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Cesium-137 in the North Atlantic Measured by Selective Absorption in Situ

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ABSTRACT

By means of selective absorption in situ, Cesium-137 was measured in November and December 1968 in two depth profiles (near 39°N, 69°W) and in ten widely scattered surface samples in the North Atlantic Ocean. The surface concentrations of Cs-137 are compatible with the surface concentrations of Sr-90 obtained by other workers in the same latitudinal range. The Cs-137 concentrations in the depth profiles, which reflect differences in the T-S structure of the water, fall off toward the main thermocline.

If it is assumed that the total amount of Cs-137 in the profiles is characteristic of the Cs-137 activity per unit surface area in the layers above the main thermocline, then a crude calculation of the residence time of Cs-137 (and presumably of water) above the main thermocline leads to times of 2.5 to 7 years; these times are in reasonable agreement with those inferred from the results of other workers.

Introduction. It has been inferred that the long half-life fallout radionuclides, Sr-90 and Cs-137, move with the water once they have entered the sea (Bowen and Sugihara 1965, Broecker 1966, Folsom and Sreekumaran 1966), and attempts have been made to use these radionuclides as tracers to follow the movement and mixing of water masses.

A method for determining the concentration of Cs-137 in seawater by selective absorption in situ (Folsom and Sreekumaran 1966) has been used extensively in the Pacific Ocean to determine the concentration of Cs-137 in surface samples and in vertical profiles down to depths of several hundred meters (Folsom et al. 1968, Folsom et al. 1970). The absorbers pick up both Cs-137 and stable cesium. The volume of water sampled is determined indirectly by measuring the amount of stable cesium absorbed. Since the ratio of stable cesium to salinity in the oceans is nearly constant (Folsom et al. 1964, Bolter et al. 1964), the volume of Cs-137 sampled may be calculated.

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This method has been used in the North Atlantic to measure Cs-137 in two vertical profiles and in ten surface samples collected on two cruises (in November and December 1968). It is believed that these observations represent the first time this technique has been used to obtain Cs-137 profiles in the Atlantic Ocean.

Methods. Open subsurface samplers similar to those of Folsom and Sreekumaran (1966) were used in the vertical profiles. Each sampler held a flat cylindrical cartridge (7.6-cm diameter) filled with about 25 g of copper ferrocyanide granules. The granules were held in the cartridge by a fine-mesh nylon netting. The samplers were attached to the hydrowire at intervals of 40 to 60 m with the axes of the cylindrical cartridges vertical; thus the roll of the ship caused water to flow through the packed mass of absorber granules. The granules were exposed to the water for several hours. Five samplers were used simultaneously at each station. The depths of the samplers were determined from the wire length and the average wire angle; the depths given in Table I are believed to be accurate to $\pm 5\%$. A pressure recorder used on the second profile, D-2, confirmed this estimate when it showed that the depth of the deepest sampler did not vary by more than $\pm 5$ m during the time the samplers were at depth.

The surface sampler, containing a flat cylindrical cartridge (2.5-cm diameter) with about 5 grams of copper ferrocyanide granules, was towed 90 m behind the ship. Again, the granules were exposed to the seawater for periods of several hours. Following exposure, the granules were returned to the laboratory and processed chemically to extract and concentrate the absorbed cesium (Folsom and Sreekumaran 1966). Cs-137 was determined by gamma-ray spectrometry, the stable cesium by flame photometry.

Since water passed through the subsurface samplers while they were being lowered and raised, it was necessary to make a correction for the cesium absorbed by each sampler during transit. This was done by making a calibration cast. Immediately before each profile cast, one or two closely spaced samplers were lowered to the same depth as the deepest sampler in the profile and then immediately returned to the surface. The amount of cesium picked up by these samplers was used in determining the corrections for the profile samplers. In Table I, the volumes sampled and the Cs-137 concentrations listed for the vertical profiles have all been corrected in this way. For a given cast, the size of the correction increased with the depth of the sampler. The largest correction amounted to $7\%$ of the Cs-137 concentration listed in Table I for the deepest sampler of cast D-2.

Results. The data obtained from these samples are listed in Table I. The volumes sampled (column 6) were calculated by using an average concentration
Table I. Cs-137 concentrations at the surface and at depth; Cruises TR-058 and TR-064.

<table>
<thead>
<tr>
<th>Date (1968)</th>
<th>Actual depth (m)</th>
<th>Location at start of sampling</th>
<th>Sampling time (hrs)</th>
<th>Ship speed (kn)</th>
<th>Volume sampled (l)</th>
<th>Conc. Cs-137 (dpm/l)</th>
<th>Salinity (%o)</th>
<th>Temp. (°C)</th>
</tr>
</thead>
<tbody>
<tr>
<td>11 Nov</td>
<td>surface</td>
<td>41°06'N, 70°53'W</td>
<td>5</td>
<td>11</td>
<td>26</td>
<td>.26 ± .04</td>
<td>32.30±</td>
<td>11</td>
</tr>
<tr>
<td>11 Nov</td>
<td>surface</td>
<td>40°37'N, 70°21'W</td>
<td>5</td>
<td>8</td>
<td>21</td>
<td>.27 ± .04</td>
<td>33.00±</td>
<td>12</td>
</tr>
<tr>
<td>12 Nov</td>
<td>surface</td>
<td>39°41'N, 69°56'W</td>
<td>3</td>
<td>7</td>
<td>30</td>
<td>.28 ± .03</td>
<td>34.50±</td>
<td>15</td>
</tr>
<tr>
<td>14 Nov</td>
<td>surface</td>
<td>40°40'N, 71°51'W</td>
<td>3.3</td>
<td>9</td>
<td>14</td>
<td>.29 ± .12</td>
<td>32.50±</td>
<td>11</td>
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<tr>
<td>14 Dec</td>
<td>surface</td>
<td>32°06'N, 64°49'W</td>
<td>6</td>
<td>10</td>
<td>51</td>
<td>.37 ± .04</td>
<td>36.65±</td>
<td>19</td>
</tr>
<tr>
<td>15 Dec</td>
<td>surface</td>
<td>31°43'N, 69°04'W</td>
<td>6</td>
<td>5</td>
<td>62</td>
<td>.42 ± .01</td>
<td>36.55±</td>
<td>19</td>
</tr>
<tr>
<td>17 Dec</td>
<td>surface</td>
<td>32°07'N, 73°17'W</td>
<td>6</td>
<td>9</td>
<td>54</td>
<td>.40 ± .01</td>
<td>36.50±</td>
<td>19</td>
</tr>
<tr>
<td>17 Dec</td>
<td>surface</td>
<td>33°05'N, 74°32'W</td>
<td>6</td>
<td>10</td>
<td>56</td>
<td>.36 ± .01</td>
<td>36.50±</td>
<td>20</td>
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<tr>
<td>18 Dec</td>
<td>surface</td>
<td>36°07'N, 72°26'W</td>
<td>6</td>
<td>10</td>
<td>54</td>
<td>.34 ± .01</td>
<td>36.05±</td>
<td>20</td>
</tr>
<tr>
<td>20 Dec</td>
<td>surface</td>
<td>39°09'N, 69°13'W</td>
<td>5</td>
<td>6</td>
<td>33</td>
<td>.37 ± .03</td>
<td>35.50±</td>
<td>13</td>
</tr>
<tr>
<td>19 Dec</td>
<td>cast D-1</td>
<td>39°10'N, 69°13'W</td>
<td>7</td>
<td>0</td>
<td></td>
<td></td>
<td></td>
<td></td>
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<tr>
<td></td>
<td></td>
<td></td>
<td>10</td>
<td>7</td>
<td>47</td>
<td>.36 ± .02</td>
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<td></td>
<td>50</td>
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<td>.39 ± .02</td>
<td>35.10</td>
<td>14.5</td>
</tr>
<tr>
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<td></td>
<td></td>
<td>100</td>
<td>7</td>
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<td>.37 ± .02</td>
<td>35.50±</td>
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<td>.27 ± .02</td>
<td>35.92</td>
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<td></td>
<td></td>
<td>201</td>
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<td>49</td>
<td>.15 ± .01</td>
<td>35.70</td>
<td>13.0</td>
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<tr>
<td>20 Dec</td>
<td>cast D-2</td>
<td>39°29'N, 69°30'W</td>
<td>4.5</td>
<td>1.5*</td>
<td>32</td>
<td>.37 ± .02</td>
<td>35.50±</td>
<td>12.5</td>
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<tr>
<td></td>
<td></td>
<td></td>
<td>9</td>
<td>4.5</td>
<td>32</td>
<td>.37 ± .02</td>
<td>35.50±</td>
<td>12.5</td>
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<td></td>
<td>46</td>
<td>4.5</td>
<td>42</td>
<td>.30 ± .01</td>
<td>35.54</td>
<td>13.5</td>
</tr>
<tr>
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<td></td>
<td></td>
<td>82</td>
<td>4.5</td>
<td>44</td>
<td>.31 ± .01</td>
<td>35.60±</td>
<td>15.0</td>
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<td>119</td>
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<td>39</td>
<td>.14 ± .01</td>
<td>35.77</td>
<td>13.5</td>
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</tbody>
</table>

* 25-kn. wind; based on Loran fixes, it has been estimated that the ship drifted westward at about 1.5 kn.

± Value estimated from Oceanographic Atlas of the North Atlantic Ocean (U.S. Naval Oceanographic Office, 1967: fig. II-57), "mean surface salinity, October through December.”

† Value obtained by linear interpolation of measured salinity values at the depths of the samplers above and below the sample listed.

†† November surface-temperature values were estimated from Oceanographic Atlas of the North Atlantic Ocean (U.S. Naval Oceanographic Office, 1967: fig. II-11), “mean surface temperature, November.” December surface-temperature values were scaled from XBT’s obtained during Cruise TR-064.
of stable cesium in North Atlantic water adjusted to the salinity at the sampling site. An average concentration of stable cesium was obtained by using the ten North Atlantic cesium values (adjusted to a common salinity) listed in the two most recently published sets of measurements of stable cesium concentration (Folsom et al. 1964, Bolter et al. 1964). This average was 0.346 µg Cs/l at a salinity of 35.00°/oo. Only a limited number of salinities were obtained at the sampling sites. Therefore, interpolated values, or values obtained from the Oceanographic Atlas of the North Atlantic Ocean (U.S. Naval Oceanographic Office 1967), were used in the absence of measured values. The salinity values used are listed in Table I.

The uncertainties listed in Table I were determined primarily by taking one standard deviation of the Cs-137 counting variation. For the first set of samples processed at the laboratory (those of November 1968), additional uncertainties were included due to problems in the processing. Uncertainties arising from the use of estimated salinity values are small with respect to the counting uncertainties.

In addition to the uncertainties listed in Table I, there is a ± 6% apparent variation in the cesium-salinity ratio in North Atlantic waters and a ± 14% uncertainty in the average value of the ratio; the second of these uncertainties derives from a systematic difference in the values obtained at two laboratories (Folsom et al. 1964, Bolter et al. 1964).

A recent review (Folsom, personal communication) of the flame photometric methods used by Folsom et al. (1964) strongly suggests that the earlier analyses were imprecise, with contaminated reagents giving too high a concentration. Improvements in this regard (Folsom, personal communication) have led to average North Pacific concentrations that are about 20% lower than those reported earlier (Folsom et al. 1964). This suggests that the North Atlantic cesium concentrations may be much more uniform than were indicated by the two 1964 reports. Most likely the North Atlantic average lies close to 0.29 ± .01 µg Cs/l, as reported by Bolter et al. (1964). Thus the Cs-137 concentrations in Table I may be biased too high by the ratio 0.35/0.29 or by a factor of 1.20. More analyses of cesium in the North Atlantic are needed to settle this point. One other set of measurements (Robertson et al. 1968), obtained by direct counting of activated samples without chemical treatment, gave an average of 0.261 µg Cs/l (at salinity = 35°/oo) for 18 surface samples collected in the North Atlantic during July and August 1968 between 9°N and 26°N. Careful replication tests have suggested that the results obtained with this type of analysis and with the most recent flame techniques can be brought into very good agreement (Folsom, personal communication). On the basis of recent measurements, Folsom has suggested that the variation in the concentration of stable cesium in the open Pacific Ocean, when normalized to constant salinity, may be less than ± 5%. Similar uniformity might be expected in other open-ocean areas.
Discussion of Results. The Cs-137 concentrations obtained from the shallowest absorbers of casts D-1 and D-2, using 25-g cartridges, and the surface sample taken on 20 December, with a 5-g cartridge, all measured within 50 miles of each other at approximately the same time, are the same, to the precision of the measurements. The six most precise surface values, all taken in December between 30° and 40°N, are in reasonable agreement with the Bowen et al. (1968) average of five Sr-90 surface measurements made during the first half of 1967 in the same latitudinal range. For a comparison of these two averages, the Sr-90 average has been multiplied by 1.26, the most recently measured value for the ratio of the concentration of Cs-137 to Sr-90 in the Atlantic (Bowen and Noshkin, personal communication); this result was then multiplied by 0.70 to correct for the decrease in the Sr-90 activity in the surface waters of the North Atlantic between the first quarter of 1967 and the end of 1968. [The factor 0.70 was obtained by extrapolating the decrease observed between 1964 and 1967 (Bowen et al. 1969) to the end of 1968.] The average value for the six Cs-137 concentrations is 0.38 dpm/l (± 7%) while the value for Sr-90, corrected to Cs-137, is 0.32 dpm/l (± 8%). The variations given with these average values are the average deviations (of the values averaged) from their mean value. They represent not only the true variation of the radio-isotope concentrations in surface waters in the 30°-40°N range but the experimental uncertainties as well. The agreement between the Cs-137 and Sr-90 results would be improved if the Cs-137 results were reduced by 20%, as suggested above in discussions of the recent flame results for stable cesium.

Of the five values for the concentration of Sr-90 listed by Bowen et al., four were obtained at Ocean Weather Station E (35°N 48°W); the six values given here were all obtained west of Bermuda (32°N 65°W). A comparison of all the 30°-40°N values listed by Bowen et al. (1968: table 2) for samples obtained since 1963 showed no appreciable difference between the averages of samples taken west of Bermuda and of samples obtained at Station E.

In Fig. 1, the Cs-137 concentrations of the vertical profiles are plotted against depth. The values have been arbitrarily connected by straight lines. The salinity values and temperature profiles obtained at the same time are also shown. The temperatures and salinities show that the structure of the water at the locations of profiles D-1 and D-2 was different, although the two profiles were taken within 30 miles of each other, on successive days. The Cs-137 concentrations appear to reflect these differences. Comparing the two profiles, the three samples at intermediate depth showed greatest differences in temperature and salinity and also the greatest differences in Cs-137. The deepest sample in each profile where the salinities and temperatures were similar had similar Cs-137 concentrations, as did the shallowest samples, in the isothermal layer. Note also that the Cs-137 concentration diminished toward the main thermocline. This behavior is in accord with the principle
that the increased stability at the thermocline acts as a barrier to mixing. Similar behavior at the thermocline was observed in Cs-137 profiles taken in the Pacific, south of 33°N (Folsom et al. 1970).

**General Discussion.** The total amount of Cs-137 activity per unit surface area in the near-surface region may be estimated by integrating the area to the left of the profiles in Fig. 1 after extending the line through the two lowest points in each profile to zero concentration of Cs-137. By employing the assumption that this activity per unit surface area is characteristic of the surface region in general, a crude estimate of the residence time of Cs-137 (and presumably of the water in the surface layer) may be obtained from the total amount of global fallout deposited in the northern hemisphere. Perhaps the simplest way to make this estimate is as follows: first assume that all the northern hemispheric fallout since 1961 (Volchok 1969) came down in 1963; second, assume that about 3.5 megacuries of Sr-90 fell on land in the northern hemisphere between 1961 and 1968 (estimated from Volchok 1969); third, assume that the ratio of Cs-137 to Sr-90 in this fallout was about 1.26 (Bowen and Noshkin, personal communication). Using these assumptions, the half-depletion-time of cesium in the upper layers of the North Atlantic approximates 7, 5, or 2.5 years, depending respectively upon the choice of 1, 1.5, or 3 as the ratio of the rate of fallout over the North Atlantic to the rate of fall-

3. In 1963, the fallout rate, from the largest series of tests by far, reached a peak with about 2.5 year full width at half maximum.
out over land. The probable ratios of fallout over the North Atlantic to fallout over land have been discussed by Bowen et al. (1969). The times obtained here are in reasonable agreement with the times that may be inferred from box-model calculations (Montgomery 1959, Broecker 1966) and from the decrease with time of the average surface concentration of Sr-90 (Bowen et al. 1969: fig. 1), mentioned above.

Although the above arguments can be defended, no great trust can be placed in a residence time based on two profiles obtained close together in one area of the ocean. However, the calculation does indicate that a series of Cs-137 profiles would make it possible to obtain a fairly direct and accurate estimate of the residence time of water in the surface layer if it were possible to resolve the uncertainties in the ocean-land fallout ratio. The technique could also be used for investigating mechanisms that have been postulated to explain the loss of water from the surface layer, such as the sinking of 18° water north of Bermuda (Schroeder et al. 1959, Worthington 1959, Istoshin 1962).

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