Seasonal variation of Beryllium-7 relative to Caesium-137 in surface air at tropical and sub-tropical latitudes

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ABSTRACT

A comparative study of the seasonal variations of cosmic-ray produced Be-7 and fission product Cs-137 has been carried out in the surface air at Bombay and other stations in India. The results show that both isotopes attain peak values during the spring months of March–April. However it is observed that the amplitude of increase is much less in the case of Be-7 compared to stratospheric Cs-137. This difference is explained as being due to only a part of Be-7 in surface air being of stratospheric origin. By comparison with Cs-137, it is estimated that the stratospheric component of Be-7 is about 60 % in spring decreasing to 25 % in the autumn.

Introduction

Several interesting features of atmospheric circulation processes such as the exchange of air between the stratosphere and troposphere, transport within the stratosphere, residence times of aerosols in the various layers of the atmosphere, etc., have been studied using the bomb produced radioactivity released into the atmosphere (Newell *et al.*, 1963; List *et al.*, 1966; Feely *et al.*, 1966). The measurement of these radioactivities in the stratosphere and upper troposphere have been carried out by balloon borne as well as aircraft carried samplers (Rep. U.N., 1964).

Since the tracer concentrations at the ground level are the end result of several atmospheric processes in the stratosphere and troposphere, surface air activity measurements will also be useful for studying these processes. For this reason measurements on fission product activity of ground level air at several stations in India were carried out by us and their meteorological significance has already been discussed in previous publications (Rangarajan *et al.*, 1968; Vohra *et al.*, 1966).

Significant features of the atmospheric circulation mechanisms can also be studied using the cosmic ray produced isotopes existing in the atmosphere since these are subject to the same meteorological processes operative in the atmosphere. The measurement of cosmogenic isotopes can thus complement the data based on fission products and a comparative study of the two types of radioactivity will reveal the similarities and differences in their seasonal behaviour and the implications of these variations. Such a study is described in this article.

Measurements have been made on the concentration of Be-7 at Bombay and a few other stations in India from latitude 19°N to 34°N. These measurements have been carried out from November 1965 through the middle of 1966 along with the measurement of fission product activity. Be-7 is produced throughout the atmosphere, although in varying amounts, by the cosmic ray bombardment of the atmosphere (Lal & Peters, 1962). Maximum concentrations have been measured in the high latitude stratosphere, the concentrations increasing with altitude (Rep. U. N., 1964). Concentrations are also considerably higher in the upper troposphere compared to groundlevel air (Rep. U. N., 1964). In this respect Be-7 distribution is somewhat similar to that of fission products in the atmosphere following high latitude–high yield tests when maximum concentrations are found in the polar latitudes. Since fission products diffuse into the troposphere from the stratosphere, their concentrations are also higher in the upper troposphere as compared to ground level. Hence it is of considerable interest to study the seasonal behaviour of Be-7 in relation to Cs-137.

Measurements of Cs-137 in surface air at

Table 1. *Be-7 and Cs-137 in Bombay Surface Air*

(Monthly Average Activities from Daily and Monthly Pooled Sample Countings)

Month	Be-7 $\mu\mu c/1000\ m^3$		Cs-137 $\mu\mu c/1000\ m^3$	
	Daily	Monthly	Daily	Monthly
1965				
December	67.2	59.5	5.4	4.9
1966				
January	107.5	100.7	10.5	8.6
February	104.4	117.4	12.5	14.5
March	142.0	144.7	25.4	20.0
April	122.5	133.4	24.1	19.5
May	77.3	86.4	13.5	12.0
June*	56.8	31.5	6.4	5.1
July*	56.2	82.3	6.7	3.9
August*	—	—	—	—
September	65.8	73.6	5.7	3.5
October	75.3	76.1	7.2	11.9
November*	110.4	135.0	9.9	8.5
December	87.1	64.7	4.9	5.1
AVERAGE	91.5	94.5	11.5	10.2

* Data likely to have large error due to presence of fresh activity from nuclear tests.

Bombay and several other stations in India have shown a seasonal variation with a regular increase in activity during the spring of each season similar to that observed in temperate latitudes (Rangarajan *et al.*, 1968) Since Cs-137 is a stratospheric source, with its behaviour as reference, the activity of Be-7 was studied to see if, from the relative variations of the two isotopes the origin of Be-7 sampled in ground level could be inferred.

Experimental

The measurement of atmospheric radioactivity is carried out by sampling about $3 \times 10^3\ m^3$ of air daily at Bombay using Hollingsworth and Vose H-70 filters. The filters are pressed into discs 2″ in diameter and ½″ thick, and counted using a 3″ × 3″ NaI (Tl) crystal and a 100-channel analyzer. A monthly composite sample is prepared from the daily filters by ashing below 400°C which is then counted once within a few weeks of collection date and again eight months later when Be-7 has completely decayed. From the second reading, the long-lived fission products Ce-144, Sb-125, Ru-106, Cs-137 and Mn-54 present in the samples are analysed by solution of the appropriate number of simultaneous equations using relevant photo-peak counts as described previously (Rangarajan and Mishra, 1966). However, Be-7 is not analysed using simultaneous equations as significant errors could result from slight instrumental drifts due to the close proximity of its photo-peak (478 kev) to the 510 kev gamma of Ru-106. Instead, Be-7 activity is calculated from the photo-peak counts B given by

$$B = P - (\mathrm{Ce}\, f_1 + \mathrm{Sb}\, f_2 + \mathrm{Ru}\, f_3 + \mathrm{Cs}\, f_4 + \mathrm{Mn}\, f_5) \quad (1)$$

Here P is the total counts in the photo-peak channels of Be-7 and Ce, Sb, etc., are the activities due to Ce-144, Sb-125, etc., and f_1, f_2, etc., are the factors which convert activities to counts in the photo-peak channels of Be-7 which can be calculated from the spectra of the sources of the isotopes concerned.

The above method involves taking two readings for each sample. This has been avoided in the analysis of the daily samples by the use of the following equation:

$$B = P - K\,\mathrm{Cs}\, F(R_1 f_1 + R_2 f_2 + R_3 f_3 + f_4 + R_5 f_5) \quad (2)$$

Here R_1, R_2, etc., are the ratios of the activities of Ce-144, Sb-125, etc., to Cs-137 activity, K the fraction of counts due to Cs-137 in Cs which is the total sample counts in Cs-137 photo-peak region and F the factor converting Cs-137 photo-peak counts to activity. R_1, R_2 etc., and K are calculated from the second reading of the monthly samples and are valid for the daily samples as they do not change much within a period of ±15 days. The fractions K and B/P are a measure of the "substractions" (due to compton interference) of other isotopes in the photo-peak regions of Cs-137 and Be-7. During the period November 1965 to May 1966, the values of these fractions were of the order of 0.8 as the activities of the other isotopes were very small and the analysis was found to be very reliable. After May 1966 however, fallout from the Chinese and French tests was present in the atmosphere, but the levels of contamination were such that except for short periods following the tests most samples could be processed for Be-7. Table 1 gives the monthly average activity calculated both from the daily measurements and the monthly pooled samples. The agreement between the two sets is quite

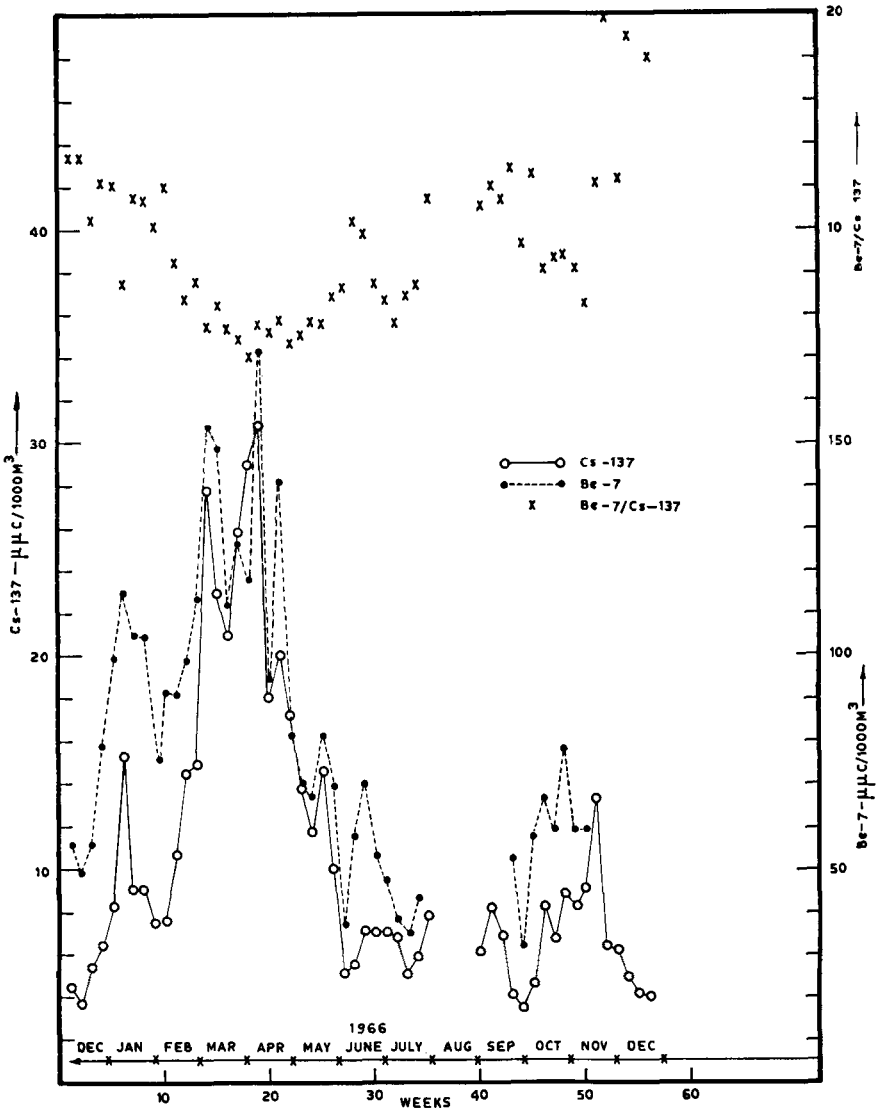

Fig. 1. Weekly average activity of Be-7 and Cs-137 in surface air at Bombay.

close showing the reliability of the method of analysis based on equation (2) for the daily samples.

Results and Discussion

Fig. 1 gives the seven-day averages of Be-7 and Cs-137 activity and their ratios. Cs-137 shows the usual spring increase and a corresponding increase is observed in the case of Be-7 also. Peak values are reached for both isotopes during the last week of March and the early part of April. The correspondence between Be-7 and Cs-137 is quite close and a calculation of the correlation coefficient (r) from the daily data for each month showed values ranging from 0.7 to 0.9. Both the isotopes are thus controlled by the same meteorological processes which evidently transfer them from the upper troposphere to ground level. However, on a long term basis an increase of Cs-137 relative to Be-7 is seen, the ratios reaching minimum values in March–April when both Cs-137 and Be-7 are at

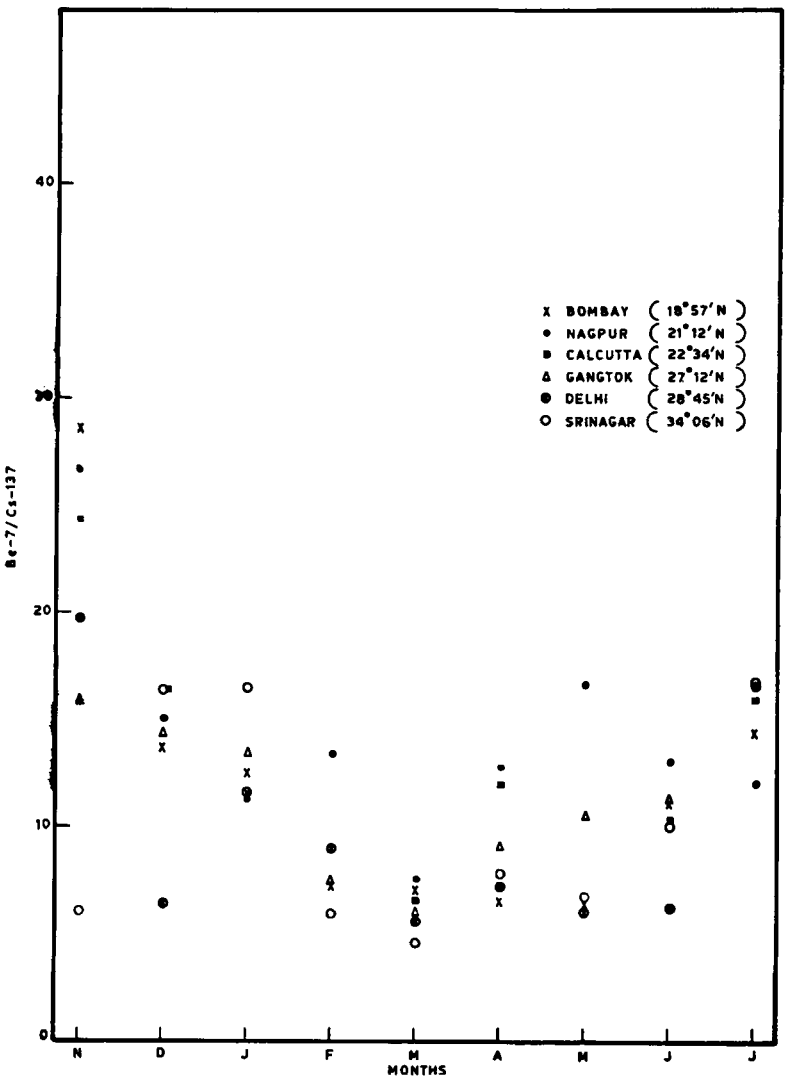

Fig. 2. Be-7/Cs-137 at different stations in India.

their highest concentrations. Hence although Be-7 increases along with Cs-137 and attains the maximum value at the same time, the amplitude of the spring increase is lesser, being about a factor of two as compared to about five for Cs-137. Monthly data on the ratio of Be-7/Cs-137 given in Fig. 2 for the other stations, show trends similar to that at Bombay. At these stations, only monthly measurements are made, as the volumes sampled are lesser, being only about 5000 m³.

On the basis of the above data, the origin of Be-7 can now be discussed. Since Be-7 is being continuously produced unlike Cs-137 which is slowly depleted from the stratosphere, it is reasonable to expect its spring rise to be comparable to that of Cs-137 if most of the Be-7 sampled in the troposphere originated from the stratosphere. The short-residence time in the lower polar stratosphere of about an year argues in favour of a transfer of large quantities of Be-7 in spite of its half-life of 53 days. However the results show the seasonal increase to be lesser in magnitude indicating that a part at least of the

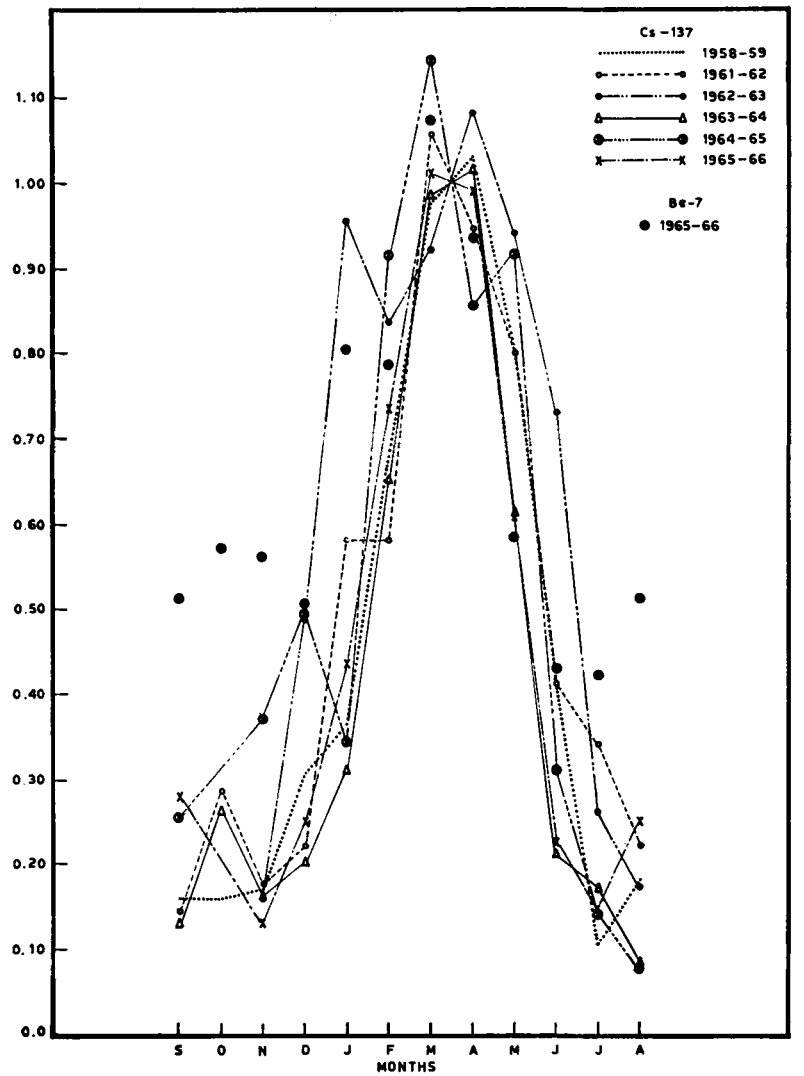

Fig. 3. Cs-137 activity in surface air at Bombay (Monthly values normalized to the average of March–April).

tropospheric activity is not of stratospheric origin. This argument is not necessarily conclusive as the amplitude of the spring increase may well depend on the stratospheric distribution of the trace substance. Lockhart (Lockhart *et al.*, 1960) observed that the ratio of Sr-89/W-185 increased during spring although both were of stratospheric origin. However, in this case Sr-89 was mainly from the polar stratosphere, while W-185 was concentrated in the equatorial stratosphere. The transfer of polar debris into the troposphere was faster than the equatorial debris. Be-7, on the other hand, is in higher concentrations in the high latitude stratosphere and its stratospheric distribution is likely to be similar to that of Cs-137 resulting from the high yield polar tests. A comparison of the Cs-137 activity during several years was made to study the extent of the variation in the spring increase. Fig. 3 gives the spring peaks observed for several years following the various high latitude tests of 1958, 1961 and 1962. In Fig. 3 the values for each spring cycle from September to August of the following year are normalized to the

average of the values of March and April when maximum activity is reached. Be-7 values for 1965–66 are also shown for comparison after normalization. Evidently the Be-7 increase in spring is less than that observed for Cs-137 during the various years when Cs-137 originally introduced into the polar stratosphere had mixed both vertically and horizontally. In view of this it is considered unlikely that Be-7 is predominantly from the stratosphere and a considerable fraction of Be-7 should have originated in the troposphere itself.

The fraction of Be-7 from the stratosphere during the spring maximum and autumn minimum can be easily calculated if it is assumed that the increase of the stratospheric component is similar to that of Cs-137 being about a factor of five and that the tropospheric component is more or less constant. On this basis stratospheric Be-7 would be about 60% in spring decreasing to 25% in the autumn.

Measurements of Be-7 in surface air have been reported by Shvedov (Shvedov *et al.*, 1962) at Leningrad (60° N), by Parker (Parker, 1962) at Sutton, U.K., (53° N), by Peirson (Peirson, 1963) at Chilton, U.K. (53° N), by Schumann (Schumann & Stoeppler, 1963) at Heidelberg (49° N) and by Gustafson (Gustafson *et al.*, 1961) at Argonne, U.S. (42° N). Most of these measurements, carried out in the middle latitudes during the moratorium of 1959–62 do show a "spring increase" somewhat similar to that observed for fission products although the amplitude of the increase varies from station to station. Junge (Junge, 1963) commenting on these differences remarks that the relative proportion of Be-7 originating from the stratosphere and that originating in the troposphere itself may vary depending on latitude since tropospheric production becomes more important at low latitudes while at latitudes greater than 30° N, lower stratospheric production becomes more important. This is consistent with our measurements which show a significant difference in the case of Be-7 from stratospheric Cs-137 measured during identical periods. In view of this all further use of Be-7 in studies of tropospheric process should take into account the presence of varying amounts of a possibly latitude dependent stratospheric component in the tropospheric air.

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СЕЗОННЫЕ ВАРИАЦИИ БЕРИЛЛИЯ-7 ПО ОТНОШЕНИЮ К ЦЕЗИЮ-137 В ПРИЗЕМНОМ ВОЗДУХЕ ТРОПИЧЕСКИХ И СУБТРОПИЧЕСКИХ ШИРОТ

В Бомбее и на других станциях в Индии в приземном слое воздуха было проведено сравнительное изучение сехонных вариаций Be-7, производимого космическими лучами, и искусственного продукта Cs-137. Результаты работы показывают, что концентрации обоих изотопов достигают максимальных значений весной в марте–апреле. Однако амплитуда роста для Be-7 много меньше, чем в случае стратосферного Cs-137. Разница объясняется тем, что только часть Be-7 в приземном слое имеет стратосфёрное происхождение. Путем сравнения с Cs-137 оценивается, что стратосферная компонента Be-7 составляет около 60% весной, уменьшаясь до 25% осенью.