Transport and fallout of stratospheric radioactive debris

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ABSTRACT

The average stratospheric residence half-time of particulate radioactive debris, as represented by strontium-90 and manganese-54, was approximately 10 months during the two years following the last atmospheric tests of high yield nuclear weapons. During this interval, the carbon-14 which was produced artificially by nuclear weapons tests showed a stratospheric residence half-time which increased with time, but averaged about 18 months. It is suggested that the stratospheric residence half-time of particulate debris did not increase significantly during 1963 until early 1965 because in the upper stratosphere, above 20 km, the rate of particle settling of the debris exceeded the rate of upward diffusion, and as a result, this debris remained highly concentrated in the lower stratosphere throughout this period. It is also suggested that the relatively more rapid meridional movement of particulate debris, compared to the movement of carbon-14, into the stratosphere of the Southern Hemisphere during the second half of 1963 resulted to some extent from the partial separation of the particulate debris from the carbon-14 in the equatorial stratosphere as a result of particle settling.

1. Introduction

The U.S. Defense Atomic Support Agency has sponsored the sampling of the lower stratosphere for radioactive fallout from nuclear weapons tests since 1957 utilizing WU-2 aircraft and, recently, RB-57F aircraft. The current phase of this program is code-named Project Stardust.

The Stardust sampling missions collect filter samples of stratospheric air using 1PC-1478 filter medium and, since August, 1963, they also collect gas samples. At Isotopes, Inc., carbon dioxide from the gas samples has been analyzed for carbon-14; the filter samples have been analyzed for fission products, such as strontium-90 and cerium-144, for products of neutron activation, such as manganese-54 and cadmium-109, and for naturally produced radionuclides, such as beryllium-7, phosphorus-32, and lead-210.

The purpose of the program has been twofold: (1) to monitor the stratospheric burdens of strontium-90, carbon-14, and other radioactive debris, and (2) to obtain evidence on the nature and rates of the processes which produce movement of radioactive debris in the stratosphere and between the stratosphere and troposphere.

Sampling is currently performed between 75°N and 55°S. The flight tracks followed during 1965 are illustrated on the map in Figure 1, and on the vertical cross-section of the atmosphere in Figure 2. The aircraft capabilities limit Stardust sampling to the lower stratosphere, below 22 km. Vertical arrows in Figure 2 represent the approximate sampling positions for the U.S. Atomic Energy Commission high altitude balloon sampling program (SALTER, 1965 A). Data from the balloon program have been used to extrapolate Stardust data concerning particulate debris to the upper stratosphere.

The radiochemical procedures employed in Project Stardust and the calibration of the filter samples used in the program have been described in reports issued by the U.S. Defense Atomic Support Agency (FRIEND *et al.*, 1961; FEELY *et al.*, 1963). It has been shown that the filter sampler used in the program has essentially a 100 per cent collection efficiency for the

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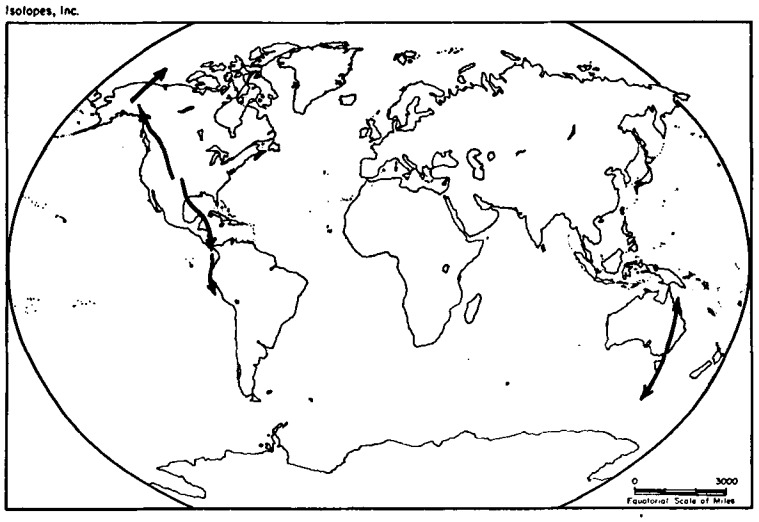

FIG. 1. Flight tracks of Stardust missions during early 1965.

particles carrying stratospheric radioactivity (VAN DEN AKKER, 1960; FRIEND *et al.*, 1961). It has been estimated that errors in sampler intercalibration, in recording of collection data (such as actual altitude and airspeed of aircraft during filter exposure, and actual duration of exposure), and in sample analysis may give a random error of ± 20 per cent in the final results for a typical Stardust filter sample analysis.

The same types of possible errors in sample collection and analysis may be expected to give a random error of about ± 20 per cent in results of radiochemical analyses of filter samples collected during the balloon program. If the filter sampler used in the balloon program has less than a 100 per cent efficiency for the collection of the particles which carry stratospheric radioactivity, however, the nuclide concentrations reported for the balloon samples may be too low.

The filter sampler used since 1961 for the collection of samples for the balloon program

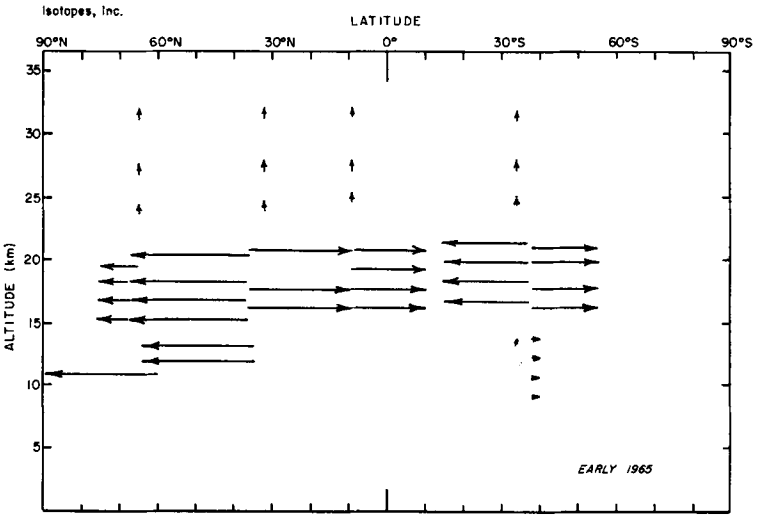

FIG. 2. Sampling corridor with Stardust and AEC balloon program flight tracks.

has been described by WOOD (1961; 1964). From the data presented by Wood, it appears that this sampler has collection efficiencies in excess of 75 per cent for particles with diameters greater than 0.026 micron. This size range should include the bulk of the stratospheric aerosol, and, probably, the bulk of the stratospheric debris from nuclear weapons tests. DREVINSKY & PECCI (1965) attempted to measure the size distribution of stratospheric particles containing fission products, and found that with few exceptions most of the activity is carried by particles of about 0.04 to 0.30 micron diameter. If the results of these measurements are correct, it can be concluded that any bias in the balloon data due to incomplete collection of the stratospheric particles is less than 33 per cent.

We have attempted to estimate the uncertainties in the calculated total stratospheric burdens of strontium-90, manganese-54 and carbon-14 which are given below, taking into account the possible errors in sampler calibrations, in collection data, and in sample analysis, and the probable representativeness of the sample collections. Generally 80 per cent or more of the total stratospheric burdens of strontium-90 and manganese-54 has been found to be in the lower stratosphere, where it has been sampled by Stardust aircraft. Thus even if the assumed collection efficiencies of the balloon filter samplers are too high, and the resulting concentration data are low by as much as 33 per cent, the calculated stratospheric burdens will be in error by less than 7 per cent. If the balloon sampler collection efficiencies are essentially 100 per cent, of course, no error will be introduced into the burden calculations from this source. The random errors resulting from errors in sampler intercalibration, in collection data, and in sample analysis will tend to cancel each other, so that if enough sample measurements are used in calculating the stratospheric burdens, the resulting net error should generally be considerably less than ± 10 per cent. An additional random error will be introduced into the calculated burden if the samples collected during a particular time interval are not truly representative of the radioactivity concentrations in the whole stratosphere. This danger is most pronounced during and immediately following periods of atmospheric testing of high yield weapons, such as much of 1962 and the first few months of 1963. This danger is also present during periods of especially sparse sampling. Considering all of these factors we would estimate that the uncertainty in the calculated stratospheric burdens of strontium-90 and manganese-54 is generally about ± 20 to ± 25 per cent. The collection and analysis of carbon-14 samples are subject to relatively small errors, and the main uncertainty in the calculated stratospheric burdens of this nuclide probably arises from questions concerning the representativeness of the sampling. We would expect this uncertainty to be less than ± 20 per cent.

Almost all of the gas samples analyzed for carbon-14 during Project Stardust have been collected between 75° N and 15° N and between 13 and 22 km. We have also used data from Argonne National Laboratory published recently (HAGEMANN *et al.*, 1965; Health and Safety Laboratory, 1966) to estimate carbon-14 distributions in the regions above 22 km., below 13 km., and south of 15° N. The results of measurements of carbon-14 in ground-level air at Westwood, New Jersey, and at Wellington, New Zealand (*Rafter*, 1965), have also been used. The data of Hagemann *et al.* have been used to estimate carbon-14 distributions in all stratospheric regions before August, 1965, when Stardust gas sampling began.

On the basis of the results of measurements of stratospheric radioactivity made during recent years, several conclusions can be made concerning the stratospheric residence time of radioactive debris and the movement of such debris from one stratospheric region to another.

2. The stratospheric residence time of radioactive debris

The stratospheric burdens of strontium-90, manganese-54, and carbon-14 have been calculated from the observed distributions of these nuclides for a series of time periods during 1963 to 1965. The stratospheric residence times of these nuclides during these years have been determined from the rates of decrease of the stratospheric burdens.

The apparent distributions of strontium-90 in the stratosphere during January–April, 1963, 1964, and 1965 are shown in Figure 3. The apparent distributions of manganese-54 during the same periods, with concentrations corrected

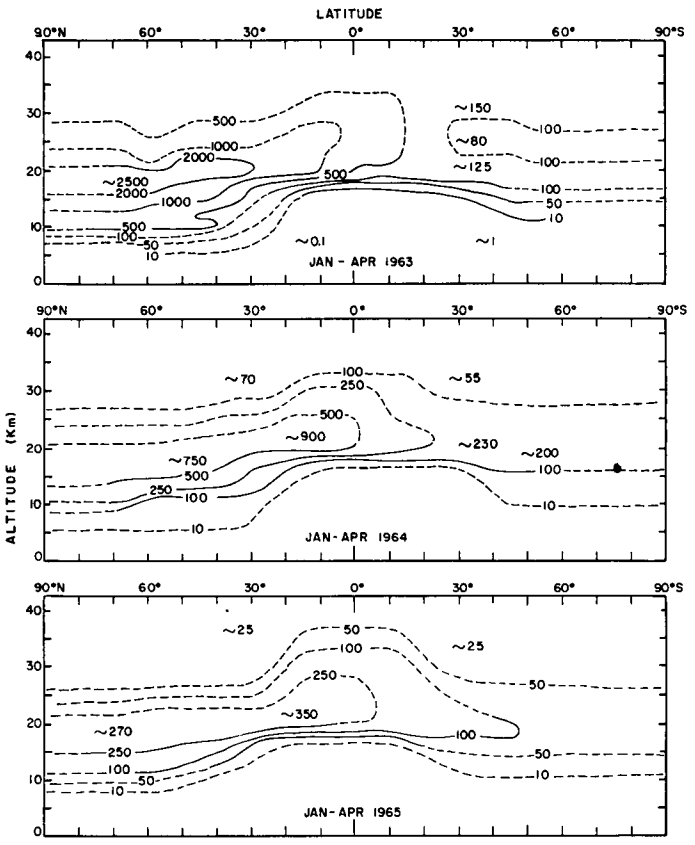

FIG. 3. Distribution of strontium-90 in the atmosphere (dpm Sr^{90}/1000 SCF).

for radioactive decay to 31 December, 1962, are shown in Figure 4. The period January–April 1963 immediately followed the last atmospheric tests of high yield nuclear weapons, which were performed in December, 1962.

The stratospheric burdens of strontium-90 and manganese-54, calculated from apparent distributions such as those shown in Figures 3 and 4, are plotted as a function of time in Figure 5. The decrease with time in the burdens of both nuclides during 1963 and 1964 corresponds to a stratospheric residence half-time of 10 months for this particulate radioactive debris.

The apparent distributions of carbon-14 in the stratosphere during January–April 1963, 1964, and 1965 are shown in Figure 6. The distribution for January–April, 1963 is based almost entirely on the data of HAGEMANN *et al.* (1965).

We have used apparent distributions such as those shown in Figure 6 to calculate the carbon-14 burdens of the stratosphere for a series of periods during 1963 to 1965. The calculated burdens are plotted as a function of time in Figure 7. The data indicate that during 1963–1964, this gaseous radioactive debris had an apparent residence half-time of about 16 months in the stratosphere of the Northern Hemisphere, and about 18 months in the entire stratosphere. Of course, part of the decrease in the burden of the Northern Hemisphere stratosphere resulted from movement of carbon-14 into the stratosphere of the Southern Hemisphere. Because of the relatively slow removal of carbon-14 from the troposphere, moreover, the tropospheric burden of this nuclide was increasing during 1963 to 1965. It should be expected that the build-up of tropospheric concentrations would result in a

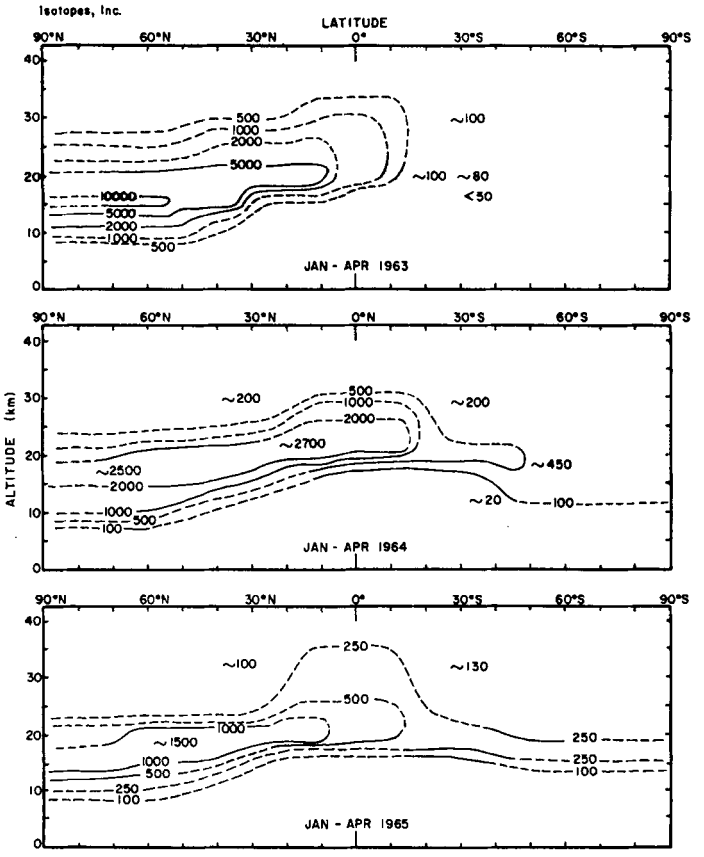

FIG. 4. Distribution of manganese-54 in the atmosphere (dpm Mn^{54}/1000 SCF corrected to 31 December, 1962).

gradual lengthening of the residence half-time of carbon-14 in the stratosphere. This phenomenon may be distinguished in the data for the total stratosphere in Figure 7, with the effective residence half-time increasing from about 15 months during 1963 to about 36 months by the end of 1964.

3. The distribution of radioactive debris in the stratosphere

It is evident from Figures 3, 4, and 6 that in early 1963, only a few months after the last atmospheric test of a high yield weapon in late December, 1962, the highest concentrations of radioactive debris, both particulate and gaseous, were present in the lower stratosphere of the Northern Hemisphere. The concentrations in the upper stratosphere, at about 30 km, were factors of 5 to 10 lower. The concentrations in the lower stratosphere of the Southern Hemisphere were factors of 10 to 100 lower. Of course, most of the radioactive debris in the lower stratosphere of the Northern Hemisphere, and much of that in the other stratospheric regions, was injected during the 1961–1962 weapons test series; the stratospheric burdens had increased between mid-1961 and early 1963 from less than 1 to more than 6 megacuries of strontium-90, from less than 2 to over 20 megacuries of manganese-54, and from about 10×10^{27} to over 50×10^{27} excess atoms of carbon-14. It is noteworthy that so much of the debris, even from the very high yield devices tested by the U.S.S.R. during 1961 and 1962, was injected into the lower stratosphere. This lends additional weight to the conclusions reached in the past (FRIEND *et al.*, 1961;

FEELY *et al.*, 1963) that very little debris from high-yield weapons tested at high latitudes before 1961 was injected into the stratospheric regions above 20 km, and that relatively little debris, even from the very high yield 1961 tests, was injected above that level.

The possibility has been discussed (MACHTA *et al.*, 1963) that a large fraction of the debris from the 1961 and 1962 U.S.S.R. weapons tests was injected above the highest balloon sampling altitudes (32 km), and was still there in mid-1963. It seems unlikely that such was the case, for measurements of balloon samples collected in the Northern Hemisphere in the stratospheric layer between 24 and 32 km during mid-1963 to mid-1965 show a rather rapid, continuing decrease in the concentrations of strontium-90 and other long-lived components of radioactive debris in this region during this period. So rapid a decrease would not be expected if large quantities of debris were reentering this region from higher atmospheric layers during this period. Moreover, debris from the very high yield U.S.S.R. events of October, 1961 appears to have contained a high ratio of certain products of neutron activation, such as manganese-54 and iron-55, to fission products, such as strontium-90 (*Feely* & BAZAN, 1965). This debris was characterized by a Mn^{54}/Sr^{90} activity ratio of about 100, when corrected for decay to October, 1961. It seems reasonable to assume that debris from these very high yield events was the most likely to be injected at high altitudes, if any debris at all from the 1961 Soviet low altitude events was so injected. Had any significant quantity of such debris reentered the region of the stratosphere between 24 and 32 km during mid-1963 to mid-1965 it should have produced a marked increase in the Mn^{54}/Sr^{90} ratio found in samples collected there, but this ratio remained below 15 during 1963 and 1964, and generally decreased with the passage of time. Similarly the fission product ratios in these samples showed no changes such as would have been produced by the reentry of old debris from the 1961 events.

It might be hypothesized that radioactive debris from the very high yield U.S.S.R. tests during 1961 and 1962 was indeed injected at altitudes above 32 km, and that most of it had not yet reentered the atmospheric layers below 32 km by early 1965. This also seems unlikely

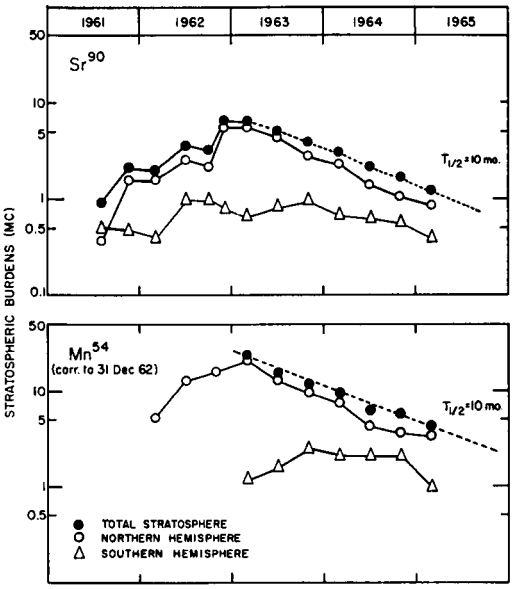

FIG. 5. Stratospheric burdens of strontium-90 and manganese-54.

for less than one per cent of the mass of the atmosphere is present in the region above 32 km, and the injection of large quantities of weapon debris into this region would produce very high concentrations of radioactivity there. Vertical mixing within the stratosphere, especially during the winter seasons, would probably, within two years, transfer most of this debris into the layer between 24 and 32 km. This layer constitutes about two per cent of the atmosphere and by 1964 contained comparatively low concentrations of radioactive debris.

Even if rates of vertical mixing in the upper atmosphere are too slow to bring down into the lower atmosphere within a year or two most of the debris originally injected at high altitudes, particle settling would probably accomplish this transfer. Rates of particle settling are quite rapid in the thin air above 32 km, even for particles as small as 0.01 micron in radius (JUNGE *et al.*, 1961). A spherical particle with this radius and a density of 2 g cm^{-3} would settle from 50 km to 30 km in less than a year. Debris from weapons tests would probably be present on much larger, much denser particles, and thus would settle much more rapidly into the lower stratosphere.

During both Project Stardust and the U.S.A. E.C. balloon sampling program, measurements

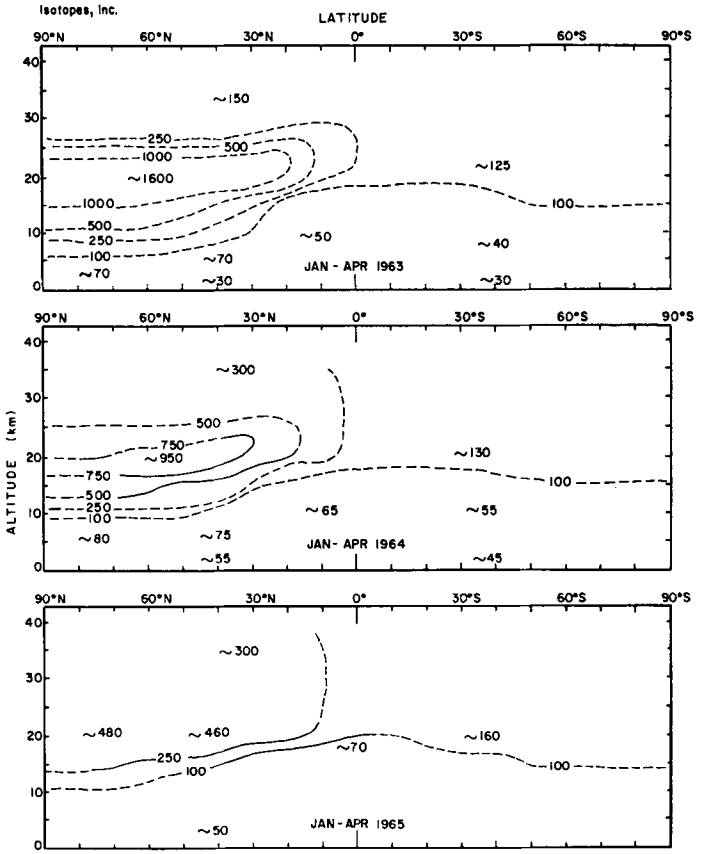

FIG. 6. Distribution of carbon-14 in the atmosphere (10^5 excess atoms C^{14}/g air).

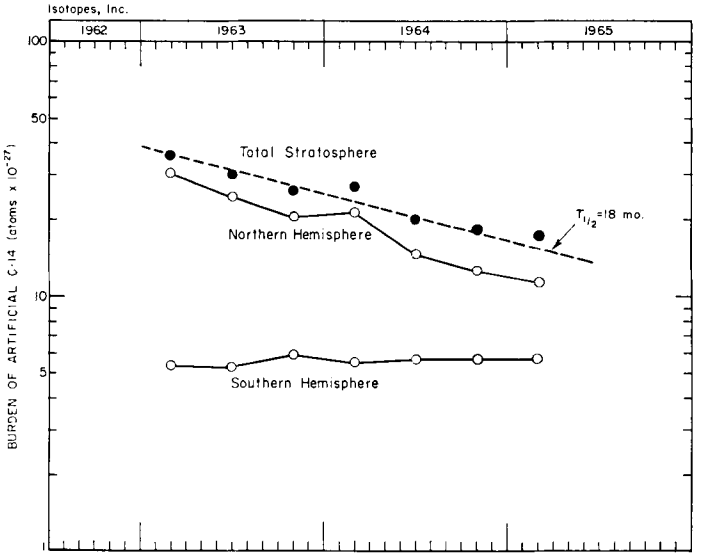

FIG. 7. Stratospheric burdens of artificial carbon-14.

have been made of the cadmium-109 tracer nuclide injected into the upper atmosphere by the Starfish Prime event, detonated at 400 km over Johnston Island on July 9, 1962. We believe that the results of these measurements indicate that much, and perhaps most, of the debris from this event was present in the stratospheric layers below 32 km by the end of 1964. If this is true it appears very unlikely that much debris from any of the low altitude events in the 1961 and 1962 test series could still be present in the atmospheric regions above 32 km.

It is also noteworthy that the maximum concentrations of both particulate and gaseous debris persisted in the lower stratosphere of the Northern Hemisphere for at least two years. Measurements made at Argonne National Laboratory (HAGEMANN *et al.*, 1965; Health and Safety Laboratory, 1966) do show, however, that during late 1964 and early 1965 the highest concentrations of carbon-14 at 35° N were found at about 24 km, rather than at 20 km where they had been found during all of 1963 and, for the most part, during the first half of 1964.

Assuming that the most important process causing the movement of radioactive debris within the stratosphere is eddy diffusion (FEELY & SPAR, 1960; NEWELL, 1961), we would expect upward movement of some debris from the region of highest concentration. We would also expect that as the stratospheric concentrations decreased, the level of maximum concentration would be found at higher and higher altitudes. This gradual change in the distribution pattern should be expected because removal of the stratospheric debris, both particulate and gaseous, can occur only at the lower boundary of the stratosphere, and the most rapid decreases in concentration should take place in the lowest of the stratospheric layers. Depletion of these layers would lower the concentrations in them to values below the concentrations in the higher layers, more distant from the tropopause, and would result in an upward migration of the level of highest concentration whether or not any of the radioactive debris was also diffusing upward.

Indeed, there was a significant increase in carbon-14 concentrations at altitudes above 24 km, at 35° N during 1963, and this may have resulted in part from the upward diffusion of carbon-14 from the layer below 24 km, which contained very high concentrations. In addition, the level of maximum concentration, which was found at about 20 km during 1963, was found at about 24 km by the second half of 1964. Thus the changes which occurred in the stratospheric distribution of carbon-14 during 1963 to 1965 were consistent with the behavior expected if the injected radioactive debris were affected mainly by eddy diffusion.

On the other hand, concentrations of strontium-90 and manganese-54 in the upper stratosphere in general, and in the upper stratosphere of the Northern Hemisphere especially, were decreasing during 1963 to 1965. In addition, the level of maximum concentration of this particulate debris remained in the lower stratosphere during this period. It has been proposed that this effect indicates that particulate debris in the upper stratosphere during 1963 to 1965 was moving downward as a result of particle settling more rapidly than it could be moved upward by eddy diffusion (FEELY & BAZAN, 1965). It is probably because particle settling has caused the level of maximum concentration of particulate debris to remain in the lower stratosphere that the apparent stratospheric residence time of such debris has not increased significantly during the past few years. Of course, the rate of particle settling decreases with increasing air density, and in the lower stratosphere, the effects are probably small compared to those produced by air motions.

4. The movement of radioactive debris into the Southern Hemisphere

While concentrations of radioactive debris in the stratosphere of the Northern Hemisphere were decreasing during 1963, concentrations in the Southern Hemisphere were increasing. Bimonthly mean concentrations of manganese-54, strontium-90, and carbon-14 at the 20 km level at 35°–40° N and 35°–40° S are shown in Figure 8. The figure shows that concentrations of particulate debris at 20 km at 35°–40° S more than doubled between May–June 1963 and September–October 1963, during the late winter and early spring of the Southern Hemisphere. Concentrations of carbon-14 at this location increased only about 11 per cent at the time, however. In Figure 9 are plotted the ra-

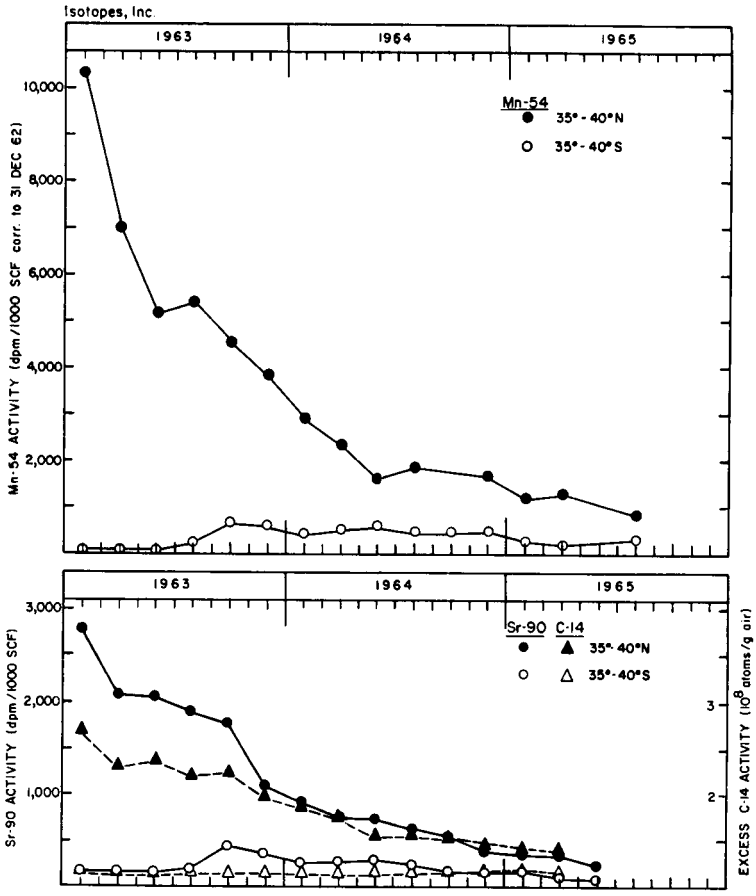

FIG. 8. Trends in activities of manganese-54, strontium-90, and carbon-14 at 35°–40° N and 35°–40° S at 20 km.

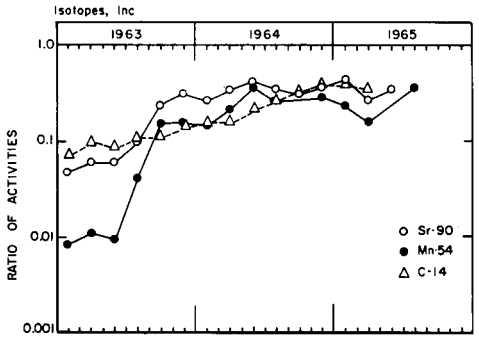

FIG. 9. Ratios of nuclide activities at 35°–40° S to the activities at 35°–40° N, both at 20 km.

tios of the nuclide concentrations at 20 km at 35°–40° S to the concentrations at 35°–40° N. These ratios are a measure of the concentration gradient across the tropical stratosphere: i.e., a steep concentration gradient is indicated by a low ratio. The ratio of carbon-14 concentrations increased from about 0.07 in early 1963 to about 0.16 in early 1964 and to about 0.40 in early 1965. This indicates only a gradual decrease of the concentration gradient across the tropical stratosphere during 1963 to 1965. The ratio of strontium-90 concentrations increased rapidly from 0.06 in mid-1963 to 0.3 by the end of 1963 but then increased only slowly with fluctuations between 0.27 and 0.43 during the next 18 months. Similarly, the ratio of manganese-54 activities increased rapidly from 0.01 in mid-1963 to 0.16 by late 1963,

but then rose only slowly, with fluctuations between 0.15 and 0.37, during the next 18 months. Thus during the Southern Hemisphere 1963 winter season there was a rapid decrease in the gradients of strontium-90 and manganese-54 concentrations across the tropical stratosphere; but subsequently, there was only gradual further decrease in these gradients.

The bimonthly mean nuclide concentrations at 20 km at 35–40° S have been normalized relative to the activities at that location during January–February 1963, and the results have been plotted in Figure 10. The normalized activities of carbon-14 show little departure from 1.0 during 1963 to early 1965. The normalized activities of both strontium-90 and manganese-54, on the other hand, increased sharply in late 1963 to values of about 3 and 8, respectively, but showed a general decrease during the subsequent 18 months. During early 1965, in fact, the strontium-90 concentrations decreased below 1.0.

It might be concluded, on the basis of the data summarized in Figures 8, 9, and 10, that in early 1963 there was between 35°–40° N and 35°–40° S at 20 km a very large difference in concentrations of manganese-54 (a factor of 100), a smaller difference in concentrations of strontium-90 (a factor of 16), and a still smaller difference in concentrations of carbon-14 (a factor of 11). Thus, across the tropical stratosphere at that time the concentration gradient of manganese-54 was very steep, that of strontium-90 was much less steep, and that of carbon-14 was still less steep. During late 1963, there was a movement of radioactive debris into the southern stratosphere which raised severalfold the concentrations of strontium-90 and manganese-54 at 20 km at 35°–40° S. During subsequent months, the ratios of the concentrations of these nuclides at 35°–40° S to their concentrations at 35°–40° N continued to rise gradually, though with fluctuations, in spite of the fact that the actual concentrations of strontium-90 and manganese-54 at 20 km at 35°–40° S were gradually decreasing. Obviously, the ratios increased only because the concentrations at 35°–40° N were decreasing more rapidly than were those at 35°–40° S.

Unlike the particulate debris, the carbon-14 showed only a small increase in concentration at 20 km at 35°–40° S during late 1963, and during

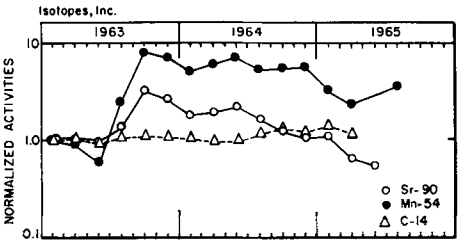

FIG. 10. Nuclide activities at 35°–40° S at 20 km normalized relative to the activities at that location during January–February, 1963.

subsequent months showed a fairly steady, rather than a gradually decreasing, concentration there. Evidently, carbon-14 was moving from the stratosphere of the Northern Hemisphere to that of the Southern Hemisphere at about the same rate as it was moving from the stratosphere to the troposphere of the Southern Hemisphere. The ratio between carbon-14 concentrations at this location and those at 35°–40° N rose gradually during 1963–1965, indicating a decreasing concentration gradient across the tropical stratosphere, but mainly because of a continuous decrease in concentrations at the Northern Hemisphere location.

This analysis of the data strongly indicates that the rate of transport of carbon-14 into the Southern Hemisphere differed significantly from that of particulate debris during the second half of 1963. A possible explanation for this difference may be obtained in terms of the difference in the concentration gradients and in the paths followed by particulate debris and carbon-14 in crossing the tropical stratosphere. The rate of eddy diffusion is directly proportional to the existing concentration gradient of material. Since it has been shown that the concentration gradients of the particulate material were considerably greater than those of the carbon-14 during the first half of 1963, it is to be expected that the rate of transport, in turn, would be more rapid. As a result, relatively more particulate debris than carbon-14 should have reached the stratosphere of the Southern Hemisphere during those months when interhemispheric transfer was accelerated. It would appear, however, that an additional factor must have affected the relative rates of diffusion of the particulate debris and carbon-14, for by late 1963, the concentration gradients across the tropical stratosphere, as indicated by the

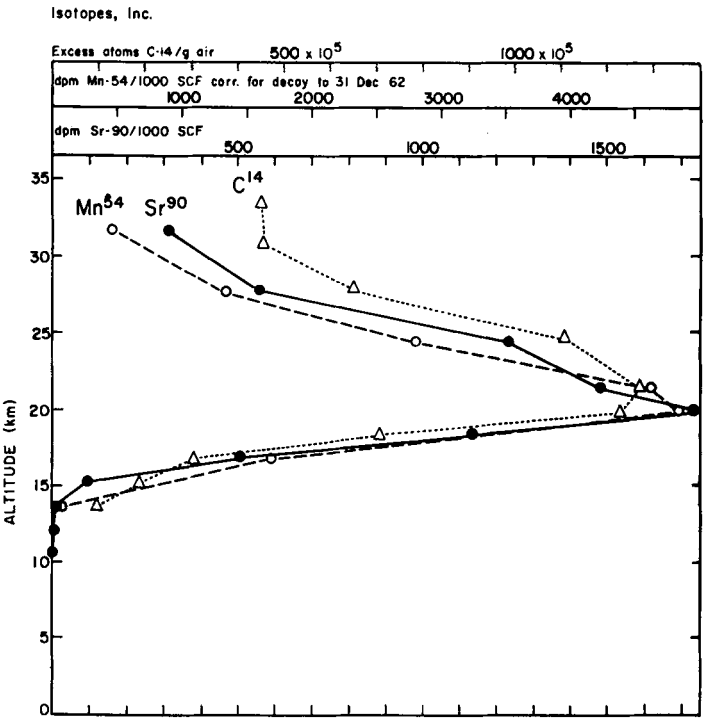

FIG. 11. Vertical profiles of strontium-90, manganese-54, and carbon-14 concentrations at about 35° N during September–October, 1963.

ratios in Figure 9, had become less steep for the particulate debris than for carbon-14.

We will hypothesize that the additional factor which was active was the separation of particulate debris from carbon-14 in the equatorial stratosphere as a result of particle settling. It has been pointed out in an earlier section that the stratospheric distribution of particulate debris in the regions above 20 km appears to have been considerably affected by particle settling during the period 1963–1965. Such particle settling could strongly influence rates of interhemispheric transfer of radioactive debris if the southward movement of the air carrying the debris from the Northern Hemisphere source region into the equatorial stratosphere is accompanied by an upward movement of the air into the regions above 20 km. The carbon-14, present as the gas carbon dioxide, would move with the air, but the particulate debris, under the influence of particle terminal velocities, would tend to follow a lower trajectory and enter the layer of air beneath that which originally contained it. In this way, the movement of air into the equatorial stratosphere could be accompanied by a partial separation of gaseous and particulate debris, with the carbon-14 in the equatorial stratosphere concentrated in the layers above 20 km, and the particulate debris concentrated in the layer between 17 and 20 km.

Based on the presentation by NEWELL (1963) of the distribution of the standard deviations of the standing eddy component and transient eddy component of the total wind vector, it is evident that the meridional transport in the equatorial stratosphere would be greater in the layer between the tropopause and 20 km than in the layer between 20 and 25 km. This difference in diffusion rates could have produced the preferential movement of particulate debris, concentrated in the lower layer, into the Southern Hemisphere during the second half of 1963.

The Argonne National Laboratory data for balloon samples collected at 35° N, provide some information on the vertical distribution of carbon-14 in the upper stratosphere, but not in the equatorial stratosphere, where we hypo-

thesize the separation of carbon-14 from particulate debris will take place. Vertical concentration profiles of carbon-14, strontium-90, and manganese-54 at about 35° N during September–October 1963 are shown in Figure 11. At that latitude, the peak concentrations of strontium-90 and manganese-54 were found at about 20 km and those of carbon-14 at about 22 km. Balloon samples of particulate debris have been collected at about 9° N since late 1964 (SALTER, 1965B). Measurements of these samples have shown that, although roughly equal concentrations of radioactive debris occur at 9° N and 31° N at about 20 km, the concentrations at about 24, 27, and 30 km are generally twice as high at 9° N as at 31° N. This is consistent with the hypothesis that large-scale eddy diffusion occurs along quasi-horizontal surfaces which slope upward comparatively steeply from 31° N to 9° N. If such sloping surfaces do indeed exist, they could have resulted in the separation of particulate debris from gaseous debris in the equatorial stratosphere in 1963.

5. Summary

Particulate radioactive debris from nuclear weapons tests, as represented by strontium-90 and manganese-54, displayed a stratospheric residence half-time of about 10 months during 1963 and 1964. The stratospheric burden of strontium-90 decreased from over 6 megacuries in early 1963, to about 3 megacuries in early 1964, and to about 1.3 megacuries in early 1965. The statospheric burden of manganese-54, corrected for decay to 31 December, 1962, decreased from about 24 megacuries in early 1963, to about 9.7 megacuries in early 1964, and to less than 5 megacuries in early 1965.

The "excess" carbon-14 produced by nuclear weapons tests displayed a stratospheric residence half-time which gradually lengthened during 1963 and 1964, but averaged about 18 months. The carbon-14 burden of the stratosphere decreased from about 36×10^{27} atoms in early 1963 to about 27×10^{27} atoms in early 1964, and to about 17×10^{27} atoms in early 1965.

The highest concentrations of carbon-14 were found in the lower stratosphere during 1963 and 1964, but the level of the maximum concentration gradually rose from about 20 km in early 1963 to about 24 km by mid-1964. The highest concentrations of strontium-90 and manganese-54 were also found in the lower stratosphere during 1963 to 1965. The level of the maximum concentration of this particular debris did not rise during 1963–1965, but remained at about 20 km. At sampled levels above 20 km, moreover, the concentrations decreased markedly during 1963 and 1964. It is concluded that these effects reflect the action of particle settling in the relatively thin air above 20 km. It is suggested that this particle settling also prevented any significant increase from occurring in the stratospheric residence half-time of particulate debris during 1963–1965.

It is observed that the rate of transport of particulate radioactive debris into the stratosphere of the Southern Hemisphere during the second half of 1963 was more rapid than the rate of transport of carbon-14. It is suggested that an explanation for this difference may be obtained in terms of the difference in concentration gradients across the tropical stratosphere in early 1963, and of the separation of particulate debris from carbon-14 in the equatorial stratosphere as a result of particle settling.

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ПЕРЕНОС И ВЫПАДЕНИЕ ОСТАТКОВ РАДИОАКТИВНОГО РАСПАДА В СТРАТОСФЕРЕ

В течении двух лет после последних атмосферных испытаний мощного ядерного оружия, средний полупериод очищения стратосферы от радиоактивных остатков, таких элементов как стронций 90 и магний 54, связанных с очень маленькими частицами, был приблизительно равен 10 месяцам. Поскольку углерод 14 образуется во время ядерных испытаний, полупериод очищения от него стратосферы увеличивался в течении этого времени, но был в среднем около 18 месяцев.

Это предполагает, что полупериод очищения стратосферы от связанных с маленькими частицами остатков не увеличивался существенно с 1963 до начала 1965 годов, потому что в верхней стратосфере (выше 20 км) скорость выпадения радиоактивных частиц превосходила скорость диффузии вверх.

В результате этого, в течение указанного времени эти радиоактивные остатки имели высокую концентрацию в нижней стратосфере. В стратосфере южной полусферы в течении второй половины 1963 г. имело место относительно более быстрое меридиональное движение этих радиоактивных остатков, по сравнению с движением углерода 14. Предполагается также, что это проистекает в некоторой степени от частичного разделения связанных с малыми частицами остатков от углерода 14 в экваториальной стратосфере в результате выпадения частиц.