Fallout Nuclides in Atlantic and Pacific Water Columns: GEOSECS Data

by


May 1985

Technical Report

Funding was provided by the United States Department of Energy under Contract DE-AC02-EV03563.

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Fallout Nuclides in Atlantic and Pacific Water Columns:
GEOSECS Data

by

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Robert B. Gagosian, Chairman
Department of Chemistry
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Abstract

This report contains results of measurements of the fallout radionuclides $^{90}$Sr, $^{137}$Cs, $^{239,240}$Pu, and $^{241}$Am in large volume seawater samples collected between 1972 and 1974 in the Atlantic and Pacific as part of Geochemical Ocean Sections (GEOSECS) program. The stations for which data are reported include both the North and South Atlantic oceans and latitudes north of $20^\circ$S in the Pacific Ocean. The $^{90}$Sr and $^{137}$Cs data set has been corrected by a procedure which estimates independently the analytical blank for the laboratory which made the analysis. When the data quality and spacing permit, water column inventory estimates were made for each nuclide over depth intervals appropriate to the nuclide's distribution.
Introduction

The GEOSECS (Geochemical Ocean Sections) program, which began to be funded by the National Science Foundation in 1971, was the first really large-scale, multi-ocean, multi-laboratory and multi-discipline attempt to combine high quality physical oceanographic measurements with chemical measurements of both natural and anthropogenic chemical substances, both stable and radioactive, in the world's oceans. Broadly stated, it had a goal of trying to understand the physical and biogeochemical controls which determine the properties and behavior of chemical substances in the ocean. Through 1979, the program produced five volumes of collected papers published in Earth and Planetary Science Letters (e.g. Craig and Turekian, 1980) and hydrographic data tables and atlases published by the National Science Foundation (Bainbridge et al., 1981-83). A further series of data reports and atlases devoted to the shore-based chemical measurements is planned for the near future.

This report has been prepared to tabulate the results obtained by the various laboratories of the concentrations and distributions in the Atlantic and Pacific Ocean of some of the bomb-derived fallout radionuclides introduced to the oceans subsequent to atmospheric nuclear weapons tests. This report contains data obtained on samples collected as part of the GEOSECS program in these oceans and is part of the continuing study of these substances in the world ocean and of their use as tracers to study a variety of physical and biogeochemical oceanic processes.

The radionuclides for which data are reported include the fission product radionuclides $^{137}\text{Cs}$ and $^{90}\text{Sr}$ and the transuranic elements Pu and Am – produced by neutron capture and decay chain processes. This suite of radioele-
ments encompasses a range of chemical reactivities with respect to particle scavenging — ranging from the soluble end ($^{137}$Cs and $^{90}$Sr), where the processes controlling the nuclide distributions are primarily physical, to the reactive end ($^{241}$Am), where uptake and transport on settling particles becomes an important process. Plutonium appears to lie somewhere in the middle of the range. Initially it was believed to exhibit relatively high particle reactivity but recent studies are beginning to point to a lower degree of particle reactivity than was originally postulated. As chemical analogues, $^{137}$Cs and $^{90}$Sr would be hard to distinguish from $^{3}$H, based on their observed oceanic distribution patterns, while $^{241}$Am appears likely to have a chemical reactivity approaching that of Th.

A discussion of distribution patterns of fallout radionuclide patterns in the Pacific, based primarily on GEOSECS data, provided a first large-scale review, in a single ocean, of the fate of these fallout radionuclides (Bowen et al., 1980). It was stated in this paper that the individual data upon which the paper was based would be published in a forthcoming report. Partly, then, this report is intended to complete this plan. In addition, it includes the available data set for fallout nuclides measured in Atlantic GEOSECS samples and a suite of $^{241}$Am data which were not discussed in the above paper.

One comment may be appropriate at this point in respect to the quality of the reported data, although this is discussed later. As would be expected, data quality deteriorates as ambient concentrations approach sensitivity limits. As the input of fallout has varied to the oceanic areas for which data are reported here, the uncertainties in the data, especially in the deeper samples, increase as the level of the fallout input decreases. The fallout
input decreased in the order North Pacific > North Atlantic > South Atlantic. An additional factor which resulted in the quality of the transuranic data in the Pacific samples being higher than that of the Atlantic samples is the fact that the Pacific samples were generally analyzed at a later time than were the Atlantic ones. Improved chemical recoveries, increased detector availability, and hence counting times, all worked to produce a Pacific transuranic data set of significantly higher precision than that obtained for the Atlantic samples.

**Sampling, Methods and Quality Control**

A fairly detailed account of the techniques used in collection and fallout radionuclide analyses of the large volume water samples from the Atlantic and Pacific GEOSECS cruises appeared in a paper discussing the Pacific data set (Bowen et al., 1980). This account, though directed at the samples collected on the Pacific GEOSECS cruises, is also generally relevant to the collection and analyses of Atlantic GEOSECS samples. We therefore include here an abbreviated outline of these procedures, supplemented by remarks which are relevant to the fuller data set presented in this report.

In both Atlantic and Pacific GEOSECS cruises, large volume sample collection for fallout nuclide analysis was made primarily by the 260 liter Gerard samplers. After $^{14}$C extraction, 60 liter samples were returned to shore-based laboratories for fallout nuclide analyses - in some cases, following Rn extraction for $^{226}$Ra analysis. Fallout radionuclide analyses were subsequently completed in a variety of combinations of radionuclides analyzed by a given laboratory. Four different laboratories performed or contracted the various radiochemical analyses.
(1) Some samples were analyzed by commercial laboratories, under contract to the (now) U. S. Dept. of Energy, Environmental Measurements Laboratory in New York City, NY. H. L. Volchok, of that laboratory, supervised the contracting and reporting. These analyses were restricted to measurement of \(^{90}\)Sr alone or for \(^{137}\)Cs and \(^{90}\)Sr.

(2) Some samples were analyzed by V. E. Noshkin's group at Lawrence Livermore Laboratories (LLL). Mostly \(^{137}\)Cs and Pu isotopes were determined by LLL although a few sample series were processed for \(^{137}\)Cs and \(^{90}\)Sr by a contractor laboratory after LLL isolated a Pu fraction.

(3) Some samples were analyzed by V. T. Bowen and H. D. Livingston's group at Woods Hole Oceanographic Institution (WHOI). These analyses typically included \(^{137}\)Cs, \(^{90}\)Sr, Pu isotopes and, for a subset, \(^{241}\)Am.

(4) A number of samples, from the Atlantic GEOSECS sample suite, were analyzed for Pu and \(^{241}\)Am by T. M. Beasley's laboratory at Oregon State University (OSU). A small number of these samples were, in addition, processed to isolate a Cs fraction at OSU, and then the radiochemistry and sample counting completed at WHOI.

The analytical methods used by these various laboratories have been described in detail elsewhere (if not proprietary) and are referenced in Bowen et al. (1980). The methods used by the OSU laboratory were generally similar to those used by WHOI or LLL with the exception of a solvent extraction based radiochemical procedure for \(^{241}\)Am.

Throughout the analytical program for these GEOSECS samples, a considerable analytical effort was devoted to activities which provided assurance of data quality. These efforts are again described in detail by Bowen et al.
(1980) and include the analyses of known 'blind' samples, duplicates, environmental blanks, reagent blanks and interlaboratory intercomparison exercise samples. In addition, both Atlantic and Pacific data sets have been subjected to a "deep ocean blank" procedure for blank correcting measured values for \(^{137}\)Cs and \(^{90}\)Sr obtained both by different laboratories and at different times. Details of this procedure have been reported for Atlantic samples by Kupferman and Livingston (1979) and essentially the same approach was followed for blank correction to the GEOSECS Pacific data set.

**Results**

Radiochemical and hydrographic data from GEOSECS Atlantic and Pacific stations are compiled in the Tables of this report. The first set of tables lists the data for most of the Atlantic GEOSECS large volume stations. The second set lists data from Pacific GEOSECS large volume stations north of 20°S. In some cases no samples for fallout nuclide analyses were collected at South Pacific stations, or were collected by methods which did not protect the integrity of the sample with respect to contamination during storage; some South Pacific large volume samples remain unanalyzed for reasons of lack of funding or scientific interest.

Each large volume station is identified by its GEOSECS station number preceded by the prefix GX. Hydrographic data for the large volume water samples were derived from the GEOSECS Atlantic and Pacific preliminary leg reports. Depths are best depths in meters. Salinities and potential temperatures are in conventional units; densities are listed as sigma theta. Concentrations of \(^{137}\)Cs, \(^{90}\)Sr, \(^{239,240}\)Pu and \(^{241}\)Am are tabulated where measured in units of disintegrations per 100 kg of seawater as of date of collection. One sigma
counting uncertainties are listed in the columns labelled by the letter e (error). $^{239,240}$Pu concentrations are tabulated under the abbreviated Pu239 heading but are conventionally the sum of $^{239}$Pu and $^{240}$Pu concentrations - unresolvable by state-of-the-art alpha spectrometry. In a few cases no uncertainty is listed with a value for a radionuclide. For a variety of reasons no estimate of the counting uncertainty was obtained and the value listed should be regarded as an upper limit (or a less than value) for the actual concentration in the sample.

The final column in the tables lists the laboratory code which has been used to identify the laboratory or laboratories which performed the radiochemical analyses of each large volume sample. As described in the previous section, there were a variety of combinations of radiochemical analyses used over the several years during which the lengthy analyses were completed. A listing of these various combinations and of the laboratories involved is tabulated in Table 1.

Also listed in the tables are the results of integration of the various radionuclide concentrations measured over all or part of the water column at each station. The result of this calculation is an estimate of the radionuclide inventory in the water column at each station over the depth intervals over which these calculations were made. The inventory units are expressed as mCi/km$^2$. Where possible, the integration was carried out over the complete water column, but in many cases this was not possible or appropriate. At some stations or for some radionuclides the available data were not complete enough to define the water column distribution well enough to permit a meaningful inventory calculation. The other limiting factor was frequently analytical sen-
sitivity. In the deep ocean, especially in areas where atmospheric fallout input was minimal, the concentrations at depths below the main thermocline were often essentially zero or at the limit of available analytical sensitivities. In this situation, small overestimates of nuclide concentrations in the generally large portion of the water column below the main thermocline would result in substantial and erroneous overestimates of the actual nuclide inventory. The inventory calculations listed are therefore data limited and the depth intervals over which integration was made is noted at each station and for each nuclide. Deep water inventory calculations were also susceptible to overestimation when concentrations were close to sensitivity limits because of the nature of the inventory integration calculation itself. Firstly, no consideration was given to the precision of a given concentration - only the actual concentration value being used. At low deep water concentrations, the uncertainties in the measurement often produced apparent concentrations substantially higher than was reasonable, but with very large uncertainties. When these situations occurred, inventory overestimates were likely to be non-trivial. Secondly, only positive concentration values were computed in the inventory calculation. Blank correction procedures often produced negative concentration values for some nuclides from deep water. Exclusion of these again tends to bias inventory estimates. The integration calculation itself used the measured concentration of a nuclide at a given depth to represent the average concentration over the depth interval between the mid-points of the distances to the sample depths above and below the given depth.

The radionuclides $^{90}$Sr and $^{137}$Cs have been found to exhibit oceanic chemistries dominated by highly soluble characteristics such that their dis-
tributions are essentially controlled by purely physical processes of mixing (Kupferman and Livingston, 1979). Their oceanic distributions thus are generally found to correlate strongly with those of other soluble tracers introduced from the atmosphere with comparable temporal and spatial input histories - such as tritium, or to lesser extent, freons. Except in oceanic areas which have received recent deep ventilation, the distribution of these species is for practical purposes confined to depths shallower than the bottom of the main thermocline. Accordingly, integration of $^{137}$Cs and $^{90}$Sr distributions at such stations was in general only made to a depth where their concentrations became undetectable. The distributions of the particle active transuranic elements $^{239,240}$Pu and $^{241}$Am were integrated where the data permitted throughout the whole water column.

Acknowledgements

The collection and publication of the data in this report was made possible through the efforts of a large group of people who are too numerous to list here. They include the officers, crew and science parties on research vessels Knorr and Melville and the many radiochemists in the several laboratories represented who generated the basic data set. S. L. Kupferman and D. E. Moore provided the capability of the 'deep ocean blank' correction technique applied to $^{90}$Sr and $^{137}$Cs data. Without all of this skilled help, this work would not have been possible and it is our pleasure to acknowledge it here.

Support for this work has been variously funded as follows: at WHOI, subcontracts from EML, and directly to WHOI from the U. S. Department of Energy under contract 73-C-02-3563; at LLL, under the auspices of the U. S. Depart-
ment of Energy under contract number W-7405-ENG-48. We are very grateful to these agencies and to the several program administrators for this support.

References


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L = Lawrence Livermore Laboratory
O = Oregon State University
T = Trape10 (now Environmental Analysis Laboratory)
t = Teledyne
W = Woods Hole Oceanographic Institution
* = Value reported is the mean of two measurements
SECTION 1

Radiochemical Data (GEOSECS Atlantic)
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**CS137 FROM 0 M TO 3390 M = 141.2 mCi/km²**

**SR90 FROM 0 M TO 3390 M = 103.1 mCi/km²**
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PU239 FROM 0 M TO 2392 M = 1.61 mCi/km²
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SR90 FROM 0 M TO 1500 M= 75.4 mCi/km²
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CS137 FROM 0 M TO 1000 M = 37.8 mCi/km²
SR90 FROM 0 M TO 1000 M = 24.7 mCi/km²
PU239 FROM 0 M TO 4401 M = 0.48 mCi/km²
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CS137 FROM 0 M TO 1200 M = 22.8 mCi/km²
SR90 FROM 0 M TO 1200 M = 17.2 mCi/km²
PU239 FROM 0 M TO 1200 M = 0.22 mCi/km²
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**CS137 FROM 0 M TO 500 M** = 4.6 mCi/km²

**SR90 FROM 0 M TO 500 M** = 3.8 mCi/km²
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SR90 FROM 0 M TO 500 M = 6.3 mCi/km²

Cruise-Sta# | Position | Collection Date | Bottom Depth
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62 | 56.5 W |

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CS137 FROM 0 M TO 3000 M = 112.8 mCi/ km²
SR90 FROM 0 M TO 3000 M = 71.4 mCi/ km²
PU239 FROM 0 M TO 5265 M = 1.40 mCi/ km²
## Cruise Data

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**Note:**
- CS137 from 0 M to 3000 M = 142.6 mCi/km²
- SR90 from 0 M to 3000 M = 76.9 mCi/km²
SECTION 2

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CS137 FROM 0 M TO 1000 M = 75.7 mCi/m²
SR90 FROM 0 M TO 1000 M = 54.5 mCi/m²
PU239 FROM 0 M TO 4800 M = 1.53 mCi/m²
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139 | 34.4 | W |

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| | PU239 FROM 0 M TO 5129 M | | | | | | | | |
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PU239 FROM 0 M TO 5877 M = 3.88 mCi/km²
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CRUISE-STA# | POSITION | COLLECTION DATE | BOTTOM DEPTH
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PU239 FROM 0 M TO 1000 M = 1.18 mCi/km²
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SR90 FROM 0 M TO 5603 M = 88.5 mCi/km²
PU239 FROM 0 M TO 5603 M = 3.27 mCi/km²
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Pu239 FROM 0 M TO 6061 M = 3.39 mCi/km²
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CS137 FROM 0 M TO 1000 M = 94.5 mCi/km^2
SR90 FROM 0 M TO 1000 M = 58.9 mCi/km^2
Pu239 FROM 0 M TO 5729 M = 2.95 mCi/km^2
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CS137 FROM 0 M TO 750 M= 32.4 mCi/km²
SR90 FROM 0 M TO 750 M= 22.3 mCi/km²
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CS137 FROM 0 M TO 600 M= 32.9 mCi/km²
SR90 FROM 0 M TO 600 M= 24.2 mCi/km²
PU239 FROM 0 M TO 600 M= 0.47 mCi/km²

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PU239 FROM 0 M TO 900 M = 0.26 mCi/km²

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| 118   | 34.430   | 24.200 | 24.696 | 20.5  | 1.1  | 12.0  | 0.7  | T   |
| 197   | 35.797   | 20.140 | 25.281 | 19.4  | 0.6  | 0.020 | 0.040 | Q   |
| 599   | 34.520   | 6.260  | 27.180 | 0.3   | 0.5  | 0.070 | 0.030 | Q   |
| 799   | 34.580   | 5.010  | 27.323 | 0.0   | 0.4  | -0.5  | 0.5  | Q   |
| 1047  | 34.531   | 4.000  | 27.450 | 1.3   | 0.4  | 0.2   | 0.8  | Q   |
| 1295  | 34.567   | 3.210  | 27.555 | 0.1   | 0.4  | -1.0  | 1.0  | T   |
| 1546  | 34.595   | 2.690  | 27.625 | -0.4  | 0.4  | -1.4  | 0.8  | 0.000 | 0.020 | Q   |

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| 2028  | 34.642   | 2.840  | 27.714 | 0.1   | 0.4  | 0.8   | 0.7  | 0.080 | 0.040 | Q   |
| 2277  | 34.660   | 1.810  | 27.747 | -0.6  | 0.4  | 0.4   | 0.7  | T   |
| 2527  | 34.670   | 1.660  | 27.766 | 0.9   | 0.5  | 1.9   | 1.0  | 0.000 | 0.020 | Q   |
| 2776  | 34.675   | 1.590  | 27.775 | 0.1   | 0.7  | -0.6  | 0.4  | T   |
| 3026  | 34.680   | 1.470  | 27.787 |       |      | 0.040 | 0.020 | L   |
| 3276  | 34.684   | 1.350  | 27.799 | 0.4   | 0.4  | 0.7   | 0.9  | T   |
| 3527  | 34.689   | 1.240  | 27.810 |       |      | 0.020 | 0.010 | L   |
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SR90 FROM 0 M TO 500 M= 7.4 mCi/km²
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PU239 FROM 0 M TO 4346 M = 1.56 mCi/km²
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| CS137 FROM 0 M TO 450 M= 39.5 mCi/km² |
| SR90 FROM 0 M TO 450 M= 25.3 mCi/km² |
| PU239 FROM 0 M TO 4262 M= 1.10 mCi/km² |

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This report contains results of measurements of the fallout radionuclides $^{90}$Sr, $^{137}$Cs, $^{239,240}$Pu, and $^{241}$Am in large volume seawater samples collected between 1972 and 1974 in the Atlantic and Pacific as part of Geochemical Ocean Sections (GEOSECS) program. The stations for which data are reported include both the North and South Atlantic oceans and latitudes north of 20°S in the Pacific Ocean. The $^{90}$Sr and $^{137}$Cs data set has been corrected by a procedure which estimates independently the analytical blank for the laboratory which made the analysis. When the data quality and spacing permit, water column inventory estimates were made for each nuclide over depth intervals appropriate to the nuclide's distribution.