lattice sites during transport or accumulation in estuarine areas.

4.3 TRANSPORT AND GEOCHEMICAL FATE OF RADIOCOBALT

Cobalt-60 and cobalt-58 are produced by neutron activation during reactor operations. In freshwater and estuarine systems, radiocobalt is geochemically similar to iron and can exist in two oxidation states, Co(II) and Co(III). The relative stabilities of divalent and trivalent radiocobalt in solution are greatly affected by complexes with common anions (Onishi et al. 1981). While cobaltous ion (Co^{+2}) is the most soluble and stable form of radiocobalt up to a pH of about 9.5, organic and inorganic complexes can stabilize trivalent cobalt in solution.

Previous work has shown that radiocobalt sorption-desorption in freshwater and estuarine systems is not affected by salinity but is affected by organic complexation and the redox cycling of Mn-Fe hydrous oxides (Jenne and Wahlberg 1968; Fukai and Murray 1974; O'Connor and Kester 1975; Olsen et al. 1981b; Murray and Mayer 1986). Organic compounds can influence radiocobalt sorption-desorption in three ways: (1) Soluble organic materials can complex radiocobalt, preventing its sorption onto particles as they move through the Susquehanna-Chesapeake Bay System, (2) insoluble organics and organic detritus can sorb radiocobalt and thus remove it from the water phase, (3) organic compounds, through biologically-mediated reactions, can reduce the valence of radiocobalt or other metals such as iron or manganese, rendering them more soluble or can also provide localized reducing conditions in an otherwise oxic system.

The redox cycling of Fe-Mn hydrous oxides into soluble and particulate phases probably plays a significant role in radiocobalt sorption-desorption. In reducing environments (characterized by low Eh and often low pH), iron and manganese occur as soluble ferrous (Fe^{+2}) and manganous (Mn^{+2}) ions. In oxidizing environments (high Eh and often high pH), iron and manganese form relatively insoluble oxide and hydroxide coatings on particles. With changes in the environmental Eh or pH, Fe and Mn can cycle between soluble and particulate phases. Such changes occur during transport from fresh water to sea water and after deposition from oxic water to anoxic sediments. Bricker et al. (1977) and Eaton (1979) have shown that massive quantities of manganese may be released from anoxic sediments and cycled into the bottom waters of Chesapeake Bay. Eaton (1979) estimated that this input via internal cycling accounts for more than four times the total annual input of manganese (soluble and particulate) entering the Bay from the Susquehanna River. Turekian (1977) has stressed that Mn-Fe cycling in estuarine and coastal waters may provide an important mechanism for both the removal of metals (via scavenging by freshly precipitated Fe-Mn hydroxide coatings) and the release of metals (when these coatings are resolubilized under reducing conditions).

In the Susquehanna-Chesapeake Bay System, particle-to-water sorption ratios for radiocobalt are relatively high ($>10^5$) and appear to be relatively unaffected by estuarine salinity (Fig. 7). This suggests that reactor-released radiocobalt will be rapidly scavenged from the water column by particles in both fresh and estuarine environments. Since the sorption ratio for radiocobalt is similar to that of ^{7}Be in Conowingo Pond and in Chesapeake Bay, the water-column-removal rates for radiocobalt in the Susquehanna-Chesapeake Bay System should likewise be similar to the ^{7}Be removal rates listed in Table 9.

Our laboratory leaching experiments indicate that only about 10% of the radiocobalt in the sediments and suspended matter of Conowingo Pond is associated with organic matter; the remaining 90% appears to be associated with Fe-Mn coatings, and only a very small amount is exchangeable with seawater cations. [It should be noted, however, that the particles from Conowingo Pond are fine-grained muds and that the proportion of radiocobalt associated with organic matter may be much greater in sandy sediment areas. Olsen (1979) has shown that organic detritus contains much of the ^{60}Co associated with sandy sediments in the Hudson estuary.] The strong association of radiocobalt with Fe-Mn phases on the particulate samples (Fig. 8) implies that the precipitation of dissolved Fe and Mn in the estuarine environment may play an important role in the removal of dissolved radiocobalt. The extent to which radiocobalt may be desorbed from particles when the Fe-Mn hydroxide coatings are resolubilized is uncertain. In areas where sediments are accumulating rapidly (>1cm/yr), the release of radiocobalt by diffusion out of the sediments with soluble Fe and Mn is probably insignificant relative to its rapid burial by deposition.

4.4 TRANSPORT AND GEOCHEMICAL FATE OF RADIOZINC

Zinc-65 exists exclusively in the Zn(II) oxidation state, and dissolved ^{65}Zn probably exists as cationic Zn^{+2} or as an organically complexed colloid (Sigleo and Helz 1981). Previous studies have indicated that the sorption-desorption fate of ^{65}Zn and stable Zn in estuarine systems is sensitive to pH changes, with sorption increasing as water pH increases from 6 to 8 (Onishi et al. 1981). Field studies concerning the geochemical fate of zinc have been controversial. Several studies have indicated that Zn is appreciably desorbed from particles in the estuarine environment (Holmes et al. 1974; Carpenter et al. 1975; Helz 1976; Grieve and Fletcher 1977), while other studies have indicated that only a small fraction of the sorbed zinc is exchangeable in seawater (Johnson et al. 1967; Cutshall et al. 1973; Evans and Cutshall 1973) and that particulate transport during high-flow conditions (rather than desorption) may account for the depleted (or diluted) Zn concentrations in estuarine sediments relative to concentrations on riverborne particles (Helz et al. 1985).

Documentation of the sorption-desorption behavior and geochemical fate of zinc in estuarine systems is important because zinc serves as a nutrient metal for aquatic plants and organisms and is therefore bioaccumulated. Data concerning the maximum concentrations of ^{65}Zn in various aquatic biota near the PB and CC reactor facilities are listed in (McLean et al. 1983 and 1986). Although concentrations of ^{65}Zn in aquatic biota were highest in Conowingo Pond, the levels reported for 1983-1984 were significantly lower than the ^{65}Zn concentrations measured in 1981-1982 for the same biotic groups (McLean et al. 1983). This decrease was attributed the low levels of ^{65}Zn released from the PB reactors during 1983-1984 relative to levels released in 1981-1982 (Appendix B) and indicates that (1) biological uptake is strongly dependent on radionuclide concentrations in the Pond, which in turn are primarily governed by variations in radionuclide inputs, and (2) radioactive decay as well as chemical exchange and biological depuration limit long-term bioaccumulation.

Some of the results from a caged fresh-water mussel study (McLean et al. 1983) near the PB reactor site are illustrated in Figure 11. Of all the reactor-released radionuclides, ^{65}Zn exhibited the greatest biological uptake, with a bioaccumulation ratio (R_b) of about 10^5:

$$R_b = \frac{\text{Nuclide Concentration per Kg of Tissue}}{\text{Dissolved Nuclide Concentration per L}}$$

It is apparent from Figure 11 that mussels collected in the Susquehanna Flats area contained about an order of magnitude less ^{65}Zn than those collected in Conowingo Pond. Although dissolved ^{65}Zn concentrations were not measured in 1982, the particle-to-water distribution data presented for 1983-1985 in Tables 1-4 indicate that most of the ^{65}Zn

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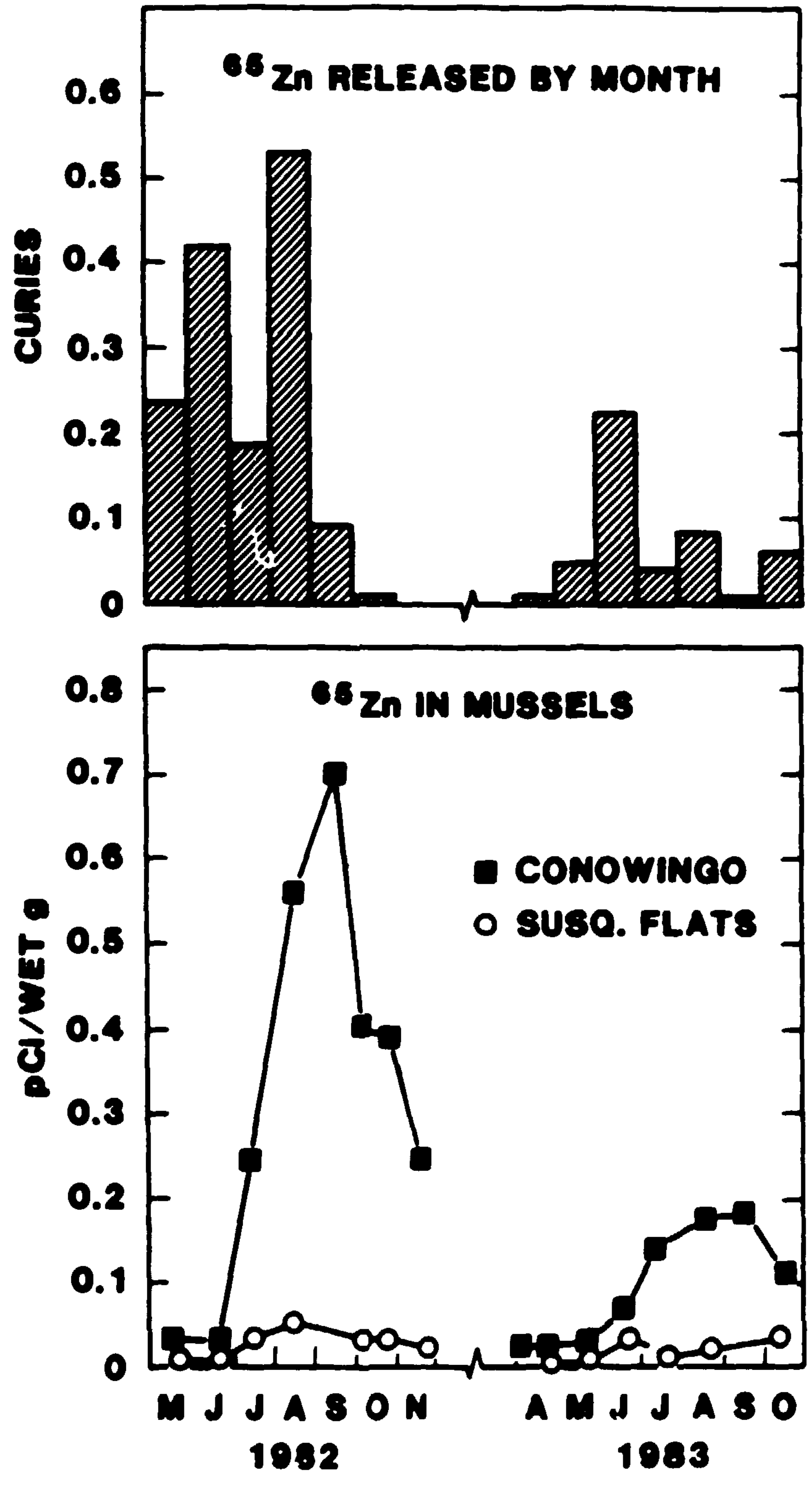

Fig. 11. Monthly releases of ^{65}Zn from the Peach Bottom Atomic Power Station and ^{65}Zn bioaccumulation in caged mussels collected and analyzed on a monthly basis (McLean et al. 1983).

released from the PB facility is rapidly removed from the water column in Conowingo Pond; and, as a result, the bioavailability of ^{65}Zn in the waters below the dam is greatly reduced as evidenced by the ^{65}Zn concentrations in mussels.

The ^{65}Zn released into Conowingo Pond is geochemically and biologically reactive, with a particle-to-water sorption ratio (R_s) averaging about 10^5. Since this particle-to-water sorption ratio is similar to that of ^{7}Be and ^{60}Co, we expect that the rapid sorption rates and water-column removal rates listed Table 9 are also applicable to ^{65}Zn. Rapid sorption from the dissolved phase and rapid removal from the Pond water by particle deposition is the most plausible explanation for the relatively low concentrations of ^{65}Zn in the water, suspended matter, and biota in the Susquehanna Flats and in Upper Chesapeake Bay.

An estimate for the amount of ^{65}Zn retained within Conowingo Pond via sorption, uptake, deposition, and accumulation can be obtained by comparing the dissolved and particulate ^{65}Zn concentrations measured in the pond waters with the waters downstream of Conowingo Dam (Susquehanna Flats). During periods of low flow (such as during October 1983 or May 1985), it appears that only 10-20% of the ^{65}Zn released into Conowingo Pond escapes the reservoir and is transported into Upper Chesapeake Bay (Table 11). However, during periods of increased flow (associated with seasonal or storm events), dissolved and/or particulate ^{65}Zn concentrations below the dam may actually exceed respective concentrations within the Pond itself (Table 11). This implies that ^{65}Zn concentrations in the Conowingo Pond may be diluted by river flow and by sediment resuspension from uncontaminated upstream areas, while ^{65}Zn concentrations in the Upper Bay are enriched by discharge from the dam and by sediment particles resuspended from the reservoir or other contaminated downstream areas. The net effects of radionuclide trapping within Conowingo Pond during normal flow conditions and radionuclide escape from the reservoir during high-flow and/or storm conditions will be documented when the inventory of radionuclides in the Pond sediments is completed.

Table 11. Biogeochemical cycling of ^{65}Zn

Date	Dissolved (fCi/L)	Particulate[a] (fCi/L)	Total (fCi/L)	Fraction[b] (%)
		Conowingo Pond		
October 1983	13.6	19.5	33.1	
April 1984	0.1	5.3	5.4	
May 1985	6.3	11.8	18.1	
October 2, 1985	5.2	7.4	12.6	
October 3, 1985	0.7	3.3	4.0 (storm)	
October 4, 1985	1.4	2.0	3.4	
		Susquehanna Flats		
October 1983	1.5	2.1	3.6	11%
April 1984	0.4	1.4	1.8	33%
May 1985	1.5	1.5	3.0	17%
October 2, 1985	—	—	—	
October 3, 1985	—	—	—	
October 4, 1985	1.8	6.3	8.1	240%

[a] Particulate ^{65}Zn concentration per liter was calculated from ^{65}Zn activity per gram of particles times the suspended matter concentration (mg/l).

[b] The fraction of ^{65}Zn activity in Conowing Pond water that escapes to Upper Chesapeake Bay. The large value for October 4, 1985, reflects the sediment resuspension and flushing effects associated with a major rainstorm event on October 3, 1985.

Our particulate leaching experiments indicated most of the ^{65}Zn was associated with Fe-Mn coatings but that 20-40% of the sorbed ^{65}Zn was exchangeable with seawater cations and NH_4^+. The field-determined particle-to-water sorption ratio, however, does not appear to be affected by estuarine salinity, as indicated by the one sample collected at 3 ppt salinity (Grove Point, Table 4). This implies that particulate ^{65}Zn may be retained long enough in Conowingo Pond to convert most of the cation exchangeable Zn into Fe-Mn-associated zinc before transport into the Upper Bay.

The geochemical fate of ^{65}Zn in the mesohaline portion of Chesapeake Bay is uncertain because the levels of ^{65}Zn released from the CC reactors (Appendix A) were too low to be detected in the water, suspended matter, or sediments (Tables 1-4). Zinc-65 is available to the biota in the region as evidenced by its occasional detection in oysters (McLean et al. (1982)).

5. CONCLUSIONS AND SUMMARY

Nuclear energy production at the Three Mile Island, Peach Bottom and Calvert Cliffs reactor facilities, has introduced several anthropogenic radionuclides into the Susquehanna-Chesapeake System via controlled low-level releases of coolant-water effluent. In this project, research has been completed to identify the important biogeochemical processes which affect the transport, sorption, distribution, and fate of these radionuclides (and by analogy, other trace substances or contaminants) in this river-estuarine system. The results from this project provide actual site-specific field data on how these radionuclides are transported and distributed among dissolved and particulate phases. This information is important for developing accurate models for radionuclide transport, biological uptake, and human dose; and for eliminating some of the uncertainties associated with extrapolating numerical simulations and laboratory data to natural environments.

Our field and laboratory measurements indicate the following conclusions with regard to radionuclide distributions and sorption behavior in the Susquehanna-Chesapeake Bay System:

- Fallout and reactor-released radiocesium (^{137}Cs and ^{134}Cs) rapidly sorb to particles in the Susquehanna River, but are much more soluble in the saline waters of Chesapeake Bay. Particle-to-water sorption ratios for radiocesium decrease by more than two orders of magnitude with increasing salinity. Field and laboratory evidence indicates that this decrease is a result of radiocesium's tendency to remain in solution in Bay water (due to its competition with seawater cations for binding sites on particles), rather than reflect its desorption from riverborne particulate surfaces (due to exchange with seawater cations). The biogeochemical fate of radiocesium in fresh-water environments is therefore governed to a great extent by the transport and accumulation of fine particles, whereas its biogeochemical fate in estuarine areas is primarily governed by water transport.

- Variations in dissolved ^{60}Co, ^{58}Co, ^{65}Zn, and ^{7}Be concentrations are strongly correlated with variations in radionuclide input, and do not appear to reflect equilibrium responses to changing chemical conditions, such as water salinity or dissolved oxygen content.

- Radiocobalt, ^{65}Zn and ^{7}Be are rapidly removed from the dissolved phase by sorption onto particulate phases in the Susquehanna-Chesapeake Bay System, and their biogeochemical fate is primarily governed by particle dynamics. Concentrations of ^{60}Co, ^{58}Co, ^{65}Zn, and ^{7}Be on suspended particles reflect short-term variations in sediment resuspension as well as variations in radionuclide input. During high seasonal flows or during rainstorms, sediment resuspension from upstream (uncontaminated) areas can dilute suspended-particulate radionuclide concentrations in Conowingo Pond. Simultaneously, sediment resuspension in the Pond or in contaminated areas below Conowingo Dam, can reintroduce radionuclide-labelled particles back into the water column and thereby increase suspended-particulate radionuclide concentrations in upper Chesapeake Bay.

- Because both dissolved and particulate radionuclide concentrations are affected by short-term variations in radionuclide input, environmental conditions, and particle dynamics; chemical equilibrium is continually adjusting and may not be attained prior to sample collection in the Susquehanna-Chesapeake Bay System. As a consequence, measured particle-to-water distributions for reactor-released radionuclides may vary over an order of magnitude. This limits the suitability of using laboratory-derived equilibrium distribution coefficients in modelling the transport and fate of radionuclides in river-estuarine environments.

- Chemical and mineralogical characterization of the particulate material and selective chemical-leaching treatments indicate that radiocobalt ^{65}Zn and ^{7}Be, but not radiocesium, are associated primarily with particulate Fe-Mn oxide phases. The geochemical cycling of these phases, and associated radionuclides, may be affected by changes in oxygen levels and pH, but are unaffected by changes in salinity. In contrast, radiocesium appears to be associated with clay minerals and its particle association may be affected by estuarine salinity, depending on its contact time with the particle. Long contact times allow radiocesium to become irreversibly fixed within clay minerals by substitution for K^+ or collapse of the hydrated lattice. Short contact times leave radiocesium on the particle surface and susceptible to desorption by exchange with seawater cations. Consequently, recently introduced radiocesium from reactors may desorb more readily and behave differently than fallout ^{137}Cs which has been in the system for more than two decades. Field evidence, however, indicates that neither reactor-released nor fallout radiocesium is desorbed from Susquehanna River particles during transport into Upper Chesapeake Bay, and that radiocesium may actually be removed from the dissolved phase by sorption onto suspended matter within the estuarine turbidity zone. This lack of radiocesium exchange with seawater cations suggests that the residence time of particle-associated radiocesium in Conowingo Pond is long enough to allow most of it to diffuse into nonexchangeable mineral lattice sites before transport into Upper Chesapeake Bay.

- The particle-to-water distribution of naturally occurring ^{7}Be (53.3-d half-life), has been used to quantify radionuclide <u>sorption rates</u> from the dissolved phase onto suspended matter and radionuclide <u>removal rates</u> from the water column by settling particles. Our results indicate that apparent <u>sorption rates</u> are dependent on suspended-matter concentration, and range from 10% d^{-1} during the winter to about 20% d^{-1} in the early spring when biological productivity and runoff increases suspended-matter concentrations from about 5 to 15 mg/L. Our results also indicate that removal rates for ^{7}Be from the water column in Conowingo Pond ranges from about 5% d^{-1} in April (high river flow) to about 10% d^{-1} in October (low river flow). During October, the removal rate from the water column was similar to the sorption rate onto particles from the dissolved phase. This implies that during low-to-normal flow conditions, the residence time of ^{7}Be (and by analogy other particle-reactive radionuclides) in the water column, is primarily controlled by sorption kinetics. During periods of high flow events and pronounced sediment resuspension, however, the residence time of particle-reactive radionuclides in the water column is primarily controlled by fine-particle dynamics rather than sorption kinetics.

- Reactor-released radionuclide concentrations in waters below Conowingo Dam were greatly reduced because of rapid sorption and removal in Conowingo Pond. The trapping efficiency of Conowingo Pond for reactor-released radiocesium, radiocobalt and radiozinc, varies with seasonal and storm-flow conditions. During low-to-normal flow conditions, it appears that greater than 80% of the ^{137}Cs, ^{134}Cs, ^{65}Zn, and ^{60}Co released from the Peach Bottom Atomic Power Station is sorbed to suspended matter and deposited in the Pond sediments. During high flow conditions, Pond sediments and associated radionuclides may be resuspended and transported into Upper Chesapeake Bay. The net affect of deposition and resuspension on the long-term trapping efficiency of Conowingo Pond have been estimated by comparing reactor-radionuclide inputs with radionuclide concentrations measured in a 5-year series of surface sediment collections. These results indicate that about 30% of the ^{60}Co and ^{65}Zn, and about 15% of the ^{134}Cs released from the PB nuclear plant has been retained within the reservoir sediments (Mclean et al. 1988).

- Particle-associated radionuclides which escape Conowingo Pond are rapidly buried by sediment deposition within the turbidity zone of Chesapeake Bay. The residence time of particle-reactive radionuclides within the turbidity zone appears to be less than a day. Rapid burial of radionuclides in the sediments of the estuarine turbidity zone helps to isolate these contaminants from exposure to humans and from contact with epibenthic and pelagic biota.

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APPENDIX A

MONTHLY DEPOSITIONAL FLUXES FOR ^{7}Be and ^{210}Pb

Table A-1. Total monthly depositional fluxes of ^{7}Be and ^{210}Pb at Norfolk, Virginia (36°35'N, 76°18'W)

Date	Total Precipitation (cm)	^{7}Be Flux^{a} (pCi/cm^{2})	^{7}Be Inventory^{b} (pCi/cm^{2})	^{210}Pb Flux (pCi/cm^{2})
15-Oct-82	18.2	0.49±0.01		0.043±0.006
15-Nov-82	7.7	0.29±0.01		0.036±0.002
15-Dec-82	12.5	0.52±0.02	0.94	0.071±0.009
15-Jan-83	4.4	0.26±0.03	0.89	0.017±0.005
14-Feb-83	16.0	0.71±0.02	1.31	0.047±0.008
15-Mar-83	13.7	0.82±0.05	1.72	0.043±0.007
15-Apr-83	16.1	0.76±0.05	1.91	0.067±0.006
15-May-83	8.2	0.33±0.02	1.62	0.021±0.004
15-Jun-83	12.8	0.51±0.03	1.60	0.027±0.007
15-Jul-83	2.7	0.22±0.03	1.30	0.014±0.003
15-Aug-83	4.1	0.35±0.02	1.22	0.027±0.003
15-Sep-83	15.3	0.20±0.01	1.02	0.024±0.005
15-Oct-83	18.0	0.41±0.04	1.10	0.023±0.006
15-Nov-83	5.2	0.25±0.02	0.98	0.015±0.003
15-Dec-83	14.3	0.59±0.03	1.26	0.031±0.005
15-Jan-84	8.0	0.32±0.03	1.16	0.019±0.003
15-Feb-84	13.3	0.58±0.04	1.36	0.025±0.004
15-Mar-84	13.0	0.73±0.07	1.66	0.061±0.004
15-Apr-84	11.6	0.78±0.03	1.89	0.056±0.005
15-May-84	17.5	0.71±0.02	1.99	0.026±0.005
15-Jun-84	2.5	0.16±0.01	1.51	0.006±0.002
15-Jul-84	23.3	0.95±0.01	1.96	0.057±0.007
15-Aug-84	13.6	0.48±0.02	1.79	0.046±0.003
15-Sep-84	5.2	0.34±0.02	1.54	0.016±0.003
15-Oct-84	1.7	0.09±0.01	1.13	0.014±0.003
15-Nov-84	16.6	0.53±0.02	1.29	0.035±0.003
15-Dec-84	2.5	0.13±0.01	1.00	0.013±0.003
15-Jan-85	19.6	0.43±0.02	1.10	0.027±0.004
15-Feb-85	11.5	0.27±0.02	1.00	0.025±0.003
15-Mar-85	4.7	0.3±0.01^{c}	0.99	0.025±0.004^{c}
15-Apr-85	11.2	0.4±0.07^{c}	1.14	0.036±0.002^{c}
15-May-85	12.8	0.58±0.02	1.45	0.029±0.002^{c}
15-Jun-85	20.2	0.74±0.03	1.72	0.018±0.002^{c}
15-Jul-85	21.1	1.09±0.03	2.00	0.058±0.003^{c}
15-Aug-85	5.6	0.29±0.02	1.62	0.046±0.002^{c}
15-Sep-85	19.5	0.82±0.04	1.94	0.040±0.002^{2}
		Rain Storm Conowingo Pond^{d}		
2-4-Oct-85	2.8	0.15±0.02	1.95	0.004±0.001

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[a] The integrated monthly data have been decay-corrected to the midpoint of the sampling period.

[b] The ^{7}Be inventory data have been calculated by consecutively decay-correcting the prior month's standing crop and adding this residual amount to the current month's flux.

[c] Norfolk samples were lost and data for ORNL has been substituted.

[d] Integrated precipitation sample collected at Conowingo Pond to determine the input of ^{7}Be during the three days of large-volume water collection (see Table 4).

APPENDIX B

HISTORY OF REACTOR RELEASES INTO THE SUSQUEHANNA RIVER AND CHESAPEAKE BAY

Table B-1. Annual liquid releases (in Curies) from the Peach Bottom Atomic Power Station from 1975 to 1985

Date	CS-137	Decay-COR 05-Oct-85	CS-134	Decay-COR 05-Oct-85	CO-60	Decay-COR 05-Oct-85	CO-58	Decay-COR 05-Oct-85	ZN-65	Decay-COR 05-Oct-85
31-Dec-75		0.00E+00		0.00E+00	2.00E-02	5.52E-03		0.00E+00	5.00E-02	2.08E-06
31-Dec-76	8.20E-01	6.70E-01	8.00E-01	4.44E-02	2.00E-02	6.30E-03		0.00E+00	2.00E-01	2.34E-05
31-Dec-77	1.30E+00	1.09E+00	1.40E+00	1.08E-01	1.20E-01	4.31E-02		0.00E+00	1.05E+00	3.45E-04
31-Dec-78	8.10E-01	6.93E-01	2.86E+00	3.07E-01	1.55E-01	6.35E-02	2.73E-02	9.20E-13	4.24E-01	3.91E-04
31-Dec-79	3.26E+00	2.85E+00	3.92E+00	5.85E-01	1.62E-01	7.58E-02	2.40E-02	2.85E-11	4.60E-01	1.19E-03
31-Dec-80	6.91E-01	6.19E-01	5.68E-01	1.18E-01	1.56E-01	8.33E-02	2.34E-02	9.91E-10	3.06E-01	2.23E-03
31-Dec-81	1.70E-01	1.56E-01	9.93E-02	2.87E-02	1.30E-01	7.92E-02	9.75E-03	1.46E-08	2.90E-01	5.95E-03
31-Dec-82	6.50E-01	6.10E-01	4.60E-01	1.85E-01	6.50E-01	4.52E-01	1.30E-01	6.85E-06	1.96E+00	1.13E-01
31-Dec-83	2.56E-01	2.46E-01	1.47E-01	8.21E-02	1.57E-01	1.24E-01	1.72E-02	3.20E-05	7.86E-01	1.27E-01
31-Dec-84	2.11E-01	2.07E-01	1.64E-01	1.28E-01	1.12E-01	1.01E-01	4.56E-03	3.02E-04	2.11E-01	9.61E-02
31-Oct-85	1.75E-01	1.75E-01	1.64E-01	1.68E-01	1.31E-01	1.32E-01	2.20E-03	2.83E-03	1.74E-01	1.87E-01
Total input (Curies)	8.34	7.89[a]	10.6	1.67	1.81	1.11	0.24	0.0013	5.91	0.34

[a] Decay corrected yearly from 1975 to 1981 and monthly from 1982 to 1985.

Table B-2. Monthly liquid releases (in Curies) from the Peach Bottom Atomic Power Station from January 1982 to November 1985

Date	CS-137	Decay-COR 05-Oct-85	CS-134	Decay-COR 05-Oct-85	CO-60	Decay-COR 05-Oct-85	CO-58	Decay-COR 05-Oct-85	ZN-65	Decay-COR 05-Oct-85
31-Jan-82	7.80E-02	7.16E-02	4.90E-02	1.46E-02	1.50E-02	9.24E-03	1.20E-03	2.43E-09	4.90E-02	1.10E-03
28-Feb-82	7.80E-02	7.18E-02	6.70E-02	2.04E-02	2.10E-02	1.31E-02	4.20E-03	1.12E-08	7.30E-02	1.77E-03
31-Mar-82	7.40E-02	6.82E-02	5.30E-02	1.66E-02	3.80E-02	2.39E-02	4.00E-03	1.44E-08	1.10E-01	2.91E-03
30-Apr-82	7.10E-01	6.56E-01	4.50E-02	1.45E-02	1.40E-01	8.90E-02	1.10E-02	5.30E-08	2.20E-01	6.33E-03
31-May-82	8.00E-02	7.40E-02	4.80E-02	1.59E-02	1.10E-01	7.07E-02	7.90E-03	5.16E-08	2.40E-01	7.54E-03
30-Jun-82	1.20E-01	1.11E-01	9.10E-02	3.10E-02	8.70E-02	5.66E-02	8.90E-03	7.78E-08	4.20E-01	1.44E-02
31-Jul-82	8.20E-02	7.62E-02	5.40E-02	1.69E-02	5.60E-02	3.68E-02	1.30E-02	1.54E-07	1.80E-01	6.72E-03
31-Aug-82	4.70E-02	4.38E-02	3.40E-02	1.22E-02	1.60E-01	1.06E-01	7.30E-02	1.17E-06	5.30E-01	2.16E-02
30-Sep-82	7.80E-03	7.27E-03	5.10E-03	1.89E-03	1.50E-02	1.01E-02	6.50E-03	1.40E-07	9.60E-02	4.26E-03
31-Oct-82	1.50E-03	1.40E-03		0.00E+00	4.60E-03	3.13E-03		0.00E+00	1.40E-02	6.78E-04
30-Nov-82		0.00E+00		0.00E+00		0.00E+00		0.00E+00		0.00E+00
31-Dec-82	1.10E-02	1.03E-02	9.30E-03	3.74E-03	7.60E-03	5.28E-03	2.00E-03	1.05E-07	2.30E-02	1.32E-03
31-Jan-83	2.40E-03	2.26E-03	1.30E-03	5.37E-04	4.40E-03	3.09E-03	1.70E-03	1.21E-07	2.70E-02	1.70E-03
28-Feb-83	4.20E-02	3.95E-02	2.10E-02	8.90E-03	1.40E-02	9.94E-03	3.50E-03	3.28E-07	1.20E-01	8.16E-03
31-Mar-83	8.10E-02	7.64E-02	4.30E-02	1.87E-02	4.20E-02	3.01E-02	7.00E-03	8.88E-07	1.20E-01	8.91E-03
30-Apr-83	2.40E-02	2.27E-02	2.10E-02	9.40E-03	5.00E-03	3.63E-03	1.60E-04	2.72E-08	2.40E-02	1.94E-03
31-May-83	1.20E-02	1.14E-02	7.70E-03	3.55E-03	9.60E-03	7.04E-03	5.00E-04	1.15E-07	5.20E-02	4.59E-03
30-Jun-83	5.30E-02	5.03E-02	2.60E-02	1.23E-02	3.00E-02	2.22E-02	2.00E-03	6.17E-07	2.20E-01	2.11E-02
31-Jul-83	7.57E-03	7.20E-03	4.67E-03	2.27E-03	7.15E-03	5.36E-03	1.84E-04	7.68E-08	4.15E-02	4.35E-03
31-Aug-83	1.32E-02	1.26E-02	8.94E-03	4.47E-03	2.31E-02	1.75E-02	1.94E-03	1.10E-06	7.82E-02	8.95E-03
30-Sep-83	1.04E-02	9.93E-03	4.02E-04	2.07E-04	2.97E-03	2.28E-03		0.00E+00	9.92E-03	1.24E-03
31-Oct-83	1.25E-02	1.20E-02	8.58E-03	4.54E-03	8.20E-03	6.36E-03	1.72E-05	1.76E-08	6.36E-02	8.65E-03
30-Nov-83	3.16E-03	3.03E-03	2.11E-03	1.15E-03	3.38E-03	2.65E-03	1.01E-05	1.39E-08	1.31E-02	1.94E-03
31-Dec-83	4.22E-03	4.05E-03	2.27E-03	1.27E-03	6.64E-03	5.26E-03	1.69E-04	3.14E-07	1.21E-02	1.96E-03
31-Jan-84	8.63E-04	8.30E-04	2.48E-04	1.43E-04	3.16E-03	2.53E-03	9.38E-05	2.36E-07	3.57E-03	6.30E-04
28-Feb-84	3.39E-04	3.27E-04		0.00E+00	1.72E-03	1.39E-03		0.00E+00	1.94E-03	3.71E-04
31-Mar-84	6.12E-05	5.91E-05		0.00E+00	5.31E-04	4.35E-04		0.00E+00		0.00E+00
30-Apr-84	1.31E-02	1.27E-02	6.15E-03	3.83E-03	2.18E-02	1.80E-02	1.36E-03	8.24E-06	5.49E-02	1.25E-02
31-May-84	1.56E-02	1.51E-02	5.04E-03	3.23E-03	1.46E-02	1.22E-02	5.41E-04	4.44E-06	2.40E-02	5.97E-03
30-Jun-84	6.33E-03	6.15E-03	4.72E-03	3.11E-03	1.04E-02	8.80E-03	1.76E-04	1.94E-06	1.83E-02	4.95E-03
31-Jul-84	4.65E-04	4.52E-04	1.22E-04	8.26E-05	5.67E-04	4.85E-04		0.00E+00	1.41E-03	4.17E-04
31-Aug-84	5.40E-03	5.26E-03	3.53E-03	2.46E-03	2.56E-02	2.22E-02	8.67E-04	1.75E-05	1.35E-02	4.35E-03
30-Sep-84	7.05E-05	6.89E-05		0.00E+00	3.66E-04	3.20E-04		0.00E+00	4.94E-04	1.73E-04
31-Oct-84	3.22E-02	3.15E-02	2.01E-02	1.48E-02	9.35E-03	8.27E-03	1.82E-04	6.65E-06	2.42E-02	9.27E-03
30-Nov-84	2.89E-02	2.83E-02	2.52E-02	1.91E-02	8.94E-03	8.00E-03	4.83E-04	2.36E-05	1.33E-02	5.55E-03
31-Dec-84	1.08E-01	1.06E-01	9.93E-02	7.72E-02	1.54E-02	1.39E-02	8.53E-04	5.65E-05	5.51E-02	2.51E-02
31-Jan-85	4.21E-03	4.14E-03	3.34E-03	2.67E-03	4.53E-03	4.14E-03	8.20E-05	7.35E-06	6.01E-03	2.99E-03
28-Feb-85	1.09E-02	1.07E-02	1.23E-02	1.01E-02	6.46E-03	5.97E-03	2.11E-04	2.49E-05	2.17E-02	1.17E-02
31-Mar-85	8.36E-03	8.26E-03	7.09E-03	5.98E-03	2.30E-02	2.15E-02	1.98E-04	3.16E-05	1.04E-02	6.11E-03
30-Apr-85	1.19E-03	1.18E-03	8.11E-04	7.03E-04	1.15E-03	1.09E-03		0.00E+00	1.74E-03	1.11E-03
31-May-85	5.81E-02	5.76E-02	5.28E-02	4.71E-02	2.54E-02	2.43E-02	1.20E-04	3.47E-05	2.45E-02	1.71E-02
30-Jun-85	3.66E-02	3.64E-02	3.21E-02	2.94E-02	2.85E-02	2.75E-02	1.72E-04	6.67E-05	3.63E-02	2.76E-02
31-Jul-85	2.01E-02	2.00E-02	1.90E-02	1.79E-02	1.15E-02	1.12E-02	2.78E-04	1.46E-04	9.86E-03	8.16E-03
31-Aug-85	4.46E-03	4.45E-03	4.28E-03	4.15E-03	1.46E-02	1.44E-02	8.18E-04	5.81E-04	2.25E-02	2.04E-02
30-Sep-85	1.54E-02	1.54E-02	1.64E-02	1.63E-02	8.37E-03	8.35E-03	2.41E-04	2.30E-04	2.26E-02	2.23E-02
31-Oct-85	1.56E-02	1.56E-02	1.57E-02	1.61E-02	7.38E-03	7.45E-03	7.63E-05	9.83E-05	1.79E-02	1.93E-02

Table B-3. Annual liquid releases (in Curies) from the Calvert Cliffs Nuclear Power Plant from 1975 to 1985

Date	CS-137	Decay-COR 05-Oct-85	CS-134	Decay-COR 05-Oct-85	CO-60	Decay-COR 05-Oct-85	CO-58	Decay-COR 05-Oct-85	ZN-65	Decay-COR 05-Oct-85
31-Dec-75	1.12E-01	8.94E-02	4.36E-05	1.74E-06	2.90E-02	8.00E-03	5.63E-01	4.28E-16		0.00E+00
31-Dec-76	3.01E-01	2.46E-01	7.31E-02	4.05E-03	5.05E-02	1.59E-02	1.88E-01	5.09E-15		0.00E+00
31-Dec-77	4.36E-01	3.64E-01	2.13E-01	1.64E-02	1.84E-01	6.61E-02	1.18E+00	1.13E-12		0.00E+00
31-Dec-78	4.65E-01	3.98E-01	3.08E-01	3.30E-02	4.62E-01	1.89E-01	1.97E+00	6.64E-11		0.00E+00
31-Dec-79	5.68E-01	4.97E-01	3.94E-01	5.88E-02	3.25E-01	1.52E-01	3.81E+00	4.53E-09		0.00E+00
31-Dec-80	1.83E-01	1.64E-01	1.06E-01	2.20E-02	4.41E-01	2.35E-01	2.00E+00	8.47E-08	2.40E-02	1.75E-04
31-Dec-81	1.00E-01	9.17E-02	5.00E-02	1.44E-02	2.10E-01	1.28E-01	1.30E+00	1.94E-06		0.00E+00
31-Dec-82	8.00E-01	7.51E-01	4.40E-01	1.77E-01	1.90E-01	1.32E-01	1.50E+00	7.91E-05		0.00E+00
31-Dec-83	3.15E-01	3.02E-01	1.81E-01	1.01E-01	8.00E-02	6.34E-02	6.05E-01	1.13E-03		0.00E+00
31-Dec-84	2.43E-01	2.39E-01	1.05E-01	8.17E-02	1.00E-01	9.05E-02	5.65E-01	3.74E-02		0.00E+00
31-Dec-85	2.58E-01	2.59E-01	1.26E-01	1.36E-01	1.03E-01	1.06E-01	9.06E-01	2.12E+00		0.00E+00
Total input (Curies)	3.78	3.26[a]	2.00	0.54	2.17	1.12	14.6	0.25	0.024	0.05

[a] Decay corrected yearly from 1975 to 1983 and quarterly from 1984 to 1985.

Table B-4. Quarterly liquid releases (in Curies) from the Calvert Cliffs Nuclear Power Plant from October 1983 to October 1985

Date	CS-137	Decay-COR 05-Oct-85	CS-134	Decay-COR 05-Oct-85	CO-60	Decay-COR 05-Oct-85	CO-58	Decay-COR 05-Oct-85	AG-110M	Decay-COR 05-Oct-85
01-Oct-83	2.33E-02	2.22E-02	3.27E-02	1.68E-02	1.97E-02	1.51E-02	1.07E-01	8.19E-05	1.22E-02	1.63E-03
01-Jan-84	5.78E-02	5.55E-02	5.83E-02	3.26E-02	1.93E-02	1.53E-02	2.35E-01	4.41E-04	9.49E-03	1.63E-03
01-Apr-84	1.28E-01	1.24E-01	5.51E-02	3.35E-02	1.36E-02	1.11E-02	2.79E-02	1.27E-04	4.05E-03	8.93E-04
01-Jul-84	8.60E-02	8.35E-02	4.18E-02	2.76E-02	4.67E-02	3.95E-02	2.47E-01	2.74E-03	3.89E-02	1.10E-02
01-Oct-84	1.01E-02	9.87E-03	1.69E-03	1.21E-03	2.36E-02	2.07E-02	5.78E-02	1.58E-03	3.16E-02	1.15E-02
01-Jan-85	1.92E-02	1.89E-02	5.91E-03	4.60E-03	1.62E-02	1.47E-02	2.32E-01	1.55E-02	2.07E-02	9.69E-03
01-Apr-85	5.49E-02	5.43E-02	1.89E-02	1.60E-02	9.59E-03	8.96E-03	1.10E-01	1.77E-02	1.53E-02	9.17E-03
01-Jul-85	3.46E-02	3.44E-02	1.67E-02	1.53E-02	1.65E-02	1.59E-02	4.21E-01	1.65E-01	5.74E-03	4.41E-03
01-Oct-85	3.20E-02	3.20E-02	1.65E-02	1.64E-02	1.87E-02	1.87E-02	4.50E-02	4.33E-02	2.34E-03	2.31E-03

Table B-5. Daily liquid releases (in mCi) from the Calvert Cliffs Nuclear Power Plant from April 1985 to October 1985

Date	^{137}Cs	^{134}Cs	^{60}Co	^{58}Co	^{65}Zn
19-Apr-85	37	12	177		194
25-Apr-85	19		336		360
25-Apr-85	1063	800	274		686
03-May-85	45		267		340
04-May-85	24		93		161
08-May-85	42	28	153		93
10-May-85	46	21	57		66
11-May-85	3581	3042	15756	67	9623
12-May-85	418	349	599		681
13-May-85	115	1059	2166		1745
14-May-85	1721	1649	673		1058
15-May-85	3999	3885	700	45	681
16-May-85	3832	3637	319		442
17-May-85	7174	11541	1353		1055
18-May-85	8107	7208	727		1521
19-May-85	9889	4254	4665		1396
20-May-85	6304	5562	420		2110
21-May-85	4994	4952	393		1316
22-May-85	4341	3888	1019		1033
24-May-85	406	362	722		390
25-May-85	660	581	436		571
28-May-85	50	75	160		275
31-May-85	4357	3828	438		379
01-Jun-85	104	67	244		505
02-Jun-85	576	534	1170		2565
03-Jun-85	1337	1220	2439		1453
03-Sep-85	200	151	300	34	560
04-Sep-85	1358	1444	915		2059
05-Sep-85	29	23	72		160
06-Sep-85	1168	1192	876	21	2094
07-Sep-85	978	1052	980	60	1375
08-Sep-85	576	531	1322		6575
09-Sep-85	417	462	432	21	859
11-Sep-85	3107	3300	834	25	1207
12-Sep-85	32	91	229	30	496
17-Sep-85	635	703	270	26	445
18-Sep-85	237	235	291		1073
19-Sep-85	378	426	339	22	963
21-Sep-85	162	571	387		1387
22-Sep-85	105	123	89		386
23-Sep-85	169	163	63		386
25-Sep-85	495	488	165		684
26-Sep-85	172	184	187		564
27-Sep-85	657	751	168		488
29-Sep-85	2324	2522	251		511
02-Oct-85	153	176	426		768
03-Oct-85	74	85	208		552
04-Oct-85	97	129	341	41	702
05-Oct-85	186	163	195	23	778
06-Oct-85	139	137	129		488
07-Oct-85	104	94	108		346
08-Oct-85	62	92	130		513
09-Oct-85	147	101	210		751
13-Oct-85	242	257	422		941

APPENDIX C

FLOW AND SUSPENDED MATTER CONCENTRATIONS FOR THE SUSQUEHANNA RIVER AT CONOWINGO DAM

Table C-1. Susquehanna River data at Conowingo, Maryland

Year	Month	Total flow (m^3/Sec)	Mean flow (m^3/Sec)	Suspended concentration (mg/L)
1982	October	5,306	171	
	November	9,595	320	
	December	20,394	658	27
1983	January	17,102	552	
	February	36,274	1,296	6
	March	44,469	1,435	
	April	104,365	3,480	49
	May	70,920	2,280	29
	June	28,164	939	
	July	15,346	495	129
	August	6,602	213	
	September	4,517	151	6
	October	6,072	196	
	November	19,249	642	9
	December	80,278	2,590	
1984	January	18,444	595	7
	February	95,063	3,279	286[b]
	March	49,011	1,581	
	April	111,252	3,710	212
	May	61,436	1,982	11
	June	45,067	1,505	
	July	33,948	1,095	23
	August	25,302	816	15
	September	8,194	273	12
	October	7,008	226	7
	November	12,900	430	7
	December	48,289	1,558	11
1985	January	26,172	844	2
	February	29,011	1,036	19[c]
	March	49,337	1,591	24[d]
	April	44,305	1,477	23[c]
	May	21,487	693	13
	June	14,342	478	27
	July	9,395	303	10
	August	6,811	220	18 [f]
	September	8,910	297	10
	October	12,540	404	
	November	45,227	1,508	
	December	47,380	1,528	

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[a] U.S. Geological Survey water-data reports MD-DE-82-1 to MD-DE-85-1.

[b] Average of seven samples taken on Feb. 17, 1984.

[c] Average of three samples, Feb 21, 27, 28, 1985.

[d] Average of two samples, March 1, 6, 1985.

[e] Average of four samples, April 3, 4, 5, and 22, 1985.

[f] Average of two samples, August 7, 22, 1985.

APPENDIX D

HYDROLOGICAL CHARACTERISTICS OF THE SUSQUEHANNA RIVER DURING SAMPLE COLLECTION

Table D-1. Environmental characteristics of the large-volume water Samples

Sample Date (YY-MM-DD)	Location	Salinity (ppt)	Susp. Concen.[a] (mg/l)	Susquehanna Discharge[b] (m^3/S)	Precip (mm)
3-Oct-83	Conowingo Pond	FW	6	183	-
3-Oct-83	Susq. Flats	FW	6	183	-
4-Oct-83	Annapolis	12	4	236	-
5-Oct-83	Calvert Cliffs	14	4	233	0.8
4-Apr-84	Conowingo Pond	FW	15	3115	17.8
3-Apr-84	Susp. Flats	Fw	19	2781	-
4-Apr-84	Annapolis	9	24	3115	17.8
5-Apr-84	Calvert Cliffs	8	11	4446	7.9
16-May-85	Conowingo Pond	FW	12	552	3.8
17-May-85	Conowingo Pond	FW	17	620	29.0
15-May-85	Susq. Flats	FW	7	708	Trace
20-May-85	Annapolis	11	5	895	-
21-May-85	Calvert Cliffs	14	2	782	10.2
2-Oct-85	Conowingo Pond	FW	5	1008	9.7
3-Oct-85	Conowingo Pond	FW	5	883	11.9
4-Oct-85	Conowingo Pond	FW	5	639	-
4-Oct-85	Susq. Flats	FW	16	639	-
8-Oct-85	Grove Pt.	3	14	646	-
9-Oct-85	Howell Pt.	6	10	538	-
8-Oct-85	Annapolis	15	4	646	-
7-Oct-85	Calvert Cliffs	20	5	537	-

[a] Suspended-matter concentration in the collected sample as recovered from the continuous-flow centrifuge.

[b] Data are from U.S. Geological Survey Water-Data Reports MD-DE-82-1 to MD-DE-85-1.

APPENDIX E

SUSPENDED PARTICLE AND SEDIMENT COMPOSITIONS

APPENDIX E

E.1. PARTICULATE STABLE-ELEMENT CHEMISTRY

The major and trace element chemistry of the suspended matter and the bottom sediments at each of our four principal collection sites is presented in Tables E-1 and E-2. The inorganic composition of the suspended matter is fairly constant throughout the year. Variations in elemental concentrations primarily reflect variations in the amount of organic matter present in the sample. Total suspended-matter carbon concentrations in Conowingo Pond range from about 3.2% during the winter and spring when freshwater flow and sediment resuspension are high to about 11% in the summer and fall when freshwater flow is low and biogenic particle production in the water column is high. Total carbon concentrations of the suspended matter in the mesohaline bay waters near Calvert Cliffs range from 16 to 34% (Table E-1). Total carbon concentrations in the bottom sediments are typically much lower, averaging about 3% but ranging as high as 8% in the fine-grained muds behind Conowingo Dam. In addition to containing less C and P, bottom sediments generally contained less Al, K, and Ca and more Si relative to suspended matter. This probably attests to the coarser-grained nature of bottom sediments and the resultant enrichment in quartz relative to aluminosilicate clay minerals.

E.2 PARTICULATE MINERALOGY

Suspended particles and sediments collected during May 1984 (Table 3) were also analyzed for their clay mineralogy by X-ray diffraction. Subsamples were transferred to a small beaker containing about 15 mL of deionized distilled water and sonified for 15 min to disperse any agglomerated particles. About 1-2 mL of the sonified solution was pipetted directly onto a glass slide and allowed to dry overnight. The slides were analyzed by X-ray diffraction using the copper K_α X-ray (1.54 A). Each sample was scanned over the 2 theta angle (from 2 to 30°) at 0.5° per minute, and the resulting spectrum was recorded on a strip chart. In order to obtain specific

Table E-1. Suspended-matter stable element chemistry[a]

Element	Conowingo Pond			Susquehanna Flats			Annapolis			Calvert Cliffs			USGS STD Sediment	
(µg/g)	May 85	Apr 84	Oct 83	May 85	Apr 84	Oct 83	May 85	Apr 84	Oct 83	May 85	Apr 84	Oct 83	Measured	(certified)
Al	73,000	100,000	80,000	71,000	100,000	78,000	53,000	91,000	51,000	16,000	27,000	36,000	92,000	(87,000)
Ba	540	620	620	540	660	590	380	480	270	1,200	160	230	470	(500)
Be	30	4.9	4.8	3.5	5.3	4.5	3.3	4.6	15	1.9	1.5	3.0	3.6	(3.1)
Ca	3,000	3,000	3,800	3,000	3,300	3,400	2,100	2,400	3,700	5,100	2,000	3,000	9,700	(10,400)
Cd	1.7	≤1.8	2.6	3.5	≤1.8	3.2	1.3	≤1.8	≤1.8	1.7	3.9	≤1.8	≤1.8	--- -
Co	64	44	68	64	40	53	23	23	22	7.1	6.4	8.9	22	(20)
Cr	77	98	88	82	100	100	81	120	120	60	66	72	110	(104)
Cu	210	82	260	140	85	270	260	170	420	640	220	470	29	(30)
Fe	45,000	55,000	85,000	44,000	57,000	64,000	32,000	47,000	58,000	14,000	14,000	39,000	49,000	(48,000)
K	20,500	24,000	20,000	19,700	25,000	19,000	15,700	22,000	13,000	5,800	7,900	9,600	30,000	(30,000)
K(K-40)	20,400	28,000	18,000	25,300	29,400	18,100	17,400	24,200	14,000	8,400	6,500	12,700	-----	-- -
Mg	6,400	8,000	6,500	6,400	8,800	6,800	5,500	8,300	7,400	7,500	5,300	6,400	17,000	(18,100)
Mn	6,400	1,500	7,800	6,800	1,300	4,400	1,300	1,400	1,100	160	220	600	770	(760)
Ni	110	83	110	120	78	100	50	69	59	35	36	26	51	(55)
P	3,200	1,600	3,900	3,900	1,700	4,100	3,500	2,900	6,600	4,100	2,500	5,700	550	(760)
Pb[b]	260	210	340	200	390	450	200	310	450	530	210	440	24	(24)
Si	240,000	230,000	190,000	230,000	230,000	200,000	210,000	200,000	150,000	99,000	78,000	180,000	220,000	(234,000)
Sr	79	80	78	73	77	69	68	330	85	95	120	78	130	(160)
Ti	3,000	4,100	2,400	2,500	4,300	2,600	2,000	3,100	1,800	770	1,100	1,400	3,600	(4,300)
V	96	140	130	97	150	120	92	150	120	33	51	92	170	(140)
Zn	3,100	570	3,400	1,100	630	2,200	1,400	1,200	4,200	2,000	240	3,200	120	(130)
Zr	90	100	68	70	98	70	110	86	52	24	29	39	83	(130)
Total C (%)	8.9	3.2	10.6	12.4	3.5	10.0	16.9	10.6	22.5	32.6	33.7	15.7	1.9	(1.5)

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[a] Samples were collected with a stainless steel centrifuge, and no precautions were taken to ensure against metal contamination.

[b] Results from Atomic Absorption Spectrometry.

Table E-2. Sediment stable element chemistry

Element	Conowingo Pond		Susq. Flats		Annapolis		Calvert Cliffs		USGS STD Sediment	
(µg/g)	May 85	Oct 83	May 85	Oct 83	May 85	Oct 83	May 85	Oct 83	Measured	(certified)
Al	60,000	87,000	33,000	74,000	68,000	84,000	28,000	84,000	92,000	(87,000)
Ba	440	560	270	480	380	370	180	310	470	(500)
Be	2.8	4.3	1.6	3.3	3.6	3.8	1.8	2.7	3.6	(3.1)
Ca	3,000	3,400	2,300	4,000	5,600	3,600	4,500	7,000	9,700	(10,400)
Cd	0.8	≤1.8	0.5	≤1.8	0.9	≤1.8	1.1	≤1.8		
Co	41	54	24	34	37	33	15	19	22	(20)
Cr	53	80	27	84	97	110	39	77	110	(104)
Cu	46	61	18	39	46	59	28	47	29	(30)
Fe	35,000	48,000	18,000	44,000	44,000	53,000	16,000	35,000	49,000	(48,000)
K	15,700	23,000	9,200	19,000	22,000	22,000	8,700	17,000	30,000	(30,000)
K(K-40)	13,900	21,600	8,300	20,100	21,800	19,600	14,400	16,700	——	——
Mg	5,100	6,600	2,900	6,700	10,000	10,000	4,600	7,700	17,000	(18,100)
Mn	1,800	2,100	1,700	2,900	3,200	3,400	260	600	770	(760)
Ni	67	90	38	72	54	59	≤24	40	51	(55)
P	860	1,300	550	920	1,400	1,400	840	550	550	(760)
Pb*	39	46	20	≤40	61	49	34	≤40	24	(24)
Si	320,000	240,000	280,000	280,000	250,000	190,000	290,000	290,000	220,000	(234,000)
Sr	69	82	40	70	120	110	63	99	130	(160)
Ti	3,300	3,600	1,500	3,600	2,800	3,100	1,700	3,300	3,600	(4,300)
V	72	120	40	110	120	160	48	110	170	(140)
Zn	200	330	100	160	270	320	110	220	120	(130)
Zr	94	120	44	110	85	93	59	110	83	(130)
Total C (%)	8.1	3.8	1.5	2.5	3.2	3.8	1.3	2.2	1.9	(1.5)

mineral identification, the slides were reanalyzed after glycolation for 24 h to test for the presence of certain expandable minerals.

Spectrograms of the untreated samples are shown in Fig. E-1. The major mineral peaks are identified in the spectrogram for Conowingo suspended matter. The same minerals (chlorite, illite, kaolinite, quartz, and feldspar) occurred in both the sediments and suspended matter in Conowingo Pond and in Susquehanna Flats. Treatment with glycol did not alter the respective d-spacing from 14A to 17A, which would have occurred if montmorillonite were present. Treatment with dilute acid (1N HCl) virtually removed the 14A spacing, confirming the presence of cholorite.

Although the mineralogy of the bottom sediments at Annapolis was similar to the mineralogy of the suspended matter and bottom sediments in the Susquehanna, the diffractogram for the suspended particles at Annapolis does not closely mimic that of the sediment. There is an enrichment of organic matter and perhaps chlorite in the suspended material and a decline in the feldspar concentration over that of the sediments.

At Calvert Cliffs the X-ray diffractograms for both the suspended matter and bottom sediments were markedly depleted in aluminosilicate clay minerals. Quartz dominated the sediment sample, followed by traces of illite and kaolinite and possibly chlorite. After heating, illite appears more predominant than kaolinite, suggesting that the minerals were highly hydrated in the sediments. Organic matter was a major component of the suspended matter in this portion of the Bay.

In summary, the elemental and mineralogical characteristics of the suspended matter and bottom were similar throughout the Lower Susquehanna River and Upper Chesapeake Bay. Differences arise from an increase in the organic content of the suspended matter at Annapolis and Calvert Cliffs and an increase in quartz relative to aluminosilicates in the Bay sediments near Calvert Cliffs. The major clay minerals included illite, kaolinite and chlorite. The presence of illite throughout the system allows for the irreversible sorption of

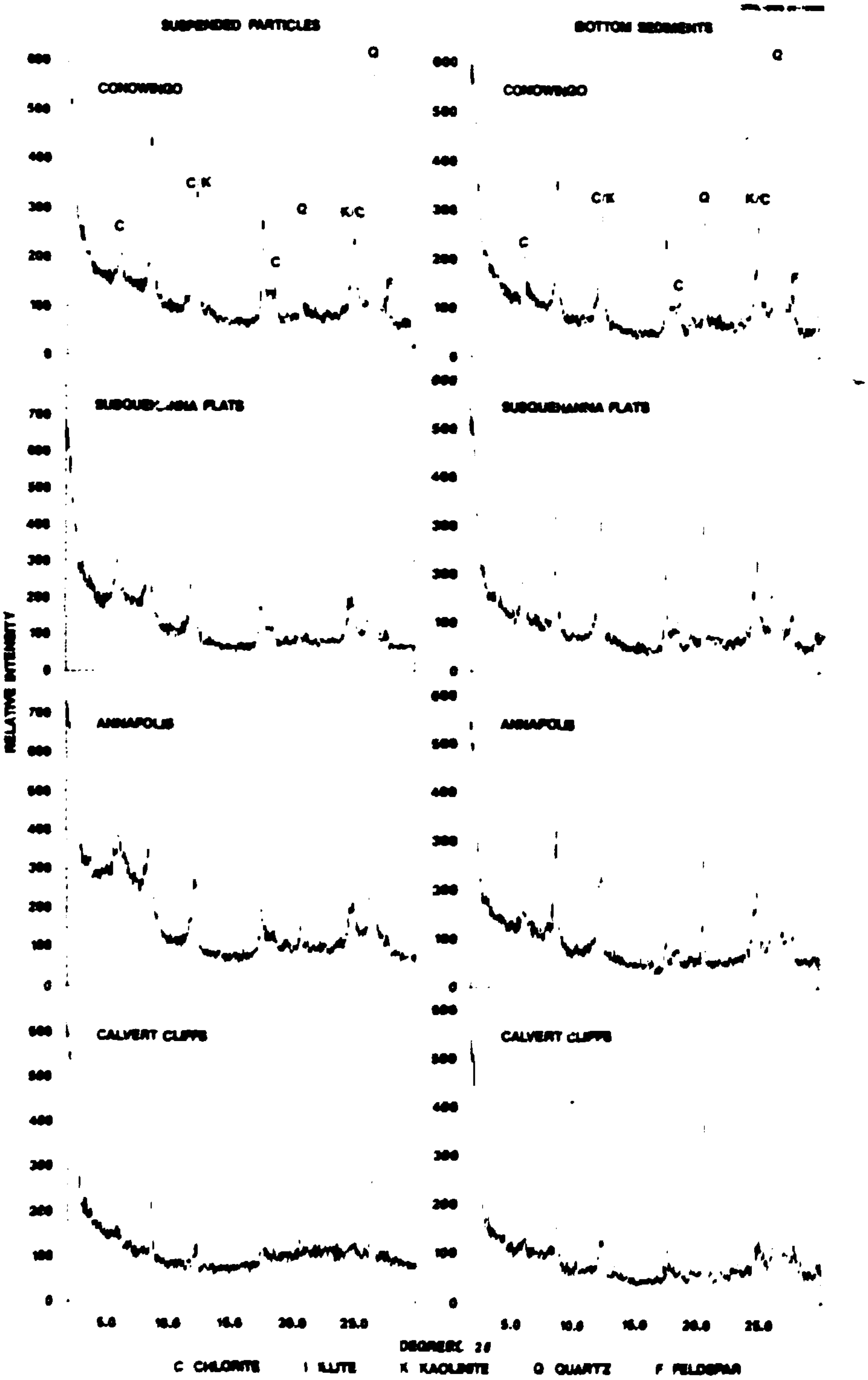

Fig. E-1. X-ray diffractograms of suspended mattter and bottom sediments collected from Conowingo Pond, Susquehanna Flats, Annapolis, and Calvert Cliffs in May 1985. The ordinate axis represents the relative diffracted intensity from the minerals present, and the abscissa represents the diffracted angle degree 2θ, a measure related through Bragg's Law to the d-spacing (Angstroms) between planar surfaces of oriented clay minerals. Thus, the spacing of the measured X-ray lines aids in determining the elements present in the sample. The intensity of each line is related to the concentration.

radiocesium (see Section 4.2), and supports our conclusion that the decrease in the radiocesium particle-to-water sorption ratio in mesohaline waters is a result of cation competition for sorption sites rather than desorption or changes in particulate chemistry. Although previous investigators have reported the occurrence of montmorillonite in subsurface sediment core borings at Annapolis (Owens et al. 1974), our analyses of surface sediment and suspended matter do not reveal the presence of montmorillonite at this location. Hathaway (1972) indicated that only trace amounts of montmorillonite occur in Chesapeake Bay sediments, suggesting that shoreline erosion from areas containing deposits of montmorillonite along the bay may not be a major contributing source.