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Cosmic ray produced Radioisotopes for studying the General Circulation in the atmosphere

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ABSTRACT. The possibility of using cosmic ray produced radioisotopes for studying large scale circulation in the atmosphere has been explored recently. The potential significance of these isotopes in the field of meteorology arises because of the following features in their production and properties—(1) Several radioisotopes are available whose half-lives are comparable to time scales involved in the atmospheric circulation. Four of the isotopes (S^{35}, Be^7, P^{33}, and P^{32}) have half-life periods ranging from three months to a fortnight, and one (Na^{22}) has a half-life of about two and a half years, and (2) Isotope production is continuous, constant, strongly dependent on the latitude and altitude in the atmosphere and independent of meteorological factors.

These isotopes can be used for investigating the nature of circulation of air in the troposphere and the detailed mechanism of exchange of air between the stratosphere and the troposphere.

Experimental work on the use of these isotopes in meteorology has so far been confined mainly to determination of the concentration in rain water by the Geophysics Research Group of the Tata Institute of Fundamental Research, Bombay. Some of the important results of these investigations are summarised in this paper.

1. Introduction

The passage of cosmic rays through the atmosphere leads to nuclear transmutations of the constituent air nuclei in a variety of modes. Several radio-nuclides produced in these transmutations have been detected including the well discussed radioisotopes C^{14} and H^3, and a few others whose half-lives are of the order of weeks or months. Removal of these isotopes from the atmosphere occurs chiefly through condensations in the troposphere. Their concentration in air between two successive condensations depends sensitively on their half-lives and also on the trajectory of motion of the air since the cosmic ray intensity in the atmosphere shows a strong latitude and altitude dependence. Because of this, the possibility exists of using these cosmic ray produced isotopes (which have half-lives comparable to the time scales involved in the circulation of air in the troposphere) for studying meteorological processes. The purpose of this article is to review briefly the work reported so far in this field.

2. Radioisotopes useful for meteorological research

The various cosmic ray produced radioisotopes, whose half-lives are greater than a few days and smaller than a few years are listed in Table 1.

The radioisotope Be^7 is produced chiefly in nuclear interactions involving atmospheric nitrogen and oxygen nuclei, while the others arise in interactions involving atmospheric argon nuclei.

3. Rate of production of various isotopes

Cosmic ray production rates of the radioisotopes S^{35}, Be^7, P^{33} and P^{32} have been calculated for all parts of the atmosphere by Lal, Malhotra and Peters (1958). Detailed calculations of the production of Be^7 have also been reported by Benioff (1956). No calculations, however, exist for the production of Na^{22}.

The rate of production of various radioisotopes can be obtained from Fig. 1, which shows the rate of nuclear disintegrations occurring in the atmosphere as a function of latitude and altitude. To obtain isotope

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TABLE 1

Radio-isotopes	Half-life	References
Na^{22}	2·6 years	Marquez *et al.* (1957)
S^{35}	87 days	Goel (1956)
Be^{7}	53 days	Arnold and Al-Salih (1955), Goel *et al.* (1956), Cruikshank *et al.* (1956)
P^{33}	25 days	Lal *et al.* (1957)
P^{32}	14·3 days	Marquez and Costa (1955), Lal *et al.* (1957)

production rates, the ordinate of Fig. 1. should be multiplied by the corresponding yield factors, σ_i , *viz.*,

$$\sigma_{S^{35}} = 4\cdot2\times10^{-4}\,; \qquad \sigma_{Be^{7}} = 4\cdot2\times10^{-2}$$

$$\sigma_{P^{33}} = 2\cdot8\times10^{-4}\,; \qquad \sigma_{P^{32}} = 2\cdot5\times10^{-4}$$

The position of the tropopause, the average nuclear disintegration rates in the stratosphere $\bar{S}_S$ and in the troposphere $\bar{S}_T$, are also shown in Fig. 1. The total number of disintegrations produced per cm^2 every second in the entire atmosphere as well as the fraction which is produced below the tropopause is shown in Fig. 2.

The variation in the production rate of the various isotopes with position in the atmosphere (Figs. 1 and 2) has the following characteristics—

(*i*) Absolute isotope production (nuclei per second per gram of air) varies strongly with altitude at a given latitude; it also varies appreciably with latitude at a given altitude.

(*ii*) Integrated isotope production in a unit cross-section column in the *troposphere* is nearly independent of the geographic location (See Fig. 2). The cosmic ray production at a given height increases as one moves toward higher latitudes, but the height of the tropopause changes in the opposite direction: the resulting total production in the troposphere remains constant, within ± 20 per cent.

(*iii*) Integrated isotope production in a unit cross-section column in the *stratosphere* is latitude dependent; it increases by a factor of ~12 as one moves from the equator toward the pole.

(*iv*) Sudden increases, and slow changes in the cosmic ray intensity which are associated with solar flares and the sunspot activity respectively, do not produce any appreciable effect on the isotope production since these changes are mainly brought about in the low energy end of the spectrum. Low energy primary protons are not very effective in isotope production since they are brought to rest mainly by ionization losses. The total changes in the global isotope production are estimated to be ± 3 per cent between the maximum and minimum of a solar sunspot cycle. The increases associated with solar flares vary largely from flare to flare, but their effect lasts only for a few hours, and can, therefore, be neglected. With good approximation, one can assume that *isotope production occurs at a constant rate.*

As we shall see later, these features in the cosmic ray production of isotopes are of great value for their application to meteorological purposes.

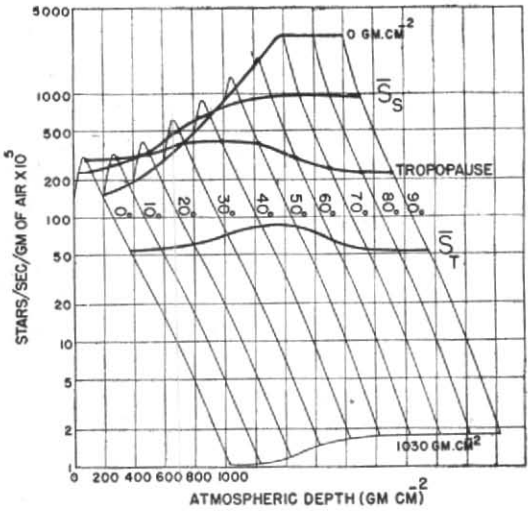

Fig. 1

The rate of nuclear disintegrations (/gm of air/sec) is plotted against atmopsheric depth. The curves for different latitudes (geomagnetic) have been displaced with respect to each other for the sake of clarity. Lines marked $\bar{S}S$ and $\bar{S}T$ represent smooth curves passing through average nuclear disintegration rates, in the stratosphere and the troposphere respectively, at different latitudes.

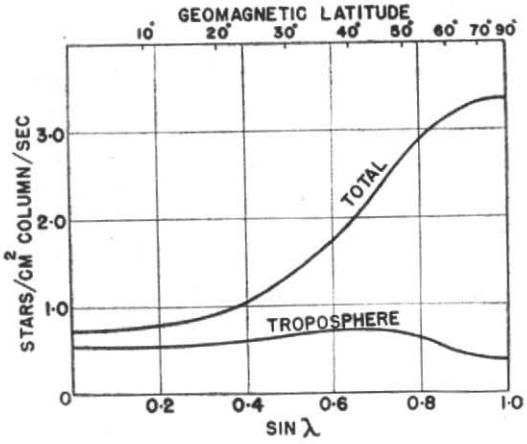

Fig. 2

The total number of nuclear disintegrations per second in an atmospheric column of cross-section 1 cm², extending up to the tropopause (lower curve) as well as up to the top of the atmosphere, are given as a function of geomegnetic latitude.

4. Concentration of isotopes in air masses calculated for a few hypothetical trajectories

The expected isotope concentrations in air for any assumed trajectory of the air mass can be calculated using the absolute isotope production rates given by Fig. 1. As an illustration, we shall discuss two simplified cases from which one may get an estimate of the magnitude of variations in isotope concentrations which one may expect under different meteorological conditions.

(*i*) An air mass remains confined within the troposphere between two successive condensations;

(*ii*) An air mass descends from the stratosphere to the troposphere and then remains within the troposphere until its activity is removed by condensation.

The expected isotope concentrations in air have been calculated for these special cases under certain simplifying but reasonably plausible assumptions (Lal *et al.* 1958). The assumptions are :

(*a*) that isotopes collect on dust or water droplets and are effectively removed from air by condensation and precipitation. After a rain or snow the air mass is supposed to be free of the isotopes in question; and

(*b*) that the troposphere is sufficiently turbulent so that the time spent by air at different heights is proportional to the pressure at these heights. It is further assumed that air masses remain in the stratosphere for time periods which are long compared to the half-lives of the isotopes under consideration. This assumption is certainly justified if the average period of residence of air in the stratosphere is of the order of several years (Libby 1956). We do not assume mixing between different layers of air within the stratosphere. The isotope production rate in an air mass moving in the stratosphere is, therefore, a function of the altitude, x, and also latitude, λ (according to Fig. 1).

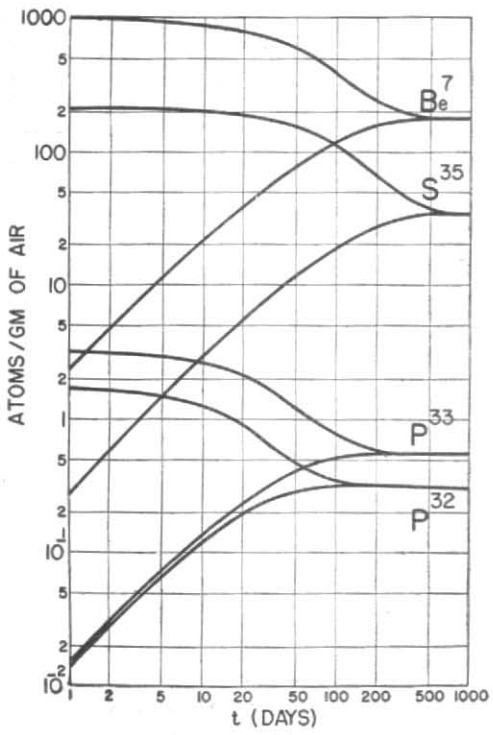

Fig. 3

The concentration of various radioisotopes in air (atoms/gm) is shown as a function of time for two cases. The lower curves refer to air which was free of radio-activity at $t = 0$ and always remained in the turbulent troposphere. The upper curves refer to air which remained just above the tropopause for a time long compared to the half-life of the various isotopes, and descended into the troposphere at time $t = 0$.

For trajectory (a), *i.e.*, air circulating entirely in the troposphere, the concentrations of various isotopes per gram of air $C_i(t)$ at any time, t, is then given by :

$$C_i(t) = \bar{S}_T \, \sigma_i \, \tau_i \, (1 - e^{-t/\tau_i}) \qquad (1)$$

if we assume $C_i = 0$ for $t = 0$. Here $\bar{S}_T$ is the average production rate of nuclear disintegrations in the troposphere and σ_i is the number of nuclei of the particular isotope, i, per nuclear disintegration, and τ_i, its mean life time.

For trajectory (b), when air descends from the stratosphere to the troposphere at time $t = 0$, the concentration of various isotopes in air at any time t is given by :

$$C_i(t) = \sigma_i \, \tau_i \left[\bar{S}_T + \left(S_S(x, \lambda) - \bar{S}_T \right) e^{-t/\tau_i} \right] \qquad (2)$$

$S_S(x, \lambda)$ represents the production rate appropriate to the altitude and latitude at which the air was exposed in the stratosphere.

The *half-lives* of the various radioisotopes under consideration are of the *order of time periods* encountered *in the troposphere* between two successive cleansings of an air mass by condensations; therefore, the rate of accumulation of isotopes in air, as given by eqns. (1) or (2) depends sensitively on time periods, t, of interest and also on the isotope half-lives. Furthermore, since the mean residence period of air in the stratosphere is long compared to the half-life of the various isotopes, the isotope concentrations in the stratospheric air are always higher compared to those in the tropospheric air. These facts can be seen from Fig. 3 which shows the calculated values of $C_i(t)$ for trajectories (a) and (b), for the four radioisotopes S^{35}, Be^7, P^{33}, and P^{32}. The value of $S_S(x, \lambda)$ used for solving equation (2) is $6\bar{S}_T$, which roughly corresponds to air irradiated in the lower regions of the stratosphere. The corresponding *ratios of the isotope concentrations* are shown in Fig. 4.

It becomes apparent that air masses which had moved in the stratosphere for periods long compared to the half-life of the radioisotopes differ appreciably in their isotope contents from those which had remained in the troposphere since the last precipitation. They remain distinguishable even after spending a few months in the troposphere. This holds even for air descending from the lower regions of the stratosphere.

The isotope concentration ratios ($Be^7 : P^{32}$, $S^{35} : P^{33}$) also differ markedly because of their widely different half-lives.

5. Experimental results

The experimental data so far are largely confined to the study of concentrations of various isotopes in rain water. No reliable measurements exist for concentrations of isotopes in air.

The concentrations of the radioisotopes Be^7, S^{35}, P^{33}, and P^{32} have been determined in wet precipitations occurring at several stations* in India during 1956—58 by the Geophysics Research Group at Bombay. In some cases samples have been analysed simultaneously for two or more isotopes.

Apart from these data, there exist some measurements on the concentrations of the radioisotope Be^7 in rain and snow at Chicago and Lafayette during 1955 (Arnold and Al-Salih, 1955).

All measurements have been made after the thermonuclear test explosions began, and one of the problems today is to ascertain whether any appreciable fraction of the various activities is due to these bomb-tests. This question has been discussed for the case of Be^7 by Rama Thor and Zutshi (1958), Peters (1958), and Lal *et al.* (1959), and for the case of P^{32} and S^{35} by Lal *et al.* (1959). Without going into details, we will summarise their conclusions :

(1) Be^7 and P^{32} activities observed in rain-water are consistent with their being of a predominantly cosmic ray origin.

(2) In the case of S^{35}, it has, however, been found that an appreciable part of its observed activity must have been contributed by bombs. Bomb contributions were found to be high especially in rains occurring at Bombay after the first week of July 1958.

(3) The available data are not sufficient to draw any conclusions about P^{33} on this question.

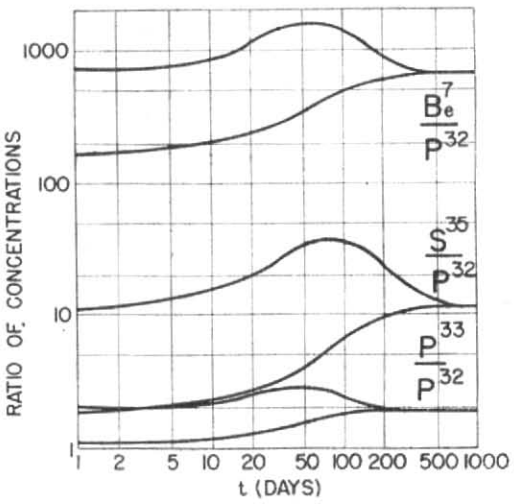

Fig. 4

The ratio in the concentration of different isotopes is given as a function of time in the troposphere for the two cases discussed for Fig. 3.

6. Annual global deposition of Be^7

Measurement of Be^7 concentrations in rain are sufficient to obtain its rate of annual deposition (atmos/cm² year) in the zonal belts 10°, 19°, (25½—34°) and 45° latitudes (Peters 1958). The measured fall-out rates were found to be nearly independent of latitude and lead to an average global fall-out of $4{\cdot}5 \times 10^5$ atmos/cm² year. It has been shown by Peters (1958) that these observations are consistent with the well-known fact that the exchange of air between the stratosphere and the troposphere is a slow process, such that most of the activity produced in the stratosphere decays before reaching the troposphere. The observed latitude independence is then in conformity with expectations if one is dealing mainly with that fraction of activity which is produced below the tropopause (Fig. 2). The measured global deposition of $4{\cdot}5 \times 10^5$ Be^7 atoms/cm² year has then to be compared

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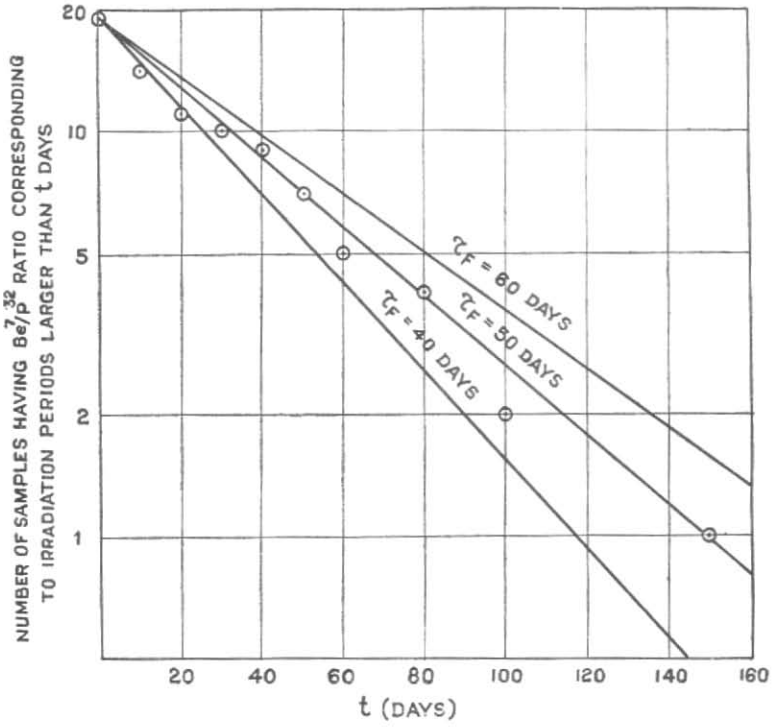

Fig. 5

The observed fraction of samples, having Be^7/P^{32} ratios greater than that corresponding to an irradiation period, t, days in the troposphere, is plotted. The solid lines show the expected behaviour for three values, 30, 40 and 50 days for τ_F, the average removal period of activity from the troposphere.

with the estimated global average production of $7{\cdot}5 \times 10^5$ atmos/cm^2 year in the troposphere. The agreement between the two figures becomes close if one takes into account that some of the Be^7 must decay between its production and deposition.

7. Average removal period of activity from the troposphere

Some tentative conclusions can be drawn regarding the average removal period of activity, τ_F, from the troposphere, from the available data on the measurements of Be^7 and P^{32} activities in rain water.

It has been shown by Goel *et al.* (1959) that whereas the absolute concentrations of the radioisotope Be^7 varied by factors of upto 40 in individual rains, the ratio Be^7/P^{32} remained confined within a much smaller factor of $\sim$4. The expected range in the extreme values of this ratio for tropospheric air under the simplifying assumption of a good mixing in the troposphere [trajectory (*a*) in Sec. 4] is

$$\frac{\tau_{Be^7}}{\tau_{P^{32}}} = 3{\cdot}7$$

This suggests that the observed variations in the ratio arise mainly from a difference in the trajectories of air masses from which the observed activities were removed. The lowest observed ratio, Be^7/P^{32} then corresponds to short irradiation periods, and should be equal to or larger than the ratio of their production rates, $\sigma_{Be^7}/\sigma_{P^{32}}$, in the atmosphere. This should be true provided that both isotopes are removed from the air by condensations with equal

TABLE 2

Period of observation	Observations			Calculated value of $\sigma_{Be^7}/\sigma_{P^{32}}$
	F_{Be^7} (Atmos/cm² year)	$F_{P^{32}}$ (Atmos/cm² year)	$\frac{C_{Be^7}}{C_{P^{32}}} = \frac{\sigma_{Be^7}}{\sigma_{P^{32}}}$	
1958	$4{\cdot}4 \times 10^5$	$4{\cdot}3 \times 10^3$	60 ± 10	54
1956–1958	$4{\cdot}2 \times 10^5$	$3{\cdot}5 \times 10^3$	60 ± 10*	63

efficiency. In that case, one can obtain the time of irradiation of air in individual cases.

The calculated distribution in the irradiation periods for 11 samples measured at Bombay during monsoon of 1957 was observed to follow an exponential corresponding to a mean removal period of 35 days (Goel *et al.* 1959). The measurements during 1958 (Lal *et al.* 1959) based on 19 rain samples collected at Bombay yield a somewhat higher value of 45 days for τ_F (see Fig. 5). The two values are consistent within the errors of measurements, and we obtain $\tau_F = 40 \pm 5$ days.

One can also deduce the value of τ_F by using the measured fall-out of the radioisotopes, Be^7 and P^{32}.

It can be easily shown that if we are dealing entirely with radioisotopes produced in the troposphere, the period, τ_F, is related to the fall-out, F, and half-life τ of an isotope by $\sigma S = F\,(1 + \tau_F/\tau)$, where S is the annual rate of cosmic ray induced nuclear disintegrations in 1 cm² column in the troposphere. For two isotopes, i and j, we can, therefore, write :

$$\frac{F_i}{F_j} = \frac{\sigma_i}{\sigma_j} \cdot \frac{\tau_i}{\tau_j} \cdot \frac{\tau_j + \tau_F}{\tau_i + \tau_F} \qquad (3)$$

Since the fall-out of Be^7 and P^{32} has been measured, $\sigma_{Be^7}/\sigma_{P^{32}}$ can be deduced from the minimum observed concentration ratio $C_{Be^7}/C_{P^{32}}$ in rain water, τ_F can be calculated. The value of τ_F is, however, very sensitive to various measured quantities and since the minimum value $\sigma_{Be^7}/\sigma_{P^{32}}$ is not known to better than $\pm$ 15 per cent in the present measurements we cannot hope to obtain an accurate value of τ_F. In order to see if the data are consistent, we will therefore use the value of 40 days for τ_F and calculate the ratio $\sigma_{Be^7}/\sigma_{P^{32}}$.

We group the measurements at Bombay separately for 1958 and the period 1956–58. These are summarised in Table 2.

The calculated value of $\sigma_{Be^7}/\sigma_{P^{32}}$ using $\tau_F = 40$ days is found to be in good agreement with that observed. It is, however,

* The lowest ratio, $C_{Be^7}/C_{P^{32}}$ during 1957 was 100 ± 15, which is higher than the value 60 ± 10 observed during 1958 series of measurements. However, the total number of samples in 1958, where this ratio was measured was only 11, and the two results are not inconsistent.

smaller than that calculated by Lal, Malhotra, and Peters (1958) by a factor of ~ 3. Since the observed fall-out of Be^7 agrees extremely well with the calculations, the discrepancy lies in the estimation of $\sigma_{P^{32}}$ which seems to have been underestimated in the calculations.

8 Acknowledgement

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