Tellus (1990), *42B*, 142–147

The use of natural radioactive tracers in a study of atmospheric residence times

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(Manuscript received 28 October 1988; in final form 21 April 1989)

ABSTRACT

The global values of the activity ratios of $^{7}Be/^{32}P$, $^{7}Be/^{35}S$, and $^{7}Be/^{22}Na$ are presented and discussed. The wide range of variations of these observed ratios and residence times derived from them indicate a varying input of a stratospheric component of these isotopes. This shows that the troposphere is not a closed system and residence times calculated from the above ratios are likely to be larger than the actual values. More consistent results are obtained for tropospheric residence times based on $^{210}Bi/^{210}Pb$ activity ratios from various locations. The $^{7}Be/^{35}S$ ratios are lower than the expected ratios by a factor of 2 to 3, calculated from cosmic ray production values, indicating other sources like bomb production. Since the ratios are less even during periods of low fallout activity, another source, such as graphite reactors is a possibility.

1. Introduction

The activity ratios of the cosmic ray spallation products ^{7}Be, ^{32}P, ^{35}S, ^{22}Na, and radon decay products ^{210}Pb, ^{210}Bi and ^{210}Po have been measured in air and rainwater to calculate tropospheric residence times (Marenco and Fontan, 1972; Martell and Moore, 1974; Marenco and Fontan, 1974a, b; Bolin and Rodhe, 1973; Georgi and Chameides, 1986). The concept of a single residence time for the whole troposphere is not valid, as it consists of different layers like the planetary boundary layer (PBL), the middle and upper troposphere, polar atmosphere etc., with different scavenging and mixing properties. The values of residence times obtained with the various radioactive tracers will therefore refer to their mean production levels, e.g., ^{7}Be, ^{32}P, for upper troposphere and ^{210}Pb, ^{210}Po, and ^{210}Bi for the PBL, etc. (Morenco and Fontan, 1972).

We present here $^{7}Be/^{32}P$, $^{7}Be/^{35}S$, $^{7}Be/^{22}Na$ and $^{210}Bi/^{210}Pb$ activity ratios from various locations of the globe to examine their utility in studying the tropospheric residence time.

2. Calculation of residence time

The build-up of an isotope by cosmic ray irradiation can be expressed as

$$N = P/\lambda(1 - \exp(-\lambda \times t)), \quad (1)$$

where N is atom concentration after irradiation for time t, λ is the radioactive decay constant and P is isotope production rate. For the atom ratios of two isotopes, the equation can be written as:

$$R = \frac{P_1/\lambda_1(1 - \exp(-\lambda_1 \times t))}{P_2/\lambda_2(1 - \exp(-\lambda_2 \times t))}, \quad (2)$$

where R is the atom ratio and the subscripts refer to the two isotopes.

The atom ratios will vary between $R_1 = P_1/P_2$ for small values of t and $R_2 = P_2\lambda_1/P_1\lambda_2$ for saturation values of t, i.e., t values much greater than the radioactive mean-life.

The above eq. (2) can be taken as indicating the growth of the ratios between successive precipitations if it can be assumed that air is washed of all activity by the previous precipitation. The range of values given by the above equations are

therefore the values to be expected for tropospheric air and rainfall if it is a closed system. The experimental ratios can be examined to see the irradiation times derived and for calculating mean residence time T_r. For a constant removal rate λ_m (inverse of residence time T_r assuming first order kinetics), the following equation holds.

$$\frac{dN_1}{dt} = P_1 - N_1(\lambda_1 + \lambda_m). \quad (3)$$

For steady state conditions ($dN_1/dt = 0$), the atom ratios of two isotopes can be derived from (3) above as

$$R_t = P_1(\lambda_2 + \lambda_m)/P_2(\lambda_1 + \lambda_m). \quad (4)$$

On simplification, it can be shown that T_r the mean residence time is given as

$$T_r = (R_t - P_t)/(P_t \times \lambda_2 - R_t \times \lambda_1), \quad (5)$$

where $P_t = P_1/P_2$. Hence, from R_t observed and P_t calculated, T_r can be derived. However the ratios of the two isotopes, in addition to irradiation time, also depend on the previous history of the concerned air mass. It can be shown (Marenco and Fontan, 1974a, b) that when an air mass from the stratosphere or upper troposphere (where production rate is higher) descends to lower levels (having lower production rates), very high values of the ratios, e.g., of $^7Be/^{32}P$, can be obtained. This will lead to erroneously high values of T_r. In addition, there are uncertainties in the calculation of isotope production rates which have undergone several revisions (Marenco and Fontan, 1974a). In view of these factors, a wide scatter in the activity ratios and errors in the calculated T_r values can be expected.

3. Results

The observed values of $^7Be/^{32}P$, $^7Be/^{35}S$, and $^7Be/^{22}Na$ are graphed in Figs. 1–3. Additional data regarding these values are also given in Tables 1–3. The tropospheric residence times calculated from the $^7Be/^{32}P$ ratios vary over a very wide range from less than 20 to more than 60 days (Fig. 1). The few values falling beyond the range of theoretical calculation limits (shown at the top of the figure) could be due to experimental errors in the case of $^7Be/^{32}P$ ratios. Alternatively, they could be due to decending air masses or production rate calculation errors as mentioned above. However, even the variations within the theoretical limits are more than what can be expected for T_r as a result of changes in frequency of rainfall and other atmospheric scavenging parameters. The comparison of ^7Be with ^{137}Cs of stratospheric origin in surface air has indicated the presence of a stratospheric component of ^7Be (Rangarajan and Gopala-

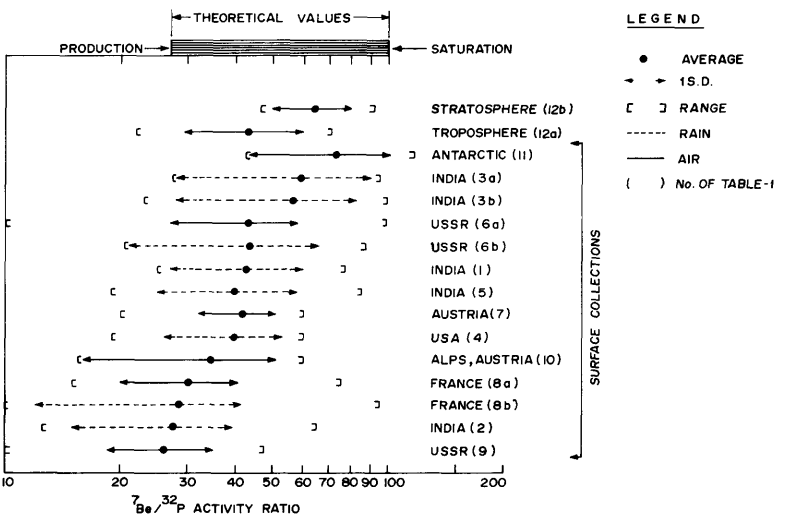

Fig. 1. $^7Be/^{32}P$ activity ratios at various locations of the world.

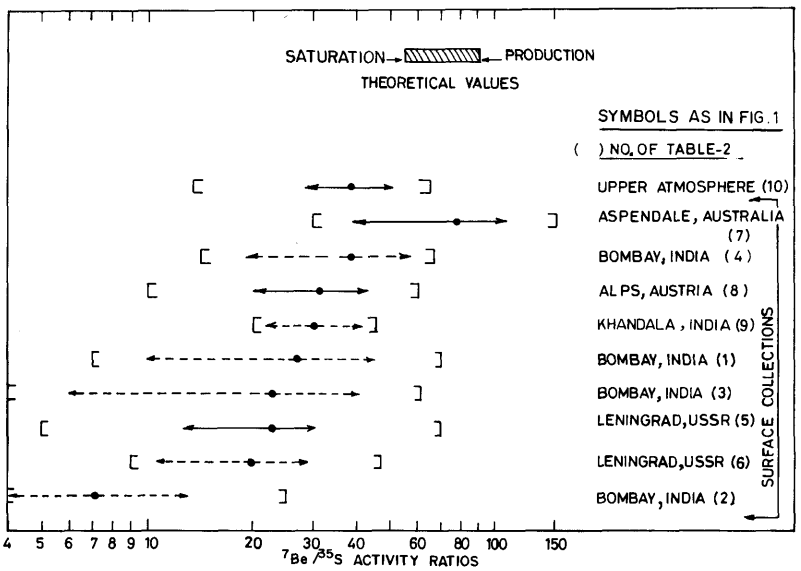

Fig. 2. $^{7}Be/^{35}S$ activity ratios at various locations of the world.

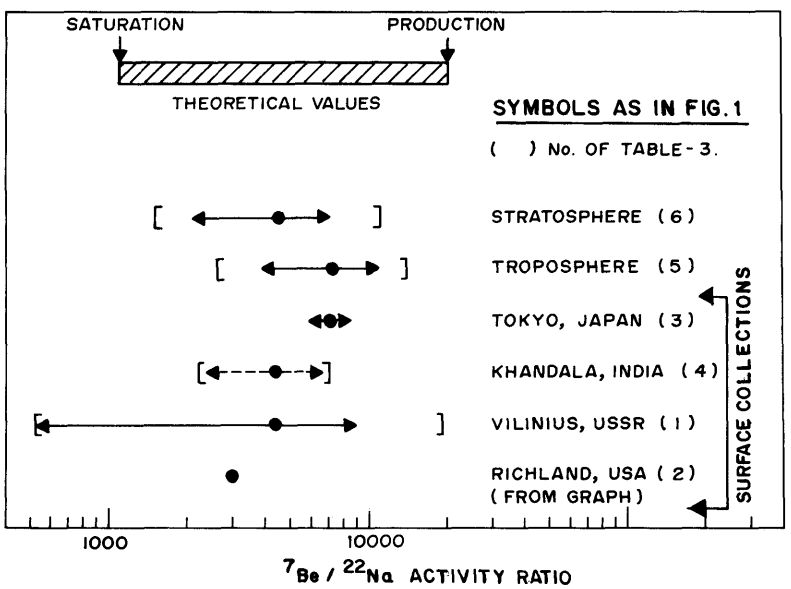

Fig. 3. $^{7}Be/^{22}Na$ activity ratios at various locations of the world.

krishnan, 1970; Feely et al., 1988), the magnitude of which is varying and time dependent. Hence the tropospheric residence times calculated from $^{7}Be/^{32}P$ ratios are not reliable. The same arguments also apply to $^{7}Be/^{22}Na$ ratios (Fig. 3 and Table 3).

The residence time values can also be obtained from $^{210}Bi/^{210}Pb$ ratios as pointed out by Martell and Moore (1974). Fig. 4 gives the average and range of residence time values calculated from the $^{210}Bi/^{210}Pb$ ratios measured at various locations. These valus refer to the PBL, as they are based on surface measurements. The higher values of residence time indicated by some rain-

Table 1. *$^{7}Be/^{32}P$ activity ratios at various locations of the world*

Sr. no.	Location	Sample type	No. of samples and year	Average activity ratio, SD and range	Reference
1	Bombay, India	rain, daily	11, 1957	43 ± 16 (25–77)	Goel et al. (1959)
2	Bombay, India	rain, daily	19, 1958	27 ± 12 (15–64)	Lal et al. (1960)
3a	Bombay, India	rain, daily	23, 1959	58 ± 30 (33–94)	Rama (1960)
3b	Kodoikanal, India	rain, daily	14, 1958	56 ± 28 (23–103)	Rama (1960)
4	New Jersey, USA	rain, daily	14, 1960	39 ± 13 (18–60)	Walton and Friend (1962)
5	Bombay, India	rain, daily	26, 1963–64	41 ± 16 (18–86)	Bhandari et al. (1970)
6a	Leningrad, USSR	air, days	117, 1963–66	42 ± 15 (10–100)	Gedeonov and Rys'yev (1969)
6b	Leningrad, USSR	rain, weekly	32, 1963–66	43 ± 22 (22–87)	Gedeonov and Rys'yev (1969)
7	Berne, Austria	air, weekly	19, 1965	41 ± 9 (20–60)	Aegerter et al. (1966)
8a	Toulouse, France	air, daily	120, 1967	30 ± 10 (15–75)	Marenco and Fontan (1974)
8b	Toulouse, France	rain, daily	Several, 1967	28 ± 15 (8–95)	Marenco and Fontan (1974)
9	Vilnius, USSR	air, days	24, 1967–69	26 ± 9.3 (8–47)	Luyanas et al. (1970)
10	Bavarian Alps, Austria	air, daily	>3000, 1970–80	34 ± 17 (20–60)	Reiter et al. (1983)
11	Antarctic Cont.	air, daily	26, 1977–81	73 ± 29 (46–132)	Sanak et al. (1985)
12a	Troposphere, 10°–70°N	air, several hours	17, 1959–65	43 ± 16 (22–71)	Bhandari et al. (1966)
12b	Stratosphere, 10°–70°N	air, several hours	21, 1959–65	64 ± 14 (47–93)	Bhandari et al. (1966)

Table 2. *$^{7}Be/^{35}S$ activity ratios at various locations of the world*

Sr. no.	Location	Sample type	No. of samples and year	Average activity ratio, SD and range	Reference
1	Bombay, India	rain, daily	11, 1957	27 ± 17 (7–70)	Goel et al. (1959)
2	Bombay, India	rain, daily	19, 1958	7.2 ± 6.0 (2.8–25)	Lal et al. (1960)
3	Bombay, India	rain, daily	13, 1959	23 ± 17 (2–62)	Rama (1960)
4	Bombay, India	rain, daily	8, 1963–64	38 ± 18 (18–67)	Bhandari et al. (1970)
5	Leningrad, USSR	air, weekly	86, 1964–66	23 ± 7.3 (5–70)	Gedeonov and Rys'yev (1969)
6	Leningrad, USSR	rain, weekly	24, 1964–66	20 ± 9.0 (9–46)	Gedeonov and Rys'yev (1969)
7	Aspendale, Australia	air, monthly	8, 1967–68	78 ± 38 (30–150)	Hicks (1969)
8	Bavarian Alps, Austria	air, daily	600, 1973–75	31 ± 10 (10–60)	Reiter et al. (1977)
9	Khandala, India	rain, daily	6, 1970	30.4 ± 9 (20–46)	Lal et al. (1979)
10	Upper atmosphere	air, few days	17, 1960–61	39 ± 10.5 (17–68)	Bhandari and Rama (1963)

Table 3. *$^{7}Be/^{22}Na$ activity ratios at various locations of the world*

Sr. no.	Location	Sample type	No. of samples and year	Average activity ratio, SD and range	Reference
1	Vilnius, USSR	air, few weeks	48, 1965–69	4400 ± 4400 (530–19000)	Luyanas et al. (1970)
2	Richland, USA	air, monthly	>100, 1968–76	3000	Reiter (1978)
3	Tokyo	rain	18, 1975–77	7100 ± 1140	Hasebe et al. (1981)
4	Khandala, India	rain, few weeks	4, 1970	4420 ± 2070 (2200–6960)	Lal et al. (1979)
5	Troposphere	air, few hours	12, 1960–61	7200 ± 3300 (2630–14280)	Bhandari and Rama (1963)
6	Stratosphere	air, few hours	15, 1960–61	4500 ± 2400 (1550–11110)	Bhandari and Rama (1963)

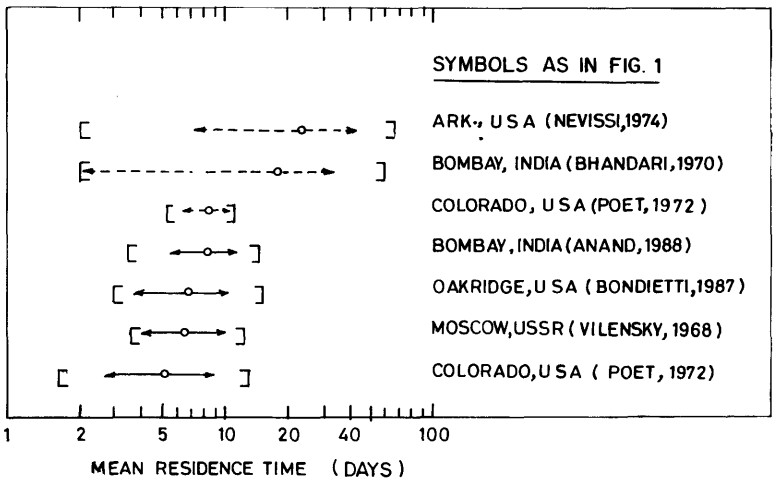

Fig. 4. Mean residence time from $^{210}Bi/^{210}Pb$ activity ratios.

fall data is possibly due to the presence of oceanic air at cloud levels at these locations (viz., monsoons and other oceanic sources). The maritime air will have a higher ratio as there is no fresh input of radon over the oceans (Martell and Moore, 1974). The T_r values based on data from various locations show a considerable degree of consistency and less scatter. The value of mean residence time for the planetary layer seems to be around 5–7 days, as data from several locations indicate (Fig. 4).

The ratios involving ^{35}S are factors of 2 to 3 below theoretical calculations, although the one set of measurements in the southern hemisphere is in the expected range (Table 2, Fig. 2). Other sources for ^{35}S, notably nuclear tests during periods of high fall-out have been suggested (Lal et al., 1960) for this difference. However, ratios are considerably lower even during periods of low fall-out (1970–1974), indicating other sources of ^{35}S, possibly graphite-moderated reactors. It is known that in graphite-moderated reactors, ^{35}S is produced by chlorine impurity by $^{35}Cl(n,p)^{35}S$ reaction. Sulphur in the lubricating oil has also been suggested as a source of ^{35}S by the reaction ($^{34}S(n,gamma)^{35}S$). Near the Hinckley reactor power station in the UK, levels up to 143 mBq/m^3 have been observed (Kluczewski et al., 1987). This is several orders of magnitude greater than the natural surface level from calculations. Hence, graphite reactors operating in Europe and USSR (Nuclear News, June 1986) could be the source of excess ^{35}S. In the southern hemisphere, there are very few reactors operating and natural levels of ^{35}S may not be affected.

4. Conclusions

The residence time calculations using cosmic ray produced isotopes are not reliable due to stratospheric input, production from other sources, etc. The best estimate appears to be those from the $^{210}Bi/^{210}Pb$ ratio. The data tabulated in this paper are also useful, as they give the range of radionuclide ratios to be expected in the atmosphere under normal conditions over the globe. This provides a reference for studying any unusual increase by inter-comparison.

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