Measurement of lead-210 in surface air and precipitation

By L. U. JOSHI, C. RANGARAJAN and Smt. SARADA GOPALAKRISHNAN, *Health Physics Division, Air Monitoring Section, Bhabha Atomic Research Centre, Bombay-74*

(Manuscript received December 28, 1967)

ABSTRACT

Lead-210 activity in ground level air and rainwater at a number of stations in India covering the latitude band 11° to 34° N has been studied and the data is presented. Seasonal variations in activity levels with maximum values in winter have been observed in the surface air at several stations. The time of occurrence of peak values of lead-210 differs from that of stratospheric cesium-137 but generally agrees with radon levels. It is therefore concluded that lead-210 is mainly controlled by radon activity in the ground level air. The possible reasons for the winter peak in radon and lead-210 are discussed.

The latitudinal variation of lead-210 and the likely reasons for these are examined. The lead-210 activity levels in India are compared with the values reported by other laboratories for temperate latitudes.

1. Introduction

In a previous publication (Rangarajan *et al.*, 1967) data on lead-210 concentrations in ground level air and precipitation at a few stations in India were presented. Therein, it was pointed out that lead-210 concentrations undergo a seasonal variation with maximum values in winter and minimum values during the summer. This was noted at Bombay and to a lesser extent at Srinagar where the variation was not so pronounced as at Bombay. It was also suggested that this seasonal variation which differed in the time of occurrence of the peak values from fission products like cesium-137 originating from the stratosphere, seemed to correlate very well with ground level radon values. In this paper extensive measurements on lead-210 made since then at the various stations listed in Table 1 are presented and their relations with other atmospheric activities like radon, fission products, etc., discussed. Activities in rainwater and their relations to air concentrations have also been explored.

2. Experimental methods

The techniques of sample collection and radioactivity measurements have already been described in detail (Rangarajan *et al.*, 1968, Vohra *et al.*, 1966, Joshi *et al.*, 1968). The sampling volume of ground level radioactive dust collection is of the order of 50×10^3 cubic meters at Bombay and about one-tenth of this at the other stations. The Hollingsworth and Vose H-70 filters used for sampling are ashed at low temperatures (less than 400°C) prior to chemical analysis. The ashed samples along with lead carrier are digested in aqua regia and then taken in 1.5 *M* hydrochloric acid. These samples are then passed through anion-exchange resin Dowex-1 × 8% (50–100 mesh). The maximum adsorption of lead takes place at this acid concentration. The adsorbed lead is then eluted with 8 *M* hydrochloric acid. It is finally precipitated as lead sulphate and counted (Joshi *et al.*, 1967). The counting of daughter product bismuth-210 and its build-up with a half-life of 5.0 days enables the purity of the samples to be checked as well as the activity to be assayed. End-window geiger counters with a back-ground of five counts per minute are used for the beta counting of the samples. With this system, samples can be counted to an accuracy better than ±5% s.d.

Rainwater collections are made in stainless steel funnels (12″ diameter) and polythene vessels. The monthly collections of rainwater are pooled together depending on the amount of rainfall to give detectable activity. Yearly

Table 1. *List of radioactivity monitoring stations in India*

Station	Altitude (Meters)	Latitude	Longitude	Type of sampling for lead-210 analysis
1. Srinagar	1598	34°06′ N	74°55′ E	Rainwater and monthly air filter analysis
2. Delhi	219	28°45′ N	77°20′ E	Do.
3. Gangtok	2000	27°12′ N	88°23′ E	Rainwater analysis. Air filter data not presented.
4. Calcutta	Sea level	22°34′ N	88°25′ E	Rainwater and monthly air filter analysis.
5. Nagpur	311	21°12′ N	79°04′ E	Do.
6. Bombay	Sea level	18°57′ N	72°55′ E	Do.
7. Bangalore	922	12°57′ N	77°30′ E	Rainwater and half-yearly air filter analysis.
8. Ootacamund	2235	11°23′ N	76°40′ E	Do.

average specific activities and depositions only are reported here. The evaporated residues of the rain samples are chemically treated as for filter ash and counted.

3. Results

Fig. 1 gives the lead-210 concentrations in air at Bombay and Srinagar from 1962 to the middle of 1966. These are a continuation of the data previously given (Rangarajan *et al.*, 1968) up-dated to the middle of 1966. Radon concentrations at Bombay not reported earlier are also included from the end of 1965 when sampling for radon was started. The radon activity is measured through its solid daughter products assuming equilibrium with radon. In view of the uncertainty in this as well as in the

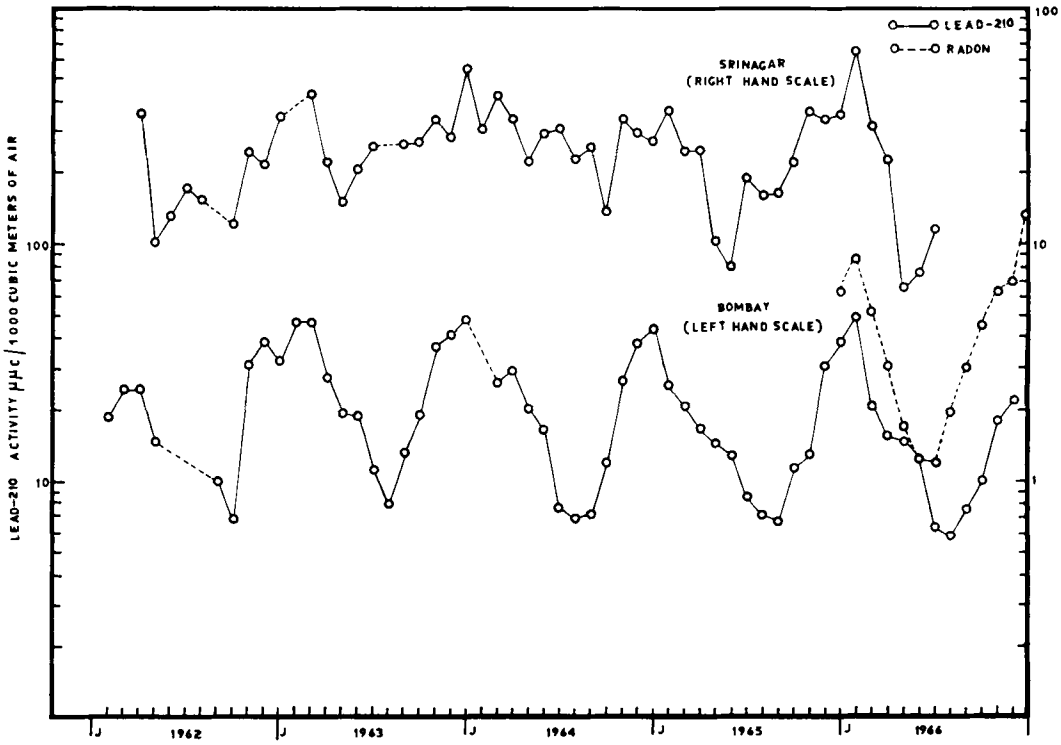

Fig. 1. Lead-210 (Ra-D) activity in ground level air at Bombay and Srinagar.

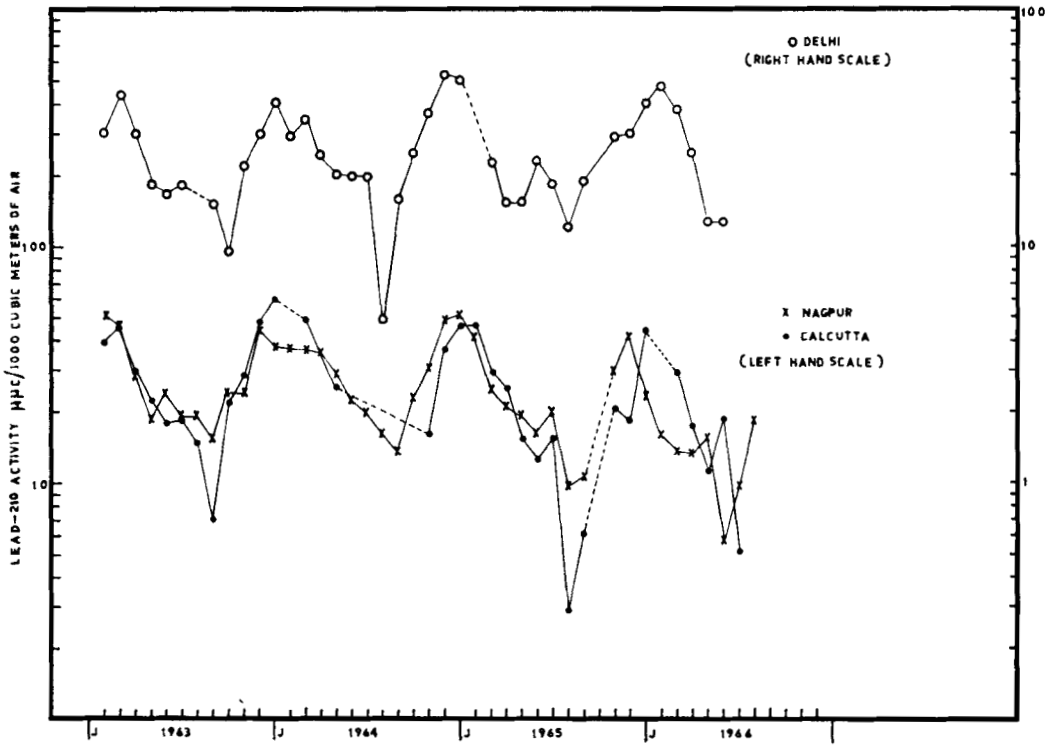

Fig. 2. Lead-210 (Ra-D) activity in ground level air at Calcutta, Nagpur and Delhi.

possibility that filter collection efficiency for the daughter products is not 100%, radon concentrations are given in relative units but if radon is in equilibrium and filter collection efficiency is taken as 100%, one unit would correspond to one micro-microcurie per cubic meter of air. The uncertainties in radon measurements are unlikely to affect the following discussions as will be shown later.

Lead-210 activities measured in surface air at three other stations, viz., Nagpur, Calcutta and Delhi are shown in Fig. 2, from 1963 onwards. These results are also based on analysis of monthly pooled filter collections as at Bombay and Srinagar.

The latitudinal variation of the average yearly concentration of lead-210 in surface air and precipitation are shown in Fig. 3 for the years 1962 to 1966. Data from two other stations, viz., Bangalore and Ootacamund are also included. The activities in the case of these two stations are based on a half-yearly sample analysis. The precipitation activities have been calculated from the total yearly deposition of lead-210 and the yearly rainfall from all the eight stations of the sampling network (Table 1). The total yearly deposition of lead-210 is summarized in Table 2.

4. Discussion of results

Seasonal variations in surface air activity

The data on air concentration of lead-210 at the stations given in Figs. 1 and 2 show that there is a seasonal variation in activity, the maximum concentration being reached in the winter months of November, December and January and the minimum values in July and August. This seasonal cycle is observed at all the five stations although it is not as pronounced at Srinagar as at the more southerly stations. (Results for another station—Gangtok—which also shows similar seasonal variation are not reported here as the values are not yet finalised).

The climatic and other reasons for this seasonal variation can be discussed on the basis of the possible sources of lead-210 in ground level air. Ground level activity could be due to local radon diffusing from the soil or it could originate

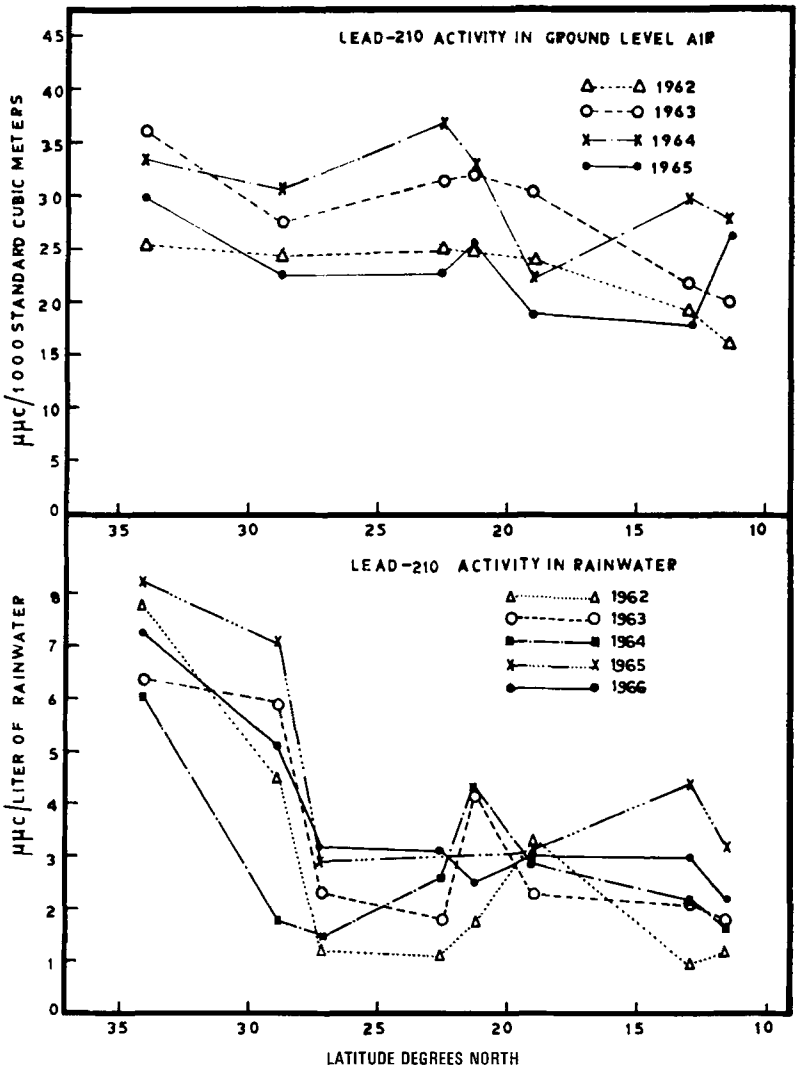

Fig. 3. Latitudinal variation of lead-210 in rain and ground level air in India.

from the upper troposphere and stratosphere where lead-210 has been sampled in significant quantities (Peirson *et al.*, 1966). The behaviour of a stratospheric source diffusing to ground level is well illustrated by the measurements on the cesium-137 activity in surface air (Rangarajan *et al.*, 1968). If lead-210 were originating from the stratosphere its peak values are likely to be observed during the spring months along with maximum values of cesium-137 in April (Rangarajan, *et al.*, 1968) rather than two to three months earlier as is the actual case. On the other hand the agreement with radon is very close as shown in Fig. 1. The possible errors in radon measurements such as nonequilibrium with radon daughters and lesser filter collection efficiencies are unlikely to result in such close agreement between radon and lead-210. The H-70 filter used is reported (Lockhart, *et al.*, 1964) to have an efficiency of nearly 95% for radium (B + C) aerosols and disequilibrium with radon daughters is not significant as what is measured is the radon daughter concentrations which actually decay to lead-210. Hence it may be concluded that lead-210 in surface air is mainly controlled by radon concentrations, at least in continental areas.

Two reasons can be advanced for the observed

Table 2. *Annual deposition of lead-210 in India (in mc/km²)*
(The values in brackets give annual precipitation in millimeters)

Station	Year				
	1962	1963	1964	1965	1966
Srinagar	4.87 (625.2)	4.98 (782.0)	2.93 (692.0)	5.36 (653.0)	6.48 (889.9)
Delhi	2.60 (577.2)	4.85 (817.5)	2.16 (1233.1)*	2.19 (420.5)*	3.31 (650.2)
Gangtok	3.77 (3170.4)	8.96 (3827.4)*	3.40 (2401.2)*	4.17 (2466.0)*	10.76 (3399.3)*
Calcutta	1.32 (1158.0)	3.03 (1678.0)	3.85 (1485.0)	—	3.25 (1025.7)*
Nagpur	2.25 (1258.8)	3.86 (932.6)	2.66 (849.9)*	—	2.17 (858.9)
Bombay	7.50 (2227.0)	5.12 (2229.0)	5.24 (1798.8)*	8.66 (2626.1)	5.79 (1952.0)
Bangalore	1.03 (1052.7)	2.09 (1040.6)	2.60 (1191.6)	3.06 (692.0)	3.04 (1021.0)*
Ootacamund	1.77 (1509.6)	2.12 (1190.0)	3.19 (1835.0)	2.14 (676.6)*	3.12 (1416.6)*

* The rainfall for these years is the total for the sampling months only but is greater than 80 % of the total annual precipitation.

annual variation in radon (and lead-210) concentrations in surface air.

A shift in surface winds from continental to maritime such as occurs when the north-east winds change into the powerful south-west monsoon over the Indian continent could result in a seasonal variation. The monsoon winds, due to lack of replenishment of radon diffusing from soil in their path across the oceans may be less radioactive than winds which have travelled over large land masses. However, the onset of the monsoon occurs some time during the period April–May when lead-210 levels have already decreased to significantly lower values compared to peak concentrations. This makes it rather difficult to correlate the fall in activity after the peak values with the onset of the south-west monsoon winds.

The occurence of frequent inversion conditions during winter could also result in high radon concentrations. Gale *et al.* (1958), found that average monthly radon concentrations showed significant correlation with the average potential temperature gradient during the period of their measurements for one year in the United Kingdom. The presence of the winter maximum at all the stations—coastal as well as inland—suggests that this could be a major factor in the seasonal variation although the effect of air mass movements cannot be ruled out. (Part of the reason for the seasonal variation not being so pronounced at Srinagar could be the lesser prevalence of the monsoon there.)

Latitudinal variation of activity

The data given in Fig. 3 shows a trend towards an increase in the specific activity of rains at high latitude stations like Srinagar although this is not so clear in the air activity data. The climate of this station (Srinagar) differs in several respects from the other lower latitude stations where precipitation is mostly in summer during monsoon when air activity—both natural and bomb produced—are at a minimum. At Srinagar, however, there is both winter and summer rains which are mostly not connected with the monsoon cycle. This difference in the precipitation system is apparently reflected in the somewhat higher specific activity at Srinagar. A similar difference in specific activity in rain was observed in the case of cesium-137 which was also attributed to the difference in the precipitation systems (Rangarajan *et al.*, 1968) between the northern stations particularly Srinagar and the other low latitude stations. In view of these results it is concluded that the specific activity of precipitation is sensitive to the origin and seasonal distribution

of rainfall with respect to the peak values in airborne activity.

The general level of activities given here for India may be compared with activities in the temperate latitudes as reported by other laboratories. Airborne lead-210 activities in India vary from about 10–50 micro-microcuries per thousand cubic meters as shown in Figs. 1 to 3, while in precipitation the activities are of the order of 1–10 micro-microcuries per litre. In general the values given here are considerably higher than the activities in the U.K. (Peirson *et al.*, 1966) of 1–10 micro-microcuries per kg of air but show agreement with the values given by Patterson (Patterson *et al.*, 1964) for the U.S. of 3–30 micro-microcuries per thousand cubic meters and by Shleien (Shleien *et al.*, 1967) for Massachusetts, U.S. of 4–40 micro-microcuries per thousand cubic meters. These differences can possibly be explained by the prevalence of maritime winds over islands like the U.K. and the presence of large land masses and high radioactive areas in the other case. The deciding factor seems to be the previous path of the air masses being sampled which determines their radioactive content.

5. Acknowledgements

Our thanks are due to Dr. K. G. Vohra, Head, Air Monitoring Section under whose guidance this work was carried out. We are grateful to Dr. A. K. Ganguly, Head, Health Physics Division for his encouragement. We also thank the following institutions for their help in the collection of samples: (1) Regional Meteorological Centres of the India Meteorological Department at Bangalore, Calcutta, New Delhi, Gangtok, Nagpur and Srinagar and (2) Cosmic Ray Research Laboratory of the Tata Institute of Fundamental Research at Ootacamund.

REFERENCES

Gale, H. J. & Peaple, L. H. J. 1958. Measurements on the near-ground radon concentrations on the A.E.R.E. Airfield, AERE HP/R 2381.

Joshi, L. U. & Mahadevan, T. N. 1967. Radiochemical determination of lead-210 concentrations in ground level air in India. *Proceedings of the Nuclear and Radiation Chemistry Symposium*, Poona.

Joshi, L. U. & Mahadevan, T. N. 1968. Seasonal variation of radium-D (Lead-210) in ground level air in India. *Health Physics, 15*, 67–71.

Lockhart, L. B. & Patterson, R. L. 1964. Characteristics of air filter media used for monitoring airborne radioactivity, NRL Report 6054.

Patterson, R. L. & Lockhart, L. B. 1964. *Geographical distribution of lead-210 (Ra-D) in the ground-level air.* The natural radiation environment. The University of Chicago press, Chicago, 383–392.

Peirson, D. H., Cambray, R. S. & Spicer, G. S. 1966. Lead-210 and polonium-210 in the atmosphere, *Tellus 18*, 427–433.

Rangarajan, C., Sarada, G., Sadasivan, S. & Chitale, P. V. 1968. Atmospheric and precipitation radiovity in India, *Tellus 20*, 269–283.

Shleien, B., Cochran, J. A., Benander, L., Bernard, L. L., Lutz., J. D. & Friend, A. G. 1967. Atmospheric radioactivity analysis and instrumentation, status report III, National Centre for Radiological Health, Public Health Service, U.S. Department of Health, Education and Welfare.

Vohra, K. G., Rangarajan, C., Sarada, G., Sadasivan, S. & Chitale, P. V. 1966. Measurements on airborne and surface fallout radioactivity in India from nuclear weapon tests, Atomic Energy Establishment Trombay (India) Report AEET-247.

ИЗМЕРЕНИЯ ИНТЕНСИВНОСТИ ВЫПАДЕНИЯ ОСАДКОВ И АКТИВНОСТИ Pb-210 В ПРИЗЕМНОМ СЛОЕ ВОЗДУХА

В данной работе проведено исследование и представлены данные об активности Pb-210 в приземном слое атмосферы и интенсивности выпадения осадков в ряде районов Индии, находящихся в пределах полосы широт от 11° до 34° с. ш. Сезонные изменения уровней активности, максимум которой приходится на зимний период, наблюдались в нескольких районах. Время распространения максимума активности Pb-210 отличается от времени распространения, соответствующего стратосферному Cs-137, но в целом согласуется с уровнями радона. Поэтому делается заключение, что активность Pb-210,(главным образом, контролируется активностью радона в приземном слое воздуха. Обсуждаются возможные причины максимальной активности радона в зимний период. Рассматривается широтное изменение активности Pb-210 и его возможные причины. Уровни активности Pb-210 в Индии сравниваются с величинами, представленными другими лабораториями в умеренных широтах.