Stratospheric circulation studies based on natural and artificial radioactive tracer elements

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ABSTRACT

The nature of large scale stratospheric circulation is studied using the cosmic ray produced isotopes P^{32}, Be^{7}, S^{35} and Na^{22} as tracers. Supplementary information obtained from observations of the distribution of the bomb-produced Na^{22} and radongenic Pb^{210} is taken into account. The activities of these tracer elements have been measured in the stratospheric air, up to altitudes of 20 km, during 1960–64. Data are fairly extensive for studying the characteristics of the mean circulation in the stratosphere as well as seasonal changes in patterns of mixing/transport of air in certain regions of the stratosphere.

The interpretation of the data on cosmic ray tracers is based on a comparison of their observed activities with the expected production rates due to cosmic rays. For this purpose, the work of Lal & Peters is extended to evaluate the variations in the relative production rates of the isotopes P^{32}, Be^{7}, S^{35} and Na^{22} in the atmosphere. These have to be taken into account when isotope data are compared for different altitudes and latitudes in the stratosphere where relative isotope production rates are different because of the markedly different prevailing energy spectrum of nucleons.

The analysis allows us to distinguish three zones in the lower stratosphere (below 20 km), well separated from the tropopause, having distinct circulation patterns. These regions are separately well mixed either vertically or horizontally; the mean time of residence of aerosols in these regions differs appreciably too. The most stable region in the stratosphere is found to be 18–20 km region at 0–30° latitude, where apparent residence times are of the order of twenty months. Polar regions are observed to exhibit an enhanced vertical mixing during November–February. Combining these results with the observations of dispersion of bomb-produced Na^{22}, which appeared in significant amounts from early 1962 onwards all over the stratosphere, we deduce that in the polar regions, vertical mixing occurs rapidly during November–February so that any activity injected in this region at 20 km or so mixes downwards at the rate of about 1.5 km $month^{-1}$. It is concluded that the observed spring peaks in the troposphere are merely the consequence of this phenomena which is triggered in upper levels (above 20 km) of the stratosphere during October–November.

The observations of concentrations of Pb^{210} in the stratosphere are discussed. The analysis reveals that an appreciable gravitational settling of Pb^{210} seems to have occurred, at least during the period over which data were collected, from the stratospheric air between 55°–75° latitude. These observations imply that the residence time of air in this region of the atmosphere is appreciably higher than that deduced from tracers which attach themselves to aerosols. Lastly, the Pb^{210} data indicate that appreciable amounts of tropospheric radon presumably enter the equatorial stratosphere; this conclusion rests on the observation that Pb^{210} concentrations are higher in this region compared to that in the surrounding air.

1. Introduction

Since the detection in 1955–57 of several cosmic ray produced isotopes of half-lives in the range of weeks to years (cf. LAL & PETERS, 1962), attempts have been made to study features of atmospheric circulation using them as tracers. With the development of high altitude air sampling techniques for studying the distribution of fission products in the atmosphere, it became possible to measure systematically the activities of all cosmic ray produced isotopes of meteorological interest, e.g. P^{32} (14 days), P^{33} (25 days), Be^{7} (53 days), S^{35} (87 days) and Na^{22} (2.6 yrs.) in air up to altitudes of ca. 20 km (FRIEND *et al.*, 1961; STEBBINS, 1961). For sam-

pling air above 20 km, balloon borne samplers have to be used; the technique is feasible but has not yet led to systematic explorations. RAMA & HONDA (1961) and FRIEND *et al.* (1961) reported the first measurements of Be^7, P^{33} and P^{32} activities in tropospheric and stratospheric air. The work was soon extended to the two other activities, S^{35} and Na^{22} by BHANDARI & RAMA (1963). These investigations showed that the four isotopes, P^{32}, P^{33}, Be^7 and S^{35} existed in near secular equilibrium with their production in all stratospheric regions, and that marked departures existed only for the long-lived isotope, Na^{22}. Essentially, the conclusion till the time of these investigations was that only Na^{22} could be used for studying time scales of mixing in the stratosphere as the half-lives of other isotopes were too short to reveal any useful information.

In this paper, we discuss the results of extensive measurements[1] of the concentrations of cosmic ray produced isotopes, P^{32}, Be^7, S^{35} and Na^{22} in the atmosphere, particularly for the stratospheric regions. In order to interpret the data, we compare their observed concentrations with those expected at secular equilibrium with their production due to cosmic ray interactions. Fairly accurate calculations to this end are available due to the work of LAL & PETERS (1962) for all isotopes except for Na^{22} for which only approximate calculations were made so far. We have now estimated the altitude-latitude variation of Na^{22} in the atmosphere. Also, for the case of Be^7, P^{32} and S^{35}, we have obtained profiles of relative variations in their production rates for all parts of the atmosphere. Curves showing this variation for Na^{22}, Be^7 and P^{32} are presented here.

A comparison of the observed and expected isotope concentrations/concentration ratios has revealed much useful information regarding the general features of stratospheric circulation. The picture is supplemented by studies of bomb-produced Na^{22} activity which appeared in the stratosphere in significant amounts around mid-1962. One of the important conclusions of our work is that contrary to earlier belief, Be^7 remains an useful tracer for several regions of the stratosphere as it does not exist in secular equilibrium with production except in the equatorial stratosphere at 18–20 km.

[1] The data presented here form part of the Ph.D. thesis of N. BHANDARI (1965).

To obtain information complimentary to above studies, we have also studied the activities of Pb^{210} in the atmosphere. Our results, combined with those of Isotopes Inc. and Health and Safety Laboratory (FRIEND *et al.*, 1961; FEELY *et al.*, 1963; HARDY *et al.*, 1961) show that the equatorial stratosphere presumably receives an appreciable supply of radon from the troposphere. Furthermore, the data seem to be of relevance for studying the possible removal of radioactivity from the stratosphere by gravitational settling of large-sized aerosols. If such aerosols are not continually present in the stratosphere, the residence times derived in this paper, particularly for the high latitude stratospheric air, represent only lower limits.

In the following we will first discuss the source functions of cosmic ray produced isotopes of interest as these are important for an evaluation of the data as well for studying the suitability/limitations of cosmic ray produced isotopes as tracer elements for stratospheric circulation.

2. Cosmic ray source functions of Be^7, P^{32}, S^{35} and Na^{22}

The cosmic ray production rate of several isotopes have been evaluated for the entire atmosphere by LAL & PETERS (1962) using cosmic ray data and directly determined production rates of isotopes in atmospheric nuclei at mountain altitudes. This method has yielded a fairly precise picture of the altitude-latitude variation of Be^7, P^{32} and S^{35} in the atmosphere. In the case of Na^{22} some uncertainty exists in its *absolute* production rate because no normalisation point exists as yet.

We will now briefly discuss the results of calculations of LAL & PETERS (1962), relevant to the present "tracer" studies and then present the altitude-latitude variation curves for Na^{22} as have been derived by BHANDARI (1965) following the star-size and α/p method developed earlier (LAL, 1958).

2.1. *Altitude-latitude variation in the production of P^{32}, Be^7, S^{35} and Na^{22}*

The results of calculations of the altitude-latitude variation in the production rates of the isotopes P^{32}, Be^7 and Na^{22} are graphically presented later in Section 3 where we discuss their experimentally measured concentrations. For

example, see Fig. 3 which shows the surfaces of iso-production of Be^7, up to 30 km altitude, for a meridional crossection of the atmosphere. The production of P^{32} can be estimated from Fig. 4 (Section 3), which gives the ratio at production in the atmosphere for Be^7 and P^{32} isotopes. Similar curves for S^{35}/Be^7 ratios have been obtained (BHANDARI, 1965). They are not presented here as our data on S^{35} are not very extensive, and furthermore, appreciable contributions due to nuclear weapons existed in the case of S^{35} during most of the period of our measurements.

The estimated rate of production of Na^{22} is presented graphically in Fig. 5 (Section 3). This isotope is a high-energy product as its production threshold (from argon) is ~ 150 MeV, in contrast to Be^7 (P^{32}) whose formation from N, O (Ar) requires nucleons of kinetic energy above 30–40 MeV only. This large difference in the production threshold explains why the relative production rates of isotopes, e.g. P^{32} and Na^{22} (see Figs. 4 and 6; Section 3) are so different for different regions of the atmosphere.

These variations were not considered earlier in the interpretation of observed ratios Na^{22}/Be^7 (BHANDARI & RAMA, 1963) and Be^7/P^{32} (FRIEND *et al.*, 1961; RAMA & HONDA, 1961) in the stratosphere.

2.2. *Suitability of cosmic ray produced isotopes for studying stratospheric circulation*

The source functions of cosmic ray produced isotopes make them ideal tracers for the study of several important meteorological processes (LAL & PETERS, 1962; BHANDARI, 1965). We will discuss here only one aspect of the source function which seems to be of relevance to their application as tracers for high-altitude circulation. In Fig. 1 we have plotted the rates of occurrence of nuclear disintegrations *per cubic meter* (ambient) as well as those occurring in a 10 cm^2 column above a certain height, separately for three latitudes 10°, 40° and 70°. Isotope production rates are roughly proportional to the disintegration rates given by the curves. For a given isotope, the constant of proportionality varies somewhat with latitude and altitude in the atmosphere as discussed earlier in Section 2.1. Nevertheless, the curves in Fig. 1 are quite adequate to see qualitatively the behaviour in the variation of source functions of various isotopes in the atmosphere.

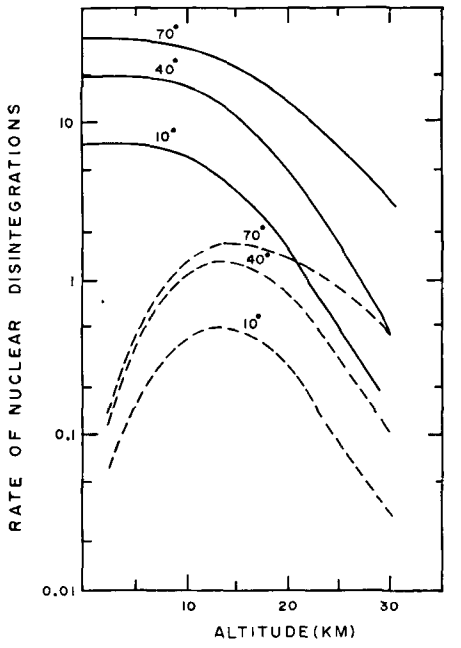

FIG. 1. Rates of nuclear disintegrations in the atmosphere at 10°, 40° and 70° latitude. The dotted curves represent nuclear disintegrations/min. m^3 air (ambient), while the solid curves give the total number of disintegrations occurring in a 10 cm^2 column of air above a given altitude.

It is seen from Fig. 1 that the total integrated production above 25 km is only about 10–15 % of that occurring in the stratosphere and thus cosmic ray produced isotopes connot serve as suitable tracers for seeing the effects of downward vertical mixing from levels above 25 km. In most stratospheric regions below this altitude, however, the production is quite appreciable and they are thus quite suitable.

It thus becomes clear that for studying vertical mixing above ~ 25 km, one must rest the information on other tracers. Fission debris injected at high altitudes is quite suitable for such studies, and we do in fact make use of the opportunity offered by a rather well defined injection of Na^{22} in the stratosphere, as discussed later.

3. Experimental results and discussion

3.1. *Observations on the cosmic ray produced P^{32}, Be^7, S^{35} and Na^{22}*

The concentrations of P^{32}, Be^7 and S^{35} have been measured in the atmosphere during Aug. 1963–April 1965. (BHANDARI, 1965). Data on

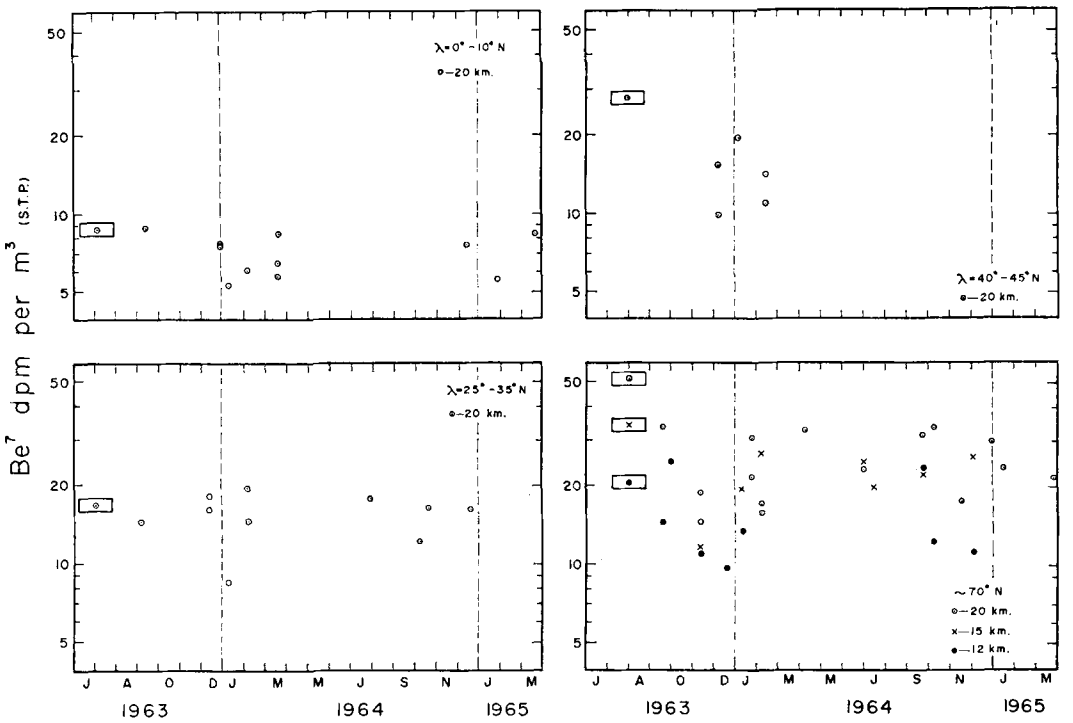

FIG. 2. Observed concentrations of Be^7 (d.p.m./10^3 m^3 air S.T.P.) in the stratosphere during 1963–65 at latitudes 0°–10° N, 25°–35° N, 40°–45° N and 70° N. The expected saturation levels are shown by points encased in boxes.

Be^7 and P^{32} are also available for the period 1959–1961 due to the work of RAMA & HONDA (1961), FRIEND *et al.* (1961) and FEELY *et al.* (1963). It has been discussed that except during late 1961 to early 1963, both Be^7 and P^{32} activities were principally of a cosmic ray origin (BHANDARI, 1965; DREVINSKY *et al.*, 1964).

In the case of Na^{22}, only the data available during Nov. 1960–Aug. 1961 are considered to represent natural cosmic ray levels (BHANDARI & RAMA, 1963; BHANDARI, 1965). Subsequently particularly from early 1962 onwards, Na^{22} concentrations shot up rapidly in the stratosphere. All measurements made in our laboratory on Na^{22} concentrations in air samples collected till the end of 1964 (BHANDARI, 1965; BHANDARI, BHAT and KHARKAR, 1966) are discussed later in this Section.

By late 1962, the concentration of Na^{22} reached values two orders of magnitude higher than the natural cosmic ray levels. How and when exactly such large amounts of Na^{22} were introduced is not clear at the moment. In the case of S^{35}, data which can be considered to represent natural levels are few. We will not discuss them in this paper as the conclusions based on their studies are similar to those derived from comparable half-life isotope, Be^7, for which data are extensive.

The salient features of the various data on the cosmogenic activities and on Na^{22} which was injected by nuclear weapon testing are summarised below:

(*a*) *Absolute Be^7 and P^{32} concentrations.* At a level of 20 km the observed Be^7 concentrations (Fig. 2) at latitudes, 0°–10° and 25°–35°, are close to the expected secular equilibrium values. (The errors in the measurements of Be^7 activity are about ± 10 %; another source of error due to uncertainties in the amount of air sampled, which may partly be systematic in nature, has to be taken into account while considering absolute concentration of Be^7.) Similar data for high latitudes at the 20 km level (Fig. 2) show that the observed concentrations are significantly lower than those expected for secular equilibrium. Both at 40°–45° and 70°, large departures occur; at 70° N where data are

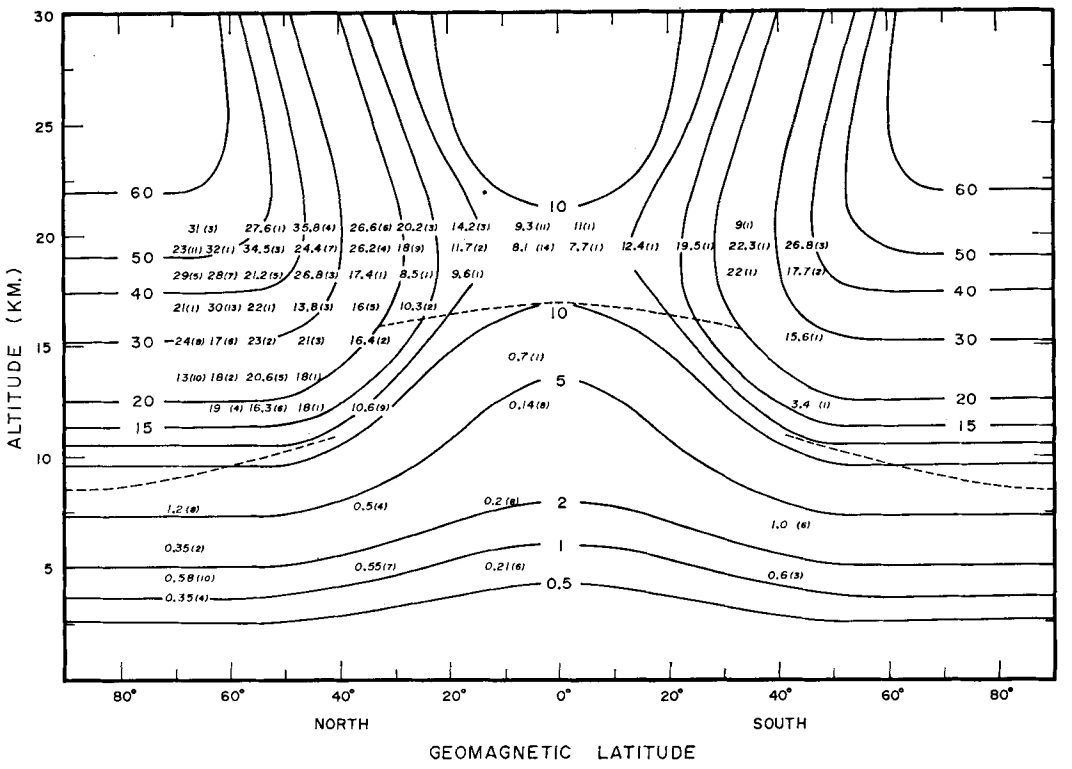

FIG. 3. Mean profile of Be^7 concentrations (d.p.m./m^3 S.T.P.) in the atmosphere. (Sources of data are given in the text.) Figures within parenthesis denote the number of samples on which the values are based. Solid lines represent surfaces of equal production rate of Be^7; numbers on these curves give the expected secular equilibrium concentrations (d.p.m./m^3 air S.T.P.). The dotted curves show the mean tropopause height.

extensive with respect to time, marked seasonal fluctuations are seen to occur.

The concentrations of Be^7 at 12, 15, and 20 km levels at 70° N show features expected to arise from a fairly good vertical mixing during October–February, and it is only during mid-months of the year when concentrations at different levels show some trend of an increase with altitude. Throughout the year, the 20 km level remains undersaturated by about a factor of 1.5 or more. The P^{32} data which are not presented here show similar features at 70° N (BHANDARI, 1965) indicating that mixing occurs fairly rapidly.

The annual global distribution of cosmogenic Be^7, obtained by combining these data with those available due to the work of RAMA & HONDA (1961), FRIEND *et al.* (1961) and FEELY *et al.*, 1963, is shown in a north-south cross-section of the atmosphere in Fig. 3. The data indicate that the concentrations of Be^7 are fairly uniform in the stratosphere at latitudes greater than 30°. Similar features are exhibited by the P^{32} data (BHANDARI, 1965).

(*b*) *Be^7/P^{32} ratios:* The observed activity ratios, Be^7/P^{32} in the stratosphere vary between about 55 and 95 (Fig. 4). High ratios are observed in the equatorial stratosphere at 18–20 km altitude. According to LAL & PETERS (1962), the expected ratio at secular equilibrium values should range between 100 and 120. It is obvious that any information derived using Be^7 concentrations, or the activity ratios, Be^7/P^{32}, depends very sensitively on the accuracy with which the expected secular equilibrium values are known. An error of 20 % in the production estimates can lead to entirely different conclusions as the half-lives of these isotopes are short. We will therefore only use the calculated relative variation and estimate from the data

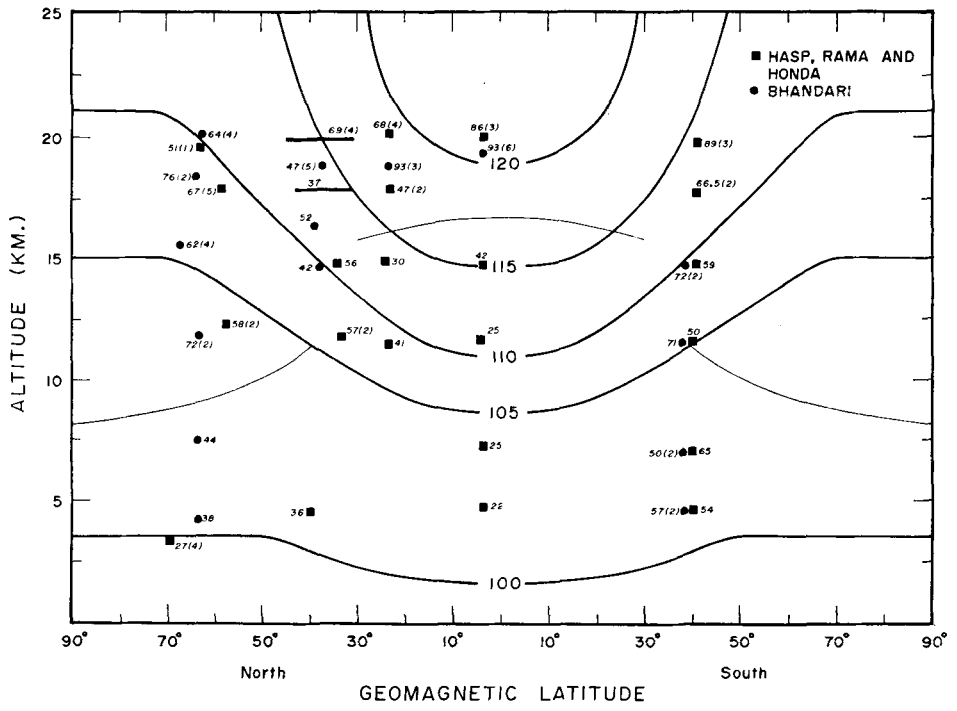

FIG. 4. Observed mean Be^7/P^{32} activity ratios in the atmosphere. Figures within parenthesis denote the number of samples on which the values are based. Solid lines represent surfaces of identical relative production rates of Be^7 and P^{32}; the numbers on these curves give the expected secular equilibrium activity ratio, Be^7/P^{32}. The thin curves show the mean tropopause height.

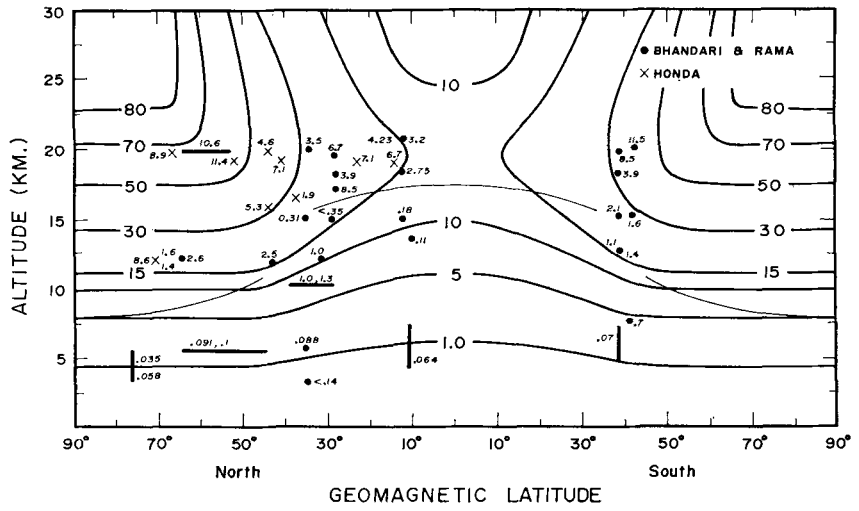

FIG. 5. Observed Na^{22} concentrations in the atmosphere (d.p.m./10^3 m^3·air S.T.P.) during November 1960–August 1961. Figures within parenthesis denote the number of samples on which the values are based. Solid lines represent surfaces of equal production rate of Na^{22}; numbers on these curves give the expected secular equilibrium concentrations (d.p.m./10^3 m^3 air S.T.P.). The thin curves show the mean tropopause height.

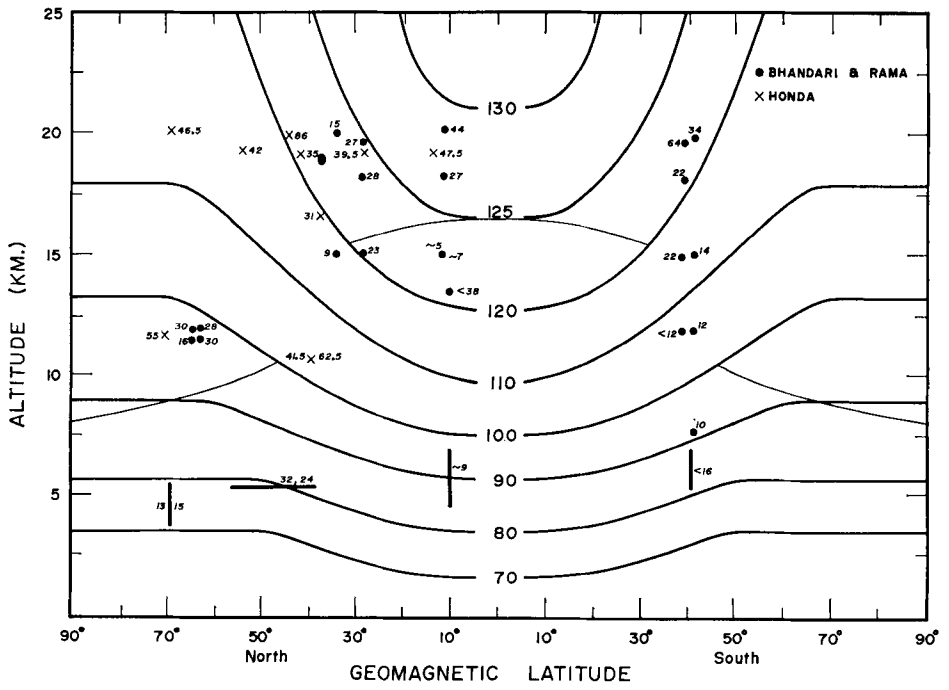

FIG. 6. Observed Na^{22}/Be^{7} activity ratios in the atmosphere during 1960–61. Figures within parenthesis denote the number of samples on which the values are based. Solid lines represent surfaces of identical relative production rates of Na^{22} and Be^{7}; the numbers on these curves give the expected secular equilibrium activity ratio, Na^{22}/Be^{7}. The thin curves show the mean tropopause height.

themselves what secular equilibrium concentration or ratio values are expected.

(c) *Na^{22} activities before Sept. 1961 (pre-bomb period):* The absolute concentrations of Na^{22} are considerably below the secular equilibrium values in all parts of the stratosphere (Fig. 5). In the equatorial regions at levels of 20 km, its activity attains only about 30 % of the saturation value; in all other regions the extent of saturation is only about 15–17 %. The degree of saturation of Na^{22}/Be^{7} ratios in all regions of the stratosphere, well separated from the tropopause, ranges from 25 to 30 % (Fig. 6).

Unlike the case of Be^{7} (P^{32}), any small uncertainty in the production estimates of Na^{22} does not cause serious errors in evaluation of time scales of mixing since its activity is considerably undersaturated.

(d) *Na^{22} activities during 1962–64 (bomb-period):* A fairly steep rise in the concentrations of Na^{22} occurred in early 1962 at all latitudes at 20 km (Figs. 7 and 8), leading to concentrations during most part of June 1962–June 1963 which are higher by about two orders of magnitude compared to those observed in 1960–61. The resulting levels were essentially similar at high and mid-latitudes, but lower by a factor of about eight at equatorial latitudes.

A well-defined peak in Na^{22} concentrations occurred at all latitudes during October–November 1963, the increase being the highest at high latitudes.

Most of the Na^{22} data mentioned above refer to 20 km altitude; concentrations at lower altitudes have been measured only at 60°–70° latitudes (Fig. 8). Throughout late 1963, Na^{22} concentrations at 12–20 km are markedly higher at higher altitudes. During 1964, this holds good only for a few months, June–August. At all other times, large fluctuations are observed. There occur rather well defined peaks in Na^{22} concentrations which occur successively later at the lower levels, with the tropospheric concentrations (at 4.5 km) reaching a maximum around June–July, 1964.

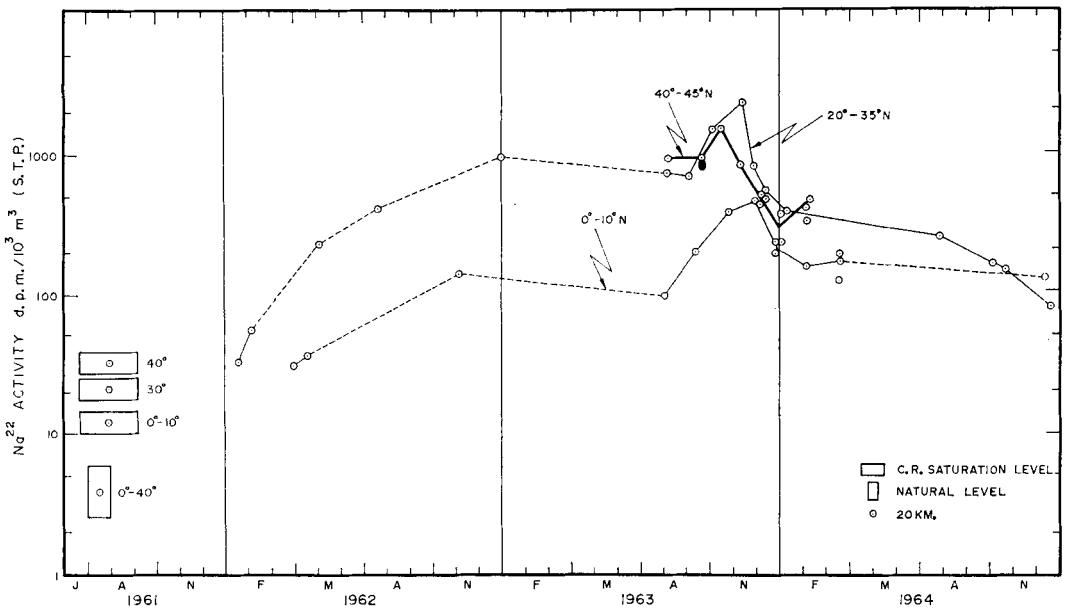

FIG. 7. Observed Na^{22} concentrations (d.p.m./10^3 m^3 air S.T.P.) during 1962–64 at the 20 km level at three latitude regions, 0°–10° N, 20°–35° N and 40°–45° N. All values are decay corrected to 1st September, 1961.

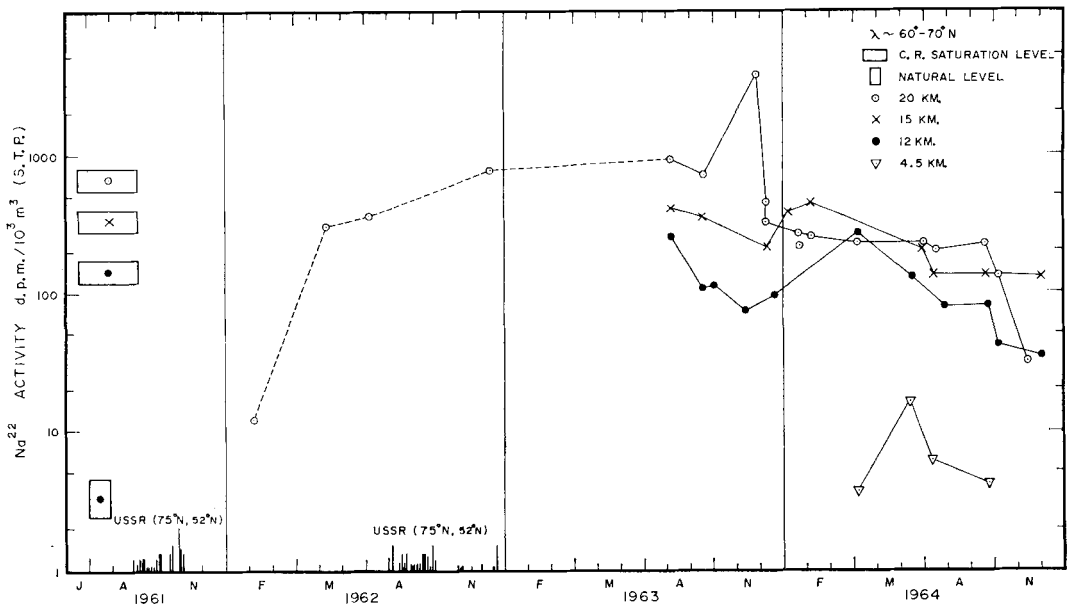

FIG. 8. Observed Na^{22} concentrations (d.p.m./10^3 m^3 air S.T.P.) during 1962–64 at 60°–70° latitude at four levels, 4.5, 12, 15 and 20 km. All values are decay corrected to 1st September, 1961.

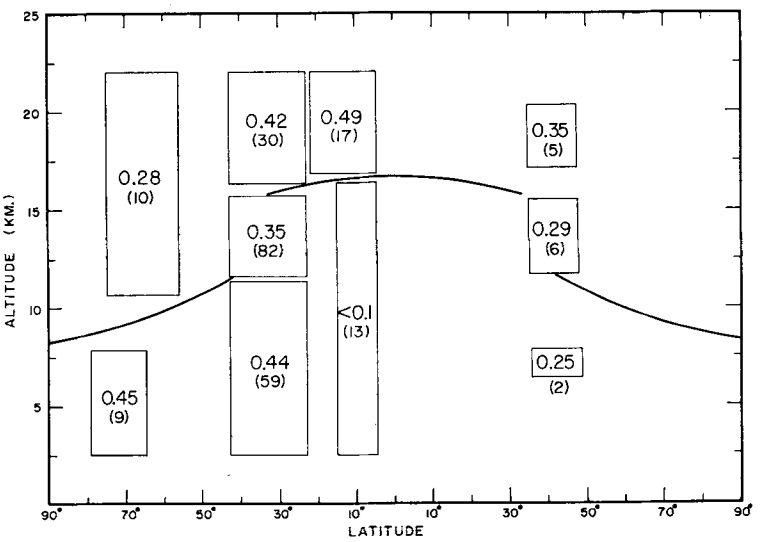

FIG. 9. Mean concentrations of Pb^{210} (d.p.m./10^3 S.C.F. air; 1 S.C.F. = 0.028 m^3 S.T.P.) in certain zones in the atmosphere. (Sources of data are given in the text.) Figures within parenthesis denote the number of samples on which the values are based. The thick curves show the mean tropopause height.

3.2. *Pb^{210} concentrations in the stratosphere*

Extensive data on the concentrations of Pb^{210} in the atmosphere are available (FRIEND *et al.*, 1961; HARDY *et al.*, 1961; RAMA & HONDA, 1961; BHANDARI, 1965). The results have been analysed from the point of view of studying the global fallout of Pb^{210} by BHANDARI, LAL & RAMA (1966). We present here the data on the mean concentrations of Pb^{210} in certain atmospheric zones (Fig. 9). The experimental errors on individual measurements on which the mean values are based are of the order of 10–15 %. Therefore, aside from any errors arising from pooling together results from various laboratories, or due to errors in ascertaining volume of air sampled, the mean concentration values shown in Fig. 9 are certainly accurate within 5 %. The observed scatter in the concentrations in any given zone leads to the conclusion that the error on the mean values is about or less than 10 %.

The Pb^{210} data presented in Fig. 9 refer primarily to the period 1959–61. No evidence exists for an appreciable production of Pb^{210} by nuclear weapons during this period. The observed distribution, in particular the absence of any excess values at high or mid-latitudes, supports such a conclusion in any case and we assume henceforth that the atmospheric Pb^{210} concentrations presented in Fig. 9 arise solely as a result of leakage of radon from the earth's crust.

The most significant features of the data shown in this figure are that, (*a*) the stratospheric air at 55°–75° latitude is lower[1] in its Pb^{210} content (0.28 d.p.m./10^3 S.C.F.) compared to the "surrounding" regions, (0.35–0.45 d.p.m./10^3 S.C.F.) and (*b*) the region of highest Pb^{210} concentration is presumably the equatorial stratosphere where the mean concentration is 0.49 d.p.m./1000 S.C.F. It is difficult to ascertain whether the errors in measurements obviate the conclusion (*b*). Further work will be necessary to confirm the validity of such a distribution occurring in the stratosphere. We will, however, assume in further discussions that the data are reliable and evaluate their implications.

4. Interpretation of data

We have just discussed the salient features of the observed distribution of radioisotopes of diverse origins in the atmosphere. We will now assess some features of large scale stratospheric

[1] It may also be noted that the Pb^{210} concentration in the "gap" region is 0.35 (82 measurements) compared to the value of 0.45 (68 measurements) for the 25°–75° tropospheric air.

circulation from the observed time and space changes in the concentrations of these tracers. No attempts will be made to integrate the information with that available from conventional meteorological measurements; thus the emphasis will be laid on what happens, and as to what are the time scales of significant processes rather than on how the processes are brought about. Tracer studies provide an integrated picture of the "net" transport and mixing processes and to seek further details one has to resort to information available from other studies.

Most data discussed under Section 3 refer to a sampling period of about four years during 1960–64. We are aware that several features, besides the seasonal effects as found for the polar stratosphere may not reproduce in time and intensity over successive years. Thus, certain unique effects observed, e.g. the sharp transients in the bomb-produced Na^{22} in the stratosphere during October–November 1963 may not recur in the manner observed for such later injections, if any. These facts have to be borne in mind in generalising the picture of stratospheric circulation to be discussed below.

4.1. *Inferences based on observations of P^{32}, Be^7 and Na^{22}*

For convenience, we will separate the discussions of Pb^{210} observations as this tracer is injected in the stratosphere from the troposphere. The interpretations based on observation of the tracers P^{32}, Be^7 and Na^{22} which are produced *in situ* in the stratosphere, and the bomb-produced Na^{22} which was placed at very high levels, will be discussed first for their implications to the upper and lower stratospheric circulations:

(i) *Upper-stratospheric circulation:* It was pointed out in Section 2.2 that, considering their source functions, the cosmic ray produced isotopes are not well suited for the study of high-stratospheric circulation (>25 km). The characteristics of this circulation in the present work emerge only from the observations of bomb-produced Na^{22}, primarily during 1963. Further, as the highest sampling altitude in present investigations is 20 km, our conclusions are limited to what one sees in way of time changes in the distribution of Na^{22} due to its downward mixing from very high altitudes, around 50 km or even higher. (The observed increases during early part of 1962 probably represent the direct injections in the lower stratosphere.) It has not yet been possible to ascertain whether Na^{22} was also injected by the major test-series carried out during 1962 (see Figs. 7, 8 in which the time of principal air-detonations has been marked). However, since the transients in the Na^{22} concentrations are observed to occur around November 1963, it is clear that this Na^{22} activity represents the part that was placed one or even two years earlier at high altitudes.

The sharp increases observed around October–November 1963 at all latitudes at the 20 km level after such an elapse of time seems to be a very significant observation. A feature of this type has not been reported previously. It could have been missed in earlier investigations because of lack of sufficient sampling in these months; alternatively it could be a non-recurring phenomena. In any case, our observations during 1963 can only be interpreted as being due to an enhanced vertical mixing between the 20 km level and higher levels (during October–November) at all latitudes (at least over the entire latitude belt 20°–70°, where the observed Na^{22} concentrations are identical during October–November 1963—see Figs. 7 and 8). We can rule out that the debris first mixed down to the 20 km level at the high latitude region and then reached other latitudes by horizontal mixing primarily because of the following consideration. The transitions in concentrations occur within about a month's interval throughout the stratosphere. Any rapid horizontal mixing assumed to achieve the observed effects throughout the stratosphere, following a principal injection at 20 km level at high latitudes, would destroy the observed well-defined gradients in the activities of the cosmic ray produced activities Be^7 and Na^{22} (Figs. 2, 3 and 5).

Thus, we observe a time delay of 1–2 yrs. between the placement of Na^{22} at high altitudes and its appearance at the 20 km level. The observations suggest that following an earlier dispersion and rather uniform mixing at high levels, the activity appeared at the 20 km level by fairly rapid vertical mixing processes (of the order of 1 month) during months of October–November. The vertical mixing either ceases or is quite complete within a period of about 2 months thereafter. The observed magnitude of

the increase at equatorial latitudes suggests that the extent of vertical mixing at high latitude regions is considerably greater than at equatorial latitudes.

(ii) *Lower-stratospheric circulation:* For evaluating features of this circulation, we have available to us the extensive cosmic ray tracer data, as well as that on the bomb-produced Na^{22} and radongenic Pb^{210} activities (these observations have been summarised in Section 3).

The pre-bomb Na^{22} data have been discussed earlier by BHANDARI & RAMA (1963). They took a mean value of 110×10^{-5} for the ratio of production of Na^{22} and Be^7 isotopes. Fig. 6 shows that there exists a significant variation at production in the stratosphere and that the expected secular equilibrium activity ratios lie in the range $(100–130) \times 10^{-5}$. The value of 110×10^{-5} used by BHANDARI & RAMA (1963) happens to be coincidentally close to that expected from detailed calculations (Section 2.1) because they adopted values lower by about a factor of two[1] for the production of both Na^{22} and Be^7. Their interpretations which are based on the measured concentrations of either of these isotopes are therefore in error. We therefore find it necessary to reconsider the implications of Na^{22} data published by BHANDARI & RAMA (1963). Also several new measurements have now been added (Figs. 5, 6) due to the work of BHANDARI (1965) and HONDA (1965). In the case of Be^7, the discussions so far have been based on rather limited number of measurements; data were particularly scanty for the equatorial regions. The present data allow one to estimate the ratio of Be^7/P^{32} at production precisely and thus study even small departures from equilibrium. Thus, we are able to use Be^7 as a stratospheric tracer, in contrast to the work carried out so far where one had to be satisfied with the statement that Be^7 exists in near secular equilibrium in the stratosphere (cf. RAMA & HONDA, 1961), except in regions close to the tropopause.

As discussed in Section 3.1, large departures from secular equilibrium exist for Na^{22} and measurable differences exist even for Be^7. These departures are indicative of the time scales of movement of air within the stratosphere. Wherever two isotopes of such different half-lives show departures in the stratosphere one can check on the validity of the mixing/transport model adopted, and thus very meaningful time scales of circulation can be deduced. One can consider two principal cases of mixing/transport of air; (1) air mixes by eddy diffusion, and (2) air moves by advection. In principle, the true situation is very complex and the circulation is not "pure"; varying proportions of the two modes being operative. In studies based on cosmic ray tracers, where cosmic ray production occurs in all regions of the atmosphere, though in a latitude-altitude dependant manner, it becomes difficult to resolve the role of the first two processes, and even to resolve the direction of advection because there exist, for instance, several directions of positive gradients at production and depending on how the air is assumed to move, different time scales are obtained. Use of isotopes of different half-lives can resolve the situation to some extent, but this does not work out in practice as measurements are not available with the required degree of precision. Furthermore, large and small scale eddy diffusion processes do play an important role complicating the situation.

With the above considerations in mind, our general approach will be not to resolve the mode of circulation. We will only deduce from the isotope data what is the dominant effective circulation pattern, and evaluate the mean time scale for mixing/transport. These time scales will be designated as "apparent". The term thus implies an ignorance of the details of the mixing processes: this parameter is, however, quite useful meteorologically as it signifies the degree of stability of the region.

The "apparent" ages for any zone in the stratosphere can be deduced in two ways, assuming in either of the cases that the environmental air which exchanges with this zone has a zero or a negligible concentration of the isotope under study, i.e. the concentrations build up in the zone essentially due to direct cosmic ray production in the zone. This assumption holds

[1] The calculations of BHANDARI (1965) yield that the stratospheric production of Na^{22} is higher by about a factor of two compared to the values used by BHANDARI & RAMA (1963). In the case of Be^7, they assumed its production to be 65 % of the values calculated by LAL & PETERS (1962), considering the observation (RAMA & HONDA, 1961) that all stratospheric measurements which were available till then were 0.65 times the values predicted by LAL & PETERS (1962), and assuming that the Be^7 concentrations were at secular equilibrium in the stratosphere.

good, for example, in the polar stratosphere where isotope concentrations are considerably higher than those in the surrounding regions, equatorial stratosphere or the polar troposphere. Similarly, it also applies for the case of the equatorial stratosphere if a Brewer-Dobson circulation exists or if the horizontal equatorward mixing/transport is slow. With this basic assumption, we write down the relation between apparent "ages", τ and the measured degree of saturation of isotope concentrations for two cases discussed below:

$$\tau_a = \frac{1}{\lambda} \ln 1/(1-S) \tag{1}$$

or

$$\tau_b = \frac{S}{\lambda} \cdot \frac{1}{1-S} \tag{2}$$

with

$$S = A/A_0, \tag{3}$$

where A_0 and A are the expected secular and the observed isotope concentrations respectively, and λ is the disintegration constant of the isotope. S as defined by equation (3) represents the degree of saturation of activity in the zone with respect to secular equilibrium conditions.

Equation (1) applies to the simplest case which is analogous to the case of target irradiation in the laboratory, the apparent age, τ_a, signifying the time required to build up activity of an isotope, under constant rate of irradiation starting with a "clean" air parcel at time $t = 0$. In the second case, τ_b gives the apparent age on a box-model concept (LAL & PETERS, 1962); the activity of an isotope is assumed to be in a steady state in the zone as a result of balance between decay, removal and production. Constant irradiation is again assumed. For small values of S, $\tau_a \simeq \tau_b$. However, when the activity has built up to a sufficient extent, i.e. $S \sim 1$, equation (1) underestimates the apparent age considerably, as in its derivation no account is taken of the depletion of the concentration due to mixing of environmental air. We will adopt the box-model concept in our treatment of data as it seems to yield more meaningful apparent "ages". In the case when apparent ages are based on the ratios of activities, τ_b is given by

$$\tau_b = \frac{S(R) - (\lambda_1/\lambda_2)}{\lambda_1} \cdot \frac{1}{1-S(R)}, \tag{4}$$

where $S(R)$ is the degree of saturation of the activity ratio of two isotopes having disintegration constants, λ_1 and λ_2.

Before discussing the stability of the various stratospheric regions, based on the above concepts, we will first evaluate the ratio Be^7/P^{32} at production from the observed data. (The values given in Fig. 4 should only be taken for considerations of relative values, as even a 10 % error can cause serious errors in the values of residence times.)

The most stable region of the lower stratosphere is found to be the 19 ± 1 km level at 0°–30° latitude, based on the pre-bomb Na^{22} data. Considering the values of τ_b (discussed later) for this region, as also the data in the troposphere and rains (BHANDARI, 1965), we deduce that the Be^7/P^{32} values given in Fig. 3 should be multiplied by 0.80 (with an estimated uncertainty of ± 7 %). It seems difficult from the available data to deduce whether the uncertainty in the production ratio, Be^7/P^{32}, arises from wholly or partly in estimation of the production rates of Be^7 and/or P^{32}. We will therefore keep this in mind in evaluating time scales based on Be^7 alone; data on its absolute concentration will be only used to see relative gradients with respect to production.

From the preceding discussions of observations of cosmic ray tracer data (Section 3.1), we first distinguish three zones in the stratosphere having distinct circulation patterns:

(A) 0°–30°, 18–20 km

(B) 30°–70°, 18–20 km

(C) 30°–70°, 12–18 km

The Na^{22}/Be^7 ratios are identical in the zones (A) and (B); however, the absolute concentrations of both Na^{22} and Be^7 (P^{32}) are markedly different, being higher for the high latitude region. Zones (B) and (C) have fairly identical Be^7 and Na^{22} concentrations. The cosmic ray source function is such that, based on the absolute concentrations alone, one cannot deduce whether this arises as a result of vertical or horizontal mixing (Figs. 3 and 5). However, if one considers the Na^{22}/Be^7 ratio data (pre-bomb Na^{22} measurements), it becomes apparent that they are indeed distinct zones, with horizontal mixing being important for zone (B) and vertical in the case of zone (C). The Na^{22}/Be^7 ratios are significantly higher in zone (B) than in zone (C).

TABLE 1

Stratospheric zone	Na^{22} concentrations		Na^{22}/Be^7 activity ratios		Mean
	S	τ_b (months)	$S(R)$	τ_b (months)	τ_b (months)
(A) 0°–30°, 18–20 km	0.32	21	0.33	18	20
(B) 30°–70°, 18–20 km	0.17	9	0.30	15	12
(C) 30°–70°, 12–18 km	0.15	8	0.25	11	10

We have listed in Table 1, the values of S and $S(R)$ for Na^{22} and Na^{22}/Be^7 ratios. The calculated values of τ_b are also given in this table.

The uncertainty in the calculated values of τ_b are estimated to be ± 20 % or less, considering the experimental errors alone; and error of 20 % in the production estimates of Na^{22} and Be^7 will lead to a systematic error of $\leqslant 25$ % in τ_b. As can be seen from Table 1, apparent "ages" estimated on the basis of $S(R)$ seem to be higher than those based on S. However, as discussed earlier, the production rate of Be^7 may have been overestimated by upto about 20 %. If we take this into account, the degree of saturation, $S(R)$ reduces and the values of τ_b based on $S(R)$ agree closer with those based on S. The values of the mean apparent "ages" which we adopt are listed in Table 1. It is seen that zones A, B, and C constitute successively lesser stable regions. These regions are well separated from the tropopause or the "break" at mid-latitudes, where both the isotope activities and ratios are considerably undersaturated and the corresponding values of τ_b are considerably smaller.

Similar apparent "age" calculations based on observed Be^7 and P^{32} activities are possible only for the 30°–70° latitudes, and here too they are less reliable because the value of "S" for these isotopes are close to 1; nevertheless the results are quite consistent with those listed in Table 1.

The only other observation relevant to the lower stratospheric circulations is that on the depletion of the inventory of bomb-produced Na^{22} in the 60°–70° latitude region. We estimate from the data given in Fig. 8, that the mean time of depletion of the polar stratospheric inventory is 9 ± 1.5 months, when considered on an annual basis. This figure is in excellent agreement with that based on cosmogenic tracer data (Table 1).

4.2 *Radongenic Pb^{210}*

Here we will consider the implications of the two significant features observed in the distribution of Pb^{210} in the northern stratosphere, namely, (i) the stratospheric air at 55°–75° latitude, 12–20 km is lower in its Pb^{210} contents compared to that in the surrounding regions, and (ii) the highest Pb^{210} concentrations occur in the equatorial stratosphere (see Section 3.2).

4.2 (i) *Gravitational settling effects*

It seems difficult to attain a Pb^{210} distribution as mentioned under (i) above, unless some fraction of it is removed by gravitational settling from the region under consideration. The half-life of Pb^{210} is sufficiently long compared to the residence times of air as estimated in Section 4.1, so that no measurable gradients are expected to occur within the stratosphere as a whole (note that except in a narrow latitude interval of 10° at the equator, the concentrations of Pb^{210} in the troposphere range between 0.35 and 0.45, and these results are based on rather large number of measurements). The observed lower Pb^{210} concentrations can be accounted for if the mean time of removal of aerosols by gravitational settling is of the order of 36 (± 9) months.

An immediate conclusion of this is that the residence time of air in the polar region is considerably longer than the estimates given in Table 1, which are the apparent values of the residence time of aerosols, as Na^{22} and Be^7 activities are expected to be attached onto them. The mean residence time of air in zone C, for example is then expected to be $1/(1/10-1/36) =$ 14 months, i.e. about forty per cent higher than that deduced on the basis of radioactive tracer data.

4.2 (ii) *Possible intrusions of tropospheric radon in the equatorial stratosphere*

It is tempting to find an explanation for the observed marginal excess Pb^{210} concentrations in the equatorial stratosphere. As wash-out processes are operative in the troposphere for Pb^{210} only and not for Rn^{222} (BHANDARI & RAMA, 1963), it is conceivable to have higher Pb^{210} concentrations in the stratosphere than in the tropospheric air. Such a situation will hold if air containing appreciable Rn^{222} entered the stratosphere. The highest concentrations of Rn^{222} occur in the continental ground level air which has about 0.2 d.p.m. Rn^{222}/g air (BHANDARI, LAL & RAMA, 1966). The average tropospheric concentrations of Rn^{222} is not known, values of $(2\text{–}8) \times 10^{-2}$ d.p.m. Rn^{222}/g air may suitably indicate the range (JACOBI & ANDRÉ, 1963; MOSES, STEHNEY & LUCAS, 1960). Decay of this much radon will lead to a concentration of about 0.4–1.5 d.p.m. Pb^{210}/1000 S.C.F. air. If one considers that the excess Pb^{210} in the equatorial stratosphere is derived due to intrusions (sporadic or continuous) of tropospheric air having Rn^{222} in such concentrations, then it does seem possible to explain the observed excess Pb^{210} concentration in the equatorial stratosphere.

The mechanisms for such intrusions may well be the highly turbulent vertical mixing in the troposphere associated with thunderstorms. Mixing across the tropopause under normal conditions is inadequate as the Rn^{222} concentrations of upper tropospheric air are expected to be of the order of 2×10^{-3} d.p.m. Rn^{222}/g air, which cannot provide enough Pb^{210}.

5. Conclusions

The salient features of the present investigations based on radioactive tracers can be summarised as follows:

(1) The high-level placed Na^{22} activity, following the nuclear weapon test series in 1961 (1962 contributions may be important), descended rapidly to the 20 km level during October–November 1963 at all latitudes.

(2) In the polar regions, further downward mixing of the bomb-produced Na^{22} continued to lower levels up to the tropopause, at a well-defined rate—about 1.5 km month^{-1}. It seems that vertical disturbances start annually around October at high-levels in the stratosphere and this is the trigger or the earliest phase of development in the sequence of events that cause the well-known spring maxima observed in the troposphere.

(3) The most stable region in the lower stratosphere (w.r.t. mixing/transport) is situated at 20–18 km at 0°–30° latitude. In the polar stratosphere (30°–70°), the 20–18 and 18–12 kilometer regions constitute two well mixed zones, with horizontal and vertical mixing being the respective dominant processes operative for their mixing. The apparent "ages" of air in the upper and lower polar zones are estimated to be 12 and 10 months respectively. The isotope data do not allow us to decide uniquely on the mechanisms of transport or mixing in this region. Irrespective of the details of transport, the estimates of apparent ages cannot be in much error, however.

(4) The 12–20 km levels at 60°–70° latitude show seasonal changes in the tracer concentrations. Highly disturbed conditions prevail during October–February (or March) as shown by cosmogenic tracers and bomb-produced Na^{22}. The vertical mixing in the 12–18 km levels seems to be fairly complete, even on the time scales of mean-life of P^{32}.

(5) Gravitational settling of radioactivity occurs from the stratosphere at 55°–75° latitudes. This evidence rests on the study of the radongenic tracer Pb^{210}, which alone provides suitable boundary conditions to observe such effects. The partial rate of removal due to gravitational settling is of the order of 36 months; this appreciably reduces the residence time of aerosols in this region.

(6) Whereas the tracer data discussed in this paper do not lead to a delineation of the importance of the role of the Brewer-Dobson circulation, it seems that appreciable amounts of tropospheric air probably enter via the equatorial tropopause to produce the highest observed Pb^{210} concentrations in the equatorial stratosphere; such intrusions, if they occur, must take place only during violent vertical disturbances in the troposphere.

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ИЗУЧЕНИЕ СТРАТОСФЕРНОЙ ЦИРКУЛЯЦИИ С ПОМОЩЬЮ ЕСТЕСТВЕННЫХ И ИССКУСТВЕННЫХ МЕЧЕНЫХ АТОМОВ

Изучается характер крупномасштабной циркуляции стратосферы с использованием в качестве меченых атомов изотопов P^{32}, Be^7, S^{35} и Na^{22}, образовавшихся под действием космических лучей. Принимается во внимание дополнительная информация, полученная из наблюдения распределений Na^{22}, образовавшегося в результате ядерных испытаний, и Pb^{210}, образовавшегося из радона. Радиоактивность этих элементов измерялась в стратосфере до высоты в 20 км в течение 1960–1964 годов. Эти данные дают довольно обширный материал для изучения характеристик средней циркуляции в стратосфере и сезонных изменений в механизмах перемешивания–переноса воздуха, в некоторых областях стратосферы. Использование радиоактивных изотопов, образующихся под действием космического излучения в качестве «метчиков», основано на сравнении данных наблюдений с ожидаемой скоростью образования этих изотопов. Для этой цели была продолжена работа Lal и Peters'a для вычисления измерения относительной скорости образования в атмосфере изотопов P^{32}, Be^7, S^{35} и Na^{22}. Полученные результаты принимались во внимание при использовании изотопных данных на различных широтах и высотах в стратосфере, где относительная скорость образования изотопов различна из-за заметного различия энергетического спектра космического излучения. Проведенный анализ позволил выделить в нижней стратосфере (ниже 20 км) три хорошо отделенных от тропопаузы зоны с различными механизмами циркуляций. В каждой из этих областей существует хорошее горизонтальное вертикальное перемешивание, а среднее время нахождения аэрозолей в них различно. (Время нахождения аэрозолей в каком-либо объеме определяется как отношение количества частиц в этом объеме к полному потоку их в этот объем.) Найдено, что наиболее устойчивой областью в стратосфере является

зона между 18–20 км по высоте и 0–30° по широте. Наблюдавшееся время нахождения аэрозолей в ней имеет порядок 20 месяцев. В полярной области в течение ноября–февраля наблюдалось усиление вертикального перемешивания. Комбинируя эти результаты с наблюдениями рассеивания Na^{22}, появившегося во всей стратосфере в результате ядерных испытаний с начала 1962 г. в значительных количествах раньше других изотопов, можно заключить, что в полярной области в течение ноября–февраля имеет место быстрое вертикальное перемешивание. При чем любая энергия, введенная в эту область на высоте около 20 км, распространяется вниз со скоростью порядка 1–5 км/месяц. Из этого заключаем, что наблюдаемый весенний максимум в тропосфере является просто распространением возмещения, которое возбуждается в верхних слоях (около 20 км) в октябре–ноябре.

Обсуждаются данные наблюдения концентрации Pb^{210} в стратосфере. Анализ собранных данных показывает, что имеет место осаждение из стратосферы Pb^{210} между 55° и 75° с. ш. Эти наблюдения показывают, что время обновления воздуха в этих областях атмосферы заметно больше чем полученное с помощью меченых атомов, которые присоединяются к аэрозолям. Из наблюдавшейся более высокой концентрации Pb^{210} в экваториальной стратосфере по сравнению с окружающим воздухом следует, что, повидимому, заметная величина тропосферного радона входит в эту область.