# Be' and P<sup>32</sup> in ground level air

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#### ABSTRACT

Weekly samples of dust collected from ground level air at Bern  $(47^{\circ} N)$  have been analysed for their contents of cosmic ray produced Be<sup>7</sup> and P<sup>32</sup> during January– September 1965. The observed ratios of Be<sup>7</sup>/P<sup>32</sup> place a limit on the spring influx of stratospheric air. It is discussed that the pronounced spring maximum in Be<sup>7</sup> concentration in ground level air, observed by some investigators at temperate and high latitudes is, to a large part, a manifestation of other meteorological factors, e.g. better vertical mixing within the troposphere, rather than due to a large influx of air from the stratosphere.

## 1. Introduction

A sharp seasonal increase in concentrations of several fission products and cosmic ray produced radionuclides, e.g. Cs<sup>137</sup>, Na<sup>22</sup>, Be<sup>7</sup>, has been observed to occur between February and July in ground level air at temperate and high latitudes. This observation has been interpreted by some authors (e.g. PARKER, 1962) in terms of an influx of considerable amount of air from the stratosphere where these nuclides are present in much higher concentrations.

The possibility of using cosmic ray produced isotopes for studying stratospheric-tropospheric exchange of air arises from the fact that there is a large difference in the concentration as well as in the ratio of their activities in the stratospheric and tropospheric air. For example, the stratospheric concentrations of Be<sup>7</sup> are about 25-50 times higher than those in the upper troposphere, and about 500 times higher compared to those in the ground level air. Any seasonal influx of comparatively small amounts of air from the stratosphere can, therefore, appreciably change the tropospheric concentrations. Further, as the ratio of activities of Be<sup>7</sup> and  $P^{32}$  in the stratospheric air is about 2-3 times that in the tropospheric air, the stratospheric influx should also cause an increase in Be<sup>7</sup>/P<sup>32</sup> ratios in the troposphere.

The measurements of Be<sup>7</sup> in ground level air at high latitudes (40-55° N) have been reported by CRUIKSHANK, COWPER & GRUMMIT (1956), PEIRSON (1963), GUSTAFSON, KERRIGAN & BRAR (1961), PARKER (1962) and SCHUMANN & STOEPPLER (1963). All these measurements show a sharp increase in Be<sup>7</sup> at ground level sometime during February to July, although the observed magnitude of increase has been found to be different at different stations. For example, GUSTAFSON, KERRIGAN & BRAR (1961) and SCHUMANN & STOEPPLER (1963) have observed an increase by a factor of 2 to 3, while PARKER (1962) reported an increase up to a factor of about 8. If all this increase is due to an influx of stratospheric activity, the Be<sup>7</sup>/P<sup>32</sup> ratios in the troposphere should also increase appreciably during such periods. It is estimated that if enough stratospheric air descends into the troposphere, so as to increase the Be<sup>7</sup> concentration there by a factor of three or more, the activity ratio of Be<sup>7</sup>/P<sup>32</sup> in it should also show an increase of about 100 %. To check on this point we undertook to measure Be<sup>7</sup>/P<sup>32</sup> in ground level air at Bern which lies at latitude 47° N where the deposition of stratospheric fission debris has been observed to be maximum (МАСНТА, 1961).

Tellus XVIII (1966), 2



FIG. 1 (a). Observed  $Be^{7}/P^{32}$  activity ratios in ground level air at Bern. The open circles represent tropospheric measurements at latitude 35° N and altitudes 3-8 km.

FIG. 1 (b). The data of Fig. 1 (a) are replotted after averaging the  $Be^7/P^{32}$  ratios for each calendar month. The solid curve shows the predicted variation in this ratio at high latitudes assuming an average stratospheric residence time of 10 months and the stratospheric-tropospheric exchange to be peaked in late spring.

## 2. Experimental

The measurements of Be<sup>7</sup>/P<sup>32</sup> in ground level air at Bern have been carried out during January-September 1965. Weekly samples of dust were collected on coarse filters on the roof of Physikalisches Institut and analysed for Be<sup>7</sup> and P<sup>32</sup>. Standard radiochemical and counting procedures (cf. BHANDARI, 1965) were employed for estimating the radioactivities of Be<sup>7</sup> and P<sup>32</sup>. The errors in the present measurements for the activity ratio Be<sup>7</sup>/P<sup>32</sup> are estimated to be  $\pm 20$  %; most of this arising from errors in the measurement of P<sup>32</sup>.

Tellus XVIII (1966), 2 14-662895

## 3. Results and discussions

The measured Be<sup>?</sup>/P<sup>32</sup> ratios are plotted in fig. 1 (a). The dark circles are the ground level measurements and the open circles refer to tropospheric air samples at latitude 35° N for altitudes 3-8 km (RAMA & HONDA, 1961; BHANDARI, 1965). No marked variation in the ratio with time stands out. The experimental error of  $\pm 20$  per cent on individual measurements makes it difficult to see small trends. We have therefore plotted the average Be<sup>7</sup>/P<sup>32</sup> ratios by taking the mean of all measurements in each calendar month (fig. 1 (b)). Here, one



FIG. 2. Predicted variations in (a) total inventories and (b) relative inventories of Be<sup>7</sup>, P<sup>32</sup> and Na<sup>22</sup> in the troposphere. The model calculations (see text) assume that the stratosphere-troposphere exchange is peaked during late spring.

sees a trend for the ratio to be high in mid-year but the range of variations is not a factor of two. Therefore, the possibility of ground level air containing Be7 and P32 activities of predominantly stratospheric origin at any time seems to be ruled out. It is, however, well known that some stratospheric-tropospheric exchange, with marked seasonal variation, does take place in the temperate and high latitude region. A recent analysis of dispersion of radiocarbon produced in the Russian nuclear tests has provided valuable information on this aspect (LAL & RAMA, 1965). An analysis of these data suggests that for considering the effective circulation in the troposphere, each hemisphere can be divided into two distinct but internally well mixed cells, extending from 0° to 30° (hereafter termed as the tropical cell) and from  $30^{\circ}$  to 90° (hereafter termed as the polar cell). The

exchange between these cells has a pronounced seasonal dependence. The stratospheric-tropospheric exchange, which also has strong seasonal dependence, occurs primarily across the tropopause of the polar cell. LAL & RAMA (1965) have evaluated the exchange parameters and find that they can be represented by the relations:

$$J = 25 + 15 \sin \left(2\pi \frac{t}{\tau} - \frac{\pi}{3}\right),$$
$$K = 1 + 0.7 \sin \left(2\pi \frac{t}{\tau} + \frac{\pi}{2}\right),$$

where  $\tau$  is 12, and t is expressed in months, J denotes the amount of air (in grams) exchanged across 1 cm<sup>2</sup> area of the tropopause of polar cell, and K denotes the fraction of air in the polar cell exchanged per month with that in tropical cell. Using these exchange parameters and probable values for the washout rate of activities from the troposphere, BHANDARI, LAL & RAMA (1965) have estimated the expected variation in concentrations and activity ratios of cosmic ray produced isotopes Na<sup>22</sup>, Be<sup>7</sup> and P<sup>32</sup> in the troposphere. These are shown in fig. 2 (a) and (b). One finds that  $Be^7$ and P<sup>32</sup> concentrations in the polar cell increase by 60 % and 25 % respectively in July-August compared to winter months; Be7/P32 ratios increase by only 35 %. The curve representing the expected variation in  $Be^{7}/P^{32}$  is reproduced in fig. 1 (b) in order to compare it with the experimental data. The two are seen to be consistent although the observed Be<sup>7</sup>/P<sup>32</sup> ratios are about 20 % systematically lower than the expected ratios. Both the model calculations and our data on  $Be^7/P^{32}$  ratio indicate that the Be<sup>7</sup> and P<sup>32</sup> activities in the troposphere should be largely of tropospheric origin. The observed large spring increase in Be<sup>7</sup> in ground level air may then be interpreted as due to enhancement by other meteorological conditions such as seasonal variations in dry and wet deposition and particularly the variation in vertical mixing with the upper layers of air (within the troposphere) where the concentrations are known to be an order of magnitude higher (GUSTAFSON, KERRIGAN & BRAR, 1961; BHANDARI, 1965).

A further support to the hypothesis of increased vertical mixing during the summer months comes from the measurements of Pb<sup>210</sup> concentrations in ground level air at Chilton (51° N), reported by PIERSON *et al.* (1965). Their observations show that the concentrations of Pb<sup>210</sup> in winter are a factor of two higher than those in summer. Since the source of Pb<sup>210</sup> is radon which has the highest concentration near the ground, the concentration of Pb<sup>210</sup> also is usually high near the ground (LIST & TELEGADAS, 1961). But due to quick vertical mixing during summer the normally high concentrations are expected to disappear.

WALTON et al. (1962) have reported a few measurements of  $Be^{7}/P^{32}$  in wet precipitation at temperate latitudes. They observe a mean ratio of 52 in summer as against 33 in winter which is again consistent with the predicted variation.

It is clear from fig. 2 that Na<sup>22</sup> should show a large variation, i.e. an increase by a factor of  $\sim 2.8$  during the period of peak influx and

BHANDARI, N., 1965, Ph. D. Thesis, University of Bombay.

- BHANDARI, N., LAL, D., and RAMA, To be published.
- CRUIKSHANK, A. J., COWPER, G., and GRUMMIT, W. E., 1956, Production of Be<sup>7</sup> in the atmosphere. *Can. J. Chem.*, 84, pp. 214-219.
- GUSTAFSON, P. F., KERRIGAN, M. A., and BRAR, S. S., 1961, Comparison of Beryllium-7 and Cesium-137 radioactivity in ground level air. *Nature*, 191, pp. 454-456.
- LAL, D., and RAMA, 1965, Characteristics of global tropospheric mixing based on man-made C<sup>14</sup>, H<sup>3</sup> and Sr<sup>90</sup> (in press).
- and Sr<sup>90</sup> (in press). LIST, R. J., and TELEGADAS, K., 1961, HASL Report 111, p. 208.
- MACHTA, L., 1961, A survey of information on meteorological aspects of world wide fallout. World Meteorological Organisation Report, 11, T-P-49, pp. 3-30.
- PARKER, R. P., 1962, Beryllium-7 and fission products in surface air. Nature, 193, pp. 967–968.

hence Na<sup>22</sup> and P<sup>32</sup> provide a more sensitive pair of isotopes for studying the stratospheric influx during spring. However accurate determination of P<sup>32</sup> is difficult and some short-term fluctuations in its concentrations may be expected. Therefore, from the point of view of accuracy in measurement and the interpretation of data, Na<sup>32</sup> and Be<sup>7</sup> provide a more convenient pair; Na<sup>32</sup>/Be<sup>7</sup> ratio during peak influx is expected to increase by 70 per cent. To this end, a study when the bomb produced Na<sup>32</sup> gets exhausted from the atmosphere may prove very useful.

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## REFERENCES

- PEIRSON, D. A., 1963, Beryllium-7 in air and rain. J. Geophys. Res., 68, pp. 3831-3832.
- PEIRSON, D. A., CAMBRAY, R. S., and SPICER, G. S., 1965, Pb<sup>210</sup> and Pc<sup>310</sup> in the atmosphere. Proceedings of the Visby Conference; this Journal.
- RAMA and HONDA, M., 1961, Natural radioactivity in the atmosphere. J. Geophys. Res., 66, pp. 3227-3231.
- RÖDEL, W., 1965, Cosmic ray produced and artificial Na<sup>22</sup> in the atmosphere. Preprint, J. Geophys. Res.
- SCHUMANN, G., and STOEPPLER, M., 1963, Beryllium-7 in the atmosphere. J. Geophys. Res., 68, pp. 3827-3830.
- WALTON, A., and FRIED, R. E., 1962, The deposition of Be<sup>7</sup> and P<sup>82</sup> in precipitation at north temperate lattitudes. J. Geophys. Res., 67, pp. 5335-5340.

### Ве<sup>7</sup> И Р<sup>32</sup> В ПРИЗЕМНОМ ВОЗДУХЕ

В течение января-сентября 1965 г. определялось содержание Ве<sup>7</sup> и Р<sup>32</sup>, образовавшиеся под действием космических лучей, в еженедельных пробах пыли, собранной в приземном воздухе в Берне (47°С). Наблюдавшиеся отношения Ве<sup>7</sup>/Р<sup>32</sup> определило границу весеннего притока воздуха из стратосферы. Обсуждается предположение, что наблюдав-

шиеся некоторым исследователями в умеренных и высоких широтах, ярко выраженный весенний максимум концентрации Be<sup>7</sup> в приземном воздухе вызван в основном другими метереологическими факторами (например лучшим перемешиванием внутри тропосферы), а не притоком воздуха из стратосферы.

Tellus XVIII (1966), 2