# Atmospheric and precipitation radioactivity in India

By C. RANGARAJAN, SMT. SARADA GOPALAKRISHNAN, S. SADASIVAN and P. V. CHITALE, *Air Monitoring Section, Bhabha Atomic Research Centre, Bombay-74, India*

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ABSTRACT

The contribution of the major nuclear weapon test series in 1961 and 1962 to the ground-level activity in India during 1962–1965, has been estimated from fission product ratios in the ground-level air and precipitation samples collected at eight stations in India. Induced activity Mn-54 produced in the 1961 and 1962 tests has also been examined in relation to the fission product activity. Fission products Cs-137, Sb-125, Ru-106, Ce-144 and Zr-95, and induced activity of Mn-54 were estimated by gamma spectrometry. These measurements have also shown that, (*a*) while the well known spring increase is observed at all these stations, the time of peak values show significant differences at the various stations which could be related to the monsoon winds, (*b*) the gradient in activity along the latitude band of 10° N–32° N shows seasonal differences which can also be attributed to changes in surface winds, (*c*) Mn-54/Cs-137 activity ratios show that the radioactive debris present at high altitudes during 1962 had started descending to ground-level by the spring of 1963.

Concentrations of natural isotope Pb-210 were also measured at all these stations. Seasonal variations in Pb-210 differed markedly from the seasonal variations in Cs-137. However, both Cs-137 and Pb-210 show higher levels at the stations in the northern latitudes in India.

On the basis of the measured ground-level concentrations of fission products in 1963, 1964 and 1965, estimates of stratospheric residence time have been discussed.

## Introduction

Several radioisotopes existing in the atmosphere have been used for the study of air mass movements and mixing processes involved. Of these, radon and thoron daughter products have their source in the earth's crust while the cosmic-ray produced isotopes are created throughout the atmosphere in varying amounts. In recent years studies have also been made on the mixing processes using the fission products and other activity produced by nuclear tests. The source of these may be taken as the polar or equatorial stratosphere depending on the latitude of testing. Although some radioactivity is left in the troposphere by the tests, this is cleared within a short time and the radioactivity existing for considerable periods after testing is due to the stratospheric debris.

A simultaneous sampling of these activities from different sources at various points along the globe will yield important results on the exchange processes involved—both in the stratosphere and the troposphere. An attempt has been made in this direction by sampling atmospheric and precipitation radioactivity at several points in India and a few preliminary results of these measurements will be reported here.

## Experimental

### (a) *Gamma-emitting isotopes in fallout*

The isotopes measured include the long-lived gamma emitters Cs-137, Sb-125, Ru-106, Ce-144, Zr-95 and Mn-54. These measurements were carried out using a 3″ × 3″ NaI(Tl) crystal and a 100-channel pulse height analyser. The "unscrambling" of the gamma spectra was done as a six component system (Rangarajan & Mishra, 1966) after allowing a suitable time-lag for the decay of shorter-lived isotopes like Ce-141, Ru-103, etc., present in fission product mixtures. (Several other radionuclides existing in the atmosphere–both natural and artificial—such as Be-7, Co-57, Y-88, Na-22, etc., have either short half-lives or are of low abundance and do not interfere with the six component system if measurements are made after a

Table 1. *Fallout monitoring stations in India*

| Station | Altitude (m) | Latitude (N) | Longitude (E) |
|---|---|---|---|
| 1. Srinagar | 1598 | 34° 06′ | 74° 55′ |
| 2. Delhi | 219 | 28° 45′ | 77° 20′ |
| 3. Gangtok | 2000 | 27° 12′ | 88° 23′ |
| 4. Calcutta | Sea level | 22° 34′ | 88° 25′ |
| 5. Nagpur | 311 | 21° 12′ | 79° 04′ |
| 6. Bombay | Sea level | 18° 57′ | 72° 55′ |
| 7. Bangalore | 922 | 12° 57′ | 77° 30′ |
| 8. Ootacamund | 2235 | 11° 23′ | 76° 40′ |

suitable time-lag.) The large initial activity of (Zr-95 + Nb-95) enables the activity of this pair to be estimated after the other isotopes with short half-lives have decayed. In addition, after new test series, the activity of Ba-140 was estimated from the 1.60 MeV peak of its daughter product La-140 since there is no interference in this region from other isotopes.

The measurements of fission products in atmospheric particulates were done using the daily air filter collections made at different stations as part of the fallout monitoring

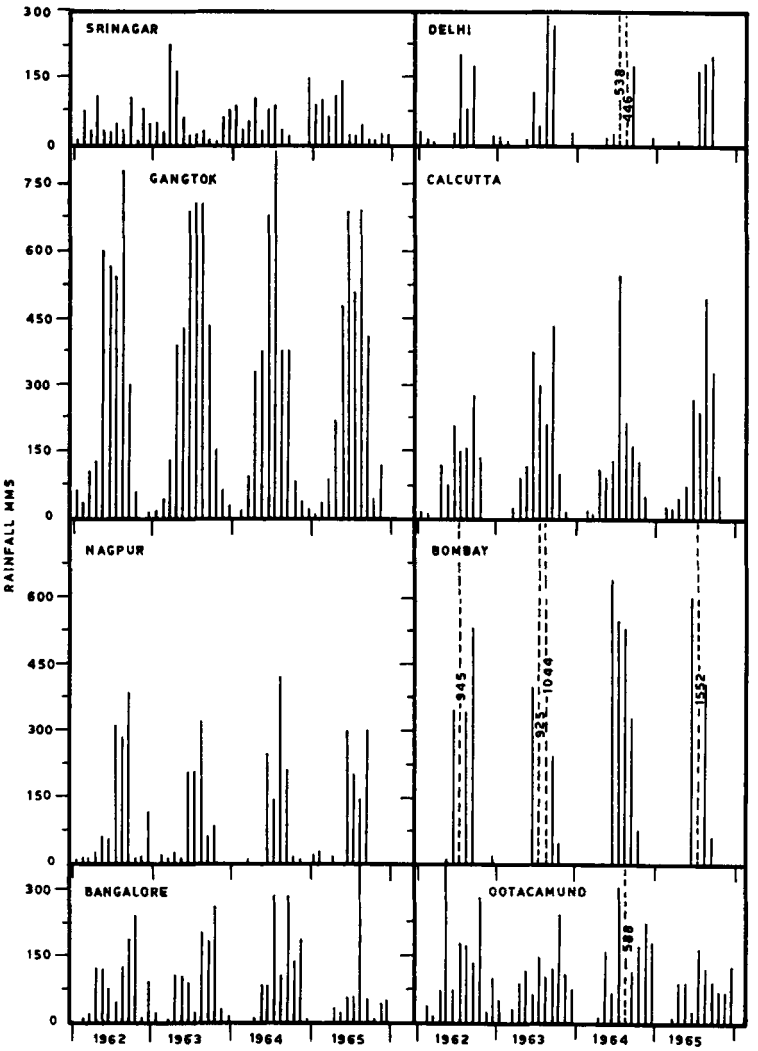


*Fig. 1.* Monthly rainfall at the fallout monitoring stations in India.

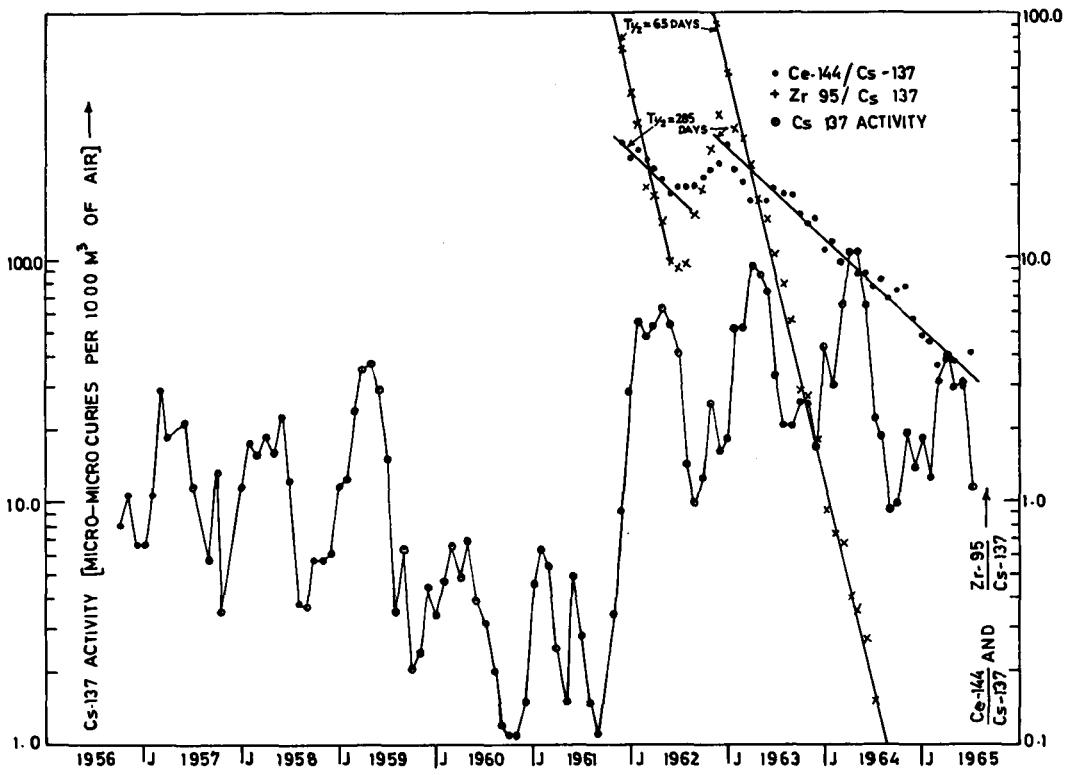


*Fig. 2.* Concentration of Cs-137 and ratios of Ce-144/Cs-137 and Zr-95/Cs-137 activity in ground level air at Bombay.

programme described previously (Vohra *et al.*, 1966; Rangarajan *et al.*, 1965*a*). Details of these stations are given in Table 1. The daily filters were counted for mixed fission product activity. The sample collections for one calender month were pooled for individual radionuclide analysis. The collected filter materials with dust were reduced to a few grams without loss of radioisotopes, by ashing below 400°C and treatment with nitric and hydrofluoric acids. The residues were transferred to perspex planchets of suitable sizes and counted for gamma activity in the scintillation spectrometer. Similar measurements were carried out on the monthly collection of rainwater and dust, made at the stations shown in Table 1, after evaporation of the water and transfer of the residue to identical planchets. The monthly rainfall at these stations is shown in Fig. 1.

(b) *Natural radioactivity*

Measurements have been made on RaD (Lead-210) activity in the surface atmosphere and precipitation. Lead-210 was chemically separated and estimated by counting for the daughter product Bismuth-210. The build-up of daughter product Bismuth-210 with a half-life of 5 days also serves as a check of the radiochemical purity of the separations. The 1.17 MeV beta of Bismuth-210 is easily counted in end window counters.

(c) *Errors*

The accuracy involved in the gamma spectrometric measurements has been estimated by several experiments which included analysis of parallel collections, repeated counting of samples with time intervals of several months and duplicate analysis of radionuclides by gamma measurements and radiochemistry (Rangarajan & Mishra, 1966). The results show that in general the sampling errors are better than a relative standard deviation of 13%, while for Cs-137 estimation the over all errors are better than 18%. For Mn-54 and Ce-144, the errors are in general less than 25% while for Ru-106

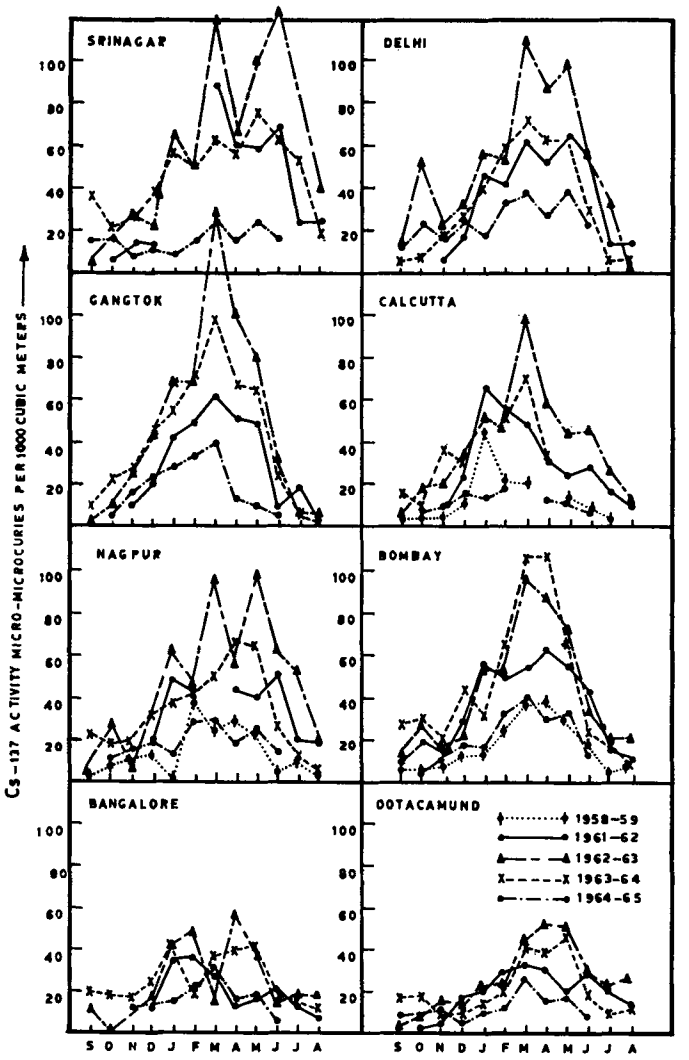


*Fig. 3.* Cs-137 activity in air at different stations in India (the spring cycle from September to August is shown).

and Sb-125 less than 30%. The relative concentrations of these radionuclides, the mutual overlap and orientation of their photo-peaks and slight instrumental drifts account for these differences in errors among the radioisotopes.

A similar calculation for the ratios of the activities of the isotopes from the various measurements shows that in general the variations are less than would be expected on the basis of the calculated errors in the individual isotopes. This is not surprising as part of the sampling error may cancel out when ratios are taken. The errors of the ratios calculated from the several repeat readings are shown in Fig. 6.

For rainwater samples, however, the above analysis does not apply. Most of the stations have seasonal rainfall depending on monsoon rains and activities of the samples vary considerably depending on rainfall amount, air activity and other factors. During several dry months or months with small precipitation, the accuracy of the measurements are low while during spring higher concentrations make more accurate analysis possible. However, when

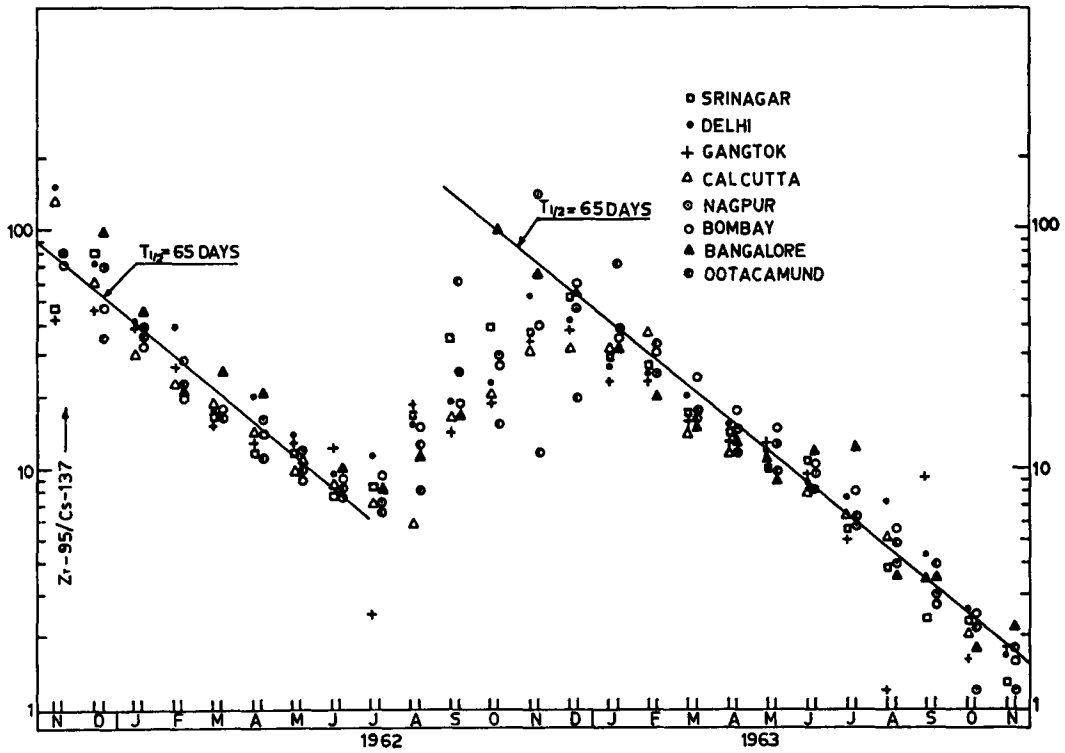


*Fig. 4.* Zr-95/Cs-137 activity ratios in ground level air at eight sampling stations in India.

yearly concentrations in rainwater are considered the larger errors of the dry months will be of little consequence in view of the small amount of activity deposited during these months.

The lead-210 samples are counted for a time long enough to give an accuracy better than $\pm 5\%$ (S.D.).

## Results and Discussions

(a) *Activity from different tests and ratios of the Isotopes*

Cs-137 activity in ground level air observed for the period 1956 to 1965 is shown in Figs. 2 and 3. In addition to several closely spaced test series up to the middle of 1958, there were three major series of tests near the Arctic in the autumns of 1958, 1961 and 1962 respectively and one test series in the equatorial regions of the Pacific during the summer of 1962 (U.S. AEC, 1964). The Cs-137 and other long-lived fission product activity from the Chinese and French tests can be neglected in view of their low yield (UNSCEAR Report, 1966). As many of these test series were not followed for several months by other tests, the ratios of selected pairs of fission products could be followed and their variations studied. Throughout the following text the terms Ba-140/Zr-95 ratio, Zr-95/Cs-137 ratio, Ce-144/Cs-137 ratio and Mn-54/Cs-137 ratio represent the ratios of activities of the isotopes.

The ratios of Zr-95/Cs-137 and Ba-140/Zr-95 in the air at the eight stations are given in Figs. 2, 4 and 5 for several months following the end of the 1961 test series. The data show that for the two periods following the autumn tests of 1961 and 1962 respectively, the ratios of Zr-95/Cs-137 decreased with the half-life of Zr-95 (65 days) for several months indicating that the activity from the various test series are in constant proportions. For the first period this trend continues only up to May, 1962 when fresh activity was introduced again by the Pacific and later by the Arctic tests. After the cessation of tests by the end of 1962, however, the ratios of Zr-95/Cs-137 as well as the Ce-144/

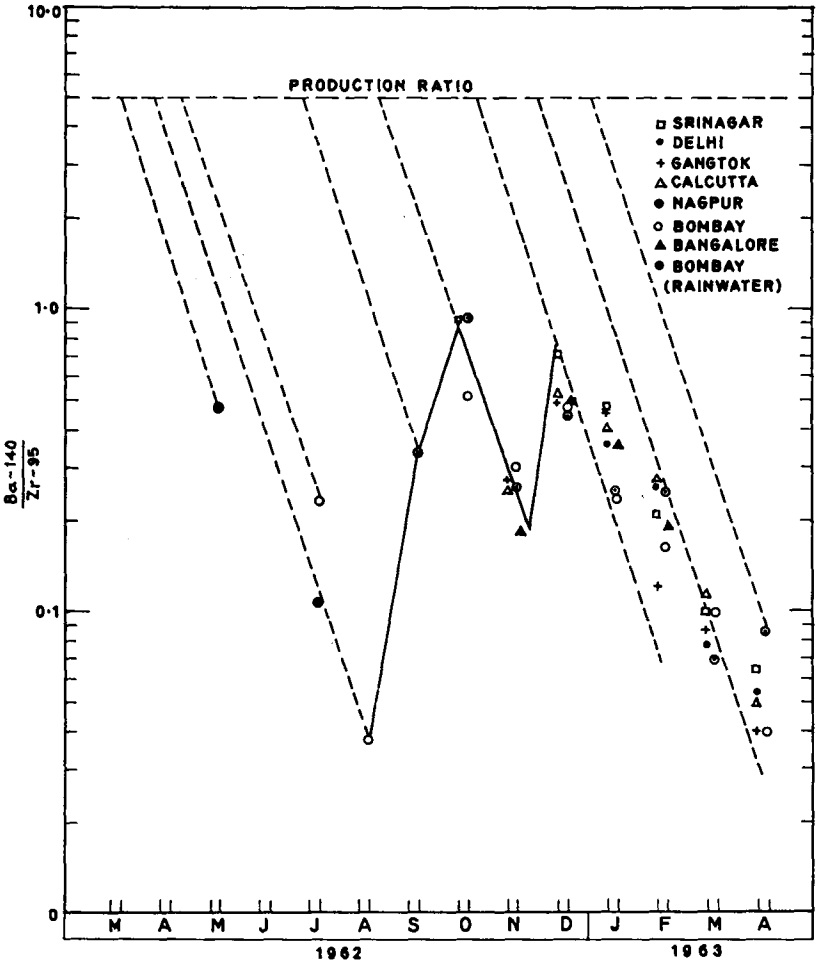


*Fig. 5.* Ba-140/Zr-95 activity ratios in air and rainwater (dashed lines indicate the theoretical decay of Ba-140/Zr-95 activity ratio).

Cs-137 indicate that no major change in the relative proportions of activity from the different sources took place for a period of nearly thirty months, i.e., through June, 1965.

This is unlike 1959 when in late summer the ratios of Zr-95/Cs-137 departed from the half-life of 65 days indicating the arrival of increasing proportions of older activity (Vohra & Bhatnagar, 1961). The absence of such a change for several months is significant considering that activities from not less than three sources, viz., pre-1961 tests, autumn tests of 1961 and 1962 and Pacific tests of 1962, were present in the atmosphere.

This difference is likely to be due to the presence in 1959 of different sources of activity at various heights in the stratosphere. The debris from the low yield 1958 autumn tests were confined mainly to the lower stratosphere (Feely & Friend, 1963). Above this, older debris from Hardtack and other tests were present in large quantities. The younger debris from the lower stratosphere was rapidly removed during the spring of 1959 and in summer increasing quantities of the older debris was descending to the lower stratosphere and entering the troposphere. During the autumn tests of 1961 and 1962 there were several explosions of very high yield in addition to low yield tests. It is likely therefore that the

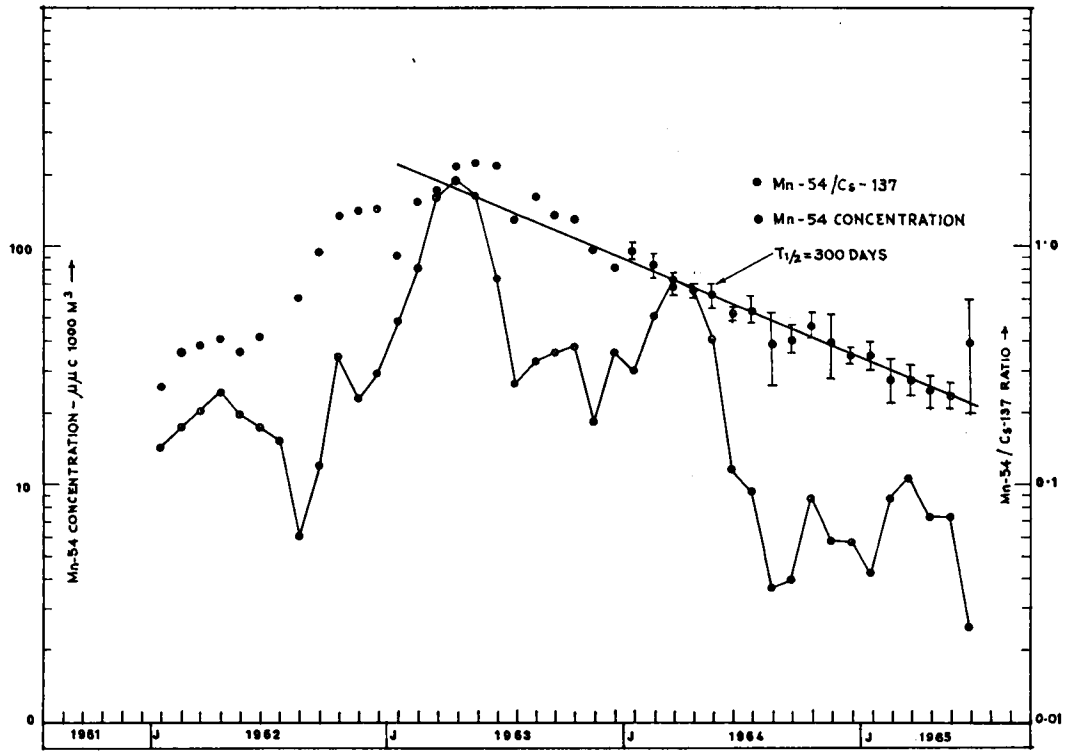


*Fig. 6.* Ratios of Mn-54/Cs-137 activity and concentrations of Mn-54 in ground level air at Bombay (vertical lines indicate standard deviation).

stratosphere was more or less uniformly contaminated with activity from the two series to very great altitudes. Hence as the debris from the upper stratosphere gradually entered the troposphere through the years, no major change in the isotopic ratios took place during 1963, 1964 and 1965.

Evidence for this is also available in the Mn-54/Cs-137 ratios, which after a more or less continuous increase throughout 1962 showed uniform decrease from spring 1963 through the summer of 1965 (Fig. 6). The increase of the ratio through 1962 is due to the appearance of radioactive materials from high altitudes. This is also supported by the data on the vertical profiles of Mn-54 and Sr-90 given by Feely *et al.* (1963) for various periods. These profiles show that in January 1962, Mn-54 was nearly two orders of magnitude greater than Sr-90 at 65,000 feet. At 40,000 feet the activities were very nearly the same. However, by July 1962 the Mn-54 activity even at 40,000 feet was nearly an order of magnitude higher than Sr-90, evidently due to the downward movement by diffusion of the debris from higher altitudes. This trend continued up to April 1963. Fresh Mn-54 was again introduced in October 1962 (Feely *et al.*, 1963) although this does not appear to have changed the relative magnitudes to any appreciable degree, the difference being about an order of magnitude from July to December 1962. Hence the increase in Mn-54/Cs-137 ratios at ground level may be explained to be due to the downward movement by diffusion of high altitude debris of 1961 tests although the 1962 tests could also have contributed partly to this increase. However, since the ratios were increasing from July 1962 onwards reaching high values by October 1962, the debris from the 1961 tests is likely to be the important, though not the only cause of the increase in Mn-54/Cs-137 ratio. Data given by Feely *et al.*, (1963) also show that in December 1962, the relative magnitudes of the two isotopes were constant to great altitudes, i.e., up to 65,000 feet suggesting that

Table 2. *Ratios of fission product activity in air and rainwater*

| Period | Stations | | Fission product activity ratios: Ce-144/ Cs-137 | Sb-125/ Cs-137 | Ru-106/ Cs-137 | Zr-95/ Cs-137 | Remarks |
|---|---|---|---|---|---|---|---|
| | | | *November 1961 to May 1962* | | | | |
| I | Srinagar | (*a*) | — | — | — | 95.6 | The individual nuclide values in the period Nov. 1961–May 1962 have been corrected back to a common date, October 15, 1961, and the ratios calculated |
| | | (*b*) | 26.4 | 1.2 | 9.3 | 88.5 | |
| | Delhi | (*a*) | — | — | — | 125.0 | |
| | | (*b*) | — | — | — | 101.9 | |
| | Gangtok | (*a*) | — | — | — | 95.5 | |
| | | (*b*) | — | — | — | 120.0 | |
| | Calcutta | (*a*) | — | — | — | 92.3 | |
| | | (*b*) | — | — | — | 73.2 | |
| | Nagpur | (*a*) | — | — | — | 108.0 | |
| | | (*b*) | — | — | — | 105.0 | |
| | Bombay | (*a*) | 33.4 | 1.0 | 7.8 | 90.3 | |
| | | (*b*) | — | — | — | — | |
| | Bangalore | (*a*) | — | — | — | 120.6 | |
| | | (*b*) | — | — | — | 118.9 | |
| | Ootacamund | (*a*) | — | — | — | 84.3 | |
| | | (*b*) | — | — | — | — | |
| | Average | (*a*) | 33.4 | 1.0 | 7.8 | 101.5 | |
| | | (*b*) | 26.4 | 1.2 | 9.3 | 101.3 | |
| | | | *January 1963 to June 1964* | | | | |
| II | Srinagar | (*a*) | 28.0 | 1.3 | 6.3 | 69.5 | The individual nuclide values in the period Jan. 1963–June 1964 have been corrected back to a common date, November 15, 1962, and the ratios calculated |
| | | (*b*) | 25.9 | 1.1 | 10.2 | 71.4 | |
| | Delhi | (*a*) | 29.2 | 1.2 | 6.4 | 75.6 | |
| | | (*b*) | 26.4 | 1.5 | 9.9 | 98.7 | |
| | Gangtok | (*a*) | 26.8 | 1.1 | 5.5 | 66.5 | |
| | | (*b*) | 25.3 | 1.8 | 12.2 | 53.3 | |
| | Calcutta | (*a*) | 26.4 | 1.0 | 6.0 | 64.8 | |
| | | (*b*) | 26.6 | 1.2 | 11.1 | 70.7 | |
| | Nagpur | (*a*) | 31.0 | 1.5 | 6.5 | 71.1 | |
| | | (*b*) | 24.6 | 1.2 | 9.1 | 75.7 | |
| | Bombay | (*a*) | 28.2 | 1.4 | 7.7 | 82.4 | |
| | | (*b*) | 24.8 | 0.8 | 10.3 | 58.2 | |
| | Bangalore | (*a*) | 30.1 | 1.6 | 6.9 | 67.5 | |
| | | (*b*) | 23.1 | 1.6 | 11.2 | 73.8 | |
| | Ootacamund | (*a*) | 29.6 | 1.6 | 6.8 | 77.1 | |
| | | (*b*) | 22.6 | 1.1 | 9.4 | — | |
| | Average | (*a*) | 28.7 | 1.3 | 6.5 | 71.8 | |
| | | (*b*) | 26.8 | 1.3 | 10.4 | 71.7 | |

(*a*) Airborne activity. (*b*) Rainwater activity.

further downward movement by diffusion of high altitude debris is not likely to disturb the ratios.

During the two periods (October 1961 to May 1962 and January 1963 to June 1965) when the activity from the various test series remained more or less constant for several months, the relative magnitudes of activity from the contributing sources can be estimated by using the ratios of selected pairs of isotopes. Table 2 gives the ratios of the several isotopes measured, both in air and in rainwater. These ratios have been derived by decay correcting the activities to the mean "date" of the last test series and then calculating the ratios of the activities.

From weapon yield considerations given in a HASL report (U.S. AEC, 1964), an average date of October 15, 1961, was estimated for the autumn 1961 tests. Derivation of an average date for the second period is difficult

Table 3. *Fission product ratios for various modes of fission and observed values in fallout*

| | Ce-144/ Cs-137 | Sb-125/ Cs-137 | Ru-106/ Cs-137 | Zr-95/ Cs-137 | Zr-95/ Ce-144 | Zr-95/ Sb-125 | Zr-95 Ru-106 |
|---|---|---|---|---|---|---|---|
| 1. U-238 14 MeV fission[a] | 22.3 | 2.6 | 16.2 | 138.0 | 6.2 | 60.0 | 8.5 |
| 2. U-238 fast fission[a] | 26.4 | 0.1 | 12.0 | 144.0 | 5.5 | 1245.0 | 11.9 |
| 3. U-235 fast fission[a] | 29.6 | 0.3 | 2.3 | 183.0 | 6.2 | 620.0 | 81.0 |
| 4. Pu-239 fast fission[a] | 22.9 | 0.6 | 29.8 | 141.0 | 6.1 | 224.0 | 4.8 |
| 5. U-235 thermal fission[a] | 33.7 | 0.06 | 1.9 | 176.0 | 5.2 | 3140.0 | 5.2 |
| 6. Samples of H. Bomb debris[b] | 32.5 | 0.8 | 13.0 | 153.0 | 4.7 | 196.0 | 11.8 |
| 7. Fallout activity, Nov. 61–May 62 | 41.0[c] | 1.3[c] | 11.8[c] | — | 3.4 | 106.0[c] | 11.7 |
| 8. Fallout activity Jan. 63–June 64 | 42.7[c] | 1.7[c] | 12.5[c] | — | 3.2[c] | 80.0[c] | 11.1[c] |

[a] Data given by R. Bjornerstedt, *Arkiv för Fysik*, Band 16, nr 28 (1959).
[b] Data given by Hallden *et al.*, USAEC Report HASL-117.
[c] The ratios are obtained after removing old activity, as explained in the text.

by the above procedure as the autumn tests were spaced over a period ranging from August to December 1962. Hence an average date of November 15, 1962, was estimated from the ratios of Ba-140/Zr-95 (Fig. 5) as these two short-lived isotopes were predominantly from the 1962 autumn tests.

A comparison of the data given in Table 2 and Fig. 4 shows no appreciable difference in ratios among the various stations and between air activity and rainwater due to the selective scavenging of the isotopes by rainfall. Only Ru-106 levels are about 40% lesser relative to Cs-137 in air as compared to rainfall activity. This difference is of no great significance for the discussions to follow. Hence the overall averages of all the stations—both rainfall and air activity—can be taken as representative of the isotopic ratios in fallout during the two periods.

(b) *Partitioning of activity from different sources*

With these data, partitioning of activity can now be done. All the Zr-95 in period one (Table 2) is from the 1961 tests as the dates of the previous tests make Zr-95 from them negligible. A major fraction of Zr-95 during the second period is also from the autumn 1962 tests as shown by Ba-140/Zr-95 and Zr-95/Cs-137 ratios (Figs. 4 and 5). The presence of the U.S. Pacific tests of 1962 is shown by the Ba-140/Zr-95 ratios in May and July in Bombay rainwater and the increase in Zr-95/Cs-137 ratios in June and July. From August onwards, the Zr-95/Cs-137 ratio increases rapidly till the end of the autumn tests in December 1962. This continuous influx of fresh activity, shown also by the high values of Ba-140/Zr-95 throughout the period, is due to the polar tests of autumn 1962. Although there were a few Pacific tests in October 1962, these were of comparatively low yield. Also the total yield of the Pacific tests was 11 MT against 60 MT of the autumn polar tests of 1962 (U.S. AEC, 1964).

To derive the fraction of Cs-137 from the recent tests, the production ratio of Zr-95/Cs-137 was taken as 138 (Rangarajan *et al.*, 1965*b*) for 14 MeV fission of U-238. This is considered the most likely mode of formation of fission products in long range fallout as will be shown later. However, the Zr-95/Cs-137 ratio varies little for other modes of fission (Rangarajan *et al.*, 1965*b*). If a Zr-95/Cs-137 value of 101 is taken for the first period and 72 for the second period (Table 2), the proportion of Cs-137 from the 1961 autumn tests would be 73% during spring 1962 and 52% from 1962 autumn tests after end of 1962. (These values differ somewhat from the previous values of 60% and 55% given by us on the basis of airborne data at Bombay only (Rangarajan *et al.*, 1965*a*).) These fractions of fresh Cs-137 are subject to the accuracy of the average date chosen. For the first period, it is unlikely that the date is in error by more than fifteen days prior to the date chosen. This will

Table 4. *Average Cs-137 concentration in ground level air at the different sampling stations ($\mu\mu c/1000\ M^3$)*

| Station | 1957 | | 1958 | | 1959 | | 1960 | 1961 | 1962 | | 1963 | | 1964 | | 1965 |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| | *a* | *b* | *a* | *b* | *a* | *b* | | | *a* | *b* | *a* | *b* | *a* | *b* | *a* |
| Srinagar | | | | | | | | | 68.6 | 20.5 | 87.9 | 41.2 | 60.9 | 20.7 | 17.3 |
| Delhi | | | | | | | | | 53.5 | 24.4 | 76.2 | 15.4 | 54.2 | 14.3 | 29.0 |
| Gangtok | | | | | | | | | 43.5 | 17.5 | 83.4 | 19.3 | 64.2 | 10.7 | 21.5 |
| Calcutta | | | | | | | | | 42.1 | 17.8 | 57.6 | 22.4 | 44.9[a] | 8.8[b] | 12.9 |
| Nagpur | | | | | | | | | 45.1 | 17.8 | 70.8 | 27.6 | 47.4 | 11.5 | 20.3 |
| Bombay | 18.1 | 8.6 | 17.1 | 6.1 | 25.5 | 3.7 | 3.4 | 6.1 | 52.3 | 16.6 | 65.4 | 26.8 | 60.8 | 14.8 | 26.2 |
| Bangalore | | | | | | | | | 25.0 | 10.6 | 35.5 | 19.4 | 31.9 | 14.1 | 18.5 |
| Ootacamund | | | | | | | | | 26.6 | 12.2 | 36.2 | 15.8 | 28.5 | 8.8 | 14.6 |

*a:* First half. *b:* Second half.

[a] Data for the period February–March only.
[b] Data for the period October–December only.

increase the proportion of recent Cs-137 to about 85%. In the second period, a larger error is possible as polar tests were spread over a period of five months. A later average date than the one chosen by us is unlikely but a date 30–40 days earlier is possible. Fresh Cs-137 in this case would be as high as 75–80%.

The above fractions of fresh Cs-137 can be used to calculate the ratios of fission products characteristic of the radioactivity released by the 1961 and 1962 tests. Following the 1961 series as 70% of Cs-137 is from the recent tests, all the activity of the other isotopes with medium half-lives may be taken as from the latest tests. The major test series prior to 1961 were in 1958 and the fraction of isotopes like Ce-144, Ru-106, etc., from them will be less than 10–15% in spring 1962. The ratios calculated on this basis are given in Table 3.

For the second period, partitioning of activity for isotopes other than Cs-137 is done using fractions of new and old Cs-137, ratio of the particular isotope to Cs-137 actually observed and extrapolated from the period prior to the arrival of fresh activity. The proportion of "new activity" for the other isotopes is 70–75%. Hence, subject to the errors in partitioning, the ratios given in Table 2 may be taken as representative of the autumn tests of 1961 and 1962. (Possible changes in the average dates discussed before will not affect the ratios by more than 10% for the first period and 20% for the second period.) The values can be compared with the ratios for the various modes of fission given in Table 3. The yields of nuclides like Ru-106, Sb-125 etc., situated in the valley of the fission yield curve, are very sensitive to the energy of the neutrons causing fission and the ratios involving these provide a good indication of the mode of fission. The data indicate that the ratios closely correspond to 14 MeV and fast fission of U-238.

(c) *Seasonal and latitudinal variation of activity*

The spring increase in stratospheric fallout mentioned previously (Rangarajan *et al.*, 1965*a*) is evident at all the stations in our monitoring net-work extending from 35° N to 11° N (Fig. 3). An examination of the data of Fig. 3 shows that peak values are reached during the months of March–April or May at all the stations except Srinagar where May and June have the highest values. The contrast during the month of June is quite striking as when the values are high at Srinagar, at all the other stations the concentrations are reduced considerably compared to the peak values.

The latitudinal variation of activity through the seasons also shows interesting results. Fig. 7 shows the latitudinal profiles of Cs-137 in air during the years 1962, 1963 and 1964 for the four seasons. There appears to be a gradient in concentration falling with latitude in the first two seasons. However, with the onset of monsoon in June, this gradient is broken down, the levels being similar except at Srinagar which continues to be higher.

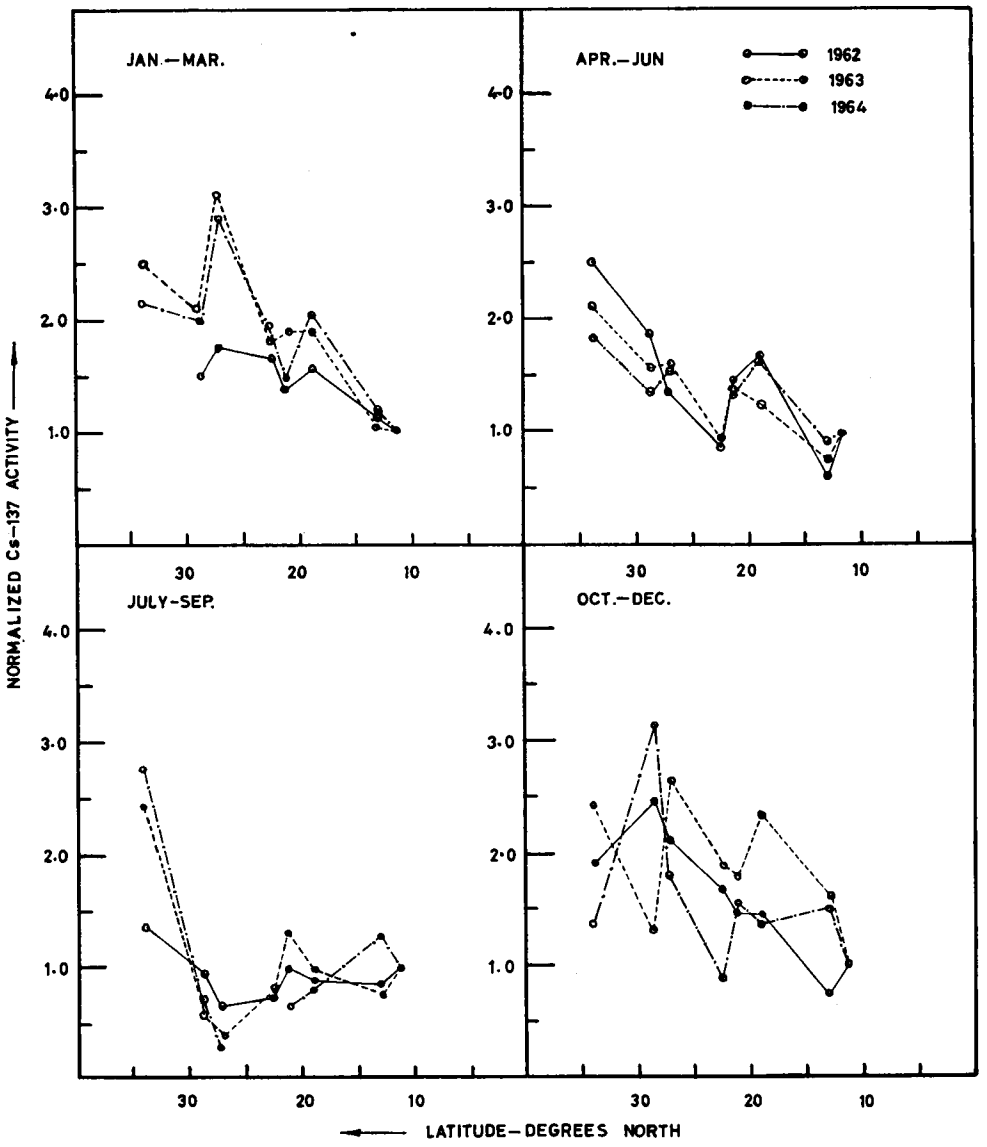


*Fig. 7.* Latitudinal variation of Cs-137 activity in air at the eight stations in India (quarterly averages of activity per standard cubic meter of air normalized to Ootacamund values).

The earlier fall in activity at the stations other than Srinagar could be due to the onset of the monsoon during the summer months. Surface winds over India are from north-west in winter and early spring (WMO, 1965) when the gradient in activity is maintained. The effect of the change into the powerful monsoon current originating near the equatorial regions and having low specific activity could be a lowering of the activity levels. Evidently the monsoon regime is not very effective at Srinagar in view of the continued higher activity during this period.

The graph of the yearly specific activity of Cs-137 in rain (Fig. 8) shows latitudinal profiles similar to the air activity during the monsoon period of June–August. The specific activities are generally of the same order showing a sharp

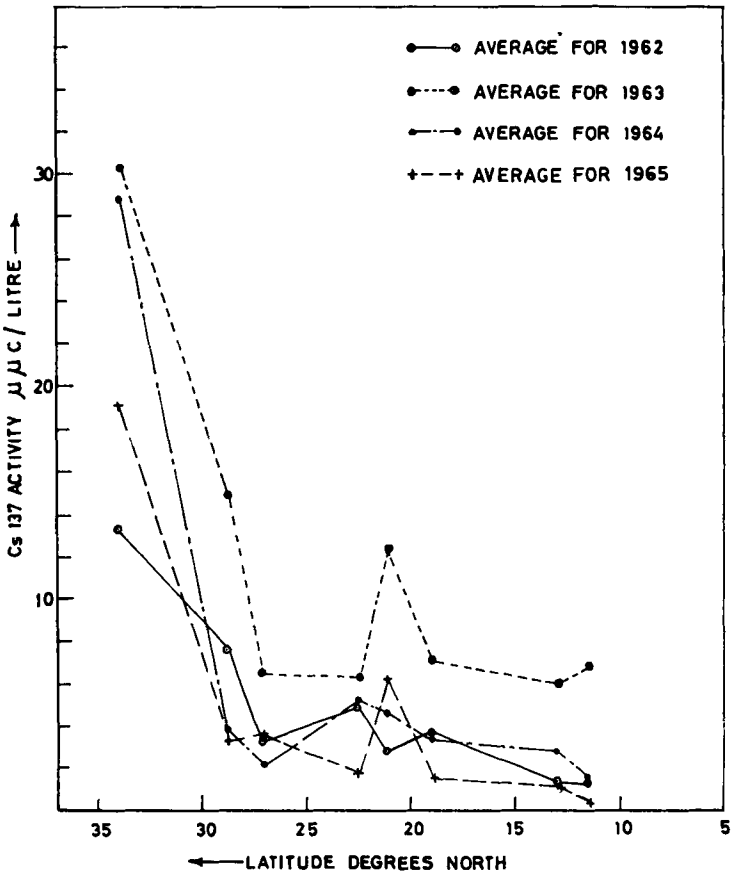


*Fig. 8.* Latitudinal variation of Cs-137 activity in rainwater at the eight stations in India.

increase towards Srinagar. This is not surprising as rainfall is mainly due to monsoon at most of the stations unlike Srinagar which has rainfall more or less throughout the year with higher rains during winter and spring when air activity levels are maximum (Fig. 1). The winter rains at Srinagar and parts of northern India are due to the movement of low pressures which originate in the mediterranean region and travel across the north of India. The air masses from near temperate latitudes could have higher associated specific activity than the rains of the monsoon current originating near equatorial regions. These factors explain the much higher specific activity of rainfall at Srinagar.

(d) *Natural activity due to Lead-210*

Lead-210 activity in Bombay air (Fig. 9) shows a seasonal variation with maximum in winter (January–February) and minimum in summer (June–August). This seasonal variation is however not similar to the variation of Cs-137 and other stratospheric sources as the peaks in the latter occur about three months later. Preliminary measurements by the authors showed that there is some evidence that RaD variations are similar to short-lived radon daughter product variations and it is likely that RaD activity at Bombay are dependent predominently on radon concentrations.

RaD in air at Srinagar (Fig. 9) also appears to show a similar variation although this is not as prominent as at Bombay. The yearly average concentrations of Lead-210 in all the eight stations both in ground level air and precipitation are given in Fig. 10. High latitude stations do show higher specific activity in rain although this is not so clear in air data.

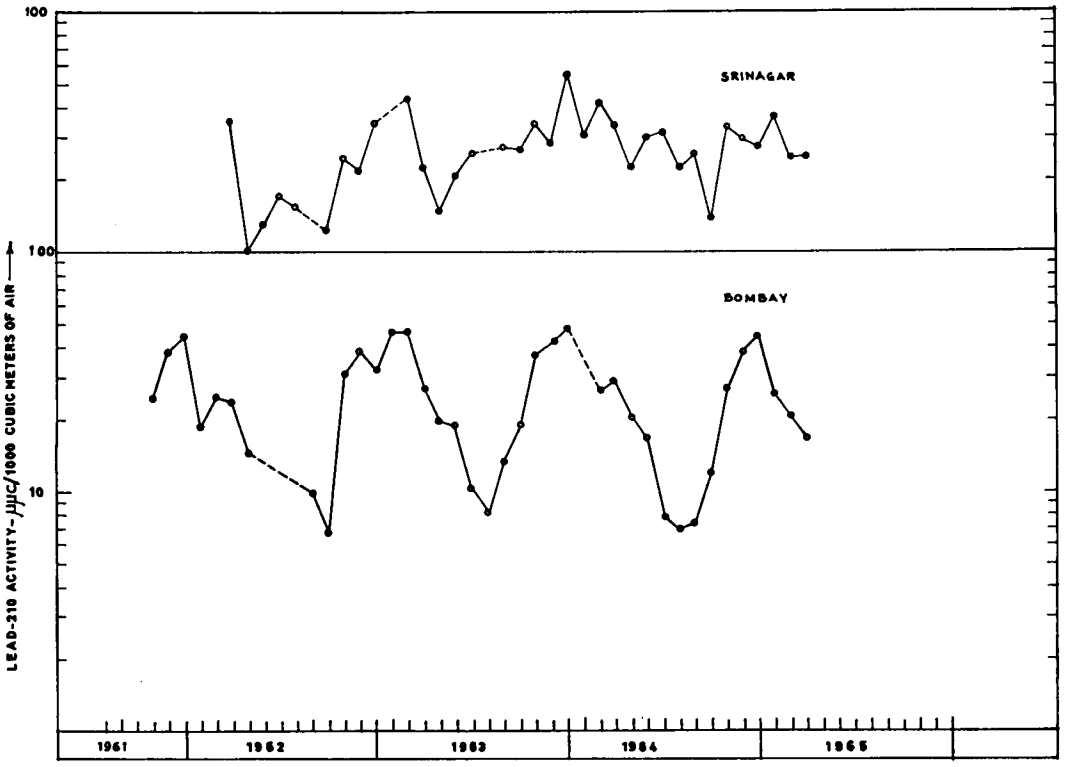


*Fig. 9.* Lead-210 (RaD) activity in ground level air at Bombay and Srinagar.

Comparison of RaD and Cs-137 activity (Table 5) shows that the relative concentrations in air and rain are of the same order implying that the scavenging processes are not very different for the two isotopes.

(e) *Estimation of stratospheric residence times*

Fallout activity at ground level is controlled mainly by stratospheric exchange processes although regional and local influences can have an effect. If a residence time is assumed for the stratospheric-tropospheric exchange of debris then the stratospheric depletion will be given by $N_t = N_0 e^{-\lambda t}$, the rate of transfer of debris into troposphere being $(dN/dt)t = -\lambda N_t$. Here $N_t$ is the stratospheric burden and $1/\lambda$ the mean residence time. Since a seasonal cycle of variations is present, the evaluation of $\lambda$ would depend on a comparison of activity levels in corresponding periods of the year to eliminate variations due to the annual oscillations. Cs-137 data given in Table 4 shows that 1964 levels are 70 to 90 % of the values in 1963 suggesting a long residence time. However, during 1965 the levels are only 30–40 % of the levels of the previous years. This small reduction in activity levels in 1964 compared to 1963 is due to the downward diffusion of activity of high altitude debris to the lower stratosphere, which feeds the troposphere. The greater reduction in 1965 probably indicates a depletion of the reservoir

Table 5. *Specific activity of cesium-137 and lead-210 at Srinagar*

| | Cs-137 | | Pb-210 | |
|---|---|---|---|---|
| Year | Air μμc/1000 m³ | Rain μμc/l | Air μμc/1000 m³ | Rain μμc/l |
| 1962 | 40.0 | 13.3 | 22.0 | 7.8 |
| 1963 | 65.0 | 30.3 | 29.8 | 6.4 |
| 1964 | 42.0 | 28.9 | 27.5 | 6.1 |

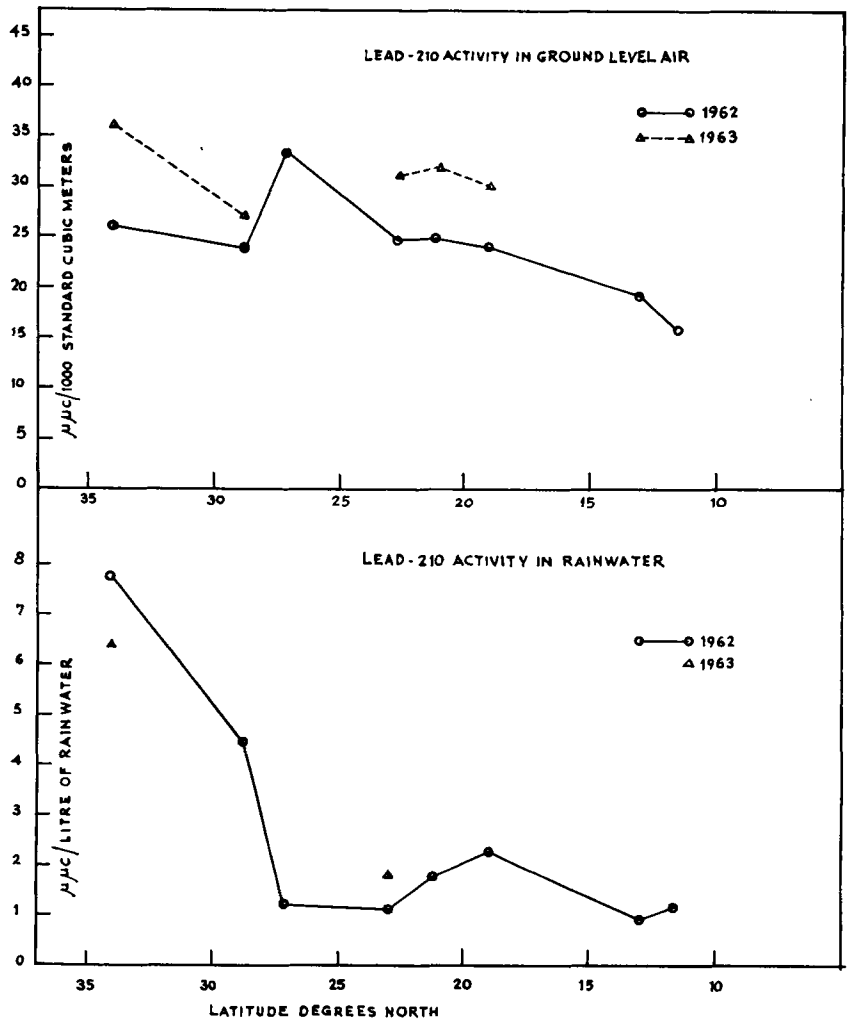


*Fig. 10.* Latitudinal variation of Lead-210 activity in rain and ground level air at the eight stations in India.

of activity in the upper stratosphere. In view of this the above equations can be modified as

$$N_l(t) = N_u(0)\frac{\lambda_1}{\lambda_2 - \lambda_1}[e^{-\lambda_1 t} - e^{-\lambda_2 t}] + N_l(0)\,e^{-\lambda_2 t}.$$

Here $\lambda_1$ and $\lambda_2$ refer to the residence times in upper and lower stratosphere and $N_l$ and $N_u$ are the corresponding stratospheric burdens. If a value of one year is taken as the residence time for the lower stratosphere from previous experience (Vohra & Bhatnagar, 1961) and $N_u = N_l$ in early 1963 (Feely *et al.*, 1963), the best fit for the data is given by a value of $1/\lambda_1 = 9$ to 10 months. Extrapolation of the expected value for 1966 shows activities of the order of about 15% of the 1963 levels. Preliminary measurements on Cs-137 in air do confirm the existence of activity levels of this order. This would imply that the transfer of material from the higher stratosphere to the lower stratosphere above the tropopause is fairly fast, being of the same order as the transfer rate into the troposphere.

The above estimates of residence times may not be valid for other cases, for instance, a large injection of activity in the equatorial regions or in the southern hemisphere.

Once in the troposphere the debris is subjected to regional and local influences as shown by the variations in time of peak values and amplitudes of air activity discussed above. That the troposphere is rather well mixed appears to be evident from the close correspondence of Zr-95/Cs-137 ratios at the various monitoring stations in India and elsewhere in the northern hemisphere (Rangarajan *et al.*, 1965*a*). For rainwater the factors are more complex with sources of rain formation, seasons of deposition, etc., influencing the activity levels.

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## REFERENCES

Feely, H. W. & Friend, J. P. 1963. *Evidence on stratospheric circulation from measurements of radioactive bomb debris injected above 20 km.* Paper presented at the XIII General Assembly of IUGG on August 21.

Feely, H. W., Davidson, B., Friend, J. P., Lagomarsino, R. J. & Leo, M. W. M. 1963. *The ninth quarterly report on Project Star Dust.* U.S. Defence Atomic Support Agency Report No. DASA 1309.

Rangarajan, C. & Mishra, U. C. 1966. Methods of standardization of fallout measurements in India. *Proceedings of the All India Symposium on Radioactivity and Metrology of Radionuclides, March 14–18.*

Rangarajan, C., Sarada Gopalakrishnan, Sadasivan, S. & Chitale, P. V. 1965*a*. *Measurements of airborne radioactive fallout in India.* Atomic Energy Establishment Trombay Report No. AEET-208.

Rangarajan, C., Mishra, U. C., Lalit, B. Y., Sarada Gopalakrishnan & Sadasivan, S. 1965*b*. *Fission Product data and its application in studying fallout from nuclear weapon tests.* Atomic Energy Establishment Trombay Report No. AEET-209.

Vohra, K. G. & Bhatnagar, V. S. 1961. Observations of fallout in India during the period of cessation of nuclear tests. *Nature 189*, 286–288.

Vohra, K. G., Rangarajan, C., Sarada Gopalakrishnan, Sadasivan, S. & Chitale, P. V. 1966. *Measurements on airborne and surface fallout radioactivity in India from nuclear weapon tests.* Atomic Energy Establishment Trombay Report No. AEET-247.

*Health and Safety Laboratory Report HASL-142*, U.S. AEC, 1964.

U.N. Scientific Committee on the effects of Atomic Radiation Report 1966, Official Records, General Assembly, 21st session, Suppl. 14 (A/6314).

WMO 1965, *Meteorology in the Indian Ocean*, WMO, No. 166 TP. 81, Geneva.

## РАДИОАКТИВНОСТЬ АТМОСФЕРЫ И ОСАДКОВ В ИНДИИ

В работе оценен вклад в радиоактивность на уровне земли в Индии от основных серий испытаний ядерного оружия в 1961 и 1962 годах в течение 1962–1965 гг. по отношениям продуктов деления в воздухе на уровне земли и по пробам осадков, проведенным на восьми станциях. Исследовалось также отношение индицированной активности Mn-54 к активности продуктов деления. Активность Cs-137, Sb-125, Ru-106, Ce-144, а также in-Cs-137, Sb-125, Ru-106, Ce-144, а также индуцированная активность Mn-54 были оценены при помощи $\gamma$-спектрометра.

Проведенные измерения показали: а) существенное различие в моменте появления пика активности на различных станциях, что можно объяснить наличием муссонных ветров; б) наличие сезонных изменений градиента °активности вдоль широтного пояса 10°N–32°N, которые также можно объяснить изменениями ветра у поверхности; в) отношение активностей Mn-54/Cs-137 показывает, что радиоактивные осколки, присутствовавшие на больших высотах в течение 1962 г. к весне 1963 г. начали опускаться до земли. На всех восьми станциях была также измерена концентрация природного изотопа Pb-210. Сезонные вариации в содержании Pb-210 значительно отличались от соответствующих изменений Cs-137, однако, для обеих изотопов на северных станциях был зафиксирован высокой уровень радиоактивности. На основе измеренных в 1963–65 гг. наземных значений концентраций продуктов деления дается обсуждение оценок времени пребывания этих продуктов в стратосфере.