Using Radioactive Tracers to Develop a Model of the Circulation of the Stratosphere

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ABSTRACT

Since 1952 a number of radioactive substances suitable for use as atmospheric tracers have been injected into the stratosphere. Information on large-scale stratospheric processes derived from measurements of strontium-90, carbon-14, tungsten-185, rhodium-102, cadmium-109 and plutonium-238 is summarized. Although the tracer data are too sparse to define an unambiguous model of the large-scale circulation features of the stratosphere, they should not be ignored in the process of constructing models from other considerations.

The tracer data indicate a summer-to-winter hemisphere flow above about 37 km and a mean descending motion in the winter stratosphere between 25° and about 70°. Ascending motion occurs near the equatorial tropopause and in the lower winter stratosphere poleward of 70°. Virtually the entire summer stratosphere and the winter stratosphere equatorward of 25° between 18 and 25 km is dominated by mixing processes with no evidence of organized circulations in the meridional plane.

1. Introduction

It is recognized that radioactive tracer measurements made to date in the atmosphere are far too incomplete to formulate an unambiguous kinematic model of the behavior of the stratosphere. This limitation will undoubtedly continue for some time. However, any model derived from other considerations must be consistent with the observed behavior of these tracers.

The ideal radioactive tracer experiments have yet to be made, if "ideal" is defined as a unique tracer injected into a precise location in sufficient quantity to be readily detected for several years, and measured virtually continuously on a global scale at all levels of the atmosphere and on the ground. Several such experiments involving different tracers injected into several atmospheric compartments and in all seasons would be needed to define the kinematics of the stratosphere, and even then some doubt would exist as to whether the radioactive material was a true tracer of the movements of the air molecules.

Of the radioactive tracers that have been injected into the stratosphere over the past two decades, six have proven to be most useful in shedding light on large-scale stratospheric behavior. Four are tracers with specific sources that can be to some extent isolated as to time and place of injection, and two are long-lived radioisotopes that are produced in all nuclear tests and have been extensively monitored.

The former include:

1) Tungsten-185 (W-185). This tracer with a half-life of 74 days was injected into the atmosphere in a series of nuclear tests conducted by the United States from April through July 1958 at 11 N in the Marshall Islands of the Pacific. The test series produced 204 MCI (megacuries) of W-185, corrected for radioactive decay to a common date of 15 August 1958 (Martell, 1968). Of this amount, about 60 MCI remained in the stratosphere for worldwide distribution (Friend et al., 1961), the remainder was contained in the larger particles associated with local and intermediate fallout. The bulk of the stratospheric material was initially injected at about 18–20 km. Aircraft up to 19 km (Friend et al., 1961; U. S. Government, 1968) and surface measurements (Lockhart et al., 1960, 1961) are available for about two years following the injections.

2) Rhodium-102 (Rh-102). Rhodium-102 was a special tracer created by neutron activation in a rocket-borne nuclear device detonated on 12 August 1958 at an altitude of 43 km near Johnston Island (17N). It is estimated that the resulting nuclear debris cloud rose to roughly 100 km. About 3 MCI of Rh-102 were produced and the isomer of principal concern has a half-life of 210 days. Some Rh-102 (about 0.3 MCI) was produced by other tests in 1958 (List et al., 1966). Limited balloon data (up to 28 km), aircraft observations (up to 19 km), and a few surface measurements are available. Additional information on the distribution of debris from the 1958 rocket tests were derived from the ratios of certain fission products in the samples (Telegadas and List, 1964).

3) Cadmium-109 (Cd-109). Cadmium-109 is also a neutron activation product produced in a rocket-borne nuclear device detonated on 9 July 1962 at 400 km. This isotope has a half-life of 470 days and the best
estimate is that 0.25 ± 0.15 MCi were produced. Although the detonation occurred at 17N (Johnston Island), the altitude was such that a substantial fraction of the ionized debris interacted with the geomagnetic field and was rapidly transported to the southern conjugate point (D’Arcy and Colgate, 1965). Considerable stratosphere data from balloon observations to 37 km, from aircraft to 20 km (List et al., 1966; Telegadas, 1968), and from surface measurements are available (Volchok and Kleinman, 1968).

4) Plutonium-238 (Pu-238). On April 1964, a navigational satellite containing an auxiliary power source with 17 kCi of Pu-238 reentered the atmosphere following a failure to attain orbit. The reentry was at about 11S, over the Indian Ocean (U. S. Atomic Energy Commission, 1968) and burn-up theoretically should have occurred somewhere between 40 and 60 km (Hansen et al., 1965). Pu-238 has a half-life of 86 years. There is a small background of Pu-238 in the atmosphere as a result of nuclear testing, but the ratio of background Pu-238 to Pu-239 is fairly constant and can be used to discriminate the satellite Pu-238 from the testing background. In all discussion to follow, Pu-238 will signify only that component resulting from the satellite reentry. More stratospheric balloon (to 37 km) and aircraft (to 20 km) data are available from this tracer than from any other. This, coupled with its relatively long half-life, has resulted in a more complete documentation of the behavior of Pu-238 in the stratosphere (Telegadas, 1968) than is available for any other specific tracer. Surface measurements are also available (Volchok and Kleinman).

Of the nonspecific tracers, i.e., those that have been produced in every nuclear test that injected debris into the stratosphere, two have proven to be of particular interest. These are:

1) Strontium-90 (Sr-90). This tracer is a fission product with a half-life of 28 years. Each megaton of fission yield creates about 0.1 MCI of Sr-90. Because of its health implications, there are extensive measuring programs to determine the Sr-90 content of the atmosphere (Telegadas, 1968).

2) Carbon-14 (C-14). Carbon-14 is both a naturally occurring cosmic-ray produced radionuclide and a product of the interaction of bomb-produced neutrons with atmospheric N2. Production of C-14 is essentially proportional to the total yield of the device from an air burst, for a surface burst up to half the neutrons produced will be absorbed by the surface and the amount of C-14 will be correspondingly reduced. Because of its long half-life, 5600 years, the naturally occurring component is assumed to be uniformly mixed throughout the atmosphere and the term C-14 will be used hereafter to designate the excess C-14 (i.e., above the natural background) that resulted from nuclear testing. Stratospheric balloon and aircraft programs have measured C-14 in the stratosphere since 1953 (Hagemann et al., 1965; U. S. Government, 1967; Telegadas and List, 1969).

All data are expressed in radioactivity concentration per mass of air, i.e., disintegrations per minute per 1000 standard cubic feet of air [dpm/(1000 SCF)−1].

2. What each tracer shows

a. Tungsten-185

The three-month average concentrations of W-185 in the stratosphere a half year and a year after the injections are shown in Figs. 1 and 2, respectively. In the

![Fig. 1. Tungsten-185 cross section, December 1958–February 1959. Isolines show average concentration [dpm/(1000 SCF)−1], decay corrected to 15 August 1958. Dots show location of data points. (Principal input occurred April–July 1958 at 11N, 18–20 km.)](image1)

![Fig. 2. Same as Fig. 1 except for June–August 1959.](image2)
tropical stratosphere the observed distributions of W-185 are consistent with an eddy mixing process as proposed by Feely and Spar (1960). However, poleward of about 25° latitude the picture is more complex. Tongues of higher concentration appear to extend poleward and downward from the equatorial maxima, giving rise to speculation concerning mixing along sloping surfaces. However, the genesis of these tongues is uncertain; adequate observational data are not available for the period during or shortly after the injections. Secondary maxima in the polar regions exist in both hemispheres but are more pronounced in the Northern Hemisphere. At the earlier time (Fig. 1) these maxima may be the result of short-term meteorological process bringing equatorial air rapidly poleward. In the Northern Hemisphere the possibility of a small contribution of W-185 from other sources cannot be ruled out. A year after input (Fig. 2) the observed pattern is remarkably similar in each hemisphere.

b. Rhodium-102

The 1958 input of Rh-102 provided the first experiment involving a unique high-altitude tracer. The downward and equatorward movement of this debris, using both Rh-102 and fission product ratios as tracers, is summarized in Fig. 3 (Telegadas and List, 1964). A mean descent of about 1.5 km month⁻¹, in the winter months, was indicated in the temperature latitude stratosphere down to about 15 km. In summer months the rate of downward propagation of the debris was <1 km month⁻¹. Although injected into the Northern Hemisphere, this tracer was seen in the Southern Hemisphere troposphere six months earlier than in the

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**Fig. 3.** Isochrones of the first appearance of Rh-102. Numbers show months after input (August 1958). Dots represent data points in the free atmosphere, crosses location of surface sampling. Spring and summer months are shaded.

**Fig. 4.** Cadmium-109 cross section, March–May 1963. Isolines show average concentration [dpm(1000 SCF)⁻¹], decay corrected to 9 July 1962. Dots show location of mean aircraft data, crosses location of individual balloon samples. (Input occurred 9 July 1962 at 17N, 400 km.)

**Fig. 5.** Same as Fig. 4 except for June–August 1963.
Northern Hemisphere troposphere, suggesting an initial movement of debris toward the winter hemisphere.

c. Cadmium-109

This tropical injection in 1962 confirmed the results suggested by the Rh-102 data. A few selected seasonal stratospheric cross sections from the complete series (Telegadas, 1968) illustrates this. Nine months after the input (Fig. 4) Cd-109 was above 25 km in both hemispheres, as shown by balloon observations at 31N and 34S. That the maxima were poleward of these stations was confirmed for the Northern Hemisphere three months later (Fig. 5), when soundings in the polar stratosphere first became available.

More complete Northern Hemisphere data in March–May 1964 (Fig. 6) bear this out. This figure illustrates the descent of the region of maximum concentration in the Southern Hemisphere and an apparent series of maxima in the Northern Hemisphere. Data to confirm the minimum concentration in the equatorial stratosphere first became available in September–November 1964 (Fig. 7). Note also the continuing descent of the Southern Hemisphere maximum. By June–August 1965 (Fig. 8), three years after the input, the primary stratospheric maxima in both hemispheres have descended to below 20 km, where they are well documented by aircraft observations. Isochrones of the first arrival of Cd-109 are shown in Fig. 9; note the similarity to the Rh-102 isochrones in Fig. 3. The relatively slow descent in spring and summer months (shaded areas) contrast with the more rapid downward progression in the fall and winter months in each hemisphere.

d. Plutonium-238

Despite the differences in the altitude and latitude of injection, Cd-109 and Pu-238 displayed remarkably similar concentration patterns in the stratosphere. The Pu-238 concentration pattern nine months after the input (Fig. 10) is virtually identical to the Cd-109 pat-
tern in Fig. 4. By September–November 1965 (Fig. 11) a maximum can be seen in the polar region of each hemisphere. The maximum is at a lower altitude in the

Southern Hemisphere, having experienced two winter seasons with the accompanying rapid descending motions vs only one winter season in the Northern Hemisphere. By June–August 1966 (Fig. 12) the maxima had descended to below 20 km. Isochrones of the first arrival of Pu-238 are shown in Fig. 13. These data also show the descent in the polar regions at the same rate as the
Rh-102 and Cd-109; about 1.5 km month\(^{-1}\) in the fall and winter and <1 km month\(^{-1}\) in the spring and summer months. The injection occurred near the start of the Southern Hemisphere winter and by the end of the winter debris had reached 27 km at 34S. It was not until the following Northern Hemisphere winter that debris was seen in that hemisphere and did not reach 27 km at 31N until the end of the first Northern Hemisphere winter.

Obviously, isochrones alone cannot specify the mode of downward propagation. However, the simultaneous downward propagation of the level of maximum concentration strongly suggests that vertical advection plays a dominant role in the fall and winter months.

The Pu-238 results from 34S are summarized in Fig. 14. Average quarterly concentrations at each altitude are shown from the time of injection through mid-1967. The altitude of the highest average concentration for each quarter is enclosed in a box; for the first year the boxes are open at the top, indicating that no data were available from higher altitudes to substantiate that this was indeed the altitude of highest concentration. The dashed line indicates the first appearance of traces of Pu-238 as deduced from the ratio of Pu-238 to Pu-239. The more rapid downward movement of the tracer in the fall and winter is clearly evident.

e. Strontium-90

The early models of stratospheric behavior based on radioactivity data were derived from observations of Sr-90, or total fission product activity, resulting from the first thermonuclear tests in 1952–54. Libby (1956) showed that the stratosphere could be considered a reservoir for the long-term storage of debris. Stewart et al. (1957) invoked the so-called Brewer-Dobson circulation, involving rising motion near the equatorial tropopause, poleward transport and ultimate descent in polar latitudes. Machta (1959) demonstrated the seasonal and latitudinal variability of fallout and related it

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**Fig. 13.** Isochrones of the first appearance of Pu-238. Numbers show months after input (April 1964). Dots represent data points in the free atmosphere, crosses location of surface sampling. Spring and summer months are shaded.

**Fig. 14.** Average seasonal Pu-238 concentration [dpm (1000 SCF)\(^{-1}\)] at Mildura, Australia (34S) as a function of time and altitude. The boxes enclosed the highest average concentration in each season. Dashed line indicates the first appearance of traces of Pu-238.
to preferred times and places for stratospheric-tropospheric exchange phenomena.

With regard to stratospheric circulation, many of the conclusions were derived from indirect evidence using measurements at aircraft altitudes and in the troposphere. It was not until October 1964 that reliable routine balloon observations became available in the equatorial stratosphere, extending our vista from the 20-km aircraft ceiling to the 30-km balloon altitude. The September–November 1964 Sr-90 cross section (Fig. 15) showed a concentration maximum in equatorial latitudes between 21 and 25 km. Six months later (Fig. 16), this maximum was even more pronounced; the observed 23-km equatorial concentration was 40% greater than any other value observed during this 3-month period. This equatorial maximum was a persistent feature of the stratosphere through March–May 1967 (Fig. 17), after which a fresh injection of significant amounts of Sr-90 into the north temperate stratosphere occurred.

Eleven megatons of fission were injected into the equatorial stratosphere by the 1962 United States tests (Salter, 1965), producing the Sr-90 maximum. That this region (approximately 18–23 km, 25N–25S) is not subject to mass exchange with the surrounding air can be seen not only by the maintenance Sr-90 of the maximum, but also by the fact that neither Cd-109 nor Pu-238 appeared in this area in as high concentrations as observed elsewhere in the stratosphere (see Figs. 8 and 12). It is concluded that this is a semi-stagnant region, with diffusive processes dominating.

f. Carbon-14

Carbon-14 exists in the atmosphere as CO₂, a gas, and, in principle, should be a better tracer of atmospheric motions than the other radionuclides discussed which are all particulate. However, except during and
shortly after periods of nuclear testing, the settling speed of Sr-90 relative to C-14 in the stratosphere below 30 km (the upper limit of reliable data) is negligible on time scales of a year or so. In addition to showing the validity of using particulate Sr-90 as a tracer for air motions in the stratosphere, Telegadas and List (1969) also showed that C-14 to Sr-90 ratios could be used to trace the flow of tropospheric air into the stratosphere. In the troposphere, precipitation scavenges the particulate Sr-90 relative to gaseous C-14, resulting in a much higher ratio. This tagged tropospheric air enters the lower tropical stratosphere during the Northern Hemisphere winter and moves poleward below 18 km, reaching 45N by late spring. Evidence of an upward movement of tropospheric air into the lower polar stratosphere was seen at the northernmost data points, about 70N.

3. Applicability of data

That the particulate tracer distributions are altered by atmospheric processes and are not the result of gravitational settling can be illustrated by the time history of the concentration of different particulate tracers at a fixed point. As an example, Fig. 18 shows the concentration of Sr-90, Cd-109 and Pu-238 at 27 km over San Angelo, Tex. (31N); the data are 3-month running means. Throughout the period shown, the Sr-90 gradient was upward and poleward. The Cd-109 gradient was downward and equatorward until the end of 1964, when the maximum at 31N was at 25 km. Clearly, the observed changes in concentration in Sr-90 and Cd-109 were out of phase until late 1964 and then became in phase. Similarly, throughout 1965 and most of 1966, the Pu-238 gradient was downward and equatorward, and again the changes in concentration of Pu-238 and Sr-90 were out of phase until the Pu-238 maximum reached the sampling altitude. Note that although the horizontal gradients of Pu-238 and Cd-109 were always directed oppositely from the Sr-90 gradient, it was when the vertical gradients were in the same direction that the out of phase relationship ended. This suggests, at 27 km and 31N, that vertical motions and/or vertical mixing are more evident in these data than horizontal advection or mixing. Similar conclusions can be drawn at other locations where adequate balloon data exist, 32 and 24 km over San Angelo and 27 and 24 km over Mildura, Australia (34S).

4. Discussion

A schematic representation of the stratospheric circulation features that can be deduced from radioactive tracer data is summarized in Fig. 19. There is evidence from Pu-238 and Rh-102 observations of a summer-to-winter hemisphere flow, as suggested by Kellogg and Schilling (1951), above the highest observational data at 37 km. Leovy (1964) has concluded from other evidence that such a thermally driven meridional circulation exists from 40° to at least 80 km. Virtually the entire summer stratosphere below 40 km and the portion of the winter stratosphere from the equator to 25° between 18 and 25 km is dominated by mixing processes, with no evidence of organized circulations in the meridional plane. Similar conclusions have been drawn by many investigations from momentum and heat transport considerations (e.g., Gudiksen et al., 1968; Vincent, 1968). In the lower equatorial stratosphere there is
evidence for ascending motion near the tropopause level and a poleward drift between the tropopause and 18 km. This drift is more pronounced in the winter hemisphere and extends to at least 40° latitude. The radioactivity data also indicate a strong mean descending motion (on the order of 0.06 cm sec⁻¹) from 25° to 70° latitude in the winter hemisphere extending from the upper limit of the observations to the lower stratosphere. Poleward of 70° there are no radioactivity data except in the very lowest stratosphere, and here an upward motion in winter is indicated by the C-14 to Sr-90 ratio.

The descending motion in the winter stratosphere from 25° to about 60° is in agreement with the models derived from meteorological data and theory (Gudiksen et al.; Vincent, 1968; Hunt and Manabe, 1968). However, poleward of 60°, these models, based on momentum and heat transport, show ascending motions. An inherent weakness in the tracer data is the difficulty in specifying unambiguously the motions that result in the observed changes in tracer distribution. In this case descending motion equatorward of 60° and subsequent horizontal mixing may have resulted in the observed distribution. However, the tracer data does demand a descending current somewhere in this region. The radioactive tracer information cannot be ignored in developing a model of the stratosphere.

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REFERENCES


