JOURNAL OF GEOPHYSICAL RESEARCH, VOL. 88, NO. C9, PAGES 5411–5415, JUNE 20, 1983

^{7}Be and ^{210}Pb Total Deposition Fluxes at New Haven, Connecticut and at Bermuda

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The total deposition fluxes of ^{210}Pb and ^{7}Be were determined at New Haven, Connecticut, and Bermuda over approximately the same annual period in 1977–1978. The ^{210}Pb flux has remained virtually constant at New Haven from 1973 to 1978, the flux in the 1977–1978 period being 1.2 dpm/cm^2/y. The ^{210}Pb flux at Bermuda is 0.69 dpm/cm^2/y. This lower flux than expected from model calculations is due to the establishment of a blocking high pressure cell during the summer which deflects continental air. The ^{7}Be fluxes at New Haven and Bermuda are 22.7 and 17.1 dpm/cm^2/y, values consistent with western North Atlantic oceanic standing crop measurements, but higher than some other estimates. Where the difference cannot be attributed to differences in sampling it is ascribable to regional differences compatible with the oceanic data.

INTRODUCTION

The natural radionuclides produced in the atmosphere provide useful tools for the study of the behavior of aerosols since the source terms are definable and the measurements generally insensitive to normal environmental contaminations. This has been recognized for some time and useful summaries of the use of cosmogenic and uranium decay series radionuclides in atmospheric studies exist [see, for example, *Lal and Peters,* 1967; *Turekian et al.,* 1977].

In this paper we will discuss the behavior of two natural radionuclides, ^{210}Pb and ^{7}Be. ^{210}Pb (22-year half-life) is produced ultimately from ^{222}Rn (3.8-day half-life) which emanates from soils. ^{7}Be (54-day half-life) is produced by cosmic rays. Although the major production of ^{7}Be is in the stratosphere, nevertheless, because of the short decay half-life of ^{7}Be and the long (~1 year) residence time of stratospheric aerosols, most of the ^{7}Be flux to the earth's surface is from tropospheric production. Only at sites and times of stratospheric air intrusion into the troposphere is the ^{7}Be flux from the stratospheric reservoir significant. The production of ^{7}Be decreases with descent through the troposphere because of the attenuation of the cosmic ray flux by interactions with atomic targets in the atmosphere.

We have made monthly total atmospheric precipitation collections for ^{7}Be and ^{210}Pb at two sites in the Atlantic Ocean basin as part of the SEAREX program (sponsored by the National Science Foundation). The aim of the quasisynoptic program was to determine the atmospheric deposition fluxes for the two radionuclides at the two sites over the same time period (1977–1978). Such a data set can then be used to answer the following questions: (1) How do the fluxes of the two radionuclides differ from the continental margin to an island 'downwind' along the prevailing westerlies? (2) What are the relations of the ^{210}Pb and ^{7}Be fluxes to the theoretical predictions for these nuclides? (3) What is the relationship of the radionuclide fluxes to precipitation? (4) How are the fluxes of the two radionuclides related to their atmospheric concentrations?

The answers to these questions should provide a framework for determining how these natural radionuclides can be useful in predicting the behavior of chemical species injected into the troposphere.

METHODS AND RESULTS

The sampling sites chosen for this study were the roof of the Kline Geology Laboratory (KGL) at Yale and the roof of the laboratory at the Bermuda Biological Station (BBS). From May through December 1977, the primary KGL collector was a bottle-and-funnel assembly having a liquid-seal trap and venting through a bubbling tube. Precipitation was estimated by using the amount of water in the bottle, assuming no evaporation. Starting in January and continuing through June 1978, parallel samples were collected at KGL in two identical polyethylene jars (25 cm internal diameter, 28 cm depth). To one jar, medicinal mineral oil was added to suppress evaporation; this sample was used for determination of precipitation. Radiochemical analyses for this period were performed on the parallel sample which contained no mineral oil. The KGL collectors were emptied about the first of each month, cleaned, and were re-deployed after the addition of dilute acid. The collector at BBS was a plastic funnel that emptied via a short plastic tube into a polyethylene bottle containing acid and which was vented through narrow-diameter tubing in order to suppress evaporation. The BBS sample bottles were changed at the beginning of each month and shipped by air to Yale. These procedures were necessary to measure the short-lived ^{7}Be. The analytical and counting procedures for both ^{210}Pb and ^{7}Be have been described earlier (^{210}Pb, *Benninger* [1976]; ^{7}Be, *Krishnaswami et al.* [1980]). The purification procedure for ^{7}Be prior to precipitation and mounting for counting is quite extensive and virtually guarantees the exclusion of other gamma emitters. The decay curves were followed and all fit a 54 day half-life characteristic of ^{7}Be.

The simultaneous sampling for ^{7}Be and ^{210}Pb at KGL began in March 1977 and terminated in June 1978; the BBS sampling started in September 1977 and terminated 1 year later. The ^{7}Be results for KGL from March 1977 through November 1977 have previously been reported by *Krishnas-*

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Paper number 2C1674.
0148-0227/83/002C-1674$05.00

TABLE 1. Monthly Fluxes of Precipitation, ^{210}Pb and ^{7}Be at the Bermuda Biological Station (BBS) and the Kline Geology Laboratory (KGL) During 1977–1978

Month	Rainfall (cm)	Pb-210 Flux,* dpm/cm^2	Be-7 Flux,* dpm/cm^2
		New Haven (KGL)	
March 1977	16.6	0.159 ± 0.002	2.82 ± 0.15
April 1977	11	0.096 ± 0.002	1.41 ± 0.07
May 1977	6.90	0.052 ± 0.002	1.45 ± 0.16
June 1977	12.1	0.131 ± 0.002	2.98 ± 0.11
July 1977	3.81	0.065 ± 0.002	1.59 ± 0.06
Aug. 1977	9.08	0.064 ± 0.002	0.81 ± 0.06
Sept. 1977	16.2	0.128 ± 0.002	2.2 ± 0.10
Oct. 1977	15.07	0.195 ± 0.002	3.12 ± 0.21
Nov. 1977	22.86	0.042 ± 0.002	3.62 ± 0.01
Dec. 1977	12.23	0.085 ± 0.002	1.73 ± 0.01
Jan. 1978	18.78	0.135 ± 0.002	1.22 ± 0.027
Feb. 1978	3.47	0.0265 ± 0.002	0.40 ± 0.04
March 1978	10.93	0.135 ± 0.002	2.49 ± 0.06
April 1978	5.13	0.051 ± 0.002	0.70 ± 0.02
May 1978	15.72	0.157 ± 0.002	2.76 ± 0.03
June 1978	4.94	0.077 ± 0.002	0.90 ± 0.03
		Bermuda (BBS)	
Sept. 1977	19.1	0.047 ± 0.001	1.68 ± 0.15
Oct. 1977	20.6	0.033 ± 0.001	1.82 ± 0.49
Nov. 1977	21.5	0.054 ± 0.001	1.65 ± 0.26
Dec. 1977	15.6	0.076 ± 0.001	2.26 ± 0.55
Jan. 1978	10.6	0.063 ± 0.001	—
Feb. 1978	15.5	0.112 ± 0.001	1.74 ± 0.35
March 1978	10.1	0.055 ± 0.002	1.12 ± 0.09
April 1978	6.9	0.050 ± 0.002	0.89 ± 0.17
May 1978	11.4	0.037 ± 0.002	0.22 ± 0.10
June 1978	17.9	0.047 ± 0.002	—
July 1978	5.7	0.043 ± 0.002	—
Aug. 1978	15.0	0.074 ± 0.006	—

*Errors are 1 sigma counting errors.

wami et al. [1980]. The results are shown in Table 1. In most cases the precipitation data are based on the actual mass of water used in the analysis (or, in the case of the jar collections at KGL, in the parallel collector). KGL estimates of precipitation made by this method for 1977 compare favorably with data published for the same time period by the National Climate Center stations in New Haven at both the airport and in town. During 1978 there was no official weather station in New Haven, but the data compare well with the Bridgeport data for that period.

Discussion

The ^{210}Pb flux patterns. *Benninger* [1976, 1978, and unpublished data] obtained ^{210}Pb and precipitation fluxes at KGL for several years prior to this study. For the 4-year period August 1973 to July 1977, annual ^{210}Pb fluxes were 0.99, 0.86, 1.21, and 1.04 dpm/cm^2/y, respectively. Monthly ^{210}Pb flux and precipitation over the 4-year period are correlated with a regression coefficient of 0.67 (significant at $P < 0.001$). The annual ^{210}Pb deposition flux for the period between March 1, 1977, and June 30, 1978, is 1.2 dpm/cm^2/y, which falls in the range of the earlier results; the correlation coefficient for the 1977–1978 data set is 0.52 (significant at $P < 0.05$, > 0.02).

At Bermuda during the year beginning September 1, 1977, the ^{210}Pb flux was 0.69 dpm/cm^2/y, significantly lower than at New Haven. There was no correlation ($r = 0.04$) between ^{210}Pb flux and precipitation.

'Total deposition velocity' of ^{210}Pb at New Haven and Bermuda. The 'total deposition velocity,' v, of a nuclide is here taken as the ratio

$$v = \frac{\text{total flux to the earth's surface}}{\text{concentration in air at some reference level}}$$

So defined, the total deposition velocity includes both wet precipitation and dry deposition. We shall be concerned with concentrations measured in surface air (reference = ground level).

^{210}Pb in surface air in New York City (100 km SW of the KGL sampling site) was measured during August 1974 through July 1976 by the Environmental Measurements Laboratory (EML, formerly Health and Safety Laboratory, HASL) of the U.S. Department of Energy. Combining these composite monthly concentrations of ^{210}Pb in air with our monthly ^{210}Pb deposition fluxes at New Haven yields a mean 'total deposition velocity' of 0.97 ± 0.36 cm/s ($\bar{x} \pm \sigma$, 24 observations). Alternatively, for an average ^{210}Pb total deposition flux of 1.03 ± 0.14 dpm/cm^2/y ($\bar{x} \pm \sigma$) for four annual fluxes at New Haven) and an average air concentration of (34.5 ± 7.7) × 10^{-3} dpm/m^3 ($\bar{x} \pm \sigma$, 24 EML observations), the mean deposition velocity at New Haven is 0.95 ± 0.25 cm/s. This indicates that the total deposition velocity in the northeastern United States is about 1 cm/s for ^{210}Pb.

Bermuda, as an oceanic site, need not show the same pattern as the northeastern U.S. continental sites. At Enewetak, a tropical island site in the western North Pacific, *Turekian and Cochran* [1981*a*, *b*] found that the total deposition velocity for ^{210}Pb varied from about 1 cm/s during the 'dry' season (April–May) to about 4 cm/s during the 'wet' season (July–August). A series of concurrent measurements of ^{210}Pb total deposition flux and air concentration at Bermuda would be required to establish the nature of the variation pattern of total deposition velocity with the seasons.

We obtained an air sample at sea aboard the R/V *Endeavor* near Bermuda during the summer of 1979 as part of the SEAREX program and analyzed it for ^{210}Pb. The concentration of ^{210}Pb was 21.2 × 10^{-3} dpm/m^3 which, if combined with our measured Bermuda ^{210}Pb total deposition flux for June, July, and August 1978 (equivalent to 0.66 dpm/cm^2/y) yields a total deposition velocity of 1.0 cm/s. This is valid only for the summer season characterized by a persistent high pressure cell (the so-called 'Bermuda high') which blocks air from North America from reaching Bermuda. Although this velocity is comparable to the northeastern U.S. value, we do not know its variability with time.

Turekian et al. [1977] constructed a hemispheric model for ^{222}Rn fluxes from the continents and the ^{210}Pb flux to the earth's surface as a function of longitude. In this model it was assumed that a mean westerly flow of air carried ^{222}Rn and ^{210}Pb and that their fluxes were homogenized at each longitude between 15° and 50° latitude in each hemisphere. No allowance was made for regional patterns which might dominate the local fluxes. The model predicted that the ^{210}Pb flux should first increase proceeding eastward from the eastern seaboard of the United States before decreasing across the Atlantic Ocean to Europe.

The ^{210}Pb flux at Bermuda by this simplified model would be equal to or slightly greater than the flux at New Haven. As our results show, this is not the case. The measured Bermuda flux is about 70% of the expected flux for that longitude. The reason for this seems to lie in the fact that

TABLE 2. Comparison of KGL and BBS ^{7}Be Total Deposition Fluxes With Those From Other Sites

Location and Time	^{7}Be Flux (dpm/cm^2/y)	^{7}Be Flux/ Precipitation (dpm/L)	Source
Fluxes Based on Precipitation Collectors			
Chilton, England (51°N) Oct. 1959–Sept. 1960	5.5	81	*Peirson* [1963]
Milford Haven, England (51°N) Oct. 1959–Sept. 1960	5.2	83	*Peirson* [1963]
Westwood, N.J. (41°N) Dec. 1960–Aug. 1961	4.3	55	*Walton and Fried* [1962]
Rijswijk, Netherlands (52°N) Nov. 1960–Oct. 1961	9.5	102	*Bleichrodt and van Abkoude* [1963]
Bombay, India (19°N) 1955–1961, 1963–1965, 1968, 1970	7.6	33	*Lal et al.* [1979]
Quillayute, Wash. (49°N) Feb. 1976–Jan. 1977	8.1	30	*Crecelius* [1981]
New Haven, Conn. (41°N)	22.7	162	this paper
Bermuda (33°N)	17.1	101	this paper
Fluxes Based on Ocean Water Inventory			
GEOSECS II (36°N)			*Silker* [1972]
Aug. 27, 1970	22.5		
Aug. 28, 1970	19.2		
Aug. 29, 1970	21.6		
Sargasso Sea			*Aaboe et al.* [1981]
T-1 (37°N) Fall, 1978	20.1		
T-2 (32°N) Fall, 1978	18.7		
T-3 (32°N) Fall, 1978	21.1		
T-4 (32°N) Fall, 1978	14.5		

during the summer the 'Bermuda high' is sufficiently strongly developed to block passage of air coming directly off the North American continent. The three winter months, December, January, and February, yield a ^{210}Pb flux similar to that for the northeastern United States (where the flux does not vary systematically over time). During these 3 months, the probability of developing a stationary high pressure air mass over the North Atlantic is very small and direct streaming of air off the continent is the common feature.

The ^{7}Be flux patterns. As ^{7}Be has its origin in cosmic ray interaction with the atmosphere, its flux to the earth's surface can be expected to have a latitudinal variation [*Lal and Peters,* 1967]. For the atmosphere as a whole there is a generally higher cosmic ray flux at the high latitudes attenuating to the low latitudes because of the Earth's magnetic field.

Most of the production of ^{7}Be, like the other spallation and reaction products of cosmic rays with atmospheric molecules, occurs in the stratosphere. However, the long residence time of aerosols in the stratosphere (~1 year) and the short half-life of ^{7}Be (54 days) means that the stratospherically produced ^{7}Be will reach the earth's surface only when advection of stratospheric air into the troposphere occurs. Thus, the tropospheric ^{7}Be production determines the major flux rate, modulated regionally by the delivery of stratospheric air. Generally, the stratosphere 'leaks' through the troposphere around 45° latitude at certain times of the year. In semi-arid locations like Richland, Washington, this is seen as a seasonal change in the ^{7}Be concentration of the surface air [*Silker,* 1972; *Young and Silker,* 1980]. At New York, however, virtually no change in the atmospheric concentration of ^{7}Be occurs over time [*Freely et al.,* 1979].

The flux of ^{7}Be at KGL is 22.7 dpm/cm^2/y and at BBS it is 17.1 dpm/cm^2/y. These two fluxes may be significantly different but one year's data can hardly be sufficient to establish that. ^{7}Be flux at both sites correlates with the precipitation on a monthly basis although the correlation is better in New Haven (correlation coefficient = 0.74, significant at $P < 0.001$, > 0.0001) than in Bermuda (correlation coefficient = 0.66, significant at $P = 0.05$). This correlation between ^{7}Be and precipitation is much better than it is for ^{210}Pb for the same time period.

In Table 2 we compare our ^{7}Be flux data with other estimates. Our ^{7}Be deposition fluxes are greater than those determined from the analyses of precipitation in other areas but about the same as the calculated flux from the standing crop of ^{7}Be in the northwestern Atlantic Ocean.

Some of the low precipitation fluxes may be due to incomplete sampling of the year's total ^{7}Be flux (as at Westwood, New Jersey), or to loss of ^{7}Be by adsorption to the surface of the collector if the sample was not acidified prior to transfer (as at Chilton and Milford Haven in England; Bombay, India; and Seattle, Washington). The flux measurement made in the Netherlands was not subject to either of these possible problems, yet the ^{7}Be flux there is about one half our measured flux.

We cannot rule out regional variations in ^{7}Be flux. The flux data of *Young and Silker* [1980] for the North Atlantic Ocean based on the estimated standing crop of ^{7}Be in the ocean column indicates a decreasing flux from west to east across the ocean. Part of this may be due to oceanographic effects, but certainly the data are compatible with the measured precipitation fluxes in Connecticut, Bermuda, and The Netherlands as well. Regional variations of ^{7}Be flux on a

worldwide scale have been suggested by *Young and Silker* [1980] based on their oceanic standing crop data. The same texture may be showing up in direct precipitation studies.

'Total deposition velocity' of ^{7}Be in New Haven. We can use the atmospheric concentration of ^{7}Be determined by *Feely et al.* [1979] for the New York area (two locations) and our precipitation ^{7}Be flux to calculate a total deposition velocity for ^{7}Be as we have done for ^{210}Pb. The atmospheric average concentration is 0.266 dpm $^{7}Be/m^{3}$, which, for 22.7 dpm $^{7}Be/cm^{2}/y$, yields a deposition velocity of 2.8 cm/s. This value is significantly larger than the deposition velocity for ^{210}Pb (0.95 cm/s) in the same area.

The comparative behavior of ^{7}Be and ^{210}Pb as aerosols. A comparison of the behavior of ^{7}Be and ^{210}Pb during the precipitation process indicates marked differences which can be explained on the basis of their modes of production and delivery to the earth's surface. The major differences between the behavior of the two nuclides are (1) ^{7}Be fluxes at both New Haven (KGL) and Bermuda (BBS) are correlated with the precipitation flux at each site whereas the correlation is weaker for ^{210}Pb, (2) on the basis of one year's sampling the fluxes of ^{210}Pb and ^{7}Be at Bermuda are lower than the fluxes at New Haven, (3) the 'total deposition velocity' of ^{210}Pb in Bermuda appears to be about the same as that in New Haven for the months for which comparison is possible, and (4) the 'total deposition velocity' of ^{210}Pb in New Haven is 0.95 cm/s compared to 2.8 cm/s for ^{7}Be.

The reason for these observations lies in the sites of production of the two nuclides. ^{210}Pb is produced from ^{222}Rn which emanates from the ground and undergoes decay with a 3.8 day half-life as it moves upward through the troposphere. The concentration of ^{210}Pb in the air over the continents decreases with elevation following ^{222}Rn as shown by *Moore et al.* [1973]. ^{7}Be, on the other hand, is produced by cosmic rays bombarding the atmosphere so that its concentration will tend to decrease as the earth's surface is approached, as is observed for bomb radionuclides descending from the stratosphere [see citations in *Young and Silker,* 1980]. ^{7}Be differs from ^{210}Pb in that the former's production is independent of geography at any particular latitude and therefore, to a first approximation, its standing crop in the atmosphere should be the same over both the continents and the oceans, whereas the latter's will be strongly dependent on longitude because the production of its antecedent, ^{222}Rn, is strictly continental.

Moore et al. [1973] have argued that for the continental atmosphere the mean residence time of aerosols is independent of height for the major part of the troposphere. This is compatible with a precipitation scavenging at each level of the troposphere proportional to the concentration there. Thus more of the ^{210}Pb is derived from the lower part of the atmosphere over continents and more of the ^{7}Be is derived from the upper part of the troposphere at all locations. The direct consequence of this is that using ground level as the reference, the 'total deposition velocity' of ^{210}Pb at a continental site will be smaller than 'total deposition velocity' of ^{7}Be, as indeed we have observed.

Application to other chemical species. The difference in behavior of ^{210}Pb and ^{7}Be provides an insight to the behavior of other chemical species in atmospheric aerosols. A general rule can be stated: if the source of the aerosol is effectively a gaseous component derived from the surface with a conversion time scale to an aerosol similar to the ^{222}Rn radioactivity decay time scale, then the resulting aerosol will behave like ^{210}Pb and its total deposition velocity will be similar to that of ^{210}Pb; if the aerosol is generated most efficiently in the upper troposphere, or is supplied from the stratosphere by settling, its behavior will resemble that of ^{7}Be if its half-life is significantly longer than the residence time of an aerosol in the air.

In the first category are the gaseous emanations such as SO_2 and NO_x (or N_2O) which are converted in the atmosphere to $SO_4^{=}$ and NO_3^{-} and thereby become aerosol components. Metals released as vapors by man or natural means from the ground level may also behave like ^{210}Pb if their half-lives of conversion to the ionic state are about the same as that of ^{222}Rn. Fine-grained particles released from ground level will also most closely be replicated by the behavior of ^{210}Pb, but large aerosols such as those emitted by automobiles and power plants will obviously have much shorter residence times.

The components behaving like ^{7}Be are stratospheric bomb fallout debris, stratospherically injected volcanic components and most cosmic-ray produced nongaseous nuclides. It is possible to approximate the flux of these nuclides by using the ^{7}Be deposition velocity at a particular site and the ground surface air concentration of the component of interest.

At Enewetak, *Turekian and Cochran* [1981*a*, *b*] used ^{210}Pb correlation with Al to determine the Asian dust flux to the North Pacific using the measured ^{210}Pb flux at the site. Similarly, *Settle et al.* [1982] determined the Pb flux to a number of tropical sites from the $Pb/^{210}Pb$ ratio in air, precipitation, and an independently determined ^{210}Pb flux.

Recently, *Jickells et al.* [1982] measured $SO_4^{=}$ in rain water from Bermuda in monthly collectors for a year starting in May 1980. Although their sampling program was started 3 years after our sampling program had started, it is evident that $SO_4^{=}$ and ^{210}Pb concentrations in rain at Bermuda increase during the three winter months (December, January, February). It is not possible to determine, quantitatively, the sulfate flux from these two unrelated data sets, but the rough correlation indicates that a precise way to obtain this value, of course, is to determine $SO_4^{=}/^{210}Pb$ ratios in air samples over an extensive sampling period.

Acknowledgments. Anthony Knap graciously tended our precipitation collector at the Bermuda Biological Station and promptly sent us the monthly collection. S. Krishnaswami read an early draft of the manuscript and offered many valuable comments. This research was started under a grant from the U.S. Department of Energy, EY-76-S-02-3573, and completed as part of the SEAREX Program supported by the National Science Foundation under grants OCE-7712847 and OCE-8113153.

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(Received June 25, 1982;
revised September 24, 1982;
accepted October 20, 1982.)