Radioactive debris as a tracer for investigating stratospheric motions

By ROBERT J. LIST, LEONARD P. SALTER[1] and KOSTA TELEGADAS, *U.S. Dept. of Commerce, Environmental Science Services Administration, Washington, D.C. (R.J.L. and K.T.) and U.S. Atomic Energy Commission Health and Safety Laboratory, New York (L.P.S.)*

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ABSTRACT

Two meteorologically interesting radioactive tracers, Cd-109 injected in 1962 at 400 km by a rocket-borne nuclear device and Pu-238 resulting from the reentry burnup in 1964 of a nuclear powered satellite in the 40–60 km altitude range, are examined and compared with the results of the Rh-102 experiment conducted in 1958. Although detonated at 17° N, the maximum concentration of Cd-109 was detected by the high-altitude balloon samples collected at 34° S. It first appeared at the highest altitude sampled, 32 km, and subsequently seemed to propagate downward at about 1.5 km per month. A second similar surge of Cd-109 was observed at 34° S about a year later. Maximum concentration of Pu-238 also occurred at 32 km at 34° S. Slightly lower concentrations were observed at this altitude in northern temperate and polar regions while concentrations near the equator were an order of magnitude lower. Although the Pu-238 distribution could result from either diffusive mixing along sloping surfaces or from meridional transport, the two surges of Cd-109 suggest poleward transport and subsequent reinforcement from a polar reservoir.

Introduction

Unique radioactive tracers injected into the upper atmosphere can yield significant information on the transport and diffusion processes within the stratosphere. The results of the tungsten-185 injection into the lower equatorial stratosphere during the summer of 1958 have shown the dominance of the eddy diffusion process in that portion of the stratosphere (FEELY & SPAR, 1960). There have been three suitable tracers injected to much higher altitudes (Table 1). The rhodium-102 results have been discussed by KALKSTEIN (1962) and by TELEGADAS & LIST (1964). The latter concluded that "debris injected above 43 km over the equatorial regions descends into the polar stratosphere and is subsequently propagated downward and equatorward. North of 35° N between 14 and 20 km the downward movement in winter months is of the order of 1.5 km per month, and it is suggested that mass movement rather than vertical diffusion is the dominant mechanism. Debris injected into the lower equatorial stratosphere [tungsten-185] appears to be governed by mixing phenomena to about 35° N, but no simple model serves to explain the distribution found over more northerly regions." The conclusions concern the Northern Hemisphere as adequate data in the Southern Hemisphere were unavailable.

Cadmium-109

On July 1962 a rocket experiment injected debris over Johnston Island, this time at 400 km. Although the detonation occurred at 17° N, the altitude was such that a substantial fraction of the ionized debris interacted with the geomagnetic field and was measured at the southern conjugate point (D'ARCY & COLGATE, 1965). The specific tracer for this debris has been estimated to be 0.25 ± 0.15 megacuries of Cd-109 (SALTER, 1964). The time history of the

[1] Mr. Salter's untimely death in a traffic accident in September 1965 was a tragic personal loss for all who knew him and an irreplaceable loss for the scientific community.

TABLE 1. *High-altitude tracers.*

Rh-102	Cd-109	Pu-238
Date		
Aug. 1958	July 1962	April 1964
Type		
Nucl. Test	Nucl. Test	Re-entry Burnup
Altitude		
43 km[a]	400 km	40–60 km
Latitude		
17° N	17° N	Indian Ocean
Source[b]		
3×10^6 curies	2.5×10^5 curies	1.7×10^4 curies
Half-Life		
210 days	410 days	86 years

[a] Cloud rose to ~100 km.
[b] Best estimate.

concentration of this debris in the stratosphere is given in Figs. 1–3 for the three latitudes for which sufficient data are available: 34° S, 31° N and 65° N. (Each number represents a single flight.) These measurements were made by the U.S. Atomic Energy Commission high-altitude balloon program (SALTER, 1965), supplemented by observations below 20 km with samples collected by Defense Atomic Support Agency aircraft at approximately the same latitude (KALKSTEIN *et al.*, 1965; FEELY, 1965). Data from a few additional balloon soundings at 9° N and 45° N are also entered on the latter two diagrams. All concentrations are expressed in units of 10^{-18} parts of the tracer produced by the device per 35 kg (1000 standard cubic feet) of air. Values have been rounded to the nearest 10 units, except for concentrations near the limits of detection. In general, this limit was about 4 or 5 units for the samples shown here. A "5" is shown when Cd-109 was definitely detected, a "0" when the concentration was below the limit of reliable detection.

The Cd-109 data from 34° S (Mildura, Australia, balloon station and below 20 km aircraft from 30 to 50° S) are shown in Fig. 1. The dashed lines delineate the first appearance of Cd-109, the shaded area indicates concentrations of more than 40×10^{-18} parts of the device per 35 kg of air. Cd-109 first appeared at the highest altitude sampled, 32 km, about 5 months after injection. Although the relatively high concentration in December 1962 implies that it may have been present somewhat earlier, no observations are available above 30 km between September and December. The Cd-109 did not reach the 20 km level until a year after the injection. The apparent rate of descent, 12 km in 7 to 9 months, is very similar to the rate at which the Rh-102 descended in north temperate latitudes between somewhat lower altitudes. Relatively high concentrations (shaded area) appeared at 32 km during the summer and fall months (December through April) and by

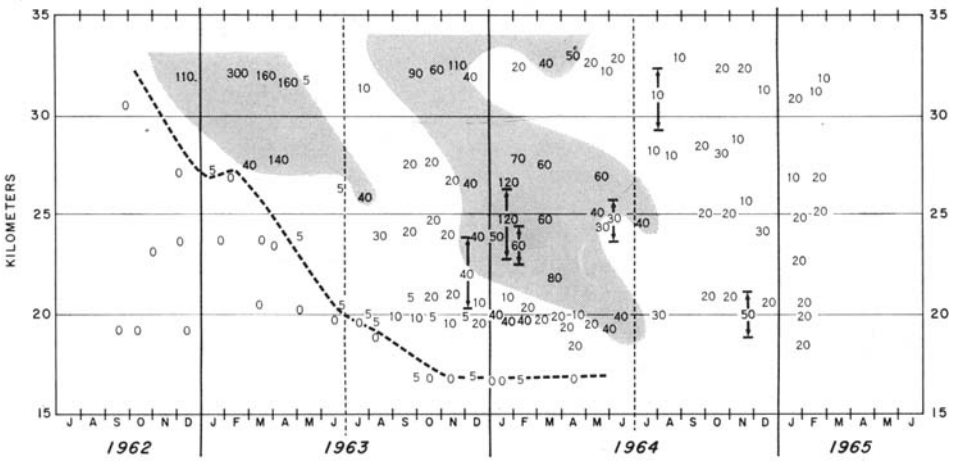

FIG. 1. Cd-109 concentrations at 34° S (Mildura, Australia, balloon station), supplemented below 20 km by aircraft data collected between 30 and 50° S. Units are 10^{-18} parts of the device per 35 kg of air. The dashed line delineates the first appearance of detectable amounts of Cd-109. Shading indicates concentrations of 40 units or more.

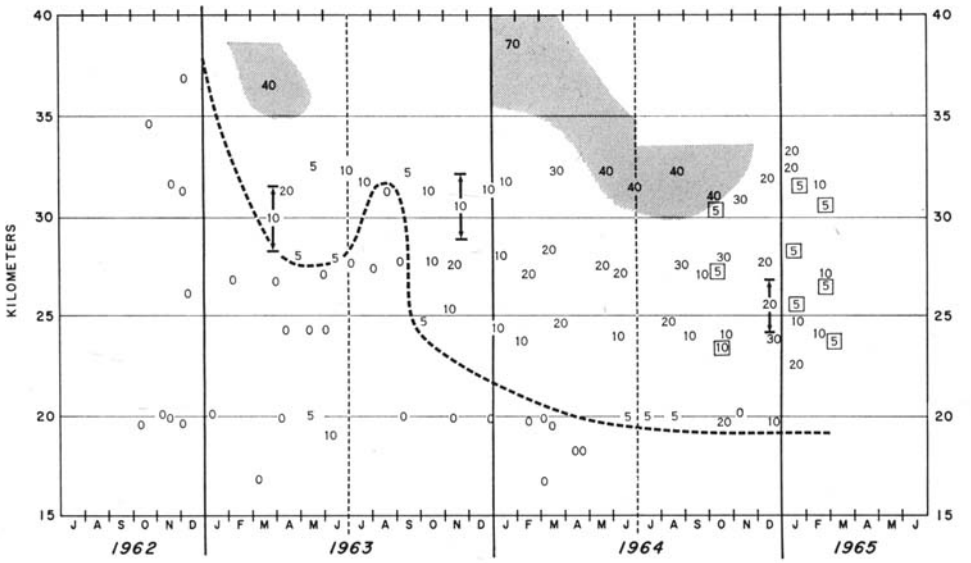

FIG. 2. Cd-109 concentrations at 31° N (San Angelo, Texas, balloon station), supplemented below 20 km by aircraft data collected between 30 and 40° N. Data in squares are from 9° N (Canal Zone balloon station). Units are 10^{-18} parts of the device per 35 kg of air. The dashed line delineates the first appearance of detectable amounts of Cd-109. Shading indicates concentrations of 40 units or more.

March had reached down to 28 km. The concentrations at the highest altitude decreased markedly in the Southern Hemisphere winter months but by spring (September) another surge of air containing higher concentrations of Cd-109 appeared at 32 km and was subsequently seen as low as 22 km. From the data available it is not possible to determine whether an actual mass movement downward was occurring or whether horizontal advection, beginning at the higher level and progressing to successively lower levels, brought material in from a polar reservoir. The apparent timing of the two surges at the highest levels suggest that advection from a polar reservoir after the early spring (September–October) period of explosive warming may play a role. Also noteworthy is the uniformity of the Cd-109 concentrations at all altitudes in the stratosphere at 34° S after 2 years from input: from mid-1964 on, most of the observable stratosphere is characterized by a Cd-109 concentration of about 20×10^{-18} parts of the device per 35 kg of air.

Fig. 2 shows the data for 31° N (San Angelo, Texas, balloon station and aircraft below 20 km from 30° to 40° N), also shown on this figure are the few data available from 9° N (Canal Zone balloon station). It is evident that despite the fact that the injection occurred in the Northern Hemisphere, the concentrations and gradients of Cd-109 observed at 31° N are smaller than at 34° S. The highest concentration at 32 km, 40×10^{-18} parts of the device per 35 kg of air, occurred approximately two years after the injection as contrasted to a value of 300 only 8 months after the injection at 34° S at the same altitude. Cd-109 did not reach the 20 km level until two years after the injection at 31° N, about twice the time it took to appear at this level at 34° S. However, by the end of 1964, concentrations and gradients are similar to those over Mildura, a relatively uniform value of about 20×10^{-18} parts of the device per 35 kg of air. Note that at 9° N, the few data available during late 1964 and early 1965 invariably indicate a concentration equal to or less than that at 31° N or 34° S.

Fig. 3 shows the data from 65° N (Fairbanks, Alaska, balloon station and aircraft below 20 km between 50° and 70° N) and the single 32 km balloon observation at 45° N. The concentrations are higher than at 31° N in July 1963 and Cd-109 reached the 20 km level about 5 months earlier. This again suggests a polar rather than an equatorial maximum of Cd-109.

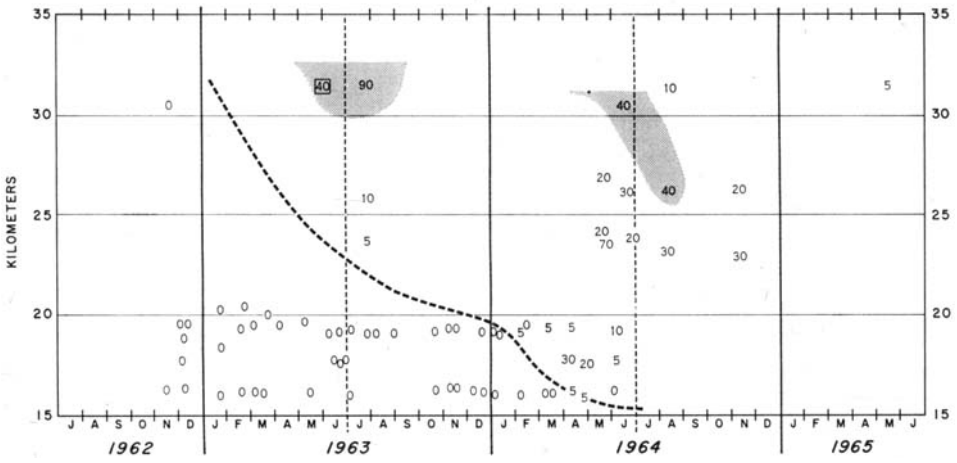

FIG. 3. Cd-109 concentrations at 65° N (Fairbanks, Alaska, balloon station), supplemented below 20 km by aircraft data collected between 50 and 70° N. Datum in square is from 45° N. Units are 10^{-18} parts of the device per 35 kg of air. The dashed line delineates the first appearance of Cd-109. Shading indicates concentration of 40 units or more.

A comparison can be made between the estimated Cd-109 input and the stratospheric content of Cd-109 in October 1964, 27 months after production. If the stratosphere is assumed to contain 15% of the atmosphere, a uniform distribution of the total device in the stratosphere would result in a concentration of 45×10^{-18} parts of the device per 35 kg. The observed concentration at this time, generally about 20×10^{-18} parts of the device per 35 kg, suggests that about one-half of the estimated total Cd-109 produced was in the stratosphere. If the "best estimate" of the source strength is correct, the discrepancies can be ascribed to underestimating the amount in the unmeasured portion of the stratosphere. It is also possible that by this time a significant fraction of the Cd-109 had entered the troposphere.

Plutonium-238

A different type of injection resulted from the re-entry burnup south of the Equator over the Indian Ocean off the coast of Africa in April 1964 of 17 kilocuries of Pu-238 contained in a SNAP-9A auxiliary power source. The burnup theoretically should have occurred somewhere between 40 and 60 km (HANSEN *et al.*, 1965). The resulting debris cloud received no explosive thrust or upward movement such as a nuclear detonation would produce.

The Pu-238 data have been obtained by the U.S. AEC high altitude balloon program and are presented in Figs. 4–7. (Each number represents a single flight.) The concentrations are again expressed in units of 10^{-18} parts of the tracer contained in the device per 35 kg of air for intercomparison with the other tracers discussed. There is a small background concentration of Pu-238 in the atmosphere as a result of nuclear testing. In the units used here, the background of Pu-238 is less than 5. The ratio of background Pu-238 to Pu-239 is fairly uniform and therefore can be used to discriminate the SNAP-9A Pu-238 from the testing background.

Fig. 4 shows the Pu-238 concentration as a function of altitude and time at 34° S; Fig. 5 shows the data at 9° N. Although the source was located between these two latitudes it appears that most of the Pu-238 moved southward. At 34° S it appeared at 32 km about 4 months after injection and concentrations of about 600×10^{-18} parts of the device per 35 kg of air were observed within 6 months.

At 9° N, no Pu-238 was found in the observation made above 30 km in October 1964. Concentrations at 32 km are more than an order of magnitude smaller than at 34° S in the first several months of 1965.

Fig. 6 shows the data at 31°; Fig. 7 at 45° and 65° N. In contrast to the very small con-

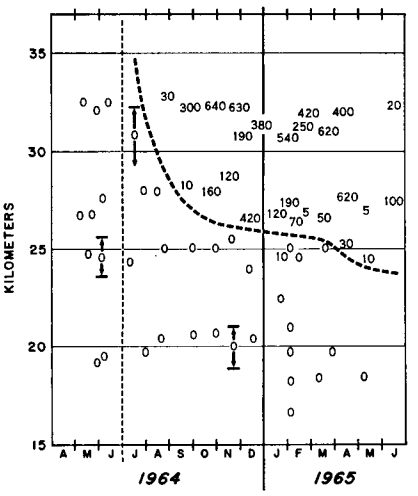

Fig. 4

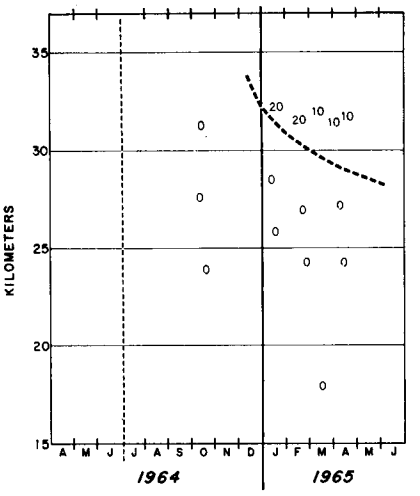

Fig. 5

FIG. 4. Pu-238 concentrations at 34° S (10^{-18} parts of the device per 35 kg of air). The dashed line delineates the first appearance of Pu-238.

FIG. 5. Pu-238 concentrations at 9° N (10^{-18} parts of the device per 35 kg of air). The dashed line delineates the first appearance of Pu-238.

centrations at the Canal Zone nine months after the burnup, concentration of Pu-238 of the order of 100×10^{-18} parts of the device per 35 kg of air were observed at 31° N at 32 km. A few months later it also appeared at 28 km. Farther poleward, the limited data in Fig. 7 show similar or higher concentrations twelve months after injection than at 31° N. Again, it seems necessary to postulate a transport mechanism toward the poles in each hemisphere,

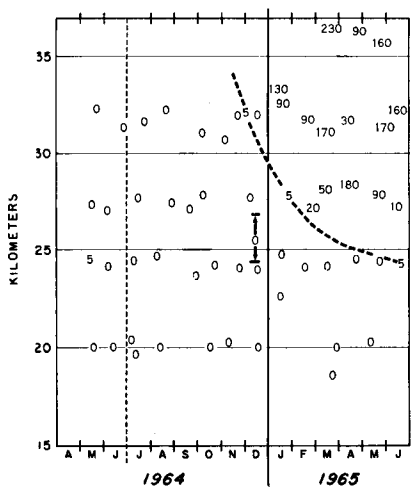

Fig. 6

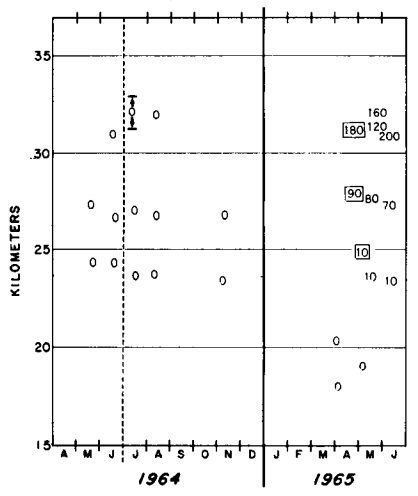

Fig. 7

FIG. 6. Pu-238 concentrations at 31° N (10^{-18} parts of the device per 35 kg of air). The dashed line delineates the first appearance of Pu-238.

FIG. 7. Pu-238 concentrations at 65° N, data in squares from 45° N (10^{-18} parts of the device per 35 kg of air).

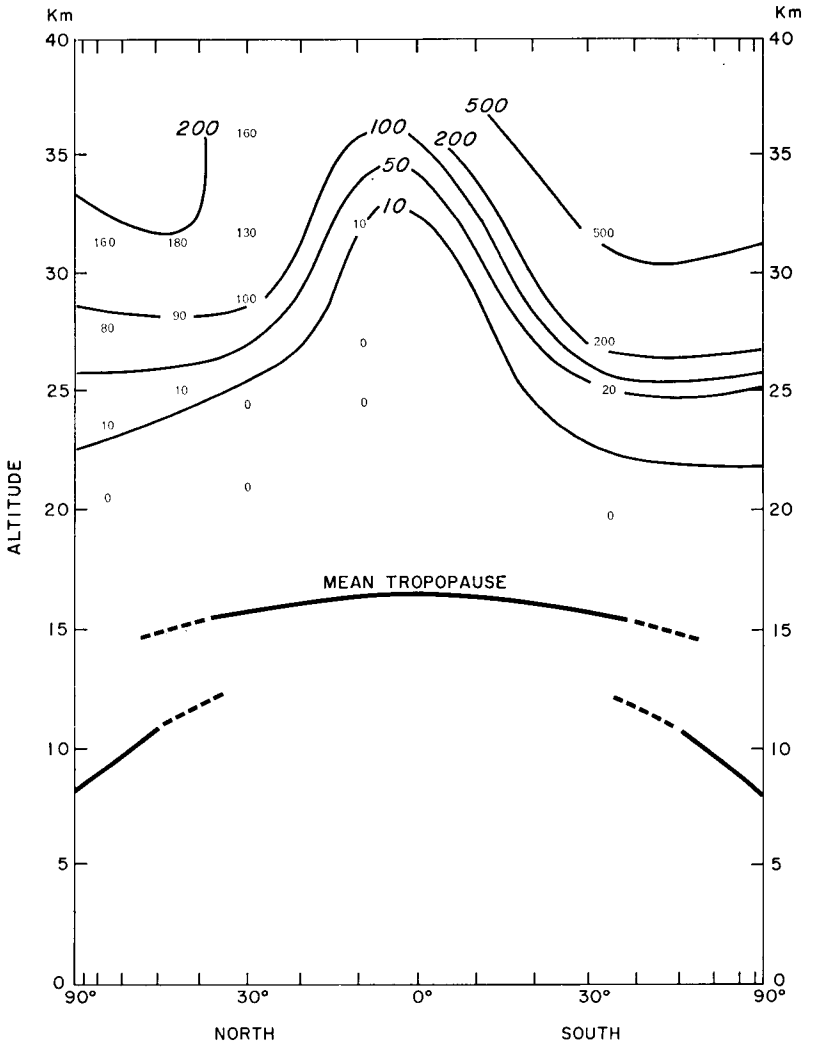

FIG. 8. Distribution of Pu-238 in the atmosphere one year after input (10^{-18} parts of the device per 35 kg of air).

with material crossing the equator at elevations in excess of the highest point sampled, 34 km.

Since observations were made at all four sampling sites at about 12 months after input, there are sufficient data to prepare a global cross-section of the distribution of Pu-238 in the stratosphere. The average concentrations are shown in Fig. 8, together with an estimate of the distribution of Pu-238 in the unmeasured parts of the atmosphere. Integration of the pattern indicates that more than half of the 17 kilocuries of Pu-238 was present in the stratosphere between 22 and 36 km at that time and none was below 22 km. Although the uncertainties in this estimate are large, it does suggest that burnup was complete and that the resulting particles were so small as to have negligible fall rates. These conditions were assumed by HARLEY (1964) and by MACHTA (1965) in their successful predictions of the concentration of Pu-238 to be expected following the SNAP-9A burnup.

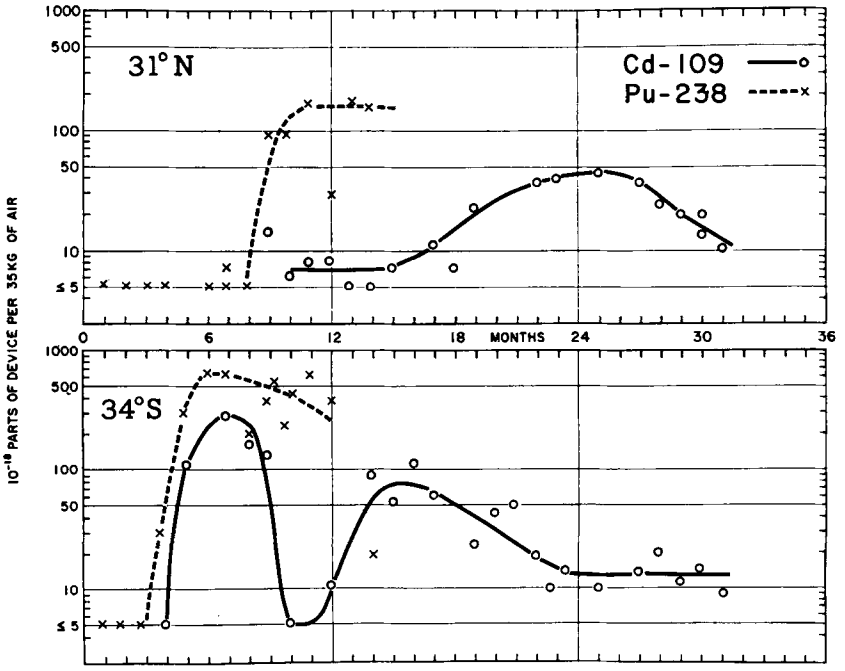

FIG. 9. Concentration of Cd-109 and Pu-238 in 30–33 km layer as a function of time after input.

Comparison of time history: rhodium-102, cadmium-109 and plutonium-238

It is interesting to compare the time history of the concentration of Rh-102, Cd-109 and Pu-238 at specific altitudes at 31° N and 34° S as a function of time since injection. Fig. 9 shows the data for Cd-109 and Pu-238 at 30–33 km, (no Rh-102 measurements were made at this altitude), the upper part for 31° N, the lower part for 34° S. Despite the large differences in the altitude and latitude of injection, at 34° S both the Cd-109 and the Pu-238 concentration reached peak values of the same order of magnitude seven months after the event. The two surges of Cd-109 and the subsequent leveling-off to a value of about 20 units at 34° S were discussed earlier. At 31° N, it is evident that both the Cd-109 and Pu-238 behaved somewhat differently. The first appearance of the plutonium was delayed by about a half-year over that observed at 34° S and the cadmium came still later, the major increase occurring about a year and a half after injection. After the second year following the injection, the concentrations of Cd-109 at 31° N and 34° S were about equal.

At 26–29 km (Fig. 10), the results were similar. At this altitude Rh-102 data were available from 31° N, beginning about 2 years after the injection. Note that this tracer, too, appears in virtually the identical normalized concentration as the Cd-109, about 20×10^{-18} parts of the device per 35 kg of air.

At the lowest level with adequate data, 19–21 km (Fig. 11), the increase in Cd-109 concentration occurred about a year earlier at 34° S than that at 31° N. Rh-102 at 31° N, however, began to increase six months sooner relative to the time of injection than did the Cd-109. At 34° S the Rh-102 data was inadequate to determine the time of arrival. Again at this altitude, after about two years following the injection, both rhodium and cadmium had the same concentration at each of the two observing points, about 20 units. This similarity lends credence to the "best estimates" of the source strengths of the two devices given in Table 1. If the best estimates are in error, it would appear that the errors are of the same sense in each.

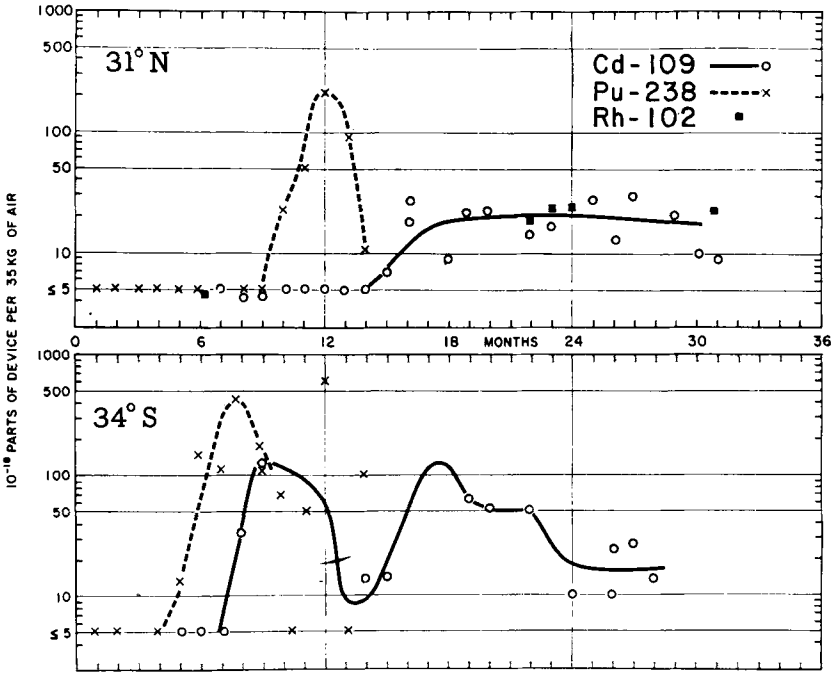

FIG. 10. Concentration of Cd-109, Pu-238 and Rh-102 in the 26–29 km layer as a function of time after input.

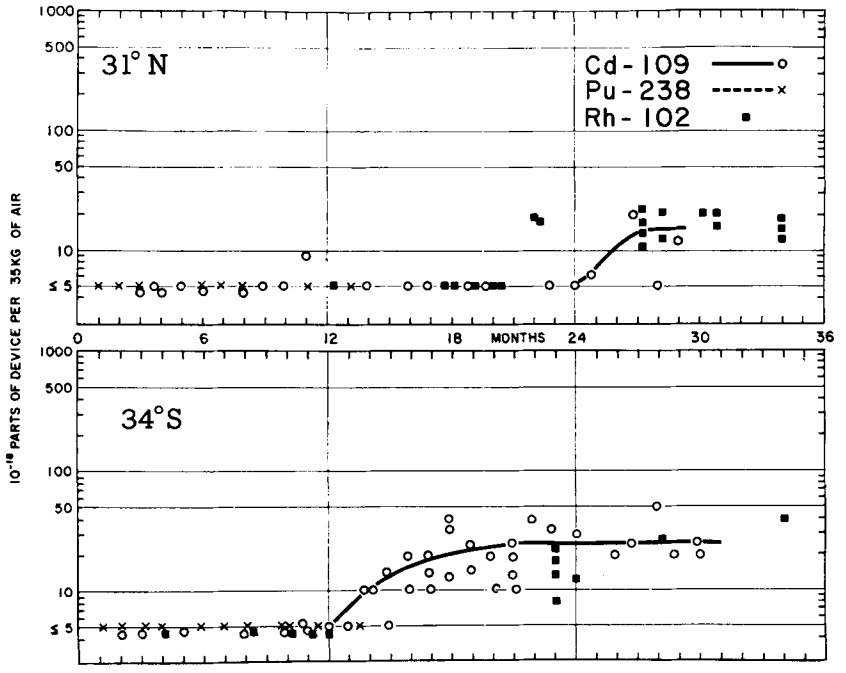

FIG. 11. Concentration of Cd-109, Pu-238 and Rh-102 in the 19–21 km layer as a function of time after input.

Conclusions

It is evident from the data presented that the meteorology of the upper stratosphere is not readily depicted by simple models. Both diffusive processes and transport phenomena seem to play important roles. Year-to-year differences in the atmosphere may also be responsible for the difference in behavior of the several tracers discussed. Another uncertainty lies in the assumption that a single station is representative of an entire latitude band.

There are unfortunate gaps in the data that tend to restrict some of the comparisons one would like to make. There is nothing in the Cd-109 data which would contradict the interpretation of the Rh-102 data in the Northern Hemisphere (TELEGADAS & LIST, 1964), but comparable data exist only at 31° N. Here, both behave similarly in the 22–30 km region although at 20 km the Rh-102 appeared relatively earlier than the Cd-109.

Somewhat surprising, is the high concentration of Cd-109 found at 34° S. This may be a result of the season, the relative latitude of the observing location (i.e., 34° S may be less "tropical" than 31° N) or the initial behavior of the particles resulting from a detonation at 400 km. The two surges of debris at 34° S about a year apart suggest that simple eddy diffusion can not be the major transport mechanism.

Although one would expect the largest Pu-238 concentration in the Southern Hemisphere, there is virtually none of this material at 9° N up to 32 km and a relatively high concentration as low as 27 km at 31° and 65° N. If diffusion along sloping surfaces is the appropriate mechanism, the slope of these surfaces near 30 km in the sub-tropics must be of the order of 5 km in less than 30° of latitude. The tungsten experiment at about 20 km indicates a slope of 2 to 3 km in 30° latitude (FEELY & SPAR, 1960). Employing the numerical model developed by MACHTA (1966) for one-dimensional horizontal diffusion on a spherical earth and confining the Pu-238 to a thin, quasi-spherical shell, a diffusion coefficient of 10^9 cm^2 sec^{-1} would produce the observed concentrations at 31° N and 34° S about a year after injection.

Both the Cd-109 and the Pu-238 data in the Southern Hemisphere at 34° S suggest the possibility that air from higher altitudes descends into the layers below approximately 30 km in the late winter and early spring (August–September) and subsequently is transported downward. The rate of downward transport seems to be similar to that found at more temperate latitudes in the Northern Hemisphere from the Rh-102 experiment, about 1.5 km per month. Data from a single observing point makes it difficult to differentiate vertical transport from mixing or horizontal advection.

Acknowledgement

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ИСПОЛЬЗОВАНИЕ РАДИОАКТИВНЫХ ОСТАТКОВ КАК ТРАССЕРОВ ПРИ ИССЛЕДОВАНИИ СТРАТОСФЕРНЫХ ДВИЖЕНИЙ

Исследовано два радиоактивных трассёра, представляющих метереологический интерес: Cd-109 инжектированный в 1962 г. на высоте 400 км с помощью ядерного механизма, помещенного на ракете и Pu-238 образовавшегося на высоте 40–60 км в 1964 г. в результате сгорания при входе в атмосферу спутника с ядерным двигателем. Эти результаты сравниваются с данными экспериментов в Rh-102 выполненных в 1958 г. Хотя инжекция произошла на 17° с. ш., максимум концентрации Cd-109 был определен в пробах взятых высотными шарами при 34° ю. ш. Сначала этот максимум появился в пробах с больших высот (32 км), и в последствии распространялся вниз, примерно, на 1,5 км в месяц. Второй подобный максимум Cd-109 наблюдался на этой же широте примерно через год. Максимум концентрации Pu-238 также имел место при 34° ю. ш. на высоте 32 км. Наблюдавшиеся на этой высоте концентрации были в северных умеренных и полярных областях незначительно меньше, а в экваториальнных областях меньше на порядок. В то время как распределение Pu-238 может быть результатом либо диффузионного перемешивания вдоль наклонных поверхностей, либо меридионального переноса, то наблюдавшиеся две волны в концентрации Cd-109 указывают на перенос в сторону полюса и последующую поддершку из полярного бассейна.