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FINAL REPORT ON PROJECT STARDUST

VOLUME I, Chapters 1 - 6

by

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VOLUME I

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CHAPTER 1. OBJECTIVES AND ACCOMPLISHMENTS OF PROJECT STARDUST

In this, the final report on Project STARDUST, a review is given of the results of analyses of filter and gas samples collected during the course of the program, 1961 to 1967, and a summary is given of the conclusions reached on the basis of these results concerning the influence of atmospheric processes on the transfer and fallout of radioactive materials injected into the stratosphere. Reference is made to results obtained during Project HASP also, for in many ways Project STARDUST was a continuation of that program.

1.1 The Objectives of Project STARDUST

Project STARDUST was initiated in 1961, during what proved to be the final year of the 1958-1961 informal moratorium on nuclear weapon tests. The project was undertaken initially to prepare a mathematical model of atmospheric transport and deposition which could be used to predict stratospheric hold-up and ultimate patterns of deposition on the surface of the earth of radioactive debris injected into the stratosphere by nuclear weapon tests, by burnup on re-entry of radioactive power sources in earth satellites, or by other causes. It was anticipated that the results obtained during Project HASP would constitute the main source of information on stratospheric radioactivity for use in work on the model, but a limited filter sampling program was begun in June 1961 to provide some additional data.

When the U.S.S.R. resumed the testing of nuclear weapons in September 1961, the decision was made to expand the STARDUST sampling program to permit the monitoring of concentrations of radioactive debris injected into the stratosphere. It was anticipated that this program would provide data needed for the estimation of the potential future hazard from such debris. Thus, during 1962 sampling missions were flown from Alaska, from the Panama Canal Zone, and finally in the southern hemisphere. Only filter samples were collected during the period June 1961 to July 1963, but during the last four years of the project, August 1963 to June 1967, gas samples were collected as well as filter samples.

In the decision as to which measurements should be made on STARDUST samples, a major consideration was the need to obtain information which would be useful in the development and testing of the STARDUST model of atmospheric transport and deposition. For this reason a variety of radionuclides from the filter samples was measured, including not only fission products but also products of neutron activation, which could be used as tracers for specific nuclear events, and natural radionuclides, which could serve as tracers with completely different source characteristics. Measurement of plutonium-238 in STARDUST samples was begun when the burnup of a SNAP-9A power source in April 1964 injected this nuclide into the stratosphere. This event provided a tracer for which the source characteristics were rather well known. Moreover, this type of event was one of those anticipated when it was decided that if the effects of the use of nuclear energy in space were to be properly evaluated, there would be a need for a model of atmospheric transport and deposition. In the development of the STARDUST model, use was made of measurements of atmospheric ractioactivity performed during Project HASP, during Project STARDUST, during the U.S. Atomic Energy Commission high altitude balloon sampling program, and during other measurement programs.

Even after the development of the model of atmospheric transport and deposition was removed from the objectives of Project STARDUST in late 1965, the acquisition of data useful to the development of this or similar models remained the main aim of the project. The measurements made of radioactive debris from pre-1963 weapon tests, of the SNAP-9A plutonium-238, and of radioactive debris from the 1966 Chinese and French weapon tests were all undertaken to provide information on the nature and rates of movement of air masses within the stratosphere or between the stratosphere and the troposphere. Such data is essential to the proper design of such atmospheric models.

1.2 The Sampling and Analytical Techniques Used

The basic aim of the STARDUST sampling program was the determination of the distribution of certain radionuclides within the stratosphere. Since it was clearly not possible to sample the entire stratosphere, it was believed that representative data would be obtained by sampling periodically within a meridional plane. It could be assumed that the zonal circulation would eventually carry all of the stratosphere through that meridional plane. Because of the limitation imposed by the operational requirements of the aircraft, however, it was necessary to perform the actual sampling within a series of meridional and quasi-meridional segments, which were generally, but not always contiguous.

The range in altitude over which collections were performed was dictated mainly by the flight characteristics of the available aircraft. It was seldom possible to obtain samples from altitudes much above 20 km. In the southern hemisphere, generally only WU-2 or RB-57F aircraft were available for use, and sampling was limited to altitudes above 14 km, since these aircraft performed most efficiently at high altitudes. When aircraft such as the RB-57C and C-130, which could be flown efficiently below an altitude of 14 km were available, they were used to sample the upper troposphere and the lowest regions of the stratosphere. These aircraft were more often available and had more bases from which they could fly in the northern hemisphere than in the southern hemisphere. As a result most sampling at altitudes below 14 km was performed in the northern hemisphere.

STARDUST filter samples were collected by means of the IPC-1478 filter medium, which was developed at the Institute of Paper Chemistry. This filter medium offers a low resistance to air flow at high velocities, but it has a high collection efficiency under flight conditions. The filter samples were shipped to Isotopes, Inc. in Westwood, N. J., where an aliquot was assayed radiometrically by beta counting, generally over the course of a few weeks, to determine the total activity and the decay rate. From these the approximate age of the fission products in the sample could be determined. At times gamma spectra of filters were measured to detect quickly the presence of new debris or to determine the presence of certain gamma-emitters, such as antimony-124 or yttrium-88. Subsequent to the total beta and total gamma measurements, aliquots of the filters were ashed and certain fission products, products of neutron activation and a few radionuclides of natural origin, chemically separated. Most STARDUST analytical results are based on the radio-assay of separated and purified elements.

Whole air samples were collected during most STARDUST sampling missions flown after July 1963. The air was compressed to 3000 psi in spherical steel bottles of about 900 cubic inches capacity. These bottles were shipped to Air Reduction Company, Inc. in Murray Hill, N. J., where the carbon dioxide was separated from each sample. The carbon dioxide and aliquots of the original whole air samples were then shipped to Isotopes, Inc., where the carbon-14 concentrations of the samples were determined. Carbon-14 in carbon dioxide samples collected from surface air in northern New Jersey during 1960 to 1967 was also measured.

A small number of samples of stratospheric particles was collected during the first few years of the program, using direct flow impactors which were contained in probes mounted on the WU-2 aircraft. These impactor samples were analyzed using a variety of techniques, but the most useful data were obtained by means of electron microscopic and electron diffraction analyses performed by Ernest F. Fullam, Inc. of

1.3 The Accomplishments of Project STARDUST

The primary objectives of Project STARDUST were accomplished satisfactorily. A large amount of relevant information was obtained on the mechanisms which transfer radioactive materials from one region of the stratosphere to another and from the stratosphere to the troposphere and this information was used to prepare and test a model of atmospheric transfer and deposition.

The electron microscope measurements of the stratospheric particles which were collected using the direct flow impactors provided some useful information on the size distribution of the stratospheric sulfate aerosol. Measurements of several fission products in radioactive debris from the 1966 Chinese and French nuclear weapon tests gave some insight into fractionation effects within the radioactive clouds from these events. Information was obtained on the presence of various products of neutron activation in radioactive debris from the 1961 and 1962 test series, and to some extent, on their presence within debris from specific events in those series. All of these data concerning the stratospheric aerosol and concerning the composition of the stratospheric radioactivity may be used in seeking to evaluate the extent to which the movement of radioactive particles within the stratosphere corresponds to the movement of the air itself. Evidence was found that the rate of particle settling within the upper stratosphere controls the stratospheric residence time of such debris. This residence time has been found to be less than one year.

Observations of radioactive debris injected into the stratosphere have shown that dispersion in the meridional direction occurs much more rapidly within the polar stratosphere than within the tropical stratosphere. They also show that the rate of meridional transport through the tropical stratosphere is seasonally dependent. STARDUST measurements showed that the radioactive debris which was injected into the lower northern polar stratosphere during the 1962 U.S.S.R. test series was transferred to the southern polar stratosphere very slowly because of the low rate of dispersion through the tropical stratosphere. Equal concentrations in the two polar stratospheres were not reached even four years after injection. It was observed in 1963 and in 1959 that the transfer of material across the equator and into the southern polar stratosphere occurs preferentially during the winter season of the southern hemisphere. Observations of the rapid southward movement of radioactive debris injected into the lower northern polar stratosphere at mid-latitudes by the 9 May 1966 Chinese nuclear weapon test suggested that the high altitude return circulation of the southwest monsoon might have been instrumental in moving this material to low latitudes. This circulation might also facilitate in other years the interhemispheric exchange of material originally situated in the lower polar stratosphere.

The rate at which radioactive materials were dispersed in the vertical direction was also observed to be strongly dependent upon the season of the year. Within the lower polar stratosphere the rate of vertical mixing increased markedly during the late autumn and winter seasons. This was reflected by the dispersion of cadmium-109 which was injected at high altitude by a nuclear weapon test in July 1962, and especially by the dispersion of plutonium-238 which was injected into the upper stratosphere in April 1964 as a result of the burnup of a SNAP-9A power source. The plutonium-238 appeared in significant quantities at 20 km in each hemisphere early in the winter season for that hemisphere, and had reached the level of the tropopause in that hemisphere by the end of the winter season.

Two sets of data obtained during Project STARDUST appear to provide confirmation that one mechanism of transport of radioactive material from the stratosphere to the troposphere involves the movement of air along surfaces of constant potential temperature which may be traced from the lower polar stratosphere into the upper tropical troposphere. During "Project SPRINGFIELD", which was carried out as part of Project STARDUST during the spring of 1963, E. F. Danielsen was able to demonstrate that thin laminae of air which exhibited high values of potential vorticity and which were found along developing frontal systems over the southwestern United States contained high concentrations of radioactive bomb debris, characteristic of stratospheric air. The presence of this debris could thus be attributed to the extrusion of stratospheric air into the troposphere in conjunction with the folding of the tropopause. Some interceptions of radioactive debris from the 9 May 1966 Chinese test were consistent with the hypothesis that the debris had recently moved from the lower polar stratosphere, through the "tropopause gap" region and into the upper tropical stratosphere along surfaces of constant potential temperature.

Using data from Project STARDUST and from the U.S. A.E.C. balloon sampling program it was calculated that the strontium-90 burden of the stratosphere reached a high value of 6.5 megacuries by early 1963, but subsequently decreased with a stratospheric residence half-time of about 10 months to a value of about 0.5 megacurie by mid-1966. Other radionuclides produced by the 1961 and 1962 test series and carried by particulate debris exhibited a similar stratospheric residence half-time. Artificial carbon-14, present in the stratosphere and troposphere in carbon dioxide, exhibited a stratospheric residence half-time which was longer than that of the particulate debris and which increased with the passage of time during 1963 to 1967. Measurements of radioactive debris from nuclear devices tested during 1966 indicated that the Chinese test of 9 May 1966 injected about 5.5 kilocuries of strontium-90 into the stratosphere, and that the French test series injected about 7 kilocuries of strontium-90.

A few radionuclides which are produced in the atmosphere by natural processes were measured during Project STARDUST to provide data on tracers which had a source configuration quite different from that of debris from nuclear weapon tests. The cosmic ray product beryllium-7 was found to have a distribution within the stratosphere which is only slightly different from the distribution expected in a stagnant atmosphere. Lead-210, a daughter product of radon, which leaks into the atmosphere from soil and rocks at the surface of the earth, was found to be almost in radioactive equilibrium with its daughter product polonium-210, suggesting that it had a stratospheric residence half-time of the order of one to two years.

The STARDUST numerical model of transport and deposition describes a general anisotropic diffusion process. Variations of the model were tested in which the principal diffusion axis was along a horizontal surface, along surfaces of constant potential temperature, and approximately along surfaces of constant potential vorticity. Only the last-mentioned of these versions successfully reproduced the observed distributions of radioactivity in the stratosphere.

1.4 Reports of Results Obtained During Project STARDUST

A number of reports have been issued during Project HASP and Project STARDUST giving results of measurements of filter samples and gas samples, or discussing the significance of these results. The following reports contain the results of radiochemical analyses of filter samples:

1. J. P. Friend, H. W. Feely, P. W. Krey, J. Spar and A. Walton, "The High Altitude Sampling Program", Defense Atomic Support Agency, report DASA 1300, Volume 2, (1961).
2. H. W. Feely, J. P. Friend, P. W. Krey, and B. A. Russell, "The Tenth Progress Report on Project STARDUST - Results of Radiochemical Analyses of Filter Samples Collected During 1961 and 1962", USAEC report HASL-153, (1965).
3. H. W. Feely, J. P. Friend, R. J. Lagomarsino, D. C. Bogen, P. E. Biscaye and J. E. Hardaway, "Appendix to the Eleventh Progress Report on Project STARDUST - Results of Radiochemical Analyses of Filter Samples Collected During 1963", USAEC report HASL-168, (1966).
4. H. W. Feely, D. C. Bogen, R. J. Lagomarsino, J. E. Hardaway and M.W.M. Leo, "The Twelfth Progress Report on Project STARDUST - Results of Radiochemical Analyses of Filter Samples Collected During 1964", USAEC report HASL-189, (1966).

5. H. W. Feely, D. Katzman, P. E. Biscaye, J. B. Panaccione and E. R. French, "The Thirteenth Progress Report on Project STARDUST - Results of Radiochemical Analyses of Filter Samples Collected During 1965", USAEC report HASL-176, (1966).

6. H. W. Feely, D. Katzman, J. B. Panaccione and E. R. French, "The Fourteenth Progress Report on Project STARDUST - Results of Radiochemical Analyses of Filter Samples Collected During January-March 1966", USAEC report HASL-173, pp III-2 - III-25, (1966).

7. H. W. Feely, D. Katzman, J. B. Panaccione and E. R. French, "The Fifteenth Progress Report on Project STARDUST - Results of Radiochemical Analyses of Filter Samples Collected During April - June 1966", USAEC report HASL-174, pp. III-2 - III-27, (1967).

8. H. W. Feely, D. Katzman, J. B. Panaccione and E. R. French, "The Seventeenth Progress Report on Project STARDUST - Results of Radiochemical Analyses of Filter Samples Collected During July-September 1966", USAEC report HASL-181, pp III-2 - III-23, (1967).

9. H. W. Feely, D. Katzman, J. B. Panaccione and E. R. French, "The Eighteenth Progress Report on Project STARDUST - Results of Radiochemical Analyses of Filter Samples Collected During October-December 1966", USAEC Report HASL-182, pp III-2 - III-29, (1967).

10. H. W. Feely, D. Katzman and J. B. Panaccione, "The Nineteenth Progress Report on Project STARDUST - Results of Radiochemical Analyses of Filter Samples Collected During January - March 1967", USAEC report HASL-183, pp III-6 - III-30, (1967).

11. H. W. Feely, D. Katzman and S. Kaminsky, "The Twentieth Progress Report on Project STARDUST - Results of Radiochemical Analyses of Filter Samples

Collected During April - June 1967", USAEC report HASL-184, pp III-2 - III-21, (1968).

The following reports contain the results of carbon-14 analyses of carbon dioxide from gas samples of stratospheric air:

1. ESSA, "Carbon-14 Measurements in the Atmosphere", USAEC report HASL-166, (1966).
2. ESSA, "Carbon-14 Measurements in the Atmosphere", USAEC report HASL-174, (1967).

The following progress reports contain discussions of the significance of the data obtained during Project HASP and Project STARDUST:

1. A. K. Stebbins, III, "Third Annual HASP Briefing", Defense Atomic Support Agency report DASA 531 (1959).
2. H. W. Feely and J. Spar, "Mixing and Transfer within the Stratosphere", Defense Atomic Support Agency report DASA 1222 (1960).
3. A. K. Stebbins, III, "Special Report on High Altitude Sampling Program", Defense Atomic Support Agency report DASA 532B, (1960).
4. J. P. Friend, H. W. Feely, P. W. Krey, J. Spar and A. Walton, "The High Altitude Sampling Program", Defense Atomic Support Agency report DASA 1300, 5 volumes, (1961).
5. A. K. Stebbins, III, "Second Special Report on High Altitude Sampling Program", Defense Atomic Support Agency report DASA 539 B, (1961).
6. J. P. Friend and H. W. Feely, "Second Quarterly Report on Project STARDUST", Defense Atomic Support Agency report DASA 1302, (1961).

7. J. P. Friend, H. W. Feely, E. L. Fisher and B. Davidson, "Third Quarterly Report on Project STARDUST", Defense Atomic Support Agency report DASA 1303, (1962).

8. J. P. Friend and H. W. Feely, "Fifth Quarterly Report on Project STARDUST", Defense Atomic Support Agency report DASA 1305, (1962).

9. J. P. Friend, H. W. Feely and M. W. M. Leo, "Sixth Quarterly Report on Project STARDUST", Defense Atomic Support Agency report DASA 1306, (1962).

10. J. P. Friend, H. W. Feely and M.W.M. Leo, "Seventh Quarterly Report on Project STARDUST", Defense Atomic Support Agency report DASA 1307, (1963).

11. J. P. Friend, H. W. Feely and M.W.M. Leo, "Eighth Quarterly Report on Project STARDUST", Defense Atomic Support Agency report DASA 1308, (1963).

12. H. W. Feely, B. Davidson, J. P. Friend, R. Lagomarsino and M.W.M. Leo, "Ninth Quarterly Report on Project STARDUST", Defense Atomic Support Agency report DASA 1309, (1963).

13. E. F. Danielsen, "Project SPRINGFIELD Report", Defense Atomic Support Agency report DASA 1517, (1964).

14. H. W. Feely, P. E. Biscaye, B. Davidson and H. Seitz, "Eleventh Progress Report on Project STARDUST", Defense Atomic Support Agency report DASA 1821, (1966).

15. H. W. Feely, D. Katzman and C. S. Tucek, "Sixteenth Progress Report on Project STARDUST", Defense Atomic Support Agency report DASA 1905, (1966).

The following papers, containing discussions of data obtained during Project HASP and Project STARDUST, have been published in the scientific literature or presented at scientific conferences:

1. J. Spar, "Strontium-90 in the Stratosphere", a paper presented at the strontium-90 symposium, Bad Kreuznach, Federal Republic of Germany, (1959).

2. H. W. Feely, "Strontium-90 Content of the Stratosphere", Science, 131, 645 - 649, (1960).

3. H. W. Feely and J. Spar, "Tungsten-185 from Nuclear Bomb Tests as a Tracer for Stratospheric Meteorology", Nature, 188, 1062 - 1064, (1960).

4. P. W. Krey, D. Bogen and E. French, "Plutonium in Man and His Environment", Nature, 195, 263 - 265, (1962).

5. A. Walton, "The Distribution in Soils of Radioactivity from Weapons Tests", Jour. of Geophys. Res., 68, 1485 - 1496, (1963).

6. H. W. Feely and J. P. Friend, "Evidence on Stratospheric Circulation from Measurements of Radioactive Bomb Debris Injected above 20 km", a paper presented at the Thirteenth General Assembly of the IUGG, Berkeley (1963).

7. H. W. Feely, "Discussion of Paper by W. F. Libby - Moratorium Fallout and Stratospheric Storage", Jour. of Geophys. Res., 68, 6215 - 6220, (1963).

8. H. W. Feely and F. Bazan, "Stratospheric Distribution of Nuclear Debris in 1962, 1963 and 1964", in "Proceedings of the Second Conference on Radioactive Fallout from Nuclear Weapons Tests", Germantown, Md., Nov. 1964, published by the USAEC as CONF-765, (1965).

9. H. W. Feely, J. P. Friend and H. Seitz, "The Role of Particle Settling in the Movement of Radioactive Debris in the Stratosphere", a paper presented at the Forty-sixth Annual Meeting of the American Geophysical Union, Washington, D. C. (1965).

10. B. Davidson, J. P. Friend and H. Seitz, "Numerical Models of Diffusion and Rainout of Stratospheric Radioactive Materials", Tellus, 18, 301 - 315, (1966).

11. H. W. Feely, H. Seitz, R. J. Lagomarsino and P. E. Biscaye, "Transport and Fallout of Stratospheric Radioactive Debris", Tellus, 18, 316 - 328, (1966).

12. J. P. Friend, "Properties of the Stratospheric Aerosol", Tellus, 18, 465 - 473, (1966).

13. H. W. Feely, C. Barrientos and D. Katzman, "Radioactive Debris Injected into the Stratosphere by the May 9, 1966 Chinese Nuclear Weapon Test", Nature, 212, 1303 - 1304, (1966).

14. H. W. Feely, "Discussion of the Paper by J. N. Beck and P. K. Kuroda - Radiostrontium Fallout from the Nuclear Explosion of October 16, 1964", Jour. of Geophys. Res., 72, 3295 - 3299, (1967).

CHAPTER 2. THE STARDUST SAMPLING PROGRAM

The sampling techniques used during Project STARDUST were generally similar to those used during Project HASP, and described in the final report on that project[1]. There were some changes, however, in the types of aircraft used, in the configuration of the filter samplers employed, and in the flight tracks followed. The probe sampler employed to collect impactor samples of the stratospheric aerosol was modified to provide a higher collection efficiency for small particles. The main addition to the sampling procedures made during Project STARDUST was the collection of whole air samples, to be analyzed for carbon-14. In this chapter the procedures used for collecting filter samples and whole air samples are summarized, and the schedules of sampling flights which were followed are reviewed. The procedures used to collect impactor samples are discussed in Chapter 5.

2.1 The Collection of Air Filter Samples

Filter samples for Project HASP were collected entirely by the 4080th Strategic Wing (SAC) generally using WU-2 aircraft capable of reaching very high altitudes. Occasional use was made of RB-52 aircraft to collect filter samples between 60° N and 90° N in the lower stratosphere. The six WU-2 aircraft especially modified for performing HASP sampling were equipped with filter samplers in the nose of the aircraft capable of exposing four filters sequentially during the course of a mission. The filter medium used was IPC-1478, developed by the Institute of Paper Chemistry. In June 1959 six WU-2 aircraft with nose samplers were equipped with a sampler capable of exposing six filters sequentially mounted in a hatchway in the fuselage. These six WU-2 aircraft could, consequently, collect ten samples. Other WU-2 aircraft with only the hatch sampler were also frequently used by 1960. An inflight calibration of both the nose and hatch samplers was performed during the last year of Project HASP to permit accurate calculation of air volumes collected. During Project HASP, between August 1957 and June 1960, a total of approximately 3700 filter samples was collected.

During the first three and one-half years of Project STARDUST, from mid-1961 to late 1964, the WU-2 continued to be the prime sampling vehicle. Particulates were sampled at low altitudes mainly below the tropopause with other aircraft, the RB-57C, the WB-50, the C-130 and the RB-52. The IPC-1478 filter medium was used in the samplers on all of these aircraft, and all of the samplers gave similar flow rates under similar flight conditions. The WU-2 was replaced as the prime sampling vehicle by the RB-57F, operated by the 58 Weather Reconnaissance Squadron, during the last few months of 1964 and the first few months of 1965. A V-1 hatch sampler, which is very similar in flow rate characteristics to the hatch sampler used on the WU-2, was employed on this aircraft. The RB-57F was used mainly at altitudes between 15 and 21 km, with most sampling at lower altitudes during 1965 to 1967 being performed by RB-57C aircraft.

On most sampling flights the filters were exposed for intervals of 30 to 60 minutes with the average time interval being approximately 45 minutes. The quantity of air sampled by a filter depended on several factors, the duration of exposure, the area of the filter, the altitude of collection, the velocity of the aircraft, and the ambient air temperature. Generally, for an exposure time of 45 minutes, samples collected at 12 or 13 km came from 2000 to 2500 SCM of air. Samples collected at 17 km came from 600 to 700 SCM, and samples collected at 19 or 20 km came from 300 to 400 SCM.

2.2 The Collection of Whole Air Samples

Beginning in August 1963 whole air samples were collected during most sampling missions flown for Project STARDUST. Compressed air samples were collected aboard WU-2, RB-57C and RB-57F aircraft using the sampling system described by Hagemann, et al[2]:

> "Whole air samples are obtained aboard aircraft by compressing air into nickel-plated spherical steel bottles about 900 in^3 in volume. Under normal operating conditions each bottle is compressed to about 3,000 psi. The bottles are not evacuated prior to sampling, hence they contain about one atmosphere of ground level air which is added to the sample. No correction is made for this background. The air sample is obtained from the compression section of the jet engine motor and fed to an oil lubricated compressor. The tubing between compressor and bottle is metal with some synthetic rubber connections. Normally four bottles are used to supply the CO_2 for a single sample. There is a positive outward pressure gradient from the bottles from the time of collection to laboratory processing."

All samples were shipped to Airco (Air Reduction Company, Inc., Murray Hill, New Jersey) by the units of the U.S. Air Force which collected them. At Airco a small aliquot of the air was drawn off into an evacuated 1,000 ml. flask before the sample was processed. The carbon dioxide after being separated from the remainder of the air sample using standard freeze-out techniques was shipped to Isotopes, Inc. with the aliquot of unprocessed air. There the carbon-14 content of the carbon dioxide sample and the carbon dioxide content of the aliquot of unprocessed air were measured.

2.3 The Sampling Plan of Project STARDUST

The STARDUST sampling plan was designed to obtain sampling within a meridional corridor. As in Project HASP, however, the ideal situation could not be obtained, and the actual sampling corridor consisted instead of a series of quasi-meridional segments. These segments were more or less contiguous between 75°N and 10°S, and during 1966 and 1967, between 75°N and 52°S. During 1962 to 1965, however, sampling between 18°S and 55°S was performed by aircraft based in Australia, so that the southernmost segments of the sampling corridor were far removed in longitude from the other segments. The various configurations of the STARDUST sampling corridor during 1961 to 1967 are summarized in Tables 1 to 7.

During the first seven months of the sampling program, June to December 1961, regular STARDUST sampling was confined to missions flown northward and southward from Laughlin A.F.B., Texas, and to "orbit" missions in the vicinity of Laughlin. Additional samples were obtained, however, from deployments of aircraft between Eielson A.F.B., Alaska and Laughlin during September 1961, from missions flown south from Hickam A.F.B., Hawaii during June 1961, from deployments of aircraft between Hickam and East Sale, Australia via Nandi, Fiji Islands, during October and December 1961, from "orbit" missions flown in the vicinity of East Sale, and from missions flown southward from East Sale during October and November 1961. All missions, except one flown north from Laughlin during September 1961, were at altitudes of 15 km or higher.

TABLE 1. Flight Tracks of Stardust Missions During 1961

	Northern Limit	Southern Limit	Altitudes (km)
Deployments between Eielson A.F.B., Alaska and Laughlin A.F.B., Texas:			
	63°40'N, 145°25'W	30°27'N, 101°25'W	20
Missions Northward from Laughlin A.F.B., Texas:			
	48°00'N, 97°25'W	27°44'N, 98°00'W	12,14, 15,17,18,19,20
Orbit Missions near Laughlin A.F.B., Texas:			
	30°00'N, 100°00'W	-----	17,18,19,20
Missions Southward from Laughlin A.F.B., Texas:			
	27°40'N, 99°07'W	14°55'N, 92°15'W	18,19,20
Missions Southward from Hickam A.F.B., Hawaii:			
	19°41'N, 158°00'W	7°00'N, 162°15'W	15,18,19,20
Deployments between Hickam A.F.B., Hawaii and East Sale R.A.A.F.B., Australia via Nandi:			
	20°00'N, 158°00'W	35°18'S, 149°11'E	20
Orbit Mission near East Sale R.A.A.F.B., Australia:			
	38°00'S, 147°00'E	-----	17,18,19,20
Missions Southward from East Sale R.A.A.F.B., Australia:			
	40°00'S, 147°30'E	60°00'S, 147°30'E	20

TABLE 2. Flight Tracks of STARDUST Missions During Early 1962

	Northern Limit	Southern Limit	Altitudes (km)
Missions to the North Pole by RB-52 Aircraft:			
	90°00'N, 120°00'W	60°00'N, 120°00'W	12
Missions Northward from Eielson A.F.B., Alaska:			
	70°11'N, 146°53'W	64°49'N, 147°30'W	12,14,15,17,18,19,20
Orbit Missions near Eielson A.F.B., Alaska:			
	64°04'N, 147°47'W	------	6,8,9,11,12,14,15, 17,18,19,20
Missions Southward from Eielson A.F.B., Alaska:			
	64°02'N, 145°42'W	48°57'N, 122°35'W	12,14,15,17,18,19,20
Missions Northward from Laughlin A.F.B., Texas:			
	48°40'N, 112°20'W	31°50'N, 100°40'W	12,14,15,17,18,19,20
Orbit Missions near Laughlin A.F.B., Texas:			
	30°00'N, 100°00'W	------	12,14,15,17,18,19,20
Missions Southward from Laughlin A.F.B., Texas:			
	29°37'N, 98°21'W	15°00'N, 92°25'W	17,18,19,20
Deployment from Laughlin A.F.B., Texas to Andersen A.F.B., Guam, via Hickam A.F.B. and Wake Island:			
	31°15'N, 104°00'W	14°06'N, 146°25'E	20
Missions Northward from Howard A.F.B., Canal Zone:			
	15°00'N, 82°30'W	08°46'N, 79°33'W	12,14,15,17,18,19,20
Orbit Missions near Howard A.F.B., Canal Zone:			
	08°00'N, 79°33'W	------	12,14,15,17,18,19,20
Missions Southward from Howard A.F.B., Canal Zone:			
	08°46'N, 79°33'W	10°00'S, 78°40'W	12,14,15,17,18,19,20
Deployments between Hickam A.F.B., Hawaii and East Sale R.A.A.F.B., Australia via Nandi:			
	20°00'N, 158°00'W	38°05'S, 147°10'E	20
Missions Southward from East Sale R.A.A.F.B., Australia:			
	40°00'S, 148°00'E	61°00'S, 147°30'E	20

TABLE 3. Flight Tracks of STARDUST Missions During August to November 1962

	Northern Limit	Southern Limit	Altitudes (km)
Missions Northward from Heyford, England:	73°40'N, 17°45'E	54°00'N, 02°14'W	18, 20
Vector Missions between Eielson A.F.B., Alaska and Laughlin A.F.B., Texas:	64°00'N, 146°00'W	31°00'N, 103°00'W	17, 18, 19
Vector Missions between Eielson A.F.B., Alaska and McClellan A.F.B. California:	65°00'N, 147°00'W	38°00'N, 121°00'W	usually 6 or 9
Deployment from Hickam A.F.B., Hawaii to Laverton, Australia via Nandi:	18°25'N, 158°25'W	37°20'S, 145°10'E	20
Missions Northward from Laverton, Australia:	15°00'S, 145°00'E	37°00'S, 145°00'E	17,18,19,20

TABLE 4. Flight Tracks of STARDUST Missions During Early 1963

	Northern Limit	Southern Limit	Altitudes(km)
Missions to the North Pole by RB-52 Aircraft:	90°00'N, 120°00'W	60°00'N, 120°00'W	12
Missions Northward from Eielson A.F.B., Alaska:	70°07'N, 143°25'W	64°35'N, 147°00'W	12,14,15,17, 18,19,20
Orbit Missions near Eielson A.F.B., Alaska:	65°00'N, 147°30'W	-----	6,8,9,11
Missions Southward from Eielson A.F.B., Alaska:	64°43'N, 147°00'W	48°55'N, 122°35'W	12,14,15,17, 18,19,20
Missions Northward from Laughlin A.F.B., Texas:	48°40'N, 112°20'W	31°20'N, 100°30'W	12,14,15,17, 18,19,20
A Mission Eastward from Laughlin A.F.B., Texas:	37°51'N, 100°00'W	30°25'N, 81°33'W	17, 20
Orbit Missions near Amarillo, Texas:	36°00'N, 102°20'W	-----	6,8,9,11
Missions Southward from Laughlin A.F.B., Texas:	31°20'N, 100°25'W	19°00'N, 95°11'W	17,18,19,20
Missions Northward from Albrook A.F.B., Canal Zone:	20°00'N, 86°25'W	09°00'N, 79°20'W	17,18,19,20
Missions Southward from Albrook A.F.B., Canal Zone:	09°10'N, 79°30'W	10°00'S, 79°00'W	17,18,19,20
Missions Northward from Laverton, Australia:	15°13'S, 145°00'E	37°00'S, 145°00'E	17,18,19,20

TABLE 5. Flight Tracks of STARDUST Missions During Late 1963

	Northern Limit	Southern Limit	Altitudes (km)
Missions to the North Pole by RB-52 Aircraft:	90°00'N, 120°00'W	60°00'N, 120°00'W	12
Missions Northward from Eielson A.F.B., Alaska:	70°10'N, 143°40'W	64°35'N, 147°00'W	12,13,15,17, 18,19,20
Orbit Missions near Eielson A.F.B., Alaska:	65°00'N, 147°30'W	-----	6,8,9,11
Missions Southward from Eielson A.F.B., Alaska:	64°43'N, 147°00'W	48°55'N, 122°35'W	12,13,15,17,18 19,20
Missions Northward from Davis-Monthan A.F.B., Arizona:	48°17'N, 122°37'W	32°07'N, 110°50'W	12,13,15,17,18 19,20
Orbit Missions near Amarillo, Texas:	36°00'N, 102°20'W	-----	6,8,9,11
Missions Southward from Davis-Monthan A.F.B., Arizona:	31°22'N, 100°27'W	20°00'N, 96°05'W	17,18,19,20
Missions Northward from Albrook A.F.B., Canal Zone:	20°00'N, 86°25'W	09°00'N, 79°20'W	17,18,19,20
Missions Southward from Albrook A.F.B., Canal Zone:	09°10'N, 79°30'W	10°00'S, 79°00'W	17,18,19,20
Missions Northward from Laverton, Australia:	15°00'S, 145°00'E	37°00'S, 145°00'E	17,18,19,20
Missions Southward from Laverton, Australia:	37°50'S, 144°45'E	50°00'S, 147°30'E	17,18,19,20

TABLE 6. Flight Tracks of STARDUST Missions During 1964 and 1965

	Northern Limit	Southern Limit	Altitudes (km)
Missions Northward from Eielson A.F.B., Alaska:			
	75°N, 143°W	67°N, 145°W	15,17,18,20
Missions between Eielson A.F.B., Alaska and Kirtland A.F.B., New Mexico:			
	67°N, 145°W	37°N, 108°W	12,13,15,17, 18,20
Missions between Kirtland A.F.B., New Mexico and Albrook A.F.B., Canal Zone:			
	36°N, 103°W	9°N, 80°W	17,18,19,20
Missions Southward from Albrook A.F.B., Canal Zone:			
	8°N, 80°W	10°S, 82°W	17,18,19,20
Missions Northward from East Sale R.A.A.F.B., Australia:			
	18°S, 147°E	36°S, 147°E	16,18,19,20
Missions Southward from East Sale R.A.A.F.B., Australia:			
	38°S, 147°E	55°S, 148°E	16,18,19,20

TABLE 7. Flight Tracks of STARDUST Missions During 1966 and 1967

	Northern Limit	Southern Limit	Altitudes (km)
Missions Northward from Eielson A.F.B., Alaska:	75°N, 143°W	64°N, 146°W	15,17,18,20
Missions between Eielson A.F.B., Alaska and Kirtland A.F.B., New Mexico:	64°N, 146°W	37°N, 108°W	12,13,15,17, 18,20
Missions between Kirtland A.F.B., New Mexico and Albrook A.F.B., Canal Zone:	36°N, 104°W	9°N, 80°W	15,17,18,20
Missions between Albrook A.F.B., Canal Zone and Mendoza Airfield, Argentina:	8°N, 80°W	30°S, 69°W	15,17,18,20
Missions Southward from Mendoza Airfield, Argentina:	35°S, 68°W	54°S, 68°W	15,17,18,20

The STARDUST sampling program was extended during January 1962 to cover more intensively a greater range of latitude and altitude. Missions were flown northward and southward from Eielson and Laughlin, and "orbit" missions were flown in the vicinity of each base. Missions were scheduled to cover each five thousand foot (1.5 km) level between 12 km and the maximum altitude 20 km, but only between 17 km and maximum altitude on missions southward from Laughlin, once every two weeks. Beginning in March 1962 a mission was flown to the North Pole at about 12 km by an RB-52 aircraft once each month, and "orbit" missions were flown at 6, 8, 9 and 11 km in the vicinity of Eielson by RB-57A aircraft once each week. Samples were received from missions flown northward and southward from Howard A.F.B., Canal Zone, and from "orbit" missions flown in the vicinity of that base during April, June and July 1962. Some samples were also received from a deployment of aircraft from Laughlin to Guam, via Hickam and Wake Island during June 1962, from a deployment of aircraft from Hickam to East Sale via Nandi, and from a mission flown south from East Sale during March 1962.

Regular STARDUST sampling during late August to November 1962 was limited to "vector" missions flown between Eielson and Laughlin by WU-2 air craft (generally at 17 km between 64°N and 40°N and at 17, 18 or 20 km between 40°N and 31°N), and between Eielson and McClellan A.F.B., California by WB-50 and C-130 aircraft (generally at 6 or 9 km). The low altitude flights continued into January 1963. Samples were also received from a series of missions flown northward from Heyford, England during August and September 1962, from a deployment of aircraft to Laverton, Australia during September 1962, and from missions flown northward from Laverton beginning in October 1962.

There was an expansion of STARDUST sampling in December 1962, and by January 1963 the regular STARDUST missions included flights northward and southward from Eielson, Laughlin and Albrook A.F.B. in the Canal Zone, and northward from Laverton. The complete set of missions was flown twice per month. One mission was flown eastward from Laughlin in December 1962 to sample variations of activity in the zonal direction. Beginning in March 1963 WU-2 aircraft were replaced by RB-57A aircraft at the lower altitudes, 12 and 13 km. Also beginning in March a series of missions at 6, 8, 9 and 11 km were flown in the vicinity of Eielson and of Amarillo, Texas once every two days. By June 1963 the frequency of these flights had been decreased to once per week at both Amarillo and Eielson. Also during early 1963 there was a resumption of monthly flights to the North Pole by RB-52 aircraft.

The home base of the 4080th Strategic Wing, S.A.C., which flew the STARDUST WU-2 missions, was changed from Laughlin to Davis-Monthan A.F.B., Arizona during early July 1963. While this move was being made several missions were flown along the WU-2 flight tracks at altitudes between 15 and about 19 km by RB-57D aircraft equipped with U-1 samplers. By the second half of July 1963 regular missions flown northward and southward from Davis-Monthan had replaced the missions formerly flown from Laughlin.

By September 1963 sampling was performed northward and southward from Eielson A.F.B., Alaska and northward from Davis-Monthan A.F.B., Arizona at 11 and 12 km by RB-57C aircraft and at 15, 17, 18, 19 and about 20 km by WU-2 aircraft. Southward from Davis-Monthan A.F.B., northward and southward from Albrook A.F.B., Canal Zone and northward and southward from Laverton R.A.A.F.B., Australia sampling was performed at 17, 18, 19 and about 20 km by

WU-2 aircraft. Sampling by the WU-2 aircraft was performed once every two weeks while sampling by RB-57C aircraft was performed once every four weeks. Once each month "orbit" missions were flown at 6, 8, 9 and 11 km in the vicinity of 65°N and 36°N by RB-57C, WB-47 or C-130 aircraft. At intervals of once each month or every two months sampling missions were flown from 60°N to 90°N by RB-52 aircraft.

During 1964 and 1965 there were numerous modifications of the sampling program. In February 1964 the number of altitudes at which WU-2 aircraft were flown northward and southward from Eielson A.F.B., and northward from Davis-Monthan A.F.B. were reduced to four, 15, 17, 18 and about 20 km. During the same month a series of "orbit" flights in the vicinity of 38°S at 9, 11, 12 and 14 km was begun and repeated once every two weeks. In May 1964 the "orbit" flights in the vicinity of 65°N and 36°N were discontinued. During the same month the flight plans were changed to replace most flights northward from Davis-Monthan and southward from Eielson by flights from Davis-Monthan to Eielson and back again. Similarly, southward flights from Davis-Monthan and northward flights from Albrook were largely replaced by flights from Davis-Monthan to Albrook and back again. In June 1964 the frequency of WU-2 sampling missions was decreased from once every two weeks to once every four weeks. The flights northward from Eielson at 11 and 12 km by RB-57C aircraft were eliminated after October 1964. In November 1964 WU-2 aircraft ceased flying the sampling missions at 15 km and higher altitudes from Davis-Monthan and Eielson. They were replaced by missions flown from Kirtland A.F.B. and Eielson by RB-57F aircraft. The RB-57F aircraft replaced the WU-2 aircraft in missions flown from Albrook A.F.B. in February 1965 and in missions flown from Laverton R.A.A.F.B.

in March 1965. Also during March 1965 the "orbit" missions at 38°S, flown by RB-57C and C-130 aircraft, were terminated. Some samples were received from missions flown during late November-early December 1963 and during April and June 1964 as part of Project SPRINGFIELD, but these were not used in Project STARDUST. These were flights by RB-57C and WB-47 aircraft searching for extrusions of stratospheric air into the troposphere.

The flights included in the STARDUST sampling program during late 1965 are portrayed in Figure 1.

During 1961 to 1965 sampling of the southern polar stratosphere for Project STARDUST was performed by aircraft stationed in Australia. By February 1966, however, these aircraft had left Australia, and sampling of the southern polar stratosphere thereafter was performed by aircraft deployed to Mendoza, Argentina. Under the final sampling plan, missions were flown at 15, 17, 18 and about 20 km from 75°N to 54°S latitude, and at 12 and 13 km between 64°N and 37°N latitude. Some samples collected in the upper troposphere and lower stratosphere (at about 5, 8 and 12 km) were received for analysis.

The flights included in the sampling program during mid-1966 are portrayed in Figure 2.

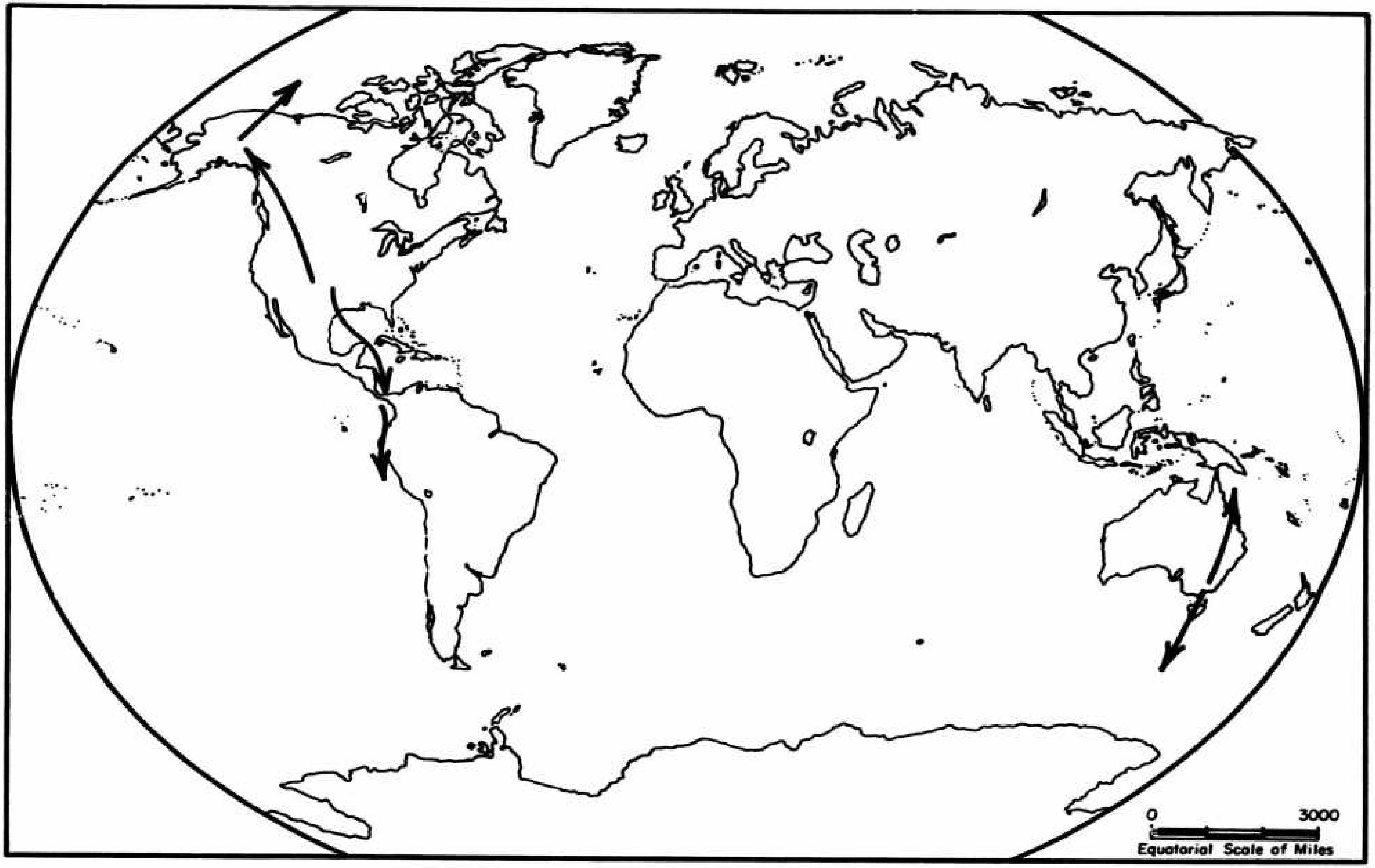

FIG 1. FLIGHT TRACKS OF STARDUST MISSIONS DURING LATE 1965

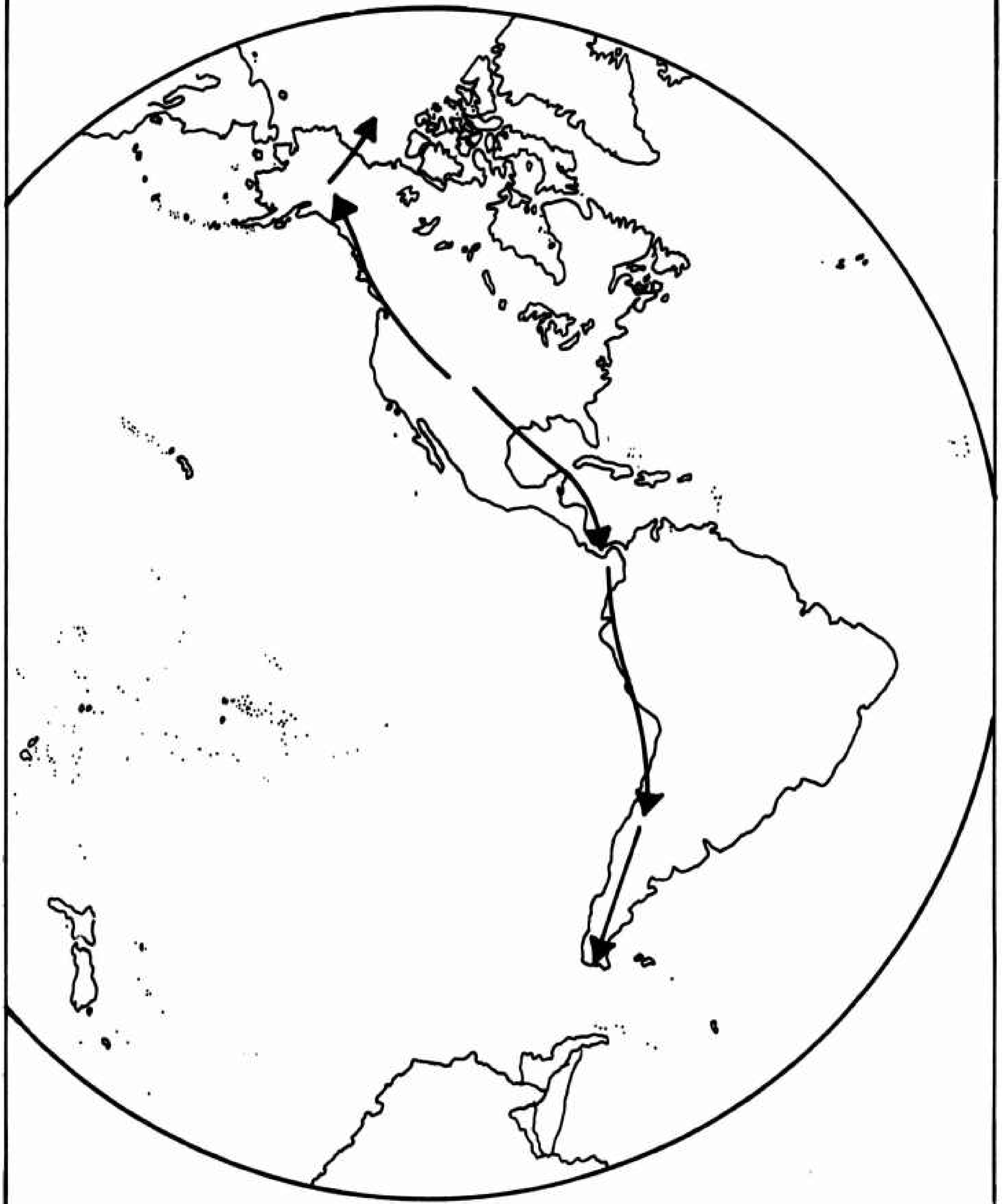

FIG. 2. FLIGHT TRACKS OF STARDUST MISSIONS DURING MID-1966

CHAPTER 3. THE ANALYSIS OF STARDUST FILTER SAMPLES

The filter samples of stratospheric and tropospheric debris collected during Project STARDUST were analyzed radiochemically to determine the concentrations of various artificial and natural radionuclides in the sampled air. The analytical plan had two main objectives: (1) the delineation of the distribution of radioactive debris in the stratosphere to permit the calculation of the stratospheric burden of such debris, and (2) the determination of the diffusion and circulation of air masses as measured with tracer radionuclides needed in the development of a model of atmospheric transport and fallout. In order to accomplish the second of these purposes a variety of radionuclides were analyzed. These included fission products, such as barium-140 and molybdenum-99, products of neutron activation, such as rhodium-102 and manganese-54, and naturally occurring radionuclides such as beryllium-7 and lead-210.

3.1 The Procedure for Analyzing Filter Samples

As soon as a filter sample was received at Isotopes, Inc., a disk, 2.86 cm in diameter, was cut from it and was beta-counted to determine the level of radioactivity which it contained. Usually this disk was counted several times over the course of a week or a month to determine the rate of decrease of the beta activity. The age of the youngest component of the fission product radioactivity in the filter could be estimated from this rate of decrease of activity, for the effective half-life of the total beta activity increased with time as the short-lived fission products decayed away. The level of activity and the age of the debris were used in determining which filters would be analyzed separately and which would be combined and in deciding which radionuclides should be measured. The radioactivity on disks from samples collected during and immediately following periods of testing of nuclear weapons in the atmosphere, most often from those with the highest total beta activities, was measured on the 400-channel gamma spectrometer. The gamma spectra revealed the relative concentrations of certain short-lived fission products, such as barium-140 and cerium-141. The presence of a significant concentration of these indicated that other short-lived fission products, such as molybdenum-99 and iodine-131, should be sought.

At times during the program certain gamma-emitting products of neutron activation, such as manganese-54, antimony-124 and yttrium-88, were monitored. During these periods samples, often selected from several stratospheric regions each month, were measured using the gamma spectrometer to obtain preliminary information on the stratospheric distribution of these activation products.

It was necessary during Project STARDUST to reach a compromise between the desire to obtain detailed information on the stratospheric concentrations of many nuclides and the necessity of keeping the cost of the analytical program within reasonable bounds. During Project HASP most samples had been analyzed individually for strontium-90, and usually for other components of radioactive debris from nuclear explosions. Only during the later stages of that program, when the radioactivity of some nuclides in single aliquots was too low to be measured were aliquots of several filters combined to provide enough radioactivity for the analyses of nuclides such as tungsten-185, rhodium-102 and phosphorus-32. During Project STARDUST, however, aliquots of several filters were combined for analysis more often than individual filters were analyzed. Generally, during the project the meridional gradients of nuclide concentrations within the stratosphere were not very steep. Consequently, insignificant error was introduced by combining filters collected at a single altitude over a range of 10° to 30° of latitude. The vertical concentration gradients frequently were rather steep; however, and since most sampling missions were made along horizontal flight tracks, filters collected at significantly different altitudes were generally not combined for analysis. On the other hand the concentration gradients were often too steep to permit combination when the sampling mission on a horizontal track passed through clouds of fresh radioactivity during the first week or so following a nuclear weapon test, or when it passed through the tropopause.

A major aim of Project STARDUST was the monitoring of the stratospheric burden of radioactive debris from nuclear explosions, and specifically of strontium-90. Thus almost all usable filter samples received were analyzed for strontium-90, and often for one or more additional fission products. Frequently two or three different samples were prepared using aliquots from the same set of filters, and these samples were generally analyzed for different groups of nuclides. Normally, however, strontium-90 was included in each group to provide a cross-check on the consistency of the data from the different samples.

During the course of the program the radiochemical analyses were divided into five basic sample groups. These were designated the "SF" group, in which mainly fission products were measured; the "SZ" group, in which were measured mainly products of neutron activation produced during the 1961 and 1962 series of nuclear weapon tests; the "SQ" series, in which plutonium isotopes and cesium-137 were measured; the "ST" series, which were analyzed mainly for products of cosmic ray activity and occasionally for other "tracer" nuclides; and the "SR" series, in which the lead-210 and polonium-210, both daughter products of radon, were measured. The specific nuclides included within the analytical scheme of each sample group varied from time to time during the program as the potential usefulness of the data obtained for them appeared to increase or diminish.

Some of the samples included in the "SF" group were analyzed only for strontium-90, but other fission products were also measured in most of these samples. Many of them were analyzed for cerium-144, strontium-89 and zirconium-95 in addition to strontium-90. During and immediately following periods of testing some analyses of molybdenum-99, barium-140 and cerium-141 and a few of iodine-131 and cesium-136 were also performed. During 1963 promethium-147, yttrium-91, and two products of neutron activation, yttrium-88 and silver-110, were analyzed in a set of samples collected at a variety of latitudes and altitudes.

When it became apparent that the 1961 Soviet test series had produced large amounts of products of neutron activation, the analytical scheme of the "SZ" group of samples was modified to include measurements of strontium-90, manganese-54, iron-55, cobalt-57, cobalt-58, cobalt-60, antimony-124, antimony-125, and thallium-204. Cadmium-109 was added to the analytical scheme following the injection of that radionuclide into the upper atmosphere by the 9 July 1962 rocket shot, STARFISH PRIME.

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During the first three years of the program a series of filters designated "SQ" samples were analyzed for strontium-90, cesium-137 and plutonium-239,240. Plutonium-238 was added to that group following its injection into the upper atmosphere in April 1964 by the burn up of a satellite containing a SNAP-9A power source. When the injected plutonium-238 began to reach the lower stratosphere in measurable quantities in early 1965, the frequency of measurement of "SQ" samples was greatly increased, and to effect economy cesium-137 was virtually eliminated from the scheme of analysis.

A number of samples, designated the "SX" group, were analyzed for some of the nuclides generally included in the "SZ" group and for some of those generally included in the "SQ" group. Typically the nuclides measured were strontium-90, manganese-54, cadmium-109, plutonium-238 and plutonium-239.

During the first few months of Project STARDUST most samples in the "ST" group were analyzed for rhodium-102 and tungsten-181 as well as strontium-90. Both the rhodium and tungsten had been injected into the atmosphere by the 1958 weapon tests. The "ST" group of samples was also analyzed for the cosmic ray product, beryllium-7, and for the radon daughter products, lead-210 and polonium-210. In late 1961 tungsten-181 was eliminated from the scheme of analysis of these samples. During early 1963 rhodium-102, lead-210 and polonium-210 were also eliminated, restricting the group after the addition of phosphorus-32, phosphorus-33 and sodium-22 to cosmic ray products. When the analysis of the daughter products of radon was begun again in 1965, the "SR" group, to be analyzed only for strontium-90, lead-210 and polonium-210, was created.

The actual radiochemical and radiometric procedures have changed considerably over the course of the project as nuclides have been added to or deleted from the scheme of analysis. Typical procedures used during Project HASP and Project STARDUST are described in the Final Report on Project HASP, DASA 1300 (Volume 1, pages 91-181), in the "Ninth Quarterly Report on Project STARDUST", DASA 1309 (pages 67-136), in the "Eleventh Progress Report on Project STARDUST", DASA 1821 (pages 15-26), and in the "Sixteenth Progress Report on Project STARDUST", DASA 1905 (pages 8-20).

As the first step in the analysis of all filter samples, the selected filter aliquots were cut up and placed in a beaker. Appropriate carriers and radioactive tracers for the determination of radiochemical yields were added to the beaker, and fuming nitric acid was added to digest the filter material. Perchloric acid and concentrated nitric acid were next added, and then hydrofluoric acid to bring any undissolved residue into solution. A few samples were found to contain excessive quantities of sulfate ion, and to form insoluble sulfates during the normal dissolution procedure. These sulfates were boiled with solid sodium carbonate as a final step in the dissolution to convert them to carbonates which could be dissolved in acids.

Procedures are given below for five representative samples. The first, representing the "SF" group, includes analyses for strontium-89, strontium-90, zirconium-95, cesium-137, barium-140, cerium-141 and cerium 144. The second, representing the "SZ" group, includes analyses for strontium-89, strontium-90, manganese-54, iron-55, iron-59, cadmium-109, cadmium-113m, cadmium-115m, cobalt-57, cobalt-58, cobalt-60, antimony-124, antimony-125 and thallium-204. The third, representing the "SQ" group, includes anlyses for strontium-90, cesium-137, plutonium-238 and plutonium-239,240. The fourth, representing the "SR" group, includes analyses for strontium-90, lead-210 and polonium-210. The fifth, representing the "ST" group, includes analyses for strontium-90, beryllium-7, sodium-22, phosphorus-32 and phosphorus-33.

3.2 The Analysis of Filter Samples for Fission Products

A typical set of analyses of fission products in filter samples includes measurement of strontium-89,90, zirconium-95, cesium-137, barium-140 and cerium-141,144. In this procedure the cesium-137 is separated from the other nuclides by precipitation of the latter as carbonates from a basic solution. It is purified by precipitation of cesium silicotungstate, and by extraction into sodium tetraphenyl boron - amyl acetate solution. It is counted as the chloroplatinate.

Zirconium-95 is separated from the sample solution by precipitation as zirconium mandelate; it is extracted into thenoyltrifluoroacetone (TTA) in xylene to separate it from its niobium-95 daughter; it is purified by precipitation as the hydroxide; and is converted again to the mandelate for counting.

Cerium-141,144 is precipitated as $Ce(OH)_3$ to separate it from the sample solution. It is purified by extraction into methyl isobutyl ketone, and is counted as cerium oxalate.

Barium-140 is isolated by precipitation as $BaCrO_4$. It is purified by precipitation as $BaCl_2$ in HCl-ethyl ether reagent, and by a ferric hydroxide scavenge. It is converted again into barium chromate for counting.

Yttrium-90 is precipitated as $Y(OH)_3$ at a pH of 7.6 to separate it from its strontium-90 parent. It is purified by extraction into tri-butyl-phosphate, and is counted as yttrium oxalate. From the yttrium-90, the strontium-90, which is in equilibrium, can be calculated. Then the strontium-89 can be determined by subtracting the strontium-90 and the yttrium-90 count from the total strontium count.

A. Sequential Separation Procedure

1. To an appropriate aliquot of the sample contained in a 150-ml beaker add 1 ml of standardized strontium carrier (20 mg Sr/ml), 1 ml of standardized zirconium carrier (5 mg Zr/ml), 1 ml of standardized barium carrier (20 mg Ba/ml), 1 ml of standardized cerium carrier (20 mg Ce/ml), and 1 ml of standardized cesium carrier (20 mg Cs/ml). (Note 1)

2. Add 1 ml of 5M $NH_2OH \cdot HCl$ dropwise from a pipette and stir carefully during the addition. (Note 2) Evaporate until a volume of approximately 5 ml is obtained.

3. Transfer the solution to a 40-ml centrifuge tube with water. Raise the pH to 8.5 - 9.0 by the dropwise addition of 50% NaOH. Digest the solution in a hot water bath for 10 minutes and add 10 ml of saturated Na_2CO_3 solution. Stir and allow the precipitate to coagulate for 10 minutes.

4. Cool to room temperature, centrifuge and decant the supernate into a 100-ml beaker. The supernate is to be saved for cesium purification.

5. Dissolve the precipitate from step 4 with 5 ml of 6M HNO_3 and boil over a flame for several minutes to remove all the CO_2.

6. Place the solution in a hot water bath, add via pipette 4 ml of 16% Mandelic acid solution with stirring, and digest the precipitate for 20 minutes. (Note 3)

7. Cool, centrifuge, decant the supernate into a 40-ml centrifuge tube and reserve the precipitate for zirconium-95 purification.

8. To the supernate from step 7 add 50% NaOH until the pH is greater than 8.5, place in a hot water bath, add 10 ml of saturated Na_2CO_3 solution and digest for 10 minutes.

9. Cool, centrifuge and discard the supernate. Dissolve the precipitate in 5 ml of 6M HNO_3 and boil the solution over a flame for several minutes to remove all the CO_2.

10. Cool, add concentrated NH_4OH until $Ce(OH)_3$ precipitates, and digest in a hot water bath for 5 minutes. Cool, centrifuge, decant the supernate into a 40-ml centrifuge tube and reserve the supernate.

11. Dissolve the $Ce(OH)_3$ precipitate from step 10 in 5 ml of 6M HNO_3 and heat the solution over a flame until the solution is clear. Cool, add concentrated NH_4OH until $Ce(OH)_3$ precipitates, and digest the precipitate in a hot water bath for 5 minutes.

12. Cool, centrifuge and combine the supernate with that from step 10. Reserve the precipitate for cerium-144 purification.

13. To the combined supernates from step 12, add 5 mg of Fe^{+3} carrier. To the resultant $Fe(OH)_3$ precipitate add concentrated HCl dropwise with stirring until the solution clears and then make basic with concentrated NH_4OH until the precipitate reappears (Note 4). Centrifuge, transfer the supernate to a 40-ml centrifuge tube and discard the precipitate.

14. To the supernate from step 13, add sufficient concentrated NH_4OH to bring the pH >8.5. Heat the solution in a hot water bath and add 10 ml of saturated Na_2CO_3 with stirring. Digest until the precipitate settles out, cool, centrifuge and discard the supernate.

15. To the carbonate precipitate from step 14, add 5 ml of 6M HNO_3 and boil over a flame for about 2 minutes to remove all the CO_2.

16. Cool the solution to room temperature and add 4-5 drops of Meta-cresol purple indicator. Add 6M NH_4OH until a color change from yellow to violet occurs.

17. Add 5 ml of "barium buffer solution", heat nearly to boiling (Note 5), add 1 ml of 1.5M $NaCrO_4$ (via pipette) with stirring and digest in a hot water bath until the $BaCrO_4$ settles out.

18. Cool, centrifuge and decant the supernate into a 40-ml centrifuge tube. Reserve the $BaCrO_4$ precipitate for barium-140 purification.

19. To the supernate from step 18, add 5 ml of concentrated NH_4OH with stirring (pH >8.5), heat in a hot water bath for several minutes and add 10 ml of saturated Na_2CO_3 with stirring.

20. Digest in a hot water bath for 15 minutes, cool, centrifuge and discard the supernate.

21. Dissolve the precipitate in 5 ml of concentrated HNO_3 and boil over a flame to remove all CO_2. Cool in an ice bath for 5 minutes (Note 6), add two to three drops of fuming nitric acid, centrifuge and discard the supernate.

22. Dissolve the precipitate in 10 ml of water, add 5 mg of Fe^{+3} carrier, and make basic with concentrated NH_4OH until $Fe(OH)_3$ precipitates. Centrifuge and transfer the supernate to a 40-ml centrifuge tube. Discard the precipitate.

23. To the supernate from step 22 add 5 mg of Fe^{+3} carrier. To the resultant $Fe(OH)_3$ precipitate add concentrated HCl dropwise with stirring until the solution clears, and then make basic with concentrated NH_4OH until the precipitate reappears. Centrifuge, transfer the supernate to a 40-ml centrifuge tube and discard the precipitate. Record the date and time of this last $Fe(OH)_3$ scavenge. (This represents starting time of Y-90 growth).

24. Acidify the supernate from step 23 with concentrated HCl and add, via pipette, 3 ml of standardized yttrium carrier (10 mg Y/3ml). Set aside for yttrium-strontium separation.

25. To the solution which has been set aside for at least 3 days, add 7-8 drops of meta-cresol purple indicator; stir well and make basic with 6M NH_4OH until one drop causes a color change from yellow to purple (pH 7.6).

26. Digest in a hot water bath for about 10 minutes. Cool to room temperature. Immediately centrifuge the $Y(OH)_3$ precipitate and decant the supernate into a 40-ml centrifuge tube. Reserve the supernate. Record the time and date of "milking", i.e. when the supernate is decanted.

27. Wash the $Y(OH)_3$ precipitate with 3 ml of water, using a stirring rod to slurry the precipitate. Centrifuge and combine the supernate with the "strontium supernate" from step 26.

Note 1. If the analysis of yttrium 88, 91, of promethium-147, or of silver-110 is required, the following insertions may be made:

A. - Analysis of Y-88,91:	Step 1.	Add 1 ml of standardized yttrium carrier (10 mg Y per ml). The Y-88,91 is removed as $Y(OH)_3$ along with the $Ce(OH)_3$. The yttrium is separated from cerium by a T.B.P. extraction.
B. - Analysis of Y-88,91 and Pm-147:	Step 1.	Add 3 ml of standardized yttrium carrier (10 mg Y in 3 ml) and 1 ml of standardized promethium carrier (20 mg Nd,Sm/ml). The combined hydroxides are separated by ion exchange chromatography.

C. - Analysis of Ag-110: Step 1. After all the carriers have been added; the sample is evaporated to fumes of $HClO_4$. Add 5 ml of 6M HNO_3, 1 ml of standardized Ag carrier (20 mg/ml) and transfer sample to a 40-ml centrifuge tube. Add 1 ml of 1N HCl to precipitate AgCl. Centrifuge, retain the supernate for the sequential separation and reserve the AgCl for Ag-110 purification.

Note 2. Effervescence should occur at this point, but if it does not, carefully add 3 to 4 drops of concentrated HNO_3 and heat for at least 20 minutes.

Note 3. Zirconium mandelate is slow in forming.

Note 4. If the precipitate is slow in forming, place in a hot water bath for several minutes.

Note 5. The pH should be 5.5.

Note 6. $Sr(NO_3)_2$ will precipitate. The fuming nitric acid step separates strontium from calcium. Care should be exercised in handling fuming nitric acid. Do all work under a fume hood.

B. Yttrium-90 Purification Procedure

1. Dissolve the $Y(OH)_3$ from step 27 of the Sequential Separation Procedure in 25 ml of 14N HNO_3 and transfer the sample to a cylindrical separatory funnel containing 10 ml of equilibrated Tri-Butyl-Phosphate.

2. Rinse the centrifuge tube with 25 ml of 14N HNO_3 and add the rinse to the separatory funnel.

3. Shake for 15 minutes. Separate and discard the lower phase. Wash the organic phase twice by shaking for 5 minutes with 50 ml of 14N HNO_3 and discard the washings.

4. Strip the yttrium out of the organic phase with three 10-ml aliquots of water, shaking for 1 minute each. Discard the organic phase.

5. Add concentrated NH_4OH to the aqueous solution until the pH >8.5 and digest the resultant $Y(OH)_3$ precipitate in a hot water bath for five minutes. Cool, centrifuge and discard the supernate.

6. Wash the $Y(OH)_3$ precipitate with 3 ml of water, using a stirring rod to slurry the precipitate. Centrifuge and discard the supernate.

7. Dissolve the $Y(OH)_3$ precipitate in 2 ml of 1M HCl. With a pipette add 1.5 ml of 6M HNO_3, heat to boiling and add 5 ml of saturated $(NH_4)_2C_2O_4 \cdot H_2O$ solution. Stir for several minutes and gradually add 10 ml more of saturated $(NH_4)_2C_2O_4 \cdot H_2O$.

8. Digest in a hot water bath for 20 minutes with intermittent stirring. Cool to room temperature and filter the $Y_2(C_2O_4)_3 \cdot 9\ H_2O$ onto a previously washed and weighed Whatman #42 filter disk using water and finally anhydrous "Anhydrol" as transfer agents. Dry in an oven at 105°C for 30 minutes and cool to room temperature in a desiccator. Weigh, record the chemical yield of $Y_2(C_2O_4)_3 \cdot 9\ H_2O$ and mount.

C. Strontium-89 Purification Procedure

1. Heat the combined "strontium supernates" from steps 26 and 27 of the sequential separation procedure almost to boiling in a hot water bath and adjust the $pH > 8.5$ with concentrated NH_4OH (check pH with pH paper).

2. Add 10 ml of saturated Na_2CO_3 solution and allow to digest in a hot water bath until the precipitate settles. Remove from the water bath and allow the mixture to cool to room temperature.

3. Centrifuge and discard the supernate. Add 10 ml of water and slurry with a stirring rod.

4. Filter the precipitate onto a previously weighed Whatman #42 filter disk using water and finally acetone as transfer agents. Dry in an oven at 100°C for 30 minutes and then cool to room temperature in a desiccator. Weigh and record the chemical yield of $SrCO_3$, and mount on a planchet for strontium-89 assay.

D. Zirconium-95 Purification Procedure

1. Dissolve the zirconium mandelate precipitate from step 7 of the sequential separation procedure in 5 ml of 6N NaOH, adjust the pH to 1.0 with 6N HCl and slowly add 1 ml of 30% H_2O_2. Heat over a flame until effervescence starts, then adjust the pH to 10.0 with 6N NaOH and heat over a flame until the effervescence stops. Adjust the pH to 9.5 and digest in a hot water bath. Cool, centrifuge, decant and discard the supernate.

2. Dissolve the precipitate in 10 ml of 6N HCl, add 10 drops of 30% H_2O_2 and boil until the effervescence stops. Transfer the solution to a 125-ml separatory funnel with a minimum of 6N HCl. Add 10 ml of 0.5M thenoyltrifluoroacetone (TTA) in xylene and shake for 15 minutes.

3. Separate the lower (aqueous) phase and record this time as the separation time of Nb from Zr. Wash the TTA phase with 10 ml of 3N HCl containing 3 drops of 30% H_2O_2 for 5 minutes.

4. Repeat the wash of the TTA phase with 10 ml 3N HCl containing 3 drops of 30% H_2O_2 for 5 minutes.

5. Add 10 ml of 0.25N HF-0.25N HNO_3 solution and shake for 5 minutes. Collect the Zr strip solution (lower) in a 40-ml glass centrifuge tube.

6. Repeat step 5 combining the strip solutions.

7. Add 5 ml of 3N NaOH to precipitate the $Zr(OH)_4$. Digest, centrifuge and discard the supernate.

8. Dissolve the hydroxide precipitate in 5 ml 3N HCl, dilute to 15 ml with water and reprecipitate the hydroxide with 3N NaOH. Digest, centrifuge and discard the supernate.

9. Dissolve the $Zr(OH)_4$ in 2 ml of concentrated HCl (added via pipette); add 1 ml of water and heat until a clear solution is obtained. Add 4 ml of 16% mandelic acid (via pipette) and heat in a hot water bath for 20 minutes with occasional stirring (Note 1).

10. Add a drop of "Aerosol" solution, cool, centrifuge and discard the supernate. Wash the precipitates with 5 ml of a 2% HCl-5% mandelic acid solution. Centrifuge and discard the supernate.

11. Slurry the Zr mandelate precipitate with "anhydrol" and filter through a previously washed, dried and weighed Whatman #42 filter disk. Air dry on filtering apparatus for several minutes, oven dry at 110°C for 10 minutes and cool in a desiccator. Weigh, record chemical yield of Zr mandelate and mount on a brass planchet.

Note 1. The zirconium mandelate forms slowly, but is quantitative in about 20 minutes.

E. Cesium-137 Purification Procedure

1. To the cesium fraction from step 4 of the Sequential Radiochemical Separation Procedure add 3 drops of meta cresol purple indicator and carefully neutralize with concentrated HCl; add an excess of 15 ml of 6M HCl.

2. Evaporate the solution on a hot plate until "salts" just begin to appear; cool to room temperature and then dilute with enough water to obtain a clear solution.

3. Transfer 30 ml of the solution to a 40-ml centrifuge tube and add 5 ml of 0.13M silicotungstic acid and 3 drops of "Aerosol" solution.

4. Digest the precipitate in a hot water bath for 10 minutes, centrifuge the cesium silicotungstate and discard the supernate.

5. Transfer, with washings, the remaining solution from step 2 to the centrifuge tube containing the previously precipitated cesium silicotungstate; proceed as in steps 3 and 4.

6. To the precipitate add 3 ml of 6M NaOH and heat over a flame until the precipitate is completely dissolved.

7. Add 20 ml of 6M HCl and digest in a hot water bath for 10 minutes; centrifuge the silica and tungstic acid and transfer the supernate to a clean 100-ml beaker.

8. Wash the precipitate twice with two 5 ml portions of 6M HCl and combine the washings in the beaker; discard the precipitate.

9. To the solution add 3 drops of meta cresol purple indicator and neutralize with 50% NaOH; transfer to a 125-ml separatory funnel containing 10 ml of citrate buffer solution (1M $Na_3C_6H_5O_{17}$, 0.5M HNO_3).

10. Add 25 ml of 0.05M sodium tetraphenyl boron-amylacetate solution to the funnel, shake for 30 seconds and allow to stand for 3 minutes. (Note 1).

11. Withdraw the aqueous (lower) phase and collect in another 125-ml separatory funnel; extract again with 15 ml of 0.05M sodium tetraphenyl boron-amyl acetate solution.

12. Withdraw and discard the aqueous phase and combine the two portions of the organic phase in a separatory funnel.

13. Strip the cesium from the organic phase by extracting twice with two 10-ml portions of 3M HCl; combine the strips in a clean 100-ml beaker and heat on a hot plate for 30 minutes to distill off all traces of the amyl acetate.

14. Remove the solution from the hot plate. Add 3 drops of meta cresol purple indicator and neutralize with 6M NaOH. Cool in an ice bath. Add 2 ml of 10% chloroplatinic acid very slowly, from a pipette, and stir vigorously during the addition. Allow to stand for 30 minutes in an ice bath.

15. Centrifuge, and discard the supernate. Mix the precipitate with 10 ml of cold anhydrol.

16. Filter the precipitate onto a previously washed and weighed glass fiber filter disk without applying suction (Note 2). Wash the precipitate with two 5 ml portions of cold water and 5 ml of cold anhydrous anhydrol applying suction each time. Oven dry at 100°C for 10 minutes. Cool in a desiccator, weigh as the cesium chloroplatinate for chemical yield and mount on a nylon planchet for counting.

    Note 1. An insoluble precipitate (cesium tetraphenyl boron) will form at the bottom of the amyl-acetate layer, obscuring the interface. The aqueous phase may be readily separated by draining until the white precipitate just begins to appear in the bore of the stopcock.

    Note 2. The Cs_2PtCl_6 precipitate is a very fine powder and filtering without suction deposits the precipitate evenly on the surface of the filter disk with no tendency for seepage around the edge of the filtering column.

F. Barium-140 Purification Procedure

1. Wash the $BaCrO_4$ precipitate from step 18 of the Sequential Radiochemical Separation Procedure with 10 ml of hot water, centrifuge and discard the wash. Dissolve the precipitate in 1 ml of 6M HCl. Cool in an ice bath.

2. Add 15 ml of cold HCl-ethyl ether reagent (1:4 ether - concentrated HCl) and stir for 1 to 2 minutes. Place in ice bath for 10 minutes. Centrifuge and decant the supernate. (Note: No open flames should be allowed in the laboratory during the preparation of the HCl-ethyl ether reagent; all handling of the reagent should be done in the fume hood).

3. Dissolve the $BaCl_2$ in a minimum of water and repeat step 2.

4. Dissolve the $BaCl_2$ precipitate in 5 ml of water and add 10 drops of Fe carrier; precipitate $Fe(OH)_3$ with 2 ml of 6M NH_4OH. Centrifuge and discard the precipitate; record the time of the $Fe(OH)_3$ scavenging as the separation time for subsequent lanthanum-140 growth calculations.

5. Neutralize the supernate with concentrated HNO_3 and add, via pipettes, 1 ml of 6M HOAc and 4 ml of 3M NH_4OAc. Heat the solution nearly to boiling and add 1 ml of 15M Na_2CrO_4 dropwise with stirring.

6. Digest in a hot water bath until the $BaCrO_4$ settles to the bottom of the tube. Cool and filter through a previously washed, dried, and weighed Whatman No. 42 filter disk. Wash with three 5-ml aliquots of water and then with two 5-ml aliquots of Anhydrol and finally with acetone.

7. Air dry for several minutes in a desiccator; weigh as $BaCrO_4$ for chemical yield determination and mount on a nylon planchet for counting.

G. Cerium 141,144 Purification Procedure

1. Transfer the cerium fraction from step 12 of the Sequential Separation Procedure to a separatory funnel containing 50 ml of freshly equilibrated methyl isobutyl ketone (Note 1).

2. Wash the glass storage vial with 6.5 ml of concentrated HNO_3, 2ml of 6M $NaBrO_3$ and 4.5 ml of water, add washings to the separatory funnel and shake for 15 - 30 seconds.

3. Withdraw the aqueous phase (bottom phase) and wash the methyl isobutyl ketone phase twice with 10 ml of 9M HNO_3 containing a few drops of 2M $NaBrO_3$ (Notes 1 and 2).

4. Back-extract the cerium by shaking the methyl isobutyl ketone phase with 5 ml of water containing 3 drops of hydrogen peroxide (H_2O_2) (Notes 1 and 3).

5. Withdraw the aqueous phase into a clean 40-ml centrifuge tube and neutralize by adding concentrated NH_4OH (3 - 5 ml) until a precipitate just appears, and accidify with 1.5 ml of 6 M HNO_3.

6. Dilute the solution to a volume of 15 ml with water and heat to boiling; add 5ml of saturated $(NH_4)_2C_2O_4$, stir for several minutes and add gradually 10 ml more of saturated $(NH_4)_2C_2O_4$.

7. Digest the precipitate in a hot water bath for about 10 minutes, cool to room temperature and filter the $Ce_2(C_2O_4)_3 \cdot 9\,H_2O$ on a Whatman No. 42 filter disk. Wash three times with 5 ml portions of water and three times with 5 ml portions of acetone. Dry in an oven at 110°C for 20 minutes and then cool to room temperature in a desiccator, mount and count.

8. The chemical yield of the cerium mount is determined, upon completion of counting, by dismounting the filter disk and pliofilm and igniting at 850°C for 1 hour in a previously weighed (brought to constant weight) porcelain crucible. Weigh and record chemical yield of CeO_2.

Note 1. The equilibration of sufficient methyl isobutyl ketone for use with ten samples is performed in the following manner: to 400 ml of methyl isobutyl keton, add 400 ml of 9 M HNO_3 containing 16 ml of 2 M sodium bromate ($NaBrO_3$) and shake or stir for five minutes.

CAUTION: In extractions of strong HNO_3 solutions (6 to 12 M) with methyl isobutyl ketone, considerable amounts of HNO_3 pass into the organic phase. It has been observed that such solutions of HNO_3 in methyl isobutyl ketone are unstable and will undergo a vigorous reaction after standing for a few hours. The methyl isobutyl ketone phases remaining after back-extraction with 5 ml of water were observed to react similarly but only after standing for about 3 days. It is recommended, therefore, that the methyl isobutyl ketone not be equilibrated with HNO_3 until just before use and that it be washed thoroughly with water (three times with an equal volume) soon after use. It is also recommended that HNO_3 solutions which have been in contact with methyl isobutyl ketone be neutralized with NH_4OH before storing or discarding.

Note 2. Combine the aqueous phase and washings and neutralize with NH_4OH before discarding.

Note 3. Wash the methyl isobutyl ketone three times with 50 ml of water before discarding. Also neutralize washings before discarding.

H. Radioassay Procedures

The strontium-89 activity is calculated based on beta measurements of the $SrCO_3$ mount. The $SrCO_3$ is first counted shortly after the separation of the yttrium-90, when most of the activity present is attributable to strontium-89 and strontium-90. It is then counted several times during the course of the following several weeks. The beta activity increases during the first week as yttrium-90 grows into equilibrium with the strontium-90 present. During subsequent weeks the beta activity decreases, due mainly to the decay of the strontium-89 component. The data points for each sample are fitted to a series of standard curves for different mixtures of strontium-89 and strontium-90, and the activities of both nuclides are estimated.

High zirconium-95 activities are determined by gamma-ray spectrometry and lower activities are determined by beta counting. The 0.76 Mev photopeak is measured using a cylindrical NaI (Tl) crystal, 3 inches deep by 3 inches diameter, coupled with a multi-channel pulse height analyzer. The area under the peak obtained from the sample is compared with the equivalent areas for radiochemically pure standard samples, and the activity is calculated. The samples are counted several times over the course of a few weeks to monitor the ingrowth of niobium-95 and the subsequent decay of the zirconium-95 - niobium-95 mixture. When too little activity is present for accurate resolution of the gamma peak, the sample may be measured by beta-assay of the 0.40 Mev beta ray. The radiochemical purity of the sample is checked when it is beta counted by counting with and without an absorber of known thickness, and comparing the ratio of these counts against the expected ratio.

Cesium-137 is radioassayed by beta counting the 0.61 Mev beta ray. The ratios of counting rates obtained with and without absorbers of known thickness are used to check the radiochemical purity of the samples.

Barium-140 is usually measured by beta counting, with monitoring of the ingrowth of the lanthanum-140 daughter, and of the subsequent decay of the barium-140 - lanthanum-140 mixture. The 1.6 Mev photopeak of lanthanum-140 may be used for gamma assay of barium-140 samples when the activities are high and sufficient time has passed since the purification of the barium to allow the lanthanum-140 daughter to grow back in.

Cerium-141 and cerium-144 may be measured by gamma spectrometry when the activities are high. The 0.145 Mev photopeak of cerium-141 and the 0.134 photopeak of cerium-144 are measured using a three-inch by three-inch NaI(Tl) crystal coupled to a multichannel analyzer. Most STARDUST samples were beta counted, with and without absorbers, to distinguish between the 0.44 Mev beta of cerium-141 and the 0.30 Mev beta of cerium-144.

3.3 The Analysis of Filter Samples for Products of Neutron Activation

During 1962 and 1963, a number of filter samples were analyzed for manganese-54, iron-55,59, strontium-89,90, cadmium-109,113m,115m, antimony-124,125, cobalt 57,58,60 and thallium-204. The analytical procedures which are given below were used for those measurements.

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A. Sequential Separation Procedure

After the addition of carriers thallium is precipitated as thallium iodide. Antimony and cadmium are precipitated and separated as sulfides; the cadmium at pH 1.5. After the separation of manganese as the dioxide, cobalt is precipitated as cobaltinitrite. The supernate from the cobalt separation is treated with ammonium hydroxide separating iron as ferric hydroxide. The supernate contains the strontium activity.

1. From the sample contained in a 25-ml volumetric flask (Note 1), remove two 2-ml aliquots for elemental iron spectrophotometric determination.

2. Pipette 20 ml of the sample solution into a 40-ml centrifuge tube and add 20 mg each of thallium, strontium, iron, cadmium, cobalt and manganese carriers.

3. Add 5 ml of 6% H_2SO_3 and 3 or 4 crystals of NaI. Stir and allow to stand at room temperature for 10 minutes. Centrifuge, decant the supernate to a clean 100-ml beaker and retain the precipitate for thallium-204 purification.

4. Heat the supernate from step 3 on a hot plate for approximately 3/4 hour to expel SO_2, then bubble H_2S through the solution for two minutes. Cool to room temperature, transfer to a 40-ml centrifuge tube, centrifuge and decant the supernate to another 40-ml centrifuge tube. Reserve the antimony sulfide precipitate for antimony-124, 125 purification.

5. Evaporate the supernate from step 4 almost to dryness, dilute with water to a pH of 1.5 (approximately 15 ml of water), heat and bubble H_2S gas through the solution for 2 minutes. If a yellow precipitate does not form, increase the volume of water. Cool, centrifuge and decant the supernate.

6. Dissolve the cadmium sulfide precipitate in 5 ml of 6M HCl, boiling until the precipitate is completely dissolved. Adjust the volume to approximately 15 ml with water and add concentrated NH_4OH to pH 9. Heat in a hot water bath for 5 minutes and add 10 ml of saturated Na_2CO_3. Digest in a hot water bath for 20 minutes, cool, centrifuge and transfer the supernate to a 40-ml centrifuge tube. Reserve the supernate for cadmium-109, 113m and 115m purification.

7. Dissolve the carbonate precipitate from step 6 in 5 ml of 6M HNO_3, boil to expel CO_2 and combine with the supernate from step 5 in the 150-ml beaker (Note 2).

8. Evaporate the combined supernate from steps 5 and 7 until "salting out" occurs. Add 20 ml of 9N HNO_3 and again evaporate until salts appear. Add 20 ml of 9M HNO_3 and repeat the evaporation.

9. Add 5 ml of concentrated HNO_3, dilute to 100 ml with water, heat almost to boiling, and add 2 ml of saturated $NaBrO_3$ with stirring. Digest for approximately 20 minutes or until the MnO_2 precipitate settles.

10. Cool to room temperature, transfer to a 40-ml centrifuge tube and centrifuge in portions. Combine the supernates in a 150-ml beaker. Retain the precipitate for manganese-54 purification.

11. Evaporate the combined supernates almost to dryness and transfer the solution to a clean 40-ml centrifuge tube with approximately 25 ml of water. Adjust the pH to 9.0 with 10M KOH, place the tube into a hot water bath for several minutes and add 10 ml of saturated Na_2CO_3. Digest the precipitate for 15-20 minutes, cool, centrifuge and discard the supernate.

12. Dissolve the precipitate with 6 to 10 drops of concentrated HNO_3 and 2 ml of 6N CH_3COOH. Boil over a flame for 2 minutes to remove the CO_2, and dilute to 20 ml with water. (If the solution does not turn clear add 2 to 3 drops of H_2O_2 and boil slowly.) Add 6 ml of 3N CH_3COOH saturated with KNO_3 and digest in an ice bath for 30 minutes. Centrifuge and decant the supernate into a clean 100-ml beaker. Wash the $K_3Co(NO_2)_6$ precipitate with 10 ml of water, centrifuge and discard the wash. Retain the precipitate for cobalt-57, 58, 60 purification.

13. Add concentrated HNO_3, cautiously with stirring, until effervescence stops and the solution turns green. Then evaporate to a small volume. Transfer the solution to a clean 40-ml centrifuge tube with water, adjust the pH to 9.0 using concentrated NH_4OH and heat in a hot water bath for five minutes. Add 10 ml of saturated Na_2CO_3, digest the precipitate for 15-20 minutes in the hot water bath, cool, centrifuge and discard the supernate.

14. Dissolve the precipitate from step 13 with 5 ml of 6N HNO_3 and boil over a flame for two minutes to remove all of the CO_2. Cool, dilute to 10 ml with water and add sufficient concentrated NH_4OH to precipitate $Fe(OH)_3$. Digest the precipitate in a hot water bath for ten minutes, cool, centrifuge and decant the supernate into a clean 40-ml centrifuge tube. Retain the $Fe(OH)_3$ precipitate for iron-55, 59 purification.

15. To the supernate from step 13 add concentrated NH_4OH until pH 9.0 is obtained. Heat in a hot water bath for five minutes, add 10 ml of saturated Na_2CO_3 solution, digest for 10 minutes in a hot water bath, cool, centrifuge and discard the supernate. Retain the $SrCO_3$ precipitate for strontium-89, 90 purification as described in Section 3.2.

Note 1. On standing, an insoluble antimony precipitate may appear (antimony carrier is added prior to ashing). This precipitate may be dissolved by the addition of concentrated HCl and heating. The precipitate must be dissolved prior to the removal of the aliquot for elemental iron determination.

Note 2. Some strontium is entrained in the cadmium sulfide. Steps 6 and 7 are designed to separate this strontium.

B. Manganese-54 Purification Procedure

The manganese dioxide precipitate from the Sequential Separation Procedure is dissolved in concentrated HCl, and manganese carbonate is precipitated. After oxidation to MnO_4^{-2} with $NaBiO_3$ and a ferric hydroxide scavenge, the manganese is finally precipitated and radioassayed as MnO_2. Chemical yield recovery is ascertained by ignition to Mn_3O_4 at 850°C.

The step-by-step procedure is as follows:

1. Dissolve the MnO_2 precipitate from step 10 of the Sequential Separation Procedure in 4 ml of concentrated HCl and evaporate the solution to near dryness. If the solution is not clear during evaporation, again add 4 ml of concentrated HCl and evaporate until the solution turns light green.

2. Dilute the solution to 10 ml with water and add solid Na_2CO_3 slowly until all the $MnCO_3$ has precipitated. Heat vigorously to insure complete precipitation. Centrifuge and discard the supernate. Wash the precipitate with 10 ml of water, centrifuge and discard the wash.

3. Dissolve the precipitate in 3 ml of concentrated HNO_3, add 1 ml of iron carrier (10 mg Fe/ml) and dilute to 15 ml with water. Cool the centrifuge tube in an ice bath and add 0.5 grams of $NaBiO_3$ (in portions, with stirring) to oxidize Mn^{+2} to MnO_4^-. Continue stirring for 1 minute and then add 2 drops of 85% phosphoric acid to stabilize the permanganate.

4. Make the solution basic with 6M NaOH (pH 9 to 10 with pH paper), centrifuge and decant the supernate into a 100-ml beaker (Note 1). Wash the precipitate with 5 ml of water containing several drops of 6M NaOH. Centrifuge and combine the wash with the supernate in the 100 ml beaker. Discard the precipitate.

5. To the combined supernate and wash from step 4 add 3 ml of concentrated HNO_3. Heat, add 2 ml of saturated oxalic acid solution and heat until the solution become colorless (Note 2).

6. Heat the solution almost to boiling, add 2 ml of saturated $NaBrO_3$ and stir vigorously until all the MnO_2 has precipitated.

7. Transfer the precipitate by portions to a 40-ml centrifuge tube. Centrifuge and discard the supernate. Wash the precipitate with 10 ml of 1N HNO_3, centrifuge and discard the wash.

8. Filter the precipitate onto a Whatman No. 42 filter disk using water and finally anhydrous "Anhydrol" as transfer agents. Dry in an oven at 110°C for 20 minutes and mount on a nylon planchet for radioassay.

9. On completion of radioassay, place the filter paper and precipitate in a previously heated and tared porcelain crucible and heat gently until the paper is completely charred, making sure that the paper does not ignite.

10. Heat the crucible in an electric muffle furnace at 850°C for 1 hour, cool in a desiccator and determine the chemical yield of Mn_3O_4.

Note 1. The solution should remain a deep purple.

Note 2. The permanganate is reduced to Mn^{+2}.

C. Iron-55, 59 Purification Procedure

The ferric hydroxide precipitate from the Sequential Separation Procedure is dissolved in 10M nitric acid and extracted into 0.6M thenoyltrifluoroacetone-xylene. The organic phase is washed with 4M nitric acid and 0.25M hydrofluoric-nitric acid mixture in order to remove zirconium-95. The iron is back-extracted with concentrated hydrochloric acid and finally plated onto a copper disk for radioassay.

1. Dissolve the $Fe(OH)_3$ precipitate from step 14 of the sequential separation in 3 ml of concentrated HNO_3, add 10 ml of 10M HNO_3 and 1.3 ml of 30% H_2O_2. Transfer the solution to a 60-ml cylindrical separatory funnel with water washings and add 15 ml of freshly prepared 0.6M 2-thenoyltrifluoroacetone (TTA)-xylene solution (Note 1). Stir the mixture for 15 minutes with a high speed motor stirrer.

2. Withdraw and discard the aqueous phase. Wash the sides of the separatory funnel with several ml of water, stir for one minute. Withdraw and discard the aqueous wash solution.

3. Add 15 ml of freshly prepared 4M HNO_3 - 3% H_2O_2 solution and wash for one minute. Centrifuge, withdraw and discard the aqueous wash solution. Wash the sides of the separatory funnel with several ml of water. Withdraw and discard the wash.

4. Repeat step 3.

5. Add 15 ml of 0.25M HI-0.25M HNO_3 solution, wash for one minute and discard the aqueous scrub solution. Repeat this step twice more.

6. Add 7 ml of concentrated HCl and stir until the organic phase is decolorized (about 10 minutes). Withdraw the aqueous phase and repeat the back-extraction with an additional 5-ml portion of concentrated HCl.

7. Combine both aqueous portions and precipitate $Fe(OH)_3$ with excess concentrated NH_4OH. Centrifuge and discard the supernate.

8. Dissolve the precipitate from step 7 in 5 drops of concentrated HCl and evaporate to dryness by gently heating over a flame. Dissolve the residue in 2 ml of water, add 1.5 ml of "Phosphate Buffer" (2.32M $NH_4H_2PO_4$) and 8 ml of "Carbonate Buffer" (Note 2).

9. Transfer the solution to the electrodeposition unit containing a tared copper disk (Note 3) with 5 ml of "Carbonate Buffer" solution. Electrodeposit for 2 hours, starting at 450 ma and increasing to 500 ma after 15 minutes.

10. On completion of plating, remove the copper disk and wash immediately with water and anhydrous "Anhydrol". Pat dry with an absorbent tissue and weigh for chemical yield (Note 4). Immediately cover the disk with a thin coating of Krylon, dry, mount and radioassay.

Note 1. 0.6M TTA-xylene: 12 gm TTA/100 ml of xylene.

Note 2. The "Carbonate Buffer" solution is made by dissolving 392.5 g $(NH_4)_2$ CO_4 in 175 ml of concentrated NH_4OH and diluting to 1 liter with water.

Note 3. The copper disk has to be thoroughly washed with a cleansing compound, distilled water and anhydrous "Anhydrol" prior to taring.

Note 4. Adjust the yield utilizing the concentration of elemental iron in the original sample, as measured by spectrophotometric analysis.

D. Iron Colorimetric Procedure (Ortho-Phenanthroline Method)

The iron is reduced with hydroxylamine hydrochloride, the orange-red Fe(II) complex of O-phenanthroline is formed and the spectrophotometric determination is made at 510 mμ. Oxidizing agents cause the major interference. The color is stable for about 12 hours after a 5-10 minute color development. The recommended range is from 25 to 100γ of iron in 25 ml of solution using a 1-cm cell.

1. Transfer an aliquot of the sample to a 25-ml volumetric flask (Notes 1 and 2), add 1 ml of 35% hydroxylamine hydrochloride solution, 1 ml of 1% ortho-phenanthroline in anhydrol solution and 15 ml of 40% ammonium acetate solution.

2. Dilute to the mark and mix well.

3. Rinse a 1-cm cell with three portions of the solution and transfer the solution to the cell.

4. Convert the photometric readings to milligrams of iron as follows: Total milligrams Fe = $\frac{A \times B \times 1000}{C}$,

   where A = original volume of sample in milliliters;

   B = micrograms of Fe in the aliquot used (from calibration curve);

   C = volume of aliquot in milliliters.

Note 1. Two aliquots of the standard solution and 5 ml of water should be transferred to separate 25-ml volumetrics and processed with the sample.

Note 2. To prepare the standard solution, dissolve 0.5000 gm of pure iron in 50 ml of HCl (2:1) and dilute to 1 liter in a volumetric flask. Dilute 100 ml of this solution to 1 liter in a volumetric flask. This solution is used for calibration. Transfer 1.0, 2.0, 3.0, 4.0 and 5.0 ml of standard solution to separate 25-ml volumetric flasks and dilute each to 25 ml. Transfer 5 ml of water to a separate 25-ml flask for use as a blank. Proceed with steps 1 through 4 of the procedure. Plot the photometric absorbance readings versus micrograms per ml of solution.

E. Cobalt-57, 58, 60 Purification Procedure

Cobalt is purified by two potassium cobaltinitrite precipitations, a palladium sulfide and copper sulfide scavenge, plus two ferric hydroxide scavenges. Finally the cobalt is plated on a copper disk and radioassayed.

1. Dissolve the $K_3Co(NO_2)_6$ precipitate from step 12 of the Sequential Separation Procedure in 5 ml of concentrated HCl and boil for several minutes to remove decomposition products. Add 10 mg of nickel carrier and dilute to 25 ml with water.

2. Precipitate nickel and cobalt hydroxides with 10M KOH. Centrifuge and discard the supernate.

3. Dissolve the precipitate in 3 ml of 6M $HC_2H_3O_2$ and heat slightly if necessary in order to dissolve the precipitate. Dilute to 25 ml with water and cool to room temperature.

4. Precipitate $K_3Co(NO_2)_6$ by adding 6 ml of 3M $HC_2H_3O_2$ that is freshly saturated with KNO_2 (Note 1). Allow 30 minutes for complete precipitation. Centrifuge and discard the supernate. Wash the precipitate with 30 ml of water, centrifuge and discard the wash.

5. Dissolve the $K_3Co(NO_2)_6$ precipitate in 5 ml of concentrated HCl, boil almost to dryness, add 2 drops of palladium carrier (10 mg Pd/ml) and 4 drops of copper carrier (10 mg Cu/ml). Dilute to 20 ml with water and add 2 ml of 1M HCl to make the solution approximately 0.1M.

6. Heat the solution almost to boiling and bubble H_2S gas through the solution for 5 minutes. Filter the sulfide precipitate through Whatman No. 42 filter paper (9 cm) contained in a 2", 60° glass funnel and collect the filtrate in a 125 ml Erlenmeyer flask. Wash the original centrifuge tube with 5 ml of water, pass the water solution through the filter funnel and combine with the filtrate, in the Erlenmeyer flask. Discard the sulfide precipitate.

7. Boil the filtrate almost to dryness (Note 2) to remove excess H_2S, add 5 ml of water and transfer the solution to a 40-ml centrifuge tube using about 10 ml of 1N HCl as a transfer agent. Add 4 drops of iron carrier (10 mg Fe/ml) and precipitate $Fe(OH)_3$ by addition of concentrated NH_4OH (Note 3). After the $Fe(OH)_3$ has been completely precipitated, add about 0.5 ml of NH_4OH in excess. Centrifuge and discard the precipitate.

8. Acidify the supernate with 6M HCl, add 4 drops of iron carrier and repeat the scavenge as in step 7.

9. Transfer the supernate from the $Fe(OH)_3$ scavenge to a clean 40-ml centrifuge tube and precipitate CoS by bubbling H_2S gas through the solution for 2 minutes. Centrifuge and discard the supernate.

10. Transfer the CoS precipitate with 5-10 ml of water to a 125 ml Erlenmeyer flask, add 10 ml of concentrated HNO_3 and evaporate to approximately 5 ml. Transfer the solution to a 100-ml beaker and wash the Erlenmeyer twice with water, adding the washes to the beaker, and evaporate to 1-2 ml.

11. Add 3 ml of concentrated H_2SO_4 and heat to SO_3 fumes. Cool, slowly add 5-10 ml of water and again cool. Neutralize the solution with concentrated NH_4OH add 2 grams $(NH_4)_2SO_4$ and electroplate the cobalt on a tared 7/8" diameter copper disk. Begin plating at 3-4 volts and 0.10 amps. After the first half-hour increase the current to 0.20 amps and plate for 4 hours. Upon completion of plating the solution should be colorless.

12. Dismount the copper disk from the plating cell, wash the disk with water and acetone, dry in a desiccator and weigh the Co metal for chemical yield. Mount on a nylon planchet for radioassay.

Note 1. Maintain a saturated solution by repeated addition of KNO_2.

Note 2. The H_2S is removed to prevent precipitation of CoS in the $Fe(OH)_3$ scavenge step.

Note 3. If, at the addition of NH_4OH, the green color of $Co(OH)_2$ appears, add HCl until the color disappears and proceed with the $Fe(OH)_3$ precipitation.

F. Cadmium-109, 113m, 115m Purification Procedure

The cadmium fraction is purified by several cadmium sulfide precipitations and iron, silver and palladium scavenges. The cadmium is electroplated on copper for radioassay.

1. To the supernate from step 6 of the Sequential Separation Procedure, containing the separated cadmium, add 6N HCl until the pH is 1.5. Bubble H_2S gas through the solution for 5 minutes, centrifuge and discard the supernate.

2. Dissolve the CdS precipitate in 5 ml of 6N HCl and boil over a flame for approximately 2 minutes. Add 2 drops of palladium carrier (10 mg Pd/ml), heat to boiling and saturate the solution with H_2S gas for 3 minutes.

3. Digest the precipitate in a hot water bath for 5 minutes, cool to room temperature, add 2 drops of "Aerosol" solution and centrifuge. Transfer the supernate to a clean 40-ml centrifuge tube and discard the precipitate.

4. Evaporate the supernate to dryness in a hot sand bath or over a flame. Dissolve the residue in 15 ml of water, add 3 drops of iron carrier (10 mg Fe/ml) and 1 ml of 6M $NH_4C_2H_3O_2$. Heat the solution to boiling over a flame and digest for 5 to 10 minutes in a hot water bath.

5. Cool the solution to room temperature, centrifuge and discard the precipitate. To the supernate add 1 ml 6M $NH_4C_2H_3O_2$, heat to boiling over a flame, add 3 drops of iron carrier (10 mg Fe/ml) and digest in a hot water bath for 5 to 10 minutes.

6. Cool the solution to room temperature, centrifuge, transfer the supernate to a clean 40-ml centrifuge tube and discard the precipitate.

7. To the supernate add 6N HCl until a pH of 1.5 is reached and heat in a hot water bath. Saturate the solution with H_2S gas for three minutes (Note 1). Cool to room temperature, centrifuge and discard the supernate.

8. Add 2 ml concentrated HNO_3 and boil over a flame until the CdS precipitate is dissolved. Dilute to 5 ml with water, add 5 drops of silver carrier (10 mg/ml), 1 ml of 1M HCl, stir and digest in a hot water bath for 5 minutes. Cool, centrifuge and discard the precipitate.

9. Add water until a pH of 1.5 is obtained and heat in a hot water bath for 5 minutes. Saturate the solution with H_2S gas for 3 minutes, cool to room temperature, centrifuge and discard the supernate.

10. Repeat steps 2 through 7 twice.

11. Dissolve the CdS precipitate in 5 ml of 6N HCl and evaporate to dryness.

12. Wash down the walls of the centrifuge tube with approximately 1 ml of water, add 6 drops of concentrated H_2SO_4 and boil over a flame until SO_3 fumes are given off. Cool, again wash the sides of the centrifuge tube with 1 ml of water and heat to SO_3 fumes.

13. Cool, add 3 ml of water to dissolve the $CdSO_4$. Again cool and add 12M NaOH, a drop at a time, until the first permanent $Cd(OH)_2$ precipitate is formed.

14. Add 10% solution of KCN until the precipitate dissolves and transfer the solution to the electrodeposition unit containing a tared copper disk (Note 2). Wash the centrifuge tube with 5 to 7 ml of 0.03% solution of gelatin and transfer the wash to the electrodeposition unit. Electroplate for 3.5 hours at 6.0 volts.

15. On completion of plating, remove the copper disk and wash for several minutes with hot water and then with anhydrous "Anhydrol". Dry the disk and weigh the cobalt metal for chemical yield. Mount on a brass planchet for radioassay.

Note 1. If a precipitate does not form, add water until it does.

Note 2. The copper disk should be thoroughly washed with a cleaning compound, distilled water and anhydrous "Anhydrol" prior to taring.

G. Antimony-124, 125 Purification Procedure

The separated antimony sulfide is dissolved and another sulfide precipitation is performed. The antimony is finally precipitated as metal with "Oxsorbent". The metal is mounted and radioassayed.

1. Dissolve the separated Sb_2S_5 precipitate in 4 ml of concentrated HCl and boil over a flame for two minutes. If the precipitate does not completely dissolve, add 4 to 5 drops of 30% H_2O_2 and boil.
2. Add 10 mg of rhodium carrier, 10 mg of ruthenium carrier and 4 drops of 30% H_2O_2. Heat the solution to boiling, cool, add 4 drops of 3% H_2O_2 and evaporate to dryness in a hot air bath.
3. While still hot add 20 ml 6N H_2SO_4, bubble H_2S gas into the solution for ten minutes, cool, centrifuge and discard the supernate.
4. Wash the precipitate from step 3 with 5 ml of 3N H_2SO_4, centrifuge and discard the wash. Add 5 ml of concentrated HCl and heat the solution to boiling. Cool, dilute to a total of 15 ml with water. Centrifuge and filter the supernate through Whatman No. 42 filter paper, collecting the filtrate in a 125 ml Erlenmeyer flask.
5. Add another 5 ml of concentrated HCl and heat the solution to boiling. Cool, dilute to 15 ml with water and filter through the same Whatman filter paper used in step 4. Combine the filtrate with the filtrate in the Erlenmeyer flask.
6. Wash the precipitate with concentrated HCl, filter through the Whatman No. 42 filter paper and combine the filtrate with the filtrates from steps 4 and 5.
7. Add 5 ml of "Oxsorbent" ($CrCl_2$ solution) (Note 1) to the Erlenmeyer flask, heat to boiling, cool and add an additional 5 ml "Oxsorbent". Cool, centrifuge and carefully decant and discard the supernate.

8. Wash with 10 ml of water, centrifuge, decant and discard the wash. Filter the antimony metal onto a tared Whatman No. 42 filter paper using water as a transferring agent. Wash the precipitate with three 5 ml portions of anhydrous "Anhydrol". Dry in an oven at 110°C for 10 minutes, cool in a desiccator, weigh and mount on a brass planchet.

Note 1. The 5 ml of "Oxsorbent" is added to the solution via a 5-ml pipette. The tip of the pipette is immersed in the solution and the "Oxsorbent" is allowed to enter the solution without coming into contact with air. A clean pipette is used for each addition to each sample.

H. Thallium-204 Purification Procedure

The separated TlI precipitate is dissolved in 6M HNO_3 and subjected to a series of tellurium metal and lanthanum hydroxide scavengings. Finally, after several TlI precipitations, the thallium activity is mounted as thallium chromate and is radioassayed.

1. Dissolve the separated TlI precipitate in 5 ml of 6M HNO_3 by boiling over an open flame. Heat until iodine vapors are no longer evident. Transfer the solution to a 125 ml Erlenmeyer flask with water as transfer agent.

2. Add 1 ml of tellurium carrier (10 mg Te/ml), 5 ml of concentrated HCl and evaporate to dryness. Add 5 ml of concentrated HCl and evaporate to dryness three more times.

3. While still warm, add 20 ml of 3 M HCl, approximately 1 ml of $N_2H_2H_2O$ (hydrazine-hydrate) and heat to boiling. Add 1 ml of 6% H_2SO_3, continue to boil and make four to five successive 1 ml additions of H_2SO_3 (Note 1).

4. While still warm filter the solution through a 2" 60° funnel using No. 42 Whatman filter paper. Collect the solution in a clean 125-ml Erlenmeyer flask (Note 2). Wash the original flask and the precipitate with dilute H_2SO_3. Combine the wash with the filtrate and discard the precipitate.

5. Bring the volume of solution to approximately 75 ml with water, add 1 to 2 g. of NaI. Transfer to a 40-ml centrifuge tube, centrifuge and discard the supernate.

6. Add 5 ml of 6M HNO_3 and heat the solution over an open flame until the I_2 vapors are no longer evident; then add 7 drops of lanthanum carrier (10 mg La/ml).

7. Dilute the solution to 20 ml with water, heat over an open flame until the solution is hot and add 1 ml of H_2SO_3. While still warm make the solution ammoniacal by the dropwise addition of concentrated NH_4OH; then add 1 ml of NH_4OH in excess.

8. Centrifuge, transfer the supernate to a clean 40-ml centrifuge tube and discard the precipitate.

9. To the supernate add approximately 1 gram of NaI, centrifuge and discard the supernate. To the precipitate add 3 ml of 6M HNO_3 and heat over an open flame until I_2 vapors are no longer evident. Transfer the solution to a 125 ml Erlenmeyer flask with water.

10. Repeat steps 2 through 9.

11. To the supernate, again add 1-2 grams of NaI, centrifuge and discard the supernate.

12. Repeat steps 6, 7 and 8.

13. To the supernate from step 12 add 5 ml of 10% Na_2CrO_4 and allow the precipitate to stand at room temperature for 10 minutes.

14. Centrifuge and discard the supernate. Filter the precipitate onto a tared Whatman No. 42 filter disk with water. Wash the precipitate with 10 ml of water and three 5 ml portions of anhydrous "Anhydrol". Dry in an oven at 110°C for ten minutes. Cool in a desiccator and weigh for chemical yield. Mount on a brass planchet for radioassay.

    Note 1. Particular care must be given to insure complete precipitation of tellurium metal. When the tellurium has been completely precipitated, the supernate is clear with no bluish tint.

    Note 2. If the solution stands too long before filtering white crystals will form. If this happens add 1 ml of H_2SO_3 and heat.

I. Radioassay Procedures

For the radioassay of manganese-54 the purified manganese dioxide precipitate, in a test tube, is placed in a NaI(Tl) well crystal and is counted for 480 minutes. The gamma pulses are fed into a 400-channel pulse height analyser, and the manganese-54 activity is calculated by integration of the area under the peak from the 0.84 Mev photon.

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Iron-55 is measured using an internal gas flow proportional counter with 10% methane and 90% argon as the counting gas. The background of the detector is reduced to approximately 1 cpm by an anti-coincidence guard and 4 inches of lead shielding. The iron carrier with the iron-55 is plated onto a disk and is placed 1/4 inch from the thin window of the proportional counter. The 6.4 kev x-rays produced by electron capture are critically absorbed by the counting gas and the counts are recorded. The purity of the sample is established by counting it with beryllium-absorbers.

Iron-59 is measured using a 3-inch by 3-inch NaI(Tl) crystal coupled with a multichannel analyzer. The areas under the 1.10 and 1.30 Mev photopeaks are used to calculate the sample activity.

Measurement of the 22.5 kev x-ray of cadmium-109 is used for the radioassay of that nuclide. The copper disk onto which the cadmium has been plated is counted using a thin crystal of NaI(Tl) with a 128-channel pulse height analyzer. The sample is counted for 480 minutes, and the cadmium-109 activity is calculated by integration of the area within the half-height of the 22.5 kev x-ray photopeak.

Cadmium-113m and cadmium-115m are measured by beta counting. The ratio of counting rates of the 0.5 Mev beta of cadmium-113m with and without absorbers is used to test the radiochemical purity of that nuclide. The apparent half-life of the cadmium-115m from its 1.6 Mev beta is compared with the true half-life to assess the radiochemical purity of that nuclide.

The antimony-124 and antimony-125 are gamma counted using a 3-inch by 3-inch NaI(Tl) crystal coupled with a multichannel pulse height analyzer.

Both nuclides have photopeaks at 0.60 Mev, but antimony-124 also has a photopeak of 1.69 Mev. The count rate represented by the 1.69 Mev photopeak can be used, therefore, to calculate the antimony-124 activity and to estimate the contribution of the antimony-124 to the 0.60 Mev photopeak. The residual count rate at 0.60 Mev after the subtraction of the antimony-124 contribution can be used to calculate the antimony-125 activity.

The cobalt isotopes are also measured by means of a 3-inch by 3-inch NaI(Tl) crystal coupled with a multichannel pulse height analyzer. The 1.17 and 1.33 Mev photopeaks of cobalt-60, the 0.81 Mev photopeak of cobalt-58, and the 0.122 Mev photopeak of cobalt-57 are measured.

The radioassay of thallium-204 is accomplished by counting the 0.77 Mev beta ray of that nuclide on a low-background beta counter.

3.4 The Analysis of Filter Samples for Plutonium Isotopes

The filter samples which were analyzed for plutonium isotopes were normally analyzed for strontium-90 as well. During the period 1961 to 1964 cesium-137 was also measured in most of the samples which were analyzed for plutonium-239, 240. During 1965 to 1967, cadmium-109 was also analyzed in these samples. The procedures given below are applicable to the analysis of a sample for strontium-90, cesium-137, plutonium-238 and plutonium-239, 240.

A. Sequential Separation Procedure

Strontium, cesium and iron carriers and strontium-85 and plutonium-236 "spikes" (for determination of chemical yields) are added and ferric hydroxide and strontium carbonate are precipitated, leaving cesium in the supernate. The precipitate is dissolved, and the plutonium is separated by a ferric hydroxide scavenge with ammonium hydroxide.

1. To an aliquot of the filter paper solution contained in a 40 ml glass centrifuge tube add 20 mg of strontium carrier, 1 ml of strontium-85 spike solution (Note 1), 20 mg of cesium carrier and 10 mg of iron carrier.

2. Make the solution basic to pH 9 with 50% NaOH, digest in a hot water bath for 20 minutes, add 10 ml of saturated Na_2CO_3 solution, digest for 10 minutes in a hot water bath, cool, centrifuge and reserve the supernate for cesium-137 purification as described in Section 3.1.

3. Dissolve the precipitate in 5 ml of 6M HNO_3, boil over a flame for two minutes to expel all CO_2, dilute to 15 ml with water and make basic to pH 8.5 with concentrated NH_4OH. Centrifuge and reserve the ferric hydroxide precipitate for plutonium-238, 239, 240 purification.

4. Reserve the supernate for strontium-90 purification as described in Section 3.1.

Note 1. Strontium-85 spike is added to determine the recovery of strontium. This eliminates the need for a gravimetric determination of $SrCO_3$. Similarly plutonium-236 spike is added to permit the determination of the recovery of plutonium.

B. Plutonium-238, 239, 240 Purification Procedure

1. Dissolve the $Fe(OH)_3$ precipitate from step 3 of the Sequential Separation Procedure in a minimum of concentrated HNO_3. Wash down the centrifuge tube walls with 1N HNO_3 and add 1 ml 5% NH_4OH-HCl. Let stand for 0.5 hour, (this should not effervesce).

2. Add concentrated HNO_3 until effervescence occurs. Heat in a very hot (but not boiling) water bath for 0.5 hour. Remove from the hot water bath and allow to stand for a minimum of 4 hours.

3. Add the solution to the reservoir of an ion-exchange column. Wash the centrifuge tube with several ml 7N HNO_3, and add to the column.

4. Collect the effluent in a 250 ml beaker. Wash the sample on column with 150 ml 7N HNO_3 and discard the effluent.

5. Strip the Pu from the column with 20 ml of 1N HNO_3 followed by 45 ml of 5% NH_4OH-HCl. Collect this effluent in a 150 ml beaker.

6. Evaporate the effluent carefully until effervescence stops. Then evaporate to near dryness and add 1 ml of 1N HNO_3 and 1 ml 5% NH_4OH-HCl. Allow to stand for ten minutes.

7. Add 8 ml of water. The pH at this point should be 1.0. Check with pH paper (0-1.5 range) and transfer to a prepared plating cell. Plate for 3 hours at 0.5 amperes.

8. Add a dropper full of concentrated NH_4OH to the cell before turning off the current.

9. Remove the disk. Wash the disk with water. Flame to redness. Cool and label for counting.

Note 1. Resin and Column Preparation:

- a. The resin used is AG 1-X2 (100-200 mesh).
- b. Wash the resin with 7N HNO_3, let it settle. Decant and discard the HNO_3.
- c. Wash twice with water as above.
- d. Wash with 5% NH_4OH-HCl as above.
- e. Wash twice with water as above.
- f. Repeat steps b-e.
- g. Put the resin on the column with 7N HNO_3.
- h. Run about 40 ml 7N HNO_3 through the column and discard it.

Note 2. Disk Preparation:

Scrub a stainless steel disk with brillo, and wash thoroughly with water.

C. Plutonium-238, 239 Radioassay Procedure

The radioassay of plutonium-238 and plutonium-239 is accomplished by means of a 480 minute alpha count of the stainless steel disc onto which the plutonium has been plated, using solid state ZnS detectors and a 128-channel pulse height analyzer. The plutonium-238 activity is calculated by integration of the area under the 5.5 Mev peak, and the plutonium-239 activity is calculated by integration of the area under the 5.1 Mev peak.

3.5 The Analysis of Filter Samples for Radon Daughter Products

The radon daughter products, lead-210 and polonium-210, were measured in "ST" samples during 1961 to 1963, with cosmic ray products and artificial tracer nuclides. From 1965 to 1967, however, special "SR" samples were prepared and were analyzed only for strontium-90. Lead-210 and polonium-210 were analyzed only when data for the radon daughter products were desired. The radiochemical procedures used with these "SR" samples are given here.

A. Sequential Separation Procedure

The sample should be contained in a 40 ml centrifuge tube, and the volume should not exceed 20 ml. It contains the Po^{208} "spike" which was added before ashing.

1. Transfer the 0.5N HCl sample to a 70-ml plexiglass cup (which contains a polished silver disk), washing with 0.5N HCl. Add 0.5 N HCl until the volume in the cup is approximately 60 ml.
2. Add about 100 mg ascorbic acid.
3. Plate for three hours at 75° - 80°C and zero potential, replacing any solution lost due to evaporation with 0.5N HCl.
4. When plating is complete (after 3 hours), decant the hot solution back into the original 150 ml beaker and wash the cup with water.
5. Remove the silver disk from the plating cup, wash it with water, air dry, and reserve for alpha counting of polonium.
6. Add Sr and Pb carriers and the Sr^{85} spike to the solution and evaporate to about 10 ml. Add 10 ml of concentrated HNO_3 and again evaporate to 5 ml to decompose the ascorbic acid previously added.
7. Add 15 ml of fuming HNO_3 and chill in an ice bath for 15 minutes. Centrifuge and discard the supernate. Add 10 ml of concentrated HNO_3. Centrifuge and discard the supernate.
8. Dissolve the precipitate in 5 ml of distilled water, add 20 ml of fuming HNO_3, chill and repeat step 7.

9. Dissolve the precipitate in a minimum of water and transfer to a 150 ml beaker. Evaporate almost to dryness. Add 10 ml of 6N HCl and again evaporate almost to dryness.

10. Dissolve the salts in about 10 ml of 2N HCl (heat if necessary) and then bring the volume up to about 50 ml with 2N HCl.

11. Transfer the sample to an anion exchange column, which previously has been equilibrated with 2N.HCl. Elute the Sr fraction with 50 ml of 2N HCl. Hold effluent for Sr purification, as described in Section 3.2.

12. Elute the Pb from the column with 75 ml of 9N HCl, collecting the effluent in a 150 ml beaker. Hold this fraction for the Pb purification.

B. Lead-210 Purification Procedure

1. To the effluent from step 12 of the Sequential Separation Procedure, add 1 ml of Bi carrier, 2 ml concentrated H_2SO_4 (carefully) and evaporate to fumes of SO_3. Cool and add 100 mg citric acid. Dilute to 20 ml with water and heat to boiling to coagulate $PbSO_4$.

2. Cool, centrifuge and discard the supernate. Add 10 ml of water. Centrifuge and discard the supernate. Record this time for Bi scavenge.

3. Dissolve the $PbSO_4$ with 2 ml of 40% ammonium acetate which has been made acid with acetic acid. Slight heating may be necessary.

4. Dilute to 15 ml with water. Add 3 ml of 1.5 Na_2CrO_4 and digest for 20 minutes in a hot water bath.

5. Filter onto a previously tared filter paper. Wash well with hot distilled water, and then twice with anhydrol. Dry at 105°C for 15 minutes. Weigh the $PbCrO_4$ for yield. Mount on a brass planchet for radioassay.

C. Lead-210, Polonium-210 Radioassay Procedure

For the radioassay of polonium-210 the silver disk onto which it has been plated is counted using the same alpha detectors and pulse height analyzer used for plutonium counting, and the activity is calculated by integrating the area under the 5.3 Mev peak.

The lead-210 activity of the lead chromate precipitate is measured by monitoring the ingrowth of the 5.0 day bismuth-210 daughter of lead-210. A series of measurements of the sample are made, each lasting 200 minutes or to a total of 1000 counts, using an end window, gas flow, Geiger-mode counter.

3.6 The Analysis of Filter Samples for Cosmic Ray Products

Between May 1963 and April 1965 a series of filter samples was analyzed for cosmic ray products using the following radiochemical procedure:

Samples which are analyzed for 53-day beryllium-7, 14.3-day phosphorus-32, 25-day phosphorus-33, 2.58-year sodium-22 and 28-year strontium-90 are wet ashed using fuming HNO_3, $HClO_4$, and HF. Aliquots of the resultant solution are taken for determination of phosphorus and sodium before carriers are added to the sample. Beryllium is separated from the solution by precipitation of $Be(OH)_2$, phosphorus is separated as ammonium phosphomolybdate, strontium is separated as $SrCO_3$ and sodium is separated as sodium uranyl acetate.

The beryllium is purified by a series of precipitations which removes several possible radioactive contaminants, and then is extracted into benzene as an acetyl acetone complex to remove it from any remaining contaminants. It is back-extracted into HCl solution, and is precipitated as $Be(OH)_2$ to be counted for beryllium-7. The phosphorus is converted to $MgNH_4PO_4$ and is passed through two cation exchange columns in succession to remove any radioactive contaminants which are present as cations. It is then reprecipitated as NH_4PMO_4 to eliminate any remaining anionic radioactive contaminants. It is then once more reconverted to $MgNH_4PO_4$ and is heated in a muffle furnace to convert it into $Mg_2P_2O_7$, a form which is most suitable for counting and for yield determination. The sodium, which is precipitated as the uranyl acetate to separate it from possible alkali metal radioactive contaminants, such as cesium-137, is redissolved and is scavenged using several successive $Fe(OH)_3$ precipitations. Any accumulated ammonium salts are then driven off by heating the solution to dryness. The sodium is dissolved and its solution is mixed with 1-butanol, which is saturated with HCl, to force the precipitation of NaCl, which is counted.

Beryllium-7 is counted on a gamma spectrometer, using the area under the peak due to the 0.48 Mev gamma ray for the assay. Phosphorus-32 and phosphorus-33 are assayed by means of a series of beta counts, using either differential absorption of the 1.71 Mev beta of phosphorus-32 and the 0.25 Mev beta of phosphorus-33, or resolution (by means of a computer program) of the decay curve into a two component system to permit calculation of concentrations of both beta-emitters from the resulting data. The 0.54 Mev position of sodium-22 is also assayed by beta counting the sample, with absorber measurements used to check the energy of the radiation, and, therefore, the radioactive purity of the sample.

Radiochemical and radiometric procedures used to measure strontium-90 were given in Section 3.2. The details of procedures used with the other nuclides are given below:

A. Ashing Procedure

1. Cut the filter paper which is to be analyzed into small squares and place it in a 600-, 800-, or 1,000-ml beaker, depending on the quantity of paper used.
2. Add 400 ml of fuming HNO_3 to each sample, cover with a watch glass and allow to digest for 30 minutes. Evaporate the sample down to 25 ml. Care must be taken to reduce the hot plate temperature if any of the reactions becomes violent.
3. Add an additional 50 ml of fuming HNO_3 to each sample and again evaporate to 25 ml. If any of the solutions are not clear, repeat the treatment with 50 ml fuming HNO_3.
4. Add 50 ml of a 1:1 solution of concentrated $HClO_4$ and concentrated HNO_3 and evaporate to a small volume; do not allow the sample to go to dryness.
5. Transfer the sample to a teflon beaker using 7N HNO_3 as the transfer agent. Lower the hot plate temperature to about 400° and evaporate the sample to <10 ml.

6. Add 1 ml of concentrated $HClO_4$ and 10 ml of HF to each sample and evaporate down to approximately 5 ml.

7. Repeat step 6 with 10 ml of HF twice. Continue the final evaporation down to drop size. If the sample is not clear at this point, repeat a single HF treatment. If the sample is still not clear, add 5 ml of H_2O_2 and evaporate down to drop size. Add 5 ml of $HClO_4$ and evaporate down to drop size.

8. Transfer the solution to a 25 ml volumetric flask with 0.01N HNO_3.

B. Sequential Separation of Beryllium-7, Phosphorus-32, 33, Sodium-22 and Strontium-90

Because the sample is to be analyzed for sodium-22 the entire sequential separation must be carried out in either teflon or polypropyleneware.

1. Remove four 1-ml aliquots from the dissolved sample in a 25-ml volumetric flask. The sample should be dissolved in approximately 0.01N HNO_3. Dilute to the mark and shake thoroughly before removing aliquots. Two 1-ml aliquots are for determination of natural phosphorus and two 1-ml aliquots are for the determination of natural sodium.

2. Pipette 20 ml of the sample solution into two 100-ml teflon beakers.

3. Add the following carriers:

   10 ml of 1 mg/ml beryllium
   2 ml of 25 mg/ml sodium
   1 ml of 7.5 mg/ml phosphorus
   1 ml of 30 mg/ml strontium

   Add 1 ml of 2 x 10^4 dpm/ml strontium spike

4. Evaporate the sample to a volume of 10 ml and transfer to a clean polypropylene centrifuge tube.

5. Add NH_4OH dropwise until the precipitation of $Be(OH)_2$ is complete (pH = 8). The NH_4OH should be added carefully because if the correct pH for $Be(OH)_2$ is exceeded, phosphorus will also precipitate. Centrifuge, and decant the supernate into a 100 ml teflon beaker.

6. Dissolve the $Be(OH)_2$ precipitate in 5 ml of 6M HNO_3 and dilute to 10 ml with water.

7. Add NH_4OH to the solution from step 6 until $Be(OH)_2$ is completely precipitated. Centrifuge and save this precipitate for beryllium purification. Combine the supernate with the supernate from step 5.

8. In a hot water bath ($\leq$ 50°C) add 5 ml ammonium molybdate reagent (saturated) to the combined supernate from steps 5 and 7. Centrifuge and retain this precipitate (ammonium phosphomolybdate) for phosphorus purification.

9. To the supernate from step 8 add NH_4OH to pH >8.5 and add solid $(NH4)_2CO_3$. Digest in a hot water bath for 5 - 10 minutes, cool and centrifuge. Save the precipitate for strontium purification as described in Section 3.2, and retain the supernate for sodium purification.

C. Purification Procedure for Beryllium-7

1. To the precipitate from step 7 of the Sequential Separation Procedure, add 5 mg of lanthanum carrier and 10.0 ml 3M NaOH and digest in a hot water bath for *not more than 5 minutes*. Digestion for longer than 5 minutes will redissolve some of the $La(OH)_3$ precipitate that results from this step. Cool, centrifuge and decant the supernate into 40 ml centrifuge tube. Discard the $La(OH)_3$ precipitate.

2. Neutralize the solution by dropwise addition of concentrated HCl, and then make the solution strongly ammoniacal with concentrated NH_4OH. The addition of HCl will cause the precipitation of $Be(OH)_2$, but this will redissolve when the solution becomes neutral.

3. Digest in a hot water bath for about 10 minutes, cool, centrifuge ($Be(OH)_2$) and discard the supernate.

4. Dissolve the $Be(OH)_2$ precipitate in 3 ml of 6M HCl and dilute to 10 ml with water. Add 10 drops each of molybdenum and tellurium scavenging reagent (10 mg Mo, Te/ml) and heat in a hot water bath for 15 minutes.

5. To the hot solution add approximately 1 gram of thioacetamide and heat for an additional 15 minutes. Filter the solution through a 9.0 cm Whatman #42 filter paper in a 60°-2" glass funnel. Collect the filtrate in a clean 125-ml Erlenmeyer flask. The thioacetamide will precipitate both molybdenum and tellurium sulfides. In order to get complete separation from the beryllium, they should be filtered while hot.

6. Evaporate the filtrate to a volume of approximately 3 ml, cool and dilute to a volume of 15 ml with water.

7. Add 2.0 ml of "acetate buffer" and 2.0 ml of 10% EDTA solution. Adjust the pH to 5.5-6.0 by dropwise addition of NH_4OH. Transfer the solution to a 60 ml cylindrical separatory funnel using water as a transfer agent. Add 2.0 ml of acetylacetone and stir with a mechanical stirrer for several minutes.

8. Add 7.0 ml of benzene and stir mechanically for two minutes. Withdraw the aqueous layer into a clean 40-ml centrifuge tube and transfer the organic phase into a second clean 40-ml centrifuge tube.

9. Adjust the pH of the aqueous solution to 5.5-6.0 and transfer the solution back into the separatory funnel.

10. Repeat step 8 and combine the benzene fractions in the separatory funnel.

11. Wash the benzene fraction twice with 10 ml of 10% EDTA.

12. Add to the benzene fraction 7.0 ml of 6M HCl and stir mechanically for several minutes and withdraw the aqueous layer (HCl) into a 125 ml Erlenmeyer flask.

13. Repeat step 12 combining the HCl fractions in the Erlenmeyer flask.

14. Evaporate the HCl fractions almost to dryness. Add 5.0 ml concentrated HNO_3 and evaporate to dryness.

15. Dissolve the residue, $Be(NO_3)_2$, in 2.0 ml of 6M HCl (heat if necessary to get all the residue into solution), and dilute with 5 ml of water. Transfer the solution to a clean 40ml centrifuge tube. Wash the flask with three 3 ml aliquots of water and add these washings to the centrifuge tube.

16. Make the solution basic with NH_4OH. Centrifuge and discard the supernate. Wash the $Be(OH)_2$ precipitate twice with 5 ml of water, centrifuge and discard the washings.

17. Dissolve the precipitate in a minimum of concentrated HCl and transfer the solution to a 10-ml test tube. Make the solution basic with concentrated NH_4OH to reprecipitate the $Be(OH)_2$. Centrifuge and discard the supernate.

18. The $Be(OH)_2$ precipitate is counted in this form (damp precipitate) in the 10-ml test tube in a well-type counter and beryllium is yielded colorometrically after radioassay.

D. Beryllium Colorimetric Procedure

A beryllium complex of 4-(p-nitrophenylazo) orcinol is formed in a buffered solution at pH 12.0 containing E.D.T.A. chelating agent. Measurement is made at 515 mμ. Interfering elements are removed prior to the measurement. The color is stable for approximately one hour. The concentration range is 20 to 200 γ/100 ml for the 1 cm cell.

1. Dissolve the $Be(OH)_2$ in 5 ml of 6N HCl and dilute to volume in a 100-ml volumetric flask.

2. Pipette a 2.00-ml aliquot of the solution into a 100-ml beaker. Adjust the volume to 15 ml with water.

3. Add 5 ml of the chelating solution (Note 1) and adjust the pH to 5.5 using 2M HCl or 2M NaOH as required.

4. Add 10 ml of beryllium buffer solution (Note 2), stir and let stand for five minutes.

5. Transfer the solution to a 100-ml volumetric flask. Add exactly 10.0 ml of dye solution (Note 3) mix and let stand for ten minutes. Dilute to mark with distilled water.

6. Record the absorbance of each solution in a 1-cm cell at 515 mμ.

7. Divide the absorbance reading by the average slope value obtained from the calibration (Note 4) and multiply by 50.0 to obtain milligrams of beryllium.

Note 1. Chelating Solution: Saturated Disodium E.D.T.A.

Note 2. Buffer solution (pH 12): Prepared by dissolving 116 grams of citric acid, 61.5 grams of sodium borate decahydrate, 216 grams of sodium hydroxide in water and diluting to 1 liter.

Note 3. Dye Solution: Prepared by dissolving 0.150 grams of 4-(p-nitrophenylazo) orcinol (Chemical Procurement Laboratories, Inc.) in 500 ml of 0.1N sodium hydroxide solution by stirring with a mechanical stirrer for five hours. Filter the solution through a sintered glass filter and finally through a Millipore HA filter (0.45μ). A new solution has to be made up for each run.

Note 4. A calibration curve must be obtained, using a standard solution, with each set of samples. To prepare the standard solution, dissolve 1.0000 gram of high purity beryllium metal in 10 ml of 6N HCl. Boil until effervescence stops. Cool, dilute to 1 liter with water in a volumetric flask and mix thoroughly. Dilute 10 ml of this solution to 250 ml in a volumetric flask and mix thoroughly. This solution, 40 γ Be/ml, is used to calibrate the 1 cm cells. Transfer 1.00, 2.00, 3.00, 4.00 and 5.00 ml of this standard beryllium solution into a 100-ml beaker. Adjust the volume to 15 ml with the water. Concurrently run a 15-ml water blank. Proceed as in steps 3 through 6 of the procedure.

E. Purification Procedures for Phosphorus-32, 33

1. Dissolve the NH_4PMoO_4 precipitate from step 8 of the Sequential Separation Procedure in 1.0 ml concentrated NH_4OH. If the precipitate does not completely dissolve, centrifuge and decant the supernate into a 40-ml centrifuge tube and discard the remaining precipitate. Add 2 ml of 1N citric acid solution.

2. Slowly add 10 ml of saturated magnesia mixture while stirring, and add concentrated NH_4OH dropwise until the solution is just alkaline (pH~7); then add 10 more drops of NH_4OH.

3. Stir for 1 minute after formation of the precipitate, and then add 4 ml of concentrated NH_4OH. Allow the mixture to stand for at least 4 hours with additional stirring.

4. Prepare two cation exchange columns using 10-mm O.D. glass tubing and 10 ml of wet 50 - 100 mesh Dowex AG-50WX 8 cation exchange resin.

5. Condition the columns by passing 150 ml of 1.03N HCl through each and do not let the resin run dry.

6. Centrifuge the precipitate ($MgNH_4PO_4$) from step 3 and discard the supernate.

7. Dissolve the $MgNH_4PO_4$ in 1 ml (pipetted) of 3N HCl and add 99 ml of water. If the solution is not completely clear, heat on a hot plate until it is.

8. Pass the solution from step 7 through one of the conditioned exchange columns at a rate of 1 drop every 5 seconds and collect the effluent in a 150 ml beaker.

9. Pass the effluent through the second conditioned exchange column at a rate of one drop every 5 seconds and collect the effluent in a 150 ml beaker.

10. By pipette add 1 ml of concentrated $HClO_4$ and evaporate the effluent until the appearance of fumes of $HClO_4$. Transfer the solution to a 40-ml centrifuge tube with 20 ml of water and by pipette add 4 ml concentrated HNO_3.

11. Add 5 ml of saturated ammonium molybdate reagent, several drops of aerosol wetting agent and heat for 5 minutes in a hot water bath ($\leq$ 50°C). Cool, centrifuge and discard the supernate.

12. Dissolve the NH_4PMoO_4 precipitate in 1 ml (pipetted) concentrated NH_4OH. Again, if the precipitate does not completely dissolve, centrifuge and decant the supernate into a 40-ml centrifuge tube. Add 2 ml 1N citric acid solution.

13. Repeat step 2 with the magnesia mixture.

14. Repeat step 3.

15. Filter the precipitate onto a Whatman #42 filter disc, wash the precipitate with three 5 ml portions of 1:20 NH_4OH and finally with anhydrous "Anhydrol". Dry in an oven at 110°C for 10 minutes and transfer the filter paper and precipitate to a porcelain crucible.

16. Char the paper in the crucible using a Bunsen burner. Ash the sample in a muffle furnace at 1050°C for one hour. Cool, transfer the precipitate to a 40 ml centrifuge tube using anhydrous "Anhydrol" as a transfer agent.

17. Carefully grind the precipitate with a glass stirring rod, taking care that no glass is broken off of the stirring rod when grinding. Slurry and filter onto a previously washed and weighed Whatman #42 filter disc using anhydrous "Anhydrol" as a transfer agent. Dry in an oven at 110°C for 15 minutes, cool in a desiccator, weigh, and mount on a brass planchet. A chemical yield correction must be made due to natural phosphorus in the filter. An aliquot of the original sample is taken and measured colorometrically for such phosphorus.

F. Purification Procedures for Sodium-22

In the sodium-22 purification procedure demineralized water, plastic tubes, teflon beakers and stirrers are to be used exclusively.

1. Transfer the supernate from step 9 of the Sequential Separation Procedure to a 100-ml teflon beaker and evaporate to dryness.

2. Cool and add 1 ml of 6M NH_4OAc and 75 ml of "sodium-precipitating reagent" (45g uranyl acetate, 300 g magnesium acetate, 60 ml acetic acid).

3. Stir on a mechanical stirrer for 20 minutes; then centrifuge by portions in a plastic centrifuge tube and discard the supernate.

4. Wash the precipitate once with 20 ml of "sodium-precipitating reagent" and once with 20 ml of "sodium wash solution" (35ml acetic acid, 405 ml anhydrous ethyl acetate, 460 ml 100% ethanol).

5. Dissolve the precipitate in a minimum of concentrated HCl and dilute to 15 ml with water.

6. Add 3 drops of iron carrier (~10 mg/ml) and precipitate iron hydroxide with concentrated NH_4OH. Centrifuge and decant the supernate into a clean plastic centrifuge tube. Wash the precipitate with 5 ml deionized water; centrifuge, decant, wash and add to the supernate above. Discard the iron hydroxide precipitate.

7. Perform two additional iron hydroxide scavenges. Dissolve the iron hydroxide from each scavenge with concentrated HCl and then precipitate it with concentrated NH_4OH. Centrifuge, wash with deionized water, centrifuge and add decanted wash to supernate.

8. Transfer the supernate and wash from the final iron hydroxide scavenge to a teflon beaker and evaporate to dryness.

9. Cool and add 4 ml of concentrated HNO_3 and 1 ml of concentrated HCl and evaporate to dryness under very low heat.

10. Repeat step 9 three times washing down the walls of the beaker with water each time.

11. Add 5 ml of concentrated HCl and evaporate to dryness.

12. Dissolve the precipitate in a minimum of water and transfer to a plastic centrifuge tube. Wash the walls of the beaker with a minimum of water and add this to the tube.

13. Place the plastic tubes in an ice bath and add 15 ml of 1-Butanol saturated with HCl gas. Stir vigorously, and let stand until the ice bath comes to room temperature. To prepare 1-Butanol saturated with HCl, transfer 1/2 pint of 1-Butanol to another bottle. Place the remaining 1/2 pint in an ice salt bath and pass HCl gas through the solution for one hour. Keep this solution ice cold until ready for use. CAUTION A closed bottle of the saturated solution is explosive at room temperature.

14. Centrifuge and discard the supernate. Add 15 ml of 100% EtOH and stir vigorously. Filter onto a tared #42 Whatman filter paper using 100% EtOH only as a transfer solution.

15. Dry at 100°C for 30 minutes. Cool in a dessicator, weigh and record weight as NaCl. Mount for sodium-22 beta counting.

G. Radioassay Procedures

The 0.48 Mev γ radiation of beryllium-7 (53 day half life) is measured to obtain the abundance of that nuclide in STARDUST samples.

The beryllium is in the form of a moist $Be(HO)_2$ precipitate in a 10-ml test tube following the purification procedure. It is counted in this form in the test tube in a NaI(Tl) well-type crystal (1-3/4" x 2"). The height of the precipitate in the tube is measured and the counting efficiency is determined from a previously constructed calibration curve on which efficiency is plotted against height. All samples are assayed twice, and normally excellent agreement is obtained between the first and second counts. Usual counting periods are approximately an hour in length. Five per cent of the samples are followed for decay to use the half-life to check sample purity. A branching ratio of 11.00% for the gamma ray is used in computing beryllium-7 content.

The β-emitting nuclides phosphorus-32 and phosphorus-33 are counted using low-level, gas-flow detectors in the geiger region. Phosphorus-32 emits a 1.71 Mev β (half-life = 14.3d) and phosphorus-33, a 0.25 Mev β (half-life = 25d). The phosphorus precipitate from the purification procedure, counted on a brass planchet, is first counted without absorbers and with a minimum distance between sample and detectors for both phosphorus isotopes. Then the sample is recounted in the same geometry but with a 71.6 mg/cm^2 aluminum absorber between it and the detectors to cut out all phosphorus-33 betas. The sample is counted in this configuration three to six times to follow the decay of 14.3 day phosphorus-32 and the disintegration rate of that nuclide is calculated. The sample is counted without the absorber for the same length of time, and the calculated phosphorus-32 disintegration rate is subtracted from the total disintegration rate of the sample without absorber to obtain the disintegration rate for phosphorus-33. Each count is normally about 7 hours in duration.

Sodium-22, in the form of the final sodium chloride precipitate from the purification procedure, is counted with a low level gas flow detector in the geiger region. Sodium-22 emits 0.54 Mev positron radiation, and samples are counted for from twoo hundred to four hundred minutes. Sample purity is checked by counting with a 14.6 mg/cm^2 aluminum absorber.

CHAPTER 4. THE ANALYSIS OF STARDUST CARBON-14 SAMPLES

During Project STARDUST measurements of carbon-14 concentrations were made on samples of stratospheric air collected by WU-2, RB-57F and RB-57C aircraft, and also on samples of ground-level carbon dioxide collected in northern New Jersey. The stratospheric samples were processed at Air Reduction Company, Inc., and a small aliquot of each air sample plus the separated carbon dioxide were shipped to ISOTOPES, INC. for measurement. All processing of the ground-level samples was performed at ISOTOPES, INC.

4.1 The Measurement of Carbon-14 in Stratospheric Air Samples

The carbon dioxide samples received from the Air Reduction Company, Inc. were sufficiently pure that they could be counted after a minimum of additional purification. This was accomplished by passing the carbon dioxide through charcoal at room temperatures. Occasional tests were made to assure that no isotopic fractionation of the carbon dioxide occured during this final purification step. These tests involved measurements of C^{13}/C^{12} ratios and carbon-14 concentrations in carbon dioxide samples before and after passage through the charcoal.

The purified samples were introduced into internal gas proportional counters of copper with 1-ml tungsten anode center wires. The active volume of each counter was approximately 250 ml. The counters were shielded by 16 inches of steel and 4 inches of paraffin to remove the soft cosmic ray component of the background activity. The counts attributable to the hard cosmic ray component, cosmic ray mesons, were removed by an anticoincidence technique using an annular multi-anode anticoincidence guard counter surrounding each sample counter. This total shielding array reduced the background of the 250-ml counters to about 2.2 cpm.

Samples were counted sufficiently long to insure that the statistical counting errors were 5 per cent or less. The counters were calibrated using standard samples of $C^{14}O_2$ supplied by Argonne National Laboratory. The efficiency of the counters for the 15.6-kev beta of carbon-14 was 92 per cent.

The data obtained from the carbon-14 measurements yielded only the carbon-14 concentrations of the carbon dioxide in the whole air samples obtained from the aircraft sampling systems. As has been pointed out by Hagemann et al.[2], some carbon dioxide may have been added to the whole air samples by oxidation of compressor lubricating oil in the aircraft sampling systems. Because this oil was derived from fossil fuels, the carbon dioxide produced from it should have added no carbon-14 to the sample, but rather would have acted to decrease the concentration of carbon-14 in the carbon dioxide of the air. This possible contamination was corrected for in STARDUST samples by measuring the actual carbon dioxide concentrations in the whole air samples as well as the carbon-14 concentration in the carbon dioxide, and then calculating the carbon-14 concentration in the whole air samples. As long as no significant increase in the volume of the air samples was produced by contaminants from the compressor, this carbon-14 concentration could be taken as equal to that in the uncontaminated air sample.

For 76 of the first STARDUST samples collected during 1963 reliance must be placed on infrared measurements of carbon dioxide abundances in the whole air samples made at Air Reduction Company, Inc. during the processing of the samples. For subsequent samples, however, measurements of carbon dioxide abundance were made at ISOTOPES, INC. on the air aliquots supplied with the separated carbon dioxide samples. An isotopes dilution technique utilizing mass spectrometer measurements was used. The measurements of abundance made were precise to within ± 2%. The advantage of this method over the infrared measurements was that its accuracy was not affected by the presence of traces of contaminants such as methane or carbon monoxide in the whole air sample.

To perform a carbon dioxide abundance measurement an accurately measured aliquot of the whole air sample was taken, and to it was added an accurately measured aliquot of a standard "spike" sample enriched in $C^{13}O_2$. The mixture was equilibrated by two successive freeze-outs in a cold finger at liquid nitrogen temperatures, and gases volatile at these temperatures, such as oxygen, nitrogen and carbon monoxide, were pumped off of the frozen sample.

The C^{13}/C^{12} ratio in the mixture was measured using a Nuclide Corporation RMS-15 multiple collector ratio mass spectrometer. The ratio measured was that of ions of mass 45 ($C^{13}O_2$) to ions of mass 44 ($C^{12}O_2$). At least ten successive determinations of the ratio in the mixture of sample and spike were alternated with at least ten determinations of the ratio in a standard sample. The computed average ratio in the mixture was precise to within ± 0.1%. Since the quantity of carbon-13 contributed to the mixture by the whole air sample was negligible compared to the quantity contributed to the mixture by the enriched carbon-13 spike sample, the amount of carbon-12 contributed by the whole air sample could be calculated from the known C^{13}/C^{12} ratio in the spike and the measured C^{13}/C^{12} ratio in the mixture. Since the aliquot of air which contributed this carbon-12 was measured accurately, this information yielded the the carbon-12 content, and therefore the carbon dioxide content, of the air aliquot.

The values of carbon-14 abundance in the whole air samples, calculated from the results of the carbon-14 and carbon-dioxide measurements, should generally have been accurate to within ± 5%. They were expressed in atoms of carbon-14 per gram of air. To correct these samples for the presence of carbon-14 produced by cosmic ray activity it was assumed, following Hagemann et al, that each sample contained 74×10^5 atoms/g of natural carbon-14, and this amount was subtracted from each result to obtain the concentration of "excess carbon-14".

4.2 The Measurement of Carbon-14 in Ground-Level Carbon Dioxide Samples

The concentration of carbon-14 in carbon dioxide in ground-level air was monitored at the Township of Washington, Bergen County, in northern New Jersey, from January 1960 to July 1967. Samples were generally obtained by placing 50 to 100 ml of 4M KOH solution in two or three petri dishes of 10 cm diameter, and exposing this solution to the atmosphere. Before using the solution all carbon dioxide was removed by the addition of barium chloride and filtration to remove the resulting $BaCO_3$ precipitate. Atmospheric carbon dioxide was gradually absorbed by the solution in the petri dishes, which were exposed to the atmosphere in a covered wire-mesh enclosure. Periodically the petri dishes were emptied into a one-liter polyethylene bottle. At the end of the desired sampling interval this bottle was taken to the laboratory and the carbonate in the solution was analyzed for carbon-14.

The length of sampling intervals used was different at different times during the program. During most of 1960 the sampling interval was generally about a week, and the solution in the petri dishes was changed about once a day. During 1961 to mid-1965 the sampling interval was about two weeks, and the solution in the petri dishes was changed once every two or three days. During mid-1965 to mid-1967 the sampling interval was about one month, and the solution was changed once or twice per week.

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During January-February 1967 a series of samples was collected representing sampling intervals of one to three days. A larger dish, containing about 1000 ml of KOH solution constituting a single sample was used.

The carbonate-hydroxide solution was placed in a flask in a vacuum system, and the system was evacuated to a pressure of approximately 200 mm of mercury. Acetic acid, containing Methyl Red indicator, was then slowly admitted to the flask, and the solution was stirred using a magnetic stirrer. The carbon dioxide which was evolved was collected in a trap at liquid nitrogen temperature. The reaction was considered complete when the solution became clear, and when the Methyl Red indicator retained its color after its addition with the acetic acid to the sample solution. When the reaction was complete the trap containing the frozen carbon dioxide was pumped for 10 to 30 minutes to remove any volatile impurities which may freeze out at liquid nitrogen temperatures.

The separated carbon dioxide was usually fairly pure, except for the presence of some water vapor and radon. To remove these the carbon dioxide was reacted with calcium oxide in a quartz bulb at 750°C to form calcium carbonate. The radon and water vapor, which did not react with the calcium oxide, were pumped away. When the reaction was complete the calcium carbonate mixture was cooled to 400°-450°C, and evacuated to a pressure of less than one micron as indicated by a Pirani gauge. The quartz bulb was then isolated from the vacuum pump and heated to 950°C to decompose the calcium carbonate. The released carbon dioxide was collected in a trap at liquid nitrogen temperature, and was then stored or counted.

The gas was counted in proportional counters constructed of electrolytic copper with active volumes just under two liters. To provide a low background they were shielded by a ring of G-M cosmic ray counters operated in anti-coincidence with the sample counter, and by four inches of paraffin wax and 18 inches of steel. The average background of the counters with this arrangement was 2.2 counts per minute. The associated electronics were specially constructed at ISOTOPES, INC. for use in the radiocarbon dating program. Each sample was counted for two periods of 6 to 15 hours each several days apart to verify the reproducibility of the measured count rates.

The U.S. National Bureau of Standards supplies an oxalic acid modern standard for radiocarbon dating laboratories. A value of 0.95 times the activity of this oxalic acid standard is used as the modern reference value for radiocarbon dating. The carbon-14 activities of all samples measured in this program were compared with this standard value. The carbon-14 from this standard (at 273°K) assayed weekly at two atmospheres averaged 22 counts per minute. The individual determinations were within the 95 percent confidence limit as calculated from counting statistics based on the total number of counts observed for a single sample. Carbon dioxide, from carbonaceous materials older than 50,000 years containing an insignificant amount of carbon-14 was counted several times a week to ensure that the background of the counter was stable. The background counts were also within the 95 percent confidence limit as determined by the standard deviation based on the total counts from a single determination.

CHAPTER 5. THE PROPERTIES OF THE STRATOSPHERIC AEROSOL

The study of stratospheric particles was undertaken initially in Project HASP in order to provide information on the physical and chemical nature of the material with which world-wide fallout is associated. This study was continued in Project STARDUST to obtain more reliable measurements of these properties, using a sampling probe available in late 1961 which was superior in several respects to that used in Project HASP. The resultant measurements have shed light on several aspects of the stratospheric aerosol.

Junge et al.[3] who used impactor surfaces carried by balloons to sample stratospheric particles found that the preponderance of particles in the 15 to 25 km altitude region contained sulfur, and had radii in the range 0.1 to 1.0 micron. Measurements performed during Project HASP, and reported in DASA 1300, the Final Report on Project HASP, indicated that the particles consisted primarily of ammonium sulfate, and possibly ammonium persulfate. The HASP measurements of the frequency distribution of the radii of the particles, as well as the measurements performed by Junge and coworkers[3,4], indicated that in the size range between 0.1 micron and 1.0 micron, the number of particles in each size class decreased as the radius increased. Few, if any, sulfate particles were observed with radii greater than 1.0 micron. Mossup[5], reporting results of measurements of samples collected in 1963 by WU-2 aircraft, found that the sulfate particles, which are water soluble, contain one or more particles which are insoluble in water, and which range in radius from 0.005 to 0.4 micron. He also found that the frequency distribution of radii for the stratospheric aerosol was peaked in the vicinity of 0.3 to 0.4 micron.

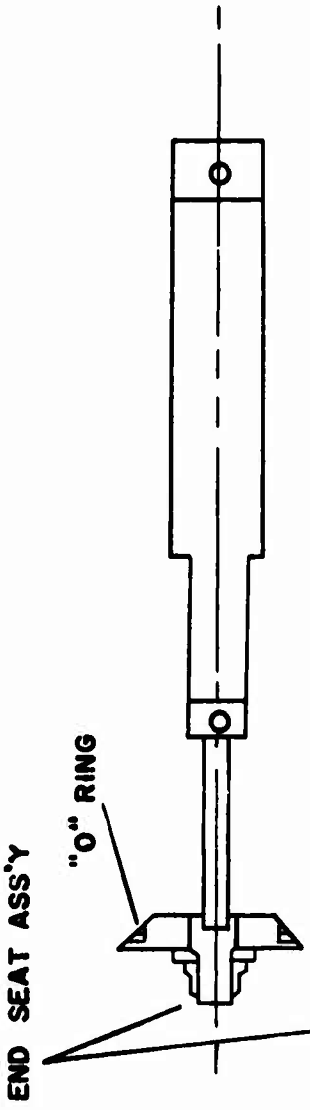
The results reported and discussed in this chapter concern samples collected between January 1962 and August 1963 by WU-2 aircraft. It is the purpose of this discussion to describe what is currently known of the physical properties of the stratospheric aerosol, and what factors are significant with regard to the origin and distribution of the particles.

The work reported here was described previously in the Ninth Progress Report on Project STARDUST, DASA 1309, was reported at the "CACR Symposium on Atomospheric Chemistry, Circulation and Aerosols" in Visby, Sweden, 18-25 August 1965, and was discussed in the Proceedings of that Symposium [6].

5.1 The Collection and Measurement of Stratospheric Particles

The methods of particle sampling and analysis used during Project STARDUST were basically the same as those used during Project HASP, which are described in DASA 1300. The particles were collected by direct flow impactors mounted in a probe on WU-2 aircraft. The main analytical tool used in the analysis of the samples was electron microscopy.

Until October 1961 only the "window" type of sampler was available for use on the WU-2 aircraft. At that time two "tip" probes (see DASA 1300, Volume 5, Part II, Figure 2.1) and three newly designed "Mark III" probes were made available. The Mark III probes, shown schematically in Figure 3, were designed by Mr. James E. Manson, then of the Air Force Canbridge Research Laboratory. In mid-1962 all aircraft collecting STARDUST samples were modified to carry the Mark III probes, and a total of six were made available for use. Between 5 January 1961 and 12 December 1963 10 "tip" and 42 Mark III samples were collected but only 12 were of suitable condition to be subjected to thorough particle size analysis.



IMPACTION SURFACE OF MARK III PROBE

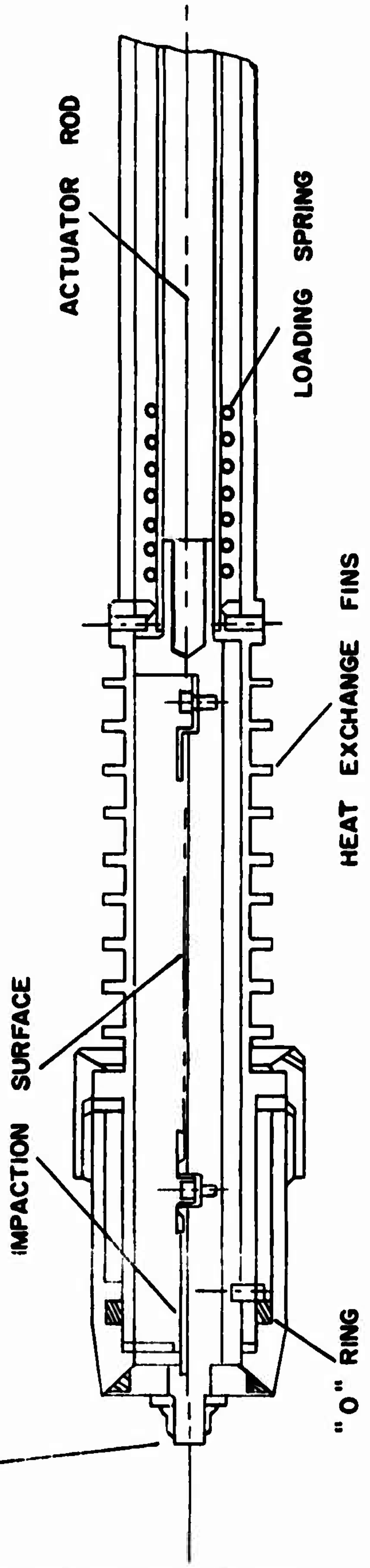


FIGURE 3. MARK III IMPACTOR PROBE ASSEMBLY

The basic design features of the Mark III impaction probe are shown in Figure 3. The impaction area had two different widths, permitting collection of larger particles on the wider portion without interference by particles smaller than the "cut-off" radius. The narrow portion had, of course, a "cut-off" at much smaller radius, and in this work, only the deposit on the narrow portion was studied. Because of their small size, the major fraction of the particles of the stratospheric sulfate aerosol were deposited upon the narrow portion of the probe.

Experience with the "window" impactor was used in the development of the improved design of the Mark III probe. An air-tight housing was provided for the new probe to minimize condensation of moisture from tropospheric air onto the hygroscopic particles of the stratospheric aerosol following the decent of the sampling aircraft below the tropopause. The main seal is provided by the "o-ring" in the end cap of the sample plunger. In addition, heat exchange fins were placed on the probe to speed the rate at which it would come into thermal equilibrium with the ambient air. This diminished the tendency for water to condense on the probe when the aircraft descended from the stratosphere into the warmer and more moist troposphere. A coating of silicone oil was applied to the outer coating of the probe before each sampling flight also to prevent icing.

The Mark III probe was also designed to have the sampling surface extend beyond the body of the probe. This eliminated the "dead air" space which caused peculiar banding of the deposit of particles in the "window" probe. The result is that the impactor surface approximates a ribbon type of impactor rather than a cylinder type. The small width of the impaction surface (0.32 cm) permits collection of particles down to 0.14 micron radius with efficiencies greater than 40 percent at an altitude of 18 km. The equivalent efficiency for the "window" probe was less than one percent.

The "tip" probe as well as the Mark III probe had narrow impaction surfaces which extended beyond the body of the probe, but there were basic differences in the overall shape and size of the extensible portions of the two devices.

The field operational aspects of the Mark III probe were essentially the same as those of the "window" probe, although there were changes in packaging for shipment.

The preparation of the probes for sampling, the unloading of the probes after exposure, and the electron microscopic and electron diffraction analyses were performed by Ernest F. Fullam, Inc., Schenectady, N. Y.

The collection surface for all samples consisted of a thin carbon film supported by a thick (0.003 to 0.005 inch) film of nitrocellulose. Three specimen grids from each sample were prepared for electron microscopic analysis by placing copper grids beneath the original thick film and dissolving the film of nitrocellulose in vapors of amyl acetate. The thin carbon film with the sample particles was left supported by the copper grid. Although there was still a "banding" or "pile-up" effect at the inboard end of the Mark III probe samples it was not as pronounced as in samples from the "window" probe. The micrographs used for determining the particle size-frequency distribution were chosen so that proper weighting for the "pile-up" was attained in each sample. Normally, about 5 to 10 micrographs, each representing 1440 square microns of sample, were counted.

Electron diffraction patterns were obtained for many of the samples to determine the chemical composition of the particles. Patterns were obtained for single particles or for a few particles in a small area of the sample.

The sulfate particles, which comprise almost all of the mass of the stratospheric aerosol, were generally flat rosettes, and often were accompanied by haloes of very small particles. Electron microscopic examination of shadowed deposits indicated that impacted particles with radii greater than 0.2 microns had thicknesses of only 0.2 microns. From the measured radius and this thickness the volume of these particles could be calculated. By assuming that all particles were originally spherical the radius prior to collection could then be calculated from the volume.

As in the case of the HASP samples, it was necessary to apply theoretical impaction efficiencies to the observed distributions of particle sizes. The impaction parameter, Ψ , was calculated for a given particle size according to the definition:

$$\Psi = \frac{C\rho_p V_o D_p^2}{18\mu D_c}$$

where: C = Cunningham correction factor for gas resistance experienced by small particles,

D_p = diameter of the particles,

D_c = width of the collector,

ρ_p = density of the particulate material,

μ = coefficient of viscosity of the air,

V_o = velocity of the free aerosol stream relative to the collector.

The theoretical relationship between $\sqrt{\Psi}$ and the impaction efficiency η for a hypothetical, infinitesimally thin ribbon, after Ranz and Wong [7], is shown in Figure 4. The values of Ψ were computed for particular values of the particle diameter, D_p, utilizing flight data to obtain C (a function of altitude and D_p), V_o and μ (a function of temperature only), and using $D_c = 0.32$ cm and $\rho_p = 2$ g/cm^3. (The actual density of ammonium sulfate is 1.78 g/cm^3 while that of ammonium persulfate is 1.98 g/cm^3.)

In this work the value of μ is 1.4222×10^{-4} poise corresponding to a temperature of 216.66°K, which is the ARDC Model Atmosphere value for the stratosphere. A 10°K temperature difference would result in less than 10% change in the calculated number concentration. The percentage changes are smaller for the larger particles which are collected with higher efficiencies.

5.2 Results of The Particle Measurements

The flight data needed for the computation of the concentrations in air of the various particle size fractions are given for each sample in Table 8.

In Table 9 are given the observed frequencies of occurrence of the particles in each size class for each sample and the number concentrations and volume concentrations calculated for the sample. The listed values of radius are the mean radii of the class intervals for which the count data were determined. The column headed $^1/_\eta$ is the reciprocal of the collection efficiency. The listed observed frequencies of particles of the various classes are multiplied by $^1/_\eta$ and divided by the volume of air sampled to give the values of $\bar{n}_i$, the concentrations of particles in the various classes. The total number concentration for a sample is found by summing the values of $\bar{n}_i$. The volume concentration, $\bar{V}$, is computed by:

$$\bar{V} = \frac{4}{3} \pi \sum \bar{n}_i \quad r_i^3 = \sum \bar{V}_i$$

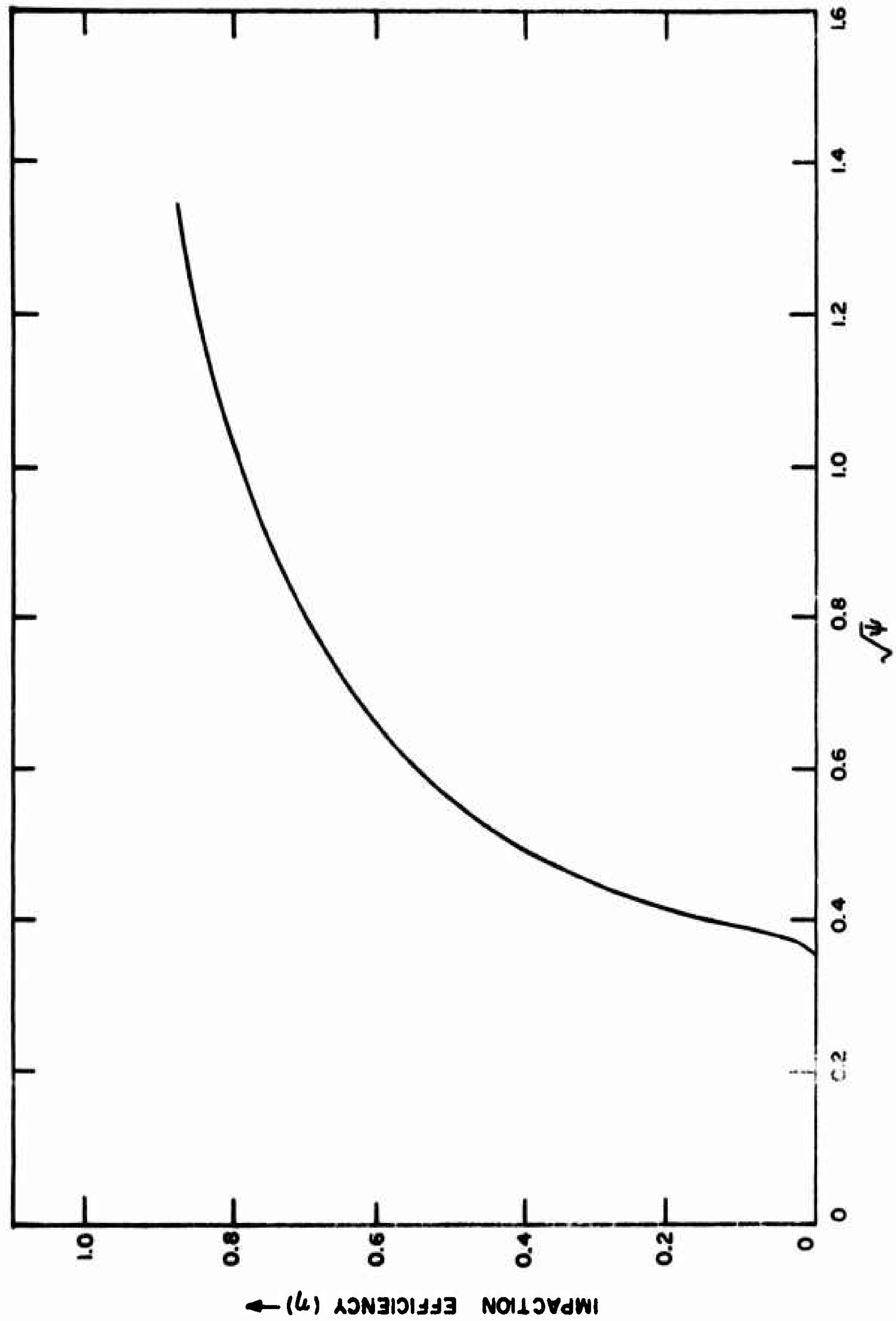


FIGURE 4 THEORETICAL IMPACTION EFFICIENCIES OF A RIBBON (AFTER RANZ AND WONG)

TABLE 8. Flight Data for STARDUST Impactor Samples

Sample Number	Collection Date	Latitude	Longitude	Altitude (km)	TAS* (Knots)	Sampling Time (Minutes)	Sample Volume (cm^3)
243	13 Feb 1962	49°-32°N	112°-100°W	18.3	410	169	2.14×10^8
244	13 Feb 1962	49°-32°N	112°-100°W	15.2	380	191	2.24×10^8
245	13 Mar 1962	49°-31°N	112°-101°W	12.2	324	209	2.09×10^8
252	19 Jun 1962	49°-31°N	112°-101°W	15.2	384	194	2.30×10^8
254	12 Jun 1962	49°-31°N	112°-101°W	16.8	403	175	2.18×10^8
257	26 Jun 1962	48°-31°N	112°-101°W	16.8	389	180	2.16×10^8
258	14 Feb 1963	45°-39°N	109°-104°W	18.3	411	60	0.76×10^8
259	19 Feb 1963	31°-20°N	100°-96°W	18.3	406	120	1.50×10^8
265	2 Apr 1963	32°-31°N	100°-99°W	18.3	407	60	0.75×10^8
266	7 May 1963	33°-31°N	100°-100°W	18.3	415	60	0.77×10^8
268	30 Jul 1963	33°-32°N	111°-111°W	18.3	415	60	0.77×10^8
271	13 Aug 1963	32°-32°N	111°-111°W	18.3	415	60	0.77×10^8

* True Air Speed

TABLE 9. Particle Size Measurements of STARDUST Impactor Samples

Sample No. [illegible]:

Radius (μ)	$\frac{1}{\eta}$ [a]	Observed Frequency $\times 10^{-4}$ (cm^{-2}) [b]	$\bar{n} \times 10^{5}$ (cm^{-3})	$\bar{V} \times 10^{5}$ (μ^{3}/cm^{3}) [d]
.138	1.82	11.88	101	1.11
.160	1.69	37.44	296	5.02
.182	1.59	51.77	385	9.66
.199	1.54	44.15	318	10.5
.224	1.46	73.06	499	23.4
.244	1.41	108.36	714	43.5
.264	1.38	98.20	633	48.7
.282	1.36	101.18	643	60.4
.300	1.33	42.18	262	29.6
.318	1.30	39.67	241	32.3
.336	1.28	16.83	100	15.9
.352	1.26	15.59	91.3	16.6
.370	1.25	11.98	70.0	14.9
.386	1.23	4.92	28.3	6.78
.402	1.22	5.32	30.3	8.24
.418	1.21	1.95	11.0	3.35
.432	1.20	4.73	26.5	8.93
.448	1.18	0.36	2.01	0.76
.462	1.18	0.45	2.48	1.02
.478	1.17	0.22	1.17	0.53
.492	1.16	0.14	0.75	0.37

Number Concentration: 4.46×10^{-2} cm^{-3}
Volume Concentration: 3.42×10^{-3} μ^{3}/cm^{3}

a Inverse Collection Efficiency
b Frequency of occurrence within size range
c Number concentrations of particles
d Volume concentrations of particles

TABLE 9. (continued)

Sample No. 244:

Radius (μ)	$\frac{1}{\eta}$	Observed Frequency x 10^{-4} (cm^{-2})	$\bar{n} \times 10^5$ (cm^{-3})	$\bar{V} \times 10^5$ (μ^3/cm^3)
.138	2.93	1.47	19.2	0.211
.160	2.41	4.96	53.6	0.908
.182	2.08	4.26	39.3	0.982
.199	1.91	6.11	51.9	1.71
.224	1.87	6.58	54.6	2.57
.244	1.68	13.90	104	6.36
.264	1.61	18.76	135	10.4
.282	1.56	23.51	164	15.3
.300	1.52	23.51	160	18.1
.318	1.48	25.08	166	22.2
.336	1.44	17.00	109	17.3
.352	1.41	14.52	92.0	16.7
.370	1.39	7.06	43.6	9.24
.386	1.37	6.74	41.1	9.87
.402	1.35	0.16	0.968	0.263
.418	1.33	0.31	1.84	0.560
.432	1.32	0.31	1.82	0.614
.448	1.30	-	-	-
.462	1.28	-	-	-
.478	1.26	-	-	-
.492	1.26	0.16	0.908	0.452
.614	1.18	0.16	0.849	0.822

Number Concentration: 1.24×10^{-2} cm^{-3}
Volume Concentration: 1.33×10^{-3} μ^3/cm^3

TABLE 9. (continued)

Sample No. 245:

Radius (μ)	$\frac{1}{\eta}$	Observed Frequency x 10^{-4} (cm^{-2})	$\bar{n}$ x 10^5 (cm^{-3})	$\bar{V}$ x 10^5 (μ^3/cm^3)
.138	-	-	-	-
.160	-	3.87	-	-
.182	-	6.58	-	-
.199	3.57	4.00	68.2	2.25
.224	2.76	3.23	42.4	1.99
.244	2.44	12.21	142	8.70
.264	2.19	11.95	124	9.57
.282	2.02	24.81	239	22.5
.300	1.90	28.89	262	29.6
.318	1.82	21.06	184	24.6
.336	1.74	24.49	203	32.1
.352	1.69	13.66	110	20.1
.370	1.63	4.00	31.1	6.59
.386	1.59	1.42	10.8	2.58
.402	1.56	1.81	13.6	3.69
.418	1.52	0.52	3.80	1.16
.432	1.50	0.13	1.05	0.36
.448	1.47	0.26	1.91	0.72
.462	1.44	0.13	1.01	0.42
.478	1.41	0.39	2.68	1.22
.492	1.40	-	-	-
.506	1.38	0.13	0.96	0.52
.520	1.37	0.13	0.96	0.57

Number Concentration: 1.44×10^{-2} cm^{-3}
Volume Concentration: 1.69×10^{-3} μ^3/cm^3

TABLE 9. (continued)

Sample No. 252:

Radius (μ)	$\frac{1}{\eta}$	Observed Frequency x 10^{-4} (cm^{-2})	$\bar{n} \times 10^5$ (cm^{-3})	$\bar{V} \times 10^5$ (μ^3/cm^3)
.138	2.93	8.93	114	1.25
.160	2.41	11.77	123	2.10
.182	2.08	13.98	127	3.16
.199	1.91	17.57	146	4.81
.224	1.87	13.73	112	5.28
.244	1.68	0.87	6.38	0.39
.264	1.61	3.86	27.0	2.08
.282	1.56	15.64	106	9.89
.300	1.52	14.38	94.7	10.7
.318	1.48	12.28	79.0	10.6
.336	1.44	9.72	60.7	9.60
.352	1.41	5.10	31.3	5.7
.370	1.39	0.19	1.11	0.24
.386	1.37	2.89	17.2	4.14
.402	1.35	4.91	28.8	7.82
.418	1.33	2.50	14.5	4.41
.432	1.32	0.29	1.72	0.57
.448	1.30	2.70	15.2	5.73
.462	1.28	0.29	1.67	0.68
.478	1.26	7.03	38.5	17.6
.492	1.26	0.19	1.00	0.50
.506	1.25	2.50	13.6	7.37
.520	1.24	2.31	12.4	7.29
.534	1.23	0.19	0.98	0.63
.548	1.22	2.31	12.2	8.39
.560	1.21	0.10	0.48	0.36
.574	1.20	-	-	-
.588	1.19	-	-	-
.600	1.18	-	-	-
.614	1.18	0.10	0.47	0.46

Number Concentration: 1.19×10^{-2} cm^{-3}
Volume Concentration: 1.31×10^{-3} μ^3/cm^3

TABLE 9. (continued)

Sample No. 254:

Radius (μ)	$\frac{1}{\eta}$	Observed Frequency x 10^{-4} (cm^{-2})	$\bar{n} \times 10^5$ (cm^{-3})	$\bar{V} \times 10^5$ (μ^3/cm^3)
.138	2.22	4.68	47.7	0.52
.160	1.94	9.24	82.6	1.40
.182	1.77	11.74	95.6	2.39
.199	1.69	5.19	40.2	1.33
.224	1.59	6.71	49.0	2.30
.244	1.54	16.72	118	7.21
.264	1.47	24.25	164	12.6
.282	1.44	31.51	208	19.6
.300	1.41	50.24	325	36.7
.318	1.38	51.80	328	43.9
.336	1.36	39.68	248	39.1
.352	1.34	21.95	135	24.5
.370	1.31	26.66	160	33.9
.386	1.30	12.88	76.8	18.4
.402	1.28	11.61	68.2	18.6
.418	1.26	2.91	16.9	5.15
.432	1.25	5.56	31.8	10.8
.448	1.24	2.78	15.9	5.97
.462	1.23	-	-	-
.478	1.22	0.13	0.73	0.33
.492	1.21	-	-	-
.506	1.20	1.39	7.68	4.16

Number Concentration: 2.22×10^{-2} cm^{-3}
Volume Concentration: 2.89×10^{-3} μ^3/cm^3

TABLE 9. (continued)

Sample No. 257:

Radius (μ)	$\frac{1}{\eta}$	Observed Frequency x 10^{-4} (cm^{-2})	$\bar{n} \times 10^5$ (cm^{-3})	$\bar{V} \times 10^5$ (μ^3/cm^3)
.138	2.22	0.87	8.89	0.10
.160	1.94	4.93	44.3	0.75
.182	1.77	10.92	89.2	2.24
.199	1.69	9.95	77.7	2.57
.224	1.59	8.01	58.8	2.76
.244	1.54	14.88	106	6.45
.264	1.47	39.24	267	20.5
.282	1.44	59.43	396	37.3
.300	1.41	52.37	342	38.6
.318	1.38	26.52	169	22.6
.336	1.36	18.62	117	18.5
.352	1.34	19.65	121	22.2
.370	1.31	12.84	77.8	16.5
.386	1.30	9.47	57.0	13.6
.402	1.28	4.93	29.1	7.92
.418	1.26	3.38	19.7	6.00
.432	1.25	3.38	19.5	6.56
.448	1.24	1.26	7.19	2.71
.462	1.23	1.06	6.03	2.49
.478	1.22	0.87	4.88	2.22
.492	1.21	1.06	5.93	2.95
.506	1.20	2.13	11.7	6.35

Number Concentration:	2.04×10^{-2} cm^{-3}
Volume Concentration:	2.42×10^{-3} μ^3/cm^3

TABLE 9. (continued)

Sample No. 258:

Radius (μ)	$\frac{1}{\eta}$	Observed Frequency x 10^{-4} (cm^{-2})	$\bar{n}$ x 10^5 (cm^{-3})	$\bar{V}$ x 10^5 (μ^3/cm^3)
.138	1.82	0.77	18.4	0.202
.160	1.69	4.90	109	1.84
.182	1.59	5.13	107	2.68
.199	1.54	6.10	123	4.07
.224	1.46	16.80	321	15.1
.244	1.41	24.80	460	28.0
.264	1.38	66.60	1,204	92.3
.282	1.36	62.90	1,120	105
.300	1.33	47.25	834	93.8
.318	1.30	43.32	738	99.1
.336	1.28	27.12	455	72.0
.352	1.26	21.90	362	65.9
.370	1.25	14.35	235	49.9
.386	1.23	17.90	289	69.4
.402	1.22	5.93	94.5	25.7
.418	1.21	10.35	164	50.2
.432	1.20	5.54	87.2	29.4
.448	1.18	1.77	27.4	10.3
.462	1.18	3.74	58.6	24.2
.478	1.17	0.97	14.8	6.78
.492	1.16	2.16	32.8	16.3
.506	1.15	0.77	11.6	6.29
.520	1.15	0.39	5.86	3.45
.534	1.14	0.39	5.81	3.70

Number Concentration: 6.88×10^{-2} cm^{-3}
Volume Concentration: 8.76×10^{-3} μ^3/cm^3

TABLE 9. (continued)

Sample No. 259:

Radius (μ)	$\frac{1}{\eta}$	Observed Frequency x 10^{-4} (cm^{-2})	$\bar{n} \times 10^5$ (cm^{-3})	$\bar{V} \times 10^5$ (μ^3/cm^3)
.138	1.82	16.3	196	2.16
.160	1.69	40.4	453	7.71
.182	1.59	27.9	294	7.36
.199	1.54	22.3	228	7.51
.224	1.46	86.0	834	39.2
.244	1.41	106.5	996	60.7
.264	1.38	83.3	760	58.5
.282	1.36	87.4	788	74.1
.300	1.33	31.9	285	32.2
.318	1.30	40.9	354	47.4
.336	1.28	18.3	156	24.7
.352	1.26	15.1	126	22.9
.370	1.25	8.6	71.2	15.1
.386	1.23	11.8	97.3	23.3
.402	1.22	8.6	69.5	18.9
.418	1.21	6.4	52.0	15.9
.432	1.20	5.9	46.8	15.8
.448	1.18	0.9	7.08	2.66
.462	1.18	0.9	7.08	2.92
.478	1.17	0.7	4.68	2.14
.492	1.16	0.5	3.48	1.74
.506	1.15	0.3	2.30	1.24
.520	1.15	0.6	4.60	2.70
.534	1.14	0.2	1.14	0.725
.548	1.13	0.5	3.39	2.33
.560	1.13	0.2	1.13	0.830
.574	1.12	0.2	1.12	0.885
.588	1.11	0.1	1.11	0.942
.600	1.11	0.1	1.11	0.997
.614	1.10	0.3	2.20	2.12
.626	1.10	0.1	1.10	1.12
.640	1.09	0.1	1.09	1.20
.652	1.08	0.0	-	-

Number concentration: 5.85×10^{-2} cm^{-3}
Volume Concentration: 4.98×10^{-3} μ^3/cm^3

TABLE 9. (continued)

Sample No. 265:

Radius (μ)	$\frac{1}{\eta}$	Observed Frequency x 10^{-4} (cm^{-2})	$\bar{n}$ x 10^5 (cm^{-3})	$\bar{V}$ x 10^5 (μ^3/cm^3)
.138	1.82	-	0	0
.160	1.69	-	0	0
.182	1.59	-	0	0
.199	1.54	3.73	76.0	2.51
.224	1.46	1.34	25.9	1.22
.244	1.41	4.87	90.8	5.53
.264	1.38	8.68	159	12.2
.282	1.36	23.55	425	39.9
.300	1.33	26.87	478	54.1
.318	1.30	13.94	240	32.2
.336	1.28	30.95	525	82.9
.352	1.26	24.46	409	74.4
.370	1.25	19.77	327	69.4
.386	1.23	18.07	295	70.7
.402	1.22	13.41	216	58.9
.418	1.21	3.08	49.3	15.1
.432	1.20	1.67	26.5	8.93
.448	1.18	0.36	5.66	2.13
.462	1.18	0.80	12.4	5.13
.478	1.17	0.27	4.22	1.92
.492	1.16	0.88	13.5	6.75
.506	1.15	0.44	6.67	3.62
.520	1.15	0.62	9.44	5.55
.534	1.14	1.15	17.3	11.0
.548	1.13	0.09	1.35	0.934
.560	1.13	0.09	1.36	0.998
.574	1.12	0.36	5.37	4.24
.588	1.11	0.9	13.2	11.3
.600	1.11	0.9	13.2	11.8

Number Concentration: 3.45×10^{-2} cm^{-3}
Volume Concentration: 5.93×10^{-3} μ^3/cm^3

TABLE 9. (continued)

Sample No. 266:

Radius (μ)	$\frac{1}{\eta}$	Observed Frequency x 10^{-4} (cm^{-2})	$\bar{n} \times 10^5$ (cm^{-3})	$\bar{V} \times 10^5$ (μ^3/cm^3)
.138	1.82	0	0	0
.160	1.69	0	0	0
.182	1.59	0	0	0
.199	1.54	1.27	25.4	0.84
.224	1.46	10.16	193	9.06
.244	1.41	21.59	396	24.1
.264	1.38	40.64	730	56.2
.282	1.36	92.71	1,636	154
.300	1.33	140.97	2,466	279
.318	1.30	130.81	2,209	296
.336	1.28	139.70	2,322	367
.352	1.26	97.79	1,598	290
.370	1.25	96.52	1,572	333
.386	1.23	77.47	1,239	297
.402	1.22	93.98	1,486	404
.418	1.21	68.58	1,081	330
.432	1.20	60.96	951	320
.448	1.18	40.64	623	234
.462	1.18	40.64	623	256
.473	1.17	45.72	696	318
.492	1.16	20.32	306	152
.506	1.15	19.05	285	154
.520	1.15	17.78	266	156
.534	1.14	12.70	188	120
.548	1.13	13.97	205	141
.560	1.13	3.81	55.9	41.0
.574	1.12	13.97	203	161
.588	1.11	5.08	73.2	62.2
.600	1.11	0	-	-
.614	1.10	1.27	18.2	17.6
.626	1.10	5.08	72.6	74.0
.640	1.09	1.27	17.9	19.7
.652	1.08	0	-	-
.664	1.08	1.27	17.8	21.8
-	-	-	-	-
.732	1.05	1.27	17.3	28.4

Number Concentration: 2.16×10^{-1} cm^{-3}
Volume Concentration: 5.12×10^{-2} μ^3/cm^3

TABLE 9. (continued)

Sample No. 268:

Radius (μ)	$\frac{1}{\eta}$	Observed Frequency x 10^{-4} (cm^{-2})	$\bar{n}$ x 10^5 (cm^{-3})	$\bar{V}$ x 10^5 (μ^3/cm^3)
.138	1.82	0	0	0
.160	1.69	12.7	279	4.74
.182	1.59	125.7	2,600	65.0
.199	1.54	247.6	4,961	164
.224	1.46	167.6	3,184	150
.244	1.41	123.2	2,256	137
.264	1.38	72.4	1,301	100
.282	1.36	58.4	1,031	96.9
.300	1.33	58.4	1,021	115
.318	1.30	64.8	1,091	146
.336	1.28	43.2	719	113
.352	1.26	55.9	913	166
.370	1.25	48.3	784	166
.386	1.23	35.6	569	137
.402	1.22	34.3	544	148
.418	1.21	20.3	320	97.9
.432	1.20	19.0	297	99.7
.448	1.18	14.0	214	80.4
.462	1.18	8.89	136	56.3
.478	1.17	6.35	96.6	44.0
.492	1.16	11.43	173	85.9
.506	1.15	6.35	94.6	51.3
.520	1.15	2.54	38.0	22.3
.534	1.14	2.54	37.6	23.9
.548	1.13	0	0	0
.560	1.13	1.27	18.6	13.6
.574	1.12	5.08	73.9	58.4
.588	1.11	5.08	73.2	62.2
.600	1.11	1.27	18.3	16.6
.614	1.10	1.27	18.2	17.6
.626	1.10	1.27	18.1	18.5
.640	1.09	0	0	0
.652	1.08	1.27	17.8	20.7
.664	1.08	1.27	17.8	21.8
.676	1.08	0	0	0

Number Concentration: 2.29 x 10^{-1} cm^{-3}
Volume Concentration: 2.50 x 10^{-2} μ^3/cm^3

TABLE 9. (continued)

Sample No. 271:

Radius (μ)	$\frac{1}{\eta}$	Observed Frequency x 10^{-4} (cm^{-2})	$\bar{n} \times 10^5$ (cm^{-3})	$\bar{V} \times 10^5$ (μ^3/cm^3)
.138	1.82	0	0	0
.160	1.69	1.59	34.9	0.60
.182	1.59	12.70	263	6.57
.199	1.54	6.38	128	4.22
.224	1.46	22.30	423	19.9
.244	1.41	17.50	321	19.6
.264	1.38	31.80	570	44.0
.282	1.36	27.00	477	44.9
.300	1.33	27.00	471	53.2
.318	1.30	33.40	564	75.6
.336	1.28	33.40	556	87.9
.352	1.26	19.10	313	57.0
.370	1.25	20.70	336	71.3
.386	1.23	9.55	152	36.5
.402	1.22	3.17	50.3	13.7
.418	1.21	3.17	50.7	15.4
.432	1.20	9.55	149	50.0
.448	1.18	9.55	146	55.1
.462	1.18	1.59	24.4	10.0
.478	1.17	1.59	24.2	11.0
.492	1.16	0	0	0

Number Concentration: 5.05×10^{-2} cm^{-3}
Volume Concentration: 6.76×10^{-3} μ^3/cm^3

Table 10 is a summary of the number concentrations and volume concentrations of the stratospheric particles in STARDUST impactor samples. The table contains information concerning sampling date and location, and the samples are grouped according to altitude of collection. The table also lists the chemical compositions of various samples as determined by electron diffraction measurements of selected areas of the samples. As indicated, some samples contained ammonium sulfate, some contained ammonium persulfate, and one contained a mixture of the two.

TABLE 10. Summary of Numbers and Volume Concentrations and of Composition of Stratospheric Particles

Sample Number	Collection Date	Altitude (km)	Latitude	Longitude	$\bar{n} \times 10^2$ (cm^{-3})	$\bar{V} \times 10^3$ (μ^3/cm^3)	Composition
241	25 Jan 1962	21.0	30°-30°N	99°-99°W	-	-	Sulfate
260	6 Feb 1963	19.8	56°-32°N	120°-101°W	-	-	Persulfate
262	21 Feb 1963	19.8	64°-49°N	147°-123°W	-	-	Sulfate
242	30 Jan 1962	18.3	48°-31°N	112°-101°W	-	-	Sulfate
243	13 Feb 1962	18.3	49°-32°N	112°-100°W	4.46	3.42	Sulfate
251	5 Jun 1962	18.3	48°-31°N	112°-101°W	-	-	Persulfate
258	14 Feb 1963	18.3	45°-39°N	109°-104°W	6.88	8.76	Sulfate
259	19 Feb 1963	18.3	31°-20°N	100°-96°W	5.85	4.98	Sulfate
265	2 Apr 1963	18.3	32°-31°N	100°-99°W	3.45	5.93	-
266	7 May 1963	18.3	33°-31°N	100°-100°W	21.6	51.2	-
268	30 Jul 1963	18.3	33°-32°N	111°-111°W	22.9	25.0	-
271	13 Aug 1963	18.3	32°-32°N	111°-111°W	5.05	6.76	-
254	12 Jun 1962	16.8	49°-31°N	112°-101°W	2.22	2.89	Sulfate
257	26 Jun 1962	16.8	48°-31°N	112°-101°W	2.04	2.42	Mixture
244	13 Feb 1962	15.2	49°-32°N	112°-100°W	1.24	1.33	Persulfate
252	19 Jun 1962	15.2	49°-31°N	112°-101°W	1.19	1.31	Sulfate
245	13 Mar 1962	12.2	49°-31°N	112°-101°W	1.44	1.69	Persulfate

The size-frequency spectrum, or size-frequency distribution is often presented in plots of $d\bar{n}/d(\log r)$ vs. r, where $\bar{n}$ is the number concentration of particles and r is radius of particles. The dimension of $d\bar{n}/d(\log r)$ is cm^{-3}, and the area under a curve (spectrum) is the number concentration. The size distribution function, as defined here, is dimensionless and is essentially the size-frequency distribution normalized by dividing by the number concentrations, namely:

$$\frac{1}{\bar{n}} \frac{d\bar{n}}{d(\log r)} .$$

In practice, in making and reporting particle size measurements on samples, the number of particles, n_i in a specified size class, say $r_a < r < r_b$, is recorded. The class intervals in this work were chosen such that $\Delta (\log r)$ is constant.

When n_i was converted to number concentration, $\bar{n}_i$, by means of flight data and corrections for impaction efficiency, the quantity $\bar{n}_i/\Delta (\log r)$ resulted. This is the experimental approximation of $d\bar{n}/d(\log r)$. The number concentration for each sample, as reported in Table 10, was obtained by:

$$\bar{n} = \sum \left[\frac{\bar{n}_i}{\Delta (\log r)} \right] \Delta (\log r) .$$

Table 11 lists for each sample the values of the size distribution function for each size class. The size classes are listed according to their mean (logarithmic) radius, in microns. The listed quantity is actually $(1/\bar{n})[\bar{n}_i/\Delta (\log r)]$. Listed in the last two columns of the table are the median and average values, respectively, of the size distribution function.

TABLE 11. Computed Values of the Size Distribution Function $\frac{1}{\eta}\frac{d\bar{n}}{d(\log r)}$

Radius	Sample Number 243	258	259	265	266	268	271	254	257	244	252	245		
(μ)	13-2 -62 18 km	14-2 -63 18 km	19-2 -63 18 km	4-2 -63 18 km	7-5 -63 18 km	30-7 -63 18 km	13-8 -63 18 km	12-6 -62 17 km	26-6 -62 17 km	13-2 -62 15 km	19-6 -62 15 km	13-3 -62 12 km	Median Value	Average
0.138	0.36	0.04	0.52	0	0	0	0	0.33	0.07	0.24	1.48	0	0.06	0.25
0.160	1.10	0.26	1.28	0	0	0.20	0.11	0.62	0.36	0.72	1.72	0	0.31	0.53
0.182	1.82	0.33	1.05	0	0	2.39	1.09	0.90	0.92	0.67	2.24	0	0.91	0.95
0.199	1.58	0.40	0.86	0.49	0.03	4.79	0.56	0.40	0.85	0.92	2.70	1.05	0.86	1.22
0.224	2.53	1.06	3.20	0.17	0.20	3.14	1.89	0.50	0.65	0.99	2.13	0.66	1.00	1.43
0.244	4.49	1.88	4.75	0.74	0.51	2.76	1.78	1.49	1.46	2.35	0.15	2.77	1.83	2.09
0.264	4.52	5.57	4.11	1.47	1.08	1.80	3.60	2.35	4.17	3.46	0.72	2.74	3.10	2.96
0.282	5.19	5.87	4.83	4.43	2.73	1.62	3.40	3.38	7.01	4.74	3.19	5.98	4.59	4.36
0.300	2.26	4.60	1.84	5.26	4.33	1.69	3.54	5.60	6.43	4.93	3.04	6.96	4.47	4.21
0.318	2.19	4.36	2.44	2.82	4.15	1.93	4.54	6.00	3.37	5.41	2.70	5.18	3.76	3.76
0.336	1.02	3.00	1.20	6.89	4.88	1.42	5.00	5.06	2.61	3.98	2.31	6.41	3.49	3.65
0.352	0.98	2.52	1.02	5.66	3.53	1.90	2.97	2.90	2.85	3.54	1.25	3.67	2.88	2.73
0.370	0.78	1.71	0.60	4.73	3.64	1.70	3.32	3.60	1.91	1.75	0.05	1.08	1.73	2.07
0.386	0.35	2.33	0.92	4.74	3.19	1.38	1.68	1.92	1.55	1.83	0.80	0.42	1.62	1.76
0.402	0.39	0.79	0.68	3.62	3.98	1.37	0.57	1.78	0.83	0.04	1.40	0.54	0.81	1.33
0.418	0.16	1.53	0.56	0.92	3.20	0.89	0.64	0.49	0.62	0.09	0.78	0.17	0.63	0.84
0.432	0.40	0.84	0.53	0.51	2.92	0.86	1.95	0.95	0.64	0.10	0.10	0.05	0.58	0.82
0.448	0.03	0.27	0.08	0.11	1.97	0.64	1.99	0.49	0.24	0	0.88	0.09	0.27	0.57
0.462	0.04	0.60	0.09	0.26	2.07	0.42	0.34	0	0.21	0	0.10	0.05	0.21	0.35
0.478	0.02	0.16	0.06	0.09	2.35	0.31	0.35	0.02	0.18	0	2.37	0.14	0.16	0.50
0.492	0.01	0.39	0.05	0.32	1.15	0.61	0	0	0.24	0.06	0.07	0	0.06	0.24
0.506	0	0.14	0.03	0.16	1.09	0.34	0	0.29	0.48	0	0.95	0.06	0.22	0.39
0.520	0	0.07	0.07	0.23	1.05	0.14	0	0	0	0	0.89	0.03	0.14	0.21

TABLE 11. (continued)

Radius (μ)	243 13-2 -62 18 km	258 14-2 -63 18 km	259 19-2 -63 18 km	265 2-4 -63 18 km	266 7-5 -63 18 km	268 30-7 -63 18 km	271 13-8 -63 18 km	254 12-6 -62 17 km	257 26-6 -62 17 km	244 13-2 -62 15 km	252 19-6 -62 15 km	245 13-3 -62 12 km	Median Value	Average
0.534	0	0.07	0.02	0.44	0.76	0.14	0	0	0	0	0.07	0	0	0.13
0.548	0	0	0.06	0.04	0.92	0	0	0	0	0	0.99	0	0	0.17
0.560	0	0	0.02	0.04	0.26	0.08	0	0	0	0	0.04	0	0	0.04
0.574	0	0	0.02	0.15	0.89	0.30	0	0	0	0	0	0	0	0.11
0.588	0	0	0.02	0.40	0.35	0.33	0	0	0	0	0	0	0	0.09
0.600	0	0	0.02	0.40	0	0.08	0	0	0	0	0	0	0	0.04
0.614	0	0	0.04	0	0.09	0.09	0	0	0	0.07	0.04	0	0	0.03
0.626	0	0	0.02	0	0.37	0.09	0	0	0	0	0	0	0	0.04
0.640	0	0	0.02	0	0.09	0	0	0	0	0	0	0	0	0.01
0.652	0	0	0	0	0	0.10	0	0	0	0	0	0	0	0.01
0.664	0	0	0	0	0.10	0.10	0	0	0	0	0	0	0	0.02

The distribution functions of the individual samples are plotted in Figures 5 to 16, and the average distribution function is shown plotted in Figure 17. The nearly parabolic shape of this function suggests a log-normal distribution. The average distribution function is shown as points in Figure 18 on a log-probability plot. The straight line drawn among the points is a log-normal distribution with geometric mean radius 0.305 micron and a geometric standard deviation of 1.30.

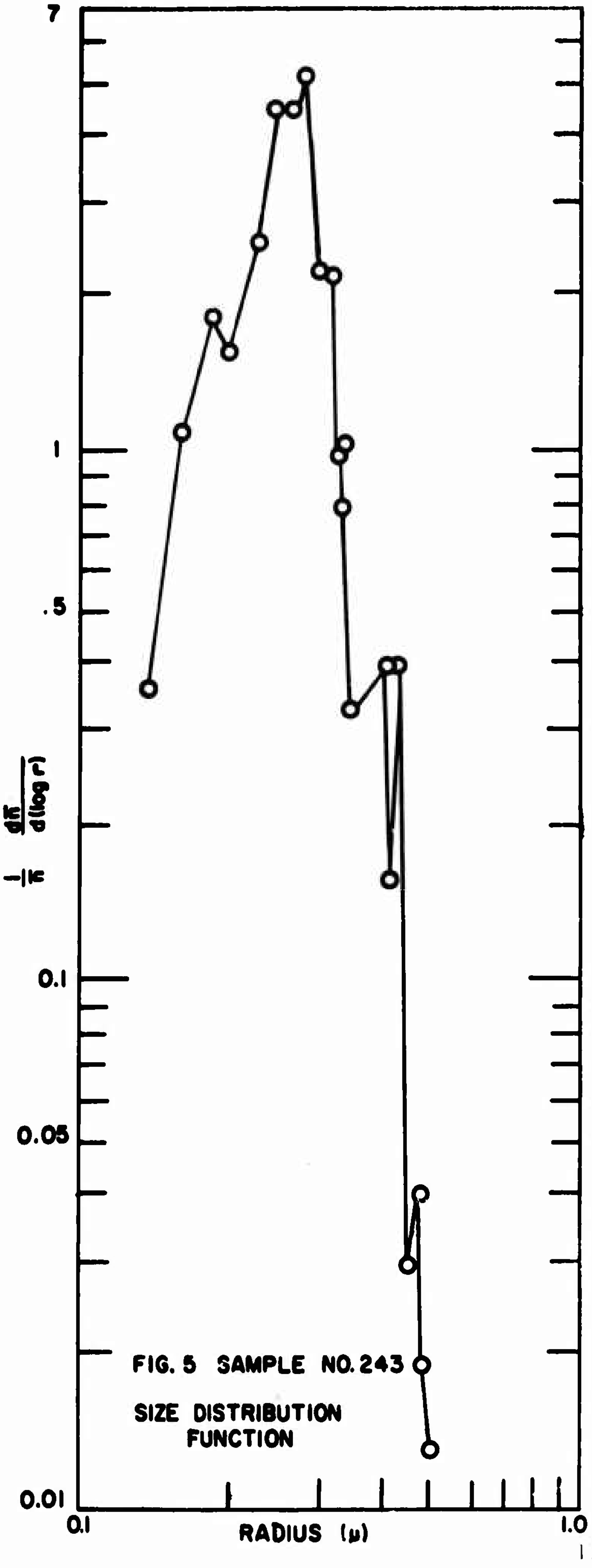


FIG. 5 SAMPLE NO. 243

SIZE DISTRIBUTION FUNCTION

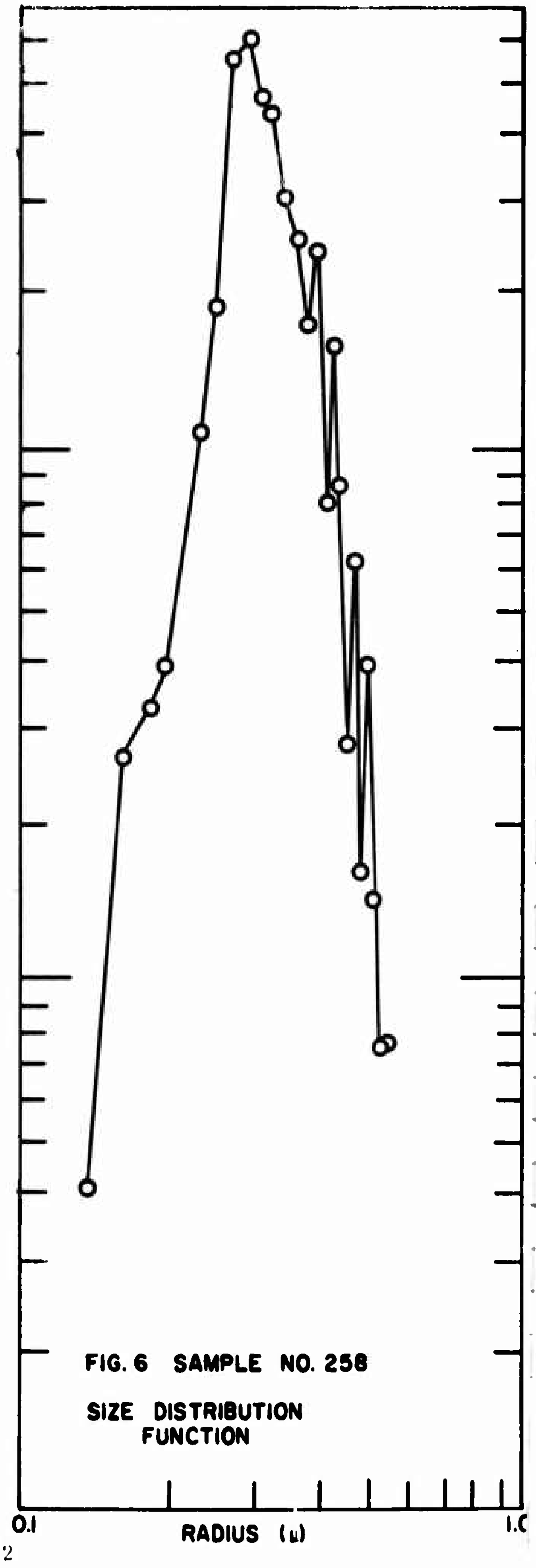


FIG. 6 SAMPLE NO. 258

SIZE DISTRIBUTION FUNCTION

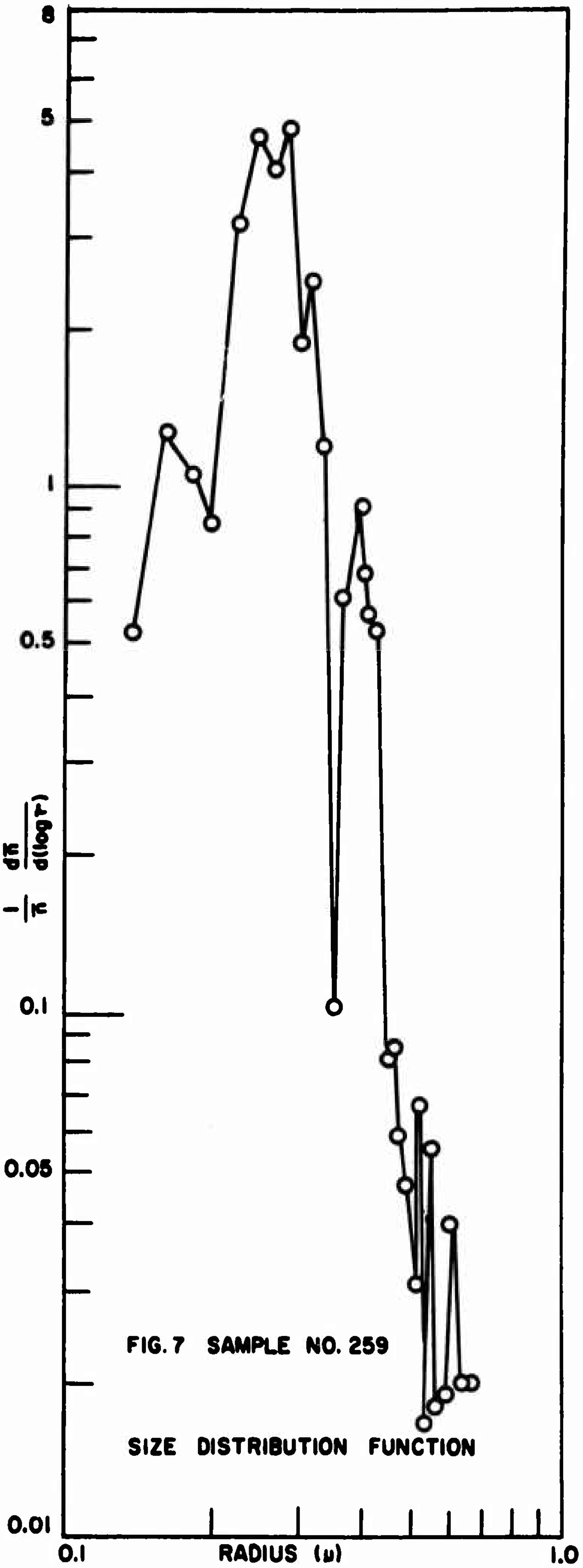


FIG. 7 SAMPLE NO. 259

SIZE DISTRIBUTION FUNCTION

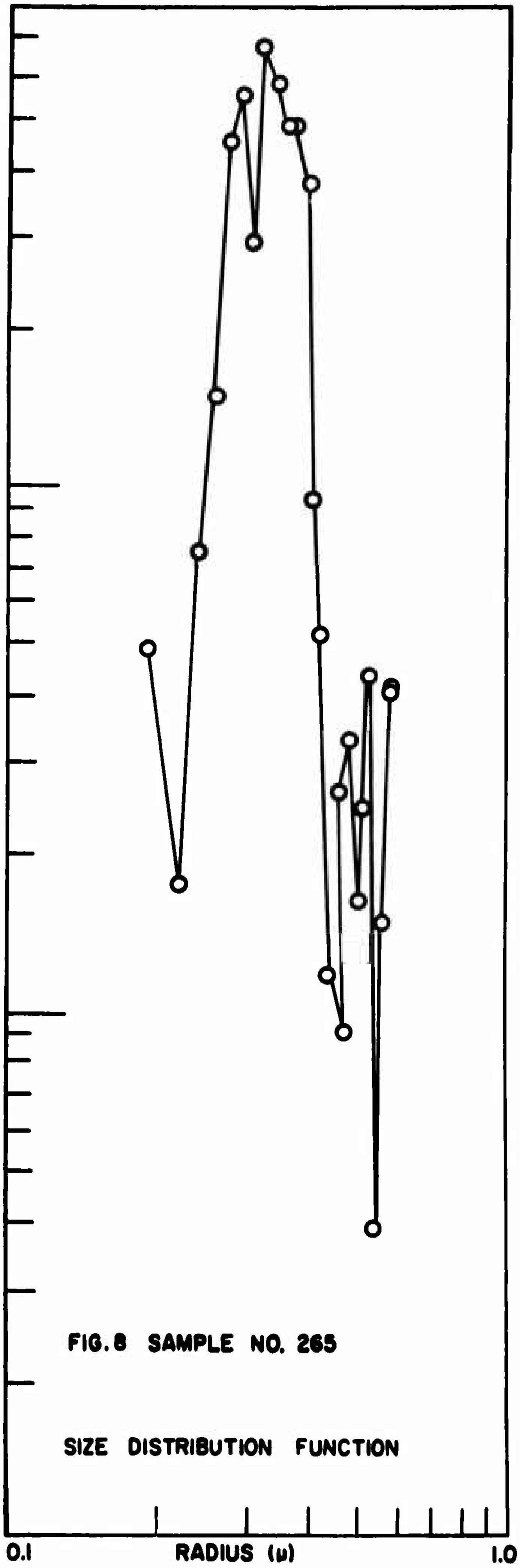


FIG. 8 SAMPLE NO. 265

SIZE DISTRIBUTION FUNCTION

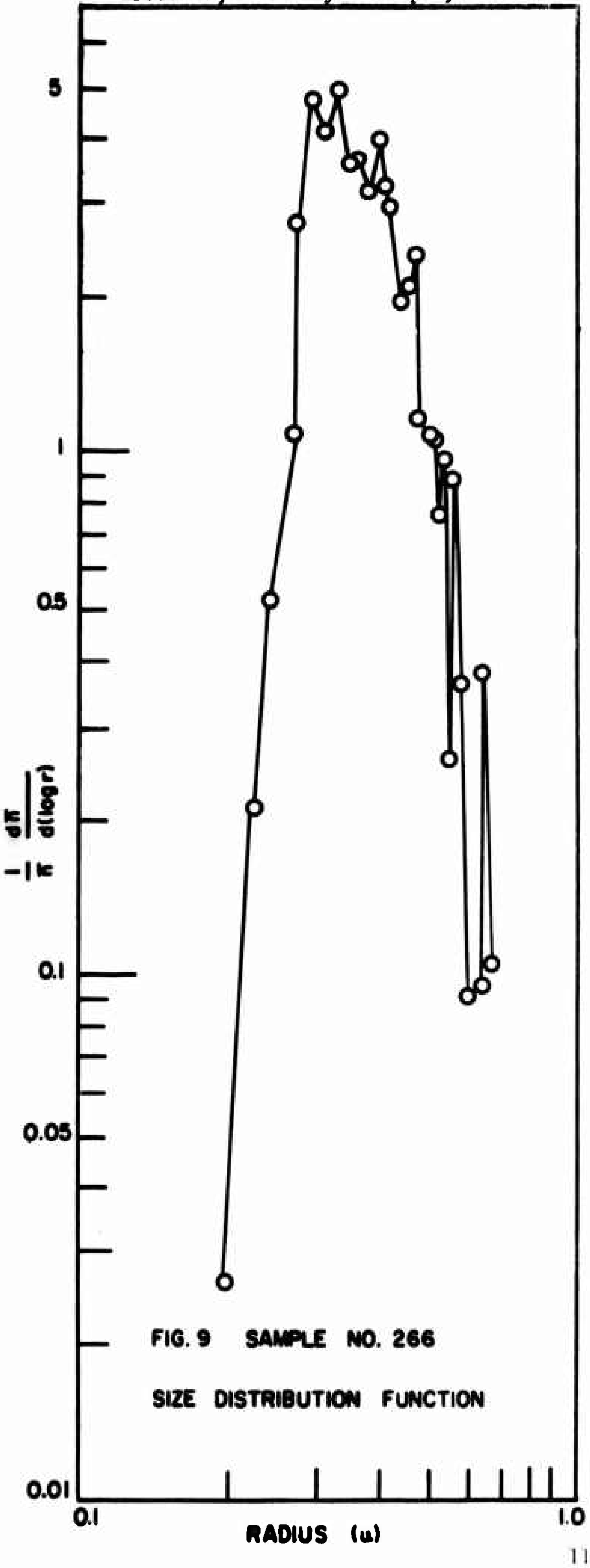


FIG. 9 SAMPLE NO. 266

SIZE DISTRIBUTION FUNCTION

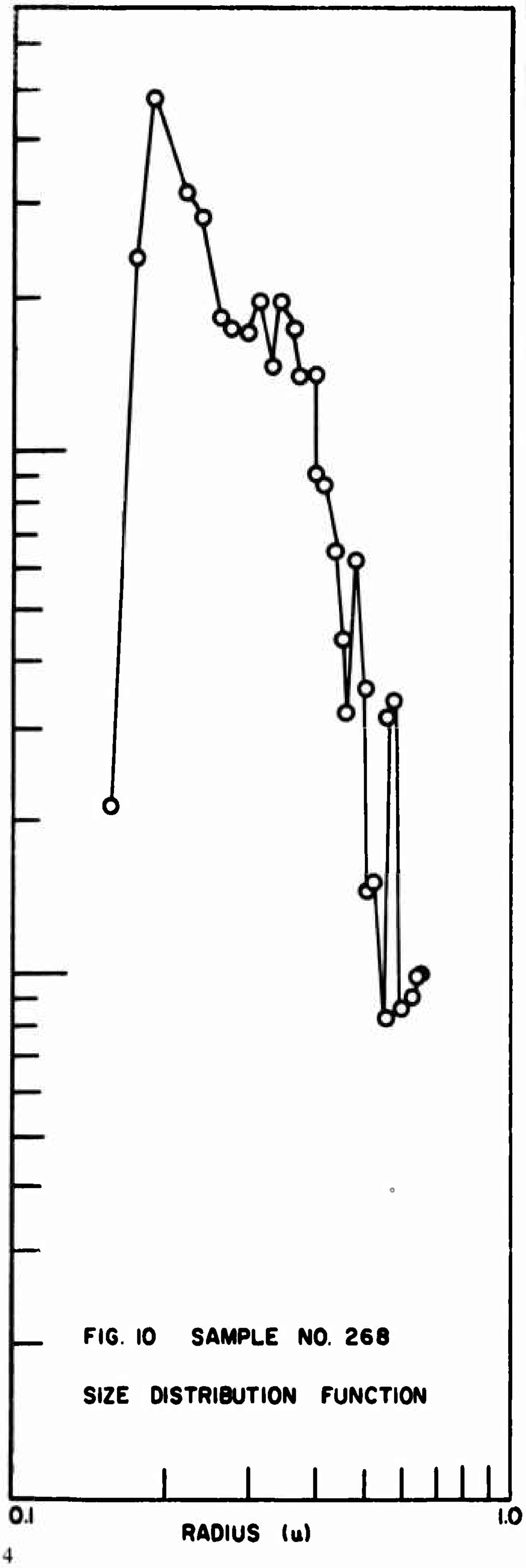


FIG. 10 SAMPLE NO. 268

SIZE DISTRIBUTION FUNCTION

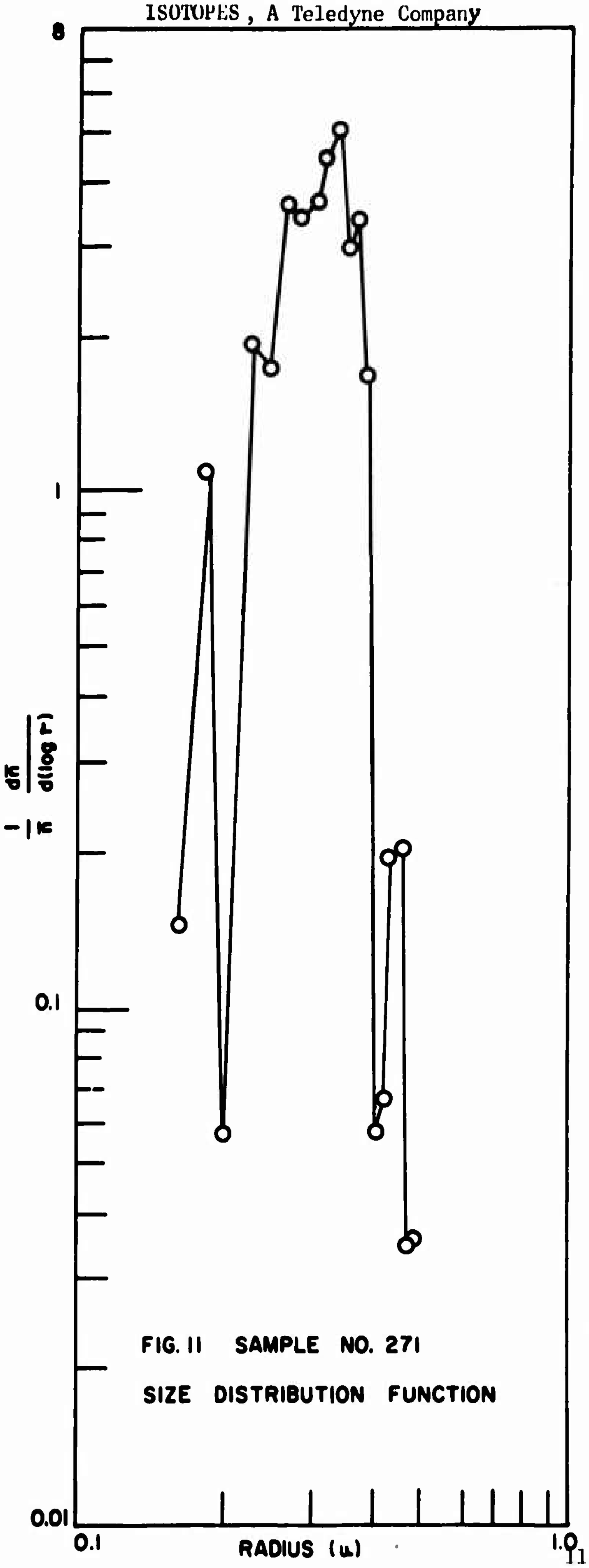


FIG. 11 SAMPLE NO. 271

SIZE DISTRIBUTION FUNCTION

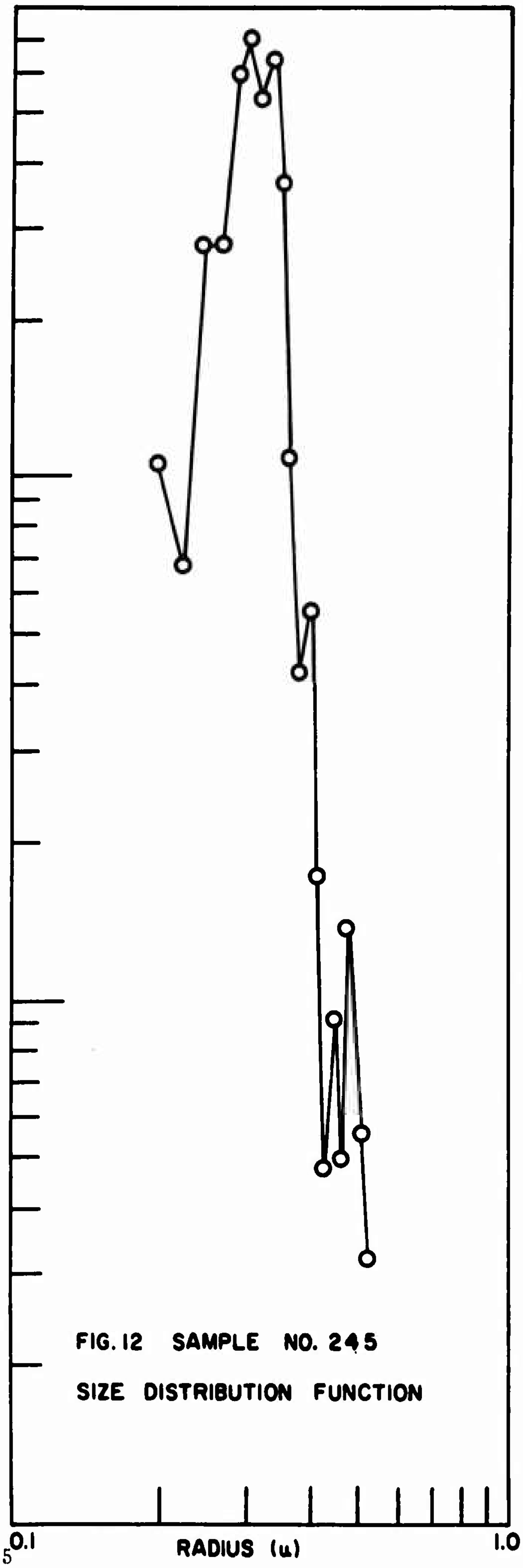


FIG. 12 SAMPLE NO. 245

SIZE DISTRIBUTION FUNCTION

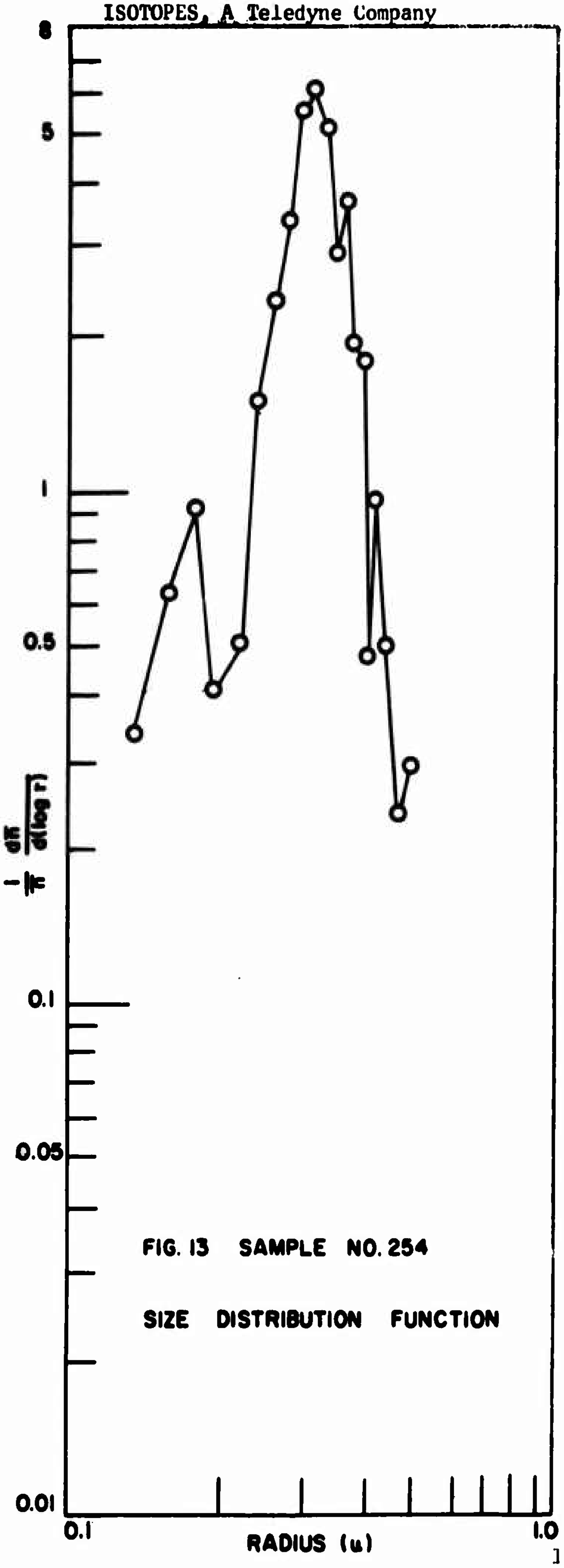


FIG. 13 SAMPLE NO. 254

SIZE DISTRIBUTION FUNCTION

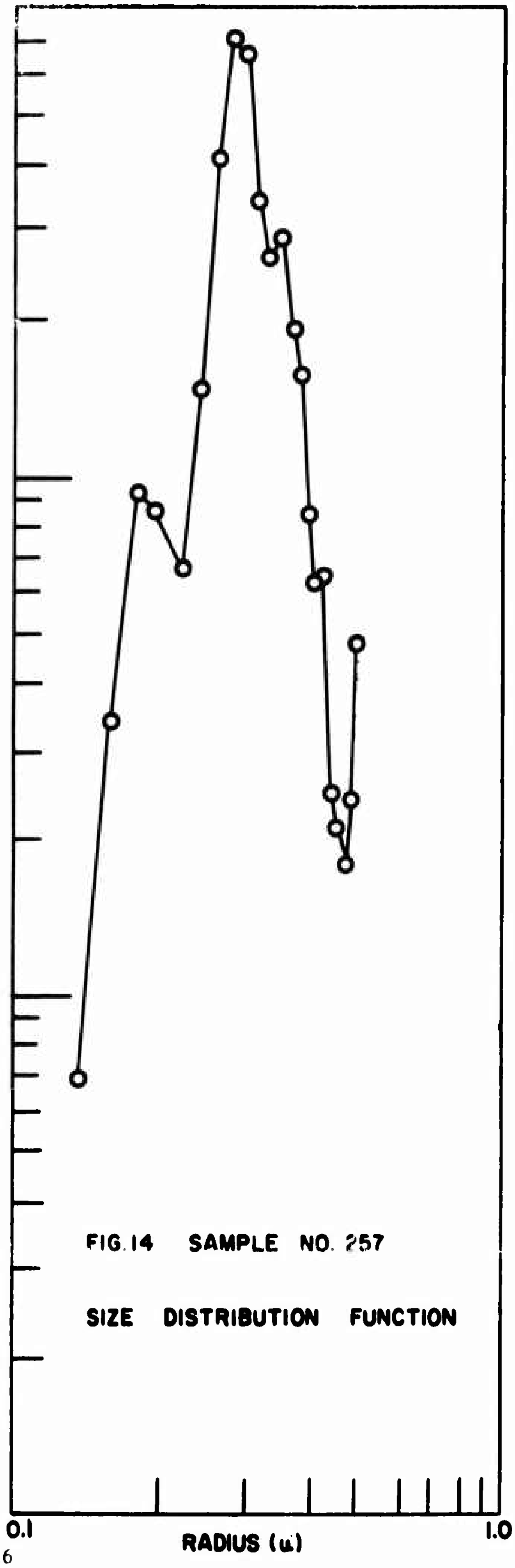


FIG. 14 SAMPLE NO. 257

SIZE DISTRIBUTION FUNCTION

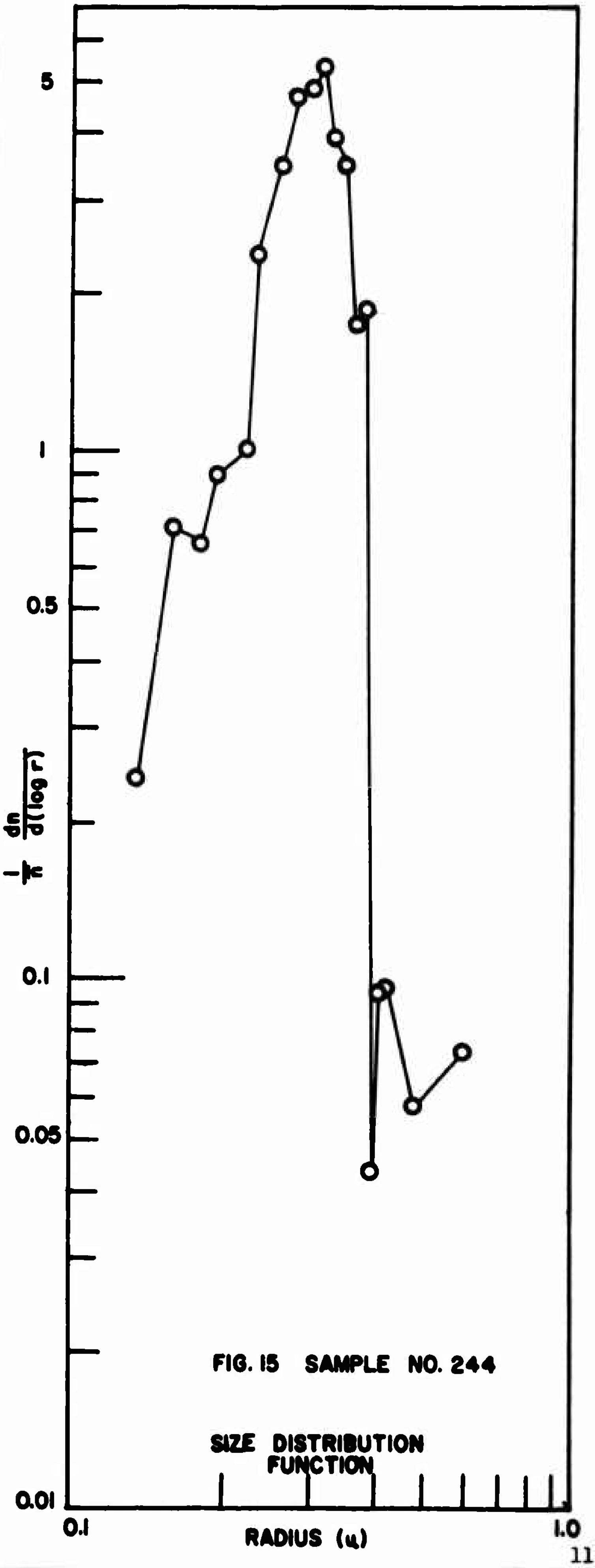


FIG. 15 SAMPLE NO. 244

SIZE DISTRIBUTION FUNCTION

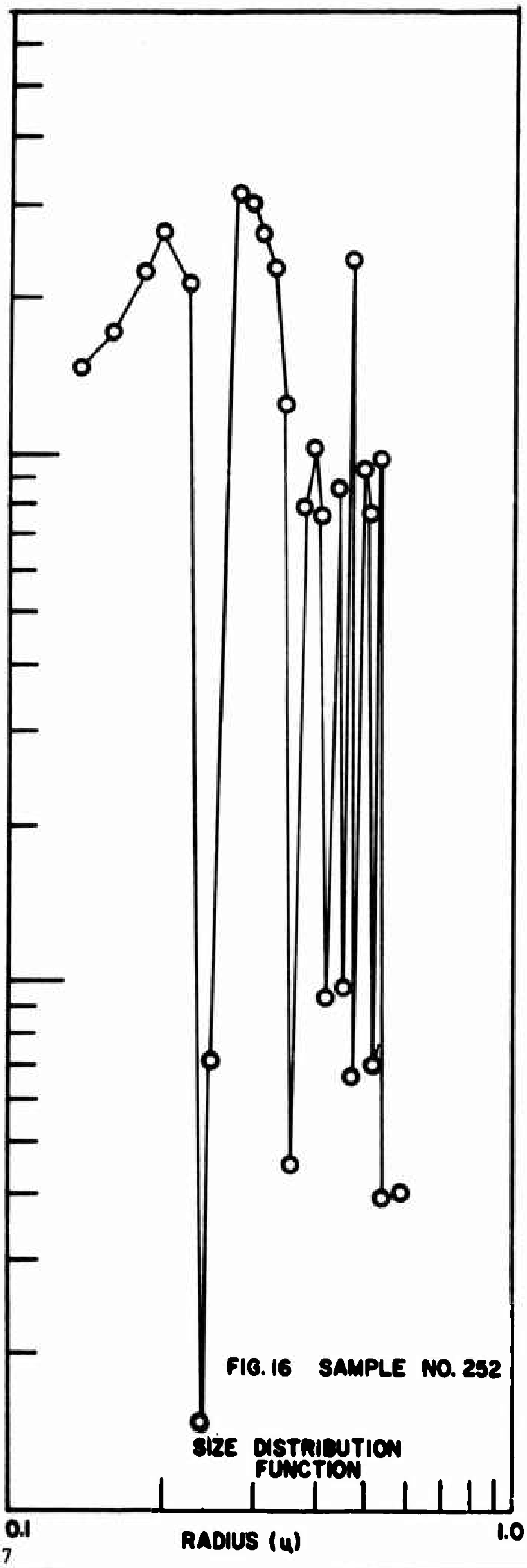


FIG. 16 SAMPLE NO. 252

SIZE DISTRIBUTION FUNCTION

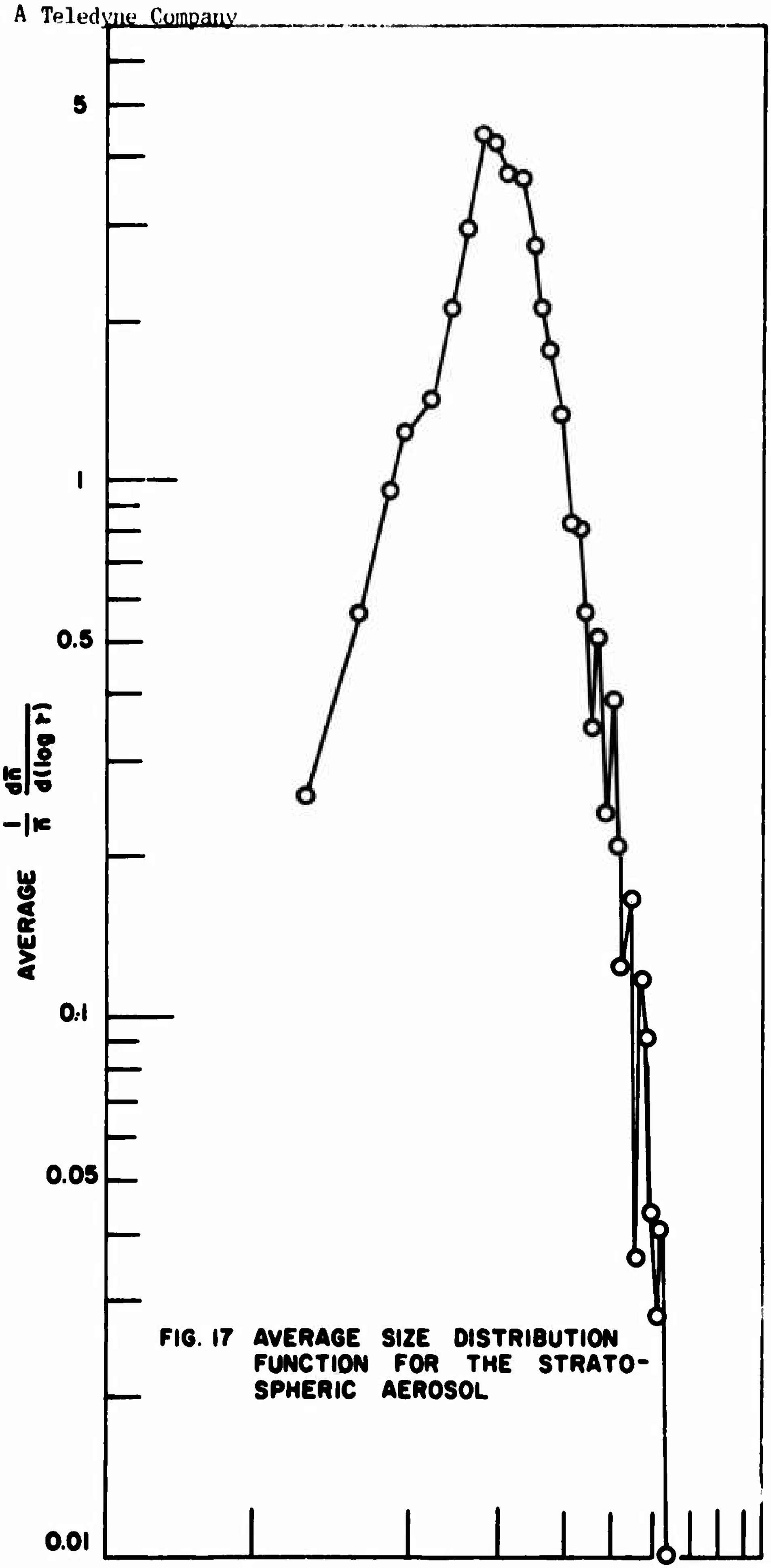


FIG. 17 AVERAGE SIZE DISTRIBUTION FUNCTION FOR THE STRATOSPHERIC AEROSOL

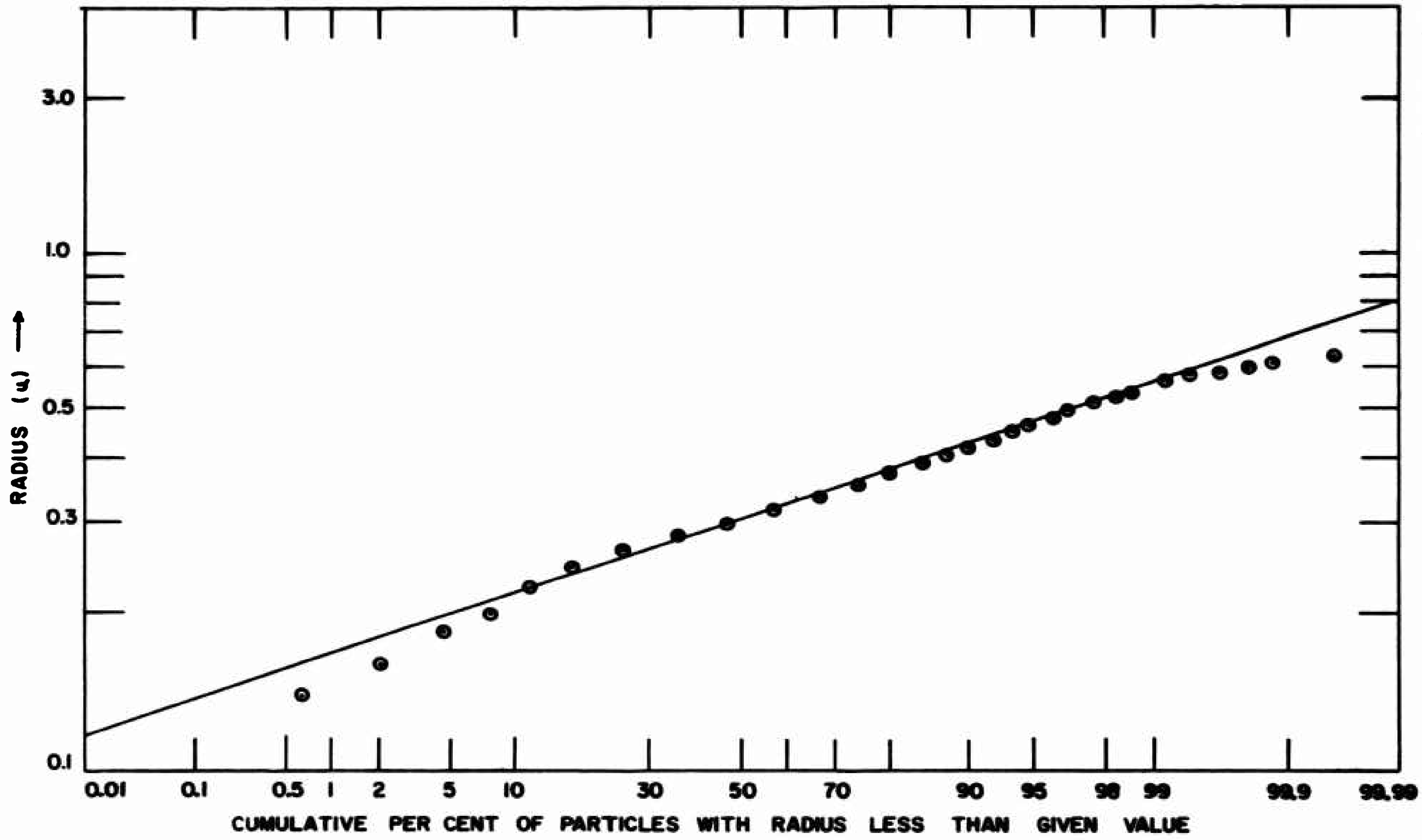


FIG. 18 AVERAGE DISTRIBUTION FUNCTION OF STRATOSPHERIC PARTICLES

5.3 Evaluation of the Measurements

Uncertainties in the data as they are given in Tables 9 and 11 stem from (a) possible non-representativeness of sample areas studied; (b) improper correction for effects of moisture (flattening) on the particles; (c) application of impaction theory for an ideal thin ribbon to the computation of probe collection efficiencies; (d) statistical fluctuations because of small numbers of particles observed in particular class intervals.

The net uncertainty due to the four sources listed above is difficult to assess quantitatively. However, items a, b and c probably cause the distribution functions to be more uncertain for the smaller than for the larger particles. To some degree all uncertainties are reflected in the variations of the experimental values of $(1/\bar{n})[d\bar{n}/d(\log r)]$ in Table 11. These latter variations in turn are reflected in differences between the median and average values of the distribution function. It must be noted, however, that variations in the values listed in Table 11 may be due to natural variations in production and removal rates of the particles. The relative proportions of real and experimental variations cannot easily be assessed.

The best established fact of the size distribution function of the stratospheric sulfate particles is the peak in the vicinity of 0.3 micron radius. It is notable that in the range of radius 0.18-0.40 micron:

(a) the value of the average distribution function increases and then decreases by a factor of four;

(b) the difference between the average and mean distribution functions varies by less than 30% of the average;

(c) the computed values of the collection efficiencies [7] at 18 km altitude vary by a factor of about 1.3;

(d) the number of observed particles in each size class was greater than in any class outside the size range (0.18-0.40 μ).

Thus all of the factors which produce uncertainty in the values of the size distribution function are minimal over the size range in which the peak is centered.

5.4 Comparison with Other Findings

JUNGE et al. [3] found that the number concentrations of particles with 0.1 <r <1.0 micron had a broad maximum at about 20 km altitude. Since the highest altitude covered in the present work was 18 km, the occurrence of peak concentrations at 20 km could not be demonstrated. However, the decrease of concentration with decreasing altitude below 18 km, as shown in Table 10, is in general agreement with Junge et al.

More recent work by NEWKIRK & EDDY [8], using light scattering measurements by a balloon-borne coronagraph, showed that the stratospheric aerosol can exist in thin cloud-like laminae. (Similar conclusions on the existence of layers of aerosols in the stratosphere were drawn by several other workers. These are reviewed by Newkirk & Eddy.) Again, the present work was not extensive enough in time or altitude coverage to demonstrate the existence of such aerosol clouds. The two high values in number concentrations (Samples 266 and 268) were obtained after the eruption of Mt. Agung in Bali, which may have contributed to the number of particles or enhanced particle formation in the stratospheric air in which the samples were taken. (See also MOSSOP [9] for discussion of the association of sulfate with the volcanic dust). However, the variations by nearly an order of magnitude in number concentrations of the samples from 18 km altitude, as shown in Table 10, are consistent with the concept of clouds of aerosols. It would be very difficult to attribute all observed variations to measurement errors. Since the sampling was not sufficiently intensive, the number concentrations in Table 10 cannot be used to give reliable time-averaged values.

In one important aspect the present work is not in agreement with previous work of JUNGE et al.[3], and JUNGE & MANSON [4]. This is in the form of the size distribution function of the stratospheric sulfate particles ($0.1 < r < 1.0$ micron). JUNGE et al.[3], from examination of particles collected by balloon-borne impactors, concluded that the radii were distributed approximately according to:

$$\frac{dn}{d\,(\log\, r)} \propto r^{-2}$$

or equivalently:

$$\frac{dn}{dr} \propto r^{-3}$$

for the range $0.1 < r < 1.0$ micron. Figure 19 shows their distribution and the distribution found in the present work. In Figure 19 the ordinate, $dn/d(\log r)$, has units of cm^{-3}. Curve B corresponds to the average distribution function of Figure 17, and the median value of $\bar{n}$ at 18 km altitude, namely 0.066 cm^{-3}. Also shown in Figure 19 are limits of uncertainty

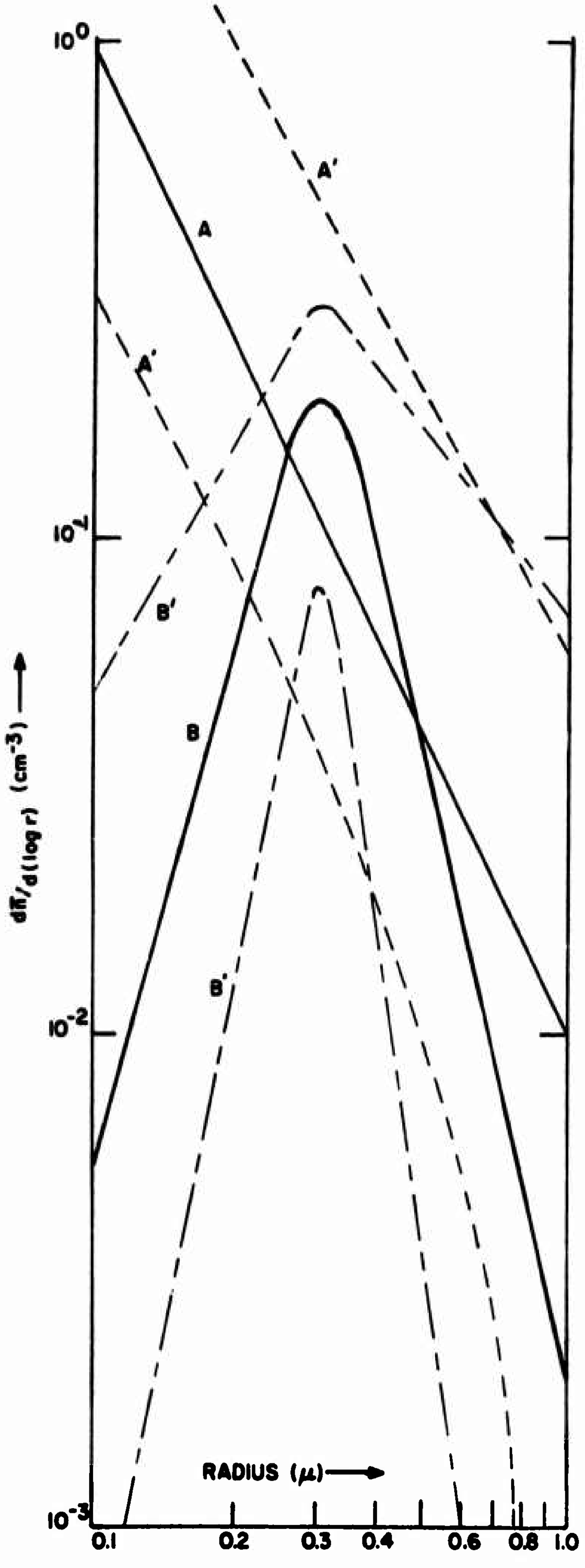


FIGURE 19.
COMPARISON OF PARTICLE SIZE-CONCENTRATION SPECTRA OF STRATOSPHERIC SULFATE PARTICLES

CURVE A: AFCRL - JUNGE ET AL
CURVE A': CONFIDENCE LIMITS OF CURVE A
CURVE B: STAR DUST
CURVE B': CONFIDENCE LIMITS OF CURVE B

estimated by Junge et al., for their curve, and estimated for the present work based on variations as illustrated in Table 11 and discussed above. It can be readily seen that the main region of discrepancy is for r <0.2 micron.

It is also to be noted that there is a real discrepancy in the shapes of the two distributions shown in Figure 19. The regions of uncertainty for the present distribution are based primarily on the variations in the absolute values of $(1/\bar{n})\ [d\bar{n}(\log r]$ about the mean value, and do not completely indicate the reliability of the shape of the distribution. The reasons for confidence in the general shape of the size distribution function were discussed at the end of the previous section. It is apparent that the discrepancy between the two distributions in Figure 17 is due primarily to one or more systematic errors. A likely cause is the correction for the effect of moisture in flattening the particles. JUNGE et al.[3] did not apply any correction for this effect and recorded the sizes of the particles as they appeared in the samples.

MOSSOP [5] reported the results of analysis of eight WU-2 aircraft impactor samples taken in the latitude range of 15° to 45°S in early 1963. The concentrations ranged from 0.017 to 0.042 cm^{-3}, and are consistent with all but two of the present results. The size distributions found by Mossop were also peaked, but at a slightly larger radius, about 0.35 micron. Figure 20 shows the present distribution in the same manner as Figure 19, and also shows Mossop's distribution corresponding to an average concentration of 0.033 cm^{-3}. The agreement between the two results is good. The slight difference in the radius at which the peak occurs may be due to different methods of correcting for flattening of the particles.

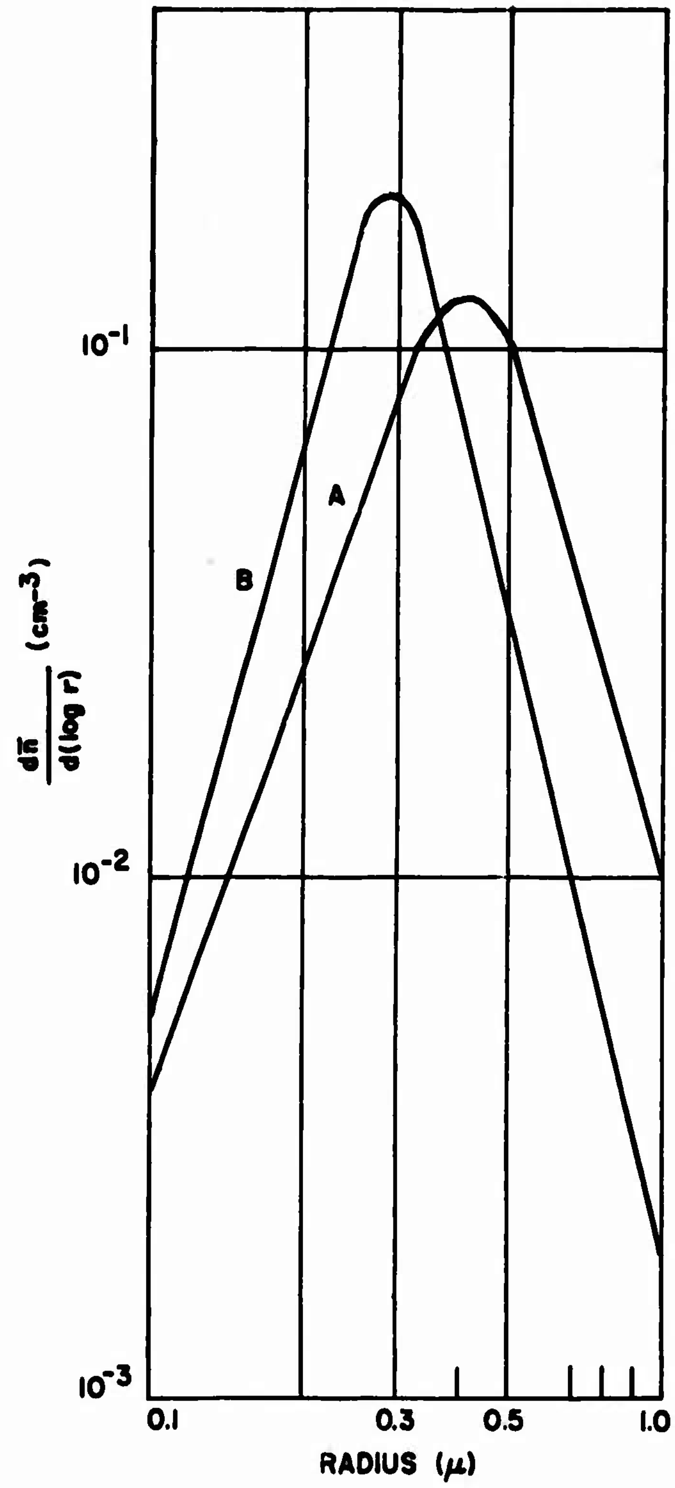


FIG. 20 COMPARISON OF PARTICLE SIZE - CONCENTRATION DISTRIBUTIONS OF STRATOSPHERIC SULFATE PARTICLES.

CURVE A: MOSSOP CURVE B: PRESENT WORK

NEWKIRK & EDDY[8] used measurements of intensity and wavelength of sky light measured by a balloon-borne coronagraph in attemps to deduce the concentrations and size distrubitons of atmospheric aerosol particles. However, they started by assuming that the functional form of the size distribution was

$$\bar{n} = \bar{n}_0 (r/r_2)^{-\delta},$$

where

$$\delta = 0 \text{ for } r < r_2, \quad \delta > 0 \text{ for } r > r_2$$

in the manner of JUNGE et al.[3]. In this equation r_2 is some fixed radius. The experimental results were then used to calculate values of $\bar{n}$ and δ, which were found to be in substantial agreement with the values reported by JUNGE et al.[3]

In his paper, MOSSOP[5] suspected that the earlier distributions reported by JUNGE et al. and in DASA 1300 might be in error because of the production of small particles in the samples following their collection by condensation of moisture and subsequent drying. Mossop noted that Newkirk & Eddy's apparently independent work did not support this suspicion. So Mossop concluded that the earlier work was also correct and that his collections of particles were made at times and places where the atmosphere was anomalously deficient in particles with $0.05 < r < 0.4$ micron.

In the light of the present results, and their favorable comparison with Mossop's, the following clarifying statements are in order:

(a) Mossop's original suspicion was probably correct.

(b) Since Newkirk & Eddy assumed the wrong functional form of the size distribution, their work does not corroborate the findings of Junge et al.

(c) Mossop's samples were "normal"; i.e. the atmosphere was not deficient in particles with $0.05 < r < 0.4$ micron.

5.5 Aitken Nuclei and Sulfate Particles

Though the present work is not directly concerned with measurement of Aitken nuclei (which consist primarily of particles with $r < 0.1$ micron in the stratosphere), combining the current results with the known properties of stratospheric Aitken nuclei leads to some interesting conclusions. JUNGE et al.[3] and JUNGE [10], in experiments with balloon-borne Aitken nuclei counters, found that the total particle concentration decreases rapidly with height above the tropopause for the first few kilometers, and then remains on the order of about 1 cm^{-3} at least up to about 33 km altitude. Since Aitken nuclei are smaller than 0.1 micron radius they would not have been collected with appreciable efficiency by the impactors. If the distribution function and number concentrations found in the present work are correct, then in the region of radius less than 0.1 micron the frequency (or number concentration expressed, e.g., as $d\bar{n}/d(\log r)$) must increase again so that the total number concentration will agree with the Aitken nuclei concentration. There is no direct evidence available concerning the average size of the Aitken nuclei. However, JUNGE et al.[3] estimated that a radius of 0.04 micron would be consisten with some theoretical considerations of mixing by turbulent diffusion from a tropospheric source. Thus it is likely that the actual distribution of particle sizes in the stratosphere is bimodal with peaks at 0.3 micron and in the vicinity of 0.04 micron radius. Such a distribution is shown schematically in Figure 21 where the portion of the curve labeled "sulfate particles" corresponds to the present findings and the portion labeled "Aitken nuclei" is conjectural. The integral under the entire curve must be such that the total number concentration is about 1 cm^{-3}. The corresponding value for the sulfate particles is 0.066 cm^{-3}, as mentioned earlier.

If the general shape of the distribution as shown in Figure 21 applies to a steady-state condition, then the following statements are true: (a) Some process in addition to coagulation must explain the distribution. (b) If the sulfate particles result from a process involving Aitken nuclei, only some of the Aitken nuclei are involved, and the process is fast compared to coagulation.

5.6 Origin of the Sulfate Aerosol

The body of data relevant to the stratospheric sulfate aerosol is by no means complete enough to determine mechanisms of its formation and removal. The following discussion shows that the sulfate particles are likely to be created in the stratosphere. For purposes of the discussion the term "large particle" refers to the sulfate aerosol particles.

JUNGE [11] and CADLE & POWERS [12] have considered that the photochemical oxidation of SO_2 can occur in the stratosphere, and that it is the probable means of production of the solid sulfate particles. The finding in this and previous work of the composition $(NH_4)_2S_2O_8$ for some of the particles lends credibility to the involvement of photochemical reactions in producing the large particles. In this compound the two SO_4 units are linked by an ozone-type bond between two oxygen atoms.

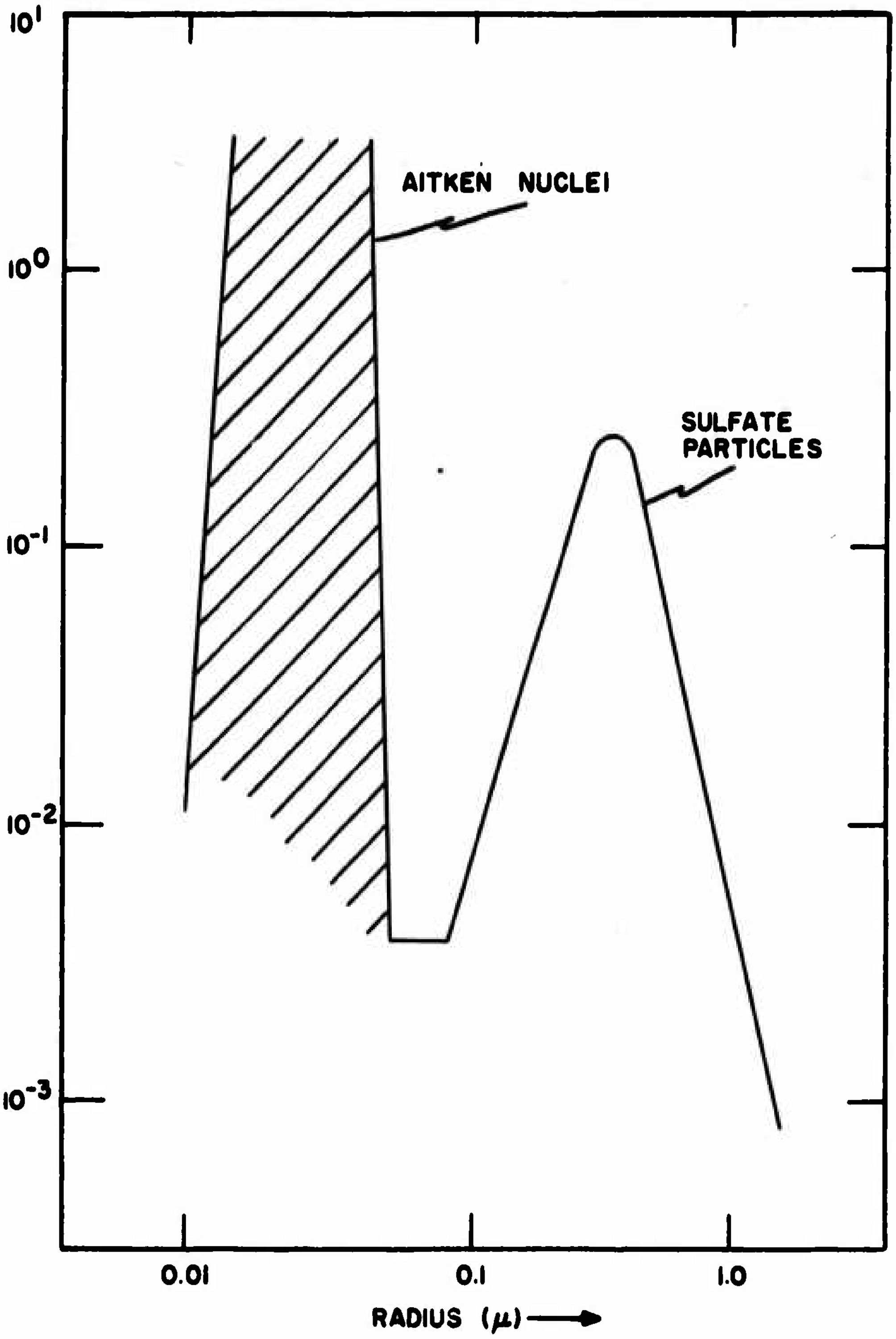


FIG. 21 SCHEMATIC SIZE DISTRIBUTION OF STRATOSPHERIC PARTICLES INCLUDING AITKEN NUCLEI.

The evidence of JUNGE et al.[3] that the large particles have peak number concentrations in the vicinity of 25 km altitude tends to preclude the troposphere as their source. Furthermore, as has been pointed out in the previous section, the size distribution function of stratospheric particles as shown in Figure 21, cannot be explained on the basis of a steady-state coagulation process. Such a process would tend to fill in the valley between two peaks. It is therefore concluded that the size distribution function deduced in this work (Figure 21) supports the concept of production in the stratosphere of the large particles. The most likely method of production is a series of chemical reactions among which is the photochemical oxidation of SO_2. The rate of reaction is fast compared to coagulation. A portion of the stratospheric Aitken nuclei provide the centers for these reactions. This latter statement is consistent also with the findings of MOSSOP [5] that insoluble inclusions of the order of 0.04 micron radius were in each large sulfate particle.

The shape of the size distribution function for $0.3 < r < 1.0$ micron might be explained by a quasi-stationary state of condensation and sedimentation processes, though this is conjectural at present.

CHAPTER 6. INFORMATION DERIVED FROM MEASUREMENTS OF RADIOACTIVITY FROM PRE-1961 NUCLEAR WEAPON TESTS

When radioactive debris in the stratosphere was first sampled for Project HASP in August 1957, relatively little was known about its distribution, and its residence time in the stratosphere, or about the atmospheric processes which influenced its movement from one stratospheric region to another or from the stratosphere to the troposphere. The measurements made during Project HASP provided an important source of data, and for some aspects the first source of data, for answering questions concerning these areas of uncertainty.

The HASP measurements showed that radioactive debris from nuclear weapon tests is not distributed uniformly within the stratosphere, but that instead, its concentration varies significantly with latitude, with altitude, and, at specific locations, with time. The measurements of tungsten-185 especially showed that eddy diffusion is the main mechanism producing the movement of radioactive debris within the stratosphere, and that the movement of debris in the meridional direction occurs within mixing layers which slope downward from the equator toward the poles. The stratospheric burdens calculated from the strontium-90 concentrations measured during Project HASP were considerably lower than were generally expected when the project began, and the stratospheric residence times calculated from the observed changes with time of the burdens were shorter than were generally expected.

A number of nuclear weapons tested during the period 1952 to mid-1957 injected radioactive debris into the stratosphere. Significant quantities of this debris were still present in the stratosphere when sampling for Project HASP began in August 1957. Moreover, several high yield weapons tested during 1957 and 1958, after HASP sampling had begun, also injected fresh debris into the stratosphere. Information of proven or potential significance was gained from HASP samples of fresh debris from these tests. Subsequently, the movement of this debris within the stratosphere was monitored through the measurement of tracer nuclides and of fission product ratios in HASP samples collected over a range of altitudes and latitudes.

6.1 Interceptions of Fresh Debris from Late 1957 Weapon Tests

During late 1957, fresh radioactive debris from at least two nuclear events was intercepted by HASP missions. The first of these interceptions was of debris from a USSR test performed in late September or early October 1957. The U. S. Government[13] announced nuclear weapon tests by the USSR on 24 September 1957 (megaton range in the Arctic), 6 October 1957 (a thermonuclear device), and on 10 October 1957 (a small explosion). Filter samples containing fresh fission products were collected on 5 November 1957 at 20 km between 55°N and 22°N. Sample 23N, collected between 49°N and 44°N, contained especially high concentrations of fresh debris. The rate of decay of the beta activity of these samples indicated an apparent age of about 20 days at the time of their collection. This estimate was based on a curve of the rate of beta decay as a function of time after formation, given by Dolan[14]. This would suggest a production date of about 16 October 1957, later than any of the announced tests. It seems most likely that the intercepted debris originated in the 6 October 1957 event, and that fractionation of the debris had occurred leading to the erroneous indication of age.

Subsequent sampling missions flown on 8 November and 12 November 1957 also encountered fresh debris from this same event, but the maxima in the activity profile at 20 km were found at different latitudes on each mission. These profiles are shown in Figure 22. A.

Series of samples collected at about 21 km on 20 November 1957, contained very little fresh debris, but a series of samples collected at 19 km on 22 November 1957 contained fresh debris from the October 1957 USSR event and fresh debris from a recent low latitude injection. The rate of beta decay of the low latitude samples, collected between 10°N and 7°S, indicated the origin of the fission products was on about 10 November 1957, agreeing well with the shot date[13] of a megaton range U.K. test near Christmas Island (2°N) on 8 November 1957. Profiles of beta activity at 19 - 21 km on 20 and 22 November 1957 are shown in Figure 23.

Meridional profiles of activity injected into the polar stratosphere by the October 1957 USSR event were quite irregular, with peak concentrations being found at a variety of latitudes, but the profile of activity injected into the equatorial stratosphere by the November 1957 U.K. event appeared to show a symmetrical distribution of activity about the latitude of injection. This difference almost certainly reflects the rather uniform zonal flow within the equatorial stratosphere, which contrasts with the meandering currents of air which are typical of the polar stratosphere.

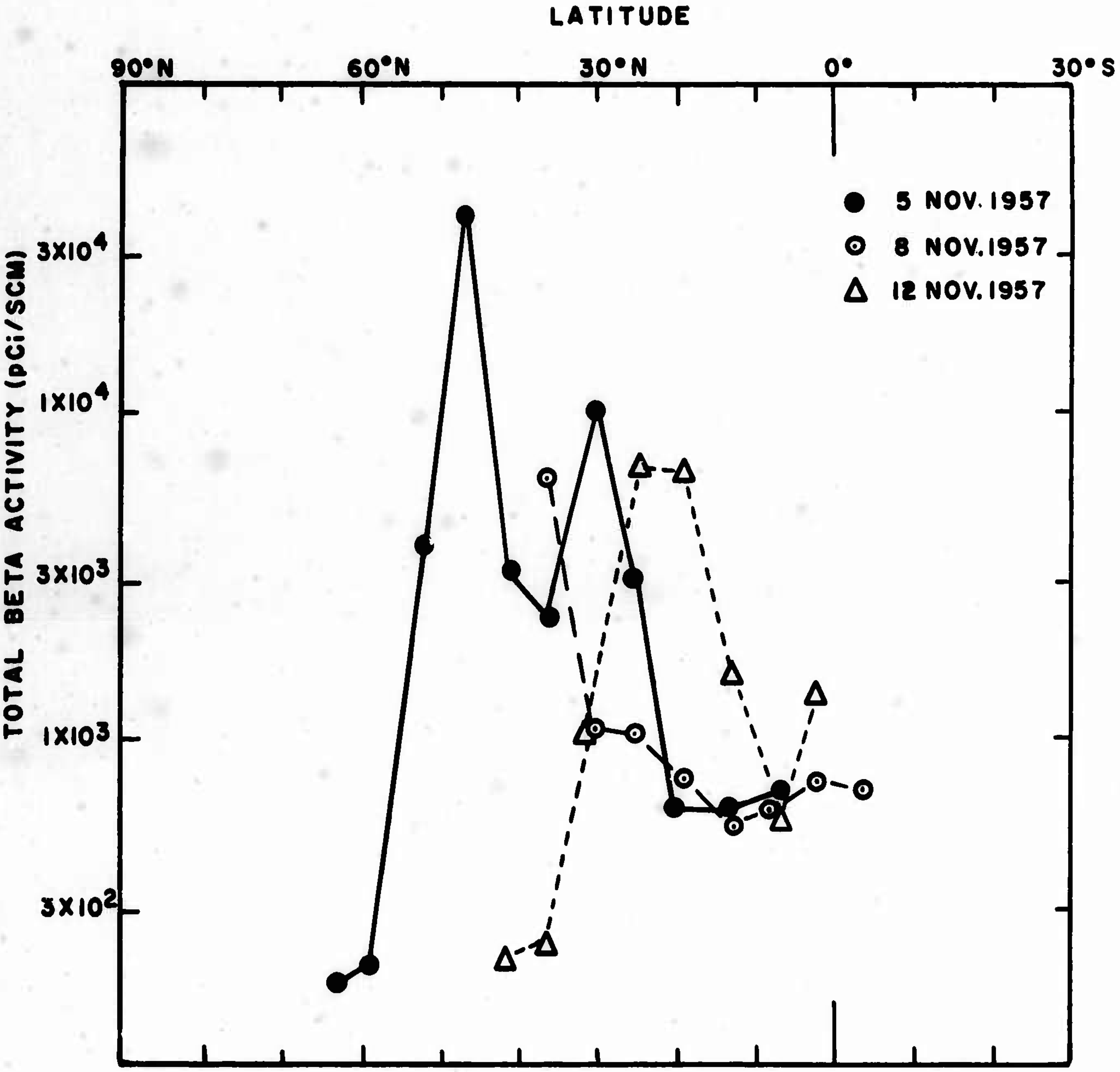


FIGURE 22 TOTAL BETA ACTIVITY AT 20 KM, 5 NOV. - 12 NOV. 1957

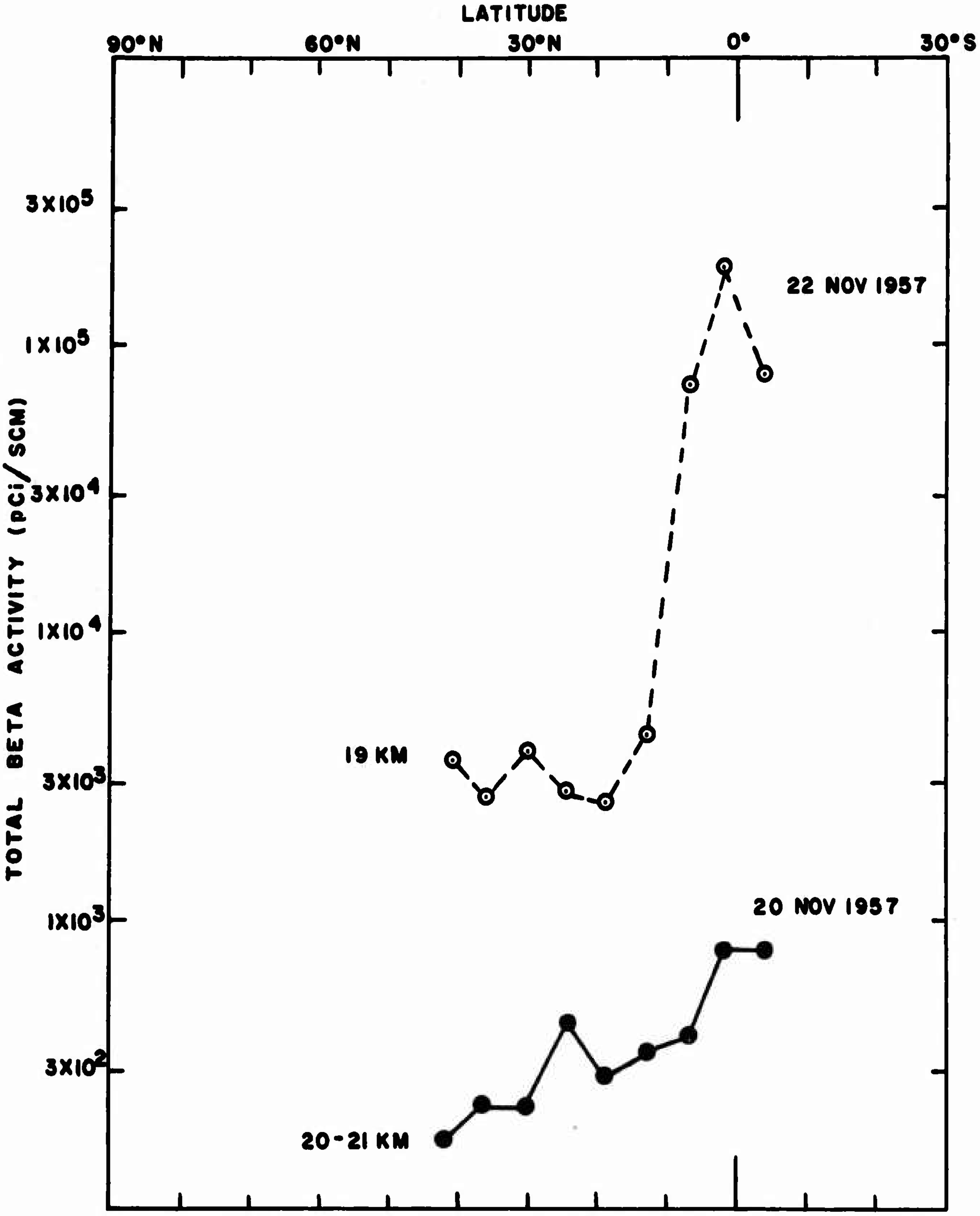


FIGURE 23 TOTAL BETA ACTIVITY AT 19-21 KM, 20 NOV. - 22 NOV. 1957

6.2 Interceptions of Fresh Debris from Early 1958 USSR Weapon Tests

Several filter samples collected during March and April 1958 contained fresh radioactive debris from a series of nuclear weapon tests performed by the USSR in February and March 1958. Announced tests[13] included explosions in the megaton range on 23 February and 27 February 1958 and moderate to low yield explosions on 14, 15, 20 and 22 March 1958. Table 12 lists HASP filter samples which contained high concentrations of beta activity from these tests. The apparent shot dates of the events which produced the fresh radioactive debris in these samples, as estimated from the rates of decay of their total beta activities, are included in the table. The apparent shot dates are obviously at least several days later than the true shot dates. One, or perhaps two, of the February events is represented, as is at least one mid- to late March event. The counting rates of the total beta activity of some of these samples are plotted as a function of time in Figure 24 to illustrate the differences between the decay curves for samples containing fresh debris from the late February events and those for samples containing fresh debris from the mid- to late-March events.

The apparent distributions of total beta activity within the HASP sampling corridor on four specific dates on which USSR radioactivity was intercepted are shown in Figure 25. In drawing the isolines in the figures, it was assumed that the clouds of radioactive debris were increasing in altitude as they moved equatorward, though only for 4 April 1958 were the locations of collection of the samples adequate to provide much evidence on the possible truth of this assumption. It is noteworthy that high concentrations of the radioactivity from these high latitude injections were intercepted

TABLE 12. Samples Containing High Concentrations of Beta Activity from the Early 1958 Tests

Sample Number	Collection Date	Latitude	Altitude (km)	pCi β SCM	Apparent Shot Date
299N	18 Mar 1958	43° - 39°N	13.8	8,900	1 Mar 1958
328N	25 Mar 1958	38° - 33°N	18.3	9,700	1 Mar 1958
356N	25 Mar 1958	38° - 33°N	18.3	10,010	1 Mar 1958
329N	25 Mar 1958	32° - 27°N	18.3	17,630	1 Mar 1958
357N	25 Mar 1958	32° - 27°N	18.3	17,000	1 Mar 1958
360N	28 Mar 1958	55° - 50°N	16.8	9,700	4 Mar 1958
361N	28 Mar 1958	61° - 55°N	16.8	9,380	4 Mar 1958
404N	4 Apr 1958	27° - 21°N	18.3	14,780	23 Mar 1958
410N	8 Apr 1958	66° - 61°N	16.8	11,440	24 Mar 1958
426N	15 Apr 1958	44° - 38°N	16.8	7,950	24 Mar 1958

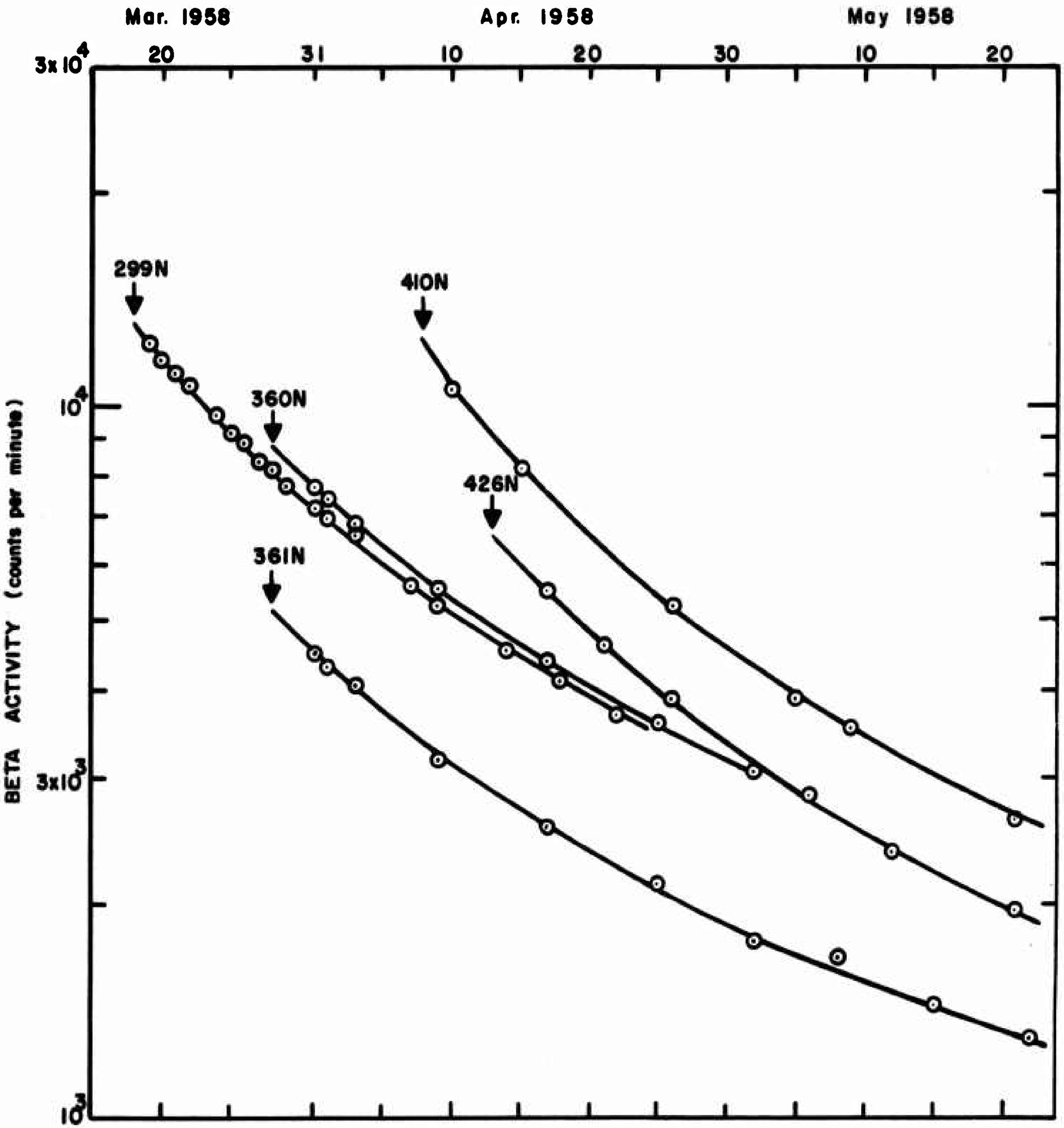


FIGURE 24. DECAY OF BETA ACTIVITY FROM EARLY 1958 USSR WEAPON TESTS

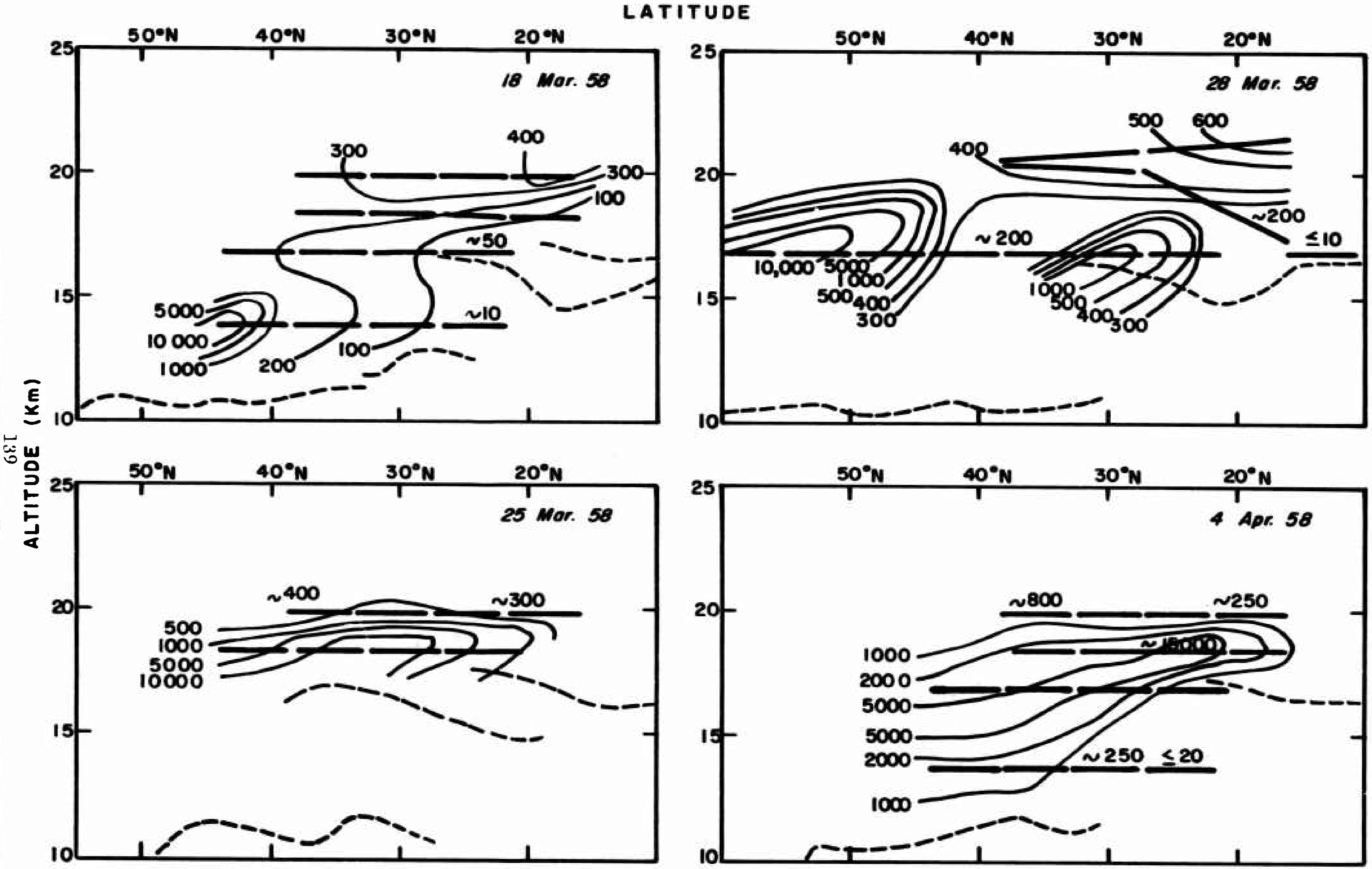


FIGURE 25. THE APPARENT DISTRIBUTION OF TOTAL BETA ACTIVITY (pCi/SCM) ON SPECIFIC DATES IN EARLY 1958 - DASHED LINES INDICATE TROPOPAUSE POSITIONS

at relatively low latitudes (south of 30°N) within a month following its production. During the spring months, air containing high concentrations of ozone, presumably from the equatorial stratosphere, enters the lower polar stratosphere. The movement of the radioactive debris injected into the lower polar stratosphere by the early 1958 USSR tests into the tropical stratosphere during the spring of 1958, and the simultaneous poleward flux of ozone may both be attributed to eddy diffusion within mixing layers which slope from the equator toward the poles within the stratosphere. It would not be reasonable to attribute the poleward flux of ozone to an organized unidirectional flow of air from the equatorial regions into the lower polar stratosphere, for the debris from the USSR tests was moving in the opposite direction.

6.3 Interceptions of Fresh Debris from 1958 U. S. Weapon Tests

Phase I of Operation Hardtack, involving a series of tests of nuclear weapons, took place at the Pacific Proving Ground between 28 April and 12 August 1958. Most of the devices were detonated on barges near the surface of water. The last of these took place on 26 July 1958. Two high altitude rocket bursts of megaton yield took place in August over Johnston Island (17°N). The Teak event took place at 252,000 feet on 1 August 1958, and the Orange event at 141,000 feet on 12 August 1958. This latter event produced a quantity of rhodium-102 radioactive tracer.

The first interception during Project HASP of debris from the U. S. tests took place on 24 May 1958, and was most clearly distinguished by the appearance of tungsten-185 in a number of HASP samples. Presumably fresh fission products accompanied the radiotungsten, but their presence in the

HASP samples was not readily detected. Martell[15] has indicated that the Hardtack tungsten-185 "was produced mainly in five surface-water nuclear explosions during May and July 1958". Evidently the tungsten-185, first intercepted by HASP sampling missions on 24 May 1958, was produced by an event during early May 1958.

Table 13 lists sampling locations, total beta activities, tungsten-185 activities, and apparent shot dates for radioactivity in some filter samples collected during May, June and July 1958. Curves representing the decay of the total beta activity of some of these samples are plotted in Figures 26, 27 and 28. The apparent shot dates listed in Table 13 were calculated from these curves. For the most part, the indicated shot dates are too early coming before the start of the U. S. test series. Because of the relatively small contribution of fission products to the samples by the U. S. tests, the beta activity contained by the samples still significantly reflects the contributions from earlier test series. This probably explains the anomalous apparent dates.

On 8 July 1958, radioactive debris containing significant amounts of fresh fission products from a mid-June 1958 event was intercepted at 19°N. The fresh debris was the main component of the activity found at 13.6, 17.1 and 18.0 km was only a minor component of the activity collected at 16.3 km, and was not present at 19.2 km and the higher altitudes sampled (up to 20.5 km). Presumably the tungsten-185 found in the 8 July 1958 samples was still that which had originated in early May 1958, and which had been sampled previously, for Martell[15] does not indicate that any June 1958 events produced important amounts of tungsten-185. Moreover, the sample collected at 13.6 km, in the troposphere, contained relatively little tungsten-185, although it did contain

TABLE 13. Some May-July 1958 Samples Containing Radioactivity from 1958 U.S. Tests.

Sample Number	Collection Date	Latitude	Altitude (km)	pCi β SCM	pCi W^{185} SCM	Apparent Shot Date
513N	23 May 1958	33° - 27°N	16.8	3,040	358	Mid-April 1958
535N	10 Jun 1958	19°N	17.0	2,830	682	Late March 1958
563N	24 Jun 1958	10° - 5°N	18.3	4,300	1,130	Mid-April 1958
571N	28 Jun 1958	10° - 5°N	18.4	2,880	570	Late April 1958
580N	1 Jul 1958	19°N	20.6	1,130	≤ 3	Late April 1958
588N	4 Jul 1958	5° - 1°S	18.5	1,700	272	Mid-April 1958
595N	8 Jul 1958	19°N	19.7	810	2	Mid-April 1958
594N	8 Jul 1958	19°N	19.2	810	26	Mid-April 1958
592N	8 Jul 1958	19°N	18.0	16,700	710	Mid-June 1958
593N	8 Jul 1958	18°N	17.1	4,490	128	Mid-June 1958
591N	8 Jul 1958	19°N	16.3	302	78	Early May 1958
590N	8 Jul 1958	19°N	13.6	572	10	Mid-June 1958

(Tungsten-185 activities are corrected for decay to 15 August 1958.)

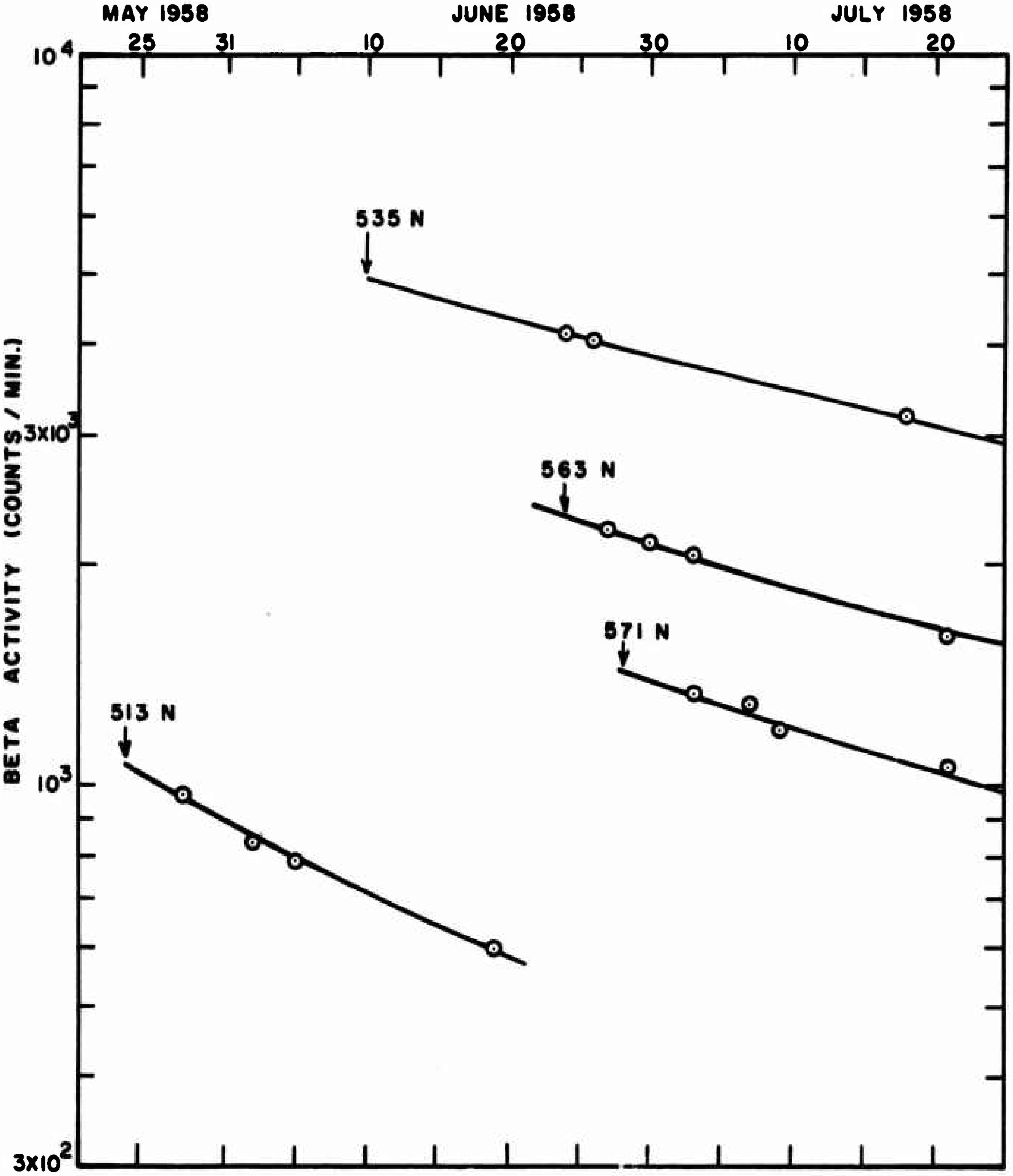


FIGURE 26 DECAY OF BETA ACTIVITY OF SAMPLES CONTAINING DEBRIS FROM 1958 U.S. TESTS

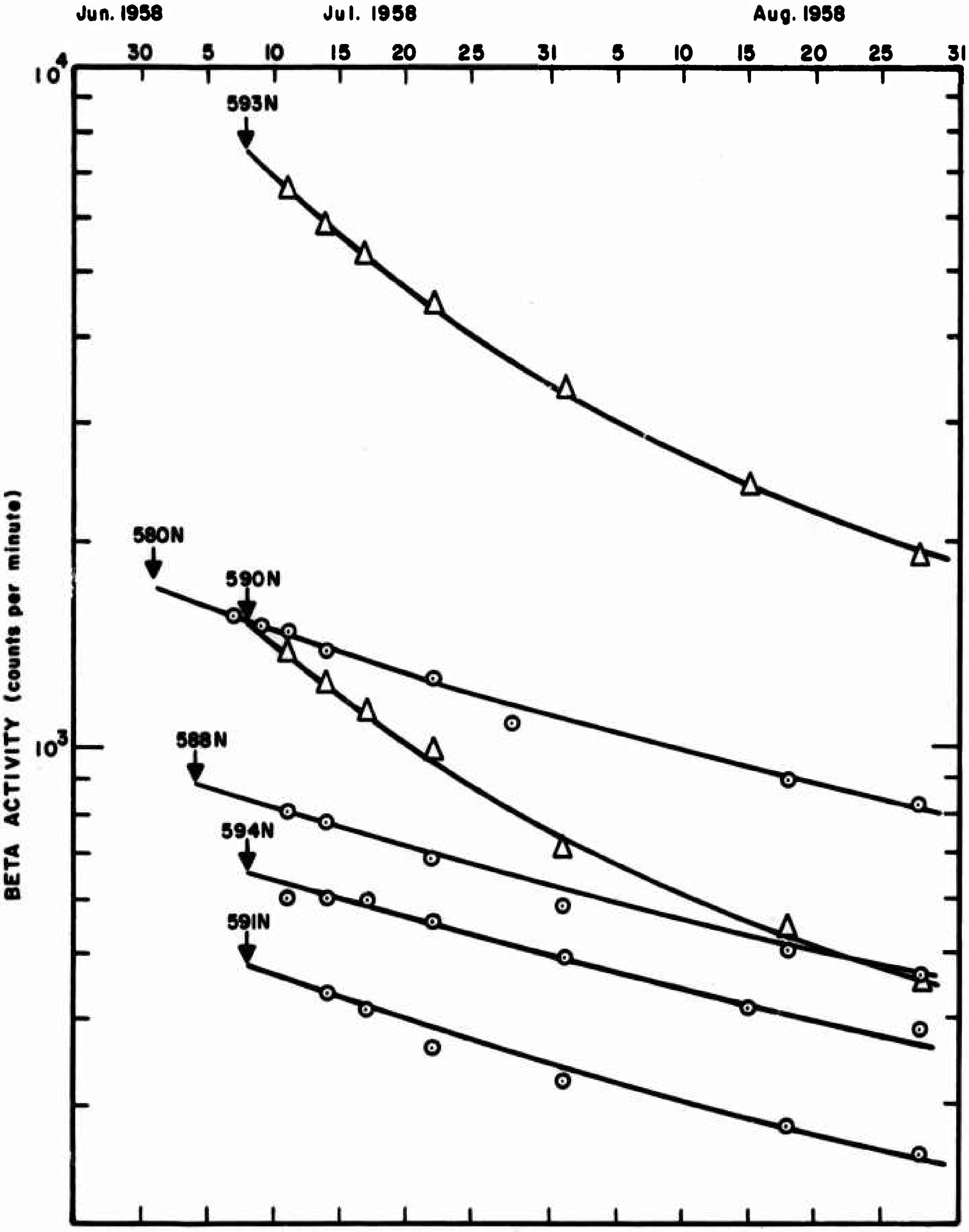


FIGURE 27. DECAY OF BETA ACTIVITY OF SAMPLES CONTAINING DEBRIS FROM 1958 U.S. TESTS

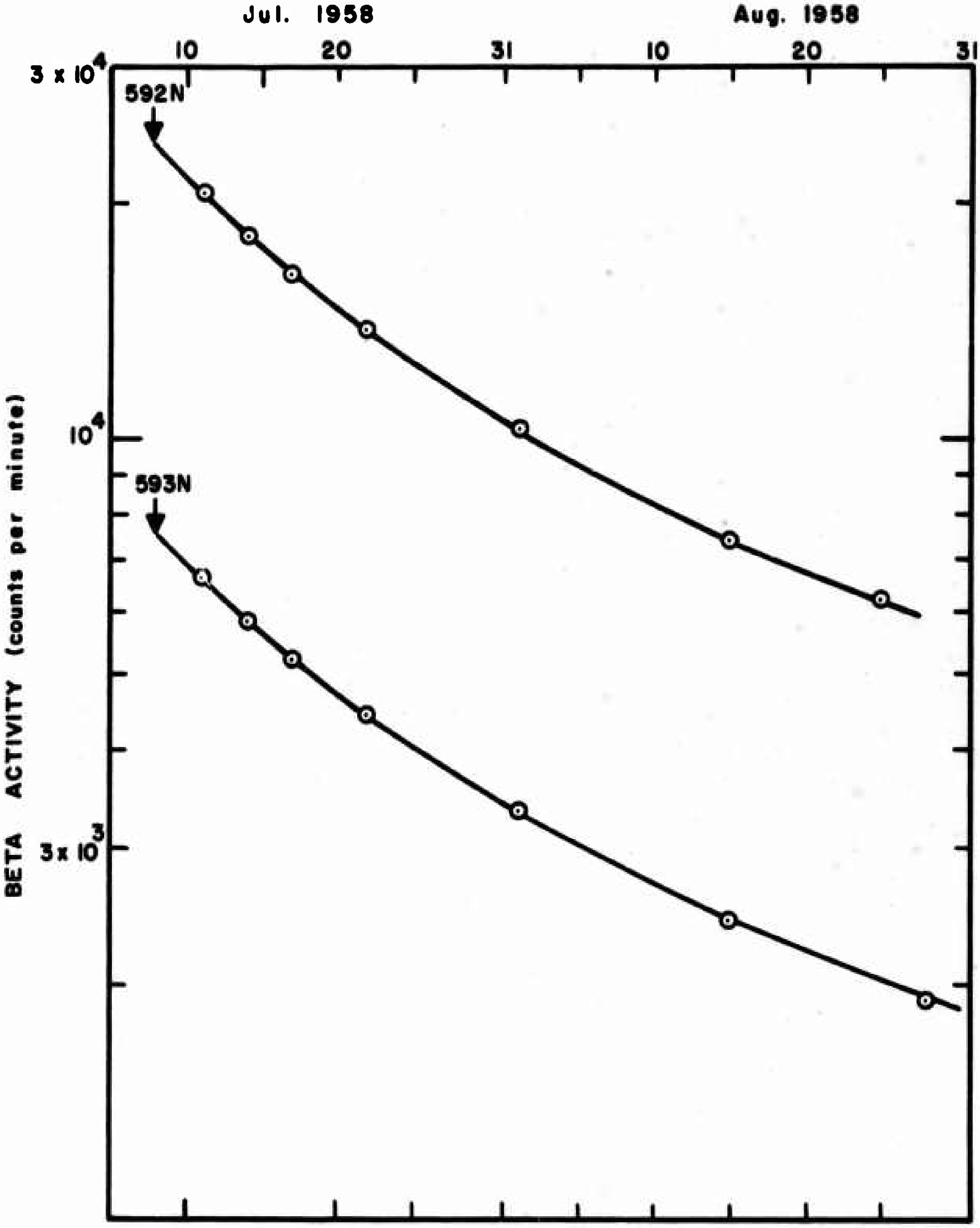


FIGURE 28. DECAY OF BETA ACTIVITY OF 2 SAMPLES CONTAINING DEBRIS FROM A JUNE 1958 U.S. TEST

fission products from the mid-June event. In Figure 29 the vertical profiles of total beta activity and tungsten-185 activity are plotted for samples collected on 3 June 1958 and on 8 July 1958. The tungsten-185 profiles are similar, but the peak in total beta activity found at about 18 km on 8 July 1958 was missing on the earlier date.

Horizontal profiles of total beta and tungsten-185 activity, in the meridional direction, are shown for samples collected on 24 May and 24 June 1958 in Figure 30. On 24 May, when the first interceptions of tungsten-185 occurred, the highest concentrations found were at about 30°N. On 24 June 1958, the highest concentrations were found at about 8°N, close to the latitude of injection. In Figures 31 and 32, flight tracks for four HASP sampling missions have been plotted on cross-sections containing zonal wind profiles along with the 75th meridian[16] in order to show any obvious relationship between the direction and velocity of the zonal wind component and the location of the radioactivity maxima. Isolines of the easterly and westerly components of the wind are plotted in knots. The tropical tropopause is plotted as a heavy line. Tungsten-185 concentrations (corrected for decay to 15 August 1958) and total beta activities are given in units of pCi/SCM. The highest concentrations found on 24 May 1958 were found near, and may be related to, the zone of strong westerlies in the jet stream. Probably the first tungsten-185 to reach the meridian of HASP sampling (65°-70°W) was carried by these westerlies. Later missions which measured meridional profiles at 18.3 km (for example on 24 June 1958: Figure 32) found the maximum at about 10°N, near the latitude of injection, but in a region of very low zonal winds. Later missions which measured vertical profiles at 10°N (for example on 6 June 1958: Figure 31, and on 8 July 1958: Figure 32) found the

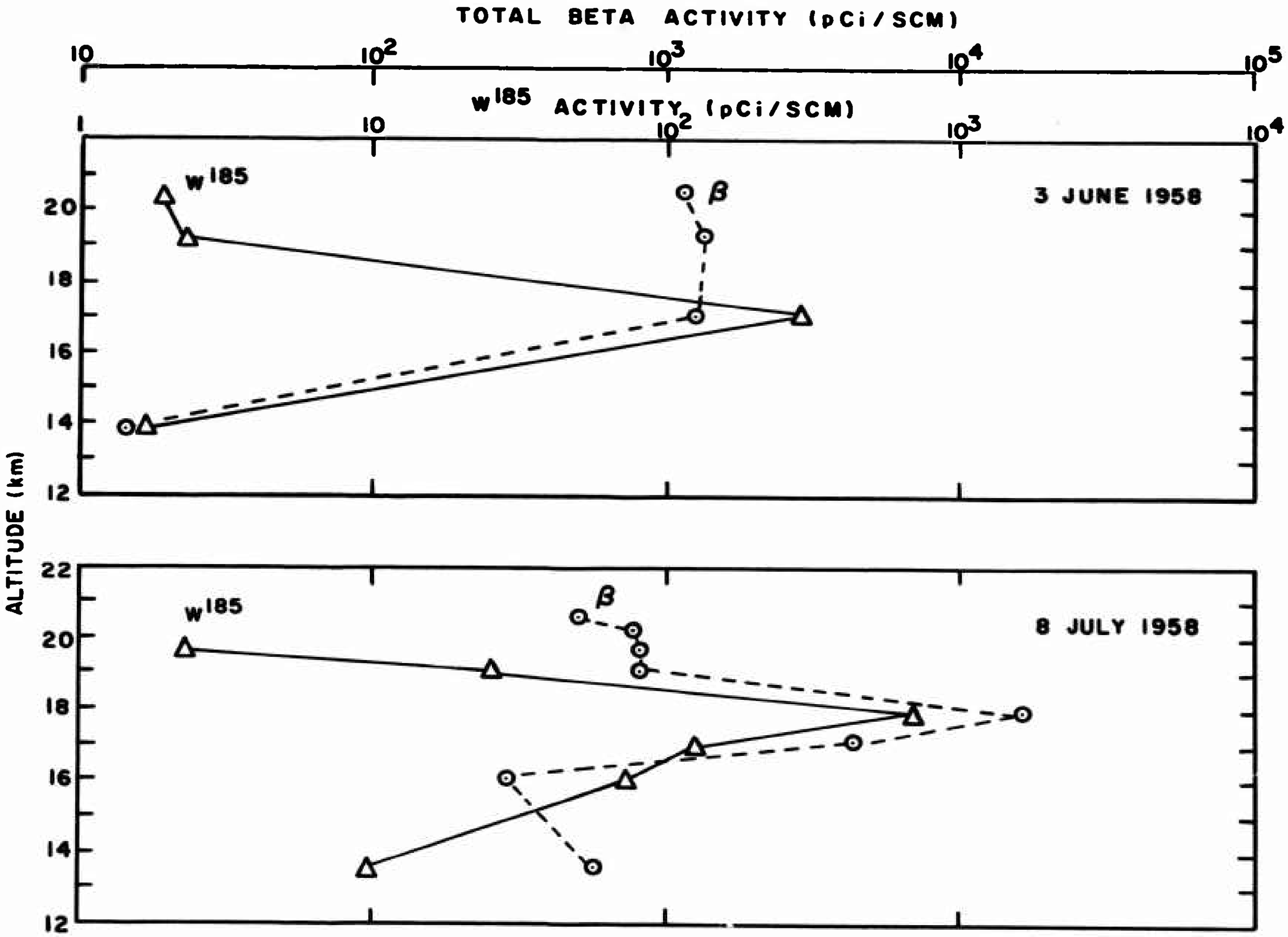


FIGURE 29 VERTICAL PROFILES OF TOTAL BETA AND TUNGSTEN-185 ACTIVITY AT 19°N ON 3 JUNE AND 8 JULY 1958.

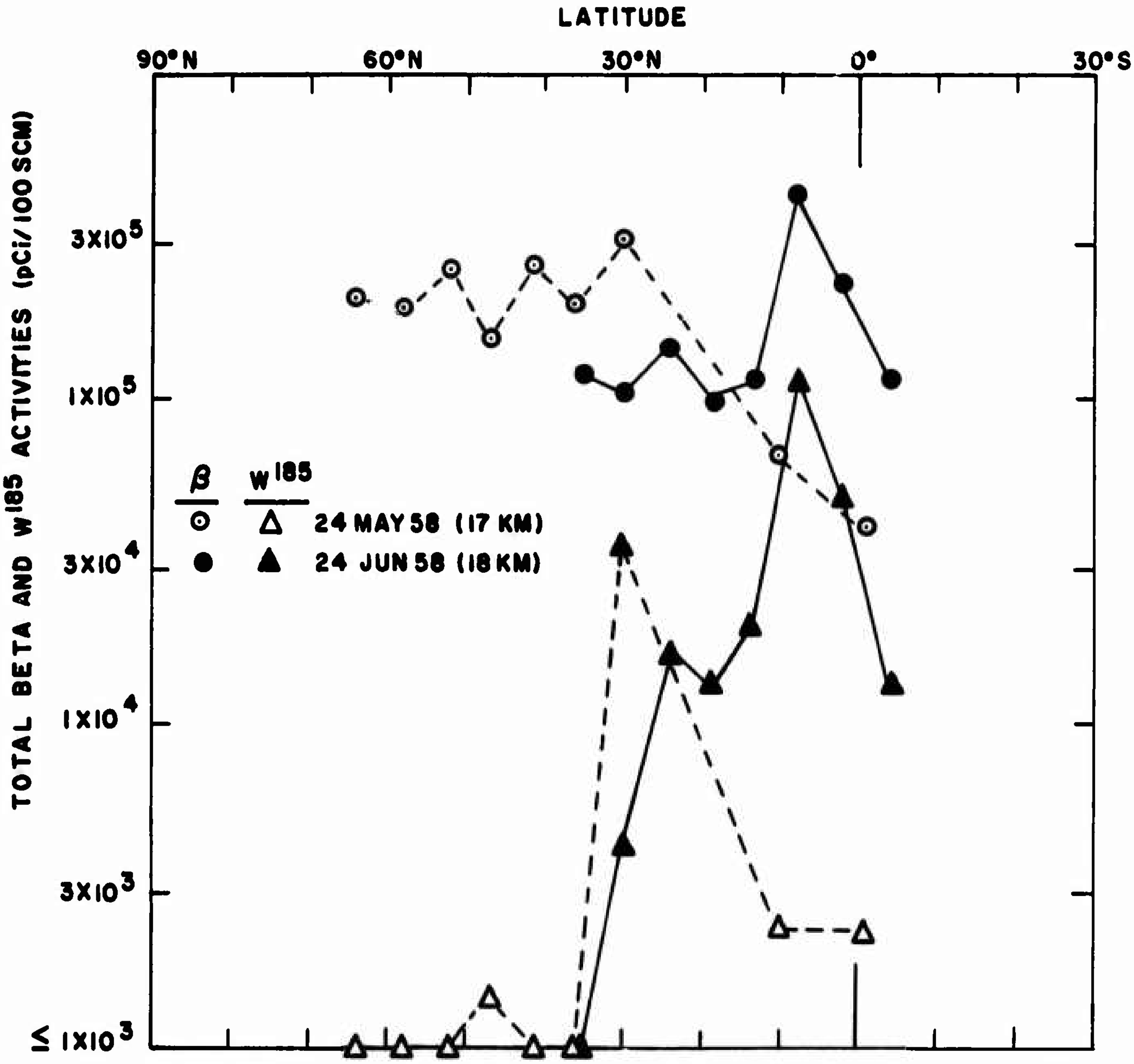


FIGURE 30. MERIDIONAL PROFILES OF TOTAL BETA AND TUNGSTEN-185 ACTIVITY ON 24 MAY 1958 AND 24 JUNE 1958

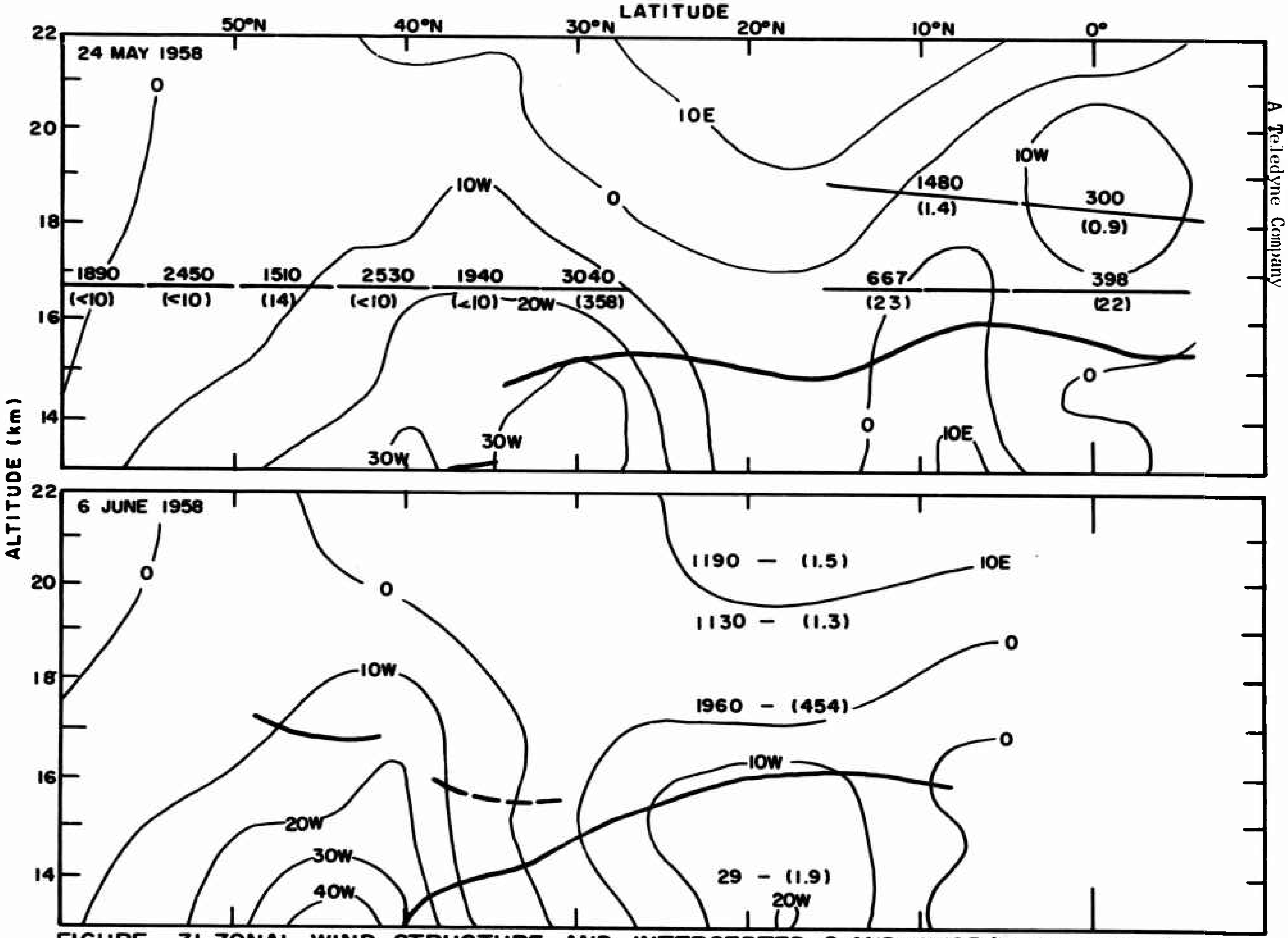


FIGURE 31. ZONAL WIND STRUCTURE AND INTERCEPTED β AND W-185 (in parentheses) ACTIVITIES ON 24 MAY AND 6 JUNE 1958

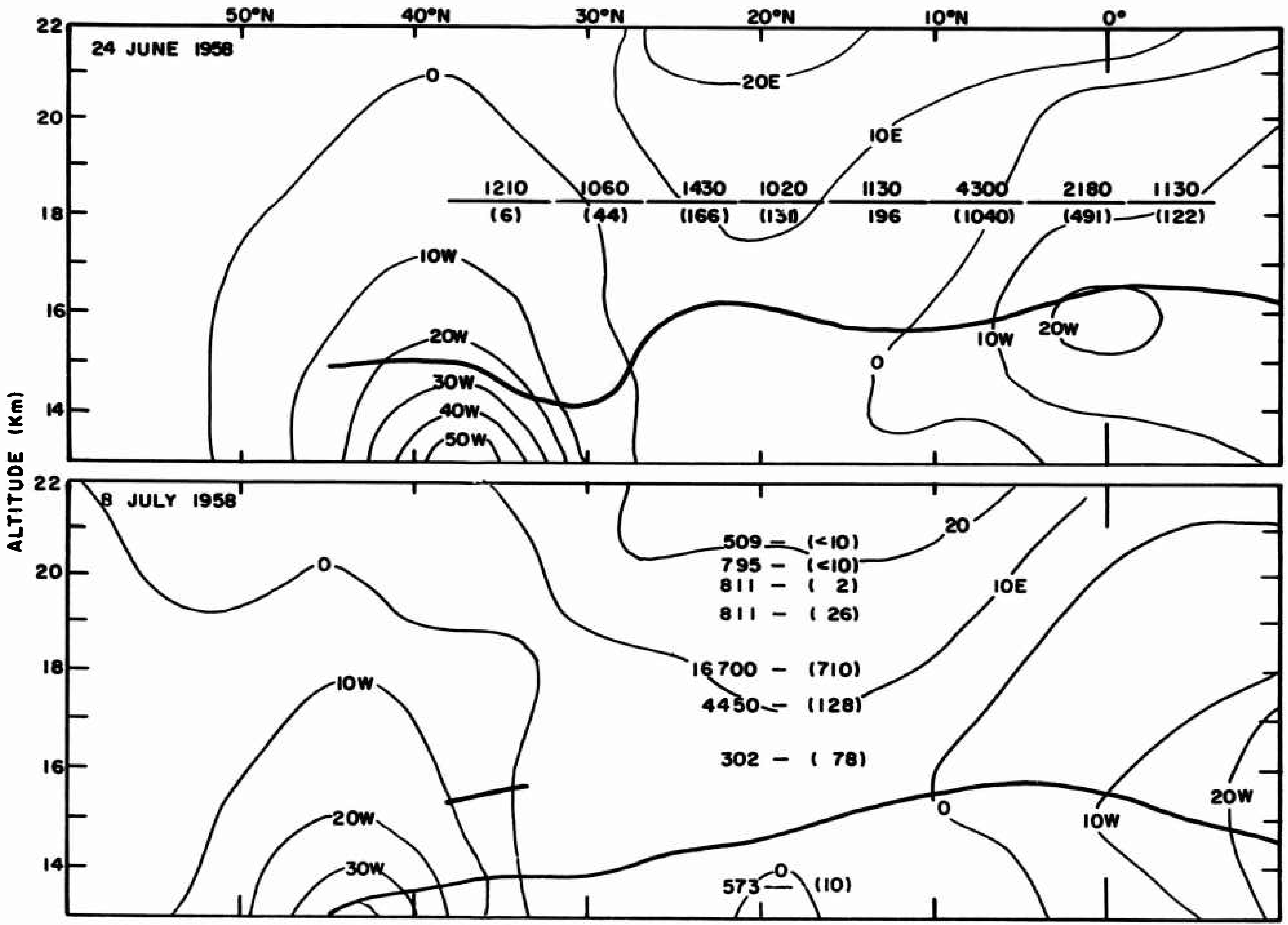


FIGURE 32. ZONAL WIND STRUCTURE AND INTERCEPTED β AND W-185 (in parentheses) ACTIVITIES ON 24 JUNE AND 8 JULY 1958

maximum concentration at 17 to 18 km, also generally in a region of low velocity zonal winds. Thus the drift of the radioactivity around the earth in the zonal direction could not be correlated with any particular wind system, except for its first appearance in the strong westerlies. The arrival of the radioactivity at the sites at which it was sampled probably involved significant north-south motion, and perhaps vertical motion, as well as movement in the zonal direction, and the frequency and location of sampling was inadequate to elucidate the trajectories followed.

The observations during May to July of the radioactivity from the U. S. tests raises two questions for which answers may be suggested but may not be completely proven. The first question is whether or not any of the weapon tests performed in April and May 1958, except for that which produced the tungsten-185 sampled during the period 24 May to 8 July 1958, injected any significant fraction of their radioactivity into the stratosphere. The available evidence indicates that none did, for the frequency and location of sampling should have been adequate to detect any significant injection of fission products into the tropical stratosphere. The second question is whether or not the vertical profiles of tungsten-185 observed during 24 May to 8 July 1958 were truly indicative of the vertical distribution of the tungsten-185 injected into the stratosphere in May 1958, or whether the radioactive cloud containing the tungsten was torn apart by wind shear and only the lower section of the cloud reached the HASP sampling corridor. The available evidence indicates that the observed vertical profiles closely corresponded to the mean vertical distribution of tungsten-185 activity, for the easterly winds at 20 km over 19°N should have carried high activities of tungsten-185 into the HASP sampling corridor well before 8 July 1958 if significant quantities of that nuclide had been injected at that altitude in early May 1958.

No HASP sampling missions were flown between 8 July and 12 September 1958. A mission flown on 12 September 1958 at 19 to 20 km used aircraft from Ezeiza Airfield, Argentina. Sampling during all subsequent missions flown in the tropical stratosphere during the next few months was done at about 18 km, so that the vertical distribution of activity injected by the U. S. tests during the second half of June and during July 1958 it not well known. Table 14 gives flight data, total beta and tungsten-185 activities, and apparent shot dates of samples collected during September and early October 1958 which contain debris from the 1958 U. S. weapon tests. The decay of the total beta activity of some of these samples is illustrated in Figure 33. The samples containing the highest concentrations had an apparent origin in about mid-July 1958. Presumably much of the tungsten-185 in these samples as well as the short-lived fission products originated in the July 1958 events[15]. On 3 October 1958, the first interception was made of radioactive debris from the September 1958 U.K. tests, and subsequent to this date this most recent debris tended to dominate the fission product content of the lower tropical stratosphere.

Meridional profiles of tungsten-185 activity (corrected for decay to 15 August 1958) and of total beta activity are shown in Figure 34 for 4 July 1958 at 18 km (before the interception of debris from the July 1958 tests) and for 12 September 1958 at 20 km and 23 and 30 September 1958 (combined) at 18 km (after debris from July 1958 tests reached the HASP sampling corridor). It appears that the July 1958 tests resulted in a substantial

TABLE 14. Some September-October 1958 Samples Containing Radioactivity from 1958 U.S. Tests

Sample Number	Collection Date	Latitude	Altitude (km)	pCi β SCM	pCi W^{185} SCM	Apparent Shot Date
598N	12 Sep 1958	10° - 4°N	19.2	7,150	1,480	Mid-July 1958
599N	12 Sep 1958	4° - 1°S	19.6	4,610	678	Mid-July 1958
603N	12 Sep 1958	13° - 18°S	20.4	1,610	12	Late May 1958
613N	16 Sep 1958	16° - 11°N	18.3	4,930	1,740	Mid-July 1958
610N	16 Sep 1958	1° - 6°S	18.3	1,700	455	Late June 1958
628N	23 Sep 1958	21° - 16°N	18.3	5,890	3,280	Mid-July 1958
652N	3 Oct 1958	21° - 16°N	18.3	3,500	975	Mid-July 1958
656N	3 Oct 1958	16° - 10°N	18.3	4,450	2,300	Late July 1958
655N	3 Oct 1958	10° - 4°N	18.3	6,840	1,750	Late August 1958
654N	3 Oct 1958	4° - 2°S	18.3	43,000	840	Early September 1958
653N	3 Oct 1958	2° - 8°S	18.3	9,700	1,390	Early September 1958

(Tungsten-185 activities are corrected for decay to 15 August 1958.)

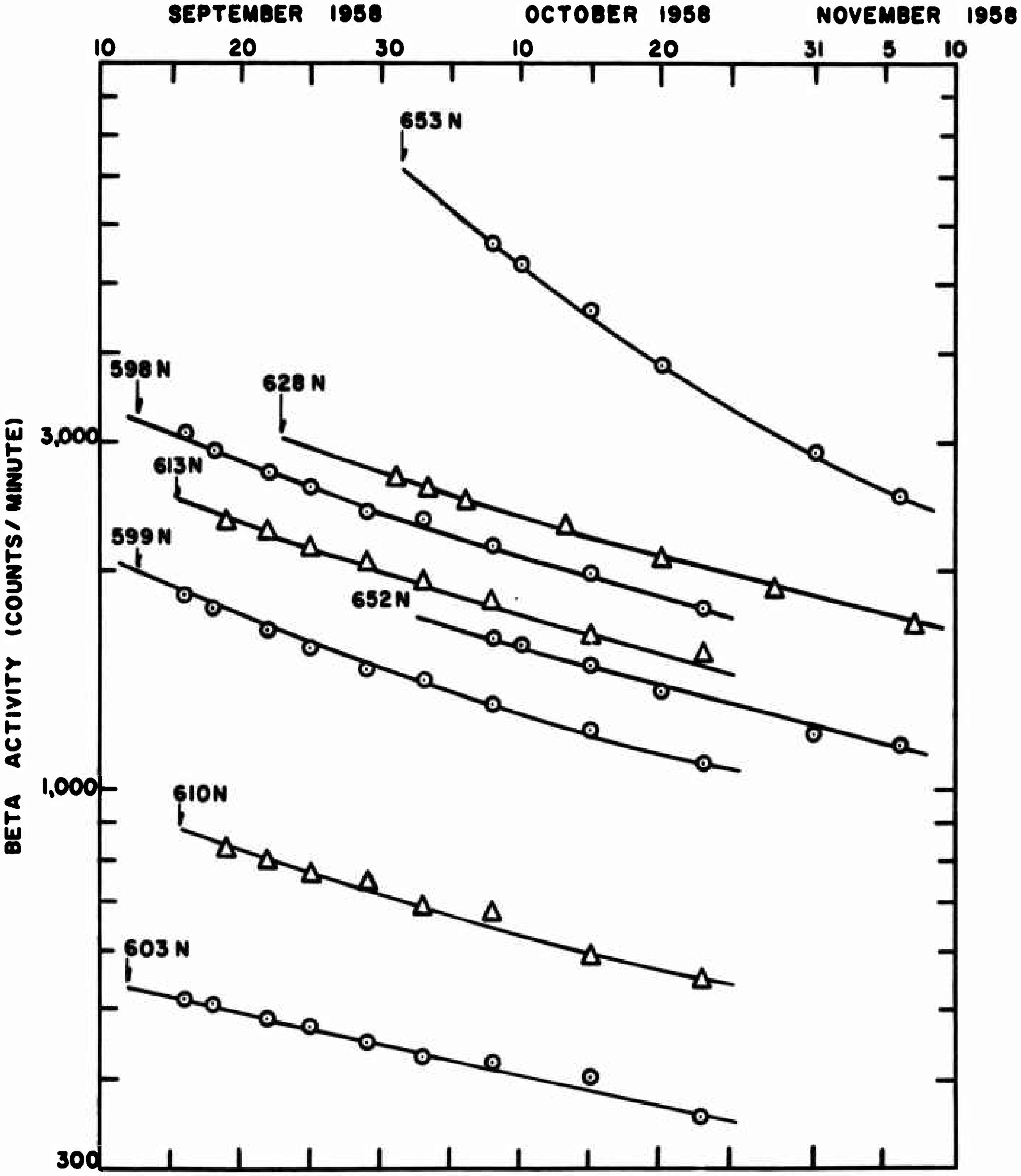


FIGURE 33 DECAY OF BETA ACTIVITY OF SOME SAMPLES CONTAINING DEBRIS FROM 1958 U.S. TESTS

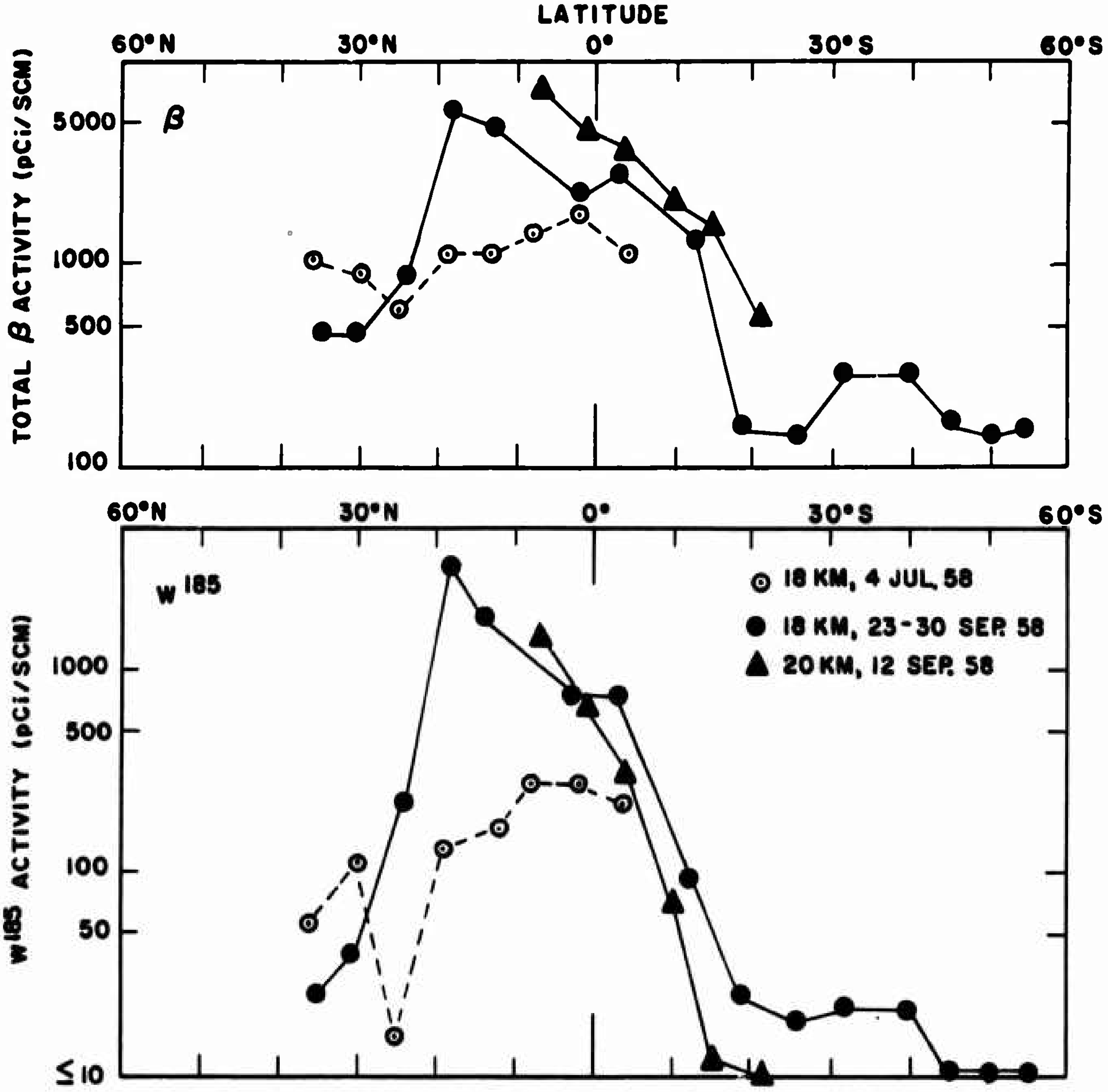


FIGURE 34. MERIDIONAL PROFILES OF TOTAL BETA AND TUNGSTEN-185 ACTIVITIES IN JULY AND SEPTEMBER 1958

increase in concentrations both of fission products and of tungsten-185 in the northern tropical stratosphere at 18 km (and doubtless also at 20 km). It is evident that the largest fraction of the fission products, and a still larger fraction of the tungsten-185, remained in the northern hemisphere, rather than entering the southern hemisphere, during the first two months following its injection. It also appears from Figure 34 that south of the equator the highest concentrations of total beta activity were probably at or above 20 km. This suggests, though the evidence is far from being conclusive, that the mean altitude of injection of total beta activity was higher than the mean altitude of injection of tungsten-185.

Martell[15] hypothesized that the radioactive debris from the U. S. 1958 Operation Hardtack tests was injected into the stratosphere as distinct layers which were well mixed internally but were effectively isolated from each other. He estimated that debris from tests with total yields in the range 0.15 to 0.5 megaton stabilized "near the tropopause", and that the W^{185}/Sr^{90} ratio, corrected to 15 August 1958, in this debris was 220. He estimated that debris from tests with total yields in the range 0.5 to 2.0 megatons stabilized near 20 km, and that it had a W^{185}/Sr^{90} ratio of 445. Finally, he estimated that debris from tests with total yields between 8 and 10 megatons stabilized near 30 km, and contained a W^{185}/Sr^{90} ratio of 42. Using data for some HASP filter samples collected at 18 km during September and October 1958, he found evidence for the existence of the layer with a W^{185}/Sr^{90} ratio of 445.

In Table 15 are summarized data for HASP samples collected during September and October 1958 which seem to contain radioactivity predominantly from the Hardtack tests. The W^{185}/Sr^{90} ratios have been calculated using

TABLE 15. W^{185}/Sr^{90} Ratios in Some Samples of 1958 U.S. Radioactive Debris.

Sample Number	Collection Date	Latitude	Altitude (km)	$\frac{\text{pCi } \beta}{\text{100 SCM}}$	$\frac{Sr^{89}}{Sr^{90}}$	pCi Sr^{90}/100 SCM		$\frac{\text{pCi } W^{185}}{\text{100 SCM}}$	$\frac{W^{185}}{Sr^{90}}$
						Total	Hardtack		
598N	12 Sep 1958	10° - 4°N	19.2	715,000	92	1,140	1,050	148,000	141
599N	12 Sep 1958	4°N- 1°S	19.6	461,000	82	954	781	67,800	87
600N	12 Sep 1958	2° - 7°S	19.9	350,000	80	695	557	33,700	60
605N	12 Sep 1958	1° - 7°S	20.1	413,000	105	760	760	31,700	42
601N	12 Sep 1958	7° - 12°S	20.0	170,000	48	431	207	7,340	35
602N	12 Sep 1958	7° - 12°S	20.4	238,000	50	680	340	6,800	20
603N	12 Sep 1958	13° - 18°S	20.4	161,000	31	363	113	1,220	11
613N	16 Sep 1958	16° - 11°N	18.3	493,000	76	764	580	174,000	300
612N	16 Sep 1958	10° - 5°N	18.3	429,000	74	684	506	104,000	205
611N	16 Sep 1958	5°N- 1°S	18.3	350,000	77	665	512	109,000	213
610N	16 Sep 1958	1° - 6°S	18.3	170,000	66	407	269	45,500	169
615N	19 Sep 1958	3° - 8°S	18.3	278,000	79	455	360	77,500	215
618N	19 Sep 1958	9° - 16°S	18.3	103,000	34	463	157	20,100	128
626N	23 Sep 1958	32° - 27°N	18.3	44,500	20	212	42	4,200	100
627N	23 Sep 1958	27° - 21°N	18.3	87,500	97	205	199	22,300	112
628N	23 Sep 1958	21° - 16°N	18.3	589,000	92	607	559	328,000	587
632N	23 Sep 1958	16° - 11°N	18.3	493,000	100	416	416	169,000	406
630N	23 Sep 1958	5° - 0°N	18.3	207,000	75	228	171	79,100	462
629N	23 Sep 1958	1° - 6°S	18.3	286,000	114	336	336	72,100	215
633N	30 Sep 1958	10° - 16°S	18.3	143,600	106	192	192	8,500	44

TABLE 15. (continued)

Sample Number	Collection Date	Latitude	Altitude (km)	pCi β / 100 SCM	Sr^{89}/Sr^{90}	pCi Sr^{90}/100 SCM Total	pCi Sr^{90}/100 SCM Hardtack	pCi W^{185} / 100 SCM	W^{185}/Sr^{90}
651N	3 Oct 1958	27° - 22°N	18.3	216,000	74	388	287	13,800	48
652N	3 Oct 1958	21° - 16°N	18.3	350,000	80	548	440	97,500	222
656N	3 Oct 1958	16° - 10°N	18.3	445,000	89	588	523	230,000	440
666N	7 Oct 1958	21° - 16°N	18.3	116,000	80	318	254	34,800	137
669N	7 Oct 1958	16° - 11°N	18.5	194,000	72	358	258	52,700	205
673N	10 Oct 1958	21° - 16°N	18.3	82,600	43	299	128	16,800	131
682N	14 Oct 1958	35° - 40°S	11.6	41,300	32	164	52	2,070	40

Martell's assumption that all strontium-89 they contain was produced by the Hardtack tests, and that the Sr^{89}/Sr^{90} ratio in Hardtack debris, corrected to 15 August 1958, was 100. In a few of the samples, most notably those with the highest tungsten-185 activities, the W^{185}/Sr^{90} ratio does approximate the 445 value Martell predicts. In most samples, however, including those collected at 19 to 20 km on 12 September 1958, the ratio is well below the expected value.

The data suggest that the height of stabilization of the radioactive clouds containing tungsten-185 was lower than predicted by Martell[15], and that probably less of the tungsten-185 was injected into the stratosphere than he estimated (204 MCi). As a result the W^{185}/Sr^{90} ratio was generally lower than he expected. It is quite possible that the tungsten-185 in the few high activity samples with high W^{185}/Sr^{90} ratios came largely from the last major tungsten-185 injection in July 1958. The observed ratios in the samples collected at 19 to 20 km can best be explained if it is assumed that there was considerable overlapping of heights of injection of debris from the events with yields in the 0.5 to 2.0 megaton range and debris from events with yields in the 8 to 10 megaton range, with material from both layers being represented at 20 km.

6.4 Interceptions of Fresh Debris from September 1958 U.K. Weapon Tests

On 30 October 1958 a HASP sampling mission intercepted fresh debris in the equatorial stratosphere at 18.3 km. The rate of decay of the total beta activity in these samples indicated that the debris originated in early September 1958. The U.K. performed two tests of megaton yield nuclear weapons in the vicinity of Christmas Island (2°N, 157°W) in early September 1958, one on 2 September and one on 11 September[13], and one or both of these events doubtless was the source of the fresh debris intercepted in the equatorial stratosphere on and after 3 October 1958.

Table 16 lists flight data, total beta activities, strontium-90 activities and apparent shot dates for a number of samples collected during October to December 1958 which appeared to contain fission products predominantly from the September 1958 events. The apparent shot dates are calculated from the rates of decay of the total beta activities. The decay rates of some of these samples are illustrated in Figure 35. Sample 719N, which contained activity mainly from an October 1958 USSR nuclear weapon test, is included in Table 16 and Figure 35 for purposes of contrast.

By 17 October 1958 the debris from the U.K. tests constituted the main contributor of fission products found between 28°N and 23°S in the tropical stratosphere. Evidently, this debris was distributed quite symmetrically about the latitude of injection. It is of interest also that on 19 October 1958 the region at 18 km between 38° and 32°N contained, as it had since 23 September 1958 when it was first sampled following Operation Hardtack; only negligible quantities of debris from any of the low latitude tests performed during 1958.

In Figure 36 are plotted the apparent shot dates of a number of HASP samples, estimated from their measured Ba^{140}/Sr^{89} activity ratios. The apparent shot dates were calculated assuming an initial activity ratio of 8.0 and a "half-life" of the ratio of 17.1 days. The five September samples gave apparent shot dates ranging from late May to late July 1958, the period of the Hardtack tests. All but one sample collected in the tropical stratosphere during October gave apparent shot dates which correlate well with the early September 1958 U.K. injections. The ratios in samples collected in the northern polar stratosphere during October generally indicated origin in the October 1958 USSR weapon tests. An exception was sample 726N, collected

TABLE 16. Some Samples Containing Radioactivity from September 1958 U.K. Tests

Sample Number	Collection Date	Latitude	Altitude (km)	pCi β SCM	pCi Sr^{90} SCM	Apparent Shot Date
654N	3 Oct 1958	4°N- 2°S	18.3	43,000	21	12 September 1958
667N	7 Oct 1958	5° - 1°N	18.3	44,500	29	12 September 1958
675N	10 Oct 1958	3°N- 2°S	18.3	29,200	17	Mid-September 1958
690N	14 Oct 1958	10° - 5°N	18.3	17,500	20	Mid-September 1958
694N	17 Oct 1958	28° - 23°N	18.3	5,720	9.2	3 September 1958
697N	17 Oct 1958	4°N- 2°S	18.3	15,400	18	15 September 1958
702N	17 Oct 1958	16° - 23°S	18.3	4,290	3.5	16 September 1958
719N	19 Oct 1958	44° - 30°N	18.0	12,200	9.2	7 October 1958
718N	19 Oct 1958	38° - 32°N	18.2	334	2.7	February 1958
717N	19 Oct 1958	32° - 26°N	18.3	4,610	6.2	3 September 1958
716N	19 Oct 1958	26° - 20°N	18.3	5,250	15	3 September 1958
771N	5 Nov 1958	27° - 22°N	18.3	2,970	6.8	8 September 1958
777N	7 Nov 1958	2° - 8°S	18.3	3,980	7.1	10 September 1958
936N	19 Dec 1958	32° - 27°N	19.8	2,460	8.2	Early September 1958
942N	19 Dec 1958	16° - 10°N	19.8	2,670	8.1	Early September 1958
940N	19 Dec 1958	4°N- 2°S	19.8	2,180	7.1	Early September 1958

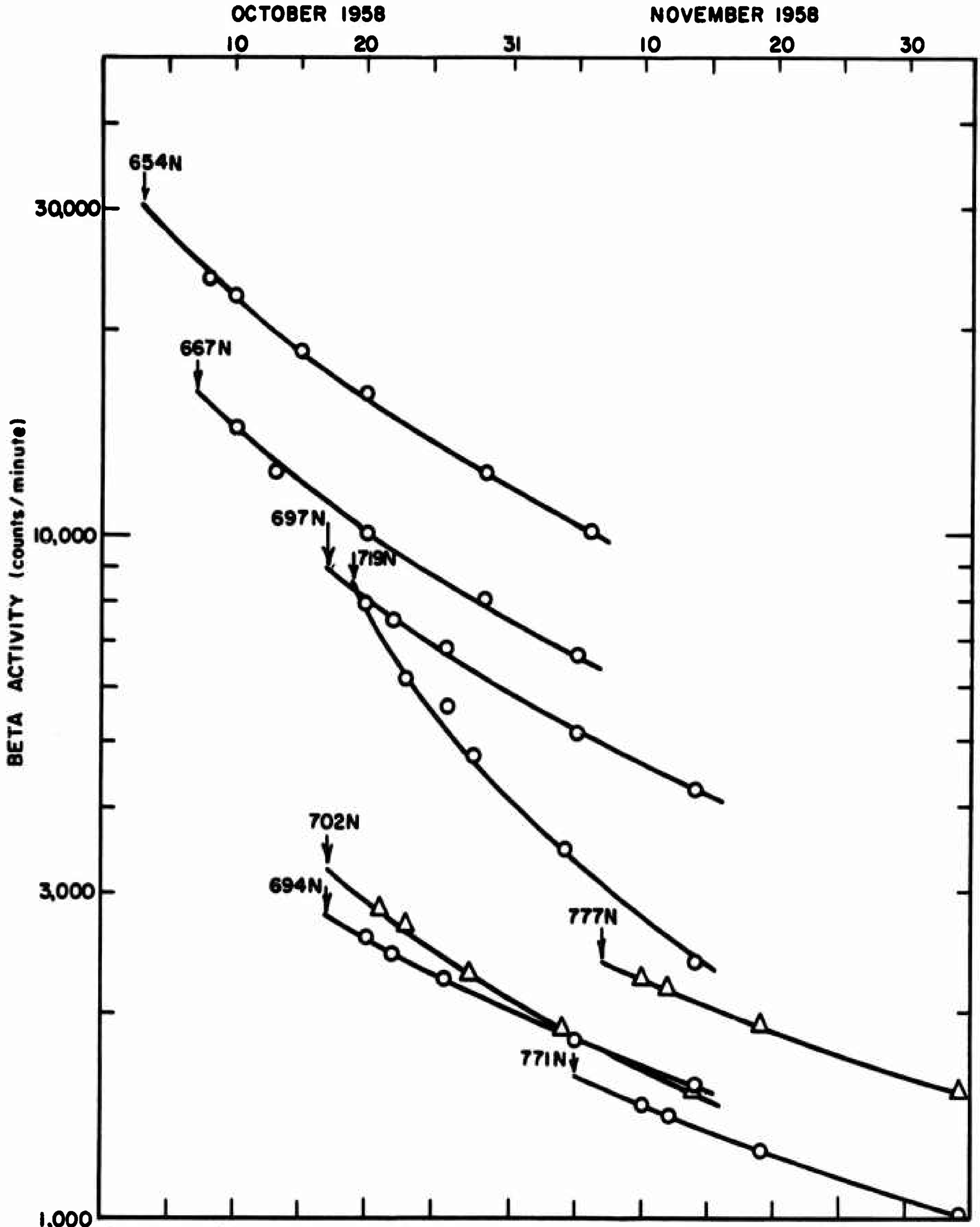


FIGURE 35. DECAY OF BETA ACTIVITY OF SOME SAMPLES CONTAINING DEBRIS FROM SEPTEMBER 1958 U.K. WEAPON TESTS

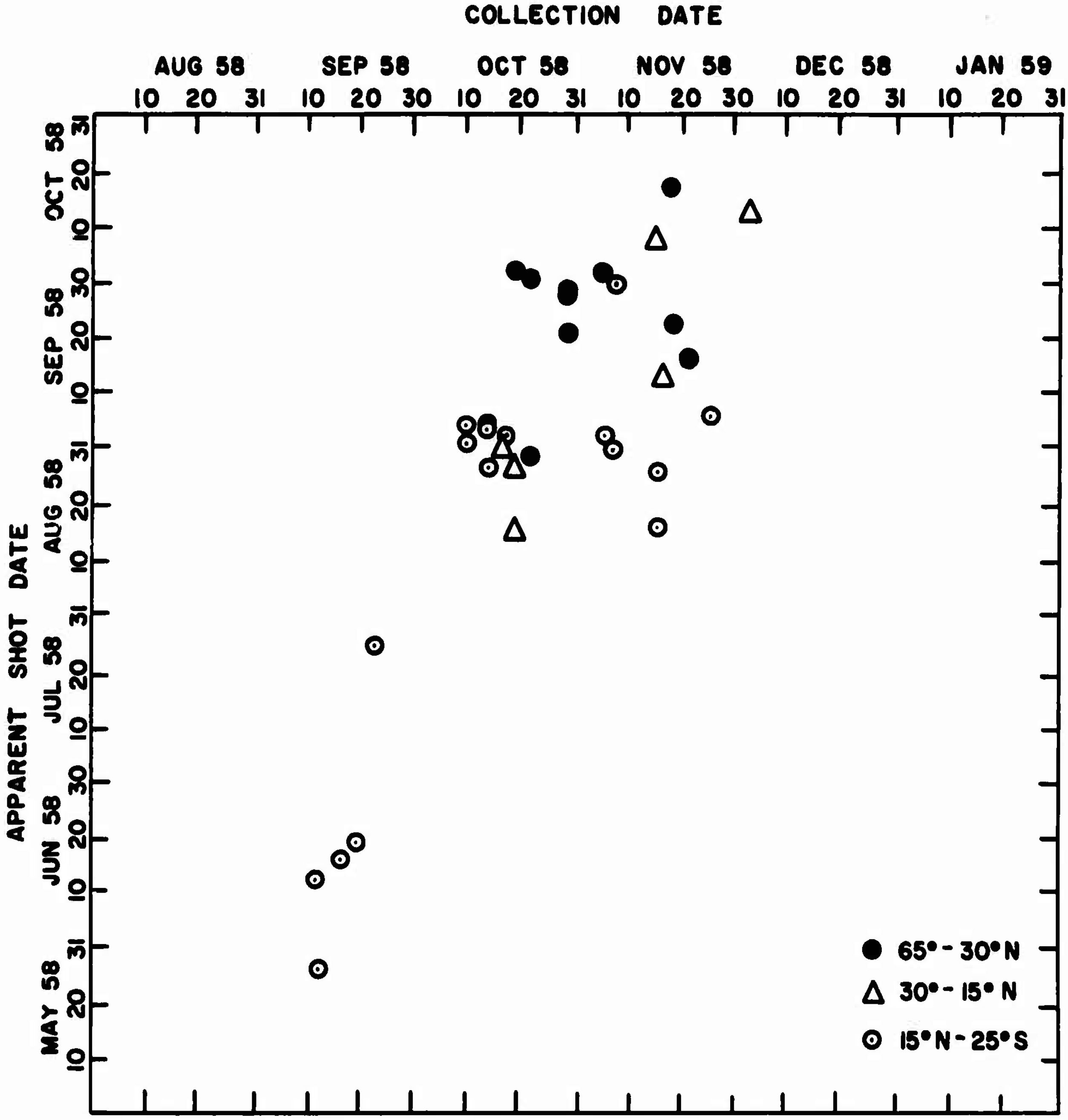


FIGURE 36. APPARENT SHOT DATES CALCULATED FROM Ba-140 / Sr-89 RATIOS

at 17 km near 50°N on 22 October 1958. The possible origins of the fission products contained by this sample are the September 1958 U.K. tests. The sample contained high concentrations of tungsten-185 indicating that debris in it had originated in the tropical stratosphere. The data for samples collected in November 1958 show more scatter, in part due to analytical errors in the measurement of the decreasing concentrations of barium-140 and strontium-89, but in part also to the intermixing of debris from the various 1958 test series. Sample 784N, collected at 18 km near 13°S on 7 November, gave an apparent shot date of 30 September 1958, but the high Ba^{140}/Sr^{89} ratio in this sample is almost certainly attributable to strontium-89 value that was too low. The presence of fission products from the October 1958 USSR tests in samples collected between 30° and 15°N during the second half of November and early December 1958 is confirmed by the rate of decay of the total beta activities of these samples.

6.5 Interceptions of Fresh Debris from October 1958 USSR Weapon Tests

The first interception of radioactive debris from the October 1958 USSR nuclear weapon tests[13] occurred on 19 October 1958. Additional interceptions occurred during the next few months. Flight data, total beta and strontium-90 activities, and apparent shot dates for samples containing this USSR debris are listed in Table 17. The apparent shot dates are calculated from rates of decay of the total beta activities, and these rates of decay are plotted for a few samples in Figure 37. Data for sample 717N, which contained debris from the September 1958 U.K. tests are included in Figure 37 for contrast.

Several features of the interceptions of the October 1958 USSR debris are noteworthy. It is surprising, for example, that all of the radioactivity

TABLE 17. Some Samples Containing Radioactivity from October 1958 U.S.S.R. Tests

Sample Number	Collection Date	Latitude	Altitude (km)	pCi β / SCM	pCi Sr^{90} / SCM	Apparent Shot Date
719N	19 Oct 1958	44° - 39°N	18.0	12,200	9.2	7 October 1958
732N	22 Oct 1958	44°N	18.3	24,600	16	11 October 1958
760N	29 Oct 1958	38° - 32°N	15.2	4,610	3.8	7 October 1958
755N	29 Oct 1958	44° - 38°N	16.8	7,000	3.8	Early October 1958
751N	29 Oct 1958	44° - 38°N	18.3	46,000	29	9 October 1958
752N	29 Oct 1958	38° - 32°N	18.3	3,820	6.5	7 October 1958
753N	29 Oct 1958	26° - 20°N	18.3	3,660	4.3	8 October 1958
798N	15 Nov 1958	27° - 17°N	18.3	9,060	11	Mid-October 1958
806N	18 Nov 1958	55° - 49°N	15.2	7,950	7.4	Mid-October 1958
831N	22 Nov 1958	44° - 38°N	16.8	2,050	3.7	Early October 1958
837N	22 Nov 1958	32° - 26°N	18.3	2,210	3.7	Early October 1958
866N	3 Dec 1958	32° - 27°N	18.3	1,940	5.4	Early October 1958
904N	12 Dec 1958	33° - 27°N	18.3	1,690	4.1	Early October 1958
927N	16 Dec 1958	38° - 33°N	16.8	1,670	4.4	Early October 1958
952N	23 Dec 1958	32° - 27°N	18.3	2,480	5.0	Early October 1958
953N	23 Dec 1958	27° - 22°N	18.3	2,060	4.0	Early October 1958
977N	6 Jan 1959	38° - 33°N	15.2	1,140	3.4	Early October 1958

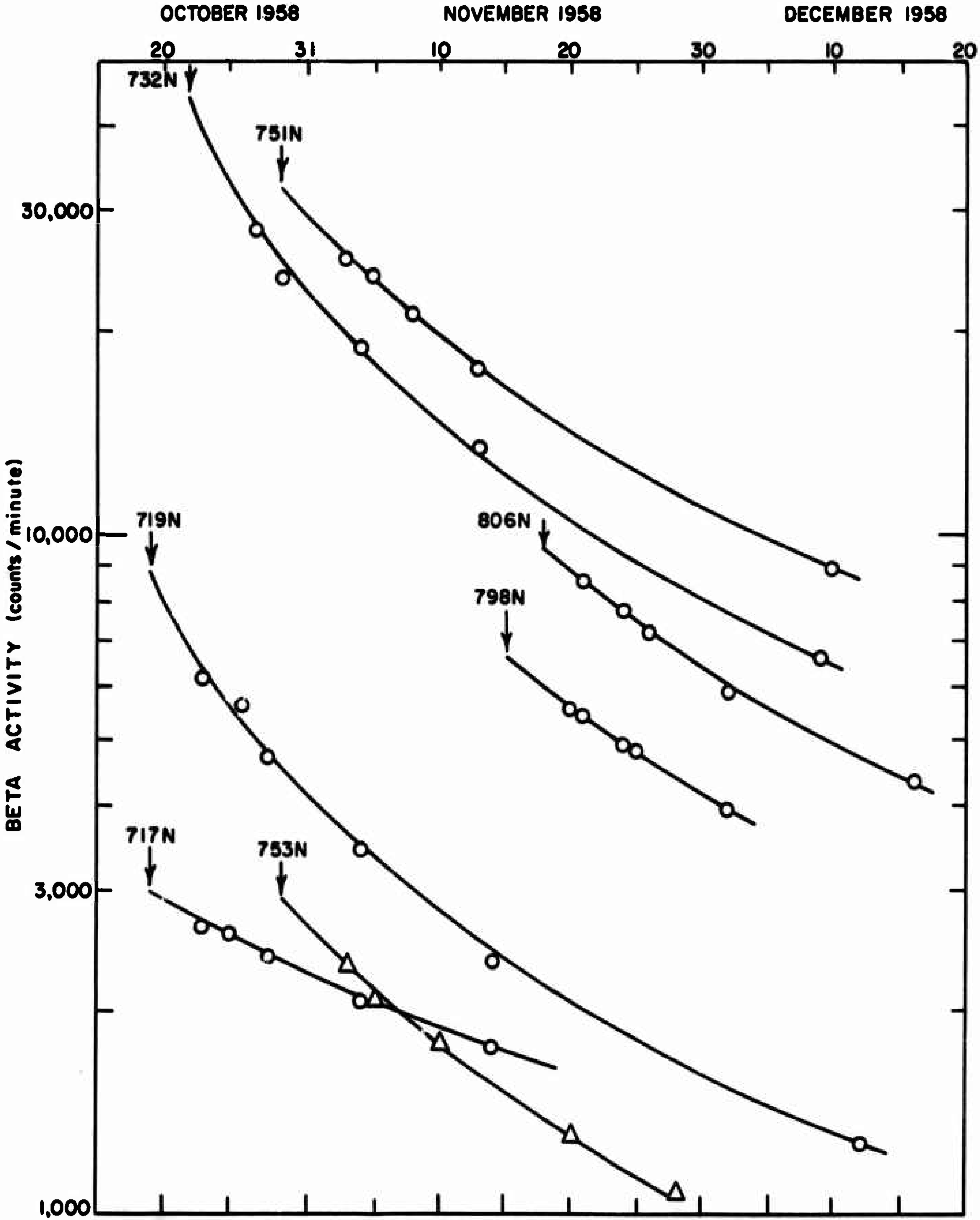


FIGURE 37. DECAY OF BETA ACTIVITY OF SOME SAMPLES CONTAINING DEBRIS FROM OCTOBER 1958 USSR WEAPON TESTS

intercepted appeared to have been produced during the first half of October, and no radioactivity from the late October tests, at least four of which had "large" yields[13], was detected. One possible explanation for this may be found in the configuration of the winter polar vortex circulation which was still developing in the northern hemisphere during October 1958. The low pressure center over the norther polar latitudes deepened considerably during the course of the month, and this in itself may have reduced the rate of eddy diffusion in the meridional direction by increasing the zonal component of the winds at the expense of the meridional component. It may also be significant that the main center of the polar low pressure system, which had been near the Queen Elizabeth Islands, north of Canada, during early October shifted across the pole and became situated near Siberia during the second half of the month[17]. It remained near Siberia until the end of 1958 at least. It would appear, therefore, that radioactivity from the late October 1958 USSR tests was injected closer to the center of the polar low pressure system than was radioactivity from the early October 1958 tests, and consequently it was carried through the HASP sampling corridor at higher latitudes than was radioactivity from the early October tests. It is likely, therefore, that the failure of the HASP sampling to intercept radioactivity from the late October tests is attributable to the general restriction of that sampling to relatively low latitudes during late 1958 and early 1959.

On several occasions radioactive debris from the early October USSR tests was intercepted as far south as 25°N, for example, on 29 October, 15 November and 23 December 1958. It is probably significant that these interceptions occurred at times when the circulation about the polar low pressure system extended farther south than usual along the meridian of HASP sample collections. In Figure 38 are shown the zonal wind structures[16] on 3 October 1958, when debris from the September 1958 U.K. tests was first intercepted, and on 19 October 1958 when debris from the USSR October 1958 tests was first intercepted. Total beta and tungsten-185 activities, the latter corrected to 15 August 1958, are given in pCi/SCM. Winds are given in knots, and the tropical tropopause is plotted as a heavy line. On 19 October at 18 km, USSR debris was intercepted north of 40°N, and U.K. debris was intercepted south of 30°N. The southern boundary of the northern hemisphere polar vortex was at about 30°N at this altitude along the 71°W meridian on this date[17]. In Figure 39 are shown the zonal wind structures on 22 October 1958 and 29 October 1958, when the highest measured concentrations of USSR debris were intercepted. Total beta and strontium-90 activities are given in pCi/SCM. It is noteworthy that on 22 October 1958, the sampling missions north of 44°N intercepted relatively little USSR debris, while on 29 October 1958, USSR debris was intercepted as far south at about 20°N at 18 km. The southern margin of the northern polar vortex at 18 km on 29 October 1958 was close to 25° or 20°N.

Murayama and Machta[18] found that data for balloon samples collected at San Angelo, Minneapolis and Sioux City during late 1958 and early 1959 indicated the movement of radioactive debris from the tropical stratosphere into the region between 31° and 45°N during the winter of 1958-1959, but that there was no evidence of significant movement into that region of USSR debris initially injected at higher latitudes. Martell[15], in discussing these observations, concludes that the "effective mixing surfaces in the polar

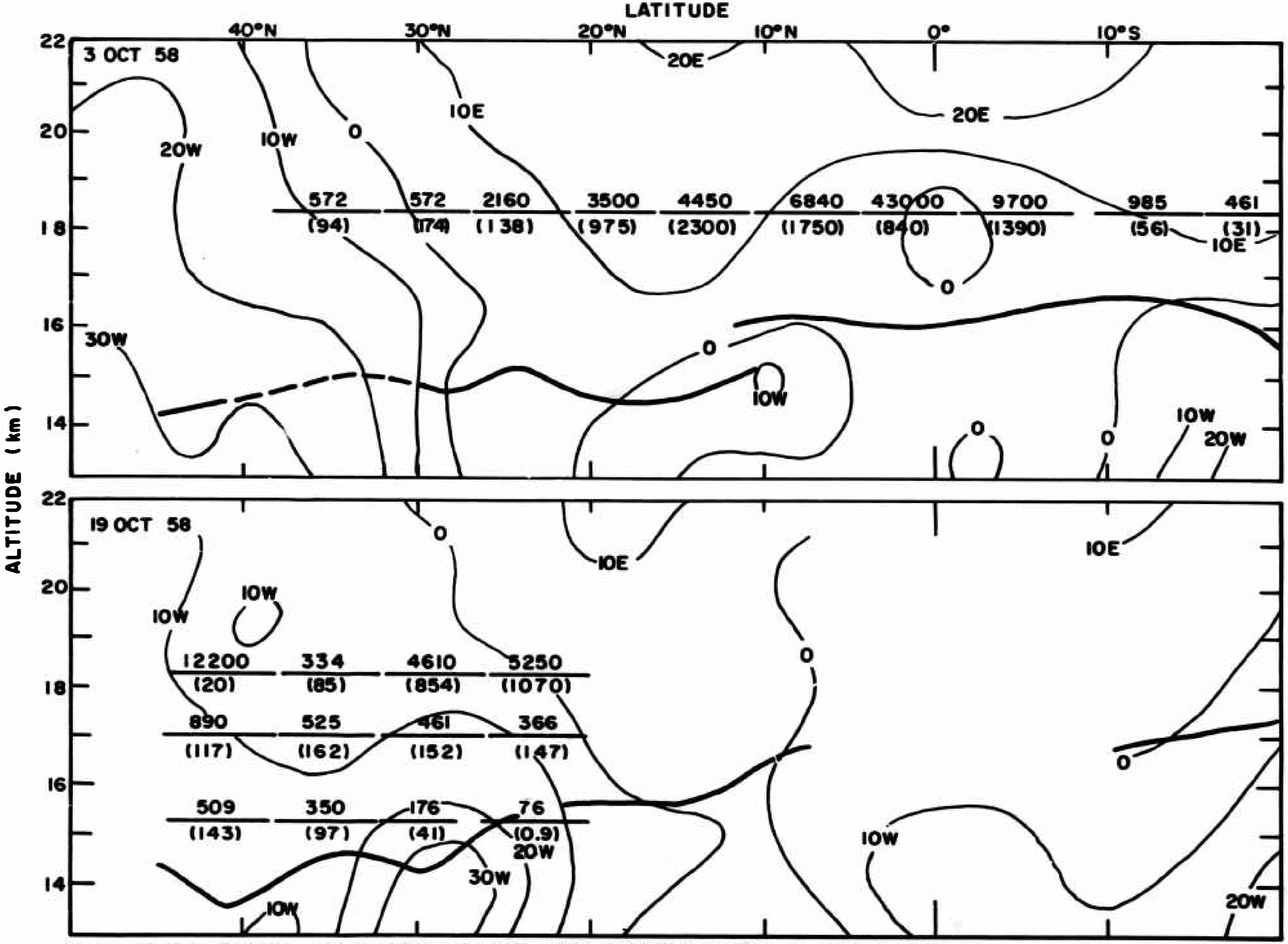


FIGURE 38. ZONAL WIND STRUCTURE AND INTERCEPTED β AND W-185 (in parentheses) ACTIVITIES ON 3 OCTOBER AND 19 OCTOBER 1958

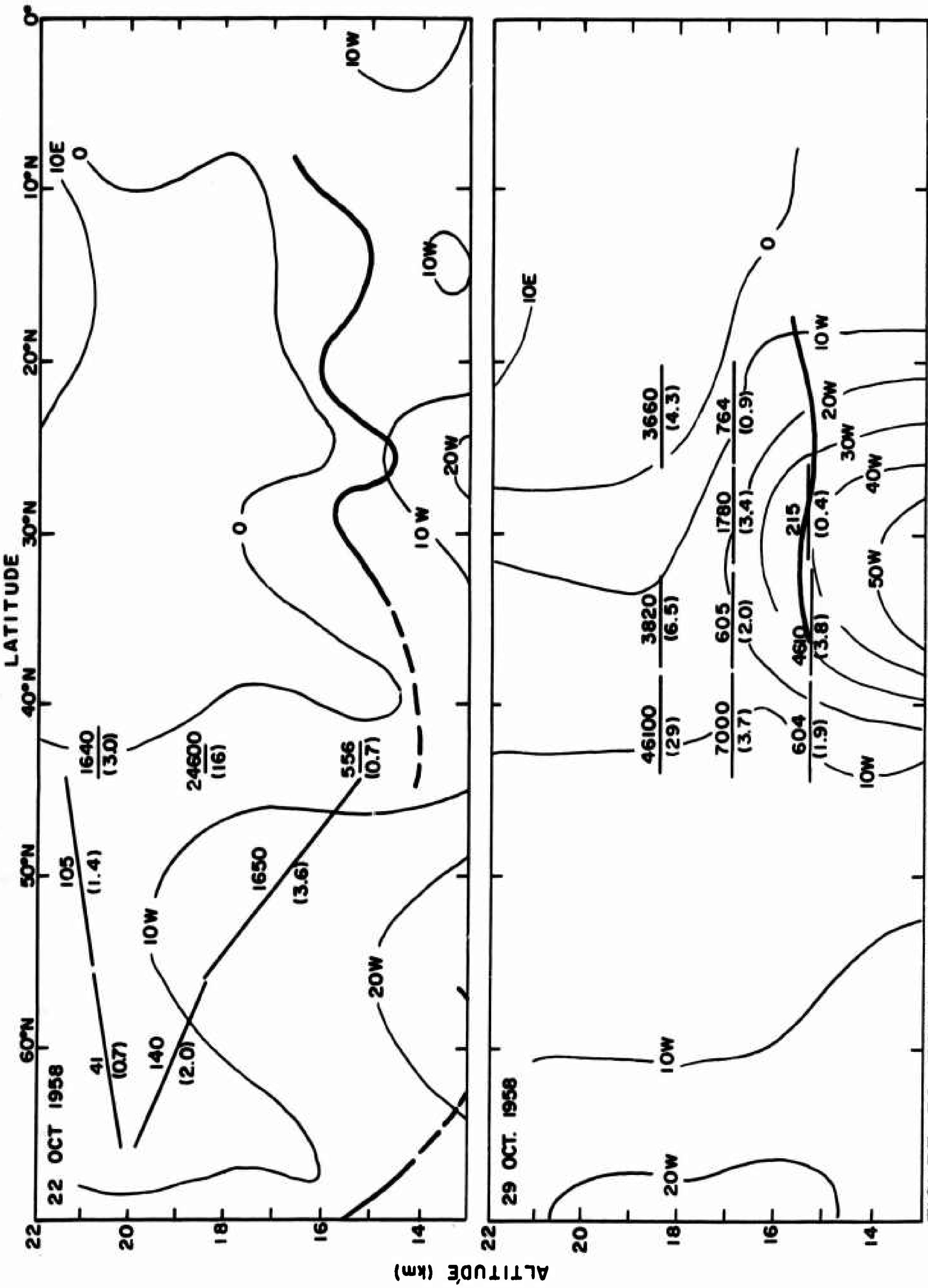


FIGURE 39. ZONAL WIND STRUCTURE AND INTERCEPTED β AND STRONTIUM-90 (in parentheses) ACTIVITIES ON 22 OCT. AND 29 OCT. 1958

stratosphere must be nearly vertical", and that debris from USSR tests "is very inappreciably mixed equatorwards at middle stratosphere altitude levels". Certainly the HASP measurements indicate that radioactive debris from the late October 1958 USSR tests did not mix equatorward appreciably in the lower stratosphere during late 1958. In Chapter 7 it will be noted also that debris from the late 1961 USSR tests did not mix equatorward appreciably either, until early 1962. It may be hypothesized that had the altitude of injection of the 1958 USSR debris been as high as that of the 1961 USSR debris, it too would have been detected at lower latitudes during the spring season following its injection, that is during the spring of 1959.

The HASP sample collections which were made at high northern latitudes during late 1958 suggested that radioactive debris from the October 1958 USSR weapons tests - at least from the early October tests - was injected mainly into the lower polar stratosphere below the 19 km level. Interceptions of USSR debris at levels above 18 km occurred far enough south - south of 45°N - for it to be assumed that the debris had risen to somewhat higher altitudes as it moved equatorward along sloping mixing surfaces[19, 20]. Even so, when a vertical profile was obtained at 44°N on 22 October 1958, relatively little USSR debris was found in the sample collected above the 19 km level. Radioactive debris sampled between 33° and 27°N at 18.3 km on 12 December 1958 appeared to have come from the USSR tests, but when debris was collected at 19.8 km in this latitude band seven days later, on 19 December 1958, it appeared to contain instead radioactivity from the September 1958 U.K. tests.

Vertical profiles of total beta and strontium-90 activities measured in the northern polar stratosphere during late 1958 are summarized in Table 18.

TABLE 18. Vertical Profiles of Radioactivity in the Northern Polar Stratosphere During Late 1958

Sample Number	Latitude	Altitude (km)	pCi β / 100 SCM	pCi Sr^{90} / 100 SCM	Sr^{89} / Sr^{90}	pCi W^{185} / 100 SCM
22 October 1958						
728N	66° - 56°N	20.4	4,140	73	1	≤ 210
727N	66° - 56°N	19.0	14,000	197	2	≤ 450
729N	56° - 44°N	21.0	10,500	140	-	660
726N	56° - 44°N	16.8	165,000	356	26	53,100
733N	44°N	20.7	164,000	297	19	829
732N	44°N	18.3	2,460,000	1,640	49	18,110
731N	44°N	15.2	55,600	67	-	9,440
730N	44°N	12.2	1,490	4	5	975
23 October 1958						
741N	66° - 56°N	20.4	6,200	92	-	920
740N	66° - 56°N	19.2	22,400	202	5	1,070
742N	56° - 44°N	20.9	42,900	121	16	138
739N	56° - 44°N	16.8	130,000	260	-	32,000
18 November 1958						
812N	55° - 44°N	20.6	19,200	178	8	369
809N	55° - 44°N	18.8	137,000	320	-	3,180
806N	55° - 49°N	15.2	795,000	741	44	19,800
805N	49° - 44°N	15.2	116,000	134	37	14,100
811N	66° - 56°N	19.8	9,370	73	11	328
810N	66° - 55°N	19.4	10,800	79	6	127
808N	66° - 60°N	15.2	382,000	415	44	-
807N	60° - 55°N	15.2	274,000	445	27	34,400
20 November 1958						
821N	55° - 50°N	19.8	24,900	165	11	364
820N	55° - 50°N	18.3	46,100	200	11	3,380
822N	50° - 45°N	19.8	46,100	288	--	985
819N	50° - 45°N	18.3	119,000	389	23	13,500

(W^{185} activities are corrected for radioactive decay to 15 August 1958.)

It is evident from these data that there were steep concentration gradients in the vertical direction in the polar stratosphere in October and November 1958. These gradients are seen in concentrations of Hardtack tungsten-185 as well as in debris from the USSR tests. It may be concluded that vertical mixing was not occurring rapidly throughout the polar region at that time. Localized regions of rapid vertical mixing may have existed, or course, but the data do not substantiate their existence within the air masses sampled.

6.6 Transport of Tungsten-185 from the 1958 U. S. Weapon Tests

Measurements of tungsten-185 during Project HASP provided evidence on the primary importance of eddy diffusion in the transfer within the lower stratosphere of radioactive debris from nuclear weapon tests[19]. The high activities of this radioactive tracer, which were injected into the lower tropical stratosphere during May to July 1958 by the U. S. Hardtack series of tests, were monitored by Project HASP sampling as this debris spread within the lower stratosphere during the second half of 1958, during 1959, and during the first half of 1960.

The distribution of tungsten-185 within the stratosphere, as evidenced by HASP measurements is shown for four intervals in Figures 40 and 41: for 14 - 29 October 1958 and 6 - 28 January 1959 in Figure 40, and for 21 July - 8 August 1959 and 5 - 15 January 1960 in Figure 41. The tungsten-185 concentrations, in pCi/SCM, are corrected for decay to 15 August 1958. A series of dots indicates the approximate locations of sample collections. It is evident that over the course of a year and a half following the injection of this tracer, the highest concentrations remained in the region of injection and

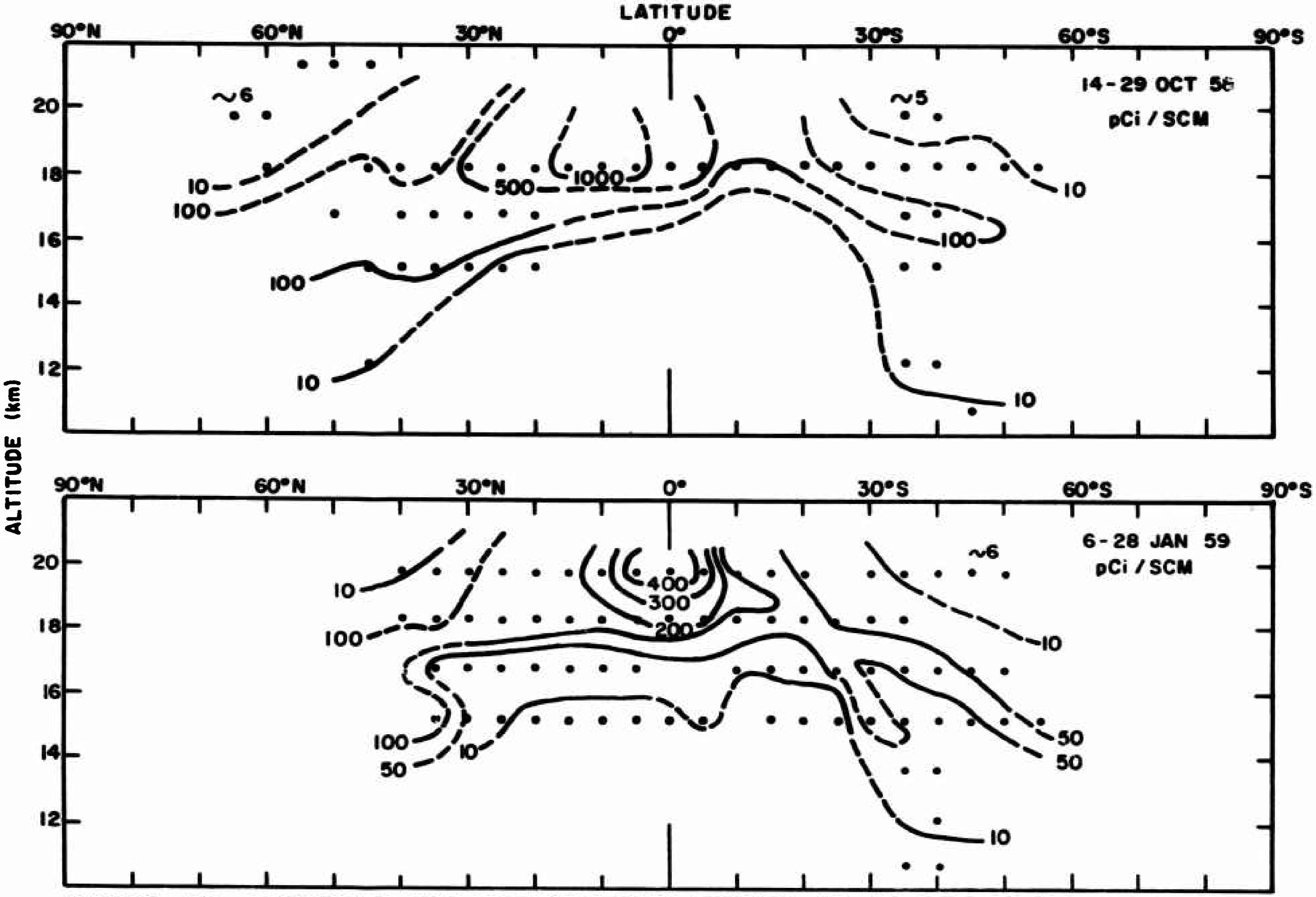


FIGURE 40. TUNGSTEN-185 DISTRIBUTIONS DURING 14-29 OCT 58 AND 6-28 JAN 59

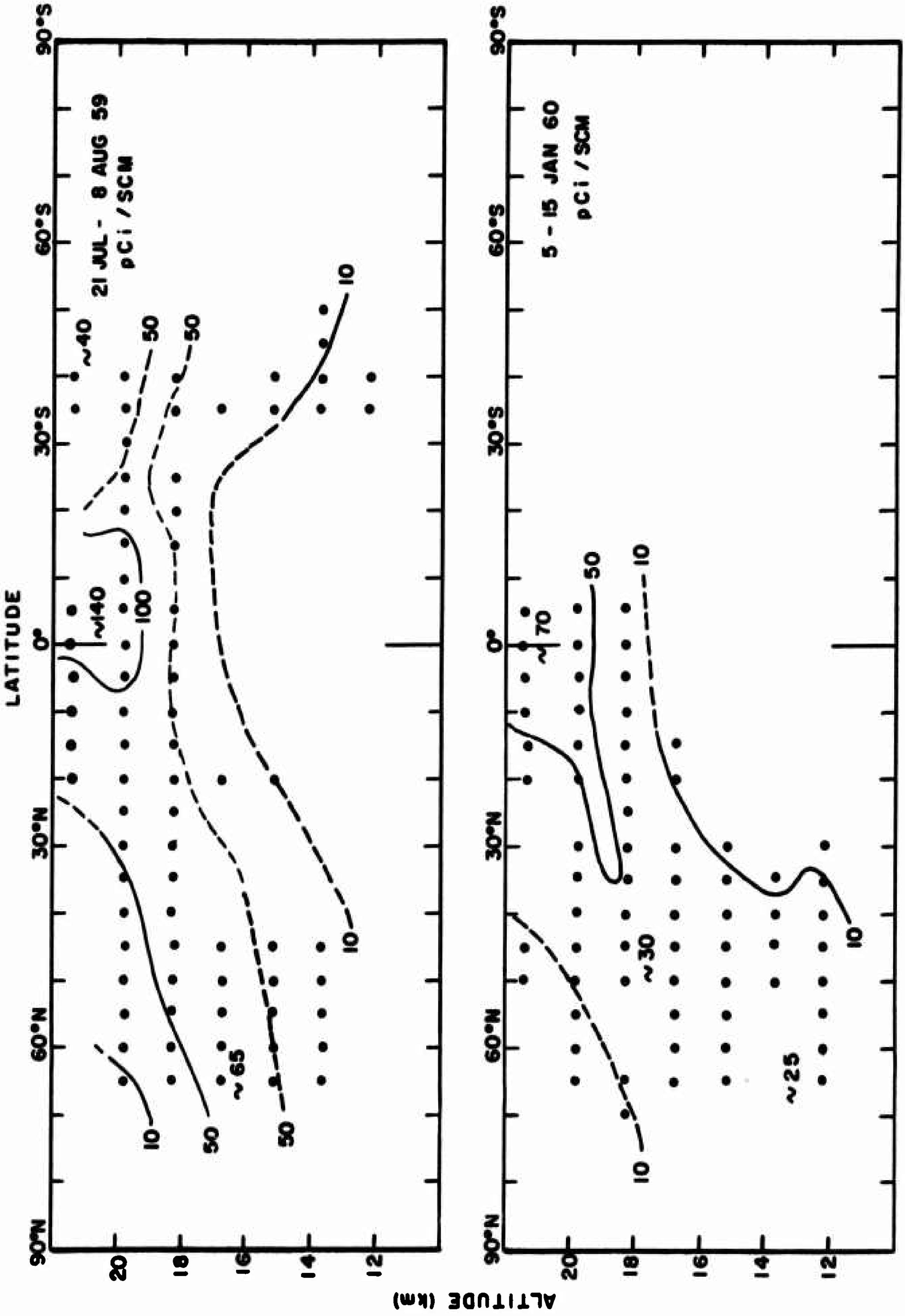


FIGURE 41. TUNGSTEN-185 DISTRIBUTIONS DURING 21 JUL - 8 AUG 1959 AND 5 - 15 JAN 1960

concentrations tended to decrease laterally away from that region, as would be expected if turbulent exchange were the transfer process operating a zone of high concentrations extended away from the tropical stratosphere, sloping downward from the equator toward both poles. Evidently the tungsten-185 sank to lower levels as it spread laterally, indicating that quasi-horizontal eddy diffusion within the lower stratosphere occurs within "mixing layers" which slope downward from the equator toward the poles. (In Chapter 7 evidence is offered that radioactive debris injected into the polar stratosphere rises to higher levels as it moves equatorward, within these same "mixing layers". In section 6.7, similar evidence is offered based on more limited measurements of radioactivity from 1958 USSR weapon tests.) Lateral mixing within the zone of higher concentrations was rapid enough to prevent destruction of the zone by vertical mixing in the polar stratosphere. The existence of this zone even in the polar regions confirms the primary importance of eddy diffusion rather than of an organized circulation even in this region. The poleward slope of the isolines, which is greater than the poleward slope of survaces of constant potential temperature, has been discussed by Newell[21], Hering[22] and others.

6.7 Transport of Radioactivity from the October 1958 USSR Weapon Tests

In section 6.5 it was noted that HASP sampling did not intercept fresh radioactivity from the USSR nuclear weapon tests which were performed during the second half of October 1958. It was also noted that measurements of the fresh radioactivity from the tests performed during the first half of October 1958 indicated that injection was mainly into the lower polar stratosphere below the 19 km level.

It is quite possible that the radioactivity from the USSR tests of late October 1958 also stabilized at relatively low levels in the polar stratosphere. Had the USSR debris stabilized in the region above 15 km in the polar stratosphere, considerable quantities of it should have spread into the tropical stratosphere by early 1959. There was no evidence of movement of significant quantities of radioactive debris from the USSR tests into the tropical stratosphere during late 1958 or early 1959, either in data from Project HASP or in data from the U. S. AEC balloon program[23]. Murayama and Machta[18], in discussing the balloon data for late 1958 and early 1959, made the simplifying assumption that all strontium-89 contained by those samples had been produced by the USSR tests, but in fact all of this strontium-89 could readily be attributed instead to the low latitude tests of May to September 1958. The Sr^{89}/Sr^{90} ratios in the balloon samples were generally similar to those measured in HASP samples collected at about the same time in the tropical stratosphere, and these samples contained no significant contribution from the USSR tests. The continuous monitoring of the tropical stratosphere during late 1958 and early 1959 which was performed during Project HASP would have revealed any significant influx of USSR debris into that region, both by an increase in the concentrations of radioactivity and by a decrease in the apparent age of the debris. Neither of these effects was observed, so it may be concluded that essentially none of the radioactivity samples south of 45° N during early 1959 was derived from the late 1958 USSR test series.

The only samples of radioactive debris collected by Project HASP during early 1959 which seemed to have been derived mainly from the 1958 USSR tests came from a sampling mission flown by an RB-52 aircraft in the lower polar stratosphere, between 60°N and 90°N at altitudes between 10.6 and 12.9 km on 14 April 1959. The estimated concentrations of total beta activity, strontium-90 and tungsten-185, together with the activity ratios Sr^{89}/Sr^{90}, Ce^{144}/Sr^{90}, and Zr^{95}/Sr^{90} are given in Table 19 for these samples and for samples collected in other stratospheric regions in April 1958.

The samples collected below 15 km in the lower polar stratosphere had higher Sr^{89}/Sr^{90}, Ce^{144}/Sr^{90} and Zr^{95}/Sr^{90} ratios (and higher ratios of total beta activity to strontium-90 activity) than those collected elsewhere in the stratosphere, indicating that they contained debris which was of more recent origin than that found elsewhere. The samples collected near 20 km in the equatorial stratosphere, and containing radioactive debris from the September 1958 U.K. tests, generally showed somewhat lower values of these ratios. Samples collected at altitudes above 15 km in the northern polar stratosphere had the lowest activity ratios, indicating that most of the debris in that region had come from tests which preceded the U.K. tests. Presumably this debris was mainly from the U. S. Hardtack test series. At about 15 km there appeared to be a minimum in the vertical profile of strontium-90 concentrations in the polar stratosphere, with largely USSR debris at lower levels and largely Hardtack debris at higher levels.

The fallout rates in the northern hemisphere during early 1959 were very high, mainly as a result of deposition of radioactivity from the 1958 USSR tests[24]. Evidently nearly all of the radioactivity injected into the stratosphere by these USSR tests had fallen out before the end of 1959[24, 25]. This observation is consistent with the finding of USSR debris in only the lowest layers of the polar stratosphere in April 1959 (Table 19).

TABLE 19. Radioactive Debris in Several Stratospheric Regions, April 1959

Latitude	Altitude (km)	$\frac{\text{pCi } \beta}{100 \text{ SCM}}$	$\frac{\text{pCi } Sr^{90}}{100 \text{ SCM}}$	$\frac{\text{pCi } W^{185}}{100 \text{ SCM}}$	$\frac{Sr^{89}}{Sr^{90}}$	$\frac{Ce^{144}}{Sr^{90}}$	$\frac{Zr^{95}}{Sr^{90}}$
10°N-10°S	19.8	64,200(5)	332(5)	24,000(5)	6.1(5)	14(2)	8(1)
66° -44°N	18.3	41,900(3)	384(3)	14,800(3)	5.1(3)	11(3)	-
66° -44°N	15.1	32,200(3)	160(3)	3,240(3)	3.9(3)	12(3)	12(2)
75° -70°N	19.6	32,700(2)	346(2)	1,110(2)	2.0(2)	11(2)	6(2)
75° -70°N	18.4	27,300(2)	330(2)	1,410(2)	1.9(2)	12(2)	6(2)
75° -70°N	15.2	17,500(2)	159(2)	5,780(2)	3.6(2)	14(2)	9(2)
85° -60°N	12.4	72,600(5)	316(5)	7,040(5)	8.9(5)	17(5)	14(5)
90° -60°N	11.2	94,600(9)	343(9)	6,740(9)	11 (8)	17(9)	20(9)

(Tungsten-185 concentrations are corrected for radioactive decay to 15 August 1958.)

(The number of samples used to calculate each average value is given in parentheses following the value.)

The data in Table 19 do not rule out the possibility that the USSR debris was initially injected at altitudes considerably higher than 12 km and that subsidence within the polar stratosphere during the winter of 1958-1959 or during the early spring of 1959 brought it down to the level where it was sampled in April 1959. It may be concluded, however, that the air mass containing the USSR debris had not undergone extensive exchange with the tropical stratosphere since the injection of the USSR debris into it, for such debris was not observed entering the tropical stratosphere. It would appear, then, that the high concentrations of tungsten-185 found with the USSR debris had entered that air mass before the injection of the USSR debris. Since tungsten-185 was essentially limited to the region of the polar stratosphere below 19 km, it seems reasonably safe to conclude at least that the USSR debris was also essentially limited to the region below 19 km.

6.8 Transport of Radioactivity from the High Yield 1958 U. S. Weapon Tests

According to Martell[15], about 1.05 megacuries of strontium-90 were produced by the U. S. 1958 Hardtack weapon tests, and about half of this was produced by weapons with total yields in the 8 to 10 megaton range. It is reasonable to expect that much of the 0.53 megacurie of strontium-90 from these high yield explosions was injected into the tropical stratosphere above 20 km, and that there was a subsequent transfer of this radioactivity into the lower stratosphere during the year which followed the completion of the U. S. tests. There is evidence that significant quantities of this radioactivity entered the northern polar stratosphere during the winter of 1958 - 1959, and the southern polar stratosphere during the winter of mid-1959.

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Although no HASP sampling missions were flown in the northern polar stratosphere during the winter of 1958-1959, this region was sampled in October and November 1958 and in April and May 1959. As a result, the changes in the distribution of radioactivity which occurred there during that season may be deduced from the available data. It is not possible, however, to determine whether these changes occurred during the early part of the winter or during the late winter and early spring.

The available data on the distribution of radioactivity in the northern polar stratosphere during the second half of 1958 and the first half of 1959 are summarized in Tables 20 and 21. In Table 20 the data are presented in the form of the vertical profiles of radioactivity at high latitudes during September, October and November 1958 and during April, May and June 1959. Results of measurements of the total beta, strontium-90 and tungsten-185 activities and of the activity ratio Sr^{89}/Sr^{90} are given. During September to November 1958, relatively low activities of all nuclides were found at 20 km, but high activities of total beta activity and strontium-90 were found at this level during the first half of 1959. The debris which reached the 20 km level, and lower levels also, during the months preceding April 1958 apparently included tungsten-185, for the activities of this nuclide found at that level in 1959 were higher than those found there in late 1958. This would indicate that the debris came from the Hardtack tests rather than from the 1958 USSR tests. The tungsten-185 activities found at 20 km during April and June 1959 were less than those found at the lower levels. This is consistent with the estimate by Martell[15] that the activity ratio W^{185}/Sr^{90} in debris from the high yield events was less than a tenth of that in the debris from the events with yields between 0.5 and 2 megatons. Since the Hardtack debris would have mixed with the residue of material from older tests when it was injected into the tropical stratosphere, and again when it was transferred to the lower polar stratosphere, the Sr^{89}/Sr^{90} ratios of about 60 (corrected for decay to 15 August 1959) found at 20 km during April and June 1959 are in reasonable agreement with the ratio of 100 expected by Martell[15] in Hardtack debris. Debris from the late 1958 USSR tests, such as was found at 11 and 12 km in April 1959, displayed a significantly higher Sr^{89}/Sr^{90} ratio. The fact that in June 1959 the Sr^{89}/Sr^{90} ratio generally increased with decreasing altitude in the polar stratosphere may be ascribed to the presence at the lower altitudes of lower concentrations of debris from the pre-Hardtack tests, and higher concentrations of debris from the late 1958 U.K. and USSR tests.

Table 21 contains meridional profiles of radioactivity at 20 km for September - October 1958 and November - December 1958 (for which few data are available), for January 1959 and late March 1959 (at which times sampling did not extend significantly into the polar stratosphere), and for April 1959 and June 1959. The data in this table confirm that the dominant source of radioactive debris at 20 km in the northern tropical stratosphere was Hardtack plus older debris during September - October 1958 and Hardtack plus late 1958 U.K. debris from November - December 1958 to June 1959. On the other hand, the dominant source of radioactive debris at 20 km in the northern polar stratosphere during late 1958 was pre-Hardtack debris, while by April 1959, it was a mixture of Hardtack plus older material.

TABLE 20. Vertical Profiles of Radioactivity in the Northern Polar Stratosphere

Altitude (km)	Late Sept. 1958	Late Oct. 1958	18 Nov. 1958	April 1959	20 May 1959	16 June 1959
Total Beta Activity (pCi/SCM):						
20	44(3)	52(2)	94(1)	327(2)	-	331(2)
19	-	182(2)	108(1)	-	-	-
18	139(2)	-	-	273(2)	-	306(4)
17	-	-	-	-	265(2)	249(2)
16	-	-	-	-	-	-
15	117(3)	-	3,280(2)	175(2)	-	227(4)
14	-	-	-	-	162(2)	-
13	-	-	-	-	-	-
12	-	-	-	766(5)	-	-
11	-	-	-	946(9)	-	-
Strontium-90 Activity (pCi/100 SCM):						
20	103(3)	83(2)	73(1)	346(2)	-	416(2)
19	-	200(2)	80(1)	-	-	-
18	258(2)	-	-	330(2)	-	373(4)
17	-	-	-	-	220(2)	277(2)
16	-	-	-	-	-	-
15	181(3)	-	430(2)	159(2)	-	188(4)
14	-	-	-	-	105(2)	-
13	-	-	-	-	-	-
12	-	-	-	316(5)	-	-
11	-	-	-	343(9)	-	-
Tungsten-185 Activity (pCi/SCM corrected for decay to 15 August 1958):						
20	1(2)	5(2)	3(1)	11(2)	-	28(2)
19	-	6(2)	1(1)	-	-	-
18	≤1(1)	-	-	14(2)	-	48(4)
17	-	-	-	-	57(2)	65(2)
16	-	-	-	-	-	-
15	19(3)	-	343(1)	58(2)	-	70(4)
14	-	-	-	-	46(2)	-
13	-	-	-	-	-	-
12	-	-	-	70(5)	-	-
11	-	-	-	67(9)	-	-

TABLE 20. (continued)

Altitude (km)	Late Sept. 1958	Late Oct. 1958	18 Nov. 1958	April 1959	20 May 1959	16 June 1959
Sr^{89}/Sr^{90} Activity Ratio (corrected for decay to 15 August 1958):						
20	2(2)	2(1)	39(1)	60(2)	-	61(2)
19	-	10(2)	22(1)	-	-	-
18	3(1)	-	-	57(2)	-	104(4)
17	-	-	-	-	128(2)	98(2)
16	-	-	-	-	-	-
15	12(2)	-	127(2)	108(1)	-	134(4)
14	-	-	-	-	154(2)	-
13	-	-	-	-	-	-
12	-	-	-	234(5)	-	-
11	-	-	-	290(8)	-	-

TABLE 21. Profiles of Radioactivity at 20km in the Northern Hemisphere

Latitude	Sept.-Oct. 1958	Nov.-Dec. 1958	Jan. 1959	Late Mar. 1959	April 1959	June 1959
Total Beta Activity (pCi/SCM):						
1°N	4,610(1)	2,180(1)	1,300(1)	453(2)	476(1)	318(1)
7°N	7,150(1)	2,500(1)	1,780(1)	572(2)	445(1)	355(3)
13°N	-	2,670(1)	1,450(1)	613(2)	589(1)	352(3)
19°N	-	1,370(1)	1,190(1)	549(2)	461(1)	318(1)
24°N	-	840(1)	870(2)	516(2)	434(3)	283(2)
30°N	-	2,460(1)	530(2)	596(2)	472(3)	323(2)
35°N	-	1,320(1)	460(2)	565(2)	434(3)	320(2)
41°N	-	-	210(1)	-	210(1)	350(1)
44°N	1,640(1)	-	-	-	-	332(1)
49°N	267(2)	461(1)	-	-	-	350(1)
54°N	-	250(1)	-	-	-	302(1)
60°N	52(2)	94(1)	-	-	-	382(1)
65°N	-	-	-	-	-	280(1)
72°N	44(3)	-	-	-	325(2)	-
Tungsten-185 Activity (pCi/SCM corrected for decay to 15 August 1958):						
1°N	678(1)	510(1)	460(1)	222(2)	195(1)	162(1)
7°N	1,480(1)	556(1)	470(1)	226(2)	212(1)	131(3)
13°N	-	416(1)	72(1)	148(2)	216(1)	118(3)
19°N	-	168(1)	182(1)	111(2)	96(1)	87(1)
24°N	-	88(1)	111(2)	85(2)	74(3)	87(1)
30°N	-	331(1)	95(2)	101(2)	81(3)	74(2)
35°N	-	166(1)	22(2)	41(2)	53(3)	65(2)
41°N	-	-	≤6(1)	-	58(1)	62(1)
44°N	8(1)	-	-	-	-	41(1)
49°N	4(2)	10(1)	-	-	-	20(1)
54°N	-	4(1)	-	-	-	48(1)
60°N	5(2)	3(1)	-	-	-	41(1)
65°N	-	-	-	-	-	14(1)
72°N	1(2)	-	-	-	11(2)	-

TABLE 21. (continued)

Latitude	Sept.-Oct. 1958	Nov.-Dec. 1958	Jan. 1959	Late Mar. 1959	April 1959	June 1959
Strontium-90 Activity (pCi/100 SCM):						
1°N	952(1)	710(1)	490(1)	232(2)	251(1)	221(1)
7°N	1,140(1)	1,100(1)	514(1)	302(2)	248(1)	361(3)
13°N	-	810(1)	497(1)	348(2)	289(1)	369(3)
19°N	-	566(1)	552(1)	362(2)	292(1)	256(1)
24°N	-	385(1)	453(2)	378(2)	291(3)	277(2)
30°N	-	816(1)	353(2)	300(2)	404(3)	313(2)
35°N	-	510(1)	296(2)	404(2)	358(3)	323(2)
41°N	-	-	221(1)	-	238(1)	376(1)
44°N	297(1)	-	-	-	-	389(1)
49°N	130(2)	288(1)	-	-	-	443(1)
54°N	-	165(1)	-	-	-	397(1)
60°N	83(2)	73(1)	-	-	-	455(1)
65°N	-	-	-	-	-	378(1)
72°N	103(3)	-	-	-	347(2)	-
Sr^{89}/Sr^{90} Activity Ratio (corrected for decay to 15 August 1958):						
1°N	79(1)	-	111(1)	118(2)	128(1)	-
7°N	89(1)	113(1)	111(1)	128(2)	172(1)	102(2)
13°N	-	124(1)	103(1)	123(2)	118(1)	96(2)
19°N	-	124(1)	-	120(1)	153(1)	108(1)
24°N	-	92(1)	107(2)	88(2)	127(3)	132(2)
30°N	-	140(1)	86(2)	108(2)	113(3)	116(2)
35°N	-	119(1)	111(2)	100(2)	107(3)	160(2)
41°N	-	-	213(1)	-	-	130(1)
44°N	47(1)	-	-	-	-	79(1)
49°N	40(2)	-	-	-	-	72(1)
54°N	-	39(1)	-	-	-	90(1)
60°N	2(2)	39(1)	-	-	-	60(1)
65°N	-	-	-	-	-	66(1)
72°N	2(2)	-	-	-	60(2)	-

The Hardtack test series was performed during mid-1958, the winter season of the southern hemisphere. If the movement of radioactive debris from the tropical stratosphere into the polar stratosphere is most rapid during the winter season, as is suggested by the observations in the northern hemisphere during 1958-1959, then it might be expected that significant amounts of radioactivity from the Hardtack tests should have entered the southern polar stratosphere during mid-1958. The HASP measurements indicate that some Hardtack debris had entered the southern polar stratosphere by September 1958, when HASP sampling of that region began. The concentrations of this debris in the southern hemisphere were much less than the concentrations at equivalent locations in the northern hemisphere, however, until mid-1959 at least. But in about June 1959, with the beginning of the 1959 winter season in the southern hemisphere, there was a renewed transfer of radioactivity into the southern polar stratosphere, and the results of HASP measurements suggest that the high yield Hardtack events were at least one source of this material.

Some of the HASP data on the distribution of radioactivity in the southern hemisphere during the second half of 1958 and the first half of 1959 are summarized in Tables 22, 23 and 24. Vertical profiles of total beta, strontium-90 and tungsten-185 activity and of the activity ratio Sr^{89}/Sr^{90} are given in Tables 22 and 23. Data for 54°S and 38°S for December 1958, and for February and March 1959 are given in Table 22 in order to illustrate the extent to which results for 38°S may be considered typical of the southern polar stratosphere. The tropopause gap region is often close to 38°S, so that concentrations found at levels below 18 km at that latitude may sometimes be lower than those found at higher latitudes, such as 54°S. At and above the 18 km level, however, results for 38°S are probably typical of those for the polar stratosphere. After March 1959 HASP sampling south of 38°S was infrequent, so that data for later periods, given in Table 23, are all from 38°S.

From Table 23, it may be seen that beginning in about June 1959 there was a significant increase in concentrations of total beta, strontium-90 and tungsten-185 activity at 20 km at 38°S. The presence of tungsten-185 in the radioactive debris which reached this location at this time, and the Sr^{89}/Sr^{90} activity at 65 to 75 (when corrected for decay to 15 August 1958), both suggest that this debris was derived from the high yield Hardtack events. The meridional profiles of radioactivity at 20 km summarized in Table 24 confirm this origin. They show that marked increases in activity occurred in the lower southern latitudes first: at 13°S in May 1959, and at 19°S, 25°S and 32°S in June 1959. At 38°S and 43°S there were significant increases in strontium-90 concentration in June 1959, but they were much less than the increases nearer the equator. At 49°S and 54°S the increases in June 1959 were even smaller. HASP sampling of the southern polar stratosphere was insufficient after June 1959 to determine whether the influx of Hardtack debris into that area continued during the months of July and August 1959, or to determine whether significant amounts of debris from the 1958 rocket shots entered the lower polar stratosphere at high latitudes at that time.

TABLE 22. Vertical Profiles of Radioactivity at 38°S and 54°S During Late 1958 and Early 1959

Altitude (km)	Mid-Dec. 1958		Early Feb. 1959		Early March 1959	
	54°S	38°S	54°S	38°S	54°S	38°S
Total Beta Activity (pCi/SCM):						
20	70(1)	108(1)	83(1)	70(1)	70(1)	73(1)
19	-	-	-	-	-	-
18	127(1)	106(1)	87(1)	83(1)	-	-
17	97(1)	269(1)	80(1)	99(1)	88(1)	60(1)
16	-	-	-	-	-	-
15	67(1)	111(1)	91(1)	41(1)	87(1)	≤7(1)
Strontium-90 Activity (pCi/100 SCM):						
20	68(1)	83(1)	78(1)	76(1)	87(1)	103(1)
19	-	-	-	-	-	-
18	168(1)	68(1)	97(1)	65(1)	-	-
17	95(1)	113(1)	67(1)	37(1)	91(1)	48(1)
16	-	-	-	-	-	-
15	52(1)	40(1)	75(1)	16(1)	62(1)	1(1)
Tungsten-185 Activity (pCi/SCM corrected for decay to 15 August 1958):						
20	2(1)	6(1)	≤3(1)	≤7(1)	13(1)	7(1)
19	-	-	-	-	-	-
18	2(1)	10(1)	8(1)	22(1)	-	-
17	10(1)	114(1)	19(1)	46(1)	28(1)	26(1)
16	-	-	-	-	-	-
15	12(1)	46(1)	35(1)	19(1)	41(1)	-
Sr^{89}/Sr^{90} Activity Ratio (corrected for decay to 15 August 1958):						
20	-	25(1)	19(1)	-	-	14(1)
19	-	-	-	-	-	-
18	6(1)	40(1)	44(1)	51(1)	-	-
17	34(1)	-	54(1)	92(1)	34(1)	56(1)
16	-	-	-	-	-	-
15	29(1)	85(1)	45(1)	82(1)	56(1)	-

TABLE 23. Vertical Profiles of Radioactivity at 38°S During 1959

Altitude (km)	Late Mar 1959	Late Apr 1959	Late May 1959	Mid-Jun 1959	Early Jun 1959	Late Jul 1959	Early Aug 1959
Total Beta Activity (pCi/SCM)							
21	-	-	-	106(1)	-	152(1)	275(1)
20	86(1)	71(1)	88(1)	102(1)	102(2)	119(1)	126(1)
19	-	-	-	-	-	92(1)	-
18	-	79(1)	87(1)	-	76(1)	72(1)	87(1)
17	49(1)	-	-	17(1)	52(1)	-	-
16	-	-	10(1)	-	-	41(1)	-
15	18(1)	37(1)	45(1)	10(1)	29(2)	13(1)	28(1)
14	-	-	30(1)	-	-	-	-
13	-	-	-	-	-	≤ 7(1)	-
12	29(1)	10(1)	25(1)	-	8(1)	-	20(1)
Strontium-90 Activity (pCi/100 SCM)							
21	-	-	-	143(1)	-	194(1)	294(1)
20	118(1)	99(1)	102(1)	133(1)	136(2)	186(1)	188(1)
19	-	-	-	-	-	131(1)	-
18	-	113(1)	106(1)	-	90(1)	103(1)	120(1)
17	38(1)	-	-	19(1)	68(1)	-	-
16	-	-	13(1)	-	-	80(1)	-
15	13(1)	29(1)	75(1)	11(1)	40(2)	19(1)	43(1)
14	-	-	40(1)	-	-	-	-
13	-	-	-	-	-	6(1)	-
12	17(1)	6(1)	36(1)	-	14(1)	-	29(1)
Tungsten-185 Activity (pCi/SCM corrected for decay to 15 August 1958)							
21	-	-	-	116(1)	-	15(1)	38(1)
20	9(1)	4(1)	7(1)	-	30(2)	35(1)	51(1)
19	-	-	-	-	-	20(1)	-
18	-	24(1)	28(1)	-	39(1)	32(1)	54(1)
17	34(1)	-	-	13(1)	27(1)	-	-
16	-	-	-	-	-	25(1)	-
15	6(1)	22(1)	25(1)	5(1)	18(2)	9(1)	38(1)
14	-	-	17(1)	-	-	-	-
13	-	-	-	-	-	4(1)	-
12	16(1)	5(1)	15(1)	-	6(1)	-	11(1)

TABLE 23. (continued)

Altitude (km)	Late Mar 1959	Late Apr 1959	Late May 1959	Mid-Jun 1959	Early Jun 1959	Late Jul 1959	Early Aug 1959
Sr^{89}/Sr^{90} Activity Ratio (corrected for decay to 15 August 1958)							
21	-	-	-	-	-	65(1)	73(1)
20	-	21(1)	65(1)	-	-	76(1)	-
19	-	-	-	-	-	142(1)	-
18	-	123(1)	32(1)	-	-	-	-
17	91(1)	-	-	-	-	-	-
16	-	-	74(1)	-	-	-	-
15	-	38(1)	39(1)	-	138(1)	-	-
14	-	-	69(1)	-	-	-	-
13	-	-	-	-	-	-	-
12	79(1)	96(1)	-	-	-	-	-

TABLE 24. Profiles of Radioactivity at 20 km in the Southern Hemisphere

Latitude	Mid-Dec 1958	Mid-Feb 1959	Late Mar 1959	Early May 1959	Late Jun 1959	Early Jul 1959	Early Aug 1959
Total Beta Activity (pCi/SCM)							
1°N	2180(1)	652(1)	430(1)	366(1)	445(1)	258(1)	283(2)
5°S	1320(1)	652(1)	366(1)	316(1)	315(1)	186(1)	268(2)
13°S	191(1)	219(1)	-	318(1)	262(1)	197(1)	235(1)
19°S	135(1)	154(1)	134(1)	127(1)	214(1)	95(1)	132(1)
25°S	153(1)	102(1)	76(1)	119(1)	204(1)	121(1)	124(1)
32°S	154(1)	76(1)	76(1)	103(1)	178(1)	99(1)	113(1)
38°S	108(1)	70(1)	86(1)	80(2)	80(1)	101(1)	200(1)
43°S	89(1)	78(1)	79(1)	49(1)	107(1)	-	-
49°S	83(1)	76(1)	83(1)	60(1)	89(1)	-	-
54°S	70(1)	83(1)	-	45(1)	70(1)	-	-
Strontium-90 Activity (pCi/100 SCM)							
1°N	710(1)	264(1)	253(1)	278(1)	474(1)	263(1)	323(2)
5°S	521(1)	307(1)	251(1)	245(1)	352(1)	235(1)	326(2)
13°S	108(1)	145(1)	-	245(1)	294(1)	216(1)	286(1)
19°S	73(1)	129(1)	106(1)	116(1)	278(1)	151(1)	175(1)
25°S	97(1)	97(1)	105(1)	119(1)	232(1)	176(1)	169(1)
32°S	89(1)	89(1)	95(1)	116(1)	240(1)	134(1)	141(1)
38°S	83(1)	76(1)	118(1)	102(2)	129(1)	132(1)	242(1)
43°S	79(1)	86(1)	95(1)	91(1)	150(1)	-	-
49°S	78(1)	100(1)	106(1)	81(1)	118(1)	-	-
54°S	68(1)	78(1)	-	102(1)	113(1)	-	-
Tungsten-185 Activity (pCi/SCM corrected for decay to 15 August 1958)							
1°N	510(1)	302(1)	223(1)	204(1)	142(1)	149(1)	115(2)
5°S	305(1)	300(1)	187(1)	158(1)	174(1)	161(1)	140(2)
13°S	30(1)	79(1)	-	187(1)	141(1)	150(1)	141(1)
19°S	16(1)	57(1)	54(1)	66(1)	140(1)	38(1)	86(1)
25°S	16(1)	28(1)	12(1)	39(1)	107(1)	34(1)	47(1)
32°S	22(1)	12(1)	17(1)	32(1)	28(1)	16(1)	44(1)
38°S	6(1)	≤ 7(1)	9(1)	7(2)	-	24(1)	44(1)
43°S	≤ 2(1)	≤ 2(1)	8(1)	≤ 3(1)	9(1)	-	-
49°S	≤ 1(1)	4(1)	4(1)	≤ 1(1)	-	-	-
54°S	2(1)	≤ 3(1)	-	4(1)	4(1)	-	-

TABLE 24. (continued)

Latitude	Mid-Dec 1958	Mid-Feb 1959	Late Mar 1959	Early May 1959	Late Jun 1959	Early Jul 1959	Early Aug 1959
Sr^{89}/Sr^{90} Activity Ratio (corrected for decay to 15 August 1958)							
1°N	-	137(1)	115(1)	94(1)	86(1)	130(1)	148(2)
5°S	128(1)	137(1)	94(1)	87(1)	86(1)	113(1)	124(2)
13°S	54(1)	66(1)	-	122(1)	-	81(1)	-
19°S	59(1)	76(1)	71(1)	98(1)	-	-	-
25°S	26(1)	46(1)	70(1)	70(1)	-	-	54(1)
32°S	44(1)	39(1)	41(1)	59(1)	-	-	-
38°S	25(1)	-	-	36(1)	-	-	79(1)
43°S	13(1)	-	16(1)	-	-	-	-
49°S	11(1)	-	-	-	-	-	-
54°S	-	19(1)	-	19(1)	-	-	-

In summary, the HASP measurements of total beta activity and of strontium-90 activity in samples collected during 1958 and 1959 show that there was an an influx of radioactive debris into the northern polar stratosphere during the winter of 1958 - 1959 and into the southern polar stratosphere during the winter of mid-1959. The tungsten-185 activity and Sr^{89}/Sr^{90} activity ratios in this debris indicate that it was produced by the high yield events during the 1958 Hardtack test series.

6.9 Transport of Radioactivity from the 1958 U. S. Rocket Shots

Two events during the 1958 U. S. test series consisted of high altitude explosions of rocket-borne devices. These were the Teak event of 1 August 1958: a megaton range device detonated at 77 km over Johnston Island (17°N, 169°W); and the Orange event of 12 August 1958: a megaton range device detonated at 43 km over Johnston Island[13].

To provide a special tracer for these high altitude injections, about 3 megacuries of rhodium-102 were produced by the Orange event[26]. A few other events in the 1958 test series also produced small amounts of rhodium-102, and about 0.3 megacurie was injected into the stratosphere by the low altitude explosions. Apparently Teak and Orange together injected about 0.4 megacurie of strontium-90 into the upper atmosphere[27], for Glasstone[28] has estimated that the mesosphere had received this much strontium-90 from weapon tests performed before the end of 1958.

Measurements by Kalkstein[26, 29, 30] of rhodium-102 in filter samples of stratospheric and tropospheric air have shown that by June 1959 detectable quantities of debris from the Orange event were present at about 27 km at 32°N.

By October 1959 it was detectable in samples collected at about 20 km at high northern latitudes. By mid-1960 it was detectable in samples collected in the southern polar stratosphere. Samples collected at about 5.5 km at 52°N showed that the first major release of rhodium-102 from the stratosphere to the troposphere in the northern hemisphere occurred in about January 1961. Thus, more than a year was required for the debris from the Orange event to reach the lower stratosphere in significant amounts, and an additional year was required for it to begin entering the troposphere.

Measurements of rhodium-102 in stratospheric samples were also performed during Projects HASP and STARDUST. Although the results generally were quite similar to those reported by Kalkstein, they were often less consistent internally. Although agreement was achieved on intercliabration samples by the Isotopes and AFCRL laboratories, the 1961 STARDUST samples which were analyzed for rhodium-102 gave results which were only about 60 percent as high as those reported by Kalkstein for comparable samples.

Most of the STARDUST samples collected during 1962 and early 1963 gave results about 20 percent higher than those obtained for 1961 samples. This apparent confirmation of Kalkstein's report that some 1962 U. S. tests had produced Rh^{102}, made further monitoring of rhodium-102 concentrations appear fruitless.

Kalkstein's measurements were mostly limited to samples collected at about 20 km, with the result that the distribution of rhodium-102 within the lower stratosphere during 1961 cannot be obtained from his results alone. This distribution which has been estimated from STARDUST rhodium-102 measurements is shown in Figure 42 for early 1960, late 1961 and early 1962.

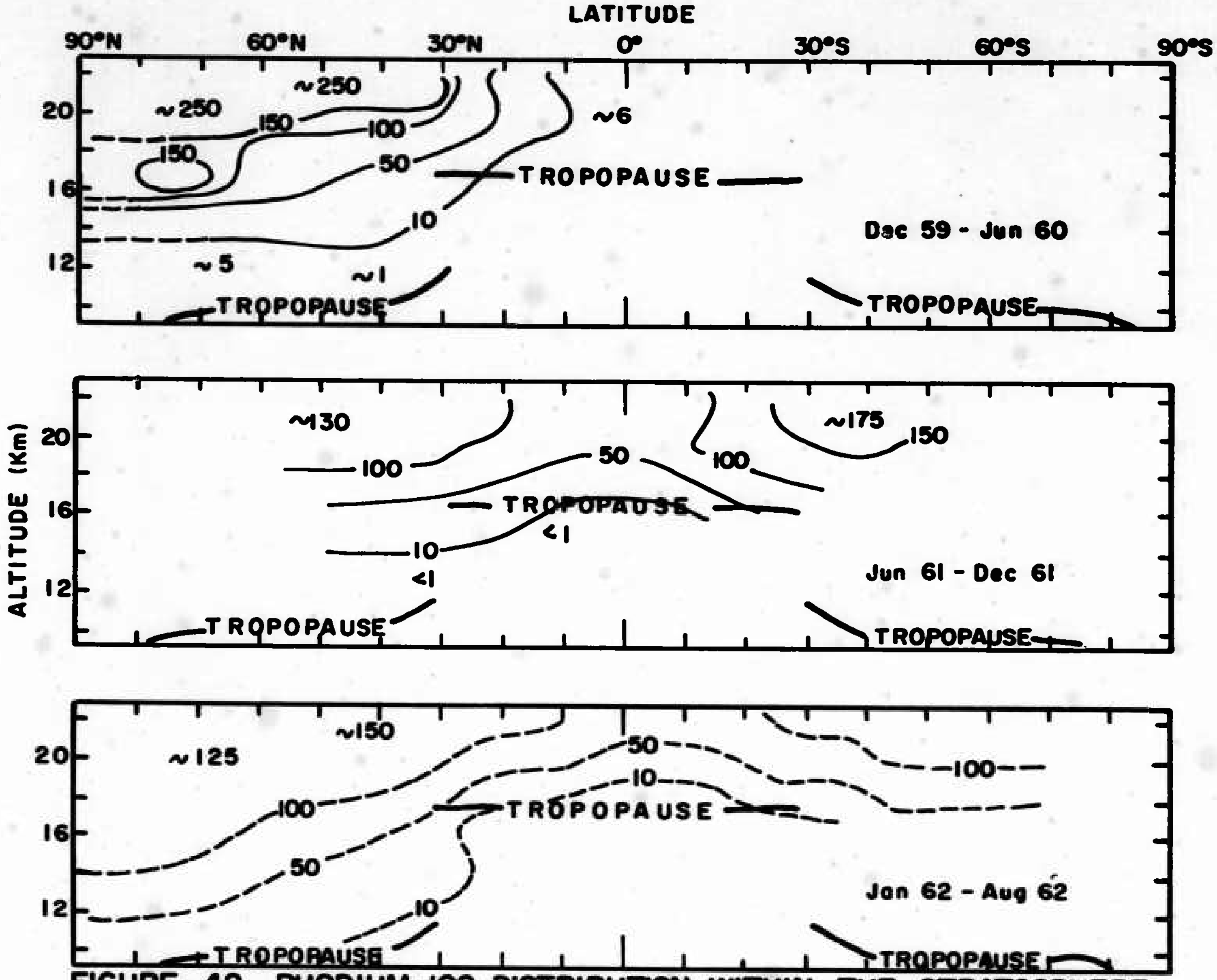

FIGURE 42. RHODIUM-102 DISTRIBUTION WITHIN THE STRATOSPHERE
(pCi/10^2 SCM, corrected 12 Aug 1958)

Kalkstein[26] estimated that the rhodium-102 which was present in the lower stratosphere during 1960 and 1961 represented about ten percent of the total injection, and therefore concluded that the "residence time" in the atmosphere of debris from the Orange event was about ten years, or at least five to twenty years. On the other hand, it was estimated in the Second Quarterly Report on Project STARDUST, DASA-1302, that approximately 40 percent of the debris from the Teak and Orange events had reached the lower stratosphere by early 1960. This would suggest that the mean "residence time" of the Teak and Orange debris in the atmosphere was probably quite a bit shorter than ten years.

The STARDUST estimates were based on measurements of strontium-90 and of the activity ratio Ce^{144}/Sr^{90} in filter samples collected during 1958 to 1960. In Table 25 are summarized bimonthly mean values of the strontium concentrations in various stratospheric regions. The Ce^{144}/Sr^{90} ratios in these same regions are given in Table 26.

In the lower tropical stratosphere, there was a gradual decrease in the strontium-90 concentration during 1958 to 1960, but the Ce^{144}/Sr^{90} activity ratio varied but little from the 23.6 mean value for this period. Evidently, this region of the stratosphere represented a well-mixed reservoir of debris from the 1958 low altitude tests performed by the U. S. and U.K. at low latitudes and from earlier weapon tests.

In the lower polar stratosphere of the northern hemisphere, strontium-90 concentrations generally decreased during the second half of 1959. A significant increase in concentrations was evident in the 19-21 km layer by January 1960, however, and in the 18 km layer by May - June 1960. Evidently, debris was

TABLE 25. Strontium-90 Concentrations in Stratospheric Regions

Interval	90°-30°N				30°N-30°S	30°-60°S			
	15km	17km	18km	19-21km	18-21km	15km	17km	18km	19-21km
Sept.-Oct. 1958	196(8)	284(6)	649(13)	143(10)	683(66)	92(2)	97(1)	122(33)	229(3)
Nov. -Dec. 1958	250(8)	383(3)	342(12)	213(6)	354(69)	64(1)	110(10)	113(34)	123(8)
Jan. -Feb. 1959	336(1)	192(1)	354(3)	320(7)	271(62)	50(22)	55(17)	108(10)	101(16)
Mar. -Apr. 1959	159(5)	216(4)	313(13)	354(10)	238(98)	27(13)	68(15)	92(6)	98(17)
May -June 1959	181(7)	230(13)	279(25)	352(25)	244(120)	42(15)	55(7)	100(7)	115(27)
July -Aug. 1959	159(6)	251(8)	270(28)	328(42)	268(185)	46(8)	96(6)	105(9)	177(15)
Sept.-Oct. 1959	104(40)	238(44)	304(61)	309(95)	246(88)	-	-	-	-
Nov. -Dec. 1959	108(50)	201(54)	264(51)	308(87)	222(101)	-	-	-	-
Jan. -Feb. 1960	124(44)	189(44)	276(53)	350(77)	157(117)	-	-	-	-
Mar. -Apr. 1960	108(41)	168(39)	257(36)	314(63)	159(85)	-	-	-	-
May -June 1960	149(15)	172(3)	322(14)	330(15)	156(72)	48(9)	-	127(11)	258(12)
Mean Jan.-June 1960	127	176	285	332	157	-	-	-	-

TABLE 26. Ce^{144}/Sr^{90} Ratios (corrected to 12 August 1958) in Stratospheric Regions

	90° - 30° N				30°N-30°S	30° - 60° S			
Interval	9-16 km	17 km	18 km	19-21 km	18-21 km	9-16 km	17 km	18 km	19-21 km
Sep-Oct 1958	21.7(3)	35.2(3)	16.1(2)	20.0(2)	24.1(11)	13.8(2)		13.9(5)	14.6(2)
Nov-Dec 1958	38.0(2)	32.3(1)	-	32.2(2)	25.8(6)	15.5(3)	19.8(5)	13.5(1)	25.9(3)
Jan-Feb 1959	29.7(1)	-	18.7(1)	22.5(1)	19.2(3)	33.0(3)	-	-	23.7(5)
Mar-Apr 1959	28.6(19)	20.7(1)	19.9(4)	22.6(4)	24.6(16)	29.0(3)	20.0(3)	-	17.0(5)
May-Jun 1959	28.7(3)	24.0(3)	22.5(3)	20.0(1)	21.4(22)	18.8(5)	-	18.7(2)	18.6(8)
Jul-Aug 1959	27.4(12)	21.1(3)	22.1(3)	22.6(3)	23.9(25)	19.9(8)	16.7(1)	27.5(3)	21.6(8)
Sep-Oct 1959	25.2(9)	26.7(2)	22.8(3)	27.7(3)	22.4(13)	-	-	-	-
Nov-Dec 1959	24.6(21)	23.1(12)	24.2(11)	26.8(11)	24.2(17)	-	-	-	-
Jan-Feb 1960	24.0(55)	24.4(20)	30.9(28)	33.9(36)	23.8(48)	-	-	-	-
Mar-Apr 1960	23.6(56)	26.9(27)	31.8(27)	33.6(50)	25.2(65)	-	-	-	-
May-Jun 1960	27.0(22)	32.2(3)	34.2(9)	33.1(17)	22.1(55)	18.9(6)	-	23.4(11)	24.6(10)
Mean Jan-Jun 1960	24.9	27.8	32.3	33.5	-	-	-	-	-
Mean Sep 1958-Jun 1960	-	-	-	-	23.6(281)	-	-	-	-

entering the lower polar stratosphere from some other region. The influx of this new debris was reflected also by increasing values of the Ce^{144}/Sr^{90} ratios. This would suggest that the new debris was either younger or less fractionated than that which was already present. Since the appearance of this new debris coincided with the appearance of rhodium-102 in the lower stratosphere, it may be concluded that the new debris was derived, in part at least, from the Teak and Orange event.

In DASA-1302 it was assumed, based on fission yields published by Katcoff[31], that the initial value of the ratio Ce^{144}/Sr^{90} in the debris from the rocket shot was 39. A ratio of 23.1, calculated from results for samples collected in the tropical stratosphere, and corrected for decay to 12 August 1958, was assumed to be the ratio in the debris present in the polar stratosphere before the influx of the debris from the rocket shots. The relative amounts of rocket-shot debris and of other debris were then calculated for each stratospheric region from the observed Ce^{144}/Sr^{90} ratios. These results were then used to divide the strontium-90 into that attributable to the rocket shots and that attributable to other sources, and the stratospheric burdens of each were calculated. Concentrations of the rocket-shot strontium-90 were assumed in the upper stratosphere which would give a total stratospheric burden of 0.4 MCi of strontium-90 from this source, to agree with the burden estimate made by Stebbins[27]. The resulting burden calculations indicated that 0.12 MCi of this strontium-90 was in the lower stratosphere (below the 40 mb level), and 0.16 MCi in the upper stratosphere of the northern hemisphere.

In the lower polar stratosphere of the southern hemisphere, at the 19 - 21 km level, the strontium-90 concentrations and Ce^{144}/Sr^{90} activity ratios increased during mid-1960 as a result of influx of debris from the high yield low altitude shots of the 1958 U. S. test series (see Section 6.8). By mid-1960, both the strontium-90 concentrations and Ce^{144}/Sr^{90} ratios had increased further in this region. Between mid-1959 and mid-1960, the rhodium-102 concentrations had also risen sharply in this region[26], indicating that there had been an influx of debris from the rocket shots.

It was assumed in DASA-1302 that the Ce^{144}/Sr^{90} ratios in the rocket-shot debris and the other debris, corrected to mid-August 1958, were 39 and 18.6, respectively. Using these assumptions, the strontium-90 was assigned to the rocket-shot debris or to "other sources", and the burdens of rocket-shot strontium-90 during early 1960 were calculated to be 0.04 MCi in the lower stratosphere and 0.08 MCi in the upper stratosphere of the southern hemisphere. No explanation was offered as to why Kalkstein[26] found essentially equal concentrations of rhodium-102 in the two hemispheres in mid-1960 while the calculations based on the Ce^{144}/Sr^{90} ratios indicated that the burden of rocket-shot strontium-90 in the lower stratosphere of the northern hemisphere was three times that in the equivalent region of the southern hemisphere.

One possible explanation of this disagreement would be that the debris from the Orange event was more or less evenly divided between the northern and southern hemispheres, while virtually all of the debris from the Teak event entered the northern hemisphere. While no reason for such behavior by the Teak debris is apparent, it cannot be ruled out on the basis of the measurements made on the STARDUST samples.

A second possible explanation of the disagreement would be that unfractionated debris from some other nuclear event was also injected into the upper stratosphere or mesosphere during 1958, perhaps during the late 1958 USSR test series. The only evidence against this hypothesis available in the STARDUST data is the lack of detection of such debris during 1959, when debris injected into the upper stratosphere by low altitude shots was reentering the lower stratosphere.

In attempting to deduce the source of the fission products which entered the lower stratosphere in the northern hemisphere in early 1960, and in the sourthern hemisphere before mid-1961, it is instructive to calculate the activity ratio Rh^{102}/Sr^{90} in the debris. This has been done for a series of intervals between early 1960 and late 1961 for which data are available. The rohdium-102 results published by Kalkstein[26] have been used, together with strontium-90 and cerium-144 results obtained during Project HASP, Project STARDUST and during other programs[32, 33, 34]. The strontium-90 concentrations to be attributed to the "rocket-shot debris" were calculated from the Ce^{144}/Sr^{90} activity ratios, as had been done in preparing DASA-1302, but using somewhat different initial values for the initial ratio in the debris from the rocket shots and in the other debris. The initial Ce^{144}/Sr^{90} ratio of 47.6, given by Harley et al[35], was used for the debris from the Teak and Orange events, since this value is presumably the most applicable to debris from megaton yield nuclear weapons. For the debris from other sources, the ratio 23.6 (derived from Table 26) was used for the northern hemisphere, and 18.6 (the same value used in DASA-1302) for the southern hemisphere. The results of the calculations are shown in Table 27.

TABLE 27. Rh^{102}/Sr^{90} in "Rocket-Shot Debris" (Corrected to 12 August 1958)

A. In The Northern Hemisphere

	% of Sr^{90} from Rocket-Shots	Total Sr^{90} (pCi/10^2SCM)	Rocket-Shot Sr^{90} (pCi/10^2SCM)	Rh^{102} (Kalkstein[26]) (pCi/10^2SCM)	$\frac{Rh^{102}}{Sr^{90}}$
Jan-Jun 1960 (HASP)	41%	332	137	225	1.6
May-Jun 1960 (USWB)	53%	283	150	225	1.5
Nov 1960 (USWB)	48%	245	116	225	1.9
May-Jun 1961 (USWB)	40%	184	73	225	3.1
Jun-Sep 1961 (STARDUST)	48%	181	86	225	2.6

B. In The Southern Hemisphere

	% of Sr^{90} from Rocket-Shots	Total Sr^{90} (pCi/10^2SCM)	Rocket-Shot Sr^{90} (pCi/10^2SCM)	Rh^{102} (Kalkstein[26]) (pCi/10^2SCM)	$\frac{Rh^{102}}{Sr^{90}}$
Jun 1960 (HASP)	21%	258	53	225	4.2
May-Jun 1960 (USWB)	20%	328	65	225	3.5
Nov 1960 (USWB)	28%	248	70	225	3.2
May-Jun 1961 (USWB)	37%	261	96	360	3.8
Nov 1961 (USWB)	40%	180	73	225	3.1
Oct-Nov 1961 (STARDUST)	55%	211	117	225	1.9

If the rhodium-102 yield of Orange was 3 MCi, as reported by Kalkstein[26], and the total strontium-90 yield of the two shots was 0.4 MCi, as suggested by Stebbins[27], the Rh^{102}/Sr^{90} ratio should be about 7.5 in "rocket-shot" debris. None of the samples had ratios approaching that value. In the northern hemisphere the ratio appeared to increase with time, approaching the values found in the southern hemisphere.

Possibly the Rh^{102}/Sr^{90} ratio in debris derived exclusively from the rocket shots was about 3.5, as suggested by the southern hemisphere samples. Perhaps the lower values originally found in norhtern hemisphere samples resulted from inclusion within them of small amounts of debris from some other high altitude injection, presumably an event in the 1958 USSR test series. Such conjectures cannot be substantiated from the data acquired during Project HASP or Project STARDUST, of course, and may be entirely erroneous. If true, it would suggest, among other things, that the assumed rhodium-102 yield of Orange was much too high or the assumed strontium-90 yields of Teak and Orange were too low.

We may conclude safely only that the debris from the rocket shots did not reach the lower stratosphere with a simple uniform composition.

6.10 Stratospheric Burdens and Residence Times of Radioactive Debris

It is possible to estimate from the data accumulated during Project HASP the approximate distributions and burdens of strontium-90 and of tungsten-185 within the stratosphere during 1958 to 1960. By noting the rate of decrease of these burdens as a function of time, it is possible to estimate a mean stratospheric residence time for each of these nuclides.

The strontium-90 distribution within the lower stratosphere was estimated for each of a series of intervals of two to four month duration for which adequate data were available. The region above 22 km was not samples, and during each of the time intervals, there were large regions of the lower stratosphere which were inadequately sampled. In estimating the distributions, it has been assumed that the distribution of radioactivity was uniform in the zonal direction, and this was probably far from true for all intervals prior to mid-1959. Because of these uncertainties, the distributions both of strontium-90 and of tungsten-185 which are given below, as well as the burdens and residence times calculated from them, are only approximate.

The estimated distributions of strontium-90 in the lower stratosphere are shown for November - December 1957 and January - April 1958 in Figure 43, for May - July and September - December 1958 in Figure 44, for January - April and May - August 1959 in Figure 45, and for September - December 1959 and January - April 1960 in Figure 46. Significant numbers of samples were collected in the southern hemisphere only between September 1958 and August 1959, and during this period, sampling of the northern polar stratosphere was quite sparse. Unfortunately, the injection of large amounts of debris into the northern polar stratosphere by the late 1958 USSR test series occurred while HASP sampling was concentrated in the tropical stratosphere and southern polar stratosphere. Most of this debris had entered the troposphere and had fallen out to the surface of the earth by the time abundant sampling of the northern polar stratosphere for Project HASP was resumed in mid-1959. As a result, HASP sampling never gave adequate data for this material, which evidently had a quite short stratospheric residence time. Thus the strontium-90

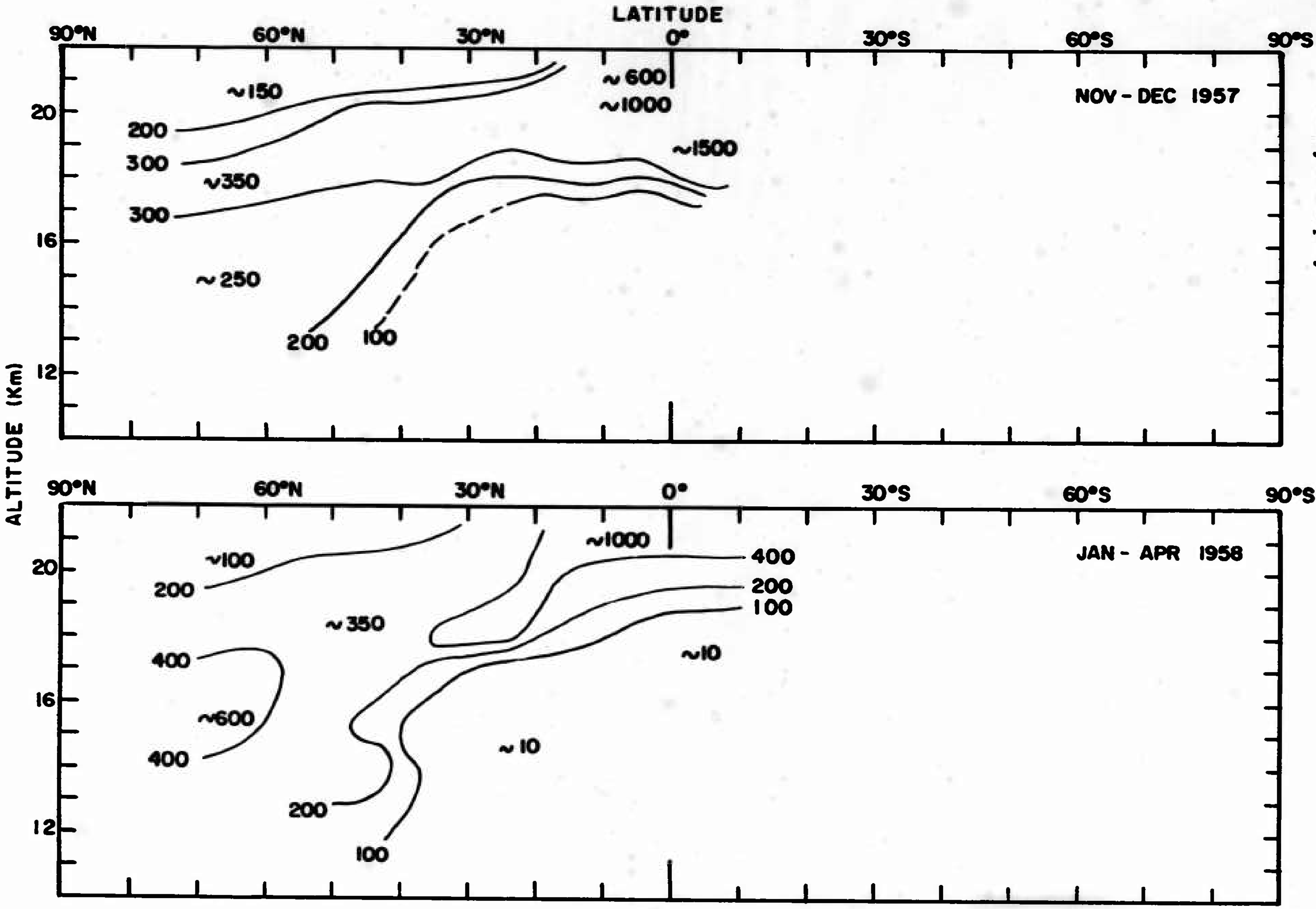

FIGURE 43. DISTRIBUTION OF STRONTIUM-90, NOV - DEC 1957 AND JAN - APR 1958

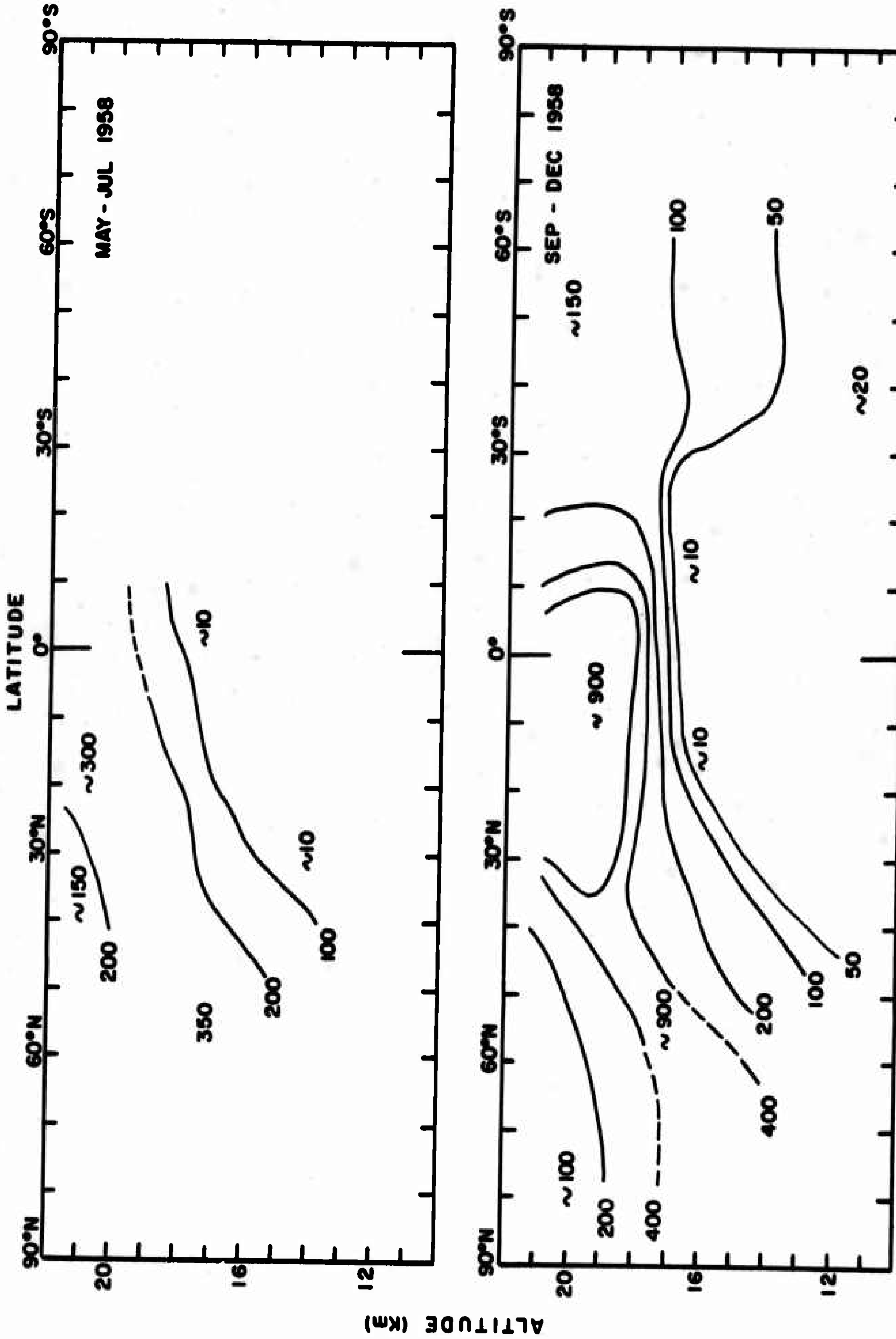

FIGURE 44. DISTRIBUTION OF STRONTIUM-90, MAY-JUL 1958 AND SEP-DEC 1958

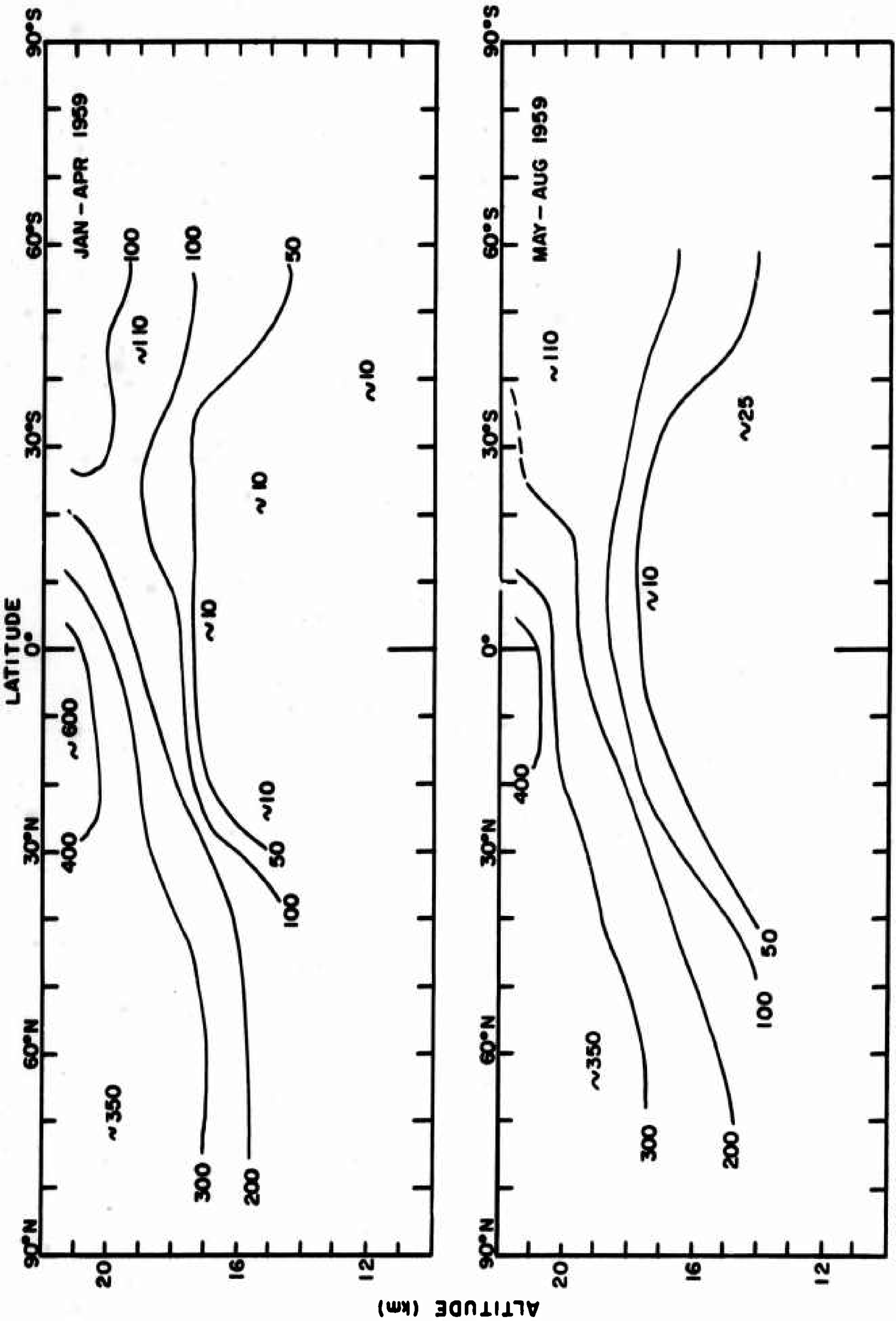

FIGURE 45. DISTRIBUTION OF STRONTIUM-90, JAN - APR 1959 AND MAY - AUG 1959

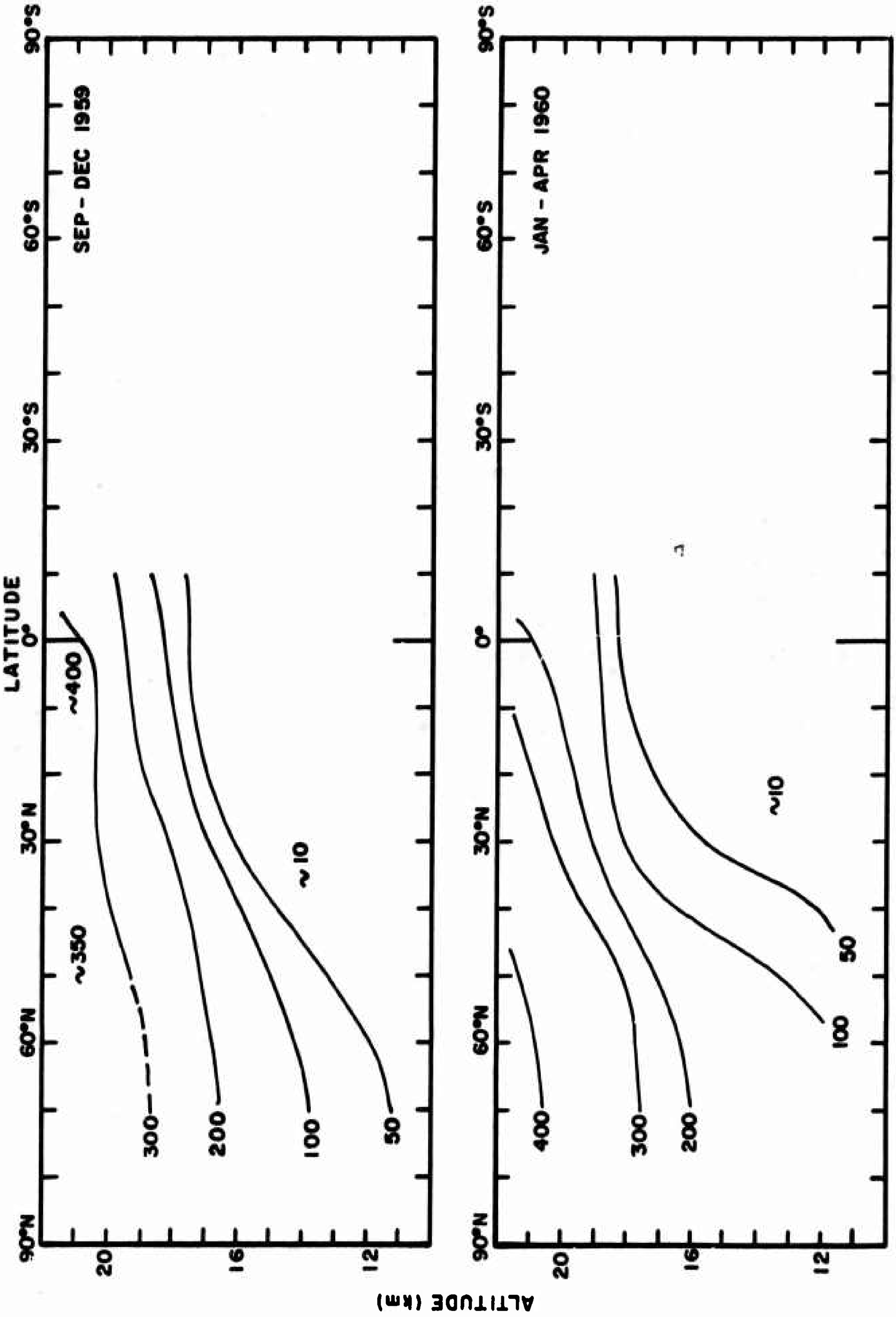

FIGURE 46. DISTRIBUTION OF STRONTIUM-90, SEP-DEC 1959 AND JAN - APR 1960

burdens for late 1958 and early 1959 which are given below are probably too low, and the residence half-time for strontium-90 which is calculated from them is probably too short.

From the strontium-90 distributions shown in Figures 43 to 46, stratospheric burdens were calculated, using the method illustrated by Tables 28 and 29. The concentrations were assumed to be uniform in the zonal direction, and the stratosphere was divided into rings by the intersection of latitude bands 10° wide and layers 40 mb thick. The mean concentration in each ring which had been sampled was estimated, and values were extrapolated into unsampled regions on the basis of earlier and later sampling of those regions. Concentrations in the upper stratosphere were estimated mainly on the basis of any subsequent movement of debris from that region into the lower stratosphere. The estimated concentrations for the interval, January - April 1959 are shown in Table 28. The high concentrations in the 0-40 mb band represent mainly the strontium-90 injected into the upper stratosphere by the August 1958 U. S. rocket shots, Teak and Orange. From the concentrations and masses of air in each ring represented in Table 28 were calculated the burdens listed in Table 29. Similar calculations gave the burdens for other intervals which are listed in Table 30, and are plotted in Figure 47.

From Figure 47, it may be seen that the apparent residence half-time for strontium-90 during late 1958 to early 1960 was about sixteen months. This represents the rate of fallout of a source that included large injections into the lower stratosphere, the upper stratosphere and the mesosphere. Had the late 1958 USSR injections been more adequately sampled, the calculated burdens during late 1958 and early 1959 would doubtless have been higher, and the calculated residence half-time shorter.

TABLE 28. Assumed Mean Strontium-90 Concentrations (pCi Sr^{90}/100 SCM) in the Stratosphere During January - April 1959

Pressure Altitude (mb)	Latitude 90°-80°N	80°-70°N	70°-60°N	60°-50°N	50°-40°N	40°-30°N	30°-20°N	20°-10°N	10°-0°N
0-40	800	800	800	850	850	850	900	900	900
40-80	350	350	350	350	350	350	400	380	350
80-120	250	250	250	250	250	220	75	25	15
120-160	170	170	170	170	150	100	25	5	5
160-200	130	130	130	130	100	30	5	-	-
200-240	100	100	100	80	50	10	-	-	-
240-280	50	50	50	40	25	5	-	-	-
280-320	30	30	30	25	10	-	-	-	-
320-360	10	10	10	10	5	-	-	-	-
360-400	5	5	5	5	-	-	-	-	-

Pressure Altitude (mb)	Latitude 0°-10°S	10°-20°S	20°-30°S	30°-40°S	40°-50°S	50°-60°S	60°-70°S	70°-80°S	80°-90°S
0-40	650	550	380	300	300	300	300	300	300
40-80	220	150	120	110	110	110	110	110	110
80-120	10	10	10	35	50	75	75	75	75
120-160	-	-	5	25	35	50	50	50	50
160-200	-	-	-	10	15	20	20	20	20
200-240	-	-	-	5	5	5	5	5	5
240-280	-	-	-	-	-	-	-	-	-
280-320	-	-	-	-	-	-	-	-	-
320-360	-	-	-	-	-	-	-	-	-
360-400	-	-	-	-	-	-	-	-	-

TABLE 29. Calculated Strontium-90 Burdens (kCi) in the Stratosphere During January-April 1959

Latitude

Pressure Altitude (mb)	90°-80°N	80°-70°N	70°-60°N	60°-50°N	50°-40°N	40°-30°N	30°-20°N	20°-10°N	10°-0°N
0-40	8.7	30.7	50.1	72.2	89.1	103.3	121.0	128.9	132.9
40-80	3.8	13.4	21.9	29.8	36.7	42.5	53.8	54.4	51.7
80-120	2.7	9.6	15.6	21.2	26.2	26.7	10.1	3.6	2.2
120-160	1.8	6.5	10.6	14.4	15.7	12.2	3.3	0.7	0.8
160-200	1.4	5.0	8.2	11.0	10.5	3.7	0.7	-	-
200-240	1.1	3.8	6.3	6.8	5.2	1.2	-	-	-
240-280	0.5	1.9	3.1	3.4	2.6	0.6	-	-	-
280-320	0.3	1.2	1.9	2.1	1.1	-	-	-	-
320-360	0.1	0.4	0.6	0.9	0.5	-	-	-	-
360-400	0.1	0.2	0.3	0.4	-	-	-	-	-
Total	20.5	72.7	118.6	162.2	187.6	190.2	188.9	187.6	187.6

Pressure Altitude (mb)	0°-10°S	10°-20°S	20°-30°S	30°-40°S	40°-50°S	50°-60°S	60°-70°S	70°-80°S	80°-90°S
0-40	96.0	78.8	51.1	36.4	31.4	25.5	18.8	11.5	3.2
40-80	32.5	21.5	16.1	13.4	11.5	9.4	6.9	4.2	1.2
80-120	1.5	1.4	1.3	4.3	5.2	6.4	4.7	2.9	0.8
120-160	-	-	0.7	3.0	3.7	4.2	3.1	1.9	0.5
160-200	-	-	-	1.2	1.6	1.7	1.3	0.8	0.2
200-240	-	-	-	0.6	0.5	0.4	0.3	0.2	0.1
240-280	-	-	-	-	-	-	-	-	-
280-320	-	-	-	-	-	-	-	-	-
320-360	-	-	-	-	-	-	-	-	-
360-400	-	-	-	-	-	-	-	-	-
Total	130.0	101.7	69.2	58.9	53.9	47.6	35.1	21.5	6.0

Northern Hemisphere = 1.32 MCi

Southern Hemisphere = 0.52 MCi

TABLE 30. Calculated Strontium-90 Burdens (MCi) in the Stratosphere Late 1957 - Early 1960

	Burden Below 40 mb			Total Burden		
Interval	90°-0°N	0°-90°S	Total	90°-0°N	0-90°S	Total
Nov-Dec 1957	0.61	-	-	0.80	-	-
Jan-Apr 1958	0.63	-	-	0.81	-	-
May-Jul 1958	0.43	-	-	0.60	-	-
Sep-Dec 1958	0.76	0.30	1.06	1.50	0.55	2.05
Jan-Apr 1959	0.58	0.16	0.74	1.32	0.52	1.84
May-Aug 1959	0.44	0.18	0.62	1.08	0.53	1.61
Sep-Dec 1959	0.37	-	-	0.95	-	-
Jan-Apr 1960	0.42	-	-	0.74	-	-

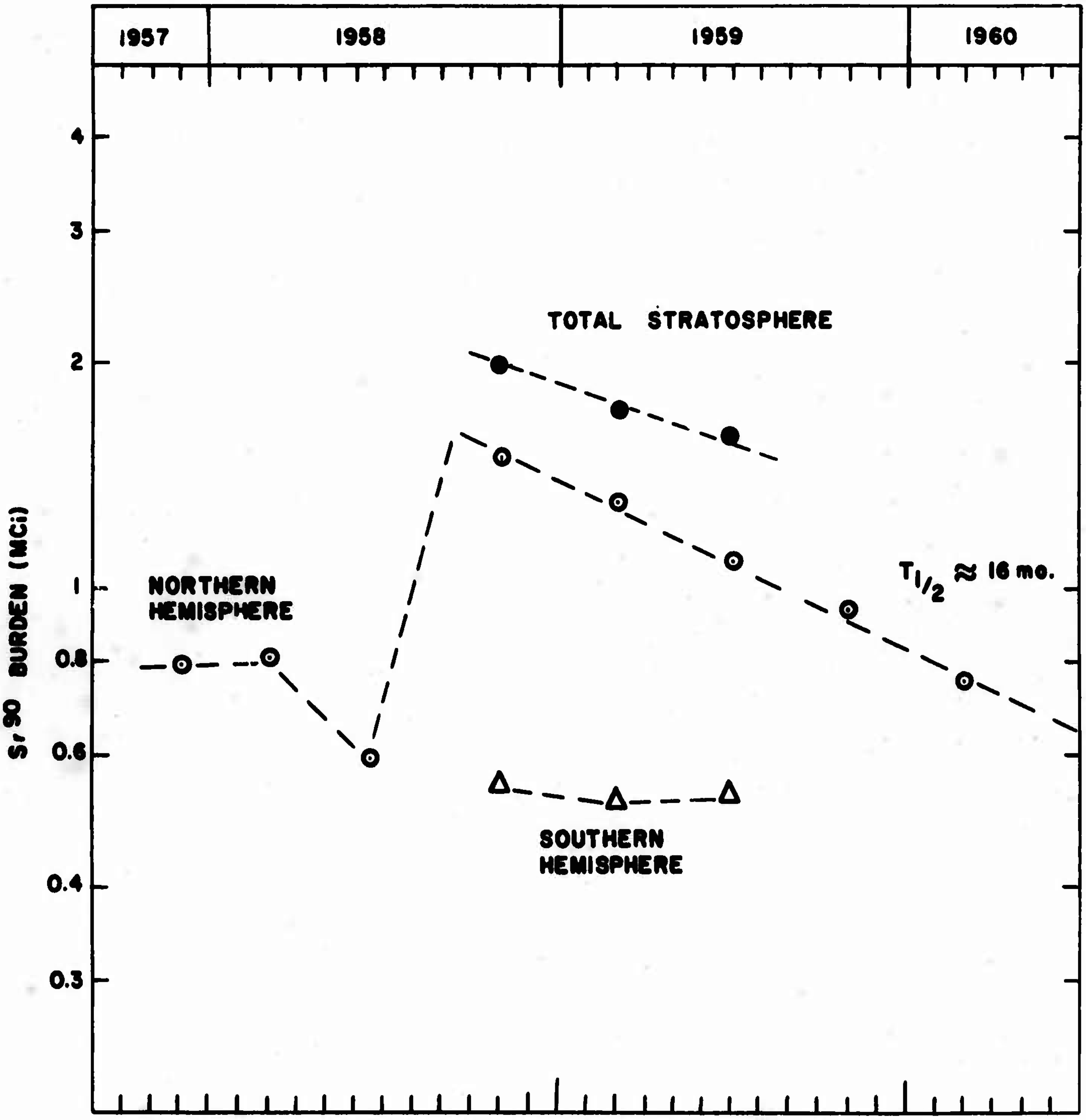

FIGURE 47. CALCULATED STRONTIUM-90 BURDENS IN THE STRATOSPHERE

The stratospheric burdens of tungsten-185 have been calculated for a series of intervals of a month or less during late 1958 to early 1960. The results are given in Table 31, and some are plotted in Figure 48. No data were available for the region above 21 km, so the concentration in the 0-40 mb layer were assumed arbitrarily to be 25 percent of those in the 40-80 mb layer on the basis of vertical profiles of concentration observed in the northern polar stratosphere.

The apparent residence half-time of tungsten-185 in the lower tropical stratosphere (30°N - 30°S) was about 5.5 months, and in the entire stratosphere, about eight months. The short stratospheric residence time of this nuclide, compared to that of strontium-90 described above, resulted no doubt from its injection mainly into the lowest layers of the stratosphere, from whence it could readily be transferred into the troposphere. It is evident that the residence time of material injected into the stratosphere is quite sensitive to the altitude of injection.

6.11 Summary of Information Obtained

Measurements of radioactivity injected into the stratosphere by pre-1961 nuclear weapon tests provided information on mechanisms and rates of mixing and transport of air within the stratosphere and between the stratosphere and troposphere.

Radioactivity injected into the equatorial stratosphere by the November 1957 and September 1958 U.K. weapon tests was distributed rather symmetrically about the equator. No doubt this resulted from the tendency of winds in the equatorial stratosphere to blow essentially parallel to the

TABLE 31. Calculated Tungsten-185 Burdens (MCi, corrected to 12 August 1958) in the Stratosphere, Late 1958 - Early 1960

Time Interval	90° - 30° N	30° - 0° N	0° - 30° S	30° - 90° S	Total
14-29 Oct 1958	12	60	27	8	107
3-23 Dec 1958	-	18.1	7.8	3.9	-
6-28 Jan 1959	-	11.4	7.8	4.1	-
1-29 Apr 1959	10.6	7.8	4.6	2.2	25.2
1-29 May 1959	8.3	7.4	6.3	2.7	24.7
2-26 Jun 1959	8.3	5.8	4.8	3.0	21.9
21 Jul-8 Aug 1959	6.2	5.0	4.5	3.6	19.3
5-15 Jan 1960	5.1	26	-	-	-

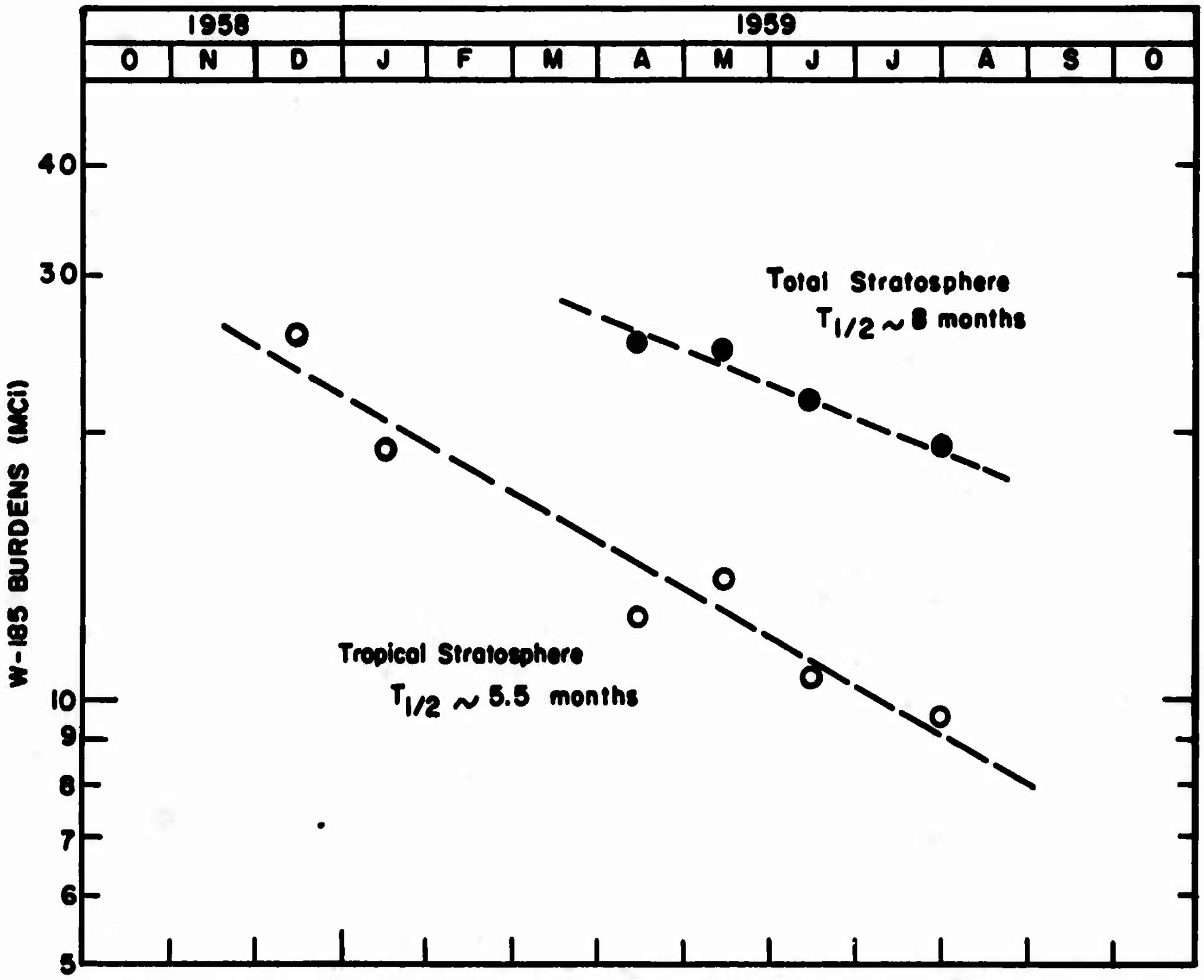

FIGURE 48. TUNGSTEN-185 BURDENS (corrected to 12 Aug 1958) IN THE STRATOSPHERE

zonal direction. Radioactivity injected into the northern tropical stratosphere by the 1958 U. S. test series remained mainly in the northern hemisphere, though significant amounts were transferred into the southern polar stratosphere during the southern hemisphere winter of mid-1959. Radioactivity injected into the upper atmosphere by the August 1958 rocket shots, Teak and Orange, was more evenly divided between hemispheres as it was transferred downward into the lower stratosphere, though one possible interpretation of the data would be that debris from Teak preferentially entered the lower atmosphere in the northern hemisphere, while that from Orange was evenly divided between hemispheres. Radioactivity injected into the northern polar stratosphere by the October 1957, early 1958, and November 1958 USSR weapon tests mainly remained in the polar stratosphere until transferred to the troposphere, but some did penetrate the tropical stratosphere, reaching as far south as 25°N within a month after injection.

The interceptions of fresh debris suggested that not all megaton yield nuclear events inject their debris into the stratosphere. For example, no evidence was found that debris from the 28 April 1958 U.K. megaton event[13] entered the stratosphere. Moreover, it appeared that debris injected into the polar stratosphere stabilizes at lower levels than debris injected into the tropical stratosphere, for very little debris from the 1958 USSR tests was intercepted at or above 19 km in the polar stratosphere during October and November 1958, although high concentrations were found below 18 km. Debris from events of comparable yield, but detonated at low latitudes, was generally found in highest concentrations at 18 to 21 km altitude.

The activity ratio, W^{185}/Sr^{90} in debris from the 1958 U. S. test series did not agree too well with the ratios predicted by Martell on the basis of yield of estimates[15]. It appears, therefore, that debris from weapons of different yields was mixed together, probably as a result of overlapping of the layers into which the debris was injected.

The failure of HASP sampling missions to intercept any debris from the 1958 USSR tests performed after the middle of October is attributed to the retention of that debris within the circulation of the winter polar vortex, and its consequent failure to reach latitudes south of 45°N over North America during the winter of 1958 - 1959. In Chapter 7 a similar phenomenon involving debris from the 1961 USSR tests is discussed.

The tungsten-185 injected into the tropical stratosphere by the 1958 U. S. test series moved downward as it moved poleward, apparently in response to eddy diffusion with its principal axis lying within mixing layers which sloped downward from the equator toward the poles. On the other hand, radioactivity injected into the polar stratosphere by USSR weapon tests appeared to move upward as it moved equatorward, presumably as a result of eddy diffusion within these same mixing layers.

The residence half-time within the stratosphere of the tungsten-185 injected into the lower layers of the tropical stratosphere by the 1958 U. S. tests appeared to be about eight months. The residence half-time of the strontium-90 injected by the U. S., U.K. and USSR weapon tests appeared to be about sixteen months. Most of this strontium-90 had been injected into the lower stratosphere and mesosphere, accounting for the longer residence time.

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Measurements of fission products attributed to the August 1958 rocket shots suggests that material from these high altitude injections had a mean atmospheric residence time at the lower end of the five to twenty-year range suggested by Kalkstein[26].

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13. ABSTRACT

In this, the final report on Project STARDUST, a review is given of the results of analyses of filter and gas samples collected during the course of the program, 1961 to 1967, and a summary is given of the conclusions reached on the basis of these results concerning the influence of atmospheric processes on the transfer and fallout of radioactive materials injected into the stratosphere. Reference is made to results obtained during Project HASP also, for in many ways, Project STARDUST was a continuation of that program. ( )

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Mathematical Fallout Model						
Mathematical Upper Atmosphere Model						
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Analysis Neutron Activation Products						
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