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Geochemistry and Deposition of ^7Be in River-Estuarine and Coastal Waters

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The atmospheric flux of cosmogenic ^7Be (53.3-day half-life) and the mode of ^7Be deposition in river-estuarine and coastal environments have been examined. The atmospheric flux of ^7Be commonly supports inventories ranging from 1.0 to 2.0 pCi/cm² (1 pCi = 0.037 Bq). Beryllium 7 concentrations in water phase samples, collected across salinity gradients in several estuaries along the eastern coastline of the United States, range from 0.03 to 0.53 pCi/L and primarily reflect variations in ^7Be supply and sorption kinetics. The major process controlling the concentration of ^7Be on estuarine suspended particles appears to be the length of time that these particles remain in the water column. Field particle-to-water distribution coefficients for ^7Be have a median value of about 4×10^4 but range over an order of magnitude reflecting short-term variations in ^7Be input, particle dynamics, and particulate iron content rather than equilibrium sorption-desorption responses to changes in water salinity or particle type. Residence times of ^7Be in the water column range from a few days in estuarine areas of rapid fine-particle deposition, to several weeks in high-energy environments where pronounced sediment resuspension reintroduces deposited ^7Be back into the water column. Inventories of ^7Be in sediments range from nondetectable to 3.3 pCi/cm², with the highest inventories in areas where fine particles are accumulating rapidly. Such sites are also major repositories for other particle-reactive substances. A ^7Be budget for the James estuary indicates that less than 5% of the expected ^7Be input is in the water column and that the short-term estuarine trapping efficiency for atmospherically derived ^7Be is somewhere between 50 and 100%.

1. INTRODUCTION

Beryllium 7 (53.3-day half-life) is produced in the earth's atmosphere by cosmic ray spallation of nitrogen and oxygen. Like other particle-reactive and atmospherically derived radionuclides, ^7Be rapidly becomes associated with aerosols, and its deposition on the earth's surface occurs by precipitation scavenging and by particle dry deposition. The contribution of the latter process (dry deposition) is difficult to evaluate by direct measurements, but is probably less than 30% in humid regions [Olsen *et al.*, 1985]. At mid-latitudes, the total (wet + dry) atmospheric flux of ^7Be commonly supports inventories ranging from 1.0 to 2.0 pCi/cm² [Young and Silker, 1974; Matsunami *et al.*, 1979; Crecelius, 1981; Robbins and Eadie, 1982; Saleh and Kuroda, 1982; Todd, 1984; Olsen *et al.*, 1985]. The highest inventories generally occur in the spring, when mid-latitude folding of the tropopause enhances stratospheric-tropospheric exchange, and the lowest inventories occur during periods of minimum precipitation, generally in late summer and early fall. Upon contact with "acidic" rainwater, ^7Be appears to be solubilized and Be^{2+} is probably the predominant species [Olsen *et al.*, 1985].

Once deposited as a constituent of rainwater, ^7Be is quickly sorbed by above-ground vegetation [Russell *et al.*, 1981] or soil particles [Brown *et al.*, 1981; Lundberg *et al.*, 1983; Monaghan *et al.*, 1983; Pavich *et al.*, 1984] in terrestrial environments, but may remain in solution in oceanic surface waters because of the lack of suspended material [Young and Silker, 1974; Bloom and Crecelius, 1983]. The potential of ^7Be as a

useful tool for examining geochemical and sedimentological processes in estuarine and coastal areas (i.e., transitional zones between terrestrial and oceanic environments) has only recently been investigated [Krishnaswami *et al.*, 1980; Aaboe *et al.*, 1981; Olsen *et al.*, 1982a; Todd, 1984].

In this study, we report ^7Be measurements for water, suspended matter, and sediment samples collected in several river, estuarine, and coastal systems during 1981 to 1984. We have used these data to examine the relationship between the atmospheric flux of ^7Be and its mode of deposition in river-estuarine and coastal environments. From this examination, we present new information concerning (1) the distribution of ^7Be between dissolved and particulate phases in estuarine and coastal areas, (2) factors that affect the concentration of ^7Be on suspended particles, (3) rates for ^7Be removal from the water column, (4) the heterogeneity of ^7Be accumulation patterns in estuarine sediments, and (5) the extent to which ^7Be is trapped within an estuarine environment. Since many chemically reactive substances become associated with particles in aquatic systems [Olsen *et al.*, 1982b], the information obtained from the distribution of ^7Be can be used to determine, understand, and predict the types and rates of processes that affect the fate of other particle-seeking substances in estuarine and coastal areas.

2. DATA COLLECTION AND ANALYSIS

Total (wet + dry) monthly atmospheric deposition data for ^7Be have been measured from October 1982 to October 1984 at Oak Ridge (Tennessee) and near the mouth of the James estuary at Norfolk (Virginia). The collection and analytical procedures used to obtain these atmospheric flux data are discussed by Olsen *et al.* [1985].

Water, suspended matter, and sediment samples were col-

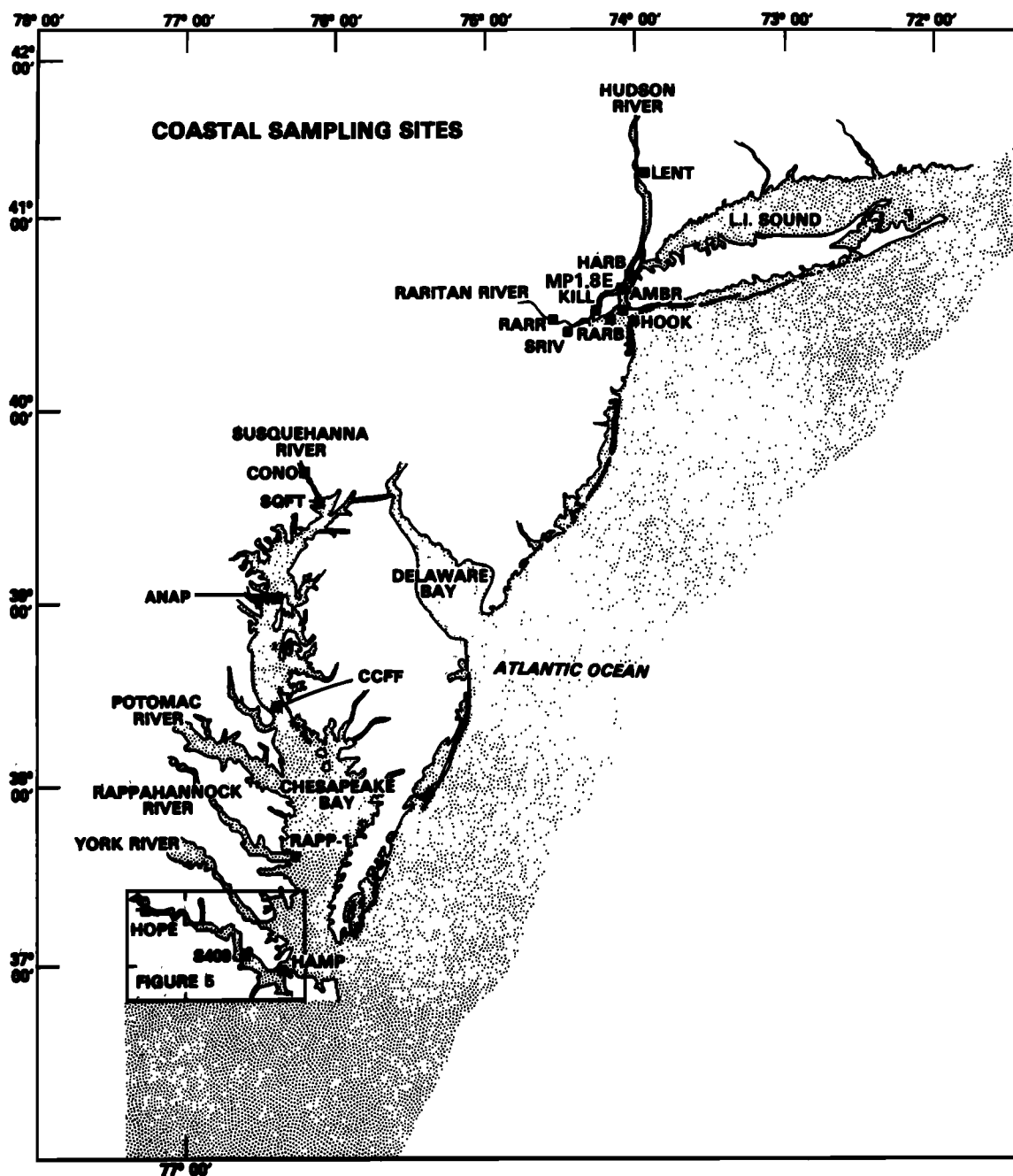


Fig. 1. Location map for large-volume water samples (squares) collected in river, estuarine, and coastal environments along the eastern shoreline of the United States. Circles indicate the location of sediment cores collected at additional sites.

lected across the salinity ranges of the James River estuary (June 16–23, 1981), the Hudson–Raritan Bay system (July 22–28, 1981), and the Susquehanna–Chesapeake Bay system (October 1–5, 1983, and April 2–5, 1984). In the James, water samples ranged from 150 to 300 L and were collected ~0.5 m below the surface. Suspended matter was removed from these samples by gravity settling for ~24 hours. In the Hudson–Raritan estuary and the Susquehanna–Chesapeake system, water samples (750–900 L each) were collected near middepth, and particulate material was removed from these samples by continuous-flow centrifugation. The locations of these large-volume water samples are illustrated in Figure 1.

After particle removal each of the water samples was acidi-

fied to pH ~2 with concentrated HNO_3 and spiked with 40 mL of a 20% FeCl_3 solution and 0.1 g of stable beryllium (in chloride form) to determine recovery yields for ^7Be . The samples were allowed to equilibrate for 4 to 8 hours after which NH_4OH was added to adjust the pH to ~10 and to precipitate $\text{Fe}(\text{OH})_3$ and beryllium as an oxide or hydroxide coprecipitate. Beryllium recovery, as determined by atomic emission spectroscopy (inductively coupled plasma), ranged from 70 to 100% except for one sample (~40%) in which a spill occurred during precipitate collection. At each site of water collection (Figure 1), a sediment core or grab sample was also taken.

Several sediment cores have also been collected in marsh, estuarine, and coastal areas at sites previously identified as

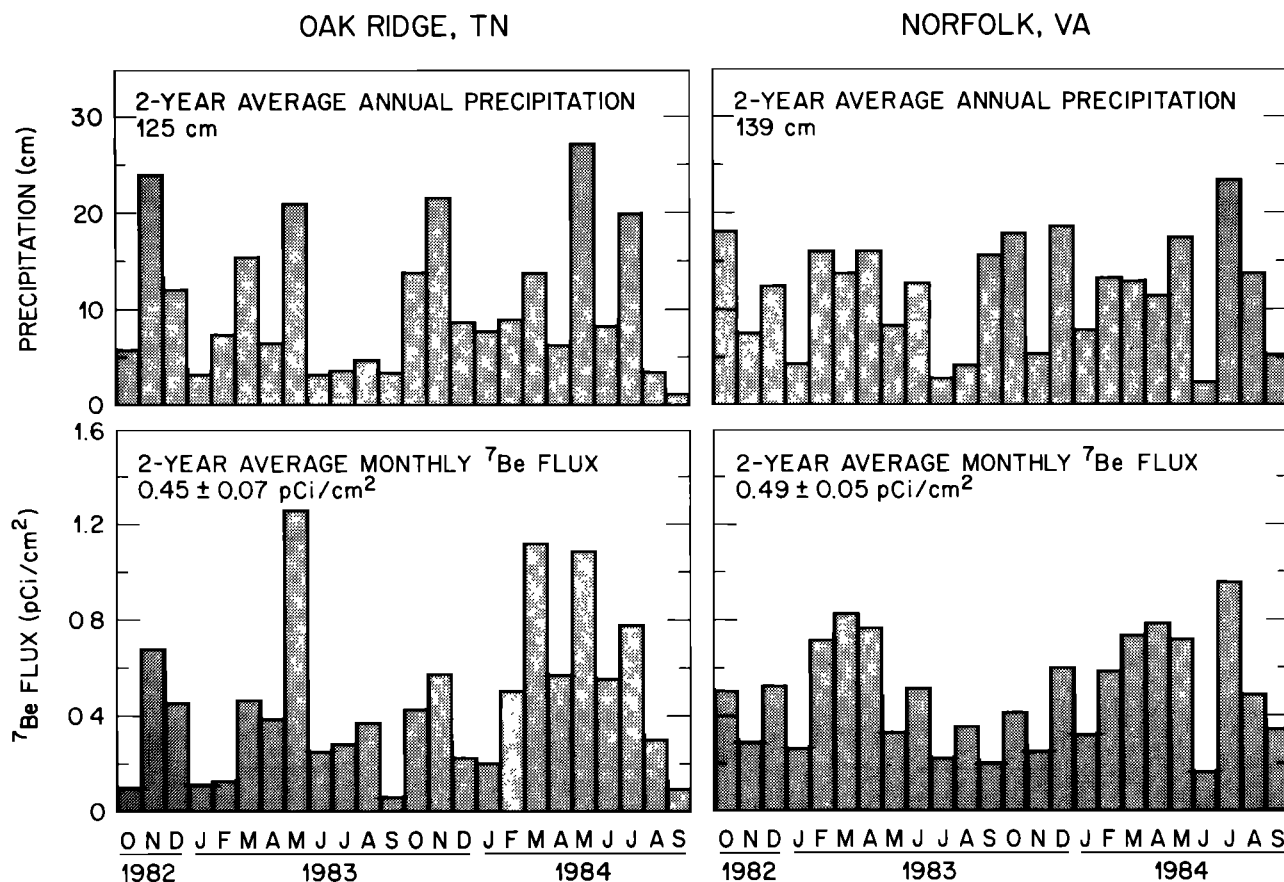


Fig. 2. Bar graph illustrating total monthly precipitation and atmospheric deposition of ⁷Be at Oak Ridge (Tennessee) and Norfolk (Virginia). The samples were collected between October 1982 and October 1984. The 2-year average monthly ⁷Be flux (± 1 standard error of the mean) is 0.45 ± 0.07 pCi/cm² at Oak Ridge and 0.49 ± 0.05 pCi/cm² at Norfolk.

fine-particle and radionuclide sinks [Olsen *et al.*, 1978, 1981a; Cutshall *et al.*, 1981; Todd, 1984]. The ⁷Be concentration and inventory data in sediments have been compared with respective suspended-matter data to evaluate factors that affect ⁷Be concentrations on suspended particles and ⁷Be removal from the water column, and to examine the relationship between the atmospheric flux of ⁷Be and its mode of deposition in these environments. In addition, ⁷Be profiles have been measured and integrated in box cores collected in different sedimentary environments along the salinity range of the James River estuary during June 16–23, 1981. The integrated ⁷Be inventories in these cores have been used to estimate the total burden of ⁷Be residing in the estuarine sediments. The sediment inventory data have been used with the water column data and atmospheric flux data to estimate the extent to which ⁷Be is trapped within the James River estuarine system.

The water-phase Fe(OH)₃ precipitate, suspended-matter and sediment samples were packed into 15- or 125-cm³ petri dishes or into 95-cm³ aluminum cans (depending on sample size) and analyzed for ⁷Be (as well as several other gamma-emitting radionuclides, such as ¹³⁷Cs, ⁶⁰Co, and ⁴⁰K) using lithium-drifted germanium detectors and a computer-based multichannel analyzer system. The calibration procedures, efficiency calculations, and activity computations for this gamma-spectrometric method have been described previously [Larsen and Cutshall, 1981]. Detector resolutions (full width at half maximum (FWHM)) at 478 keV (the ⁷Be photopeak region) were about 1.4 keV. All reported errors are statistical

counting errors only, and are expressed in terms of 1 sigma. The chemical (total carbon, silicon, aluminum, and iron) characteristics of the suspended particles were determined using inductively coupled plasma (ICP) spectroscopy. The total potassium concentrations of the particulate samples have been determined by flame photometry and compared with concentrations determined from the ⁴⁰K activity.

3. RESULTS

The measured-total (wet + dry) monthly flux of ⁷Be ranges from 0.1 to 1.3 pCi/cm² at Oak Ridge and from 0.2 to 1.0 pCi/cm² at Norfolk, with the highest fluxes occurring during March through May at both sites (Figure 2). Beryllium 7 inventories or steady state concentrations, calculated by consecutively decay-correcting the prior month's standing crop and adding this residual amount to the current month's atmospheric flux, are similar at both sites [Olsen *et al.*, 1985]. The inventory data commonly range from 1.0 to 2.0 pCi/cm², with the highest inventories occurring in the spring and the lowest in the fall.

In river-estuarine and coastal waters, along the northeastern seaboard of the United States, total ⁷Be concentrations range from 0.05 to 0.86 pCi/L (Table 1). The highest ⁷Be water-column concentration occurred in a sample collected from the turbidity zone of Chesapeake Bay during a spring rain. The lowest concentration occurred at the same location during early fall when atmospheric inputs and freshwater flow were

TABLE 1. Salinities, Particulate Chemical Characteristics, and ⁷Be Activities in River-Estuarine and Coastal Waters

Location (Date of Collection)	Salinity, ‰	Particle Concentration, mg/L	Total Carbon, %	Total Silicon, ^a %	Total Aluminum, mg/g	Total Potassium, ^b mg/g	Total Iron, mg/g	Particle Activity, pCi/kg	Water Activity, pCi/L	Distribution, ^c Field K_d	Total Activity, pCi/L
<i>Conowingo Reservoir (Susquehanna River)</i>											
CONO (Oct. 3, 1983)	FW	6	10.6	19	80	20(18)	85	11,800 ± 600	0.06 ± 0.01	2.0 × 10 ⁵	0.13 ± 0.01
CONO (April 3, 1984)	FW	15	2.3	23	100	24(28)	55	8,700 ± 320	0.38 ± 0.01	2.3 × 10 ⁴	0.51 ± 0.01
<i>Upper Chesapeake Bay</i>											
SQFT (Oct. 3, 1983)	1	6	10.0	20	78	19(18)	64	11,700 ± 500	0.06 ± 0.01	2.0 × 10 ⁵	0.13 ± 0.01
ANAP (Oct. 4, 1983)	12	4	22.5	15	51	13(14)	58	4,600 ± 630	0.03 ± 0.01	1.5 × 10 ⁵	0.05 ± 0.01
CCFF (Oct. 5, 1983)	14	4	15.7	18	36	10(13)	39	4,500 ± 580	0.07 ± 0.01	6.4 × 10 ⁴	0.09 ± 0.01
SQFT (April 2, 1984)	FW	19	3.2	23	100	25(29)	57	13,700 ± 200	0.22 ± 0.01	6.5 × 10 ⁴	0.48 ± 0.01
ANAP (April 4, 1984) ^d	9	24	10.6	20	91	22(24)	47	12,800 ± 300	0.55 ± 0.01	2.3 × 10 ⁴	0.86 ± 0.01
CCFF (April 5, 1984)	8	11	33.7	8	27	8(9)	14	11,100 ± 300	0.34 ± 0.01	3.3 × 10 ⁴	0.46 ± 0.01
<i>James River Estuary</i>											
HOPE (June 19, 1981)	FW	31	4.3	23	96	17(19)	51	1,580 ± 265	0.10 ± 0.02	1.6 × 10 ⁴	0.15 ± 0.02
S409 (June 23, 1981)	7	22	3.5	17	81	17(16)	41	835 ± 365	0.11 ± 0.04	7.6 × 10 ⁵	0.13 ± 0.04
HAMP (June 23, 1981)	22	10	1.7(15)	...	1,680 ± 705	0.13 ± 0.04	1.3 × 10 ⁴	0.15 ± 0.04
<i>Hudson-Raritan Estuary</i>											
RARR (July 24, 1981)	FW	3(10)	...	8,040 ± 1120	0.05 ± 0.01	1.6 × 10 ⁵	0.07 ± 0.01
SRIV (July 24, 1981)	FW	29	8.6(19)	...	6,760 ± 610	0.09 ± 0.01	7.5 × 10 ⁴	0.29 ± 0.01
LENT (July 26, 1981)	4	13	5.6(22)	...	1,300 ± 330	0.18 ± 0.01	7.2 × 10 ³	0.20 ± 0.01
HARB (July 26, 1981)	24	8	7.3	16	56	17(20)	39	2,760 ± 290	0.21 ± 0.01	1.3 × 10 ⁴	0.23 ± 0.01
KILL (July 23, 1981)	26	12	3.8(15)	...	2,900 ± 330	0.06 ± 0.01	4.8 × 10 ⁴	0.09 ± 0.01
RARB (July 22, 1981)	28	7	8.3	14	50	17(17)	37	11,300 ± 360	0.29 ± 0.01	3.9 × 10 ⁴	0.37 ± 0.01
<i>Coastal Shelf</i>											
HOOK (July 22, 1981)	30	4	9.4(10)	...	10,800 ± 530	0.16 ± 0.01	6.8 × 10 ⁴	0.20 ± 0.01
AMBR (July 23, 1981)	25	6	4.0	7	17	12(11)	12	3,350 ± 360	0.11 ± 0.01	3.0 × 10 ⁴	0.13 ± 0.01

FW is fresh water.

^aLiBO₂ fused before ICP analysis to obtain total silicon. A comparison with standards indicates that the reported analyses may be between 5 and 7% low.

^bN numbers in parentheses were determined from the total ⁴⁰K activity in the sample, and are listed for comparison with the concentrations determined by flame photometry.

^cField K_d is (particle activity per kilogram)/(water activity per liter); 1 pCi = 0.037 Bq.

^dSample collected during a spring rain.

TABLE 2. ⁷Be Activities in River-Estuarine and Coastal Sediments

Sediment Sample (Collection Date)	Depth, cm	Dry Density, g/cm ³	⁷ Be Activity, pCi/g	⁷ Be Inventory, pCi/cm ²	Approximate Sedimentation Rate, ^a cm/yr
<i>James River Estuary</i>					
T-233 (HOPE) ^b (June 19, 1981)	0-1	0.94	0.20 ± 0.07	0.2	< 1
	1-2	1.00	ND ^c		
T-14 (June 19, 1981)	0-1	0.36	0.48 ± 0.27	0.2	1-3
	1-2	0.37	ND		
T-13 (June 19, 1981)	0-1	0.40	0.71 ± 0.26	0.3	1-3
	1-2	0.44	ND		
T-12 (June 18, 1981)	0-1	0.32	1.14 ± 0.21	2.6	> 3 (dredged)
	1-2	0.37	1.14 ± 0.25		
	2-4	0.39	1.06 ± 0.33		
	4-6	0.42	0.67 ± 0.17		
	6-8	0.43	0.44 ± 0.18		
	8-10	0.46	ND		
T-232 (June 18, 1981)	0-1	0.66	0.91 ± 0.24	1.6	> 3
	1-2	0.77	0.43 ± 0.11		
	2-4	1.07	0.29 ± 0.14		
	4-6	0.94	ND		
T-10 (June 18, 1981)	0-1	0.39	0.50 ± 0.28	1.2	1-3
	1-2	0.46	0.38 ± 0.17		
	2-4	0.46	0.87 ± 0.18		
	4-6	0.52	ND		
T-9 (S409) ^b (June 18, 1981)	0-1	0.38	0.74 ± 0.16	1.6	1-3
	1-2	0.45	1.08 ± 0.15		
	2-4	0.54	0.74 ± 0.16		
	4-6	0.57	ND		
B-6 (Aug. 12, 1982)	0-1	0.37	0.86 ± 0.11	0.3	1-3
	1-2	0.41	ND		
Wreck shoals (Aug. 12, 1982)	0-2	0.47	0.29 ± 0.07	0.3	< 1
	2-4	0.53	ND		
JR mudflat (Aug. 12, 1982)	0-1	0.35	1.27 ± 0.22	0.7	1-3
	1-2	0.35	0.42 ± 0.15		
	2-4	0.33	0.11 ± 0.10		
	4-6	0.57	ND		
JR marsh (Aug. 12, 1982)	grass	0.09	1.32 ± 0.13	1.8	< 1 ^d
	0-1	0.11	1.65 ± 0.28		
	1-2	0.16	1.28 ± 0.22		
	2-4	0.23	0.58 ± 0.37		
	4-6	0.30	0.24 ± 0.10		
	6-10	0.37	ND		
T-8 (June 17, 1981)	0-1	0.42	1.03 ± 0.31	0.6	1-3
	1-2	0.50	0.23 ± 0.13		
	2-4	0.51	ND		
T-7 (June 17, 1981)	0-1	0.46	0.52 ± 0.15	0.3	< 1
	1-2	0.55	ND		
T-6 (June 17, 1981)	0-2A ^e	0.56	0.41 ± 0.14	0.5	< 1
	0-2B ^e	0.58	0.41 ± 0.13		
	2-4	0.60	ND		
T-102 (HAMP) ^b (June 16, 1981)	0-2	0.42	1.54 ± 0.10	3.3	> 3 (dredged)
	2-3	0.54	2.00 ± 0.29		
	3-4	0.53	1.28 ± 0.39		
	4-5	0.47	0.45 ± 0.27		
	5-6	0.47	ND		
<i>Chesapeake Bay</i>					
Rapp-1 (June 25, 1981)	0-1	0.45	3.29 ± 0.52	2.9	> 3
	1-2	0.51	2.39 ± 0.34		
	2-3	0.59	0.41 ± 0.28		
	3-4	0.59	ND		
SQFT (Sept. 28, 1981)	0-2	0.60	0.42 ± 0.09	0.5	< 1
	2-4	0.81	ND		
CONO (Sept. 28, 1981)	0-2	0.30	0.71 ± 0.17	0.6	> 3
	2-4	0.39	0.23 ± 0.09		
	4-6	0.44	ND		

TABLE 2. (continued)

Sediment Sample (Collection Date)	Depth, cm	Dry Density, g/cm ³	⁷ Be Activity, pCi/g	⁷ Be Inventory, pCi/cm ²	Approximate Sedimentation Rate, ^a cm/yr
<i>Hudson-Raritan Estuary</i>					
MP2.6E (HARB) ^b (July 26, 1981)	0-2	0.26	1.90 ± 0.40	2.3	> 3
	2-4	0.34	0.72 ± 0.37		
	4-6	0.41	0.95 ± 0.33		
	6-8	0.40	ND		
MP1.8E (July 26, 1981)	0-2	0.21	2.06 ± 0.34	1.7	> 3
	2-4	0.30	1.44 ± 0.44		
	4-6	0.32	ND		
South River (SRIV) ^b (July 24, 1981)	0-1	0.76	1.20 ± 0.11	0.9	1-3
	1-2	0.70	ND		
Raritan River (RARR) ^b (July 24, 1981)	0-1	0.98	0.53 ± 0.10	0.5	< 1
	1-2	0.92	ND		
Raritan Bay (RARB) ^b (July 22, 1981)	0-1	0.61	ND	0.0	< 1
<i>Coastal Shelf</i>					
Mud Pit (AMBR) ^b (July 23, 1981)	0-2	0.36	0.59 ± 0.18	0.4	1-3
	2-4	0.77	ND		
Sandy Point (HOOK) ^b	0-1	1.08	ND	0.0	< 1

1 pCi = 0.037 Bq.

^aThe approximated sediment accumulation rates have been estimated from the measured vertical distributions of fallout and reactor-released radionuclides in the cores. These data have been presented elsewhere [Olsen et al., 1984; Zucker et al., 1984].

^bSynergistic suspended-matter samples (Table 1), which have been plotted with the sediment core data in Figure 3.

^cNot detectable: < 0.1 pCi/g.

^dThe vertical distribution and inventory of ⁷Be in this core are discussed by Olsen et al. [1985].

^eDuplicate samples collected from the surface of the same box core.

low. Field particle-to-water distribution coefficients for ⁷Be range from 7×10^3 to 2×10^5 and have a median value of about 4×10^4 (Table 1), indicating that ⁷Be is particle reactive and should, thus, be rapidly scavenged by particulate matter in turbid coastal waters. As will be discussed later, these field distribution coefficients should not be considered to represent simple equilibrium distributions because chemical equilibrium may not have prevailed at the time of sample collection.

Beryllium 7 concentrations, vertical distributions, and inventories in sediment cores collected throughout the salinity range of the James River estuary and in other estuarine and coastal sites (Figure 1) are presented in Table 2. The extent of ⁷Be accumulation in sediments is extremely variable, and inventories range from nondetectable (< 0.1 pCi/cm²) to ~3.3 pCi/cm². The highest ⁷Be inventories occur in areas that have been previously identified as sites of rapid fine-particle accumulation (> 3 cm/yr) and radionuclide sinks, such as in the inner harbor area of New York City [Simpson et al., 1976; Olsen et al., 1981a], in estuarine turbidity zones [Nichols, 1972; Cutshall et al., 1981], behind constructed barriers [Olsen et al., 1981b], and in dredged areas [Olsen et al., 1984; Zucker et al., 1984]. In addition, by comparing the sediment concentration and inventory data for ⁷Be in different estuarine and coastal areas (Table 2) with the ⁷Be concentration data on suspended particles, collected at approximately the same time and location (Tables 1 and 2), it is evident that ⁷Be concentrations on suspended matter are lowest (and almost identical to surface sediment concentrations) in areas where ⁷Be

sediment inventories are highest, that is, in areas where fine particles are accumulating most rapidly. This relationship between the concentration of ⁷Be on suspended particles and the accumulation of ⁷Be in bottom sediments is illustrated in Figure 3.

4. DISCUSSION

4.1. Geochemistry in River-Estuarine and Coastal Waters

Although ⁷Be is particle reactive, most of the ⁷Be in the water column is in the water phase rather than on particulates because of the low suspended-particle concentrations in northeastern rivers, estuaries, and coastal waters. Water-phase ⁷Be activities range from 0.03 to 0.55 pCi/L, appear to primarily reflect variations in ⁷Be supply, and generally account for 60 to 90% of the total ⁷Be concentration in the water column (Table 1). Previous work by Measures and Edmond [1983] has shown that the solubility of stable ⁹Be in river water is strongly dependent on pH, with dissolved ⁹Be concentrations increasing significantly at pH values below 6. Consequently, one might expect the dissolved concentrations of ⁷Be to be slightly higher in fresh water (pH about 6) relative to seawater (pH about 8). If such a trend exists in the water-phase ⁷Be concentrations listed in Table 1, it is not resolvable because of the large variations in ⁷Be supply. In fact, "dissolved" ⁷Be concentrations tended to be lower (rather than higher) in freshwater areas, attesting to the ability of drainage basin veg-

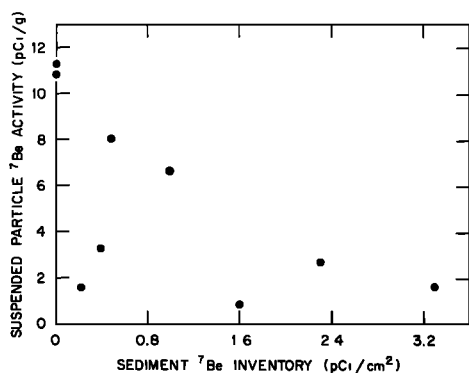


Fig. 3. Plot of the concentration of ^7Be on suspended particles as a function of its total accumulation in the sediments. Although each point represents a different river-estuarine and coastal area, only synergistic suspended-matter and sediment samples (i.e., samples collected at approximately the same place and at the same time) have been plotted and identified in Table 2. This figure helps illustrate that the primary factor governing the concentration of ^7Be (and by analogy other particle-reactive substances) on suspended particles is the length of time that particles remain in the water column acting as scavengers.

etation and soils to sequester precipitation-derived ^7Be and its subsequent runoff. Drainage basin trapping will also be evident in the next section where we show that the river input of ^7Be to the James estuary is less than 5% of the atmospheric input to the estuary surface.

The effects of atmospheric input on dissolved ^7Be concentrations are also evident from a comparison of the October and April data for the Susquehanna-Chesapeake Bay system (Table 1). The average dissolved ^7Be concentration in the four water samples collected during October was 0.06 pCi/L; the average dissolved concentration at the same four sites in April, however, was 0.37 pCi/L. The highest "dissolved" ^7Be concentration was measured in a water sample collected about 2 m below the surface in Chesapeake Bay (ANAP in Figure 1 and Table 1) during a spring rainstorm. Within a day after this storm, the dissolved ^7Be concentration decreased by about a factor of 2 in a similar sample collected at nearby station CCFF (Figure 1; Table 1), approaching dissolved values measured in the system prior to the rainstorm. These results are probably best explained by ^7Be sorption kinetics. Bloom and Crecelius [1983] have shown that about 50 hours were required for ^7Be to reach its equilibrium particle-to-water distribution at suspended-matter concentrations between 20 and 100 mg/L.

Beryllium 7 concentrations on suspended-particulate matter range over an order of magnitude from 0.8 to 13.7 pCi/g (Table 1). From the data presented in Table 1, there do not appear to be any significant correlations between suspended-particulate ^7Be activities and salinity, particle concentration (between 5 and 30 mg/L), or particle composition (total carbon, silicon, aluminum, potassium, and iron). This lack of correlation with particulate chemistry or type suggests that the tendency for ^7Be to become associated with specific particle types, such as organics or clays, is overwhelmed by bulk sorption (perhaps in association with Fe-Mn phases) onto the relatively large amount of particles supplied by sediment resuspension in turbid river-estuarine and coastal waters. Consequently, it appears that one of the major processes controlling the concentration of atmospherically derived ^7Be on

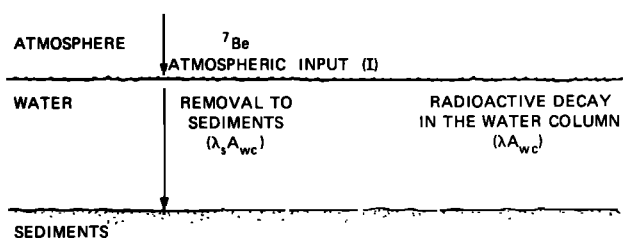
suspended particles in these coastal systems is the length of time that particles remain in the water column and are able to scavenge ^7Be rather than the variations in water salinity or particle type and concentration.

This conclusion is supported by the data illustrated in Figure 3 which shows the relationship between the concentration of ^7Be on suspended particles and the amount of ^7Be accumulation in sediments. Concentrations of ^7Be on suspended particles are highest in high-energy areas where sediment inventories are lowest (Figure 3) and where fine particles are continually being resuspended by wave and current action, such as in wide shallow areas (RARB, SQFT) and sandy sediment areas (HOOK, CCFF) (Figure 1 and Table 1). Previous studies have shown that these high-energy areas are characterized by little or no net accumulation of fine particles and when low ^7Be sediment inventories occur in these areas they probably result from sediment-mixing processes [Olsen *et al.*, 1984]. The lowest concentrations of ^7Be on suspended matter occur in areas where fine particles are accumulating at rates greater than 3 cm/yr (Table 2), such as in estuarine zones near the landward limit of saltwater flow (S409), and dredged harbor areas (HAMP, HARB, AMBR), that is, where sediment ^7Be inventories are greatest (Figure 3).

Anomalies to this general inverse relationship between the concentration of ^7Be on suspended particles and the inventory of ^7Be in sediments (Figure 3) certainly occur, and appear most often in freshwater areas. For example, both the inventory and suspended-particulate concentration of ^7Be are relatively low in the freshwater reach of the James estuary near Hopewell (HOPE). Although there are several possible explanations for this anomaly, including (1) the occurrence of unidentified sinks for ^7Be in unsampled areas, or (2) a depletion of the ^7Be concentration on particles in response to the pH-solubility effect discussed earlier, or as a result of a dilution of the suspended matter with older (^7Be -depleted) particles derived from erosion in upstream areas, the paucity of data makes any specific explanation speculative. The important point, however, is that the general trend in Figure 3 implies that the major process governing the concentration of ^7Be on suspended matter is the length of time that particles remain in the water column.

Field particle-to-water distribution coefficients for ^7Be range from 7×10^3 to 2×10^5 and are not significantly correlated with variations in salinity, or suspended-matter concentrations between 5 and 30 mg/L (Table 1). Although the concentration of ^7Be on suspended particles does not appear to be correlated with particulate type or iron content, the particle-to-water distribution coefficients for ^7Be do appear to be directly correlated with the amount of iron on the particulate samples (Table 1) and inversely correlated with the total aluminum content after the correlation with iron is accounted for. This implies that the precipitation of dissolved iron may play an important role in ^7Be sorption and removal from the water phase in these river-estuarine and coastal systems. This implication is consistent with unpublished laboratory results which indicate that ^7Be is associated with Fe-Mn phases on particulate samples (C. R. Olsen, unpublished data, 1985). Although ^7Be sorption may be associated with the precipitation or flocculation of dissolved Fe and Mn, the lack of correlation between the total iron and the total ^7Be concentration on our suspended particulate samples (Table 1) supports our earlier conclusion that the major factor affecting the total concentration of ^7Be on suspended matter is the length of time particles remain in the water column. The inverse correlation

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



ASSUME:

- (1) STEADY-STATE FLUX CONDITIONS RANGING FROM 0.013 pCi cm⁻²d⁻¹ TO 0.026 pCi cm⁻²d⁻¹
- (2) NO EXCHANGE OF WATER WITH SIGNIFICANTLY DIFFERENT ⁷Be CONCENTRATIONS OVER THE 77 DAY MEAN-LIFE (1/λ) OF ⁷Be

THEN:

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

OR:

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

AND:

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

Fig. 4. Box model used to calculate the ranges in ⁷Be removal rates and residence times listed in Table 3. *I* is steady state atmospheric flux of ⁷Be (pCi cm⁻² d⁻¹) calculated from measured monthly fluxes; *A_{wc}* is average water-column ⁷Be inventory (pCi/cm⁻²) calculated from total water-column ⁷Be concentrations and mean water depths; *λ* is radioactive decay constant for ⁷Be (day⁻¹); *λ_s* is first-order removal rate constant (day⁻¹) from the water column; and *τ* is residence time of ⁷Be in the water column.

with aluminum (after the iron correlation is factored out) suggests that the particle-to-water distribution coefficient can be diluted by eroded (⁷Be depleted) inorganic particles but that this dilution is not manifested at suspended-matter concentrations between 5 and 30 mg/L. The relatively high particle-to-water distribution implies that ⁷Be should be rapidly scavenged by suspended particulate matter and removed from the water column by settling particles.

4.2. Water Column Removal Rates and Residence Times

The rate of ⁷Be removal from the water column via deposition with settling particles, *λ_s*, can be estimated by integrating the ⁷Be standing crop throughout the water column, *A_{wc}*, and relating this value to its input function *I* and its rate of radioactive decay *λ*, using the simple box model assumptions and equations illustrated in Figure 4. Beryllium 7 removal rates and residence times calculated from this box model are listed in Table 3. The total inventory of ⁷Be delivered to the surface of Conowingo Reservoir on the Lower Susquehanna River and to the surface of Upper Chesapeake Bay was 1.10 pCi/cm² at the beginning of October 1983 and 1.66 pCi/cm² at the beginning of April 1984 when our respective water samples were collected (Figure 2). Assuming that the mean depth of Conowingo Reservoir is 9 m [McLean et al., 1983] and assuming a total water column ⁷Be concentration of 0.13 pCi/L during October and 0.51 pCi/L in April 1984 (Table 1), then the total amount of ⁷Be in the water column is 0.12 pCi/cm² (or about 10% of the total input) in October and 0.46 pCi/cm² (or about 25% of the total input) in April. Using the box model in Figure 4, the calculated length of time that atmospherically derived ⁷Be remains in the reservoir water is 9 days in October and 25 days in April (Table 3). Similar

calculations can be made for Upper Chesapeake Bay (upstream of sample CCFF, Figure 1) assuming a mean depth of 8 m [Cronin, 1971], and an average ⁷Be water column concentration of 0.09 pCi/L (for samples SQFT, ANAP, and CCFF) in October 1983 and 0.60 pCi/L in April 1984 (Table 1). Input ranges, based on the lowest and highest 1982–1984 ⁷Be inventories measured at Norfolk, Virginia [Olsen et al., 1985], were used to calculate the range in ⁷Be removal rates and residence times for the James estuary, Hudson estuary, Raritan Bay and coastal shelf waters in Table 3, because water samples from these areas were collected prior to our atmospheric input measurements.

By analogy, the range in ⁷Be removal rates and residence times, listed in Table 3, should be characteristic of other particle-reactive substances, such as Kepone in the James, PCBs in the Hudson, and various metals or other radionuclides in Chesapeake Bay. Note, however, that these removal rates and residence times (Table 3) assume steady state fluxes and are merely “snapshots” providing a picture of removal over a relatively short time period (during the 77-day mean lifetime of ⁷Be) prior to sample collection. In addition, these calculated ranges reflect the length of time that ⁷Be remains in the water column and not the length of time that ⁷Be remains in solution. Short-term processes such as sediment resuspension (associated with high seasonal flows, storms, or human perturbations such as dredging) can reintroduce deposited ⁷Be back into riverine-estuarine and coastal waters and, thus, prolong the length of time ultimately required to completely remove ⁷Be from the water column. Sediment resuspension in association with high seasonal flow is one possible explanation for the large increase in suspended-matter concentrations and in the calculated residence times

TABLE 3. ⁷Be Removal Rates and Residence Times in the Water Column

Sample Date	Input Inventory, ^a pCi/cm ²	Water-Column Inventory, ^b pCi/cm ²	Removal Rate, day ⁻¹	Residence Time, days
<i>Conowingo Reservoir</i>				
(Oct. 1983)	1.1	0.12	0.11	9
(April 1984)	1.7	0.46	0.04	25
<i>Upper Chesapeake Bay</i>				
(Oct. 1983)	1.1	0.07	0.19	5
(April 1984)	1.7	0.48	0.03	33
<i>James River Estuary</i>				
(June 1981)	1.0–2.0	0.05	0.51–0.25	2–4
<i>Hudson River Estuary</i>				
(July 1981)	1.0–2.0	0.19	0.12–0.06	8–17
<i>Raritan Bay</i>				
(July 1981)	1.0–2.0	0.17	0.14–0.06	7–17
<i>Coastal Shelf</i>				
(July 1981)	1.0–2.0	0.24	0.10–0.04	10–25

^aInput inventories were determined from measured atmospheric fluxes of ⁷Be in monthly deposition samples collected from October 1982 to October 1984 [Olsen et al., 1985]. An input range, spanning the lowest and highest measured ⁷Be inventories, was used for samples collected prior to actual input measurements. The steady state atmospheric flux of ⁷Be (pCi cm⁻² d⁻¹) was assumed to equal the measured inventory divided by the mean life of ⁷Be (77 days).

^bThe average water-column inventories were calculated using the water-column ⁷Be concentrations in Table 1 and the mean depths of each system.

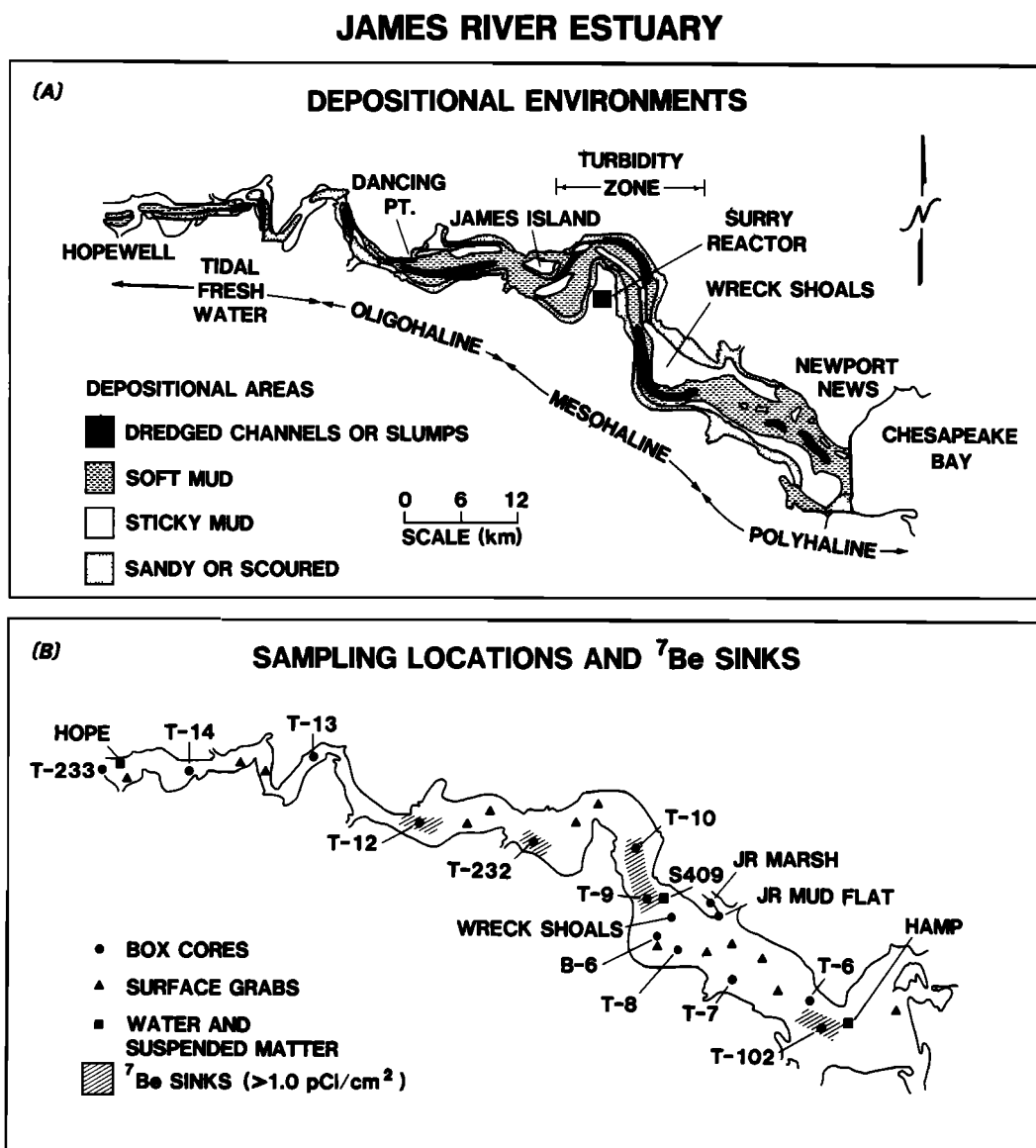


Fig. 5. (a) Map of the James River estuary illustrating the various deposition environments used in determining the total burden of ^7Be in the estuarine sediments. (b) Location of sediment sampling sites in the James River estuary. The shaded zones represent areas where the ^7Be inventory exceeds 1.0 pCi/cm^2 . These zones primarily occur in dredged-channel (slump) areas and in the turbidity zone of the estuary and are areas of rapid fine-particle, Kepone, and reactor-released radionuclide accumulation, as determined from previous studies [Nichols, 1972, 1979; Huggett *et al.*, 1980; Huggett and Bender, 1980; Cutshall *et al.*, 1981; Nichols and Cutshall, 1981; Zucker *et al.*, 1984].

for ^7Be in Conowingo Reservoir and Upper Chesapeake Bay during April relative to October (Table 3).

In addition to sediment resuspension, the removal rates calculated from the box model in Figure 4 neglect exchange effects with surrounding waters. Consequently, ^7Be removal by particle scavenging and settling is not valid in shallow or narrow water bodies, such as the Raritan River (RARR), South River (SRIV), or Arthur Kill (KILL), where strong currents and water exchange dominate. In the James, Hudson, Raritan, and Chesapeake systems, where surface areas are large and flushing times are slow relative to the mean life of ^7Be , such exchange is insignificant in relation to the atmospheric input of ^7Be (see discussion concerning ^7Be budget for the James). In addition, incoming waters in these systems have nearly the same water-phase ^7Be concentration as waters at the estuary mouth (Table 1), so the calculated range in remov-

al rates actually reflects the length of time ^7Be remains in the water column rather than reflects the physical flushing time of ^7Be from one water body to another.

Much more detailed work involving (1) sampling at different water depths and during different flow and weather conditions, (2) determining ^7Be removal rates immediately after precipitation events, (3) integrating these results with physical circulation models, and (4) applying two- or three-dimensional, nonsteady state models (similar to those used for ^{234}Th and ^{210}Pb removal by Tanaka *et al.* [1983]), will be required to estimate ^7Be and, by analogy, contaminant removal rates with more certainty. The ranges in removal rates and residence times presented in Table 3, however, are consistent with those determined by other investigators using thorium isotopes in the saline waters of the New York Bight [Li *et al.*, 1979; Kaufman *et al.*, 1981] and Narragansett Bay [Santschi *et*

TABLE 4. ^7Be Inventory for the James River Estuary

	Volumes and Areas	^7Be Budget, curies ^a
Input		
Atmospheric deposition ^b	(475 km ²)	4.8 to 9.6
Freshwater runoff		0.2
Total input		5 to 10
Water column inventory		
Water phase	(1.7×10^{12} L)	0.20
Suspended-particulate phase	(20 mg/L)	0.05
Total water column		0.25
Sediment inventory^c		
Dredged-channel areas	(25 km ²)	0.8 ± 0.1
Soft-mud accumulation areas	(210 km ²)	3.2 ± 2.1
Sticky-mud low accumulation areas	(160 km ²)	0.5 ± 0.2
Sandy areas	(80 km ²)	ND ^d
Total sediments		4.5 ± 2.1

^a1 Ci = 3.7×10^{10} Bq.

^bThe range is calculated using the lowest ($0.013 \text{ pCi cm}^{-2} \text{ d}^{-1}$) and the highest ($0.026 \text{ pCi cm}^{-2} \text{ d}^{-1}$) average daily fluxes required to support the lowest and highest ^7Be inventories for Norfolk, Virginia [Olsen et al., 1985].

^cThe average ^7Be inventory (± 1 standard deviation from the mean) is $3.0 \pm 0.5 \text{ pCi/cm}^2$ in dredged-slump areas; $1.5 \pm 1.0 \text{ pCi cm}^{-2}$ in soft mud areas; $0.3 \pm 0.1 \text{ pCi/cm}^2$ in sticky mud areas; and nondetectable in sandy or scoured areas.

^dNot detectable.

al., 1979, 1980]. In addition to quantifying first-order transfer rates, the data in Table 3 provide insights concerning both the types and the spatial and temporal heterogeneity of the various environmental processes that affect contaminant removal in river-estuarine and coastal waters.

4.3. Accumulation Patterns and Inventories in the James River Estuary

The James River estuary is a partially mixed tributary to Chesapeake Bay (boxed area in Figure 1). Its water volume from Hopewell to Newport News (Figure 5) is $\sim 1.7 \times 10^{12}$ L at mean low water (MLW), its average depth over the same span is ~ 3.5 m, and its MLW surface area is $\sim 4.8 \times 10^8 \text{ m}^2$ [Cronin, 1971]. Freshwater inflow averages $211 \text{ m}^3/\text{s}$ at Richmond, and the normal upstream limit of salt intrusion is near James Island where a pronounced turbidity maximum also occurs (Figure 5). Approximately 1.7×10^9 kg of suspended sediment is annually supplied to the estuary, primarily in short pulses during the spring [Huggett et al., 1980].

Sediment accumulation in the James River estuary is extremely heterogeneous. Zucker et al. [1984] have measured the vertical distribution of fallout and reactor-released radionuclides in 29 box cores collected throughout the estuary (including the 12 box cores located in Figure 5) and shown that greatest accumulation occurred in dredged-channel or slump areas (such as near Dancing Point and Newport News) and in the turbidity zone of the estuary (Figure 5). Although dredged-channel areas represented less than 5% of the total estuarine surface area, they accounted for more than 40% of the total sediment burden of reactor-released ^{134}Cs and ^{60}Co in the James estuary [Zucker et al., 1984]. Areas of rapid sediment accumulation are also major sinks for particle-associated contaminants, such as Kepone, released near Hopewell [Cutshall et al., 1981]. Since particle dynamics and patterns of sediment and particle-associated contaminant accumulation have been

extensively studied in the James [Nichols, 1972, 1979; Huggett et al., 1980; Huggett and Bender, 1980; Cutshall et al., 1981; Nichols and Cutshall, 1981; Zucker et al., 1984], this estuary is an ideal site for obtaining information concerning the geochemistry of ^7Be , its inventory, and its potential use as a tracer for particle-reactive contaminants.

The ^7Be inventory data for the James River estuary (Table 4) indicate that the highest values occur in the same dredged-channel areas previously identified as fine-particle and radionuclide sinks [Zucker et al., 1984]. Although the heterogeneity of trace-substance accumulation in coastal areas has been attributed to variations in the reactivity of different particle types, to the extent of surface sediment mixing, and to the net rate and pattern of fine-particle accumulation [Turekian, 1977; Aller et al., 1980; Benninger and Krishnaswami, 1981; Bopp et al., 1981; Bothner et al., 1981; Olsen et al., 1982b], our results indicate that fine-particle deposition is the most important factor affecting the accumulation pattern and vertical distribution of ^7Be in this estuarine system.

Using a river-estuary surface area of $4.75 \times 10^{12} \text{ cm}^2$ and assuming that the atmospheric flux of ^7Be ranges from 0.013 to $0.026 \text{ pCi cm}^{-2} \text{ d}^{-1}$ at Norfolk, Virginia, near the estuary mouth [Olsen et al., 1985], the calculated steady state standing crop of ^7Be in this river-estuarine system would range from 5 to 10 Ci. The total freshwater flow, as gaged at Richmond, during the 77-day period (or mean life of ^7Be) prior to the date when most of our box cores were collected, was 9.3×10^{11} L. Additional freshwater input from the Appomattox and Chickahominy Rivers would raise this integrated flow by less than 20% to $\sim 1.2 \times 10^{12}$ L. This 77-day, mean-life integrated flow from all tributaries would replace only $\sim 50\%$ of the river-estuary water volume. Calculations using the water-column concentration data at Hopewell (HOPE in Table 1) and the above integrated flow indicate that the water phase and particulate ^7Be input to the estuary from river runoff is less than 5% (or ~ 0.15 Ci) of the ^7Be standing crop from atmospheric deposition, which attests to the ability of drainage basin vegetation and soils to retain ^7Be . Although exchange with Chesapeake Bay may also be a source or a sink for ^7Be , the low concentration of ^7Be in the water phase (HAMP in Table 1), the short (2- to 4-day) residence time of ^7Be in the water column (Table 3), and the slow flushing time of this river-estuarine system all make it unlikely that such exchange will be significant in light of the atmospheric flux.

The ^7Be inventory in the water column (Table 4) was calculated assuming that (1) the average concentration of ^7Be in the water phase is 0.12 pCi/L (Table 1), (2) the average concentration of ^7Be on suspended particles is 1.6 pCi/g (Table 1), and (3) the average suspended matter concentration over the entire estuary is 20 mg/L . It is apparent from the data in Table 4 that during June 1981 less than 5% of the ^7Be standing crop is in the water column. This is consistent with earlier work [Aaboe et al., 1981] which showed that virtually no ^7Be exists in the water column of Long Island Sound.

To determine the total burden of ^7Be in the sediments, the James River estuary was subdivided into several depositional environments (Figure 5), using National Oceanographic and Atmospheric Administration (NOAA) charts, sedimentation maps [Nichols, 1972], the observed sedimentary characteristics from core and grab samples, and rates of accumulation based on previously reported fallout and reactor-released radionuclide profiles [Zucker et al., 1984]:

1. Dredged-channel or slump areas are characterized by channel dredging that has caused unusually high rates (> 3

cm/yr) of fine-particle accumulation. These areas comprise less than 5% of the total estuarine surface area in Figure 5.

2. Soft mud areas have surface sediments highly liquefied and rates of accumulation that are on the order of 1 to 3 cm/yr. These areas comprise about 45% of the total estuarine surface area.

3. Sticky mud areas have a sediment surface characterized by cohesive muds or oyster shell pavements. Fallout and reactor-released radionuclides are confined to the top few centimeters of sediment, and their vertical distribution is primarily a result of sediment mixing by currents and organisms. These areas comprise about 35% of the total estuarine surface area in Figure 5.

4. Sandy and scoured areas have particles greater than 62 μm in diameter comprising more than 75% of the total sediment weight or where river flow and tidal currents have formed a natural erosional feature, sometimes in cohesive sticky muds. These areas comprise about 15% of the total estuarine surface area and were ignored in our ^7Be inventory calculations because no appreciable ^7Be activities were detected in sandy and scoured areas of this estuarine system.

The sediment ^7Be inventory (Table 4) was calculated by averaging the ^7Be standing crop in box cores collected within each depositional zone (Figure 5) and assuming that this average (± 1 standard deviation of the mean) was representative for the entire zone. Although the surface grab samples, located in Figure 5, were not used in the budget calculations, the ^7Be concentrations measured in these samples were used to help guide extrapolation of sediment ^7Be inventories over areas of the estuary where cores were not taken. In addition, intertidal and marsh areas were ignored in our budget calculations because the emphasis of this work has been placed on the depositional behavior of ^7Be in river-estuarine and coastal waters, and these areas serve as effective atmospheric traps [Olsen et al., 1985].

All of the inventory estimates listed in Table 4 are, of course, approximate, but we believe they are accurate within a factor of 2. Since atmospheric fluxes of ^7Be were not measured prior to our 1981 sediment sampling cruise in the James estuary, we can only estimate the atmospheric flux of ^7Be into this coastal system (and, thus, the extent to which this flux is trapped within the estuarine sediments) by indirect evidence: using atmospheric fluxes measured during 1981 along the same 34° – 36° latitude band as the James estuary by other investigators at Fayetteville, Arkansas, and Sakai, Japan, or using the range in average atmospheric fluxes required to support the lowest and highest ^7Be inventories measured at Norfolk, Virginia (at the mouth of the James), during 1982 to 1984 [Olsen et al., 1985]. The 1981 average daily flux of ^7Be measured at Fayetteville, Arkansas, during the 3 months prior to our June 1981 sampling was $0.011 \text{ pCi cm}^{-2} \text{ d}^{-1}$ [Saleh and Kuroda, 1982]. The 1981 average daily flux of ^7Be measured at Sakai, Japan, during the 3 months prior to our June 1981 sampling was $0.018 \text{ pCi cm}^{-2} \text{ d}^{-1}$ (T. Matsunami, Radiation Center of Osaka Prefecture, unpublished data, 1983). These average fluxes would support ^7Be inventories of about 0.85 and 1.39 pCi cm^{-2} , respectively. When extrapolated over the entire surface area of the James estuary, these 1981 fluxes would support an estuarine ^7Be standing crop of 4 to 7 Ci, respectively.

Although the average ^7Be fluxes measured during the spring of 1981 in Arkansas and Japan are similar to the 2-year average daily flux ($0.016 \text{ pCi cm}^{-2} \text{ d}^{-1}$) calculated from the total depositional data at Norfolk (Figure 2), they are near the

low end of the range (0.013 to $0.026 \text{ pCi cm}^{-2} \text{ d}^{-1}$) required to support the 1983–1984 ^7Be inventories at the Norfolk site. This might be expected since the total amount of precipitation at Norfolk during the 4 months prior to our 1981 sediment sampling trip was less than half the amount of precipitation during the same 4 months in either 1983 or 1984. In addition, the annual amount of precipitation recorded at Norfolk during 1981 was about 100 cm which is considerably lower than the 2-year annual average of 139 cm recorded for 1982–1984 (Figure 2).

By assuming that the flux of ^7Be to the James River estuary was $0.013 \text{ pCi cm}^{-2} \text{ d}^{-1}$ during the mean-life period prior to the collection of our box cores in June 1981, then about 5 Ci of ^7Be would have been input to the estuary surface (Table 4). Such an input would be consistent with the average ^7Be fluxes actually measured during 1981 in Arkansas and Japan. Such an input would also be in good agreement with the total ^7Be standing crop measured and calculated to reside in the water column and sediments of the James River estuary (Table 4), indicating that this estuary is an effective trap for ^7Be and, by analogy, for any particle-reactive pollutant that may be introduced into the estuarine waters. If, however, we assume that the atmospheric flux of ^7Be to the James River estuary was $0.026 \text{ pCi cm}^{-2} \text{ d}^{-1}$ during the mean-life period prior to box core collection, then about 10 Ci of ^7Be would have been input to the estuary surface (Table 4). Such an input would be consistent with the flux required to support the highest ^7Be inventories measured during the springs of 1983 and 1984 [Olsen et al., 1985] and would indicate that the estuary traps about one half of the ^7Be input. The actual short-term trapping efficiency of the James estuary for particle-reactive substances having particle-to-water distributions between 10^4 and 10^5 probably falls somewhere between 50 and 100%.

5. CONCLUSIONS

The atmospheric flux of ^7Be and the mode of ^7Be deposition in estuarine environments have been examined to determine, understand, and predict the types and rates of processes that affect the fate of ^7Be and, by analogy, other particle-seeking substances in estuarine and coastal areas. The information presented in this study suggests the following conclusions.

1. The mid-latitude atmospheric flux of ^7Be commonly supports inventories ranging from 1.0 to 2.0 pCi/cm^2 , with the highest inventories generally occurring in the spring and the lowest in the fall. The average daily fluxes required to support these inventories range from 0.013 to $0.026 \text{ pCi cm}^{-2} \text{ d}^{-1}$.

2. Water-phase ^7Be concentrations range from 0.03 to 0.53 pCi/L and appear to primarily reflect variations in ^7Be supply and sorption kinetics.

3. The major process controlling the concentration of ^7Be on suspended matter in river-estuarine and coastal waters appears to be the length of time that particles remain in the water column and able to scavenge ^7Be .

4. Field particle-to-water distribution coefficients for ^7Be range from 7×10^3 to 2×10^5 and appear to reflect short-term variations in ^7Be input, sorption kinetics in association with Fe-Mn phases, and particle dynamics rather than equilibrium sorption-desorption processes in response to changes in water salinity or particle type.

5. There is an apparent correlation between the field determined particle-to-water distribution coefficient for ^7Be and the amount of iron on the particulate samples. We suggest that this correlation implies that the precipitation of dissolved iron

may play an important role in the removal of atmospherically derived ^7Be from estuarine and coastal waters.

6. The lack of correlation between the concentration of ^7Be on suspended matter and particulate chemistry suggests that bulk sorption (in association with Fe-Mn phases) and the availability of suspended particles (in turbid river-estuarine and coastal waters) overwhelm thermodynamic tendencies for ^7Be to become associated with specific particle types.

7. Residence times of ^7Be in the water column range from 1 to 3 days in areas where fine particles are deposited rapidly, such as in estuarine turbidity zones. Sediment resuspension, associated with high seasonal flows, storms, or human perturbations, can reintroduce deposited ^7Be back into the water column and, thus, prolong its residence time in estuarine waters to as much as 50 days.

8. Although the heterogeneity of trace-substance accumulation in estuarine and coastal areas has been attributed to variations in the reactivity of different particle types, the extent of surface sediment mixing, and the net rate and pattern of fine-particle accumulation, our results indicate that fine-particle deposition is the most important factor affecting the accumulation pattern of ^7Be .

9. The short-term trapping efficiency of the James estuary for ^7Be and other particle-reactive substances (having particle-to-water distributions between 10^4 and 10^5) probably falls somewhere between 50 and 100%.

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