DASA 1222

MIXING and TRANSFER within the STRATOSPHERE

by

HERBERT W. FEELY

DR. JEROME SPAR

31 Dec. 1960

ISOTOPES, INC.

ERRATA SHEET

For

DASA 1222

Page No.	Delete	Insert
1	3.2×10^5	5.7×10^7
56	$\pm$ 2%	< 2%
59	Table I (Sr90)	New Table I (below)
60	P-3	P-4
60	$(7.0 \pm 3.0) \times 10^9$	$(4.6 \pm 1.5) \times 10^7$
60	$(5.0 \pm 2.0) \times 10^4$	$(3.4 \pm 1.2) \times 10^2$
60	40%	35%
61	P-1 and P-2	P-1, P-2, and P-4
62	Second Sentence	New Sentences (below)
62	40%	35%
63	7×10^9	4.6×10^7
63	3.2×10^5	5.7×10^7
63	2.2×10^5	4.0×10^7

For Second Sentence on Page 62, Insert:

The relatively high volume concentration in sample P-3, the only sample collected at 40,000 feet in thepolar stratosphere, might be due to production of sulfate particles in this region of the stratosphere. Such production could logically occur in the spring season when the ozone content of the lower stratosphere is at ist highest. In view of the combined uncertainties in the estimation of the particle volume concentration and the average strontium-90 concentrations, it does not seem worthwhile to pursue the interpretation of a simple sample any further.

All Strontium-90 concentrations shown on Table I Page 59 are incorrect. The following values should be substituted:

Sample No.	Sr-90 Concentration ($dpm/cm^3 \times 10^{-7}$)
P-1	4.92
P-2	4.24
P-3	3.84
P-4	2.71
T-1	2.23
T-2	3.63
T-3	2.70
T-4	6.18
T-5	3.12
T-6	2.84

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MIXING AND TRANSFER WITHIN THE STRATOSPHERE

by

Dr. Herbert W. Feely
and
Dr. Jerome Spar

A progress report on HASP conducted under Contract DA-29-044-XZ-609
prepared for the Defense Atomic Support Agency
Washington 25, D.C.

31 December 1960

ADMINISTRATIVE COMMENT

This document contains material submitted by Isotopes Inc. in their 1 October 1960 Quarterly Progress Report on the DASA High Altitude Sampling Program. In addition, an appendix contains information on the size distribution and composition of stratospheric particles. The latter has been submitted for publication in Science. The duct flow charts on pages 6 and 7 were constructed at Hq. DASA using data obtained by Professor Elliott G. Reid of Stanford University. Isotopes, Inc. 123 Woodland Ave., Westwood, N.J. is the principal contractor charged with the radiochemical analysis and interpretation of the HASP data. In addition, they have performed subsidiary studies on such subjects as stratospheric particulate, radioactivity in surface air , rainfall, and soil, and filter paper collection efficiency.

ABSTRACT

Preliminary data on Phases 4 and 5 of Crowflight are given. The recalibrated U-2 duct curves are given. Tritium and Carbon-14 concentrations in New Jersey rain and air samples during 1959 and 1960 respectively are reported. The manner of mixing and transfer of Strontium-90 in the stratosphere is described. The seasonal in crease in mixing from the tropical to polar regions is noted. Tungsten-185 distributions are discussed. Measurements of Rhodium-102 indicate that Teak and Orange debris has entered the lower stratosphere. A mean residence time of less than 10 years for this debris is suggested. Concentrations of Beryllium-7, a cosmic-ray product, are reported. Strontium-89 and Cerium-144 data is discussed in the light of the HASP mixing model. The size distribution and composition of stratospheric particles collected on electron microscope probes exposed in the stratosphere is described. Particles in the size range 0.1 to 1.5 microns appear to be composed of ammonium persulphate. A stratospheric inventory of 3.2×10^5 Kg of SO_4 is deduced.

TABLE OF CONTENTS

INTRODUCTION

The collection of samples for the High Altitude Sampling Program has been completed. Phase 4 ended with a mission flown from Minot AFB, North Dakota on 6 May 1960 and one flown from Ramey AFB, Puerto Rico on 9 May 1960. During Phase 5 samples were collected during missions flown from Ezeiza Airfield, Argentina and from a few bases in the Northern Hemisphere. This Phase ended on 10 June 1960 with the return of the aircraft from Ezeiza to Laughlin AFB, Texas. The total number of samples collected during the HASP reached almost 4000.

A recalibration of the U-2 nose and hatch samplers was carried out at Edwards AFB, California during the last weeks of July 1960. When the results of this recalibration become available all HASP data will be recalculated.

The analysis of HASP samples for strontium-90, cerium-144 and tungsten-185 has almost been completed. Similarly all analyses for beryllium-7 and for phosphorus-32 have been finished. Some older HASP samples are yet to be analyzed for rhodium-102, though most analyses of recent samples are complete. Analyses of about 100 recent samples for cesium-137 and plutonium have been begun.

Progress has been made in the collection and analysis of soil samples for the delineation of the vertical distribution of certain nuclides through the upper layers of soil.

Analyses have been made of the carbon-14 concentrations in a number of samples of surface air and of the tritium concentration in a number of samples of rain and snow to provide more information on the behavior of gaseous debris from nuclear tests.

Studies of the stratospheric particulates collected on probes by Crowflight aircraft have been continued. A few characteristics of these particles have become evident.

Some data have become available from samples collected in the Southern Hemisphere during Phase 5 of Crowflight. It is evident that the concentration of nuclear debris in the southern stratosphere is much higher now than it was in early 1959, though only slightly higher than it was by mid-1959 when Phase 3 was terminated.

The HASP data may be used to obtain an estimate of the manner in which stratospheric transfer and mixing occur and of the rates at which these processes proceed. The data on stratospheric concentrations of water vapor and ozone corroborate the existence of these processes.

THE SAMPLING PROGRAM

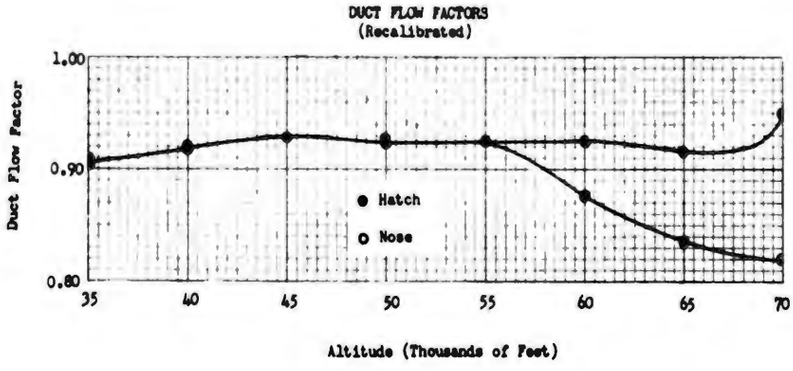
Phase 4 of Crowflight, during which sampling was carried out by aircraft based at Minot AFB, North Dakota and at Laughlin AFB, Texas - with monthly deployments of the aircraft from Laughlin to Ramey AFB, Puerto Rico - was terminated after a mission flown from Minot on 6 May 1960 and one flown from Ramey on 9 May 1960. A total of 1685 useful samples were collected during Phase 4.

Phase 5 began with the deployment of three aircraft from Ramey to Ezeiza Airfield, Argentina on 12 May 1960. Six missions were flown out of Ezeiza. Normally two aircraft flew during each mission. On 8 June 1960 the aircraft returned to Ramey and on 10 June 1960 they returned to Laughlin. During Phase 5 samples were received from several other collection sites. Those collected in the vicinity of Eielson AFB, Alaska should be quite useful but those collected at Laughlin and at Ramey generally represented such short exposure times that few can be of much value. Since many samples received from Ezeiza and from Eielson also represented short exposure times they were commonly combined to produce composite samples. Including these composites, a total of 22 samples were obtained from Ezeiza, a total of 35 from flights between Ezeiza and Laughlin and a total of 17 from Eielson.

SAMPLER CALIBRATION

A recalibration of the nose and hatch samplers used by U-2 aircraft during the HASP was carried out, under the direction of Prof. Elliot G. Reid, at Edwards AFB, California during the last weeks of July 1960. When the results of this recalibration become available they will be used to recalculate all of the HASP data. These data, in their final form, will be presented in the final report for the program (DASA 1300, due in the Fall of 1961).

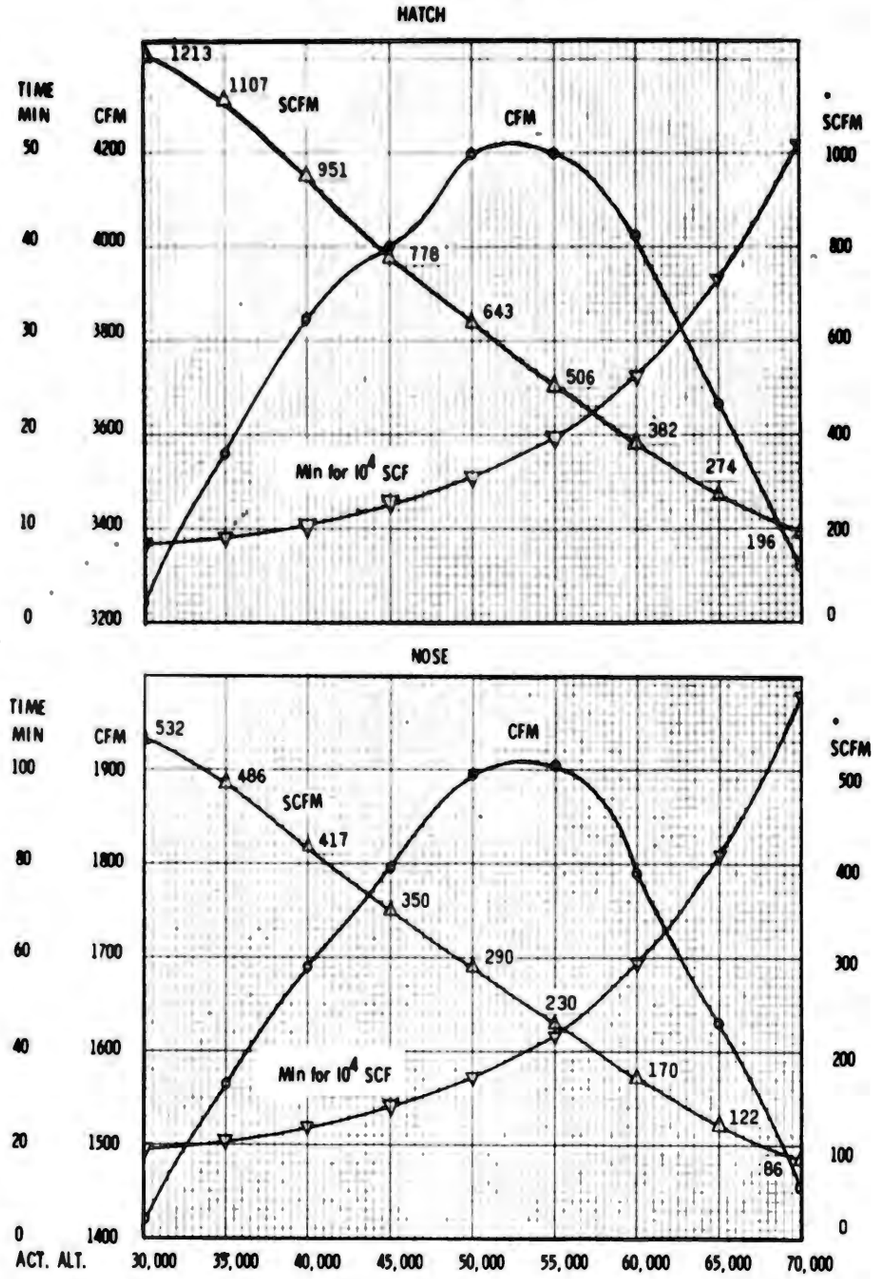
A preliminary estimate of the effect which the new calibration will have on the HASP data indicates that the calculated volumes will be lower, and the calculated nuclide concentrations higher, especially at the higher altitudes. The figure below indicates the factor by which previously computed flow rates must be reduced to correspond to the values one would obtain using the new calibration charts. It should be pointed out that none of the radiochemical data obtained from HASP filter papers reported in this document have been corrected to the new flow rates*; consequently, these values (expressed as dpm/100 scf) should be increased from 8 to 15% to obtain the latest "best value". The latest duct flow curves prepared by Maj. Albert K. Stebbins, III of Hq. DASA using the data obtained by Prof. Reid are shown on the next page.



* The material in the Appendix starting on page 53 has made use of radiochemical data corrected to the new flow rates.

U-2 DUCT FLOW RATES

Based on E.G. Reids curves Sept 60
and IAS schedule in use 59-60



ACT. ALT.	30,000	35,000	40,000	45,000	50,000	55,000	60,000	65,000	70,000
IAS CORR.	7%	7%	8%	8%	9%	10%	11%	13%	15%
TEMP. CORR.	4%	4%	5%	5%	5%	5%	6%	6%	7%
IAS	175	173	170	165	160	150	135	121	108
TAS	280	303	330	361	391	410	414	416	420
IND. TEMP.	-34	-42	-42	-39	-36	-34	-34	-33	-33
MACH NO.	0.48	0.53	0.58	0.63	0.68	0.71	0.72	0.73	0.73

FOR EVERY 10 KNOTS IAS HIGH, ADD IAS CORR. % TO CFM AND SCFM
FOR EVERY 10° COLDER IND. TEMP., ADD TEMP. CORR. % TO SCFM ONLY

• 760 mm Hg 15°C

RADIOCHEMICAL ANALYSES

The routine analysis of HASP samples for strontium-90, cerium-144 and tungsten-185 has been completed. The reanalysis of a number of samples for strontium-90 has been begun. The original analyses of these samples gave results which appeared inconsistent with the other activities observed in the samples or with the activities of other samples collected in the same regions. Experience has indicated that at least half of these discrepancies can be eliminated by reanalysis of the samples.

The analysis of cesium-137 and plutonium has been begun on about 100 samples collected since July 1959. A number of composite samples have been analyzed for beryllium-7 and rhodium-102 and a few of these have also been analyzed for phosphorus-32. It is hoped that the stratospheric distribution of the nuclides produced by cosmic rays and of the tracer introduced at high altitudes in the tropical stratosphere during Hardtack will shed additional light on the rates of mixing within the stratosphere. All of these composites consist of samples collected since 1 December 1959. A series of composites of samples collected before December 1959 will be analyzed for rhodium-102 in an effort to document the changes which have occurred with time in the concentration of this tracer in the lower stratosphere. Rhodium-102 data reported in the Third HASP Technical Report were based upon the beta counting of purified rhodium. These data are subject to error, however, because of the difficulty of separating rhodium from ruthenium, though absorber measurements were used to check the purity of all samples. Rhodium-102 activities are now obtained by gamma spectrometer measurement of the characteristic X-ray and are not subject to interference by ruthenium. There is question as to the true branching ratio for the decay of Rh^{102} and as to the possible existence of an isomer of that nuclide with a long half life, however. As a result of certain inconsistencies in HASP rhodium-102 data, the procedures for chemical preparation of samples are to be checked using a rhodium tracer to be certain that there is no loss of rhodium during the chemical preparation.

SOIL SAMPLING PROGRAM

Preliminary work has been done on the soil sampling program, the objective of which is the determination of the vertical distribution of several nuclides - strontium-90, cesium-137, cerium-144, ruthenium-106 and plutonium - through the upper two feet of soil. The data obtained from this program should be useful in the calculation of the increment in environmental gamma radiation due to fallout, in the estimation of the location of strontium-90 and cesium-137 in soil relative to the root zone of plants and in the estimation of the potential hazard from inhalation of resuspended dust.

The New Jersey and Kansas offices of the U. S. Department of Agriculture have aided in the selection of sampling sites and their personnel will furnish descriptions of the chemical, mineralogical and biological characteristics of the soil samples, in so far as these are available. The samples will display a wide range in texture and composition and should reveal any dependence of vertical penetration of fallout upon such parameters.

Sampling equipment has been designed and constructed or purchased. Chemical procedures have been worked out to permit a sequential analysis for the five nuclides to be measured.

ANALYSES OF GASEOUS DEBRIS

A number of samples of carbon dioxide from surface air, collected since January 1960, has been analyzed for carbon-14. The results, given in Table 1, indicate that surface air, during early 1960, was enriched in carbon-14 by about 20 percent over the concentration which would have existed had there been no production of that radionuclide by nuclear weapons testing. This is in agreement with data obtained by Munnich and Vogel at Heidelberg, Germany, where the excess carbon-14 reached +30 percent during mid-1959 but then declined to about +20 percent by the end of 1959 and has remained near that concentration since. It also agrees with data obtained by W. Broecker for 3 sites in the Northern Hemisphere during late 1959 and early 1960. Concentrations in the Southern Hemisphere may still be a few percent lower than those in the Northern Hemisphere according to data obtained both by Munnich and by Broecker. Evidently during early 1960 the rate of escape of carbon-14 from the stratosphere into the troposphere equalled its rate of movement into the oceans.

A number of samples of rainwater have been analyzed for tritium and the data which are available are given in Table 2. A number of other samples have been enriched by electrolysis and are ready to count, but their analysis has been delayed by difficulties with the tritium counter. The data in Table 2 probably indicate that tritium from Soviet tests does not exhibit the rapid fallout which Libby and others have detected for tritium from United States surface tests in the Pacific. Evidently air bursts inject tritiated water into the stratosphere and it then behaves in much the same way as particulate debris, exhibiting a "spring high" in fallout.

Table 1: Carbon-14 Concentration in Surface Air, Washington Township, N. J.

Sampling Interval	Concentration relative to 0.95 x N. B. S. Standard
19 Jan - 25 Jan 1960	+ 20.9 ± 1.5%
2 Feb - 9 Feb 1960	+ 20.0 ± 1.5%
23 Feb - 1 Mar 1960	+ 18.5 ± 1.5%
1 Mar - 8 Mar 1960	+ 22.4 ± 1.5%
29 Mar - 5 Apr 1960	+ 22.4 ± 1.5%
5 Apr - 12 Apr 1960	+ 16.0 ± 2.5%
26 Apr - 3 May 1960	+ 18.2 ± 1.5%
24 May - 31 May 1960	+ 20.8 ± 1.5%
Leaves (collected 28 May 1960)	+ 17.8 ± 1.5%
21 Jun - 26 Jun 1960	+ 19.7 ± 1.5%
26 Jul - 2 Aug 1960	+ 20.8 ± 1.5%
23 Aug - 6 Sept 1960	+ 18.1 ± 1.5%

Table 2: Tritium Concentration in Rainwater, Westwood, N. J.

Sampling Date	T/H x 10^{18}
28 Apr 1959	1440*
1 May 1959	1370
13 Jun 1959	843
14 Jul 1959	650
9 Aug 1959	140
2 Sep 1959	39
24 Oct 1959	87
25 Nov 1959	47

* All measurements have standard error of about 10 percent

MEASUREMENTS OF STRATOSPHERIC PARTICULATES *

Studies have been begun of samples of stratospheric particles collected by impactors carried by the U-2 aircraft probe. The majority of the particles collected lie within the size range between 0.2 and 2 microns in diameter, above the cut-off size for impaction. Two types of particles appear to be present in abundance. One of these has been identified as ammonium peroxydisulfate by means of electron diffraction measurements. Most commonly the larger particles occur surrounded by halos of smaller particles which are arranged in circular patterns. These suggest that the particles, at some time following collection, formed water droplets about themselves, probably dissolving in them at least partially, but that the water droplet subsequently evaporated leaving the halo-like residue of small particles.

Although quantitative estimates of particle concentrations from these samples are subject to several types of errors it does appear that samples collected in the tropical stratosphere contain more particles, and particles with a smaller mean diameter, than do samples collected in the polar stratosphere.

* A more complete report on this topic appears in the Appendix on page 53.

RESULTS FROM PHASE 4 AND PHASE 5

The Distribution of Strontium-90 in the Stratosphere

The stratospheric distribution of strontium-90 constitutes the best criterion available in the HASP data for judging the behavior of the total stratospheric burden of nuclear debris remaining from all past injections. Strontium-90 data from samples collected during Phase 4 and Phase 5 have been used in the preparation of diagrams to illustrate the mean distribution of strontium-90 in the stratosphere during 1959 and 1960. The mean distribution during 1958 is shown in Figure 1, that during 1959 is shown in Figure 2 and that during early 1960 is shown in Figure 3. Regions of high strontium-90 concentration existed in the lower stratosphere during 1958 as a result of weapons tests which were then still being carried out by the United States, the United Kingdom and the Soviet Union. The regions of low strontium-90 concentration which then existed at high altitudes in the polar stratosphere are of interest. It must be assumed either that relatively little debris from previous tests had yet reached these regions or that, if it did reach them, much of it (probably more than half) had already fallen out. The first assumption would imply slow mixing between the tropical and polar stratosphere, slower than subsequent changes in strontium-90 distribution have indicated, while the second would imply a short stratospheric residence time, probably less than 3 years, for debris injected during Castle and Redwing.

During 1959 (Figure 2) the highest strontium-90 concentrations were encountered in the tropical stratosphere. By early 1960 (Figure 3) the concentrations in the tropical stratosphere had decreased but the concentrations in the high polar stratesphere had increased.

These changes are indicated by Figures 4, 5, and 6 in which strontium-90 concentration in each of three latitude bands is plotted as a function of time. Strontium-90 data for samples collected within the 10° to 15° North band (Figure 4) show that, since late 1958

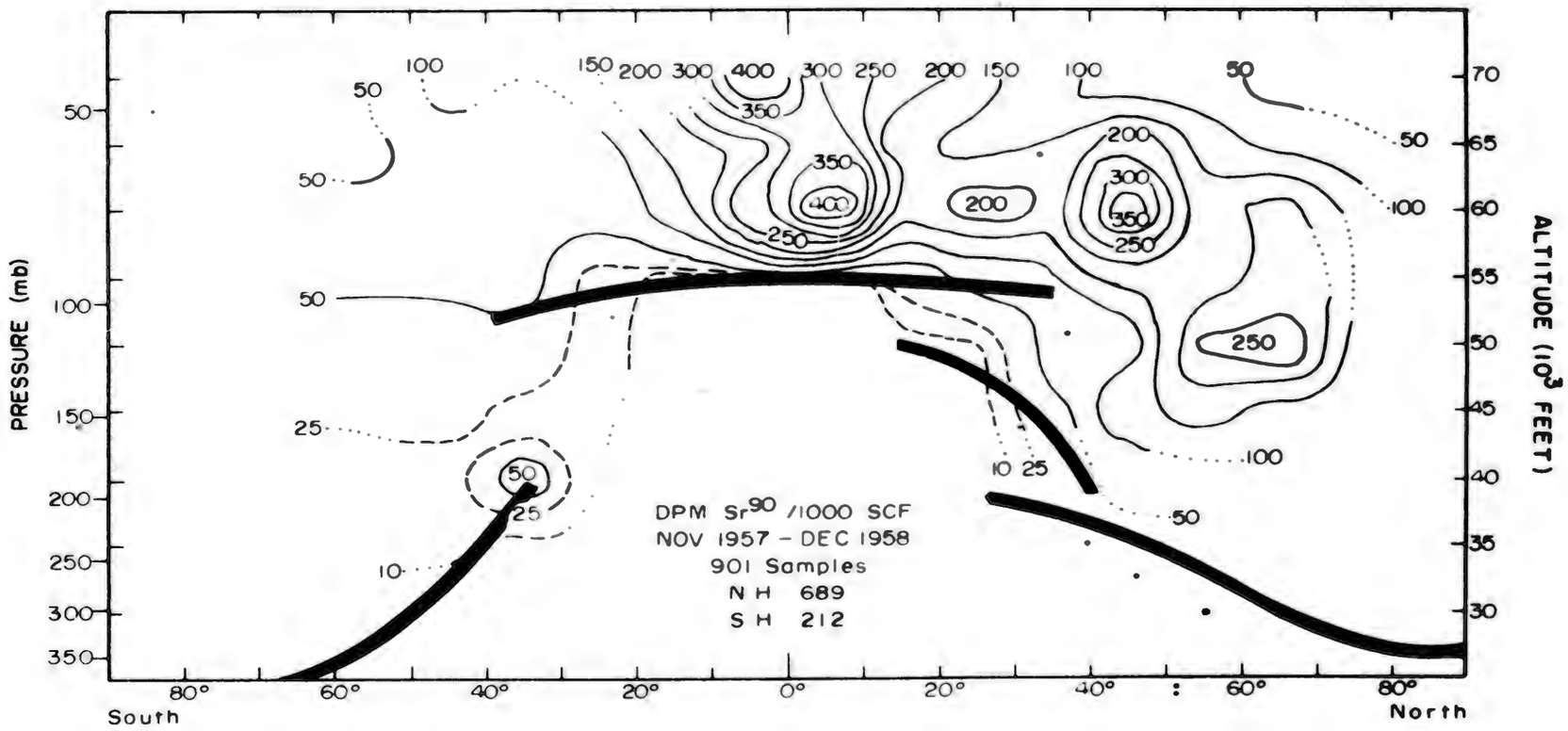


FIGURE 1 MEAN DISTRIBUTION OF STRONTIUM-90, NOVEMBER 1957 - DECEMBER 1958

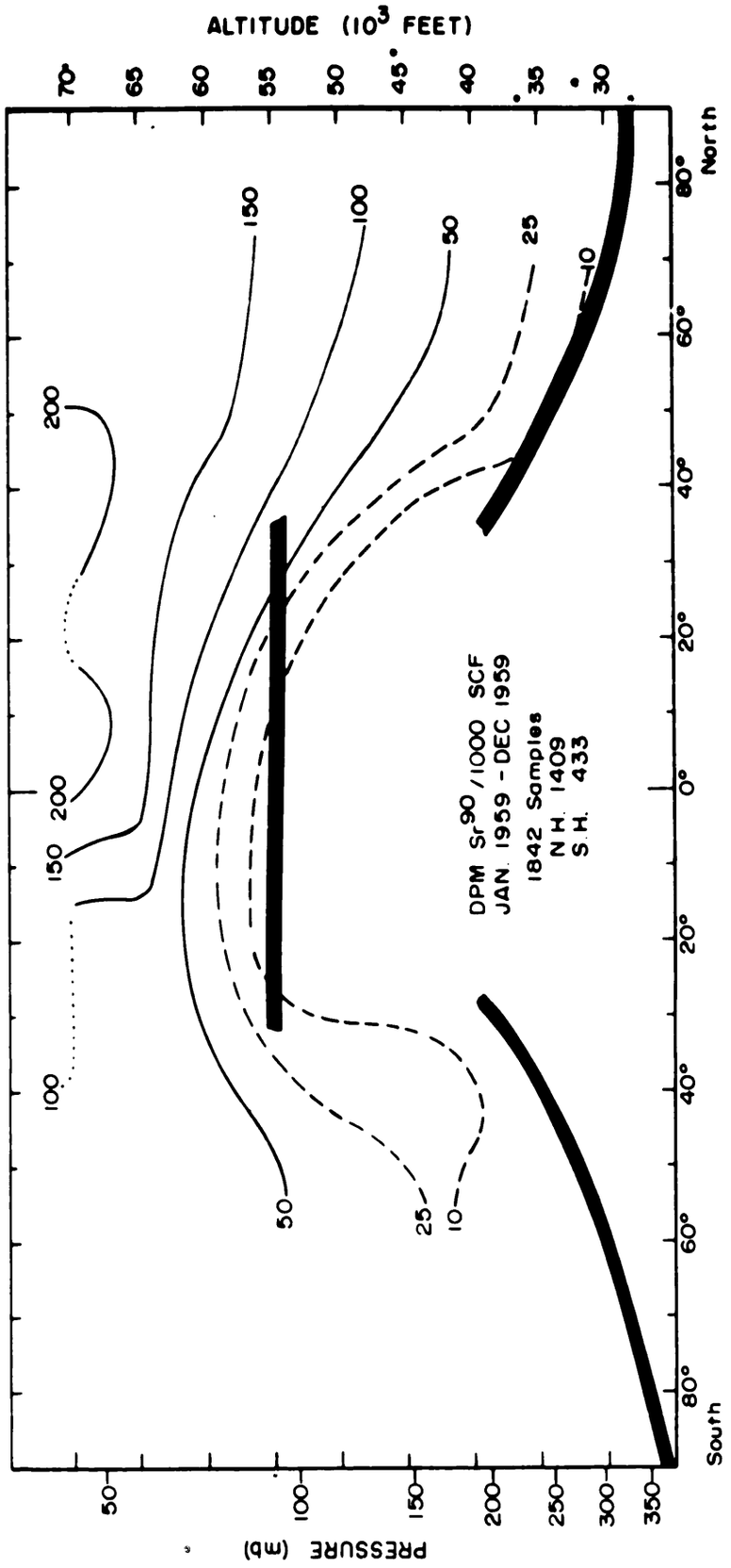


FIGURE 2 MEAN DISTRIBUTION OF STRONTIUM-90, JANUARY - DECEMBER 1959

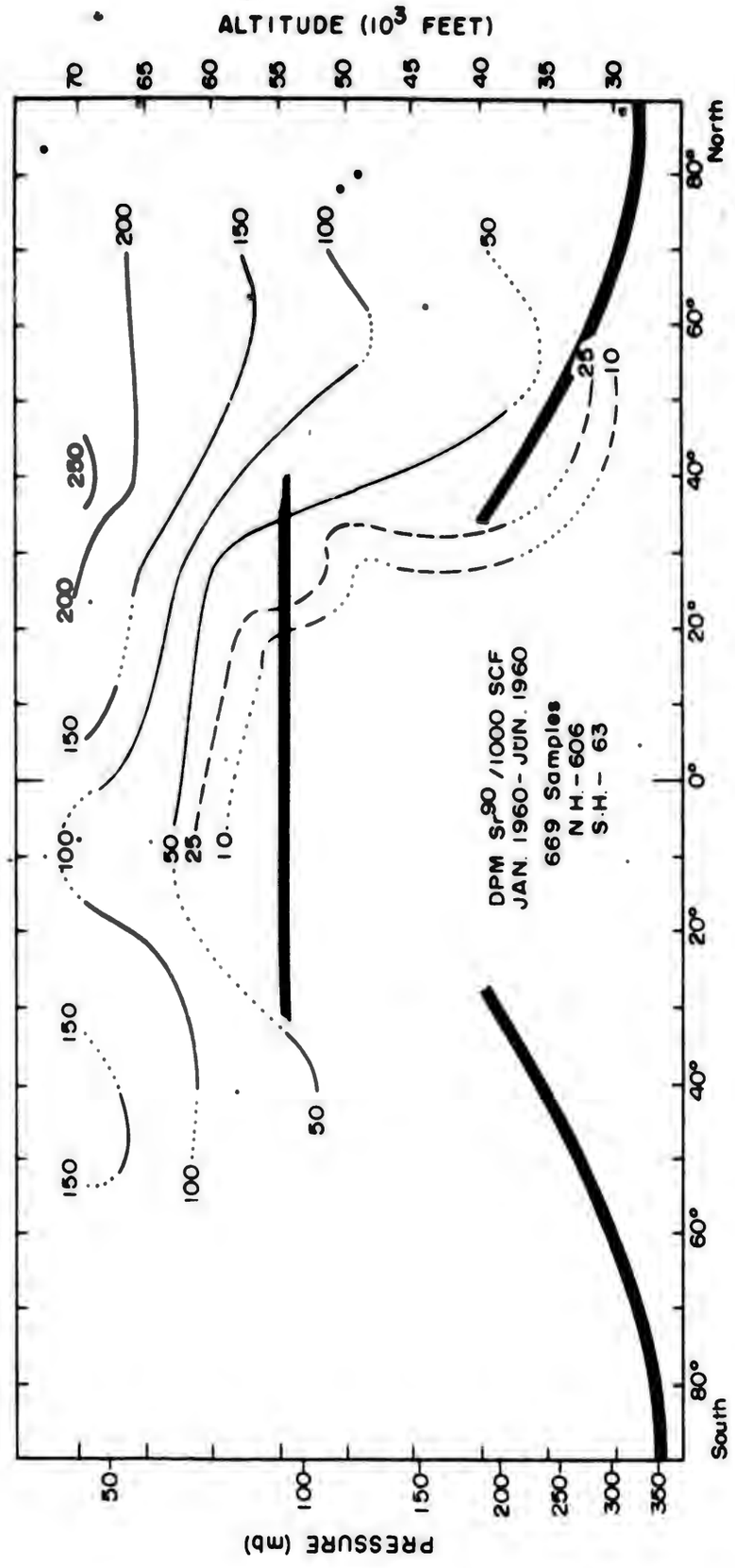


FIGURE 3 MEAN DISTRIBUTION OF STRONTIUM-90, JANUARY 1960 - JUNE 1960

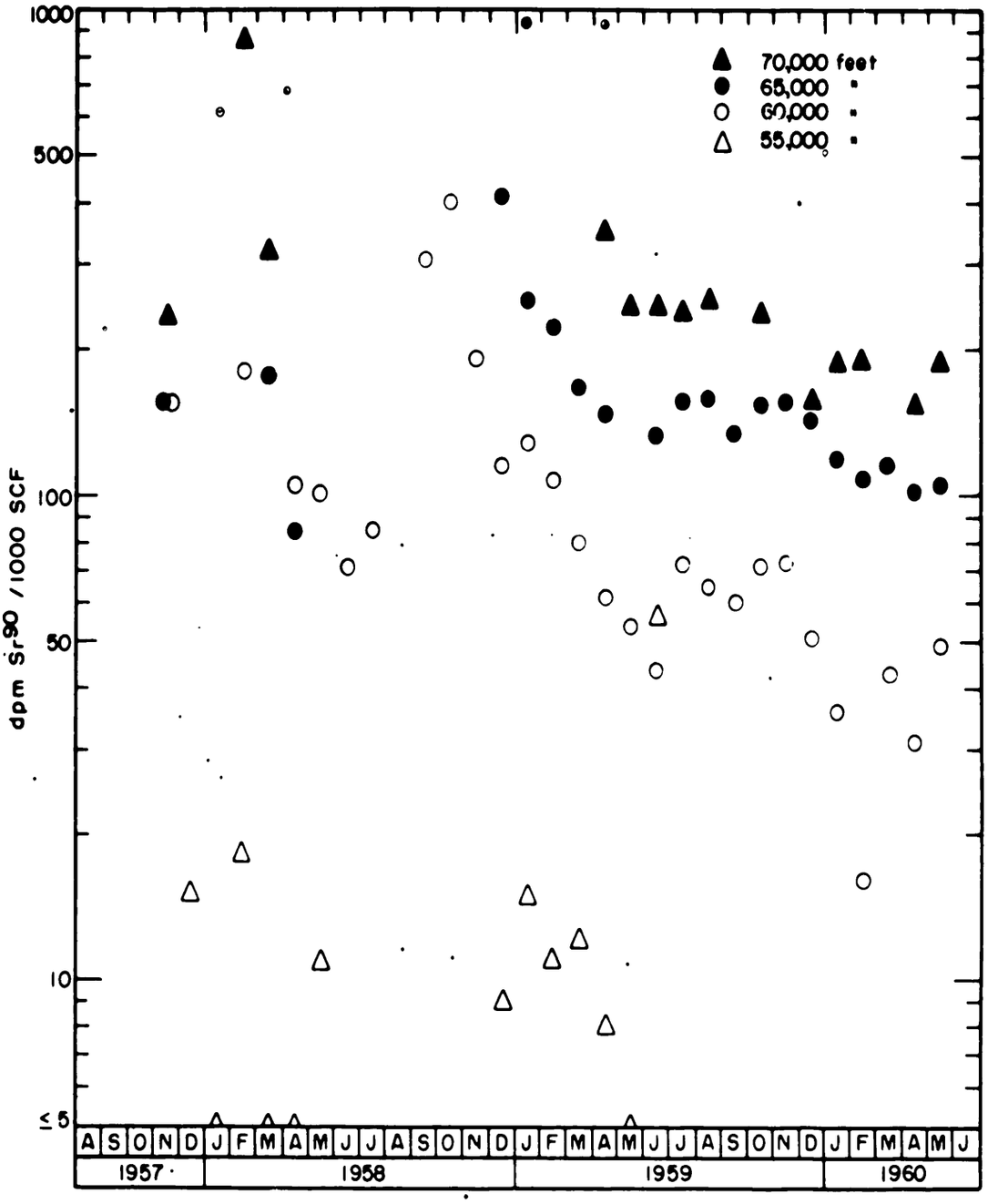


FIGURE 4 THE VARIATION WITH TIME OF THE MONTHLY AVERAGE OF STRONTIUM-90 CONCENTRATION AT EACH ALTITUDE BETWEEN 10° AND 15° NORTH

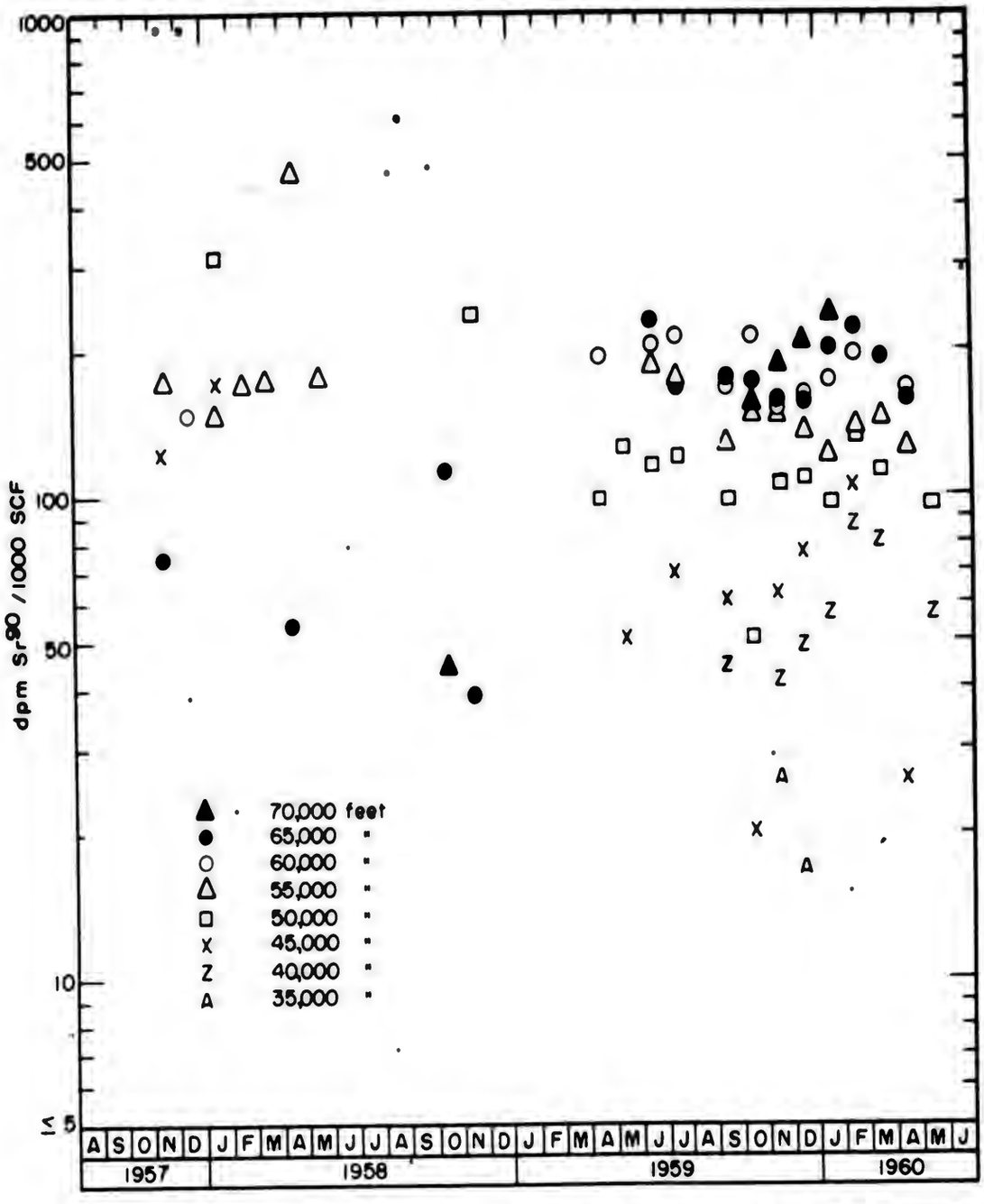


FIGURE 5 THE VARIATION WITH TIME OF THE MONTHLY AVERAGE OF STRONTIUM-90 CONCENTRATION AT EACH ALTITUDE BETWEEN 60° AND 65° NORTH

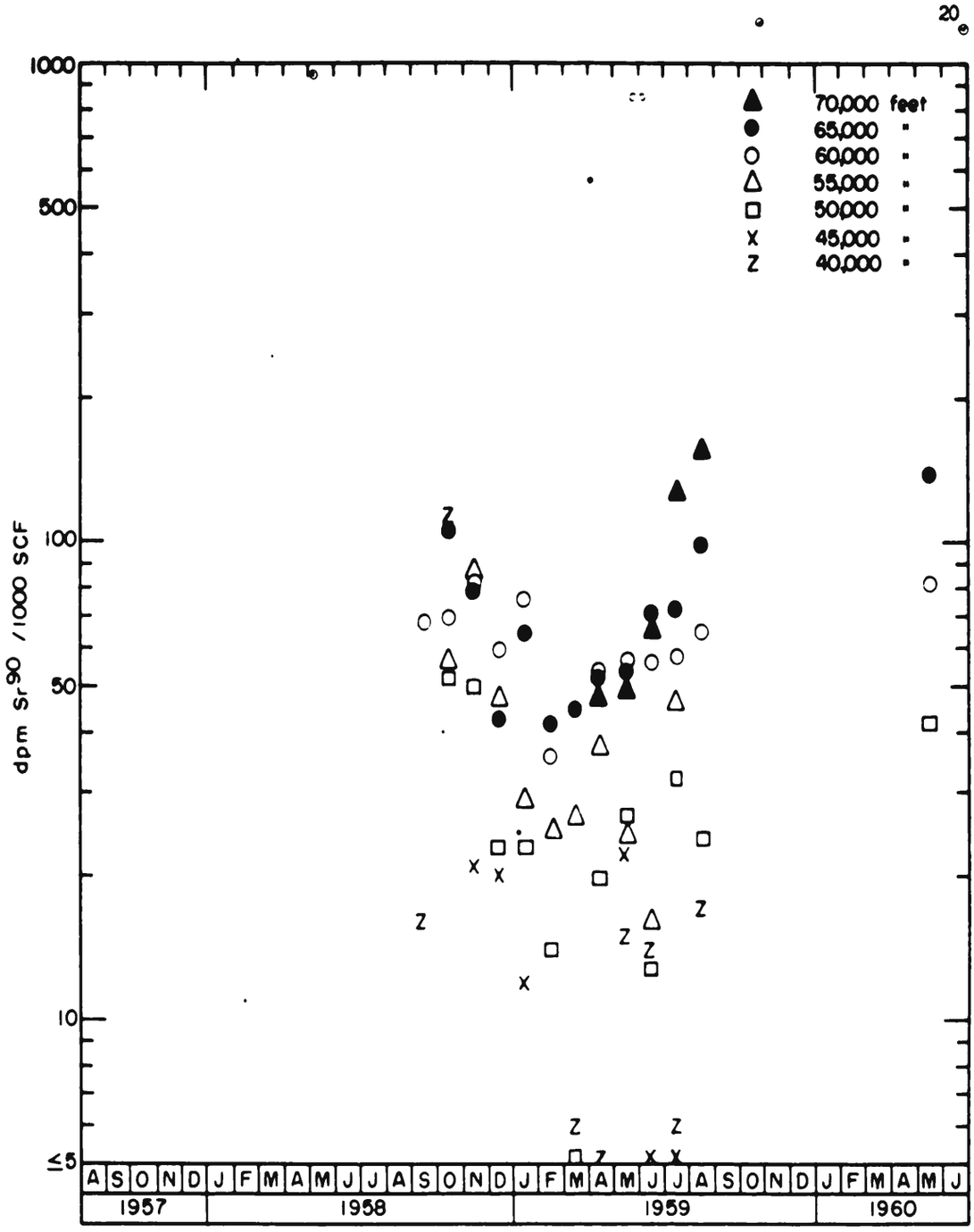


FIGURE 6 THE VARIATION WITH TIME OF THE MONTHLY AVERAGE OF STRONTIUM-90 CONCENTRATION AT EACH ALTITUDE BETWEEN 35° AND 40° SOUTH

when new injections ceased, the concentrations at and above 60,000 feet have decreased substantially only during the late fall, winter and early spring seasons in the Northern Hemisphere. This behavior can be explained by an increased rate of meridional mixing in the stratosphere during the "winter" season.

The vertical concentration gradient in the 10° to 15° North band was much steeper than that in the 60° to 65° North band (Figure 5) during 1959 and 1960 confirming the fact that vertical mixing in the tropical stratosphere is slower than that in the polar stratosphere. The great increase in strontium-90 concentrations at 65,000 feet and above in the polar stratosphere during the winter of 1958-1959 indicates an influx of debris from the high tropical stratosphere or an injection of debris at these altitudes by the autumn 1958 Soviet tests. If the former is the case the isotope ratios should suggest an age of about July 1958 for this debris. If the latter is the case, the isotope ratios should indicate an age of about October 1958 for this debris. If it is Soviet debris we must assume that previous Soviet tests had not injected much debris above 60,000 feet. If it is Hardtack debris we must assume that the injections from Hardtack raised the concentrations in the high tropical stratosphere by about a factor of three, and, therefore, that little Castle or Redwing debris remains in that part of the stratosphere. Unless this assumption is made, it is difficult to explain why Castle and Redwing debris had not migrated into the polar stratosphere fast enough during 1957 and early 1958 to keep the concentrations of strontium-90 in the high polar stratosphere as high as the Hardtack debris kept them in 1959. As is discussed below, both strontium-89 and cerium-144 data indicate that this debris was produced by United States, not Soviet, tests.

There is a suggestion in the strontium-90 data for 60° to 65° North that the concentrations above 55,000 and below 50,000 passed through a maximum in December 1959-January 1960. Perhaps the rate of meridional mixing between the polar and tropical

stratospheres reached a maximum at this time. The subsequent drop in concentrations probably resulted from an increased rate of loss of debris to the troposphere and a decreased rate of meridional mixing with the tropical source area of the debris. The failure of concentrations at 50,000 and 55,000 feet to increase significantly may have resulted from loss of debris from these altitudes to the troposphere by way of the tropopause gap.

The strontium-90 data for 35° to 40° South definitely indicate a seasonal effect in concentrations in the southern polar stratosphere, again probably due to an increase in the rate of mixing between the tropical and polar stratosphere during the "winter" season. During the Southern Hemisphere spring of late 1958, concentrations were high in this latitude band, doubtless due to an influx of Hardtack debris. The concentrations passed through a minimum in the summer of 1958-1959 but rose again in the autumn and winter of 1959, probably due to increasing exchange with the tropical stratosphere. The decreasing concentrations in this latitude band during the early summer of 1958-1959 were probably caused by mixing with higher latitudes in the southern polar stratosphere which contained lower concentrations of debris. Apparently by the autumn of 1960 concentrations in this latitude band had increased somewhat again. The vertical concentration gradient at this latitude was fairly steep by mid-1959 suggesting that source areas in the lower tropical stratosphere which had supplied debris during 1958 had been exhausted and that source areas in the higher tropical stratosphere had become relatively more important.

The Distribution of Tungsten-185 in the Stratosphere

A series of diagrams in the Third HASP Technical Report portrayed the stratospheric distribution of tungsten-185 during late 1958 and early 1959. In Figures 7 through 10 is shown the stratospheric distribution of tungsten-185 during bimonthly intervals from September 1959 through April 1960. The distribution of this nuclide in the stratosphere since its injection in the summer of 1958 provides evidence both on the mixing processes which occur within the stratosphere and on the rate of fallout of debris from the stratospheric layers immediately above the tropopause.

Through 1959 and 1960 the highest concentrations of strontium-90 in the stratosphere were located at altitudes above 70,000 feet but vertical mixing was occurring rapidly enough to prevent depletion of the lower stratosphere by loss of debris through the tropopause. The observed configurations could be explained also, however, by invoking a meridional circulation pattern such as that postulated in the so-called "Brewer-Dobson" model. The decrease in concentration in the tropical stratosphere and the increase in the polar stratosphere which occurred from 1959 into 1960 are the effects which would be predicted for such meridional circulation. They would also be predicted, however, by the HASP model which included periods of rapid mixing between the lower tropical and lower polar stratosphere and rapid vertical mixing in the high polar stratosphere during the winter season. This latter model is preferred over that which invokes meridional circulation because of the observed distribution of stratospheric tungsten-185. As illustrated in Figures 7 through 10 the tungsten-185 concentrations encountered at 50,000 feet in the polar stratosphere have consistently been higher than those encountered at 65,000 to 70,000 feet. The tungsten-185 was originally injected into the tropical stratosphere and the highest atmospheric concentrations have always been found in that

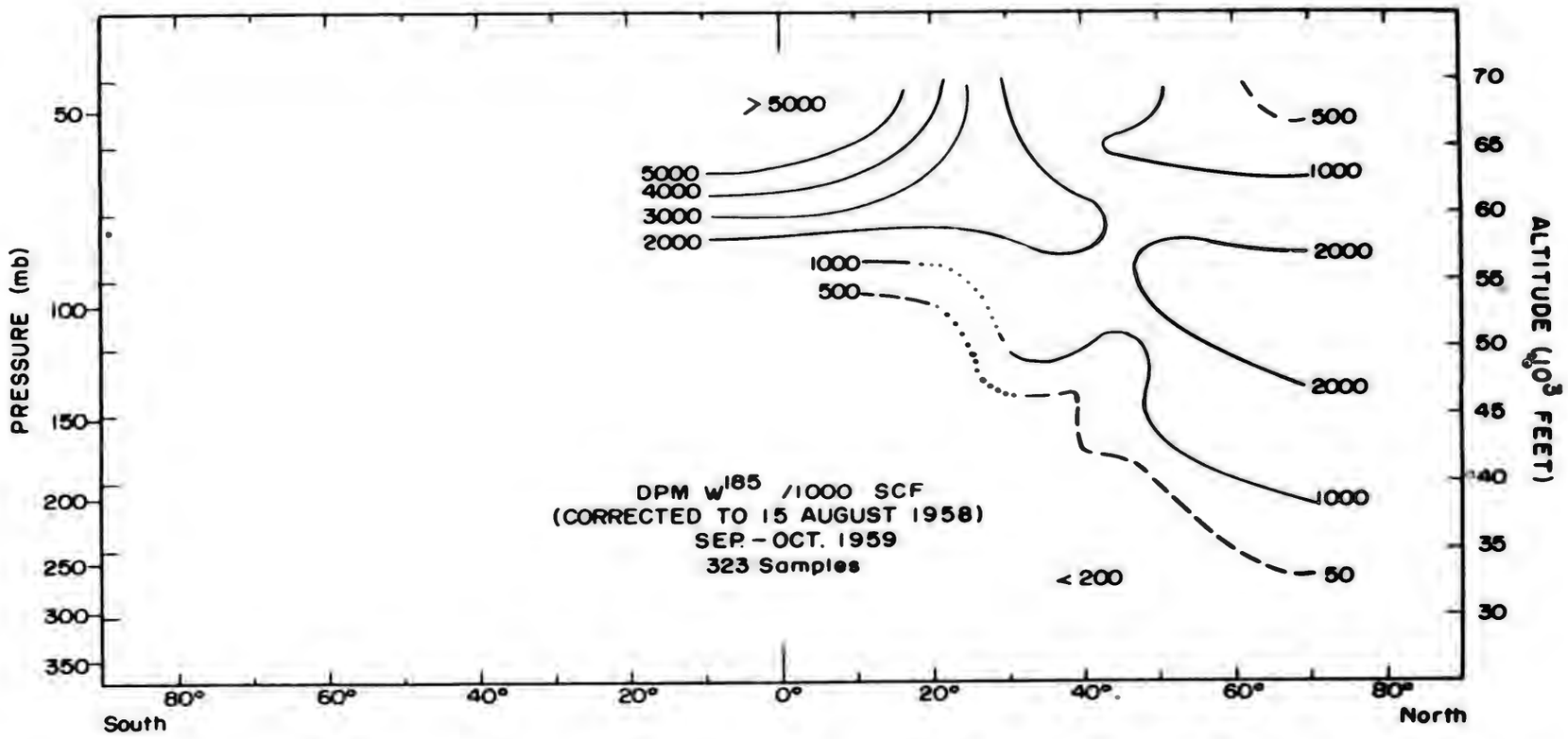


FIGURE 7 MEAN DISTRIBUTION OF TUNGSTEN - SEPTEMBER - OCTOBER 1959

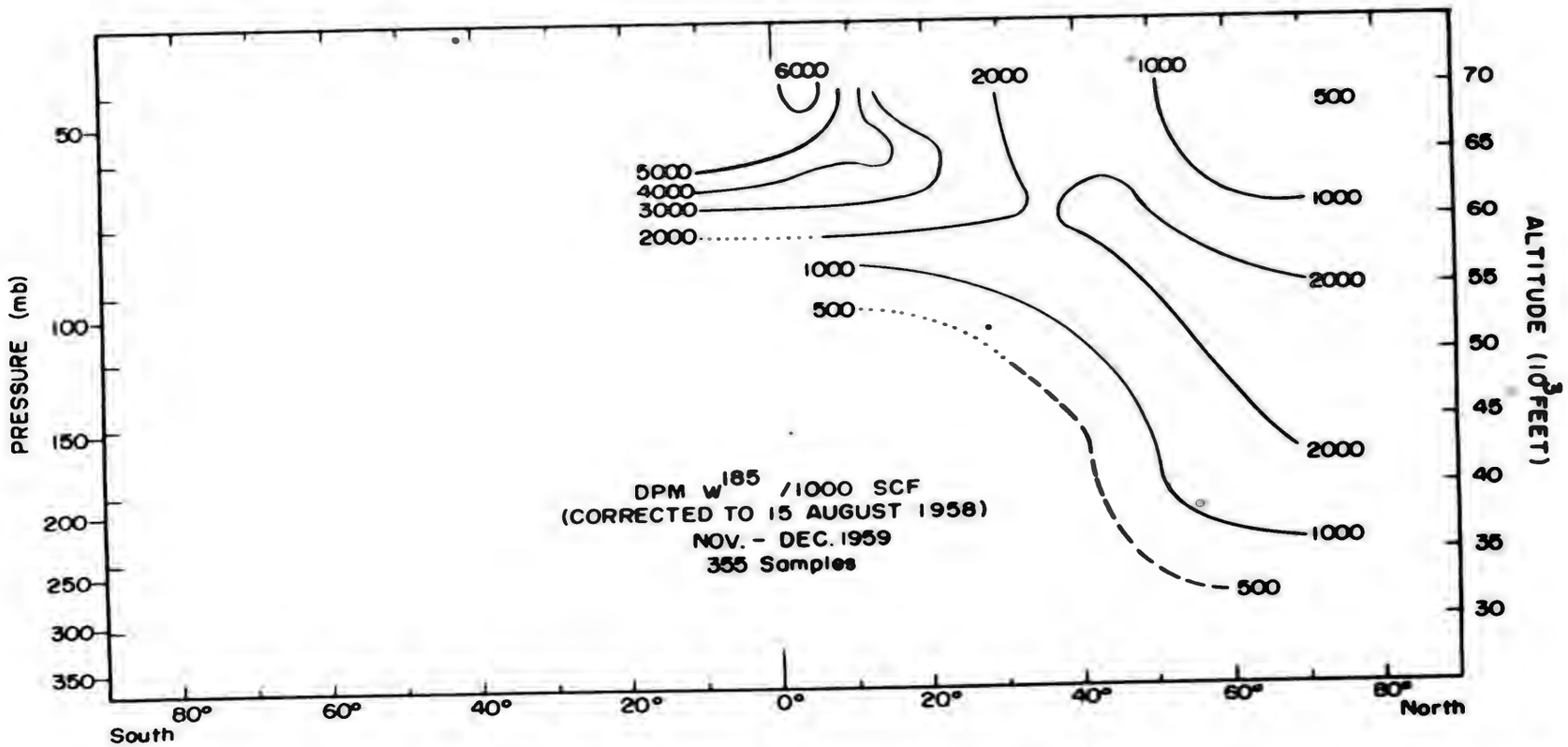


FIGURE 8 MEAN DISTRIBUTION OF TUNGSTEN-185, NOVEMBER-DECEMBER 1959

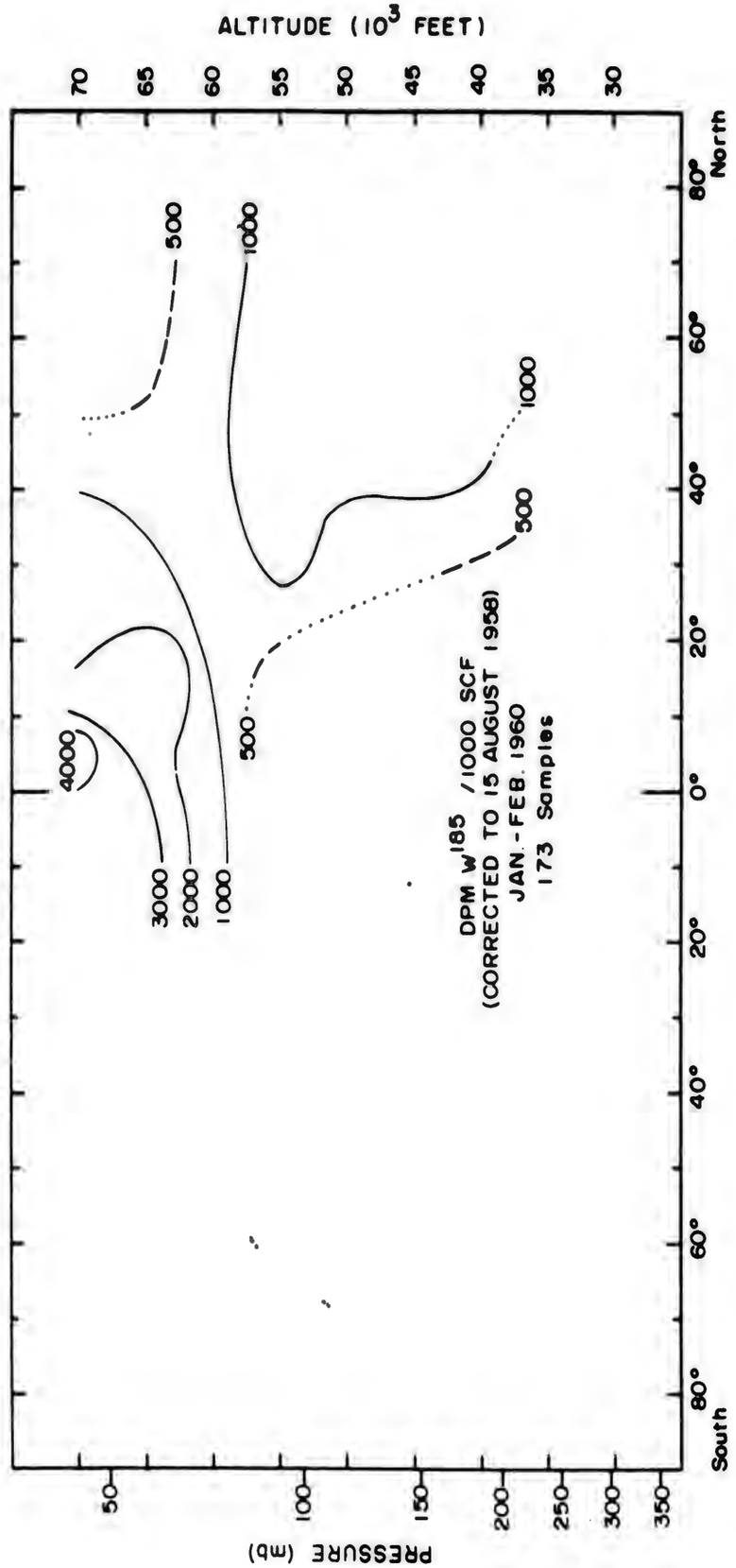


FIGURE 9 MEAN DISTRIBUTION OF TUNGSTEN-185, JANUARY - FEBRUARY 1960

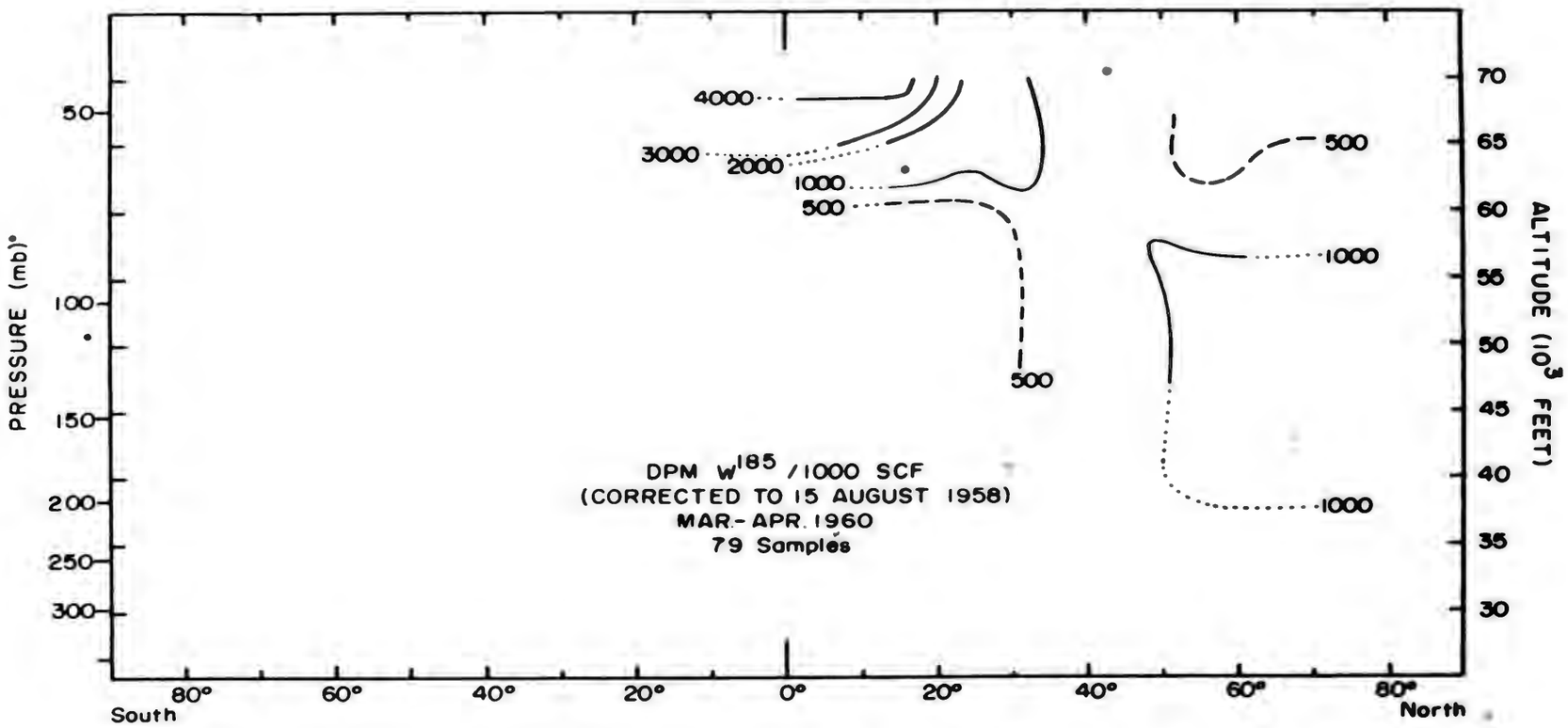


FIGURE 10 MEAN DISTRIBUTION OF TUNGSTEN-185, MARCH-APRIL 1960

region at altitudes above 60,000 feet. Thus the tungsten-185 in the lower stratosphere of the polar regions has spread there from the tropical injection site without passing through the higher layers of the polar stratosphere. In the polar stratosphere it has undergone vertical mixing but the rate of upward mixing has been sufficiently slow compared to the rates of horizontal mixing that the zone of high concentrations in the lower stratosphere has persisted. This indicates that any hypothesized meridional circulation through this region of the stratosphere has operated too slowly to offset the effects of turbulent mixing and casts doubt on the existence of such a circulation. It also shows, however, that horizontal mixing occurs within layers more or less parallel to the tropopause and that this mixing can produce rather rapid diffusion of injected debris through a wide latitude band. Thus by September 1958 quite significant quantities of tungsten-185 had spread into both the northern and southern polar regions. Because these mixing surfaces slope toward the poles it is possible for air from the high tropical stratosphere to be carried into the lower polar stratosphere by meridional mixing. Such mixing can be used to explain the observed appearance of ozone, which is produced in the high tropical stratosphere, in the polar regions during each spring, for an increase occurs in the rates of meridional mixing during the winter. This theory also explains why air in the lower polar stratosphere, which mixes readily with the very cold lower tropical stratosphere, as well as air in the high stratosphere, which once passed through some portion of the lower stratosphere, has a very low water vapor pressure.

In Figure 11 are meridional profiles of the activities in the zone of maximum tungsten-185 concentration for each of five bimonthly intervals. To prepare these curves the data for the altitude with the highest tungsten-185 concentration was selected for each 5 degree latitude band for each bimonthly interval. The curves commonly show a minimum in the middle latitudes of each hemisphere. These are probably attributable to

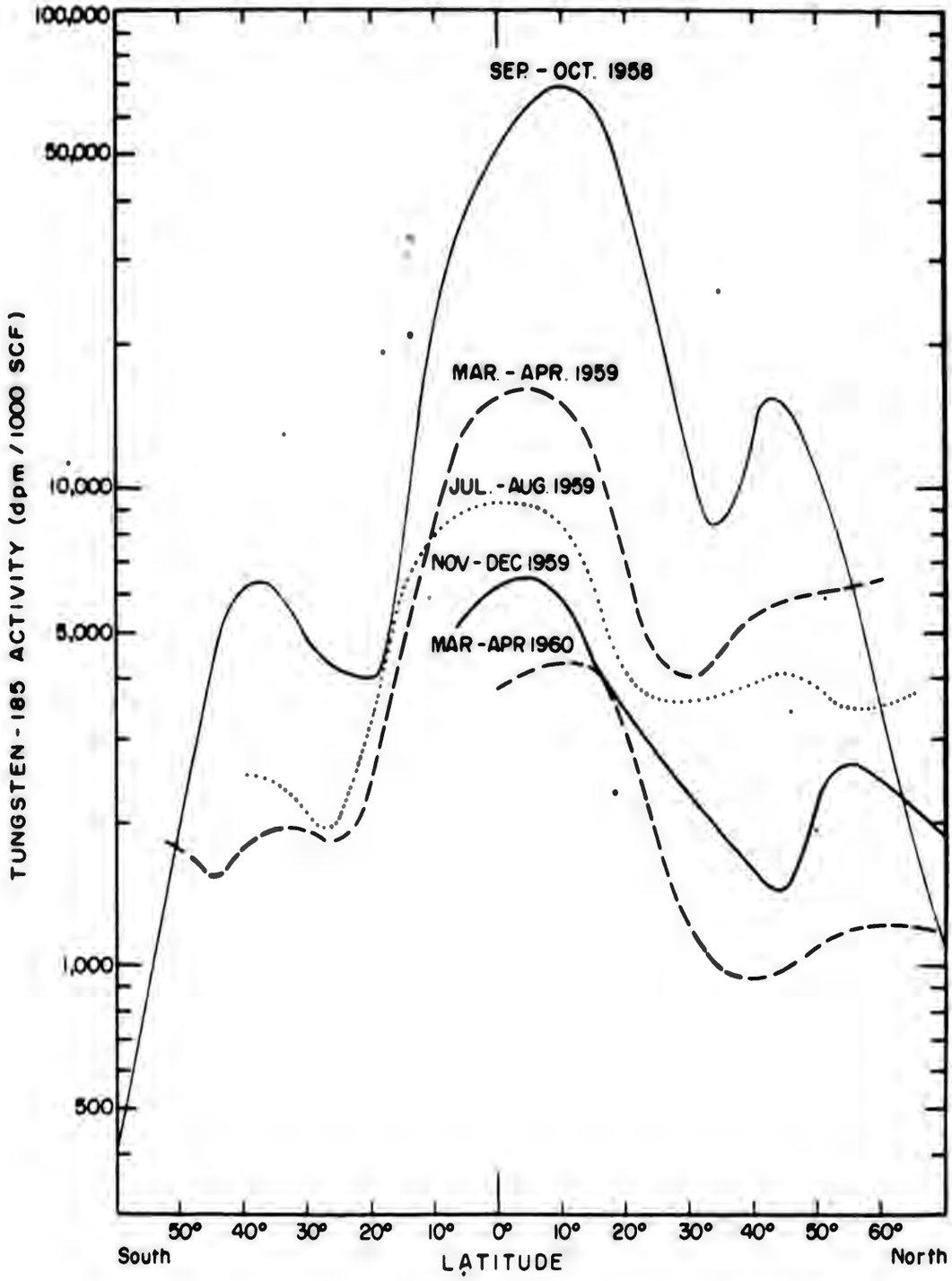


FIGURE 11 TUNGSTEN-185 ACTIVITIES (Corrected to 15 AUG. 1958) IN THE ZONE OF MAXIMUM TUNGSTEN-185 CONCENTRATION

mixing of stratospheric and tropospheric air in the vicinity of the tropopause gap. Evidently, atmospheric conditions under which mixing occurs between the lower tropical and lower polar stratospheres alternate with conditions under which the lower polar stratosphere mixes with the upper tropical troposphere.

In Figure 12 are plotted the highest tungsten-185 values found in the tropical and in the northern and southern polar stratospheres during each bimonthly interval. (Each of these numbers is the average for all samples collected within a particular 10 degree latitude band and 5000 foot altitude layer during the bimonthly intervals. The limits of the tropical stratosphere were taken as 30° North and 30° South.) The residence half time in the tropical stratosphere appeared to be about 5 months (as a minimum) during this time. The ratio of the maximum activities in the tropical to these in the northern polar stratosphere during more than a year remained close to 2. This suggests a uniform rate of mixing between these two regions regardless of season. This is in disagreement with other evidence which suggests an increase in the mixing rate during the winter. The ratio between the maximum concentrations in the northern and southern polar stratospheres passed through a maximum in January-February 1959. This is consistent with a maximum in the tropical-polar mixing rate in the winter and a minimum in the summer. Of course, any conclusion based on these "maximum concentrations" is suspect because of the arbitrary manner in which these values were chosen. It would appear from the data that the mixing phenomena are rather complicated, however, and until they are understood any selection of data will be arbitrary.

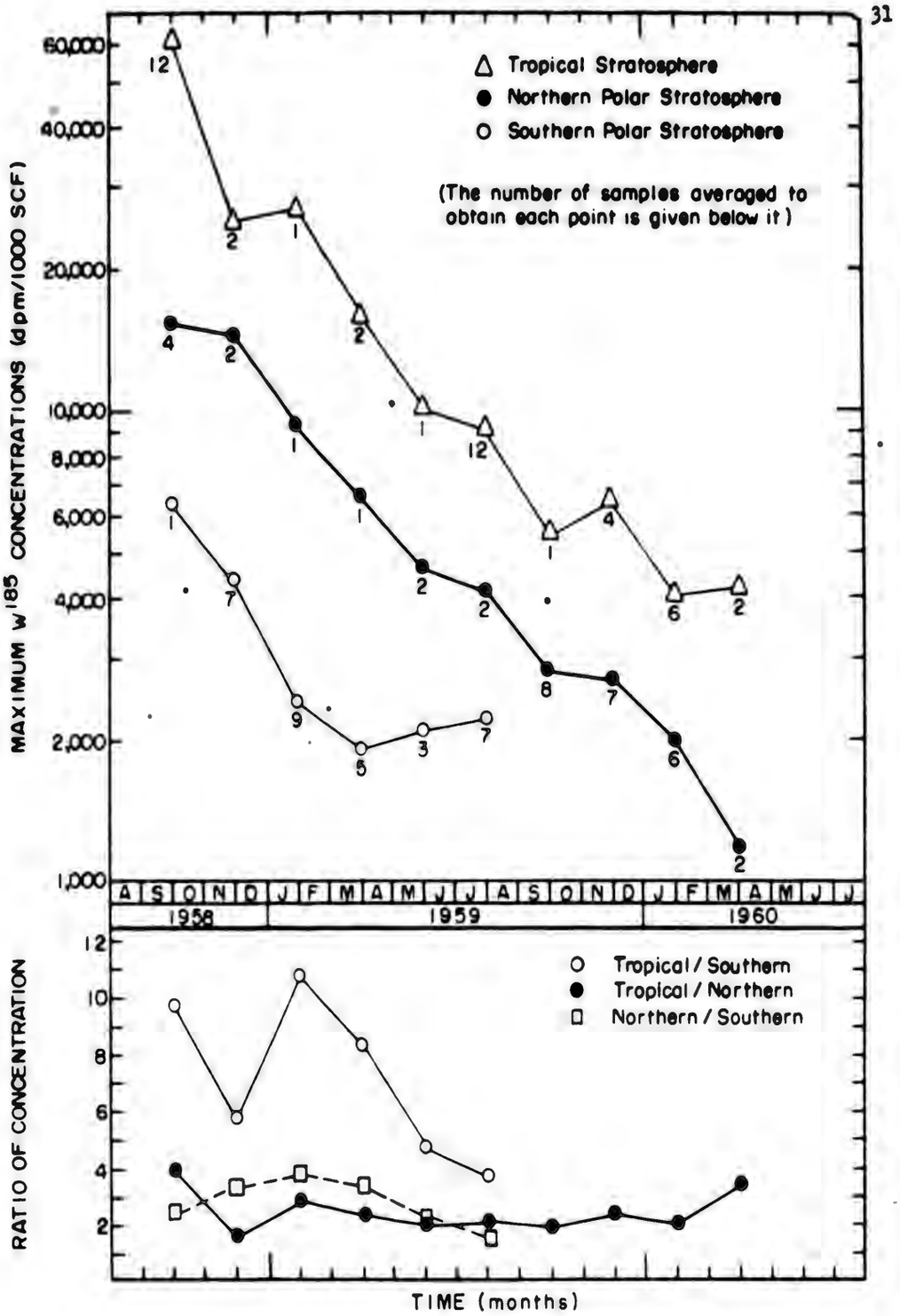


FIGURE 12 MAXIMUM CONCENTRATIONS OF TUNGSTEN-185 (corrected to 15 August 1958) FOUND IN THE STRATOSPHERE DURING EACH TWO MONTH SAMPLING INTERVAL

The Distribution of Rhodium-102 in the Stratosphere

The injection of "about 3 megacuries" of rhodium-102 into the high atmosphere (above 100,000 feet by a megaton yield nuclear weapon detonated in a rocket over Johnston Island in August 1958 provided a tracer for the behavior of high altitude debris. It seems reasonable that the mixing and fallout rates of this debris represents a limit on the rates for debris from surface bursts. Thus the stratospheric residence time of debris from Castle, Redwing and Hardtack should be shorter than, or at least no longer than, those indicated by the rhodium-102.

According to data obtained by Kalkstein at AFCRC through the analysis of filter samples of stratospheric air, the stratospheric concentrations of rhodium-102 in the Northern Hemisphere increased during the autumn of 1959 to values above those attributable to injections of rhodium-102 by Hardtack surface bursts. Measurements of stratospheric air in the Southern Hemisphere by Kalkstein suggested that similar increase may have occurred there in the autumn (May, June) or winter (July, August) of 1959 and that by the autumn (June) of 1960 the rhodium-102 concentrations in the Southern Hemisphere equalled those in the Northern Hemisphere.

A number of HASP samples collected during the winter of 1959-1960 in the Northern Hemisphere have been analyzed for rhodium-102. The mean concentrations indicated by these samples, arranged according to the 5000 foot altitude layer and according to the region (south of Ramey, south of Laughlin, between Laughlin and Minot, or north of Minot) in which they were collected, are given in Figure 13. It is evident that the highest concentrations occur at the highest latitudes and highest altitudes. This latitudinal distribution of activities is confirmed by the AFCRC data.

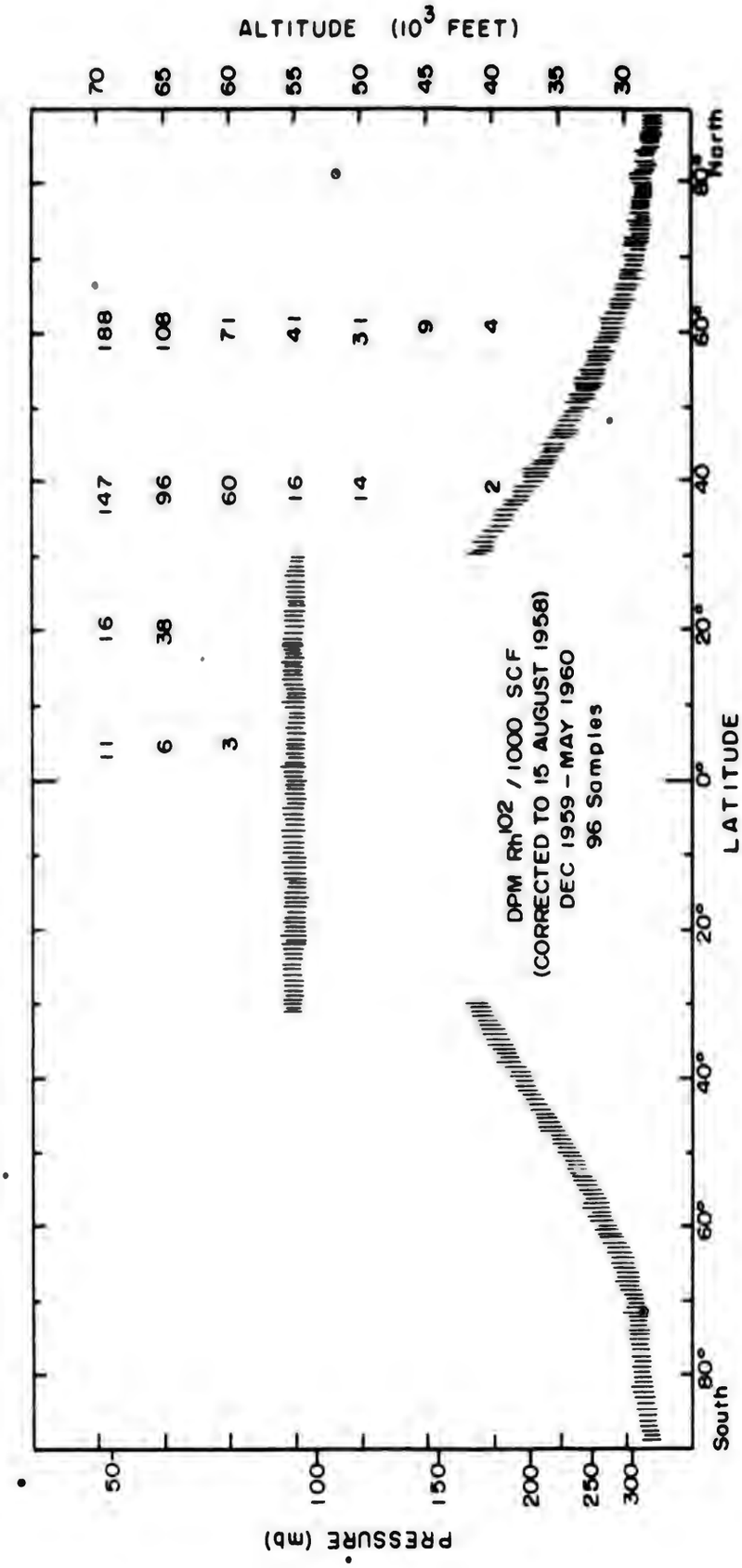


FIGURE 13 MEAN DISTRIBUTION OF RHODIUM - 102, DECEMBER 1959 - MAY 1960

It is evident that the rhodium-102 is being brought down from its original injection site by strong vertical mixing in the polar stratosphere during the "winter" season (late autumn to early spring). The vertical concentration gradient suggests that vertical mixing rather than subsidence of air is the main factor in the appearance of rhodium-102 below 70,000 feet. The rhodium-102 in the tropical stratosphere probably reached there through downward mixing into the polar stratosphere and then lateral mixing into the tropical stratosphere. The total amount of rhodium-102 in the stratosphere of the Northern Hemisphere during the winter of 1959-1960 was about 0.23 megacurie (corrected for decay to August 1958). Thus during the second year after its injection at high altitude about 8 percent of the rhodium-102 activity was carried down into the lower stratosphere. Quite probably it required a year following the injection of the rhodium for large quantities to reach the top of the stratosphere by particle settling and then mixing through the ozone layer. If this is so, the rate of transfer of rhodium-102 into the lower stratosphere (and into the troposphere) could increase rather than decrease during this coming year.

It may be noted, on the basis of the rhodium-102 data that if Teak and Orange, which are described as "megaton" weapons, each injected 0.1 megacurie of strontium-90 into the high atmosphere, the amount of strontium-90 from these weapons which had reached the lower stratosphere by mid-1960 was probably less than 0.02 megacurie.

Unfortunately, there are several possible errors in the rhodium-102 data. The exact yield of rhodium in the high altitude shot is not known, the branching ratio of rhodium-102 is not known exactly, and Kalkstein has discovered the presence of an apparent long lived isomer of rhodium-102 which introduced uncertainties in the correction of rhodium-102 measurements for decay. In addition some recent HASP measurements of rhodium-102 have been in error because of deterioration of the scintillation crystal used to count them. Some worry has also existed that rhodium might be lost from HASP

samples during the ashing procedure. Theoretically none should be lost but until now it was not possible to check this experimentally. A "hot" spike has been obtained recently, however, and this potential loss is being checked.

The Distribution of Beryllium-7 in the Stratosphere

A number of HASP samples, collected during the winter of 1959-1960, has been analyzed for beryllium-7, a product of the interaction of cosmic rays with the atmosphere. The same samples that were analyzed for rhodium-102 were also analyzed for beryllium-7. The mean concentrations, with data arranged according to the latitude and altitude of collection of the samples in the same manner as are the rhodium-102 data in Figure 13, are given in Figure 14, expressed in dpm Be^7/1000 SCF, and in Figure 15, expressed in atoms of beryllium-7 per gram of air. The observed distribution of decay rates is similar to the distribution of production rates which was predicted, based on measurements of cosmic ray activities and on the calculations of the rate of production of beryllium-7 by cosmic rays. The differences between the observed concentrations and those which would be predicted if there was no mixing at all can be attributed to loss of beryllium-7 to the troposphere through the tropopause, and especially through the tropopause gap. The value of these data for detecting mixing within the stratosphere is limited by the tendency of isopleths of production rates to parallel the directions of high mixing rates (as derived from tungsten-185 data). As a result they cannot be used to prove that there is rapid meridional mixing in the stratosphere but they do put limits on the possible rates of vertical and horizontal mixing.

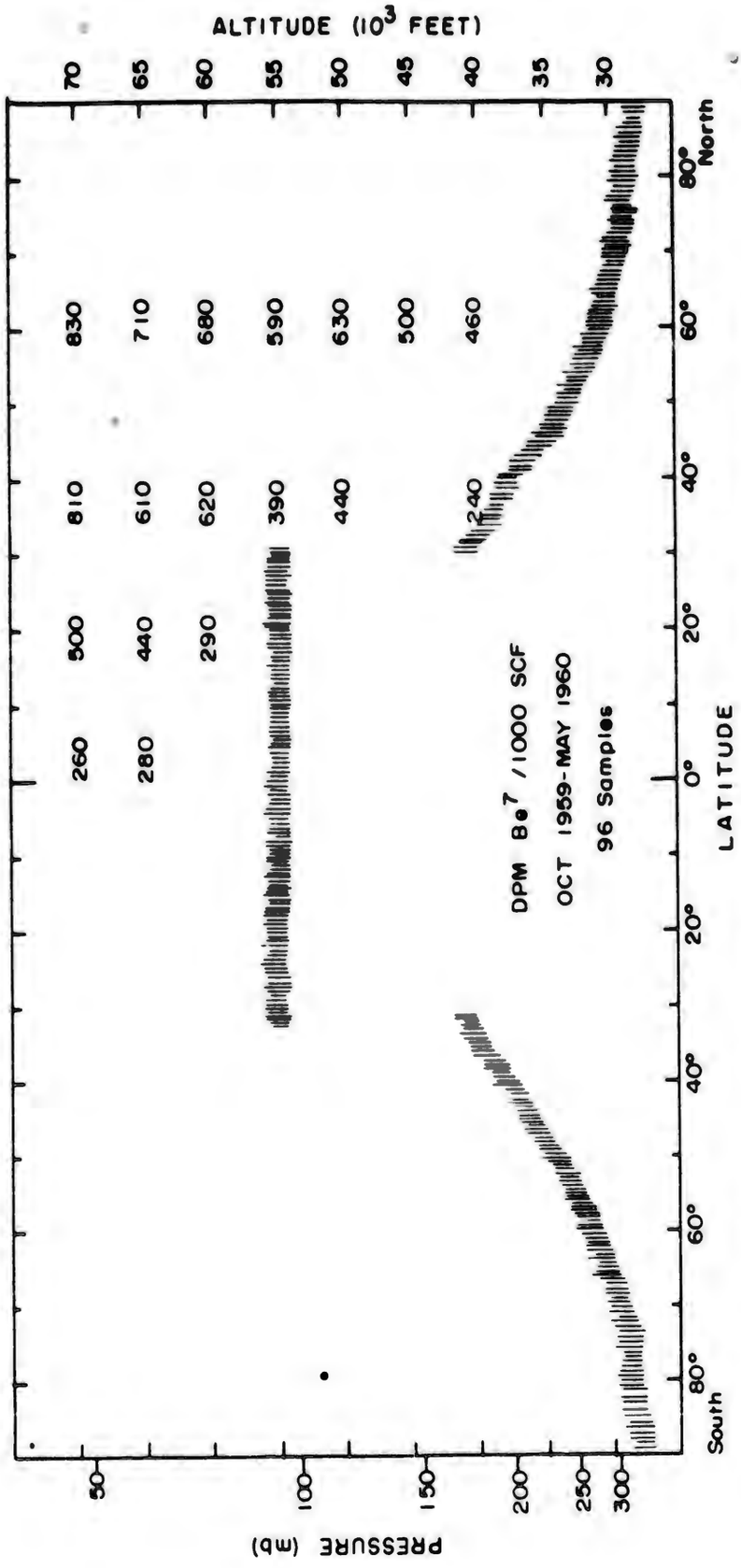


FIGURE 14 MEAN DISTRIBUTION OF BERYLLIUM-7, OCTOBER 1959 - MAY 1960

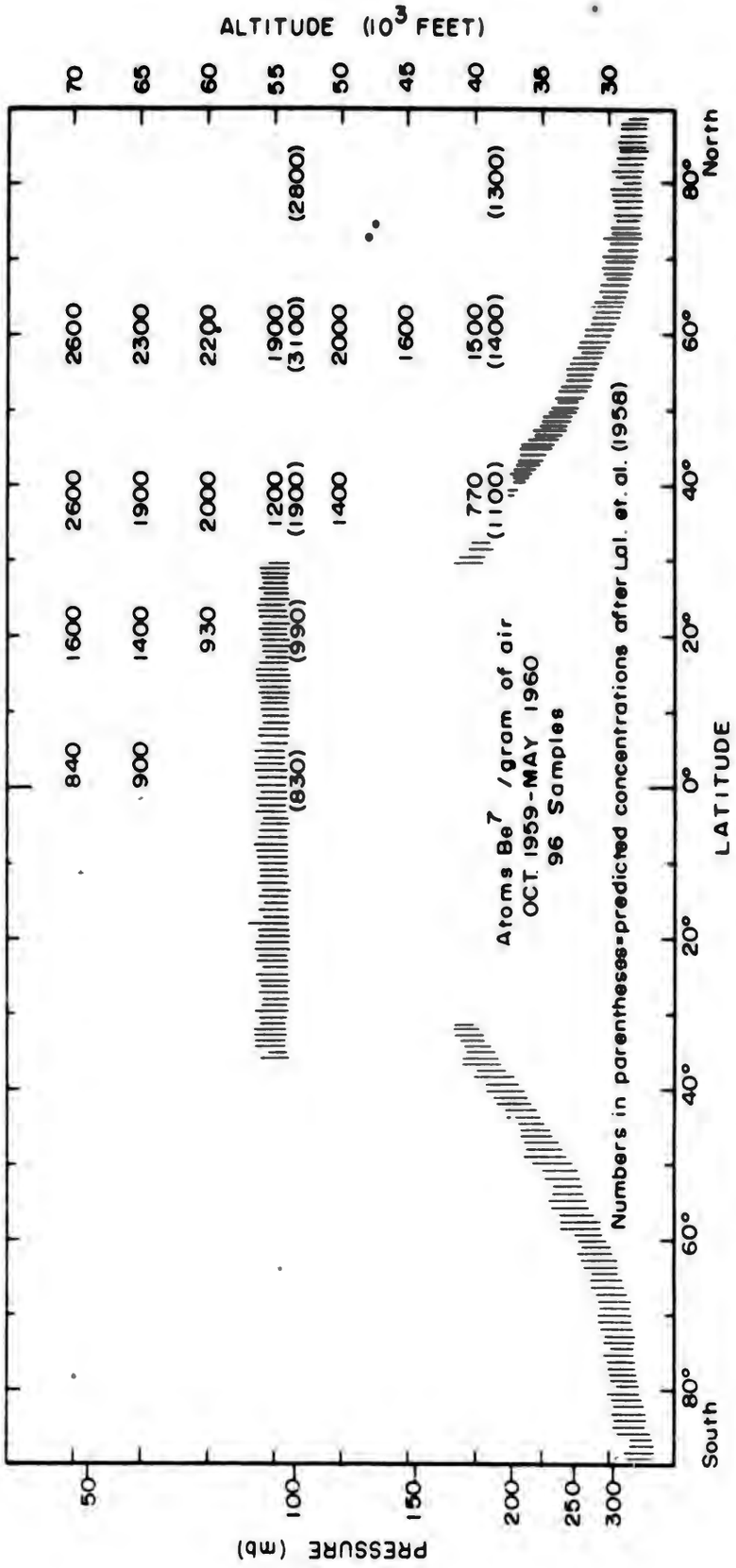


FIGURE 15 MEAN DISTRIBUTION OF BERYLLIUM – 7, OCTOBER 1959 – MAY 1960

The Distribution of Strontium-89 in the Stratosphere

The mean strontium-89 - strontium-90 ratio has been calculated, for each month for which there are data, for each of three regions of the stratosphere: the tropical stratosphere (30° North - 30° South), the northern polar stratosphere (north of 30° North) and the southern polar stratosphere (south of 30° South). For the tropical stratosphere (Figure 16) only samples collected at 60,000 to 70,000 feet were used. For the northern polar (Figure 17) and southern polar stratosphere (Figure 18) the data were divided into three sets: one for samples collected at 30,000 or 35,000 feet, a second for samples collected at 50,000 to 55,000 feet, and a third for samples collected at 60,000 to 70,000 feet. A curve has been drawn through the points for the tropical stratosphere in Figure 16 and this curve has been repeated in Figure 17 and Figure 18 for purposes of comparison.

Between November 1958 and September 1959 the ratio in debris in the tropical stratosphere fell in response to the decay of strontium-89. The apparent date of origin of this debris, obtained by extrapolating the line back to a ratio of 180, is July 1958, in good agreement with its derivation from Hardtack. After September 1959 the ratio ceased to fall, either because of the influx of younger material from some other region of the stratosphere or, more likely, because the strontium-89 could not be measured accurately in the low concentrations present in the tropical stratosphere in late 1959.

By January 1959, the debris sampled in the northern polar stratosphere had the same apparent age as that sampled in the tropical stratosphere. This correspondence in apparent age between the polar and tropical stratosphere continued until at least September 1959. The only exception was found in data for samples collected at 30,000 to 35,000 feet during April 1960 (Sea Fish Special No. 3). These gave a higher Sr^{89}/Sr^{90} ratio than samples collected at higher altitudes.

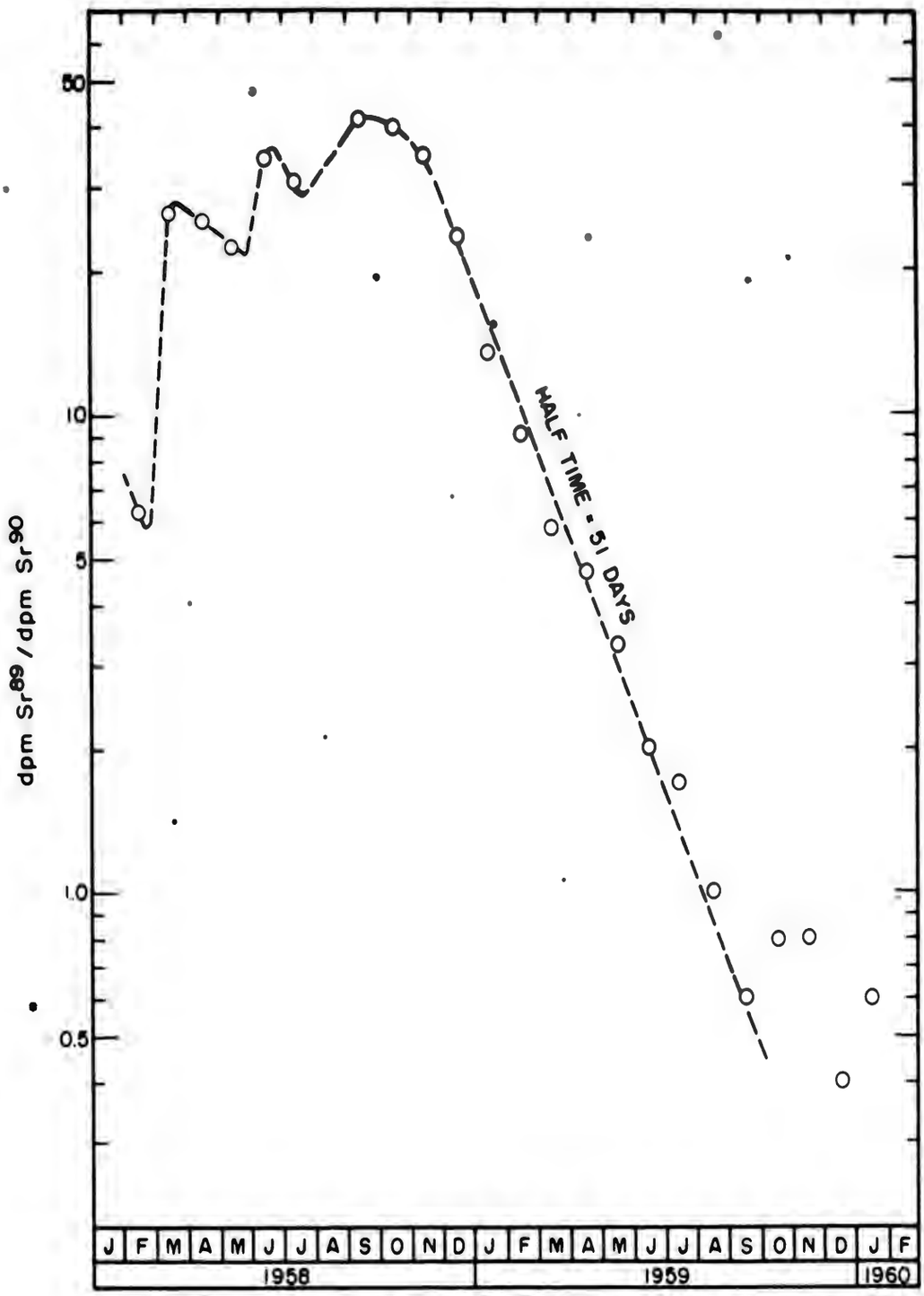


FIGURE 16 VARIATION WITH TIME OF THE MONTHLY AVERAGE FOR Sr^{89}/Sr^{90} IN SAMPLES COLLECTED BETWEEN 60,000 AND 70,000 FEET IN THE TROPICAL STRATOSPHERE (30°N - 30°S)

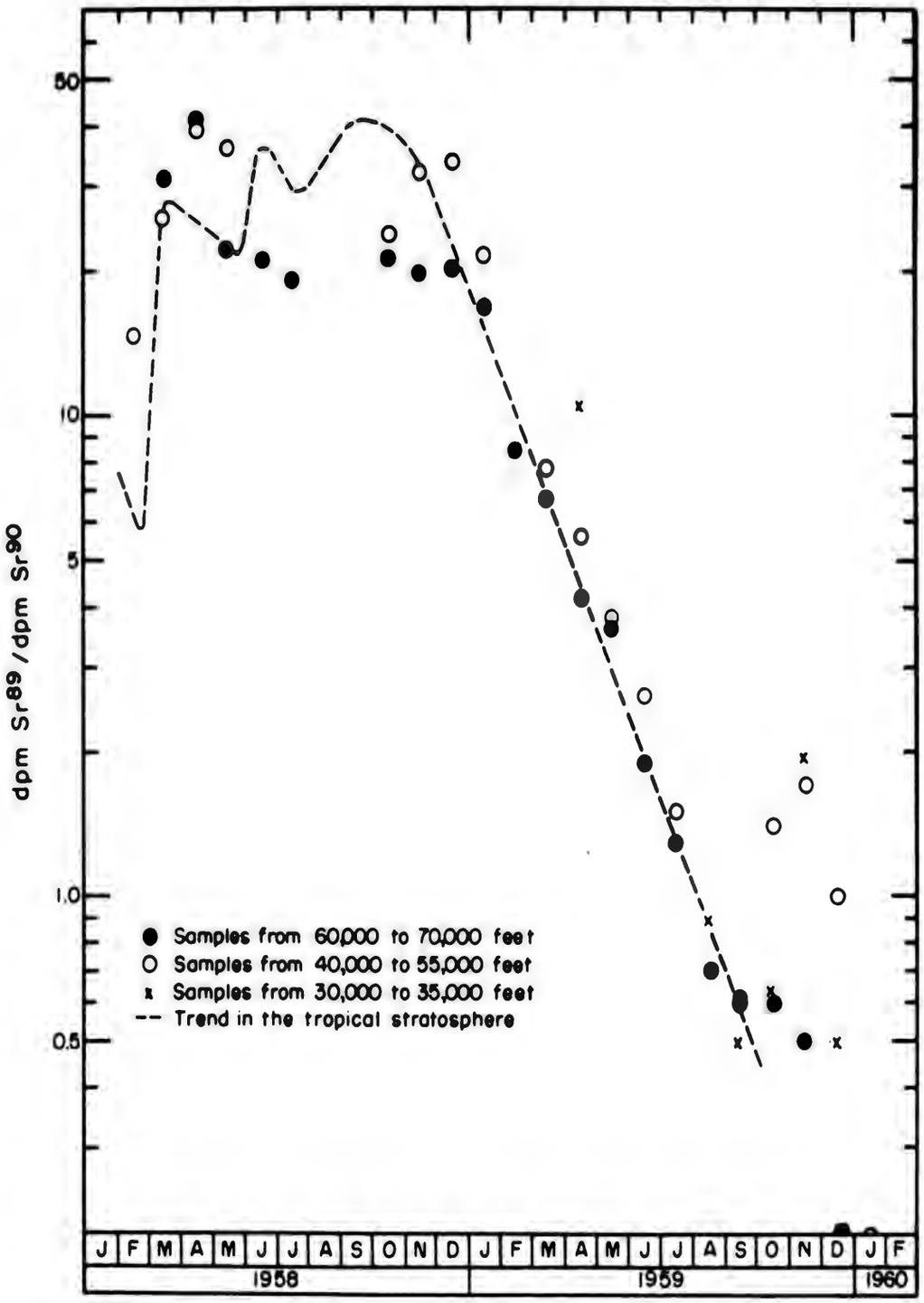


FIGURE 17 VARIATION WITH TIME OF THE MONTHLY AVERAGE FOR Sr^{89}/Sr^{90} IN THE SAMPLES COLLECTED IN THE NORTHERN POLAR STRATOSPHERE (30°N - 90°N)

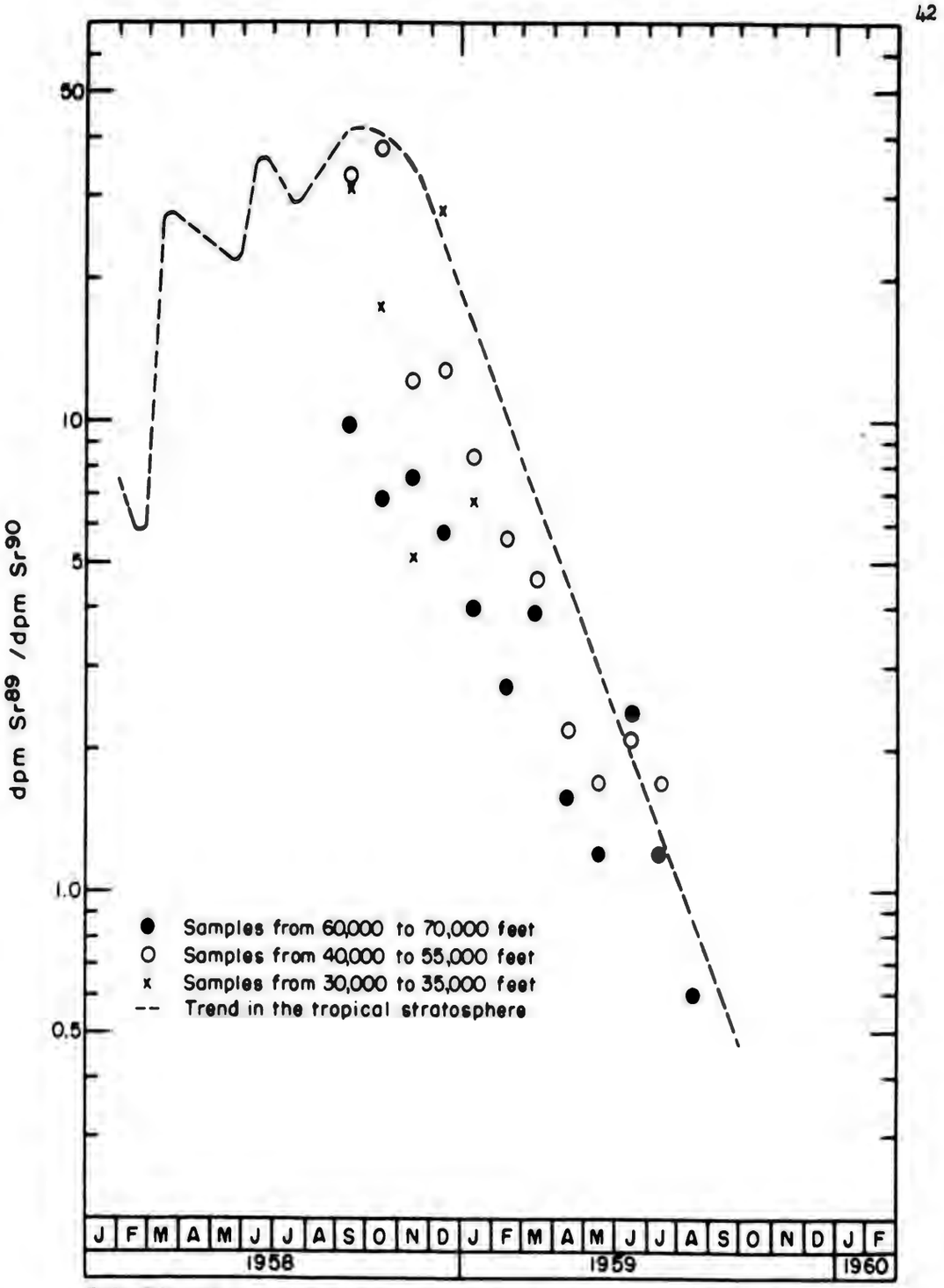


FIGURE 18 VARIATION WITH TIME OF THE MONTHLY AVERAGE FOR Sr^{89}/Sr^{90} IN SAMPLES COLLECTED IN THE SOUTHERN POLAR STRATOSPHERE (30°S - 60°S)

There are two explanations for the apparent age of the debris in the northern polar stratosphere. The first is that it represents a mixture of debris from the October 1958 Soviet tests with older debris. If it is assumed that the older debris contained no strontium-89, the Soviet debris contributed about 27 percent of the strontium-90 in the polar stratosphere. If the old debris did contain strontium-89, less than 27 percent of the strontium-90 came from the October 1958 Soviet tests. The second explanation for the apparent age assumes that virtually all debris from Soviet tests had fallen out, or at least had not been mixed into the part of the stratosphere which HASP was sampling, and that the apparent age of the debris was caused by the transfer of large quantities of debris from Hardtack into the polar stratosphere. Perhaps some Soviet debris remained in the very low stratosphere below 40,000 feet but if it did it was certainly present even there in only low concentrations.

The change in Sr^{89}/Sr^{90} ratio which occurred in the northern polar stratosphere in October 1959 is similar to that which occurred in the tropical stratosphere at the same time. Again it seems more likely that the failure of the ratio to continue falling is due to analytical error rather than to the influx of young material, especially since the ratio for debris from 40,000 to 55,000 became higher than that for debris from 60,000 to 70,000 feet and the only likely source of younger material lay above 70,000 feet.

The Sr^{89}/Sr^{90} ratio in debris in the southern polar stratosphere was lower than that in debris in the tropical and northern polar stratosphere throughout early 1959. However, the ratio increased steadily during this interval and by mid-1959 it was the same (or almost the same) in the southern polar stratosphere as it was in the tropical stratosphere. Even in September and October 1958 the apparent age of debris in the Southern Hemisphere was only two months older, for debris at 40,000 to 55,000 feet,

or five months older, for debris at 60,000 to 70,000 feet, than that of debris in the tropical stratosphere. The variation with altitude of the apparent age of this debris suggests that either Hardtack debris mixed preferentially into the low southern polar stratosphere or that Hardtack debris from higher altitudes mixed with more older debris (either in the tropical or in the southern polar stratosphere) than did Hardtack debris from lower altitudes.

The Distribution of Cerium-144 in the Stratosphere

The mean cerium-144 - strontium-90 ratio has been calculated, for each month for which there are data, for each of three regions of the stratosphere, the tropical, northern polar and southern polar stratosphere. The data for this ratio are plotted in Figures 19, 20 and 21 in much the same way as Sr^{89}/Sr^{90} data are plotted in the preceeding figures.

For samples collected in the tropical stratosphere (Figure 19) since September 1958, the data fit the theoretical decay curve for this ratio quite satisfactorily. For samples collected in the northern polar stratosphere during this same interval the data show more scatter and there is a distinct difference between the ratio in samples collected at 55,000 feet and below and that in samples collected at 60,000 feet and above. The data for samples collected in the southern polar stratosphere show even more scatter though there is no obvious altitude effect. In all regions of the stratosphere which were sampled the apparent age of the debris is older according to this ratio than to the Sr^{89}/Sr^{90} ratio, probably because the bulk of the debris present in these regions was derived from United States ground shots in the Pacific which produced fractionation of strontium-90 relative to cerium-144. Only two important sources exist which could contribute debris with a Ce^{144}/Sr^{90} ratio higher than that found in debris in the tropical stratosphere; material from the October 1958 Soviet and material from the August 1958 United States high altitude rocket shots. The ratio should be higher in debris from these sources since they probably produced no fractionation.

The debris present at 40,000 to 55,000 feet in the northern polar stratosphere at the beginning of 1959 had a Ce^{144}/Sr^{90} ratio slightly higher than that in the tropical stratosphere, probably because of the admixture of material from the October 1958 Soviet tests. If it is assumed that this Soviet material was mixed with very old

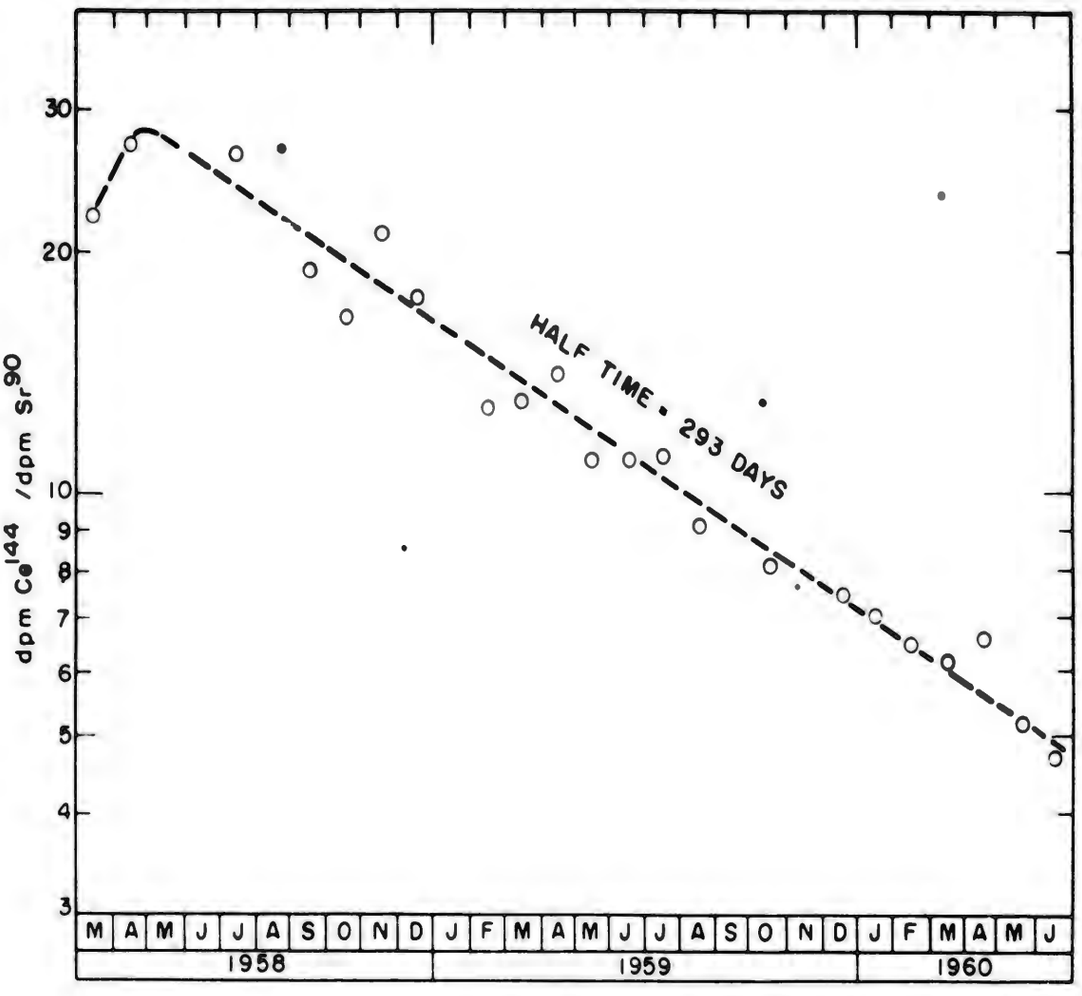


FIGURE 19 VARIATION WITH TIME OF THE MONTHLY AVERAGE FOR Ce^{144}/Sr^{90} IN SAMPLES COLLECTED BETWEEN 60,000 AND 70,000 FEET IN THE TROPICAL STRATOSPHERE (30° N - 30° S)

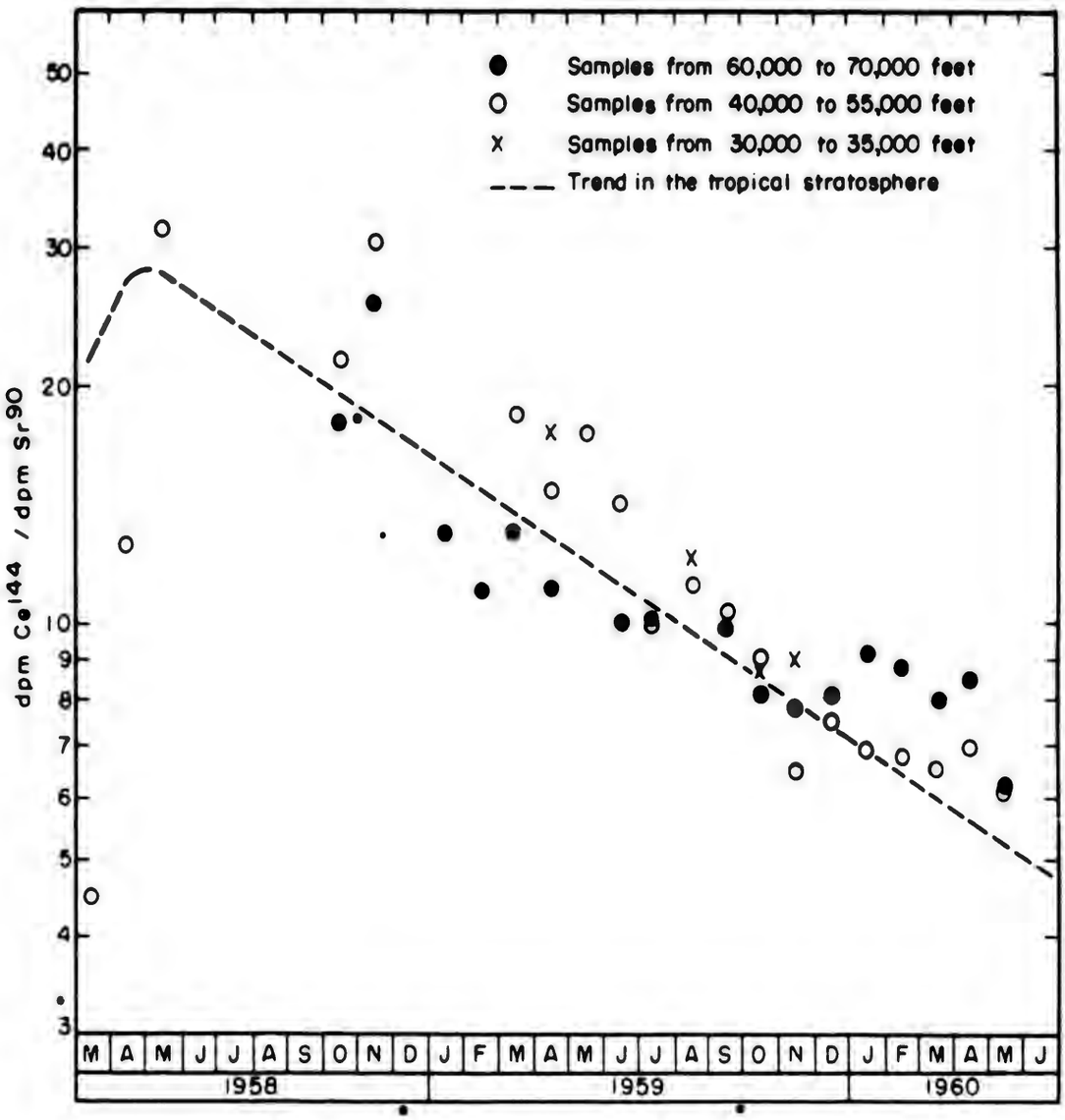


FIGURE 20 VARIATION WITH TIME OF THE MONTHLY AVERAGE FOR Ce^{144} / Sr^{90} IN SAMPLES COLLECTED IN THE NORTHERN POLAR STRATOSPHERE (30°N-90°N)

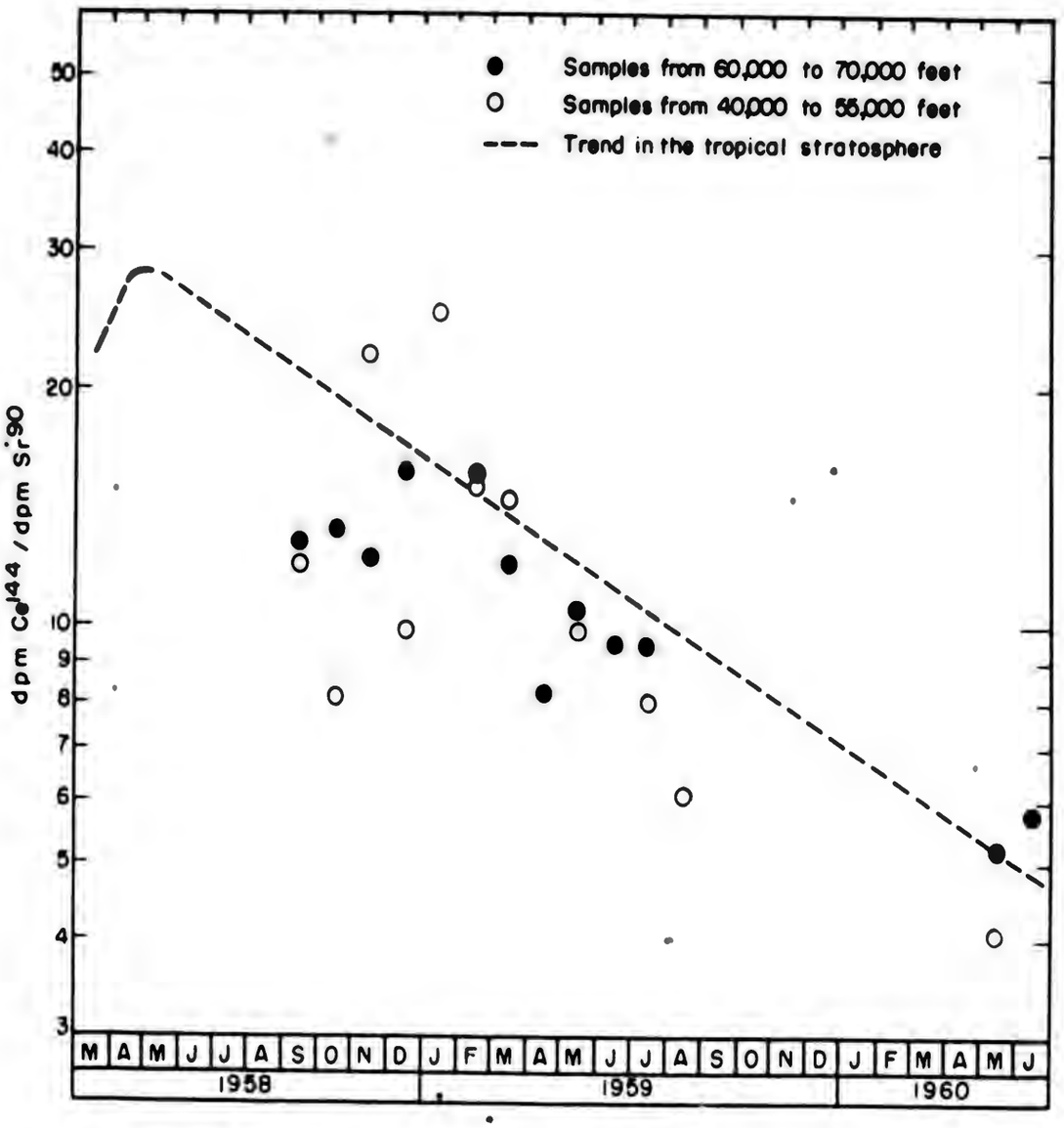


FIGURE 21 VARIATION WITH TIME OF THE MONTHLY AVERAGE FOR Ce^{144} / Sr^{90} IN SAMPLES COLLECTED IN THE SOUTHERN POLAR STRATOSPHERE (30°S-60°S)

debris which contained almost no cerium-144, the Soviet contribution may have been almost 50 percent. If, as is more likely, the Soviet material was mixed with debris which had the same nuclide ratios found in the tropical stratosphere, the Soviet contribution was less than 20 percent. The debris present at 60,000 to 70,000 feet in the same region at the same time had a slightly lower Ce^{144}/Sr^{90} ratio than was found in the tropical stratosphere, probably because of a small contribution of older material (from Castle, Redwing, etc.).

By the spring of 1960 the Ce^{144}/Sr^{90} ratio was slightly higher at 60,000 to 70,000 feet than it was at 40,000 to 55,000 feet and all debris in the northern polar stratosphere had a higher ratio than that in the tropical stratosphere. This was most likely due to the transport of debris from Teak and Orange into this region during the winter of 1959-1960. The concentration of strontium-90 from these rocket shots which was present between 60,000 and 70,000 feet by early 1960 may be calculated, by assuming a Rh^{102} injection of about 3 megacuries and a total strontium-90 injection of about 0.2 megacurie for Teak and Orange, to be about 7 dpm/1000 SCF, or about 3.5 percent of the total strontium-90 concentration. Using the cerium-144 data and assuming that debris from Teak and Orange, unmixed with any older debris, was added to debris with the same Ce^{144}/Sr^{90} ratio as was present in the tropical stratosphere, it may be calculated that between 25 and 30 percent of the debris above 60,000 feet in the polar stratosphere in March 1960 came from the rocket shots. The true strontium-90 contribution from the rocket shots probably lay between 3.5 and 30 percent.

The tendency for Ce^{144}/Sr^{90} ratios in the southern polar stratosphere to be less than the ratios in the tropical stratosphere is doubtless due to the mixture of Hardtack debris with older debris as it was transported southward. To explain the ratios found in the southern polar stratosphere we could assume that by mid-1959 about 85 percent of the debris present in the southern polar stratosphere had been derived from the lower tropical stratosphere (primarily from Hardtack injections) and that the remaining approximately

15 percent was from debris so old that the cerium-144 concentration was negligible. By June 1960 mixing between the tropical and southern polar stratosphere had apparently been sufficient to give a uniform Ce^{144}/Sr^{90} ratio through both regions. Actually, there may have been a significant proportion of old material still present in the southern polar stratosphere but its effect on the Ce^{144}/Sr^{90} ratio may have been offset by the presence of debris from Teak and Orange.

Conclusions Based on Data for Phase 4 and Phase 5

The data obtained from Phase 4 of the sampling program have served to delineate the distribution of nuclear debris through the stratosphere during late 1959 and to indicate the changes which occurred in this distribution during the winter of 1959-1960. The data obtained from Phase 5 give some idea of the changes which had occurred in the distribution of debris through the stratosphere of the Southern Hemisphere during the interval August 1959- May 1960.

A number of conclusions may be drawn from these data including the following:

(1) Since the spring of 1959 the distribution of nuclear debris through the stratosphere has changed only very slowly as evidenced by the strontium-90 data. The net effect of those changes which have occurred has been a decrease in concentrations in the lower tropical stratosphere, little or no change in the concentrations in the lower northern polar stratosphere (i.e. below 70,000 feet) and an increase in the concentrations in the southern polar stratosphere (at least between 60,000 and 70,000 feet).

(2) Changes in the distribution of nuclear debris within the stratosphere appear to occur mainly as a result of turbulent mixing. It is evident that from the tungsten-185 data that, if there is a meridional circulation affecting stratospheric air, it must operate quite slowly compared to the two year interval which has passed since the injection during Hardtack of tungsten-185 into the stratosphere. Evidently mixing takes place parallel to concentration gradient and, since vertical mixing id quite slow in the stratosphere, it is the rate of vertical mixing which determines the stratospheric residence time of that debris which has been injected well above the tropopause.

(3) The changes which have occurred in the stratospheric distribution of debris appear to have resulted mainly from an increase in the rates of horizontal mixing between

the tropical stratosphere and the polar stratosphere and from increased rates of vertical mixing within the polar stratosphere during the winter season of each hemisphere. Both strontium-90 and tungsten-185 data bear this out. In the Southern Hemisphere these increased rates of mixing appear to have become manifest by May and to have extended through October. In the Northern Hemisphere there are indications that they have prevailed from about October through March. Thus each hemisphere appears to experience a "summer" season during which mixing coefficients are small and a "winter" season during which they increase. This is consistent with the existence of the "winter polar vortex" which has been observed in the stratosphere of the Northern Hemisphere during the winter season.

(4) The changes which have been observed in the distribution of debris in the polar stratosphere during the winter do not indicate an important role either for subsidence of air within the "winter vortex" or for subsidence of air as a result of early break-up of the vortex during the late winter. The changes which occur seem to reflect rapid mixing between the lower tropical stratosphere and the lower polar stratosphere along surfaces which slope downward toward the poles rather than the vertical displacement of polar air. A decrease in the vertical concentration gradient does occur within the polar stratosphere during the winter but this can be explained most reasonably by an increase in the coefficient of vertical mixing.

(5) Detectable quantities of debris from Orange shot, originally injected above 100 kilometers in the tropical stratosphere, had descended to below 70,000 feet in the northern polar stratosphere by (or during) the winter of 1959-1960. According to the Rh^{102} data obtained by Kalkstein at AFCRC the first debris clearly derived from the high altitude injection appeared in the lower stratosphere of the Northern Hemisphere during the autumn of 1959 and in that of the Southern Hemisphere either during the autumn of 1959 or during that of 1960. The cerium-144 concentrations in debris in the polar stratosphere by mid-1960 suggest that as much as thirty percent of the debris in this region may have been derived from Teak and Orange.

H. W. Feely
J. Spar

APPENDIX
SIZE DISTRIBUTION and COMPOSITION of STRATOSPHERIC PARTICLES

Samples of stratospheric particles collected by impactor probes show two general types of size distribution, one characteristic of tropical and the other of polar stratospheric air. The vast majority of particles with radii less than 1.5 microns were found, by electron diffraction measurements, to consist mostly of ammonium persulfate plus some ammonium sulfate. It was possible to correlate either the surface area concentration or the volume concentration with the strontium-90 concentration of the sampled air with a standard deviation of about 40%. Only tentative conclusions can be drawn about the physical meaning of the relationship of nuclear debris to stratospheric sulfate particles.

Data on stratospheric strontium-90 concentrations are used to perform an approximate calculation of the quantity and residence time of sulfate in the stratosphere.

Size analyses have been performed on samples of stratospheric particles which were collected at altitudes between 40,000 and 70,000 feet by U. S. Air Force U-2 aircraft during the course of the High Altitude Sampling Program. (1)

Impaction probes for collection of stratospheric particles were installed in two of the aircraft.

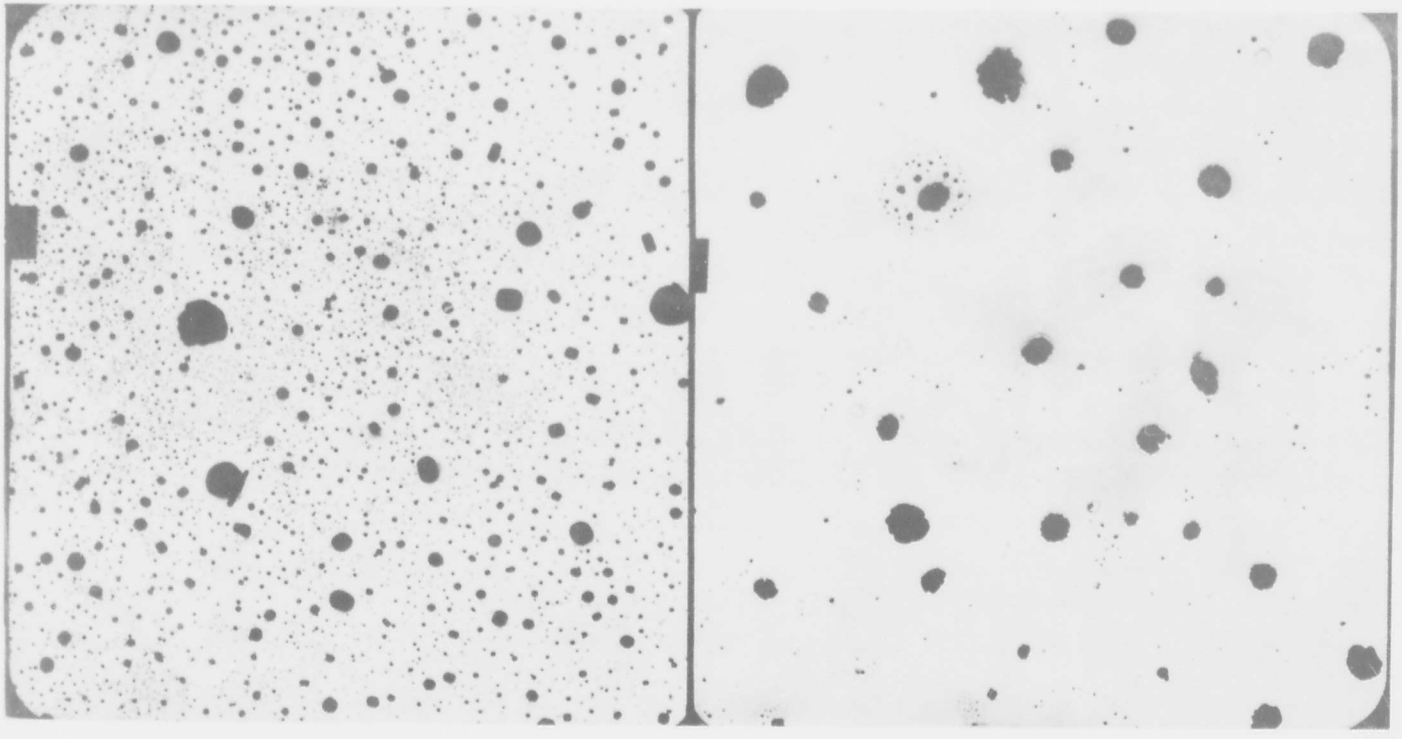
The probe, which was designed by Junge and co-workers at Air Force Cambridge Research Laboratories, has a diameter of 1.5 cm and an impaction surface with an area of 1.2 cm^2. Volumes of ambient air sampled ranged from (2.5 to 6) x 10^8 cm^3 during exposure periods of 3 to 7 hours. The impaction surfaces consisted of formvar films coated on 200 mesh gold screen. From each sample four 1/8 inch discs were punched for use as electron microscope specimens (2). Several electron micrographs of each specimen were made in an effort to ensure representativeness for particle size - frequency measurements. Electron micrographs of some typical stratospheric particles are shown in Figure 1.

Most of these particles, which have radii in the range from less than 0.10 micron to 1.5 microns, gave electron diffraction patterns corresponding to ammonium persulfate, $(NH_4)_2S_2O_8$. Some of the particles were shown to be ammonium sulfate, $(NH_4)_2 SO_4$, by measurement of crystal angles and by electron diffraction.

Electron Micrographs of Stratospheric Particles

Sample T-2 (Tropical)

Sample P-2 (Polar)



Note: Rectangular marks are 1 micron long

Figure 1

The sulfate and persulfate particles were hygroscopic and were found to be somewhat volatile in the electron beam. The identification of sulfate and persulfate as constituents of stratospheric particles represents an independent corroboration of the work of Junge and co-workers (3), who used electron microprobe analysis to determine the presence of sulfur in aerosol samples collected by means of high altitude balloon and aircraft flights.

Few of the particles with radii greater than 1.5 microns contain sulfate. Most of these large particles have a higher electron optical density and more irregular outlines than the sulfate particles. Occasionally spheres of high density with diameters smaller than one micron were found. It is possible that some of these non-sulfate particles are extra terrestrial in origin.

The particles as they appeared in the electron micrographs, were counted and classified according to radius. The number of particles in each class was corrected for impaction efficiency for a cylindrical surface according to the method of Ranz and Wong (4) using a particle density of 2 gm/cm^3. The density of $(NH_4)_2 S_2O_8$ is 1.982, that of $(NH_4)_2SO_4$ is 1.769. Because the impaction efficiency for particles smaller than 0.1 micron radius is very small ($\pm$2%)

the lower limit of radius of particles classified was 0.1 micron. Figure 2 shows two average distributions of particle radii plotted as $\left(\frac{dn}{d(\log r)}\right)$ cm^{-3} (concentration) vs r (radius), where dn is the number of particles per cm^3 of air with radii in the interval d(logr) (5) Table 1 lists the data pertinent to the individual samples from which the average distributions were computed. The curve marked "tropical" is composed of data from all samples which were taken from air above the tropical tropopause plus two samples from the vicinity of the tropopause gap (30° to 50° latitude, 40,000 to 60,000 feet altitude) The "polar" curve represents three samples of air from the vicinity of the gap and one sample(No. P-3) from above the polar tropopause The two distributions are clearly distinguishable in the region of 0.1 to 0.3 micron radius where there is no overlap between the two groups and the concentrations in polar air are about a factor of four lower than those in tropical air. For radii from 0.3 micron to 0.5 microns the curves lie close to each other with considerable overlap of individual distributions from the two groups. The disparity between the curves from 0.5 to 1.5 microns radius is probably the result of considerable variation of the number concentrations of particles in this range from sample to sample; thus no significance

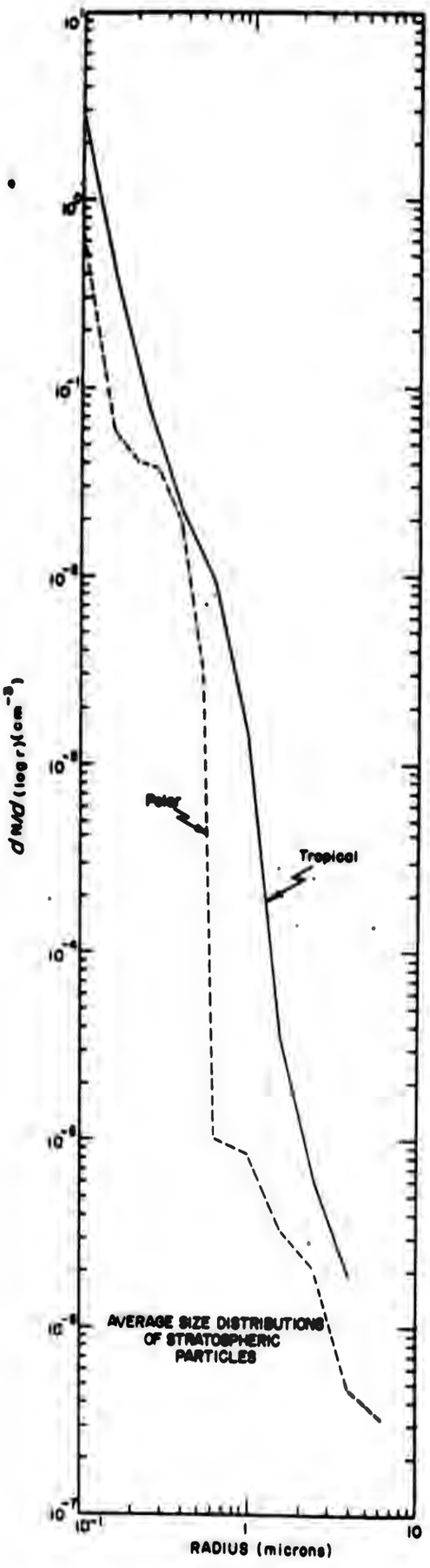

Polar
Tropical
AVERAGE SIZE DISTRIBUTIONS
OF STRATOSPHERIC
PARTICLES
RADIUS (microns)
10
10⁻¹

Figure 2

TABLE I

Sample No.	Date Collected	Altitude (Thousands of feet) & Duration	Latitude	Number Concentration (cm^{-3})	Area Concentration (cm^2/cm^3)	Volume Concentration (cm^3/cm^3)	Sr90 Concentration (dpm/cm^3)
		40 (60 min)	Orbit				
P-1	3/29	55→65(319 min)	45:45N	0.19	2.58×10^{-10}	1.04×10^{-15}	6.50×10^{-5}
P-2	3/31	55→64(406 min)	37:00→48:25N	0.16	3.36×10^{-10}	2.52×10^{-15}	5.8×10^{-5}
P-3	4/12	40.5 (171 min)	67:00→50:25N	0.21	6.94×10^{-10}	5.64×10^{-15}	6.59×10^{-6}
P-4	5/19	50 (377 min)	36:40⇆40:00S	0.09	1.72×10^{-10}	1.13×10^{-15}	1.15×10^{-5}
Average Polar				0.17	3.65×10^{-10}	2.58×10^{-15}	
T-1	2/25	60 (181 min) 65 (186 min)	27:42⇄13:42N	0.28	5.62×10^{-10}	3.25×10^{-15}	3.65×10^{-5}
T-2	3/16	66.8 (357 min)	27:42⇄22:00N	1.23	2.04×10^{-9}	9.95×10^{-15}	7.73×10^{-5}
T-3	4/5	64.5→68.6 (401 min)	15:00N⇆8:00S	0.38	8.16×10^{-10}	8.38×10^{-15}	5.30×10^{-5}
T-4	4/21	60→65 (210 min)	Orbit 48:00 N	1.33	2.73×10^{-9}	2.10×10^{-14}	8.83×10^{-5}
T-5	4/21	50 (193 min) 55 (181 min)	27:43⇄48:00N	0.48	8.57×10^{-10}	5.22×10^{-15}	1.66×10^{-5}
T-6	5/12	63→67 (442 min)	16:25N⇄29:20S	0.33	8.83×10^{-10}	9.35×10^{-15}	5.67×10^{-5}
Average Tropical				0.67	1.31×10^{-9}	9.52×10^{-15}	

is attached to the difference in the curves in this region. In addition to the effect of population variations, the difference between the curves for radii greater than 1.5 microns is of little or no significance because of the small number of particles observed in each sample.

Table 1 also lists the strontium-90 concentrations (in dpm/cm^3) of the air sampled by the probes and filters averaged over the flight paths. The strontium-90 concentrations were obtained from the HASP filter paper samples, most of which were exposed simultaneously with the particle collections. No filter samples were exposed during the collection of samples P-2 or T-2 and the strontium-90 concentrations had to be estimated from HASP collections in the same vicinity a few days prior to and a few days following the particle collections. From the data listed in Table 1 it is observed that (excluding samples P-1, P-2, and P-3) the average ratio of strontium-90 concentration to volume concentration is $(7.0 \pm 3.0) \times 10^9$ dpm/cm^3 of aerosol and the average ratio of strontium-90 concentration to surface area concentration is $(5.0 \pm 2.0) \times 10^4$ dpm/cm^2 of aerosol surface. The 40% standard deviation in each ratio is considered to be reasonable in view of an estimated 20% standard deviation in the averaged strontium-90 concentrations and the problem of obtaining consistent particle size distributions from

electron micrographs.

Two conclusions may be reached on the basis of the particle size concentration spectra and the particle chemical compositions.

1) There appears to be a relatively persistant aerosol layer in the stratosphere extending to an altitude above 70,000 feet (to about 80,000 feet according to Junge (3)).

2) Particles composed of ammonium persulfate and ammonium sulfate comprise almost all of the aerosol in the radius range from 0.1 - 1.5 micron which, in turn, comprises more than 90% of the total mass of the aerosol.

The most likely source of the sulfate particles is H_2S and SO_2 of terrestrial origin which enter the stratosphere by some mixing process and are subsequently oxidized to SO_4 by ozone and ultraviolet radiation (3).

It is probable that the strontium-90 associated with particles in all samples except P-1 and P-2 originated primarily from lower stratospheric injections of nuclear debris and especially from the Hardt ack test series. The high strontium-90 content of samples P-1 and P-2 may be due to the descent into the lower polar stratosphere of nuclear debris from the high altitude rocket shots of August 1958 during the month preceeding sample collection (6)

This debris from two high yield shots would be associated with relatively few particles. The low strontium-90 content of sample P-3, the only sample collected at 40,000 feet in the polar stratosphere, is probably due to dilution of the original particles bearing debris with freshly formed particles in the low stratosphere during the spring season when the ozone content was higher than during the winter.

The strontium-90 had been in the stratosphere long enough to have undergone vertical mixing (with a coefficient of vertical diffusion of 10^4 cm^2/sec) throughout the altitude range in which the aerosol exists (tropopause to 80,000 feet) (7). It then is quite likely that the strontium-90 had become incorporated into the stratospheric sulfate particles and that the distribution of strontium-90 in the stratosphere in the spring of 1960 was determined by the distribution of the sulfate aerosol.

Since the standard deviations of the mean ratios of strontium-90 concentration to particle surface area and particle volume concentrations are both about 40%, it is not possible to determine whether the mechanism of incorporation of nuclear debris with sulfate particles was one of coagulation of small particles of debris by attachment to the surface of larger sulfate particles or by incorporation of small particles of debris into growing sulfate particles.

Since the debris was more than a year old when sampled, it is reasonable to suspect that both mechanisms were involved.

The total sulfate burden of the stratosphere can be estimated using the HASP strontium-90 inventory of 0.60 megacurie (6) and the average strontium-90 concentration to aerosol volume concentration of 7×10^9 dpm/cm^3. Assuming the composition of the aerosol to be ammonium persulfate (density 2.0 gm/cm^3) the mass of sulfate in the stratosphere is estimated to be 3.2×10^5 kilograms. This calculation neglects the possible effect of production of particles in the polar stratosphere, which would dilute the strontium-90 content of the aerosol in that region. The estimated sulfate content, therefore, may be low. However, not enough data exist at present to justify more refined calculations.

On the assumptions that the strontium-90 and sulfate concentrations are related as described above and that the residence half-time in the stratosphere of strontium-90 of tropical origin is 1.0 year, as found in HASP (7), the stratospheric production rate and removal rate may be estimated to be approximately 2.2×10^5 kilograms of SO_4 per year.

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References

1) Contract No. DA-29-044-XZ-609

2) Sample preparation, electron microscopy and electron diffraction were performed by Ernest F. Fullam, Inc., Schenectady, New York.

3) Junge, C E., C. W. Chagnon and J. E. Manson, "A World-Wide Stratospheric Aerosol Layer", Science in press.

4) Ranz, W. E. and V. B. Wong, Ind. Eng. Chem. 44, 1371 (1952).

5) cf. Junge, C E., "Atmospheric Chemistry" Advances in Geophysics 4, Academic Press Inc., New York, (1958).

6) Feely, H. W., HASP Quarterly Progress Report, October, 1960. (DASA 1222)

7) Spar, J., Personal Communication

8) Feely, H. W., Science, 131, 645 (1960)

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