AGE ESTIMATES OF STRATOSPHERIC AIR

By
Elmar. R. Reiter

Department of Atmospheric Science
Colorado State University
Fort Collins, Colorado

Colorado
State
University

Department of
Atmospheric Science

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ABSTRACT

It is shown that careful consideration of P^{32}/Be^{7} ratios (both isotopes being generated in the atmosphere by cosmic radiation) not only yields information on the stratospheric or tropospheric origin of air masses. These ratios may also be used to determine the residence time in the stratosphere of tropospheric air that underwent upward transport through the tropopause and re-entered the troposphere at a later date.

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The Institut für Atmosphärische Umweltforschung at Garmisch-Partenkirchen presently conducts routine measurements of P^{32} and Be^{7} concentrations. Both are cosmogenetic radionuclides with a half-life of 14.31 d and 53.3 d respectively.

The behavior of a nuclide which undergoes continuous production, such as for instance by cosmic radiation, is given by

$$\frac{dN}{dt} + \lambda N = S \tag{1}$$

N is the number density of the radionuclide, λ its radioactive decay constant, and S its production rate. A solution to eqn. (1) is given by (Kamke, 1944, p. 17; Lal, 1958)

$$N = e^{-\lambda t} \cdot \int_{o}^{t} e^{\lambda t} \circ S \cdot dt \tag{2}$$

Integration of eqn. (2) yields

$$N = e^{-\lambda t} \left(\frac{1}{\lambda} \bar{S} e^{\lambda t} + C \right) \tag{3}$$

$\bar{S}$ indicates a time-averaged production rate. (A time-variable production rate also yields an analytical solution. See Reiter 1973, 1974). C is an integration constant to be determined by the initial conditions.

Let us assume that tropospheric air which enters the stratosphere either through convective cloud systems or through large-scale ascending motions in the jet-stream has been cleansed completely of radioactive debris by washout and rainout processes. Hence, at $t = o$ we adopt the value $N_o = o$. It follows from eqn. (3) that

$$C = - \frac{1}{\lambda} \bar{S} \tag{4}$$

and

$$N = \frac{1}{\lambda} \bar{S}_s (1 - e^{-\lambda t}) \tag{5}$$

The "clean" tropospheric air, which enters the stratosphere is now assumed to be exposed to stratospheric production levels $\bar{S}_s$ so that an equilibrium concentration N_e is reached asymptotically

$$N_{es} = \frac{1}{\lambda} \bar{S}_s \tag{6}$$

Stratospheric air which has reached this equilibrium concentration level will, if removed into the troposphere at $t' = o$, follow the equation

$$N = N_{es} e^{-\lambda t} = \frac{1}{\lambda} S_s e^{-\lambda t} \tag{7}$$

Eqns. (5), and (6) describe the behavior of completely clean tropospheric air entering the stratosphere and acquiring equilibrium concentrations of a particular cosmogenetic radionuclide. Eqn. (7) gives the behavior of such a "saturated" air mass when re-entering the troposphere. These two extreme conditions are described in Fig. 1 for P^{32} and Be^7 in terms of concentrations normalized with respect to the production rate, $\frac{N}{S}$.

In reality, tropospheric air entering the stratosphere might not be completely clean, but may contain traces of the radionuclide that were produced by weak cosmic radiation in the troposphere. On the other hand, the air mass now in the stratosphere may re-enter the troposphere <u>before</u> equilibrium concentration levels are reached.

Mathematically, these processes can be described as follows:

Tropospheric equilibrium concentrations are given by

$$N_{et} = \frac{1}{\lambda} \bar{S}_t \tag{8}$$

If, at time t = o, tropospheric air with this equilibrium concentration enters the stratosphere, the concentration of the radionuclide under consideration is given by

$$N = \frac{1}{\lambda} S_s(1 - e^{-\lambda t}) + N_{et} \cdot e^{-\lambda t} \tag{9}$$

because the constant C in eqn. (3) assumes the value

$$C = N_{et} - \frac{1}{\lambda} \bar{S}_s \tag{10}$$

We can assume that "old" tropospheric air, at best, shows the concentrations given by eqn. (8). Most likely, observed concentrations will be less because of washout and rainout processes (Lal, 1958), and will lie between

$$0 \leq N \leq \frac{1}{\lambda} \bar{S}_s \tag{11}$$

Normalizing eqn. (9) with respect to stratospheric production rates yields

$$\frac{N}{\bar{S}_s} = \frac{1}{\lambda}\left(1 - e^{-\lambda t}\right) + \frac{1}{\lambda} \frac{\bar{S}_T}{\bar{S}_s} e^{-\lambda t} \tag{12}$$

The factor $\frac{\bar{S}_T}{\bar{S}_s}$ is assumed to have values ranging approximately from 10^{-1} to 10^{-2} (see Haxel and Schumann, 1962).

In Fig. 1 we plotted the concentrations $\frac{N}{\bar{S}_s}$ of Be^7 and of P^{32} following eqn. (5) [curves ①], showing the build-up of concentrations to equilibrium levels. Curve ② indicates the decay of P^{32} from such an equilibrium level according to eqn. (7) Curve ③ shows the solution to eqn. (12)for P^{32}, assuming an extreme value of 10^{-1} for the factor $\frac{\bar{S}_T}{\bar{S}_s}$. This curve, thus, predicts the concentration $\frac{N}{\bar{S}_s}$ for already contaminated tropospheric air that enters the stratosphere.

If "young" stratospheric air, which has not yet reached equilibrium levels of radionuclide concentrations re-enters the troposphere, we can describe this process as follows: At time t' = o the concentration is $N_o \leq N_{es}$, hence

$$N = N_o \cdot e^{-\lambda t} \qquad (13)$$

For an arbitrary value of $\frac{N_o}{\bar{S}_s} = 6$, values of this function are plotted as curve ④ in Fig. 1.

Let us assume that, at time t = o, "clean" tropospheric air enters the stratosphere, where concentration levels build up following curve ①. After 7 days of stratospheric residence the same air mass moves back into the troposphere. Its P^{32} concentration will now follow curve ⑤ if the cosmogenetic production of this nuclide ceases upon re-entry into the troposphere. A small correction will have to be applied to this curve allowing for production of P^{32} by weak cosmic radiation in the troposphere. Such a correction is shown by curve ⑥ in Fig. 1, assuming a value of $\frac{\bar{S}_T}{\bar{S}_s} = 10^{-1}$ which, most likely, is an overestimate of real conditions.

The same reasoning, yielding a similar set of curves, can be applied to Be^7. By considering P^{32}/Be^7 ratios we can estimate, at least theoretically, the time which tropospheric air spends in the stratosphere before it re-enters the troposphere. For the time being we will have to neglect the level of contamination present in the original tropospheric air mass that intrudes at t = o into the stratosphere. We assume that this air, at $t = o$, is "clean" of cosmogenetic radionuclides. Also we assume that production of radionuclides ceases after the same air mass leaves the stratosphere.

As we have shown above, these two assumptions do not quite conform to reality, hence will introduce an amount of uncertainty into our estimates. In view of the fact that ascending motions of tropospheric air into the stratosphere, which occur mainly near jet-streams (Reiter, Glasser and Mahlman 1969, Reiter, 1972), are most likely associated with cloudiness and precipitation, the assumption of $N = o$ at $t = o$ appears justifyable. Because well-defined extrusions of stratospheric air into the troposphere, also occurring near jet streams (see Reiter, 1972), usually take only about three days to reach ground-based measurement stations, the assumption of zero tropospheric production of radionuclides also appears to be of no great detriment.

$\frac{P^{32}}{Be^{7}}$ ratios for "old" stratospheric air that has been in radiative equilibrium and is entering the troposphere at time $t = o$ can be determined from eqn. (7) as

$$\frac{P^{32}}{Be^{7}} = \frac{\lambda_{Be}}{\lambda_{p}} \frac{\bar{S}_{s,P}}{S_{s,Be}} e^{(\lambda_{Be} - \lambda_{P})t} \tag{14}$$

where $\frac{\lambda_{Be}}{\lambda_{P}} = 0.2685$, $\frac{\bar{S}_{s,P}}{\bar{S}_{s,Be}} = \frac{1}{210}$. (see Lal, 1958, and Reiter, 1974).

At $t = o$ this gives a ratio $P^{32}/Be^{7} = 1.2785 \times 10^{-3}$ or $\frac{Be^{7}}{P^{32}} = 782.2$ (see Lal, 1958). Other values are given in Table 1 and Fig. 2.

"New" stratospheric air which has come from a "clean" troposphere at time $t = o$ and is now slowly reaching radiative equilibrium levels shows the following isotope ratios [see eqn. (5)]

Table 1: $\frac{P^{32}}{Be^{7}}$ ratios for <u>old</u> stratospheric air.

t	P^{32}/Be^{7}
0	1.2785×10^{-3}
1	1.2340 "
2	1.1911 "
4	1.1096 "
8	0.9630 "
10	0.8971 "
15	0.7515 "
20	0.6295 "
30	0.4417 "
50	0.2174 "
80	0.0751 "
100	0.0370 "
125	0.0153 "

$$\frac{P^{32}}{Be^7} = \frac{\lambda_{Be}}{\lambda_P} \frac{\bar{\bar{S}}_P}{\bar{S}_{Be}} \frac{(1 - e^{-\lambda_P t})}{(1 - e^{-\lambda_{Be} t})} \qquad (15)$$

Values are given in Table 2 and Fig. 2. It is clearly seen that "new" stratospheric air which has recently come from a clean troposphere can have $\frac{P^{32}}{Be^7}$ ratios which are up to a factor of four higher than the ratio for stratospheric air in radiative equilibrium. This equilibrium ratio is approached asymptotically by "new" stratospheric air. Stratospheric residence times of originally tropospheric "clean" air of 50 days still indicate ratios twice as high than those prescribed by equilibrium considerations.

From this it appears that the accurate determination of P^{32}/Be^7 ratios in massive and rapid extrusions of stratospheric air (especially those linked to jet maxima and cyclogenetic processes) should offer a powerful tool in determining quantitatively and directly the residence times of air masses in the lower stratosphere, especially in the polar front belt of middle latitudes. The rapid downward transport of stratospheric air in cyclogenetic processes should alter the P^{32}/Be^7 ratios shown in Fig. 2 for "new" stratospheric air only very insignificantly within two or three days after the stratospheric extrusion occurred.

Preliminary data obtained in Garmisch-Partenkirchen show relatively high $\frac{P^{32}}{Be^7}$ ratios in summer, relatively low ratios in spring (oral communication by Drs. Sládkovič, Kanter, and Carnuth, June 1973). The tentative interpretation of such data would be that in the stratospheric air extrusions of summer a higher proportion of "young" stratospheric air is involved than in spring. This is in agreement with the fact that during spring the stratosphere "shrinks" in size. Well-organized vertical

Table 2: $\frac{P^{32}}{Be^{7}}$ ratios for New stratospheric air.

t	P^{32}/Be^{7}
1	4.6776×10^{-3}
2	4.5975 "
3	4.5197 "
4	4.4425 "
5	4.3683 "
8	4.1567 "
10	4.0253 "
15	3.7257 "
20	3.4638 "
30	3.0325 "
50	2.4372 "
80	1.9362 "
100	1.7435 "
∞	1.2785 "

transport processes also lead to massive infusions of ozone and nuclear debris from bomb tests from the strasosphere to the troposphere. During summer and autumn the stratosphere increases its mass content by a gradual lowering of the tropopause. Extrusions of stratospheric air near jet-streams, during these seasons, most likely involve air masses which have been incorporated into the stratosphere only recently.

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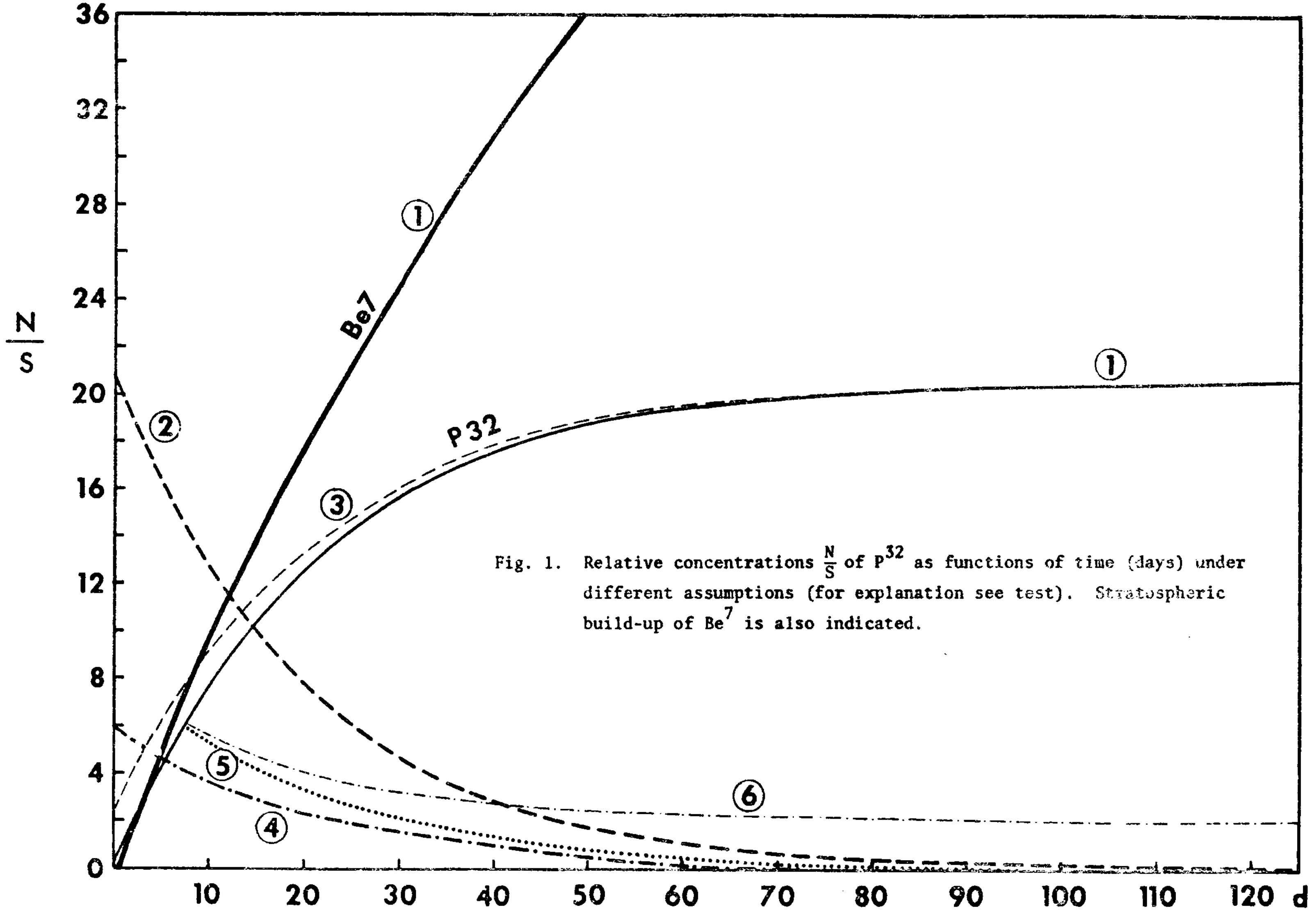

Fig. 1. Relative concentrations $\frac{N}{S}$ of P^{32} as functions of time (days) under different assumptions (for explanation see test). Stratospheric build-up of Be^7 is also indicated.

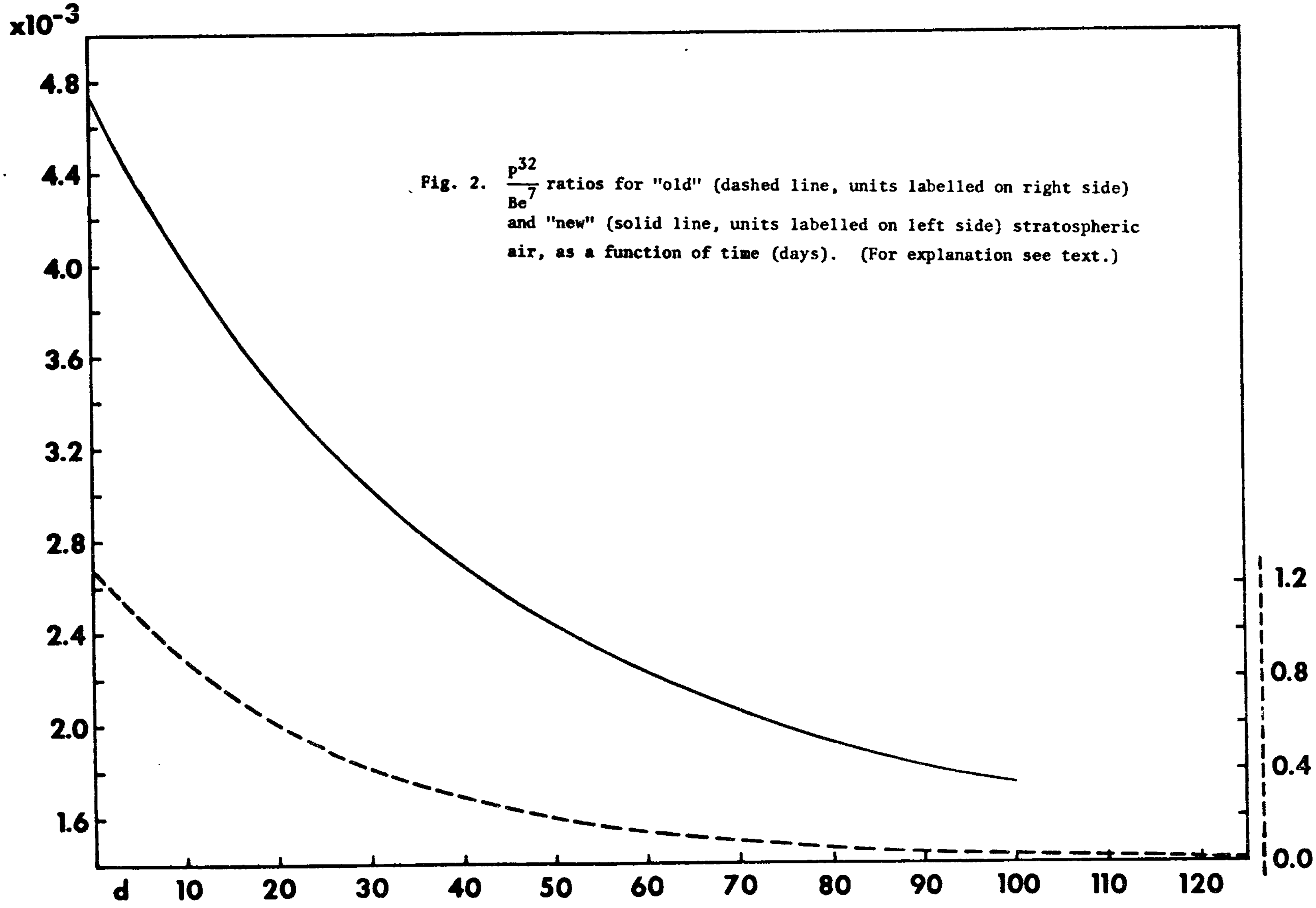

Fig. 2. $\frac{P^{32}}{Be^{7}}$ ratios for "old" (dashed line, units labelled on right side) and "new" (solid line, units labelled on left side) stratospheric air, as a function of time (days). (For explanation see text.)