Mean Residence Time of ^{7}Be-Bearing Aerosols in the Troposphere

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Measurements of ^{7}Be specific activities in ground level air have been carried out at Fullerton, California, for a 23-month period from September 1973 through July 1975. The mean specific activity observed at this relatively dry location (less than 37 cm of annual total precipitation) was 0.216 pCi/m^{3}. From this result a mean tropospheric residence time for ^{7}Be-bearing aerosols of 35.4 days is obtained. This result is in very good agreement with residence times obtained from earlier measurements of ^{7}Be and ^{7}Be/^{32}P ratios in rainwater.

The usefulness of ^{7}Be and other cosmic ray produced nuclides as atmospheric tracers has been well established [*Machta*, 1959; *Bolin*, 1964; *Lal and Peters*, 1967; *Lal and Suess*, 1968], and several series of measurements of ^{7}Be specific activities in rainwater and air have been reported [*Arnold and Ali Al-Salih*, 1955; *Cruikshank et al.*, 1956; *Goel et al.*, 1956; *Rama Thor and Zutshi*, 1958; *Anderson et al.*, 1960; *Gustafson et al.*, 1961; *Shvedov et al.*, 1962; *Parker*, 1962; *Lindblom*, 1962; *Schumann and Stoeppler*, 1963; *Peirson*, 1963; *Gold et al.*, 1964; *Perkins et al.*, 1964; *Rangarajan and Gopalakrishnan*, 1970; *Kolb*, 1970]. The results of these measurements have indicated that washout by rainfall efficiently removes ^{7}Be-bearing aerosols from the lower troposphere. In addition, the values of ^{7}Be specific activity and ^{7}Be/^{32}P specific activity ratios in the rainwater measurements are consistent with mean residence times for ^{7}Be-bearing aerosols in the troposphere of approximately 40 days [*Lal and Peters*, 1967]. In general, measurements of ^{7}Be specific activities in ground level air are distorted by washout, so that mean residence times cannot be determined from these data. The low specific activities observed (generally under 0.070 pCi/m^{3}), however, are consistent with the assumption that washout and radioactive decay are the major removal processes for the isotope in the troposphere.

Recently, a number of mean residence times for the troposphere have been derived from measurements of ^{222}Rn, ^{210}Pb, ^{210}Bi, and ^{210}Po in air [*Moore et al.*, 1973, 1974; *Rangarajan et al.*, 1975] and in rainwater [*Gavini et al.*, 1974]. The mean residence times that are derived from ^{222}Rn/^{210}Pb ratios and ^{210}Bi/^{210}Pb ratios in air generally are under 7 days, while the residence times resulting from ^{210}Po/^{210}Pb ratios generally are much longer, 20–65 days. *Moore et al.* [1973] consider the residence times derived from the ^{210}Po/^{210}Pb suspect because of the possibility that at least part of the ^{210}Po in the troposphere may be due to sources other than the decay of ^{222}Rn. This conclusion, however, is in disagreement with measurements by *Marenco and Fontan* [1972, 1973] and calculations by *Hartwig* [1973]. In addition, *Gavini et al.* [1974] found reasonable agreement between their ^{210}Bi/^{210}Pb and ^{210}Po/^{210}Pb residence times in about 70% of their observations.

Because of the marked difference between the mean residence times obtained from the ^{7}Be data and those obtained from ^{222}Rn/^{210}Pb and ^{210}Bi/^{210}Pb ratios in air, we have redetermined the mean residence time for ^{7}Be-bearing aerosols by an alternate experimental method. The results, shown below, are in very good agreement with the value of 40 days obtained from the earlier rainwater measurements.

EXPERIMENTAL PROCEDURE

Beginning in September 1973, high-volume air samples were obtained at a sampling station located on the campus of California State University, Fullerton (33°52'N, 117°55'W), on a weekly basis. Initially, an Interex model 19-100 high-volume sampler was used to obtain 300-m^{3} samples on a single paper or fiber glass filter (S&S 589/1 and Reeve Angel 934AH). After February 16, 1975, a weather measure APS-1 sampler was used to obtain 900-m^{3} samples on three fiber glass filters (Reeve Angel 934AH) run simultaneously.

Shortly after collection, the filters were counted in a high-resolution gamma ray spectrometer to determine the specific activities of a number of short-lived radionuclides. Approximately a week later, each sample was recounted for 1000 min to determine the specific activity of ^{7}Be and other long-lived isotopes collected on the filter. The specific activity of ^{7}Be was determined from the intensity of the 477.6-keV gamma ray emitted in its decay. With the 300-m^{3} samples the detection limit for ^{7}Be was less than 0.030 pCi/m^{3}, while for the 900-m^{3} samples a detection limit of 0.010 pCi/m^{3} obtained.

The lithium-drifted germanium counter used for these measurements has a resolution width of 2.7 keV for the 1336-keV line of ^{60}Co. As a result, the ^{7}Be gamma ray line is well resolved from other nearby peaks. Details of the counting system and the calibration of the spectrometer have been given previously [*Shapiro and Forbes-Resha*, 1975].

The sampling location is one of relatively light precipitation. For the years 1974 and 1975, precipitation totals of 37.1 and 36.0 cm were recorded. Most of this rainfall occurred during a small number of major storms in the winter months. As a result, it was always possible to avoid taking the weekly air sample during the time when rain was falling. The results reported in this paper are based on 91 samples taken over a 23-month period. In all but eight of these measurements, ^{7}Be specific activities above the detection limit were observed. The eight samples with activity levels below the detection limit were all taken within 48 hours after a significant rainfall. Because of the relatively small amount of precipitation at the sampling location, the specific activities of ^{7}Be observed can be considered to be undistorted by washout.

RESULTS AND DISCUSSION

Monthly average ^{7}Be specific activities for the period from September 1973 through July 1975 are shown in Figure 1, together with the total precipitation for each month during this period. In computing monthly averages, individual values below the detection limit have been set equal to 0.0 pCi/m^3. The average ^{7}Be specific activity obtained from the entire sequence of data is 0.216 pCi/m^3. If the eight samples yielding 0.0 pCi/m^3 are omitted, the average is increased to 0.237 pCi/m^3. These average values are shown as the lower and upper dashed lines, respectively, in Figure 1.

There are two notable features present in the data obtained at this sampling location. First, the mean ^{7}Be specific activity is between 2 and 3 times that observed at sampling locations with more normal precipitation levels. Second, there are no pronounced seasonal variations. The lack of strong seasonal variation in the data implies that the ^{7}Be observed at this location is predominantly tropospheric in origin. This conclusion is supported by the fact that specific activities of fission products observed on the same filters show typical seasonal peaks.

Mean tropospheric residence times for ^{7}Be-bearing aerosols have been computed under the following assumptions:

1. All ^{7}Be observed at this sampling location is tropospheric in origin.
2. The troposphere can be considered a reasonably well mixed reservoir. Values of the vertical and north-south eddy diffusivities for both the lower and upper troposphere are sufficiently high to make this assumption reasonable for the 54-day half-life of ^{7}Be [*Bolin*, 1964].
3. The principal mechanisms by which ^{7}Be is removed from the atmosphere are washout and radioactive decay. In particular, we have neglected dry deposition of ^{7}Be. While this assumption may be open to some question, the fact that the mean residence times calculated from ^{7}Be alone in rainwater and from ^{7}Be/^{32}P ratios in rainwater are both quite close to 40 days [*Lal and Peters*, 1967] provides evidence that the dry deposition rate for ^{7}Be must be a reasonably small fraction of the production rate in the troposphere.

Under these conditions the activity of ^{7}Be in tropospheric air is given by

$$a_T = (P_T/H)(1 - e^{-\lambda t_r}) \quad (1)$$

where a_T is observed mean ^{7}Be activity from our data, P_T is the production rate of ^{7}Be in the troposphere at 34°N latitude, H is the mean height of the tropopause at this latitude, $\lambda = 1.31 \times 10^{-2}$/d is the decay constant for ^{7}Be, and t_r is the corresponding mean residence time for the ^{7}Be-bearing aerosol between washouts. (The t_r values referred to in this paper, strictly speaking, are apparent irradiation times. Apparent residence times derived from ^{7}Be/^{32}P ratios in rainwater average approximately 0.66 × the apparent irradiation times [*Martell and Moore*, 1974].) For P_T we have taken the value 3.2×10^{-2}/s/cm^2 given by *Lal and Peters* [1967], and for H we have taken an average value of 15 km. If the mean value of all our data is used in the above equation, a value of 35.4 days results for t_r. If the eight samples taken within 48 hours of significant local rainfall are excluded from the mean a_T, then t_r is increased to 39.9 days. This range is in very good agreement with the results obtained from measurements of ^{7}Be and ^{7}Be/^{32}P ratios in rainwater [*Lal and Peters*, 1967]. (These results also are in reasonably good agreement with tropospheric residence times obtained by the measurement of fission product ratios in rainwater [*Beck and Kuroda*, 1966; *Cooper and Kuroda*, 1966; *Thein and Kuroda*, 1967].)

It is not possible on the basis of the data reported here, or on the basis of previous ^{7}Be and ^{7}Be/^{32}P data, to exclude completely a small dry deposition rate. If such a process is present, however, even longer mean residence times for the ^{7}Be-bearing aerosol would be required to fit the present and previously reported data. If one makes the simplifying assumption of a constant loss rate of, say, Λ atoms per second per square centimeter, (1) must be modified to read

$$a_T = [(P_T - \Lambda)/H](1 - e^{-\lambda t_r}) \quad (2)$$

From (2) a dry deposition rate of 10% of the production rate

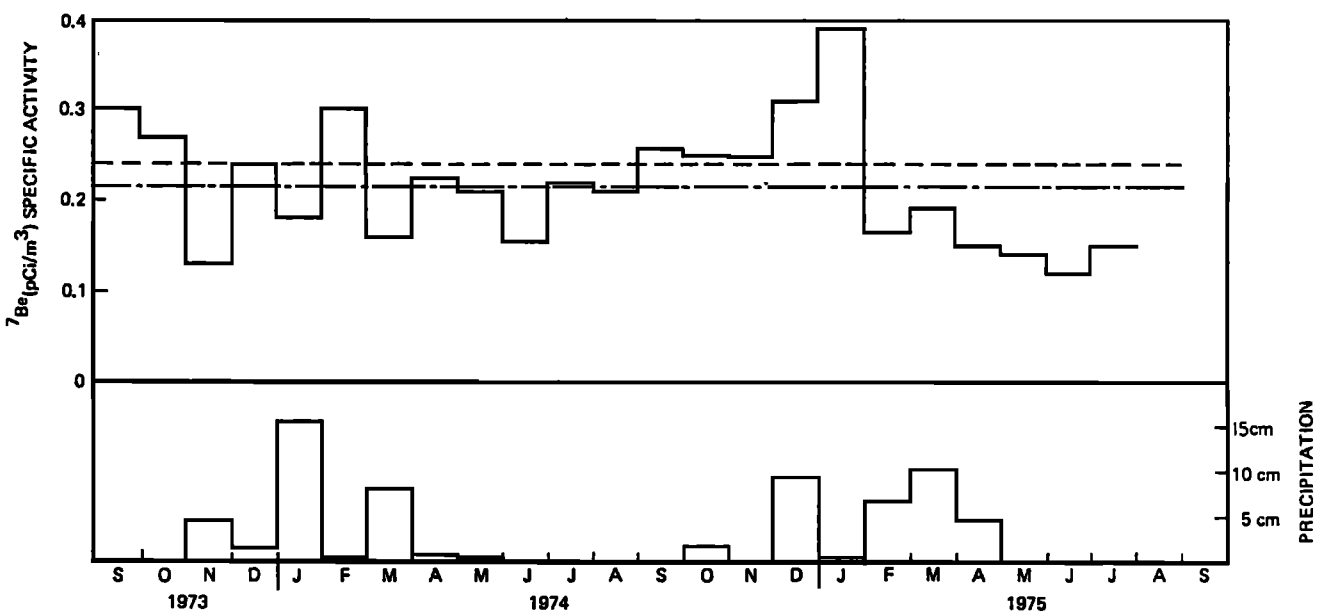

Fig. 1. (Top) Monthly average specific activities of ^{7}Be in ground level air at Fullerton, California, from September 1973 through July 1975. The lower dashed line indicates average specific activity obtained from all data taken. The upper dashed line represents the average specific activity obtained if eight samples below the detection limit are omitted. (Bottom) Monthly total rainfall at Fullerton, California, during the period from September 1973 to July 1975.

P_T would require a t_r of approximately 45 days to fit our observed ^{7}Be specific activity.

While our results support the earlier ^{7}Be data, great care must be exercised in interpreting any result obtained from a specific tracer. In the case of ^{7}Be the sources and sinks are well determined. Nearly all the ^{7}Be observed at ground level is produced in the upper troposphere well above cloud level, and the tropospheric production rate is almost independent of latitude [*Lal and Peters*, 1967]. The sinks are primarily washout and radioactive decay with perhaps a small dry deposition component. As a result, the t_r value observed (35–40 days) can be considered typical for aerosols in the upper troposphere, although any particular air mass may have an aerosol residence time considerably different from this mean value.

In addition, the ^{7}Be results are not necessarily inconsistent with the shorter residence times observed with radon daughters. The source of these tracers is at the surfaces of the continents mainly, and the shorter residence times observed with these tracers may be typical of the lower troposphere.

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