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

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The atmospheric flux of cosmogenic $^7$Be (53.3-day half-life) and the mode of $^7$Be deposition in river-estuarine and coastal environments have been examined. The atmospheric flux of $^7$Be commonly supports inventories ranging from 1.0 to 2.0 pCi/cm$^2$ ($1$ 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 $^7$Be supply and sorption kinetics. The major process controlling the concentration of $^7$Be 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 $^7$Be have a median value of about $4 \times 10^4$ but range over an order of magnitude reflecting short-term variations in $^7$Be input, particle dynamics, and particulate iron content rather than equilibrium sorption-desorption responses to changes in water salinity or particle type. Residence times of $^7$Be 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 $^7$Be back into the water column. Inventories of $^7$Be in sediments range from nondetectable to 3.3 pCi/cm$^2$, with the highest inventories in areas where fine particles are accumulating rapidly. Such sites are also major repositories for other particle-reactive substances. A $^7$Be budget for the James estuary indicates that less than 5% of the expected $^7$Be input is in the water column and that the short-term estuarine trapping efficiency for atmospherically derived $^7$Be 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, $^7$Be 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 it is probably less than 30% in humid regions [Olsen et al., 1985]. At mid-latitudes, the total (wet + dry) atmospheric flux of $^7$Be commonly supports inventories ranging from 1.0 to 2.0 pCi/cm$^2$ [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, $^7$Be appears to be solubilized and $^7$Be$^{2+}$ is probably the predominant species [Olsen et al., 1985].

Once deposited as a constituent of rainwater, $^7$Be 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 $^7$Be 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 $^7$Be 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 $^7$Be and its mode of deposition in river-estuarine and coastal environments. From this examination, we present new information concerning (1) the distribution of $^7$Be between dissolved and particulate phases in estuarine and coastal areas, (2) factors that affect the concentration of $^7$Be on suspended particles, (3) rates for $^7$Be removal from the water column, (4) the heterogeneity of $^7$Be accumulation patterns in estuarine sediments, and (5) the extent to which $^7$Be 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 $^7$Be 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 $^7$Be 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 HNO₃ and spiked with 40 mL of a 20% FeCl₃ 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 NH₄OH was added to adjust the pH to ~10 and to precipitate Fe(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
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 ¹³³Cs, 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>
<thead>
<tr>
<th>Location</th>
<th>Particle Concentration, mg/L</th>
<th>Salinity, %</th>
<th>Total Carbon, %</th>
<th>Total Silicon, a</th>
<th>Total Aluminum, mg/g</th>
<th>Total Potassium, b</th>
<th>Total Iron, mg/g</th>
<th>Particle Activity, pCi/kg</th>
<th>Water Activity, pCi/L</th>
<th>Distribution, c</th>
<th>Total Activity, pCi/L</th>
</tr>
</thead>
<tbody>
<tr>
<td>CONO (Oct. 3, 1983)</td>
<td>FW 6</td>
<td>10.6</td>
<td>19</td>
<td>80</td>
<td>20(18)</td>
<td>85</td>
<td>11,800 ± 600</td>
<td>0.06 ± 0.01</td>
<td>2.0 × 10⁴</td>
<td>0.13 ± 0.01</td>
<td></td>
</tr>
<tr>
<td>CONO (April 3, 1984)</td>
<td>FW 15</td>
<td>2.3</td>
<td>23</td>
<td>100</td>
<td>24(28)</td>
<td>53</td>
<td>8,700 ± 320</td>
<td>0.38 ± 0.01</td>
<td>2.3 × 10⁴</td>
<td>0.51 ± 0.01</td>
<td></td>
</tr>
<tr>
<td>Upper Chesapeake Bay</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>SOFT (Oct. 3, 1983)</td>
<td>FW 1</td>
<td>10.0</td>
<td>20</td>
<td>78</td>
<td>19(18)</td>
<td>64</td>
<td>11,700 ± 500</td>
<td>0.06 ± 0.01</td>
<td>2.0 × 10⁴</td>
<td>0.13 ± 0.01</td>
<td></td>
</tr>
<tr>
<td>ANAP (Oct. 4, 1983)</td>
<td>12</td>
<td>22.5</td>
<td>15</td>
<td>51</td>
<td>13(14)</td>
<td>58</td>
<td>4,600 ± 630</td>
<td>0.03 ± 0.01</td>
<td>1.5 × 10⁵</td>
<td>0.05 ± 0.01</td>
<td></td>
</tr>
<tr>
<td>CCFF (Oct. 5, 1983)</td>
<td>14</td>
<td>15.7</td>
<td>18</td>
<td>36</td>
<td>10(13)</td>
<td>39</td>
<td>4,500 ± 580</td>
<td>0.07 ± 0.01</td>
<td>6.4 × 10⁴</td>
<td>0.09 ± 0.01</td>
<td></td>
</tr>
<tr>
<td>SOFT (April 2, 1984)</td>
<td>FW 9</td>
<td>3.2</td>
<td>23</td>
<td>100</td>
<td>25(29)</td>
<td>57</td>
<td>13,700 ± 200</td>
<td>0.22 ± 0.01</td>
<td>6.5 × 10⁴</td>
<td>0.48 ± 0.01</td>
<td></td>
</tr>
<tr>
<td>ANAP (April 4, 1984)d</td>
<td>24</td>
<td>10.6</td>
<td>20</td>
<td>91</td>
<td>22(24)</td>
<td>47</td>
<td>12,800 ± 300</td>
<td>0.55 ± 0.01</td>
<td>2.3 × 10⁴</td>
<td>0.86 ± 0.01</td>
<td></td>
</tr>
<tr>
<td>CCFF (April 5, 1984)</td>
<td>8</td>
<td>33.7</td>
<td>8</td>
<td>27</td>
<td>8(9)</td>
<td>14</td>
<td>11,100 ± 300</td>
<td>0.34 ± 0.01</td>
<td>3.3 × 10⁴</td>
<td>0.46 ± 0.01</td>
<td></td>
</tr>
<tr>
<td>James River Estuary</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>HOPE (June 19, 1981)</td>
<td>FW 3</td>
<td>4.3</td>
<td>23</td>
<td>96</td>
<td>17(19)</td>
<td>51</td>
<td>1,580 ± 265</td>
<td>0.10 ± 0.02</td>
<td>1.6 × 10⁴</td>
<td>0.15 ± 0.02</td>
<td></td>
</tr>
<tr>
<td>S409 (June 23, 1981)</td>
<td>7</td>
<td>3.5</td>
<td>17</td>
<td>81</td>
<td>17(16)</td>
<td>41</td>
<td>835 ± 365</td>
<td>0.11 ± 0.04</td>
<td>7.6 × 10⁴</td>
<td>0.13 ± 0.04</td>
<td></td>
</tr>
<tr>
<td>HAMP (June 23, 1981)</td>
<td>22</td>
<td>1.7</td>
<td>10</td>
<td>17</td>
<td>17(15)</td>
<td>...</td>
<td>1,680 ± 705</td>
<td>0.13 ± 0.04</td>
<td>1.3 × 10⁴</td>
<td>0.15 ± 0.04</td>
<td></td>
</tr>
<tr>
<td>Hudson-Raritan Estuary</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>RARR (July 24, 1981)</td>
<td>FW 3</td>
<td>...</td>
<td>...</td>
<td>...</td>
<td>...</td>
<td>...</td>
<td>8,040 ± 1120</td>
<td>0.05 ± 0.01</td>
<td>1.6 × 10⁴</td>
<td>0.07 ± 0.01</td>
<td></td>
</tr>
<tr>
<td>SRIV (July 24, 1981)</td>
<td>FW 29</td>
<td>8.6</td>
<td>...</td>
<td>...</td>
<td>(19)</td>
<td>...</td>
<td>6,760 ± 610</td>
<td>0.09 ± 0.01</td>
<td>7.5 × 10⁴</td>
<td>0.29 ± 0.01</td>
<td></td>
</tr>
<tr>
<td>LENT (July 26, 1981)</td>
<td>4</td>
<td>5.6</td>
<td>...</td>
<td>...</td>
<td>(22)</td>
<td>...</td>
<td>1,300 ± 330</td>
<td>0.18 ± 0.01</td>
<td>7.2 × 10³</td>
<td>0.20 ± 0.01</td>
<td></td>
</tr>
<tr>
<td>HARB (July 26, 1981)</td>
<td>24</td>
<td>7.3</td>
<td>16</td>
<td>56</td>
<td>17(20)</td>
<td>39</td>
<td>2,760 ± 290</td>
<td>0.21 ± 0.01</td>
<td>1.3 × 10⁴</td>
<td>0.23 ± 0.01</td>
<td></td>
</tr>
<tr>
<td>KILL (July 23, 1981)</td>
<td>26</td>
<td>3.8</td>
<td>12</td>
<td>50</td>
<td>17(15)</td>
<td>37</td>
<td>2,900 ± 330</td>
<td>0.06 ± 0.01</td>
<td>4.8 × 10⁴</td>
<td>0.09 ± 0.01</td>
<td></td>
</tr>
<tr>
<td>RARB (July 22, 1981)</td>
<td>28</td>
<td>8.3</td>
<td>14</td>
<td>50</td>
<td>17(17)</td>
<td>37</td>
<td>11,300 ± 360</td>
<td>0.29 ± 0.01</td>
<td>3.9 × 10⁴</td>
<td>0.37 ± 0.01</td>
<td></td>
</tr>
<tr>
<td>Coastal Shelf</td>
<td></td>
<td></td>
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<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>HOOK (July 22, 1981)</td>
<td>30</td>
<td>4.9</td>
<td>...</td>
<td>...</td>
<td>(10)</td>
<td>...</td>
<td>10,800 ± 530</td>
<td>0.16 ± 0.01</td>
<td>6.8 × 10⁴</td>
<td>0.20 ± 0.01</td>
<td></td>
</tr>
<tr>
<td>AMBR (July 23, 1981)</td>
<td>25</td>
<td>4.0</td>
<td>7</td>
<td>17</td>
<td>12(11)</td>
<td>12</td>
<td>3,350 ± 360</td>
<td>0.11 ± 0.01</td>
<td>3.0 × 10⁴</td>
<td>0.13 ± 0.01</td>
<td></td>
</tr>
</tbody>
</table>

FW is fresh water.

aLiBO₃ fused before ICP analysis to obtain total silicon. A comparison with standards indicates that the reported analyses may be between 5 and 7% low.
bNumbers 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 K₄ 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

<table>
<thead>
<tr>
<th>Sediment Sample (Collection Date)</th>
<th>Depth, cm</th>
<th>Dry Density, g/cm³</th>
<th>7Be Activity, pCi/g</th>
<th>7Be Inventory, pCi/cm²</th>
<th>Approximate Sedimentation Rate, cm/yr</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>James River Estuary</strong></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>T-233 (HOPE) (June 19, 1981)</td>
<td>0-1</td>
<td>0.94</td>
<td>0.20 ± 0.07</td>
<td>0.2</td>
<td>&lt;1</td>
</tr>
<tr>
<td></td>
<td>1-2</td>
<td>1.00</td>
<td>ND</td>
<td></td>
<td></td>
</tr>
<tr>
<td>T-14 (June 19, 1981)</td>
<td>0-1</td>
<td>0.36</td>
<td>0.48 ± 0.27</td>
<td>0.2</td>
<td>1-3</td>
</tr>
<tr>
<td></td>
<td>1-2</td>
<td>0.37</td>
<td>ND</td>
<td></td>
<td></td>
</tr>
<tr>
<td>T-13 (June 19, 1981)</td>
<td>0-1</td>
<td>0.40</td>
<td>0.71 ± 0.26</td>
<td>0.3</td>
<td>1-3</td>
</tr>
<tr>
<td></td>
<td>1-2</td>
<td>0.44</td>
<td>ND</td>
<td></td>
<td></td>
</tr>
<tr>
<td>T-12 (June 18, 1981)</td>
<td>0-1</td>
<td>0.32</td>
<td>1.14 ± 0.21</td>
<td>2.6</td>
<td>&gt;3 (dredged)</td>
</tr>
<tr>
<td></td>
<td>1-2</td>
<td>0.37</td>
<td>1.14 ± 0.25</td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>2-4</td>
<td>0.39</td>
<td>1.06 ± 0.33</td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>4-6</td>
<td>0.42</td>
<td>0.67 ± 0.17</td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>6-8</td>
<td>0.43</td>
<td>0.44 ± 0.18</td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>8-10</td>
<td>0.46</td>
<td>ND</td>
<td></td>
<td></td>
</tr>
<tr>
<td>T-232 (June 18, 1981)</td>
<td>0-1</td>
<td>0.66</td>
<td>0.91 ± 0.24</td>
<td>1.6</td>
<td>&gt;3</td>
</tr>
<tr>
<td></td>
<td>1-2</td>
<td>0.77</td>
<td>0.43 ± 0.11</td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>2-4</td>
<td>1.07</td>
<td>0.29 ± 0.14</td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>4-6</td>
<td>0.94</td>
<td>ND</td>
<td></td>
<td></td>
</tr>
<tr>
<td>T-10 (June 18, 1981)</td>
<td>0-1</td>
<td>0.39</td>
<td>0.50 ± 0.28</td>
<td>1.2</td>
<td>1-3</td>
</tr>
<tr>
<td></td>
<td>1-2</td>
<td>0.46</td>
<td>0.38 ± 0.17</td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>2-4</td>
<td>0.46</td>
<td>0.87 ± 0.18</td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>4-6</td>
<td>0.52</td>
<td>ND</td>
<td></td>
<td></td>
</tr>
<tr>
<td>T-9 (S409) (June 18, 1981)</td>
<td>0-1</td>
<td>0.38</td>
<td>0.74 ± 0.16</td>
<td>1.6</td>
<td>1-3</td>
</tr>
<tr>
<td></td>
<td>1-2</td>
<td>0.45</td>
<td>1.08 ± 0.15</td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>2-4</td>
<td>0.54</td>
<td>0.74 ± 0.16</td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>4-6</td>
<td>0.57</td>
<td>ND</td>
<td></td>
<td></td>
</tr>
<tr>
<td>B-6 (Aug. 12, 1982)</td>
<td>0-1</td>
<td>0.37</td>
<td>0.86 ± 0.11</td>
<td>0.3</td>
<td>1-3</td>
</tr>
<tr>
<td></td>
<td>1-2</td>
<td>0.41</td>
<td>ND</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Wreck shoals (Aug. 12, 1982)</td>
<td>0-2</td>
<td>0.47</td>
<td>0.29 ± 0.07</td>
<td>0.3</td>
<td>&lt;1</td>
</tr>
<tr>
<td></td>
<td>2-4</td>
<td>0.53</td>
<td>ND</td>
<td></td>
<td></td>
</tr>
<tr>
<td>JR mudflat (Aug. 12, 1982)</td>
<td>0-1</td>
<td>0.35</td>
<td>1.27 ± 0.22</td>
<td>0.7</td>
<td>1-3</td>
</tr>
<tr>
<td></td>
<td>1-2</td>
<td>0.35</td>
<td>0.42 ± 0.15</td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>2-4</td>
<td>0.33</td>
<td>0.11 ± 0.10</td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>4-6</td>
<td>0.57</td>
<td>ND</td>
<td></td>
<td></td>
</tr>
<tr>
<td>JR marsh (Aug. 12, 1982)</td>
<td>grass</td>
<td>0.09</td>
<td>1.32 ± 0.13</td>
<td>1.8</td>
<td>&lt;1⁴</td>
</tr>
<tr>
<td></td>
<td>0-1</td>
<td>0.11</td>
<td>1.65 ± 0.28</td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>1-2</td>
<td>0.16</td>
<td>1.28 ± 0.22</td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>2-4</td>
<td>0.23</td>
<td>0.58 ± 0.37</td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>4-6</td>
<td>0.30</td>
<td>0.24 ± 0.10</td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>6-10</td>
<td>0.37</td>
<td>ND</td>
<td></td>
<td></td>
</tr>
<tr>
<td>T-8 (June 17, 1981)</td>
<td>0-1</td>
<td>0.42</td>
<td>1.03 ± 0.31</td>
<td>0.6</td>
<td>1-3</td>
</tr>
<tr>
<td></td>
<td>1-2</td>
<td>0.50</td>
<td>0.23 ± 0.13</td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>2-4</td>
<td>0.51</td>
<td>ND</td>
<td></td>
<td></td>
</tr>
<tr>
<td>T-7 (June 17, 1981)</td>
<td>0-1</td>
<td>0.46</td>
<td>0.52 ± 0.15</td>
<td>0.3</td>
<td>&lt;1</td>
</tr>
<tr>
<td></td>
<td>1-2</td>
<td>0.55</td>
<td>ND</td>
<td></td>
<td></td>
</tr>
<tr>
<td>T-6 (June 17, 1981)</td>
<td>0-2A⁶</td>
<td>0.56</td>
<td>0.41 ± 0.14</td>
<td>0.5</td>
<td>&lt;1</td>
</tr>
<tr>
<td></td>
<td>0-2B⁶</td>
<td>0.58</td>
<td>0.41 ± 0.13</td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>2-4</td>
<td>0.60</td>
<td>ND</td>
<td></td>
<td></td>
</tr>
<tr>
<td>T-102 (HAMP) (June 16, 1981)</td>
<td>0-2</td>
<td>0.42</td>
<td>1.54 ± 0.10</td>
<td>3.3</td>
<td>&gt;3 (dredged)</td>
</tr>
<tr>
<td></td>
<td>2-3</td>
<td>0.54</td>
<td>2.00 ± 0.29</td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>3-4</td>
<td>0.53</td>
<td>1.28 ± 0.39</td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>4-5</td>
<td>0.47</td>
<td>0.45 ± 0.27</td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>5-6</td>
<td>0.47</td>
<td>ND</td>
<td></td>
<td></td>
</tr>
<tr>
<td><strong>Chesapeake Bay</strong></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Rapp-1 (June 25, 1981)</td>
<td>0-1</td>
<td>0.45</td>
<td>3.29 ± 0.52</td>
<td>2.9</td>
<td>&gt;3</td>
</tr>
<tr>
<td></td>
<td>1-2</td>
<td>0.51</td>
<td>2.39 ± 0.34</td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>2-3</td>
<td>0.59</td>
<td>0.41 ± 0.28</td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>3-4</td>
<td>0.59</td>
<td>ND</td>
<td></td>
<td></td>
</tr>
<tr>
<td>SQFT (Sept. 28, 1981)</td>
<td>0-2</td>
<td>0.60</td>
<td>0.42 ± 0.09</td>
<td>0.5</td>
<td>&lt;1</td>
</tr>
<tr>
<td></td>
<td>2-4</td>
<td>0.81</td>
<td>ND</td>
<td></td>
<td></td>
</tr>
<tr>
<td>CONO (Sept. 28, 1981)</td>
<td>0-2</td>
<td>0.30</td>
<td>0.71 ± 0.17</td>
<td>0.6</td>
<td>&gt;3</td>
</tr>
<tr>
<td></td>
<td>2-4</td>
<td>0.39</td>
<td>0.23 ± 0.09</td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>4-6</td>
<td>0.44</td>
<td>ND</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>
low. Field particle-to-water distribution coefficients for ⁷Be range from $7 \times 10^3$ to $2 \times 10^5$ and have a median value of about $4 \times 10^4$ (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 *Be 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 in river water 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-
length of time that particles remain in the water column acting as the primary factor governing the concentration of $^7$Be (and by analogy other particle-reactive substances) on suspended particles is the length of time that particles remain in the water column as scavengers.

The effects of atmospheric input on dissolved $^7$Be concentrations are also evident from a comparison of the October and April data for the Susquehanna-Chesapeake Bay system (Table 1). The average dissolved $^7$Be 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" $^7$Be 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 $^7$Be concentration decreased by about a factor of 2 in a similar sample collected at nearby station CCFF (Figure 1; Table 1), approximating dissolved values measured in the system prior to the rainstorm. These results are probably best explained by $^7$Be sorption kinetics. Bloom and Crecelius [1983] have shown that about 50 hours are required for $^7$Be 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 $^7$Be 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 $^7$Be 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 $^7$Be on suspended particles in these coastal systems is the length of time that particles remain in the water column and are able to scavenge $^7$Be 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 $^7$Be on suspended particles and the amount of $^7$Be accumulation in sediments. Concentrations of $^7$Be 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 $^7$Be sediment inventories occur in these areas they probably result from sediment-mixing processes [Olsen et al., 1984]. The lowest concentrations of $^7$Be 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 $^7$Be inventories are greatest (Figure 3).

Anomalies to this general inverse relationship between the concentration of $^7$Be on suspended particles and the inventory of $^7$Be in sediments (Figure 3) certainly occur, and appear most often in freshwater areas. For example, both the inventory and suspended-particulate concentration of $^7$Be 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 $^7$Be in unsampled areas, or (2) a depletion of the $^7$Be 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 ($^7$Be-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 $^7$Be on suspended matter is the length of time that particles remain in the water column.

Field particle-to-water distribution coefficients for $^7$Be range from $7 \times 10^2$ to $2 \times 10^2$ 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 $^7$Be on suspended particles does not appear to be correlated with particulate type or iron content, the particle-to-water distribution coefficients for $^7$Be 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 $^7$Be 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 $^7$Be is associated with Fe-Mn phases on particulate samples (C. R. Olsen, unpublished data, 1985). Although $^7$Be 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 $^7$Be concentration on our suspended particulate samples (Table 1) supports our earlier conclusion that the major factor affecting the total concentration of $^7$Be on suspended matter is the length of time particles remain in the water column. The inverse correlation...
4.2. Water Column Removal Rates and Residence Times

The rate of 7Be removal from the water column via deposition with settling particles, \( \lambda_w \), 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 \( \lambda \), 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 is 9 m [McLean et al., 1983] and to the surface of Upper Chesapeake Bay was 1.10 pCi/cm\(^2\) at the beginning of October 1983 and 1.66 pCi/cm\(^2\) at the beginning of April 1984 when our respective water samples were collected (Figure 2). Assuming that the mean depth of Conowingo Reservoir on the Lower Susquehanna River and to the surface of Upper Chesapeake Bay was 1.10 pCi/cm\(^2\) at the beginning of October 1983 and 1.66 pCi/cm\(^2\) 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/cm\(^2\) (or about 10% of the total input) in October and 0.46 pCi/cm\(^2\) (or about 25% of the total input) in April. Using the box model in Figure 4, the calculated length of time that 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 SQT, 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.

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

<table>
<thead>
<tr>
<th>Sample Date</th>
<th>Input # Province</th>
<th>Water Column Inventory</th>
<th>Removal Rate</th>
<th>Residence Time</th>
</tr>
</thead>
<tbody>
<tr>
<td>Conowingo Reservoir</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>(Oct. 1983)</td>
<td>1.1</td>
<td>0.12</td>
<td>0.11</td>
<td>9</td>
</tr>
<tr>
<td>(April 1984)</td>
<td>1.7</td>
<td>0.46</td>
<td>0.04</td>
<td>25</td>
</tr>
<tr>
<td>Upper Chesapeake Bay</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>(Oct. 1983)</td>
<td>1.1</td>
<td>0.07</td>
<td>0.19</td>
<td>5</td>
</tr>
<tr>
<td>(April 1984)</td>
<td>1.7</td>
<td>0.48</td>
<td>0.03</td>
<td>33</td>
</tr>
<tr>
<td>James River Estuary</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>(June 1981)</td>
<td>1.0-2.0</td>
<td>0.05</td>
<td>0.51-0.25</td>
<td>2-4</td>
</tr>
<tr>
<td>Hudson River Estuary</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>(July 1981)</td>
<td>1.0-2.0</td>
<td>0.19</td>
<td>0.12-0.06</td>
<td>8-17</td>
</tr>
<tr>
<td>Raritan Bay</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>(July 1981)</td>
<td>1.0-2.0</td>
<td>0.17</td>
<td>0.14-0.06</td>
<td>7-17</td>
</tr>
<tr>
<td>Coastal Shelf</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>(July 1981)</td>
<td>1.0-2.0</td>
<td>0.24</td>
<td>0.10-0.04</td>
<td>10-25</td>
</tr>
</tbody>
</table>

*Input 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).

The average water-column inventories were calculated using the water-column 7Be concentrations in Table 1 and the mean depths of each system.
JAMES RIVER ESTUARY

DEPOSITIONAL ENVIRONMENTS

- DANCING PT. JAMES ISLAND
- TIDAL PT. JAMES ISLAND
- FRESH WATERS
- OLIGOHALINE
- TURBIDITY ZONE
- SURRY REACTOR
- WRECK SHOALS
- OLIGOHALINE
- MesoHALINE
- NEWPORT NEWS
- CHESAPEAKE BAY
- POLYHALINE

DEPOSITIONAL AREAS

- DREDGED CHANNELS OR SLUMPS
- SOFT MUD
- STICKY MUD
- SANDY OR SCOURRED

SAMPING LOCATIONS AND \( ^7 \text{Be} \) SINKS

- BOX CORES
- SURFACE GRABS
- WATER AND SUSPENDED MATTER
- \( ^7 \text{Be} \) SINKS (>1.0 pCi/cm\(^2\))

Fig. 5. (a) Map of the James River estuary illustrating the various deposition environments used in determining the total burden of \( ^7 \text{Be} \) in the estuarine sediments. (b) Location of sediment sampling sites in the James River estuary. The shaded zones represent areas where the \( ^7 \text{Be} \) 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; Hullett and Bender, 1980; Cutshall et al., 1981; Nichols and Cutshall, 1981; Zucker et al., 1984].

...for \( ^7 \text{Be} \) 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, \( ^7 \text{Be} \) 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 \( ^7 \text{Be} \), such exchange is insignificant in relation to the atmospheric input of \( ^7 \text{Be} \) (see discussion concerning \( ^7 \text{Be} \) budget for the James). In addition, incoming waters in these systems have nearly the same water-phase \( ^7 \text{Be} \) concentration as waters at the estuary mouth (Table 1), so the calculated range in removal rates actually reflects the length of time \( ^7 \text{Be} \) remains in the water column rather than reflects the physical flushing time of \( ^7 \text{Be} \) 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 \( ^7 \text{Be} \) 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 \( ^234 \text{Th} \) and \( ^210 \text{Pb} \) removal by Tanaka et al. [1983]), will be required to estimate \( ^7 \text{Be} \) 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...
and particle-associated contaminant accumulation have been
et al., 1981]. Since particle dynamics and patterns of sediment
accumulation are also major sinks for particle-associated con-

channel areas represented less than 5% of the total estuarine
is \(3.0 \pm 0.5\) pCi/cm² in dredged-slump areas’ 1.5 \(\pm 1.0\) pCi cm\(^{-2}\) in
sediment burden of reactor-released 3'Cs and 6øCo in the
surface area, they accounted for more than 40% of the total
accumulation occurred in dredged-channel or slump
areas (Figure 5). Approximately \(1.7 \times 10^9\) kg of suspended
occurs (Figure 5). During June 1981 less than 5% of the Be stand-

column concentration data at Hopewell (HOPE in Table 1)
flow from all tributaries would replace only \(~-, 50\)% of the
than 20% to \(~-, 1.2 \times 10^2\) L. This 77-day, mean-life integrated
lived assuming that (1) the average concentration of Be 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
assumed that (1) the average concentration of 7Be in the
water phase was 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 stand-
ning 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
radio nuclide profiles [Zucker et al., 1984]:

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

extended studies 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 geo-
chemistry 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 radio-
uclide 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 \(4.75 \times 10^{12}\) cm² and
assuming that the atmospheric flux of 7Be ranges from 0.013
to 0.026 pCi cm\(^{-2}\) 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 \(9.3 \times 10^{11}\) L.
Additional freshwater input from the Appomattox and
Chickahominy Rivers would raise this integrated flow by less
than 20% to \(~-, 1.2 \times 10^{12}\) L. This 77-day, mean-life integrated
flow from all tributaries would replace only \(~-, 50\)% 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 drain-
age basin vegetation and soils to retain 7Be. Although ex-
change 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 3.7 \(\times 10^{10}\) Bq.

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

The average 7Be inventory (±1 standard deviation from the mean)
is 3.0 ± 0.5 pCi/cm² in dredged-slump areas; 1.5 ± 1.0 pCi cm\(^{-2}\) in
soft mud areas; 0.3 ± 0.1 pCi/cm² in sticky mud areas; and nonde-
tectable in sandy or scoured areas.

<table>
<thead>
<tr>
<th>TABLE 4. 7Be Inventory for the James River Estuary</th>
</tr>
</thead>
<tbody>
<tr>
<td>Input</td>
</tr>
<tr>
<td>Atmospheric deposition(^b)</td>
</tr>
<tr>
<td>((475 \text{ km}^2))</td>
</tr>
<tr>
<td>Freshwater runoff</td>
</tr>
<tr>
<td>(0.2)</td>
</tr>
<tr>
<td>Total input</td>
</tr>
<tr>
<td>(5 \text{ to } 10)</td>
</tr>
<tr>
<td>Water column inventory</td>
</tr>
<tr>
<td>Water phase</td>
</tr>
<tr>
<td>((1.7 \times 10^{12} \text{ L}))</td>
</tr>
<tr>
<td>Suspended-particle phase</td>
</tr>
<tr>
<td>((20 \text{ mg/L}))</td>
</tr>
<tr>
<td>Total water column</td>
</tr>
<tr>
<td>(0.25)</td>
</tr>
<tr>
<td>Sediment inventory(^c)</td>
</tr>
<tr>
<td>Dredged-channel areas</td>
</tr>
<tr>
<td>((25 \text{ km}^2))</td>
</tr>
<tr>
<td>Soft-mud accumulation areas</td>
</tr>
<tr>
<td>((210 \text{ km}^2))</td>
</tr>
<tr>
<td>Sandy areas</td>
</tr>
<tr>
<td>((60 \text{ km}^2))</td>
</tr>
<tr>
<td>Total sediments</td>
</tr>
<tr>
<td>(4.5 \pm 2.1)</td>
</tr>
</tbody>
</table>

\(^{b}\) Ci = \(3.7 \times 10^{10}\) Bq.

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

\(^{c}\) The average 7Be inventory (±1 standard deviation from the mean)
is 3.0 ± 0.5 pCi/cm² in dredged-slump areas; 1.5 ± 1.0 pCi cm\(^{-2}\) in
soft mud areas; 0.3 ± 0.1 pCi/cm² in sticky mud areas; and nonde-
tectable in sandy or scoured areas.

\(^{c}\) Not 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 re-
moval 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 \(\sim 3.5\) m, and its MLW surface area is \(\sim 4.8 \times 10^8\) m²
\(\text{[Cronin, 1971]}\). Freshwater inflow averages 211 m³/s at Rich-
mond, and the normal upstream limit of salt intrusion is near
James Island where a pronounced turbidity maximum also
occurs (Figure 5). Approximately \(1.7 \times 10^9\) 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 ex-
tremely heterogeneous. Zucker et al. [1984] have measured
the vertical distribution of fallout and reactor-released radio-
uclides in 29 box cores collected throughout the estuary (in-
cluding 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 con-
taminants, such as Kepone, released near Hopewell [Cutshall
et al., 1981]. Since particle dynamics and patterns of sediment
and particle-associated contaminant accumulation have been

\(\text{NOTES}

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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 almost 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 pCi cm−2 d−1 [Sakeh 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 pCi cm−2 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 pCi cm−2 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 pCi cm−2 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 pCi cm−2 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 pCi cm−2 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 104 and 105 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/cm², 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 pCi cm−2 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 x 103 to 2 x 105 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 \(^7\)Be from estuarine and coastal waters.

6. The lack of correlation between the concentration of \(^7\)Be 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 \(^7\)Be to become associated with specific particle types.

7. Residence times of \(^7\)Be 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 \(^7\)Be 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 \(^7\)Be.

9. The short-term trapping efficiency of the James estuary for \(^7\)Be 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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