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Oceanographic Implications of Radioactive
Fall-out Distributions in the Atlantic Ocean:
From 20°N to 25°S, from 1957 to 1961

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Woods Hole, Massachusetts

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Clark University
Worcester, Massachusetts

ABSTRACT

The patterns of Sr$^{90}$ distribution in the equatorial Atlantic are described for the years 1957, 1958, 1960, and 1961. Evidence is drawn from analysis of surface samples and of vertical profiles. The concentration distributions bear no resemblance to patterns of fall-out delivery on land; it is concluded that hydrographic processes are responsible for seawater concentration patterns. The data provide evidence for two otherwise unobserved, high fall-out incidents over the Atlantic Ocean. The rapid vertical penetration of Sr$^{90}$ in the ocean, and the relatively rapid attainment of horizontal and vertical uniformity of concentration in the equatorial Atlantic are consistent with patterns of current flow derived from hydrographic considerations, only if these are complicated by very active unoriented mixing processes within water masses. Integration of vertical profiles yields estimates of fall-out delivery to the equatorial ocean surface that are much higher than estimates of delivery to land surfaces at comparable latitudes.

Vertical profiles of Ce$^{144}$ and Pm$^{147}$ show secondary concentration maxima at various depths; some of these maxima are interpreted as being local concentrations of sinking particles whereas others appear to represent retention in the bodies of plankton swarms. In most cases these secondary maxima are enriched in Pm$^{147}$ vs. Ce$^{144}$.

Introduction. For several years we have been studying the geochemical tracer experiment provided in the Atlantic Ocean by some longer-lived radioisotopes delivered to the ocean surface in the form of world-wide fall-out from nuclear test explosions. Much of our material has been published previously (Bowen and Sugihara 1958, 1960, 1963, Sugihara and Bowen 1962). With data from more recent cruises available, it now seems desirable to summarize the changing patterns observed in several more or less discrete areas in the Atlantic Ocean.

1. Accepted for publication and submitted to press 9 April 1965.
The Atlantic Ocean between 20°N and 20°S exhibits a reasonably coherent, though seasonally changing, pattern of easterly or westerly surface currents (Neumann 1960). In addition, there is a well-developed Equatorial Undercurrent (Metcalf et al. 1962, Cochrane 1963). During the various phases of the EquaLANT multiship, multination program, the oceanography of the Equatorial Atlantic was very extensively resurveyed (Austin 1963).

Procedures for Sampling and Analysis. Surface seawater samples were collected by dipping with polyethylene buckets, or, after confirmation that the ship's fire system introduced no measurable error, water was drawn from this system after it had been thoroughly flushed. Since 1958, the latter method has been routine.

All subsurface samples collected during 1957 were taken in a collapsible neoprene sampler (Bowen and Howland 1964). From mid-1958 to the present, the large-volume sampler designed by Bodman et al. (1961) has been employed. In all cases, two salinity measurements per sampling were made—on sub-samples from the large-volume sample and from a Nansen bottle tripped simultaneously. Agreement between these two values and those at a comparable depth inferred from a hydrographic cast closely preceding the large-volume station has been taken as a criterion of good performance.

The basic radiochemical procedures employed have been described (Sugihara et al. 1959). A summary of the modified procedures currently in use is now being prepared for publication elsewhere. The results represent the sum of the dissolved and the particulate forms of each isotope; the unfiltered volume of seawater was acidified to below pH 1.5, appropriate carriers were added, and the sample was stirred with a stream of nitrogen bubbles for no less than 12 hours. In the cases of strontium\(^90\), cerium\(^144\), and promethium\(^147\), our methods could not detect radioactivity from the insoluble residue after treatment as above.

Contrary to the experience described by the other U.S. group reporting fission-product concentrations in the Atlantic (Rocco and Broecker 1963), we have found no systematic radiochemical blank ascribable to either Sr\(^90\) or Cs\(^137\), whether in our reagents, in simulated samples, or in real samples. In the case of real samples, we continue to find some seawater samples from which we can extract Sr or Cs fractions that contain less than our detection limits of Sr\(^90\) or Cs\(^137\) (from 0.2 to 0.5 dpm for Sr\(^90\), and from 0.1 to 0.3 dpm for Cs\(^137\)). In the case of reagents or simulated samples, a comparable statement could be made about Ce\(^144\) or Pm\(^147\). For some years we have not found a seawater sample that was below our detection limits (Ce\(^144\) 0.2 to 0.4 dpm; Pm\(^147\) 0.2 to 0.5 dpm) for these two fission products. Since contamination from the reagents and further contamination in processing (described by Rocco and Broecker 1963) have not been observed in our laboratory, and since we believe that other investigators engaged in low-level measurements of fission products...
have not found such contamination, we apply considerable caution to the interpretation of reported values to which these blanks have made a significant contribution; see Sugihara (1961, 1962).

Measurements. The samples discussed here derive from three IGY cruises of R. V. CRAWFORD, during which latitudinal trans-Atlantic hydrographic profiles were made at 24°S, 16°S, 8°S, 0°, and 16°N; from one Caribbean cruise of R. V. CRAWFORD (Fuglister 1960); and from two cruises of R. V. CHAIN. Although in no case is the coverage adequate, information is available for the years 1957, 1958, 1960, and 1961.

In Table I are presented the Sr⁹⁰, Ce¹⁴⁴, and Pm¹⁴⁷ concentrations in surface samples; in Fig. 1 the values for Sr⁹⁰ are also shown, in position. In Table II are collected the Sr⁹⁰ values for six vertical profiles made in 1957 and for


<table>
<thead>
<tr>
<th>Coll. Date</th>
<th>Lat.</th>
<th>Long.</th>
<th>Temp. °C</th>
<th>Salinity ‰</th>
<th>Sr⁹⁰*</th>
<th>Ce¹⁴⁴*</th>
<th>Pm¹⁴⁷*</th>
</tr>
</thead>
<tbody>
<tr>
<td>1957</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>IV-1</td>
<td>15°49’S</td>
<td>35°50’W</td>
<td>27.2</td>
<td>37.112</td>
<td>4.1 ± 0.5</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>IV-21</td>
<td>15°40’S</td>
<td>7°00’E</td>
<td>23.6</td>
<td>36.046</td>
<td>3.1 ± 0.5</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>III-20</td>
<td>8°18’S</td>
<td>31°22’W</td>
<td>27.9</td>
<td>36.47</td>
<td>3.4 ± 0.5</td>
<td>8.8 ± 0.5</td>
<td>2.2 ± 0.5</td>
</tr>
<tr>
<td>III-7</td>
<td>8°19’S</td>
<td>7°43’W</td>
<td>27.6</td>
<td>35.98</td>
<td>5.0 ± 1.0</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>V-18</td>
<td>8°15’N</td>
<td>49°15’W</td>
<td>26.72</td>
<td>36.024</td>
<td>5.4 ± 1.0</td>
<td>18.1 ± 1.3</td>
<td>2.0 ± 0.3</td>
</tr>
<tr>
<td>V-8</td>
<td>8°17’N</td>
<td>17°22’W</td>
<td>27.17</td>
<td>35.628</td>
<td>4.6 ± 1.0</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>XI-23</td>
<td>16°14’N</td>
<td>45°01’W</td>
<td>26.79</td>
<td>36.634</td>
<td>6.3 ± 0.6</td>
<td>33.3 ± 2.8</td>
<td>6.5 ± 1.0</td>
</tr>
<tr>
<td>II-13</td>
<td>21°08’N</td>
<td>33°20’W</td>
<td>21.84</td>
<td>37.077</td>
<td>4.7 ± 1.0</td>
<td>14.7 ± 1.2</td>
<td>2.2 ± 0.3</td>
</tr>
<tr>
<td>1958</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>X-2</td>
<td>23°58’S</td>
<td>40°32’W</td>
<td>22.89</td>
<td>36.845</td>
<td>7.8 ± 1.8</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>X-24</td>
<td>24°12’S</td>
<td>6°52’E</td>
<td>16.79</td>
<td>35.386</td>
<td>5.3 ± 0.3</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>XI-26</td>
<td>0°10’S</td>
<td>33°40’W</td>
<td>26.57</td>
<td>36.130</td>
<td>7.1 ± 0.7</td>
<td>21.0 ± 0.7</td>
<td>3.6 ± 0.9</td>
</tr>
<tr>
<td>XI-12</td>
<td>0°23’S</td>
<td>9°41’W</td>
<td>23.36</td>
<td>35.855</td>
<td>3.5 ± 0.5</td>
<td>9.0 ± 0.5</td>
<td>1.8 ± 0.3</td>
</tr>
<tr>
<td>XII-11</td>
<td>4°36’N</td>
<td>49°43’W</td>
<td>27.3</td>
<td>-</td>
<td>7.7 ± 0.7</td>
<td>18.0 ± 0.5</td>
<td>-</td>
</tr>
<tr>
<td>XII-12</td>
<td>8°25’N</td>
<td>54°04’W</td>
<td>27.6</td>
<td>-</td>
<td>7.0 ± 0.4</td>
<td>14.5 ± 0.9</td>
<td>4.8 ± 0.6</td>
</tr>
<tr>
<td>1960</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>II-5</td>
<td>16°12’N</td>
<td>57°30’W</td>
<td>26.19</td>
<td>36.057</td>
<td>15 ± 1.1</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>II-15</td>
<td>15°32’N</td>
<td>66°36’W</td>
<td>27.12</td>
<td>35.865</td>
<td>15 ± 2.0</td>
<td>1550 ± 93</td>
<td>-</td>
</tr>
<tr>
<td>II-17</td>
<td>17°24’N</td>
<td>71°20’W</td>
<td>27.3</td>
<td>-</td>
<td>14 ± 1.4</td>
<td>-</td>
<td>337 ± 17</td>
</tr>
<tr>
<td>1961</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>IV-24</td>
<td>00°13’S</td>
<td>18°34’W</td>
<td>29.27</td>
<td>35.008</td>
<td>11.1 ± 0.5</td>
<td>25 ± 1.3</td>
<td>24 ± 1.4</td>
</tr>
<tr>
<td>IV-28</td>
<td>5°15’N</td>
<td>23°30’W</td>
<td>28.16</td>
<td>35.257</td>
<td>10.9 ± 0.4</td>
<td>8.5 ± 0.6</td>
<td>11 ± 1.6</td>
</tr>
<tr>
<td>IV-30</td>
<td>8°05’N</td>
<td>26°14’W</td>
<td>26.91</td>
<td>35.979</td>
<td>10.3 ± 1.0</td>
<td>11.9 ± 0.6</td>
<td>6.5 ± 0.7</td>
</tr>
<tr>
<td>V-2</td>
<td>11°01’N</td>
<td>29°39’W</td>
<td>24.98</td>
<td>36.122</td>
<td>11.9 ± 0.8</td>
<td>8.3 ± 0.5</td>
<td>5.3 ± 0.6</td>
</tr>
<tr>
<td>V-6</td>
<td>19°18’N</td>
<td>40°47’W</td>
<td>24.3</td>
<td>37.022</td>
<td>11.6 ± 2.2</td>
<td>78 ± 4.1</td>
<td>164 ± 10.5</td>
</tr>
</tbody>
</table>

* In disintegrations per minute per 100 L; confidence limits, 2 sigma on basis of counting statistics; on samples from 1958 on, uncertainty includes errors inferred from reproducibility of duplicate samples, variations in chemical-yield determination, and analysis of standard Sr⁹⁰ from National Bureau of Standards.
TABLE II. VERTICAL PROFILES OF STRONTIUM\(^{90}\) CONCENTRATION,** 1957 AND 1958.

<table>
<thead>
<tr>
<th>Lat.</th>
<th>Long.</th>
<th>Month</th>
<th>Depth (m)</th>
</tr>
</thead>
<tbody>
<tr>
<td>15°49'S</td>
<td>35°45'W</td>
<td>IV</td>
<td>0</td>
</tr>
<tr>
<td>8°18'S</td>
<td>31°22'W</td>
<td>III</td>
<td>50</td>
</tr>
<tr>
<td>8°19'S</td>
<td>7°43'E</td>
<td>III</td>
<td>100</td>
</tr>
<tr>
<td>8°15'N</td>
<td>49°15'W</td>
<td>V</td>
<td>300</td>
</tr>
<tr>
<td>16°15'N</td>
<td>17°22'W</td>
<td>V</td>
<td>1000</td>
</tr>
<tr>
<td>0°10'S</td>
<td>43°38'W</td>
<td>XI</td>
<td>2000</td>
</tr>
<tr>
<td>0°23'S</td>
<td>33°40'W</td>
<td>XI</td>
<td>2500</td>
</tr>
<tr>
<td>3°22'W</td>
<td>9°41'W</td>
<td>XI</td>
<td>3000</td>
</tr>
<tr>
<td>0°10'S</td>
<td>43°38'W</td>
<td>XI</td>
<td>3500</td>
</tr>
<tr>
<td>0°23'S</td>
<td>33°40'W</td>
<td>XI</td>
<td>4000</td>
</tr>
<tr>
<td>8°15'N</td>
<td>17°22'W</td>
<td>V</td>
<td>4500</td>
</tr>
</tbody>
</table>

** In disintegrations per minute per 100 L; confidence limits, 2 sigma on basis of counting statistics; on samples from 1958 on, uncertainty includes errors inferred from reproducibility of duplicate samples, variations in chemical-yield determination, and analysis of standard Sr\(^{90}\) from National Bureau of Standards.

† Surface value from 7°43'W.

‡ Extrapolated from 16°14'N, 45°01'W.

* Reassigned value, shown at the depth determined from salinity rather than tripping depth. The tripping depth is indicated by an asterisk in parentheses.

†† Depths falling between those tabulated in first column are typed on intermediate lines; this sample was from 400 m.

two made in 1958. In Table III are collected comparable values for two vertical profiles made in 1960 and for three made in 1961. In Table IV are collected the Ce\(^{144}\) concentrations for vertical profiles obtained in 1958, 1960, and 1961; in Table V, similarly, are collected the corresponding profiles of Pm\(^{147}\). The analytical data for Ce\(^{144}\) and Pm\(^{147}\) have been adjusted for radioactive decay that occurred after the collection date of each sample.

Details of each vertical profile, with all hydrographic data, have also been prepared and are deposited with the American Documentation Institute.²

2. To obtain these detailed data, order Document 8375 from ADI Auxiliary Publication Project, Photoduplication Service, Library of Congress, Washington 25, D.C. When ordering, cite the Document number and remit $2.50 for photoprints, or $1.75 for 35-mm microfilm. Advance payment is required. Make your check payable to: Chief, Photoduplication Service.
### TABLE III. Vertical Profiles of Strontium$^{90}$ Concentration, ** 1960 and 1961.

<table>
<thead>
<tr>
<th>Lat.</th>
<th>16°12'N</th>
<th>15°32'N</th>
<th>0°13'S</th>
<th>5°15'N</th>
<th>11°01'N</th>
</tr>
</thead>
<tbody>
<tr>
<td>Long.</td>
<td>57°30'W</td>
<td>66°36'W</td>
<td>18°34'W</td>
<td>23°30'W</td>
<td>29°39'W</td>
</tr>
<tr>
<td>Month</td>
<td>II</td>
<td>II</td>
<td>IV</td>
<td>IV</td>
<td>V</td>
</tr>
<tr>
<td>Depth (m)</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>0</td>
<td>15 ± 1.1 (26 ± 1.2)*</td>
<td>15 ± 2 (22.2 ± 2.5)*</td>
<td>11.1 ± 0.5</td>
<td>10.9 ± 0.4</td>
<td>11.9 ± 0.8</td>
</tr>
<tr>
<td>100</td>
<td>15.2 ± 1.0 (22.2 ± 2.5)*</td>
<td>14.7 ± 1.5</td>
<td>3.9 ± 0.4</td>
<td>4.5 ± 0.2</td>
<td>4.9 ± 0.4</td>
</tr>
<tr>
<td>300</td>
<td>8.3 ± 0.8</td>
<td>3.6 ± 0.3</td>
<td>3.9 ± 0.3</td>
<td>2.9 ± 0.3</td>
<td>5.6 ± 0.4</td>
</tr>
<tr>
<td>500</td>
<td>3.9 ± 0.4</td>
<td>2.9 ± 0.3</td>
<td>3.9 ± 0.3</td>
<td>3.9 ± 0.3</td>
<td>29 ° 39 ' W</td>
</tr>
<tr>
<td>700</td>
<td>(*)</td>
<td>3.6 ± 0.3</td>
<td>3.7 ± 0.4</td>
<td>2.9 ± 0.3</td>
<td>3.2 ± 0.3</td>
</tr>
<tr>
<td>1000</td>
<td>3.2 ± 0.2</td>
<td>4.5 ± 0.4</td>
<td>(*)</td>
<td>1900 ± 95</td>
<td>2.5 ± 0.4</td>
</tr>
<tr>
<td>1500</td>
<td>3.2 ± 0.5</td>
<td>2.8 ± 0.3</td>
<td>(*)</td>
<td>1900 ± 95</td>
<td>2.5 ± 0.4</td>
</tr>
</tbody>
</table>

** In disintegrations per minute per 100 L; confidence limits, 2 sigma, based on counting statistics, reproducibility of replicates, variations in chemical-yield determination, and analysis of standard Sr$^{90}$ from National Bureau of Standards.

* Reassigned values, shown at 0 and 100 m, determined from salinities rather than tripping depths. The respective tripping depths are indicated by an asterisk in parentheses.

### TABLE IV. Vertical Profiles of Cerium$^{144}$ Concentration, ** 1958, 1960, 1961.

<table>
<thead>
<tr>
<th>Lat.</th>
<th>0°10'S</th>
<th>0°23'S</th>
<th>16°12'N</th>
<th>15°32'N</th>
<th>0°13'S</th>
<th>5°15'N</th>
<th>11°01'N</th>
</tr>
</thead>
<tbody>
<tr>
<td>Long.</td>
<td>33°40'W</td>
<td>9°41'W</td>
<td>57°30'W</td>
<td>66°36'W</td>
<td>18°34'W</td>
<td>23°30'W</td>
<td>29°39'W</td>
</tr>
<tr>
<td>Month</td>
<td>XI</td>
<td>XI</td>
<td>II</td>
<td>II</td>
<td>IV</td>
<td>IV</td>
<td>V</td>
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<tr>
<td>Depth (m)</td>
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<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>0</td>
<td>21 ± 0.7 (193 ± 2)*</td>
<td>9.0 ± 0.5</td>
<td>1550 ± 100</td>
<td>25 ± 1.3</td>
<td>8.5 ± 0.6</td>
<td>8.3 ± 0.5</td>
<td></td>
</tr>
<tr>
<td>100</td>
<td>10 ± 0.5 (193 ± 2)*</td>
<td>7.3 ± 0.8</td>
<td>970 ± 30</td>
<td>1360 ± 70</td>
<td>1.6 ± 0.3</td>
<td>5 ± 0.5</td>
<td></td>
</tr>
<tr>
<td>300</td>
<td>12.8 ± 0.6 (193 ± 2)*</td>
<td>2.0 ± 0.1</td>
<td>3.2 ± 0.7</td>
<td>1900 ± 95</td>
<td>2.5 ± 0.4</td>
<td>4.5 ± 0.4</td>
<td></td>
</tr>
<tr>
<td>500</td>
<td>(*)</td>
<td>12 ± 1.7 (193 ± 2)*</td>
<td>1900 ± 95</td>
<td>2.5 ± 0.4</td>
<td>4.5 ± 0.4</td>
<td>4 ± 0.4</td>
<td></td>
</tr>
<tr>
<td>700</td>
<td>23.5 ± 0.7 (193 ± 2)*</td>
<td>4.2 ± 0.2 (193 ± 2)*</td>
<td>1900 ± 95</td>
<td>2.5 ± 0.4</td>
<td>4.5 ± 0.4</td>
<td>4 ± 0.4</td>
<td></td>
</tr>
<tr>
<td>1000</td>
<td>3.7 ± 0.5</td>
<td>2.8 ± 0.3</td>
<td>1900 ± 95</td>
<td>2.5 ± 0.4</td>
<td>4.5 ± 0.4</td>
<td>4 ± 0.4</td>
<td></td>
</tr>
<tr>
<td>2000</td>
<td>8 ± 0.5</td>
<td>4.2 ± 0.2</td>
<td>1900 ± 95</td>
<td>2.5 ± 0.4</td>
<td>4.5 ± 0.4</td>
<td>4 ± 0.4</td>
<td></td>
</tr>
<tr>
<td>2500</td>
<td>1.9 ± 0.3</td>
<td>2.8 ± 0.3</td>
<td>1900 ± 95</td>
<td>2.5 ± 0.4</td>
<td>4.5 ± 0.4</td>
<td>4 ± 0.4</td>
<td></td>
</tr>
</tbody>
</table>

** In disintegrations per minute per 100 L, corrected for decay since collection; confidence limits, 2 sigma on basis of counting statistics; on samples from 1958 on, uncertainty includes errors inferred from reproducibility of duplicate samples, and variations in chemical-yield determination.

* Reassigned value, shown at the depth determined from salinity rather than tripping depth. The tripping depth is indicated by an asterisk in parentheses.
### TABLE V. VERTICAL PROFILES OF PROMETHIUM\(^{147}\) CONCENTRATIONS, ** 1958, 1960, 1961.**

<table>
<thead>
<tr>
<th>Lat.</th>
<th>0°10'S</th>
<th>0°23'S</th>
<th>16°12'N</th>
<th>15°32'N</th>
<th>0°13'S</th>
<th>5°15'N</th>
<th>11°01'N</th>
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<tr>
<td>Long.</td>
<td>33°40'W</td>
<td>9°41'W</td>
<td>57°30'W</td>
<td>66°36'W</td>
<td>18°34'W</td>
<td>23°30'W</td>
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<tr>
<td>Month</td>
<td>XI</td>
<td>XI</td>
<td>II</td>
<td>II</td>
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<tr>
<td>0</td>
<td>3.6 ± 0.9</td>
<td>1.8 ± 0.2</td>
<td>24 ± 1.4</td>
<td>11 ± 1.6</td>
<td>5.3 ± 0.6</td>
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<tr>
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<td></td>
<td>200 ± 8</td>
<td>3800 ± 190</td>
<td>5 ± 1</td>
<td>3.4 ± 0.5</td>
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<tr>
<td>300</td>
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<td>(*)</td>
<td>7.3 ± 0.7</td>
<td>7 ± 1.4</td>
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<tr>
<td>500</td>
<td>(*)</td>
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<td></td>
<td>8.6 ± 1.3</td>
<td>243 ± 12</td>
<td>10 ± 1.5</td>
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<td>3560 ± 180</td>
<td>3.2 ± 0.6</td>
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<td>1.7 ± 0.4</td>
</tr>
</tbody>
</table>

** In disintegrations per minute per 100 L, corrected for decay since collection; confidence limits, 2 sigma on basis of counting statistics; on samples from 1958 on, uncertainty includes errors inferred from reproducibility of duplicate samples, and variations in chemical-yield determination.

* Reassigned value, shown at the depth determined from salinity rather than tripping depth. The tripping depth is indicated by an asterisk in parentheses.

These data provide evidence of the good operation of the samplers as well as of the fine structure of the water column in some places. It will be seen that four samples, two from 1958 stations and two from 1960 stations, have been assigned, on the basis of their salinity, to depths shallower than those indicated by the wire out at the time of sending the messenger. Technical improvements in the handling of the water sampler have steadily reduced the frequency of this type of malfunction.

### Discussion (Strontium\(^{90}\)).

Estimates of total Sr\(^{90}\) delivery, inferred from soil analyses, are available for various dates from a world-wide network of collecting points (Alexander et al. 1960, Hardy et al. 1962). These values are supplemented and extended by another world-wide network that collects precipitation data (Hardy et al. 1964). From the early days of study of fall-out deposition, it has been the consensus that these data best fit an array of latitudinal bands, generally with one or more minima at low latitudes. Although the variability among stations within latitudinal bands, coupled with the sparse station coverage in low latitudes, does not justify very precise estimates of the position of a minimum, one is apparent at low latitudes—in the curve for total Sr\(^{90}\) delivered per unit land area—for each year summarized (Alexander et al. 1960, Hardy et al. 1962). In 1956 the minimum lay at about 10°S, in 1958 between 0°...
and 10°S, in 1959 at about 20°S, in 1960 between 10°S and 20°S, and in 1961 between 0° and 10°S. Hardy et al. (1962) summarized the total Sr90 accumulated up to September 1962, in latitudinal bands, using a somewhat different method of estimation. Their curve shows three maxima and two minima: 1.1 megacuries at 40°–50°N, a minimum of 0.3 megacuries at 10°–20°N, 0.35 megacuries at 0°–10°S, a minimum of 0.2 megacuries at 10°–30°S, and 0.35 megacuries at 40°–50°S. It may be argued that the changes in slope of their curve from 20°N to 10°S are much less than the uncertainty of the numbers on which the curve is based.

Measurements of isotope concentrations in tropospheric air (List and Tegadas 1961, Collins 1963, 1964) rarely show the minimum fission-product concentration north of 0° or south of 15°S.

The shifts in position of the Sr90 minimum must be viewed in relation to the complicated curve for annual delivery of this isotope; in temperate latitudes of the northern hemisphere the annual Sr90 increment increased each year from 1954 to 1959; for New York City, taking the 1954 delivery as 100, the deliveries from 1955 to 1962 appear as follows: 1955, 130; 1956, 160; 1957, 160; 1958, 225; 1959, 315; 1960, 60; 1961, 90; 1962, 310.

There is some evidence from the Sr90 values obtained from soil (Alexander et al. 1960, Hardy et al. 1962) that the Equatorial Atlantic experience of fall-out delivery was, during 1955–1959, unlike that in the northern temperate zone (exemplified by New York City). From 1955 to 1957, the Canal Zone showed only a 50% increment whereas Belem showed a percentage increment that was very close to that shown in New York. From 1956 to 1958, the increase at Leopoldville also closely approached that in New York, whereas the increase at Dakar was almost twice that in New York. Most of these increases are difficult to argue from, since local variability, especially in the less humid areas, is considerable. The varying position of the Sr90 minimum, as mentioned above, may also be taken as evidence of a different experience south of the equator.

World-wide averages, then, would indicate that the area from 25°S to 20°N, possibly second only to the areas of the poles, experienced the lowest fall-out delivery. Furthermore, should seawater concentrations exhibit the effect primarily of local fall-out experience, a sharp latitudinal gradient, decreasing southward, would be observed. We might expect this effect to be exaggerated by the hydrographic patterns, since surface circulations are dominated by strong eastward or westward currents, with the meridional currents or components moving largely from south to north (Sverdrup et al. 1942, Neumann 1960, von Arx 1962). In fact, in 1957, as shown in Table I and Fig. 1, all of our surface samples between 15°S and 21°N gave the same Sr90 concentration, within reasonable confidence limits. (The Sr90 concentrations shown in Table I for 1957 and 1958 are estimated only from counting statistics.)
Figure 1. Surface ocean strontium$^{90}$ concentrations, Equatorial Atlantic. Disintegrations per minute per 100 L.
We have given in some detail (Bowen and Sugihara 1963) our arguments for believing that Sr\textsuperscript{90}, soon after reaching the sea surface, is mixed with the large amount of available stable strontium, and that there are no processes that separate large enough amounts of strontium from solution to provide significant vectors of this element in the ocean; this leads to our conclusion that Sr\textsuperscript{90} provides a good tracer for movement of seawater itself.

The Atlantic populations of Acantharia have been studied extensively on material from our various cruises. These organisms are of interest because they build their skeletal spicules of celestite, crystalline strontium sulfate; although these organisms are often surprisingly numerous, no samples from the open Atlantic have shown more than a few milligrams of acantharian skeleton per cubic meter (Massera Bottazzi and Vannucci 1964, 1965). It does not appear that the Acantharia can act as an efficient strontium pump. We regard with some reserve a recent report on stable strontium concentration maxima at intermediate depth (MacKenzie 1964). Examination of the mechanism proposed by MacKenzie has shown that it is ineffective in moving the quantities of Sr required (Siegel, in prep.). None of the studies by previous investigators (Chow and Thompson 1955) has shown such maxima; furthermore, our own curves of vertical distribution of Sr\textsuperscript{90} as well as those of Rocco and Broecker (1963) or Miyake (1962) do not at all resemble curves of isotopes such as Ce\textsuperscript{144} or Pm\textsuperscript{147}, which we believe to move on particles. We therefore continue to believe that the changing distributions of Sr\textsuperscript{90}, both horizontally and vertically, are to be interpreted in hydrographic terms: that concentrations decrease by mixing with water of lower specific activity, and that, when they do increase, this has resulted from advection and admixture of water of higher specific activity, whenever such an increase cannot be explained in terms of direct new fall-out delivery.

It is evident (i) that shallow vertical profiles of Sr\textsuperscript{90} concentration are to be related to the idea of the thermocline as a barrier to vertical mixing, (ii) that all vertical profiles are to be related to the possibility of net downward water movement by sliding along surfaces of uniform density, (iii) that concentrations in deep-water masses are to be related to phenomena of convective down-mixing in Arctic-Antarctic areas and to the various deep currents fed by these regions, and (iv) that horizontal homogeneities and inhomogeneities of surface concentrations are to be related to advection by surface-current systems or to processes of upwelling, as well as to direct local delivery. Data in Tables I–V bear on all these aspects of oceanography.

Neither the pattern of fall-out delivery to the land surface up to 1957 nor the ocean-current fields summarized by Neumann (1960) or by Sverdrup et al. (1942) and von Arx (1962) contribute to an explanation of the Sr\textsuperscript{90} uniformity shown for 1957 (Table I, Fig. 1). The concentrations for 8°S and 15°S should represent water labeled at these latitudes or still farther south; those for 8°N, representing the Equatorial Counter Current, should be water from farther
north, appreciably diluted by contributions from the South Equatorial Current and from upwelling (von Arx 1962: 200) along the margin between the Counter Current and the North Equatorial Current; the 16°N and 21°N samples should be North Equatorial Current water, relatively unmodified. If the difference between the sample from 21°N and the more westerly one from 16°N proved to be significant, we could postulate that the difference is related to the contribution of upwelled water of low specific activity, off the African coast; in this situation, there should be an east-to-west gradient of increasing specific activity, in samples taken at a time of steady fall-out delivery. It is difficult to escape the conclusion that, up to 1957, the over-ocean pattern of fall-out delivery differed from the delivery pattern observed on land.

Unfortunately, the 1957 vertical profiles, summarized in Table II, offer little further clarification of this difference. With a view to C¹⁴ analysis, most of the samples were collected from considerable depths, and these provided blank analyses for Sr⁹⁰. If we postulate that the rate of decrease in concentration with depth at 16°15'N, 43°38'W is applicable to the station at 8°19'S, 7°43'E, then the concentration found at the latter station at 300 m extrapolates to about 5 dpm/100L at the surface, which is actually the amount found in the surface sample at 8°19'S, 7°46'W. Only two of the six samples from about 1000 m and none of those from greater depths showed detectable Sr⁹⁰ concentrations. As was pointed out previously (Bowen and Sugihara 1958), it is gratifying that the apparent rate of reduction in Sr⁹⁰ concentration with depth at 8°19'S, 7°43'E, well within the great South Atlantic Gyre, closely corresponds to that observed by Begemann and Libby (1957) for tritium in the Sargasso Sea. Reasonable arguments also relate this station to that at 16°15'N, 43°38'W (see above).

In 1958 the pattern had shifted markedly; a uniform surface concentration, about 60% higher than in 1957, was indicated for the western basin from 24°S to 8°N, and the values for the two eastern-basin sampling points were essentially unchanged. The western surface samples, from 24°S, 4°N, and 8°N, represent substantially the same current system in Neumann's summary (1960), and the surface sample from 0°10'S, 33°40'W represents water that is strongly influenced by offshoots of this system; these relationships are by no means so clear in other current summaries (von Arx 1962: figs. 6–19, 6–20; also see Sverdrup et al. 1942). On the eastern side, a relatively minor shift of Neumann's current pattern would put the sample from 0°23'S, 9°41'W within the same gyral pattern as the sample from 24°12'S, 6°52'E. The shift of the surface pattern is then perfectly consistent with a sharp increase in the rate of fall-out delivery south of the equator and west of about 30°; at the high current speeds cited by Neumann, less than six months would be required to spread the effect of such an increase over the north-south span of our samples. It is unfortunate that no land observations give evidence of an increase of the magnitude required in late 1957 or early 1958. The northward shift of the
land-delivery minimum between 1956 and 1958, although indicative, is not based on large enough increments to explain the change in water concentrations.

Conception of the distribution in space and time of this postulated accelerated rate of fall-out delivery is made difficult by the horizontal extension of the uniformity in surface Sr\(^{90}\); the farther south we place the area of high fall-out, the longer must be postulated to be the time span over which it occurred. The most favorable situation in terms of space and time seems to be a uniform pattern from about 10\(^\circ\)S to about 40\(^\circ\)S; in this case, the time of high fall-out must have been no more remote than September 1958 to produce the concentrations observed from October to December 1958. The duration of fall-out need not have been more than about a month; as we shall see below, within this pattern it could not have exceeded about three months. Reduction of the horizontal extent of the high fall-out area, or the placement of this area farther south, would require a considerable increase in its postulated duration.

The connections between the two equatorial stations (Table II) require a more complicated analysis. At some times of the year these stations are connected by the westward-flowing South Equatorial Current, as indeed they were during the cruise on which the stations were made; they are also connected by the eastward-flowing Equatorial Undercurrent (Neumann 1960, Metcalf et al. 1962), reported to have velocities that approach 50 nautical miles per day at 100 m and may well average one-quarter of that figure over the range from 70 to 300 m. The passage time between these two stations for water at intermediate depths may be expected to be less than four months; the time ought to increase significantly between 100 and 300 m, and should be much greater at 700 m. Within the framework of our understanding of the currents in this area, we may then interpret the difference in Sr\(^{90}\) profiles at these two stations as revealing that the large western-basin injection, postulated in the previous paragraph, had been introduced early into the Equatorial Undercurrent but had not passed 10\(^\circ\)W prior to November 1958. In view of Cochrane’s (1963) and Metcalf’s (unpublished) hypotheses of the origin of Undercurrent water, either at approximately 4\(^\circ\)N, 45\(^\circ\)W, or at approximately 10\(^\circ\)S, 30\(^\circ\)W, this interpretation is plausible as a time relationship. Under this circumstance, however, we would not have expected uniform Sr\(^{90}\) vs. depth down to 300 m, as observed at 33\(^\circ\)40\('\)W. Such profiles were not observed during 1958 in our other sampling areas (Bowen and Sugihara 1960, and unpublished). That they are also not typical of the equatorial region is shown by the 0\(^\circ\)13\('\)S profile for 1961 (Table III); this station, taken at a time when the rate of fall-out delivery was at a minimum, would be expected to show in exaggerated form the effect of hydrographic processes tending to vertical homogeneity. Again we are left in the uncomfortable position of explaining the 1958 equatorial pattern as the result of unique timing in observing later effects of a fall-out event not observed as it occurred. There is no apparent
reason to connect this event with the striking increase in Sr\textsuperscript{90} concentrations at 1000 m and below.

The Caribbean Sr\textsuperscript{90} values for 1958 are illustrated in Fig. 1 but not tabulated; these were reported earlier by Bowen and Sugihara (1963). To us it seems to be simple coincidence that these values often were the same as those in the band from 25°S to 8°N. It is quite impossible, considering both travel time and mixing of surface currents, that these Caribbean samples could represent direct delivery via the Guianas Current.

The 1960 surface samples (Table I, Fig. 1) confirm a further increase in concentration. The samples do not quite replicate any of the earlier stations; they do, however, lie along the axis of the North Equatorial Current (von Arx 1962) as it is prolonged into the surface drift of the Caribbean. The approximate doubling of the Sr\textsuperscript{90} concentration during 1959 is not surprising, considering the rapid rate of delivery during that year. The still higher values for the two samples from 16°12′N, 57°30′W, reassigned shallower depths on the basis of their salinities, are taken as evidence of a very recent local fall-out event that occurred so close to the sampling area that processes of horizontal and vertical mixing had not smoothed out the increase. If we regard this increment as having been added to the concentration observed in the nominal surface and 100-m samples from this station (a concentration also found at 17°24′N, 71°20′W [Table I]), then we are impressed that down-mixing brought the 100-m increase to 60% of the increase at the surface. This seems to indicate an originally very considerable horizontal extension for this area of high local fall-out. The time difference between collection of the two surface samples and the two 100-m samples at 16°12′N did not exceed six hours, indicating that the gradient at the edge of the mass of high-concentration water was still very sharp. In both cases (see data referred to in footnote 2), the surface-water salinity was about 1 °/oo less than that at 100 m, a fact that makes the apparent speed of down-mixing even more surprising. A similar uniformity of Sr\textsuperscript{90} concentration in the face of a reasonably sharp salinity gradient was seen in the 1960 Caribbean station sample (15°32′N, 66°36′W; Table III). Here the salinity maximum, 1.1 °/oo higher than the surface salinity, was at 150 m, as opposed to about 110 m at 16°12′N. For argument, we may accept the frequently made assumption that vertical water movement is most commonly achieved by "gliding" along isopycnal surfaces. High-salinity surface water was found in 1958 at the southerly end of Caribbean profiles (Fuglister 1960) along 64°W and 74°W, but not at similar latitudes in the December profile along 66°W. Surface salinities of 37 °/oo or more were found in the North Atlantic (Fuglister 1960) at about 26°40′N, 50°W, frequently along 28°N between 21° and 31°W, and along 24°N from 26°W to 53°W. These observations and the argument accepted above could lead to the conclusion that the Sr\textsuperscript{90} observed at 100 m (salinity about 37 °/oo) at 16°12′N, 57°30′W originated as a surface delivery at least 8° farther north and 5°
farther east. That this delivery, which must then have been in transit for at least some months, had arrived at the sampling site, by simple coincidence, just at the time when its Sr$^{90}$ corresponded so closely to the surface Sr$^{90}$ concentration in the surface water, is a distasteful conclusion. Even more distasteful is the application of such reasoning to the sort of horizontal gradient of Sr$^{90}$ concentration that is indicated by the pairs of values for surface and 100 m at the 16°12'N station. It is obvious, however, that the alternative hypothesis, that salinity gradients over so wide an area are maintained in the face of rapid vertical mixing, is equally distasteful to physical oceanographers.

The 1961 samples were planned in relation to both surface currents and possible mechanisms of vertical penetration. Surface Sr$^{90}$ concentrations (Table I, Fig. 1) were found to be uniform along the track from 0°13'S, 18°34'W to 19°18'N, 40°47'W for samples representing a salinity gradient ranging from 35.008%/oo at the equator to 37.022%/oo at 19°18'N. The five sampling positions span the three great surface-current systems in this area; on the basis of the temperature-salinity profile and the bathythermogram section, the first position (0°13'S) was assigned to the South Equatorial Current, the second and third (5°15'N and 8°05'N) to the Equatorial Counter Current, the fourth (11°01'N) to the North Equatorial Current, and the fifth to a location that appears to be across the boundary into the southern part of the North Atlantic Gyre.

Although none of the usually charted water-mass boundaries is presumed to fall about 14°N, 33°30'W, on the cruise in question a very distinct change in many parameters was observed at about this point. The biological aspects of this phenomenon are discussed by Backus et al. (1965). On the basis of the 200-m temperature, the down-turning of the 12°-to-15° isotherms, the drop in surface temperature, and the rise in surface salinity, this appears to have been the transition into the mass of North Atlantic Central water. It is curious that this transition should be strongly evidenced in the mesopelagic fish populations (Backus et al. 1965) but not in the surface Sr$^{90}$ concentrations or in either the Foraminifera (R. Cifelli, in prep.) or Acantharia (Massera Bottazzi and Vannucci 1964). In the case of Sr$^{90}$, the sampling points were widely distributed, but it is clear that the transition took place north of 19°18'N; the surface concentration was 16.5 dpm/100 L at 24°53'N, 49°34'W, and 23.8 dpm/100 L at 29°15'N, 57°30'W (Bowen and Sugihara, in prep.). Clearly, in the case of these 1961 samples, the possible east-west gradient suggested above for the 1957 samples was not evident.

The mean surface value for 1961, 11.2 dpm/100 L (Table I), is reduced from that inferred for 1960 in the same area (about 15 dpm/100 L). Yet the values for 100 m and below are uniformly less than half of the surface value, even when, as in a comparison of 100-m water at 11°01'N, 29°59'W with surface water at 5°15'N, the salinities are quite similar. Development of these three profiles from those found at the equator in 1958 requires only that the
rate of fall-out delivery observed on land be reduced (see p. 129) and that vertical mixing in this section of the North Atlantic be much more vigorous below about 60 to 70 m than across this level. As we shall demonstrate in a subsequent report, a comparable change in shape of vertical profiles was not seen in the Sargasso Sea over the same time interval. For the equatorial section from 0° to 14°N, as discussed above, the density distribution strongly inhibits mixing downward from the shallow isothermal layer; in the stations referred to in footnote 2, the isotherm that is 10°C below the surface temperature is about 65 m deep at the equator, about 60 m at 2°39’N, about 75 m at 5°15’N, about 90 m at 8°05’N, and about 90 m at 11°01’N. In contrast, at 19°18’N the comparable isotherm lies below 200 m, and at 29°15’N, below 600 m.

Both the dilution of equatorial surface water and the changes in specific activity of deeper layers should be related to the southward flow of Wüst’s “North Atlantic Deep Current” (1958). This flow, at maximum speed, is calculated to reach 15 cm/sec just north of the equator at depths from 1400 to 4000 m. Unless the net southward flow is the resultant, or is at least accompanied by randomly directed movements of much greater velocity, producing both horizontal and vertical mixing in this water mass, we would expect the “age” differences, both along its axis and vertically, to be expressed in measurable differences in Sr90 specific activity. It is evident (Table III) that, at 500 m and below, no systematic trend in Sr90 exists in either direction.

Comparison With Other Analyses. Recently, Russian investigators have reported analyses of Sr90 in Atlantic Ocean samples collected on cruises of the R.V. Lomonosov during 1959 and 1961 (Shvedov et al. 1964a). Their samples include some from the surface that are close in both space and time to the surface samples discussed above; also, their data include, for 1961, a small number of shallow vertical profiles. A comparison of their results with ours is of considerable interest.

In October 1959 the Russians obtained one sample from the equatorial area, from 15°42’N, 23°39’W, in the middle of the Cape Verde Islands, close to the origin of the North Equatorial Current; for this area they have given the Sr90 concentration as 14 dpm/100 L. This agrees amazingly well with our early 1960 estimate of 15 dpm for this current, arrived at by extrapolation of analyses of samples taken much farther west.

As might be expected in an extensive series of analyses, the agreement among the 1961 samples analyzed by the Russians is by no means so satisfactory. Such differences as do appear must be interpreted in the light of the qualifying statement that accompanies the report (Shvedov et al. 1964a: 31): “in the worst case, the error was a factor of two, and in the best case, the results of two determinations are practically in good agreement with each other.”
Of eight Russian analyses of surface samples from south of the equator, \(00^\circ 21' S\) to \(25^\circ 16' S\), two are high (11, 13 dpm) and six are low (3, 6, 6, 6, 7 dpm); of the six dpm values, three are replicated samples to which we believe somewhat closer confidence limits should be applied. The localities for the two high values are very close together, at \(16^\circ 25' S, 30^\circ 04' W\) and \(17^\circ 17' S, 27^\circ 13' W\), and aligned in a NW-to-SE direction; these positions do not seem to correspond to any major current boundary, lying well within the western margin of the great South Atlantic Gyre.

In a bathythermogram section made on Cruise 247 by R.V. ATLANTIS in March 1959, approximately along \(29^\circ W\) from \(16^\circ S\) to \(20^\circ S\), there seems to be evidence of a breakdown of the thermocline at \(16^\circ 30' S, 28^\circ 24' W\); it is possible, therefore, that a minor and local hydrographic perturbation had indeed brought to this area, or retained in it, higher concentrations of soluble fall-out isotopes.

The "low" Russian values agree well with the only other reported surface value for this area and time (Rocco and Broecker 1963)—5.5 dpm \(\text{Sr}^{90}/100\text{L}\) at \(21^\circ 49' S, 35^\circ 93' W\) in July 1961. These values, compared with ours for 1957 (Table I), indicate that, from 1957 to 1961, south of the equator, there was an increase in surface \(\text{Sr}^{90}\) of less than a factor of 2. This appears to be reasonable in the light of the estimated rates of addition and of the speeds of down-mixing we have observed elsewhere. As we argued above, the high concentrations observed in 1958 do indeed appear to have been transitory.

The very low value (3 dpm) observed by the Russians just south of the equator in the Gulf of Guinea (\(00^\circ 21' S, 4^\circ 26' W\)) may well be real. This value should have been close to, or just at, the southern edge of the South Equatorial Current, where considerable upwelling of low-concentration water might be expected. That this was indeed the case is indicated by their low salinity (34.19\%\text{oo}), which must have been balanced by a low temperature, since they reported the density to be the same as that observed by them farther west on the equator, where salinities were between 35.87\%\text{oo} and 36.19\%\text{oo}. On this basis, the Russian value compares well with our 3.9 dpm (Table III), but rather less well with their own 100-m value (7 dpm) at \(00^\circ 17' N, 20^\circ 19' W\).

Salinity comparison, however, shows that their 100-m sample represents water that is somewhat closer to the core of the Undercurrent than the water in our 100-m sample.

As discussed above and shown in Table I and Fig. 1, surface samples obtained by us in the spring of 1961 showed essentially uniform \(\text{Sr}^{90}\) from the equator to \(20^\circ N\). Of seven values reported by the Russians for the same area and time (Shvedov et al. 1964a), two are lower than ours (6, 6), two agree well (10, 13) and three are perhaps significantly higher (15, 15, 16). One of the low values, for \(6^\circ 20' N, 30^\circ 06' W\), may well lie close to the boundary between the South Equatorial Current and Equatorial Counter Current (see Neumann 1960); the very shallow thermocline reported by them (45 m) may indicate this. Conventional interpretation (von Arx 1962: 200), however,
indicates that this boundary is a region of convergent sinking, and thus offers no hydrographic support for a lower Sr\(^{90}\) concentration than elsewhere.

Of the three shallow vertical profiles included by the Russians in their report, two consist of samples from 0, 30, 50, and 100 m. The profile at the equator (\(00^\circ 17'N, 20^\circ 19'W, \) April 1961) reveals concentrations of 13, 16, 17, and 7 dpm Sr\(^{90}/100\)L, respectively. We are unable to judge whether this apparent higher concentration in the core of the Undercurrent may be real; the 50-m sample exhibits the high salinity that was found to be characteristic of the Undercurrent core by Metcalf (1962, and in prep.). Our 100-m value (3.9 dpm Sr\(^{90}\), at salinity 35.526\(^{\circ}/oo\)) could be taken to indicate that the 7-dpm value at salinity 35.84\(^{\circ}/oo\) represents a reasonable mixture of the 60-m water (and Sr\(^{90}\) concentration) with water of lower salinities beneath the Undercurrent axis. It is a source of some regret that our shallow samples at this station were not more closely spaced.

A second profile by the Russians (\(20^\circ 01'S, 30^\circ 03'W, \) May 1961) gave values of 6, 7, 8, and 7 dpm Sr\(^{90}\) (depths as above); the value of 6 for the surface concentration represents two determinations—of 5 and 7. It is not possible to argue that this profile indicates a vertical change in the Sr\(^{90}\) concentration. In contrast, Rocco and Broecker (1963) have reported that, for \(21^\circ 49'S, 35^\circ 43'W\) in March, they obtained a concentration for 100 m that was twice that for the surface, and a concentration for 200 m that was the same as at surface; unfortunately, they have given no hydrographic information with their analyses. At the Russian station, salinity was uniform in the upper 50 m and had dropped by 0.46\(^{\circ}/oo\) at 100 m; the upper 100 m was reported as having been isopycnal. This is not in serious disagreement with the ATLANTIS 247 bathythermograms in this area (Fuglister 1960: 161), although most of the BTs show the isothermal depth to have been less than 50 m in March 1959. The Russian report that the thermocline depth was 85 m at this station is in disagreement with all available data, including their own salinity-vs.-density data. Uniform Sr\(^{90}\) values to 100 m or deeper are what would be expected in this area, by comparison with our profiles in the North Atlantic Gyre (to be published); also, see p. 132. No explanation seems available for Rocco and Broecker’s high 100-m value, unless we interpret it as an expression of the same sort of situation as that seen in our 1960 station at \(16^\circ 12'N, 57^\circ 30'W\); a reasonable increase in the interval between the sampling time at the surface and at 100 m would have given a curve of the shape found by Rocco and Broecker. A similar situation was reported without comment for our station at \(34^\circ 30'N, 65^\circ 42'W, \) November 1958 (Bowen and Sugihara 1960).

The third Russian vertical profile (\(11^\circ 01'N, 29^\circ 56'W, \) June 1961) consists of samples at 0, 30, and 100 m. The concentrations reported for Sr\(^{90}\)—16, 9, and 7, respectively—are higher than those we found (Table III), but perhaps not outside of the uncertainties; their 100-m concentration is 44\(^{\circ}/oo\) of the surface value, compared with 41\(^{\circ}/oo\) for our station.
It should be noted that we have intentionally omitted discussion of the various studies of fall-out isotopes in the waters of the equatorial Pacific. We believe that introduction to the Pacific has been so complicated by varying amounts of close-in fall-out and by direct introduction in surface and under-water bursts, that oceanographic interpretation of distribution patterns has to be on other bases than those that apply to the Atlantic.

**Integrated Fall-out Deliveries.** We have emphasized previously that rough integration of vertical profiles of Sr\(^{90}\) concentrations in the Atlantic indicates that considerably higher amounts of fall-out have been delivered to the sea surface (Bowen and Sugihara 1960, 1963) than to the land surface in comparable latitudinal bands. Of the stations included in the present report, those in Table II have been used in previously reported calculations. Only four of the stations in Table III are complete enough to justify integration.

In making the calculations tabulated in Table VI, we have assumed that sharp inflections of the Sr\(^{90}\) curve will usually occur only with thermal or kinetic gradients, gradual inflections occurring elsewhere, and that, in the absence of positive analytical indications, we cannot assume the presence of Sr\(^{90}\) below 1000 m. In Table VI, the figures in parentheses in the fourth column (5°15'N) are those derived by extrapolating to comparable depths the 1500-m and 2500-m concentrations for the two neighboring stations; we think this is the preferable way to use the available information. The curve for the station at 15°32'N has been extrapolated to 1000 m by comparison with similar curves for the Sargasso Sea (Bowen and Sugihara, in prep.).

**TABLE VI.**

<table>
<thead>
<tr>
<th>Position</th>
<th>15°32'N</th>
<th>00°13'S</th>
<th>5°15'N</th>
<th>11°01'N</th>
</tr>
</thead>
<tbody>
<tr>
<td>Year</td>
<td>1960</td>
<td>1961</td>
<td>1961</td>
<td>1961</td>
</tr>
<tr>
<td>Month</td>
<td>II</td>
<td>IV</td>
<td>IV</td>
<td>IV</td>
</tr>
<tr>
<td>Max. depth analyzed</td>
<td>700</td>
<td>2500</td>
<td>1000</td>
<td>2500</td>
</tr>
<tr>
<td>Integrated delivery mc/mi(^2)</td>
<td>80</td>
<td>110</td>
<td>49(105)</td>
<td>125</td>
</tr>
<tr>
<td>(10^3) dpm/m(^2)</td>
<td>66</td>
<td>96</td>
<td>42(90)</td>
<td>106</td>
</tr>
</tbody>
</table>

The available data on soil Sr\(^{90}\) (Hardy et al. 1962) show that the land delivery averaged about 13 mc/mi\(^2\) at the end of 1960, over the latitudinal band 0° to 15°N; the increment in the first quarter of 1961 for ocean Sr\(^{90}\) was much less than the analytical uncertainty of the samples represented, so we feel justified in comparing with this the data for our 1961 stations. The average for the three ocean stations, using what we take to be the best estimates, is 113 mc/mi\(^2\). This “ocean excess” is in much the same proportion to the land delivery as was found for the same area in late 1958 (Bowen and Sugihara 1963), adjusting the integrations presented then to the assumptions made above. Both on land and in the ocean, the equatorial Sr\(^{90}\) accumulation appears
to have slightly more than doubled between late 1958 and early 1961; in contrast, the accumulation at New York City (p. 129) appears to have increased by only about 50% in the same period. In sharp contrast is the report by Shvedov et al. (1964b) that Sr99 in surface waters of the Black Sea increased by four times from the third quarter of 1959 to the third quarter of 1960, and diminished only slightly by the middle of 1961.

No meteorological basis for higher rates of fall-out delivery over the ocean than over the land has yet been accepted. We intend to discuss this question at more length in a forthcoming report on data that appear to bear directly on the question of the mechanism of delivery of fall-out to the ocean.

**Lanthanides.** In an earlier report (Sugihara and Bowen 1962), summarizing analyses for Ce144 and Pm147, we laid the basis for interpretation of these data in terms of movement of the lanthanides, largely by association with the surfaces of sinking particles. In Tables IV and V are presented vertical profiles of these isotopes based on the seven stations that have since then been completed for the Atlantic area under discussion here. Comparable profiles for the 1957 stations whose Sr99 is presented in Table II were included in the earlier report (Sugihara and Bowen 1962: table II). Surface-water analyses of these isotopes, some of which have previously been discussed, appear in Table I.

Recent discussions of our 1962 paper (Osterberg et al. 1963, Sanengen, personal communication) have shown that we were insufficiently specific in one aspect of the discussion. We believed that the sinking rates deduced for lanthanide-labeled particles represented only those populations still observable in the upper parts of the water column. Unfortunately, in the statement “the content in the water columns [does not] appear to correspond to the total that might have been added,” it was left implicit that appreciable fractions of the labeled material must have sunk so rapidly as not to appear in our samples at all. We wish to state specifically here (as in our 1963 discussion) that we believe that a large fraction of lanthanide fall-out to the open ocean has left the water column; in fact, we are now measuring these isotopes in sediment samples.

The data in Tables IV and V illustrate several of the sort of secondary maxima on which our earlier estimates of sinking rates were based. The discussion of the Sr99 data above should make it clear that we are not in position to make any firm estimates of the time or place of delivery of fall-out observed in the vicinity of the equator. The peaks at 700 m at 15° 32'N in 1960 and at 1000 m at 0°13'S in 1961 fit reasonably well with the fact that 1959 (see p. 129) was the year of maximum rate of delivery to the northern hemisphere; thus the previously estimated slow sinking rate of about 1000 m in two-and-a-half years seems to be applicable in the case of these two peaks.

The very high concentration of lanthanides at 100 m, at 0°13'S in 1961, represents a new phenomenon, which will be discussed much more extensively
in a forthcoming report on our fall-out measurements between 20°N and 40°N. In 1961, prior to the end of the moratorium on atmospheric testing, shallow samples from the Atlantic frequently contained such high lanthanide concentrations as to indicate delivery during 1958 or 1959—periods of very high rates of fall-out delivery. On the other hand, the very shallow depth at which these concentrations were found indicates that the lanthanides, after introduction, can have had no significant vertical velocity other than that due to water movement. In fact, as is well exemplified by the low Sr90 in the water at 100 m at the equator in 1961, often the soluble fall-out materials in these cases seem to have left the “insolubles” behind.

Thus far no concomitant data are available for these samples to indicate where an explanation of the phenomenon is to be found. As was pointed out earlier, we do not know of any geochemical parameter that systematically resists both the physical movement of the medium and the pull of gravity, except for the active movement of organisms (Ketchum and Bowen 1958). The early reports of association of fall-out lanthanide radioactivity with organisms have often proved to be based on bad radiochemistry; in other cases, as summarized by Lowman (1960, 1963), this has seemed to be an association of fresh fall-out only, transient either as a result of surface-cleansing by the organisms, or of population turnover.

As a basis for the insolubility of diatom frustules in seawater, Lewin (1961) has reported the association of trivalent cations with the hydroxyl end-groups of the opaline silica of these structures. Her experiments showed that many of the lanthanide elements could enter linkages of this sort. Freiling and Ballou (1962) have reported that as much as 35\% of the fission-product lanthanide radioactivity could be found in filtrable, even dialyzable, form at some intervals following production and passage into seawater. Thus a reasonable argument can be developed that the lanthanide fraction of a fall-out event could be accumulated in the skeletons of a diatom population, and be held in suspension so long as grazing proceeded at a minimum rate.

The same chemical processes postulated earlier (Sugihara and Bowen 1962) for the discrimination of Ce vs. Pm in adsorption to sinking particles could operate to reduce the Ce144: Pm147 ratio in lanthanides associated with diatom silica; such discrimination did indeed occur in 1961 at the equator in the mixture at 100 m; the ratio observed is 0.36, although the ratio Ce144: Pm147 at 1000 days postproduction approximates 1 (Hallden et al. 1962). This is, in fact, the lowest Ce144: Pm147 ratio found in our 1961 equatorial samples, with the exception of the 700-m sample at 5°15′N, where the ratio was 0.2.

It may well be no more than coincidence that the Ce: Pm discrimination seen in the 1960 700-m sample from 15°32′N, if adjusted for radioactive decay since 1958, is identical to that seen in the 1961 100-m sample from the equator. Such large discrimination factors do not appear to be reflected in the high Ce: Pm ratios in the 1961 surface water, as we observed in earlier years.
The very high 1960 Ce$^{44}$ concentration at 100 m at 15° 32'N, coupled with the observed ratio Ce: Pm = 4.8, offers one possible location of the Ce$^{44}$ excess. The 1961 surface ratios (Table I) range from a low of 0.48 to 0.78, 1, 1.5, and 2—probably not an average differing significantly from the 1 to be expected simply from production in 1958 followed by radioactive decay. We may, however, take the water column at 11° 01'N (Tables IV and V) to be typical of ocean areas not exhibiting either sinking-particle populations or diatom retention of lanthanide radioactivities; of the seven depths represented, five show ratios above 1—1.6, 1.5, 1.5, 4, and 1.5; these values, when integrated over the 2500-m depth of water represented, account for a very considerable residual excess of Ce$^{44}$ over Pm$^{47}$.

Conclusions. In 1957, surface concentrations of Sr$^{90}$ were uniform in the Atlantic Ocean from 16°S to 16°N; only two of six samples from 1000 m and none from below this depth yielded detectable Sr$^{90}$.

In 1958, surface Sr$^{90}$ concentrations had increased markedly and uniformly in the western Atlantic from 24°S to 8°N; no increase was seen in the eastern basin at 24°S or at the equator. Four samples, from the equator at depths of 700 m to 2000 m, showed readily detectable Sr$^{90}$ at a concentration that was about 60% higher than that in the positive samples of 1957. At the equator, surface concentration levels extended to 300-m depth, but not to 700 m.

The 1960 surface samples from the eastern Caribbean and western Atlantic at about 15°N showed Sr$^{90}$ concentrations that were approximately twice the concentrations found in 1958.

In 1961, surface Sr$^{90}$ concentrations were uniform from the equator to 20°N; in vertical profiles taken at the equator, at 5° 15'N and at 11°N, the concentration at 100 m was less than half of the surface concentration, with little further reduction in Sr$^{90}$ below 100 m, even at depths of 2500 m.

These concentration distributions bear no resemblance to patterns of fallout delivery on land; it is concluded that hydrographic processes are responsible for seawater concentration patterns. The data provide evidence for an otherwise unobserved high fallout incident in September 1958 in the southern hemisphere, probably between 10° and 40°S, west of 30°W. Another very high fallout incident in the North Atlantic, between 10° and 20°N and east of 55°W, is indicated as having occurred in late 1959.

The rapid vertical penetration of Sr$^{90}$ in the ocean and the relatively rapid attainment of horizontal and vertical uniformity of concentration in the equatorial Atlantic are consistent with the patterns of current flow (derived from hydrographic considerations), only if these are complicated by very active, unoriented mixing processes within water masses.

In general, reasonably good agreement is found with recently published Russian Sr$^{90}$ analyses for the equatorial Atlantic.
Integration of four vertical profiles—one from 1960 and three from 1961—yields estimates of fall-out delivery to the equatorial ocean surface that are much higher than estimates of delivery to land surfaces at comparable latitudes.

Vertical profiles of Ce\textsuperscript{144} and Pm\textsuperscript{147} show, as previously observed, secondary maxima at various depths; some of these maxima are interpreted as being local concentrations of sinking particles whereas others appear to represent retention in the bodies of plankton swarms. In most cases these secondary maxima are enriched in Pm\textsuperscript{147} vs. Ce\textsuperscript{144}.

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