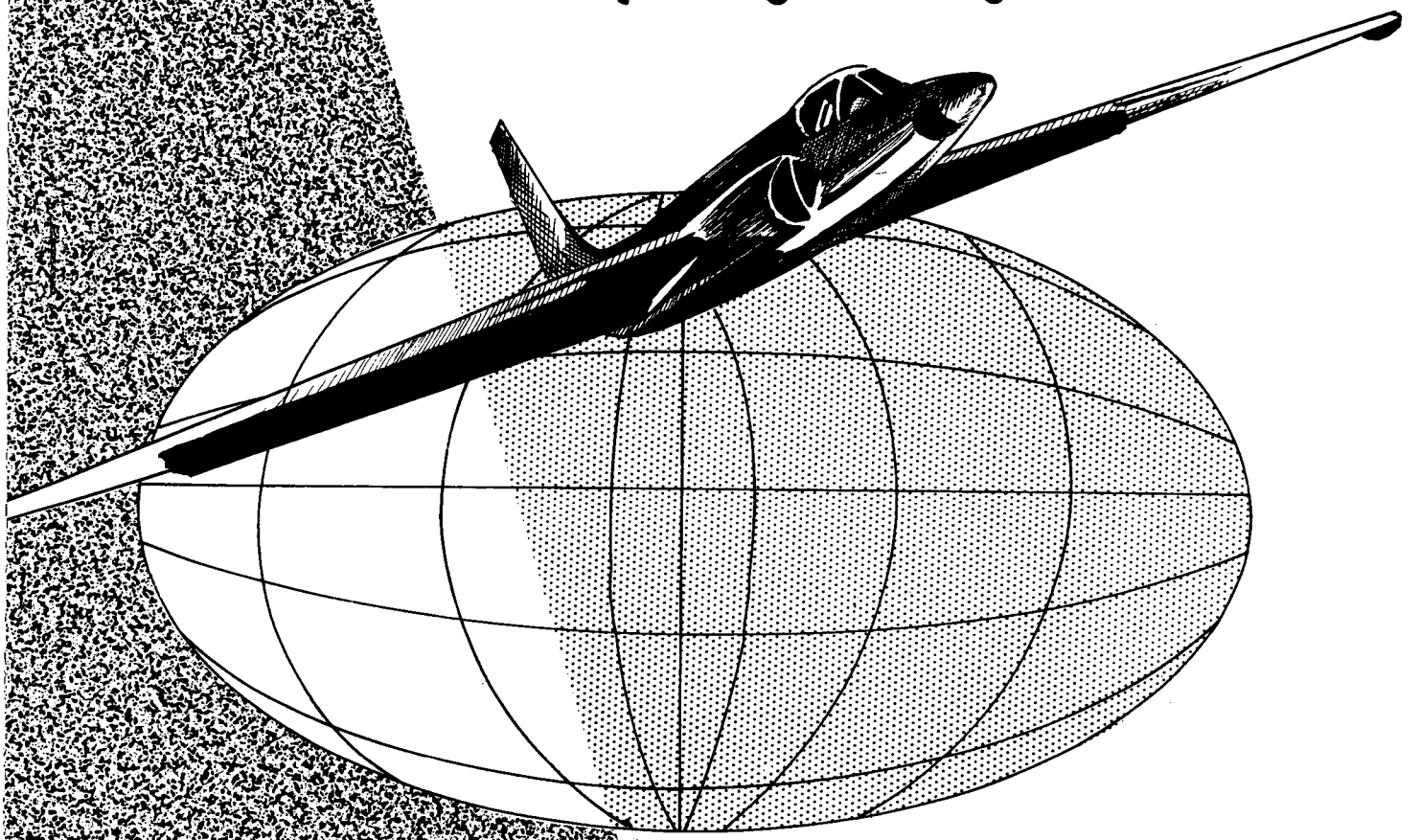
DASA 532B

MASTER

HASP

Special Report on

High Altitude Sampling Program



Maj. Albert K. Stebbins, III Ed.

RADIATION DIVISION

1 JUNE 1960

DISCLAIMER

TECHNICAL ANALYSIS REPORT

DASA-532B

A SPECIAL REPORT on the HIGH ALTITUDE SAMPLING PROGRAM

by

Major Albert K. Stebbins, III, Ed.

RADIATION DIVISION

This document was previously published as DASA 532, "A Special Report to the Government of Argentina."

1 June 1960

HEADQUARTERS, DEFENSE ATOMIC SUPPORT AGENCY
WASHINGTON 25, D. C.

ABSTRACT

This report outlines the results of the HASP program in its effort to determine the role played by the stratosphere in the world-wide distribution of radioactive fallout from nuclear weapons tests. The program has operated since the fall of 1957. The sampling network using U-2 aircraft has collected 10^8 standard cubic feet of air from 57°S to 71°N up to 70,000 feet. Ashcan data is used for upward extrapolation. IPC Paper 1478 of near 100% efficiency is used. Stratospheric matter sampled is in the 0.01 micron range. The uneven distribution of material in the stratosphere has been noted. Stratospheric inventories of Sr-90 have been calculated for the periods November 1957 to December 1958, January to August 1959, and September to November 1959 to be respectively 0.95, 0.81, and 0.7 megacuries. Concentrations have been greater in the Northern Hemisphere by a factor of 2 or 3 over the Southern Hemisphere. The Sr-90 maximum occurs in the equatorial regions around 90,000 feet and slopes down to around 70,000 feet in the polar regions. Little fractionation is noted in stratospheric debris. Cs-137 to Sr-90 ratios are 1.8 ± 0.5. A semi-empirical application of Gaussian diffusion is described which suggests that hot clouds injected in the equatorial stratosphere spread in the North-South direction with mixing coefficients near 5×10^8 cm^2/sec. Vertical mixing is slower with coefficients of 4×10^3 and 2×10^4 cm^2/sec suggested for tropical and polar regions respectively. An Injection-Depletion model is offered which indicates that as much as 50% of the material produced in a megaton ground surface burst comes down in local fallout. Removal from the stratosphere occurs at different rates depending on altitude and latitude of injection and season of the year. Effective half-residence times of respectively 5, 10, and 20 months for polar, low equatorial and high equatorial debris is suggested. Surface concentrations of Sr-90 are displayed as a function of latitude and time. The Northern Hemisphere carries 3/4 of the burden. The maximum burden of Sr-90 is predicted to occur in 1961; however, concentrations in food have probably reached their maximum value. The radiation hazard from fallout is summarized. Fallout has increased the dose man receives from the natural radiation background by about 2%.

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PREFACE

Nature of Report

This document is a special report on the High Altitude Sampling Program (HASP) being conducted by the Defense Atomic Support Agency. The main purpose of the report is to inform all people interested in world-wide radioactive fallout of the results garnered to date from this program. HASP was originally conceived in 1956 to determine the role played by the stratosphere in the distribution of radionuclides from the nuclear weapons testing programs that several nations had embarked upon. Significant progress has been made in this program and the point has been reached where a rather clear picture is emerging on how particulate matter is injected into the stratosphere, how it is circulated, and how it is subsequently deposited upon the ground.

Organization of Report

This report is organized into two parts. Part A or Method describes the sampling experiment in detail and discusses some of the results of corollary investigations which form the foundation upon which the main experiment rests. These investigations include a study of the nature of stratospheric particulates, the efficiency of collection of these particles, and duct and paper flow rates. Part B or Results describes the basic patterns evolved by the stratospheric inventory and a meteorological interpretation of their meaning. In order that the contribution of the HASP program may be placed in proper perspective,

several chapters have been included which attempt to account for all the radioactive debris introduced into the atmosphere from past tests and an evaluation of the effect this material may have upon man.

Acknowledgements

The HASP program, like many others of its nature, has been the joint effort of many people and organizations. Special acknowledgement is made to the Government of Argentina for providing base rights at Ezeiza Airport and to the Governments of Canada, Mexico, Venezuela, Brazil, Bolivia, and Paraguay for overflight rights. Much of the material in this report has appeared in contract reports from various organizations. The work on IPC Paper 1478 is that of Dr. J. A. Van den Akker of the Institute of Paper Chemistry. Members of the staff of Isotopes, Incorporated have made contributions in the following areas: Dr. H. W. Feely, Stratospheric Distribution of Radionuclides; Dr. J. P. Friend, Stratospheric Particulates; Dr. P. W. Krey, Radiochemistry; Dr. J. Spar, Meteorological Model; Drs. J. L. Kulp and A. Walton, Surface Distribution and Radiation Effects. Professor E. G. Reid of Stanford University has conducted the duct calibration program. Many helpful conversations on world-wide fallout have been held with Dr. L. Machta of the U. S. Weather Bureau, Dr. E. A. Martell of the U. S. Air Force, and Mr. J. Z. Holland of the U. S. Atomic Energy Commission. In addition, stimulating discussions have been held in Scandinavia with J. Ambrosen, T. Hvinden, S. Small, R. Bjornerstedt, and K. Edvardson.

Personnel of Headquarters, DASA who have played a key role in the HASP program include Col R. Maxwell, LtCol H. Rose, Major L. E. Trapp and Dr. F. H. Shelton. Finally, special mention must be made of the officers and men of the 4080th Strategic Reconnaissance Wing and the 64th Air Rescue Squadron of the U. S. Air Force. Operating over formidable routes on a routine basis and weathering such events as a flame-out south of the southern tip of South America and a mid-winter dead-stick landing on a frozen lake in Canada, these people have turned in a remarkable record of reliability and flight safety.

ALBERT K. STEBBINS, III, Ed.

NOTE: This document was previously issued under the title "A Special Report to the Government of Argentine." Two appendices have been removed and various typographical errors have been corrected.

PART A

METHOD

CHAPTER 1

INTRODUCTION

Historical Background

With the advent of thermonuclear weapons in 1952, two new quantities were introduced into the national defense equation. One was that test or use of these weapons would cause the production of large amounts of fission products in the open atmosphere, the other was that there would be world-wide distribution of this material through stratospheric circulation.

Obviously the magnitude of the potential hazard associated with this radioactive fallout had to be determined in order that it might be placed into proper perspective with the other hazards of our modern technological age. The Joint Chiefs of Staff, realizing the seriousness and complexity of this problem, requested in the early fall of 1954 that the Defense Atomic Support Agency (DASA), formerly the Armed Forces Special Weapons Project (AFSWP), study and evaluate this problem on a continuing basis. After considerable study of the problem within DASA during 1955, it was determined that the largest uncertainty in the prediction of the distribution and concentration of world-wide fallout debris on the surface of the earth was the quantity of fission products in the stratospheric reservoir and the rate and mode of their transfer. Therefore, DASA, early in 1956, initiated a research program to define and delineate the stratospheric reservoir of fission debris. This program became known as the High Altitude Sampling Program or "Project HASP."

Objectives of HASP

Briefly, the HASP project has the following specific objectives:

1. the determination of the quantities of various radionuclides in the stratospheric reservoir and the distribution of this debris as a function of latitude, altitude and time,

2. the estimation of the residence times of these radionuclides in the stratospheric reservoir, and

3. the estimation of the mechanisms and rates of mixing and transfer of this debris within the stratosphere and into and through the troposphere.

In addition to the pursuit of these objectives, the program has included investigations of other aspects of radioactive fallout, such as the physical nature of radioactive stratospheric particulates, evaluation of studies of concentrations in rainwater and soil, the quantity of stratospheric strontium-90 which has already fallen out, and the potential hazard to the human population from strontium-90 and from other radioactive nuclides present in world-wide fallout.

Method of Achieving the Objectives

In order to sample the stratosphere adequately, a system had to be devised which could cover a wide band of latitude, obtain samples through an altitude range extending from the troposphere well into the stratosphere, permit frequent sampling intervals, and collect enough material for accurate radiochemical analysis. Sampling with manned aircraft proved to

be the most attractive approach because of the integrated nature of the sample obtained, the tremendous potential coverage, the control of the sample path, and the potential coverage per unit cost.

The development and availability to DASA of the Lockheed U-2, using a new filter medium of low resistance to air flow at high velocities, yet with high collection efficiency under flight conditions, plus experimental and theoretical aerodynamic investigations showing that a volume of air sampled under flight conditions could be accurately determined, all made Project HASP a real possibility.

In light of these developments, certain operational considerations, and the need for establishing scientific facilities and contracts to support the Project, the following five-phase concept was conceived:

a. Phase I

1. Development of an atmospheric sampling mechanism.

2. Flight operation planning and training.

3. Establishment of scientific facilities and contracts

b. Phase II

1. Establishment of a meridional sampling net in the Northern Hemisphere.

2. Flight profiles to measure precision of sampling.

3. Calibration of the AEC Balloon Sampler.

c. Phase III

1. Extension of the meridional sampling net to the Southern Hemisphere.

2. Study rates of lateral mixing of HARDTACK (1958 Pacific Test) debris.

d. Phase IV

1. Continue the meridional sampling net in the Northern Hemisphere.

2. Detailed study of mixing and transfer processes.

3. Conduct atmospheric sampling to the North Pole.

e. Phase V

1. Spot check in Southern Hemisphere.

2. Recalibration of ducts.

3. Continued spot checks in Northern Hemisphere.

The sampling program has been designed to provide as great a density of sampling as is possible along a single meridional cross section. The normal zonal circulation of the atmosphere should eventually carry the entire atmosphere across the meridian selected. During normal HASP missions the aircraft covers 22 degrees of latitude at altitudes as high as 70,000 feet. The range limitation of the aircraft makes it impossible to sample the entire length of the meridional corridor at any one time. Moreover, the U-2 may be flown safely only through regions wherein navigation may be performed by means of the magnetic compass and only from bases meeting rather strict requirements concerning normal wind velocities and directions. Nevertheless sampling was carried out as far south at 57° South latitude during Phase 3 of the program and as far north as 71° North latitude during Phase 4. About

90 percent of the atmosphere of the earth lies between 57° South and 71° North latitude. Extrapolation to the higher stratosphere can be made by use of the Atomic Energy Commission Weather Bureau Project ASHCAN Balloon Data.

Scientific Program

During 1956 a team of organizations capable of performing the scientific program was established. These included Isotopes, Inc., the Institute of Paper Chemistry, and Stanford University. See Fig. 1.

A contract was let in February 1957 with Isotopes, Incorporated, Westwood, New Jersey, to provide scientific direction and interpretation, and to perform the radiochemical analysis.

Isotopes, Incorporated effort, under the scientific direction of Dr. J. Laurence Kulp, and Dr. Herbert W. Feely, has been devoted to:

a. Planning, in coordination with DASA, flight profiles for the sampling missions which weave the strands of the meridional sampling net.

b. Performing radiochemical analysis.

c. Research and correlation of fallout information bearing on the objectives of the HASP.

d. Interpretation of results and drawing conclusions. All radiochemical data are reduced to disintegrations per minute per 1,000 standard cubic feet prior to interpretation.

The analytical program is based on a sequential radiochemical analysis of all filters. The total beta activity of each sample is

HIGH ALTITUDE SAMPLING PROGRAM

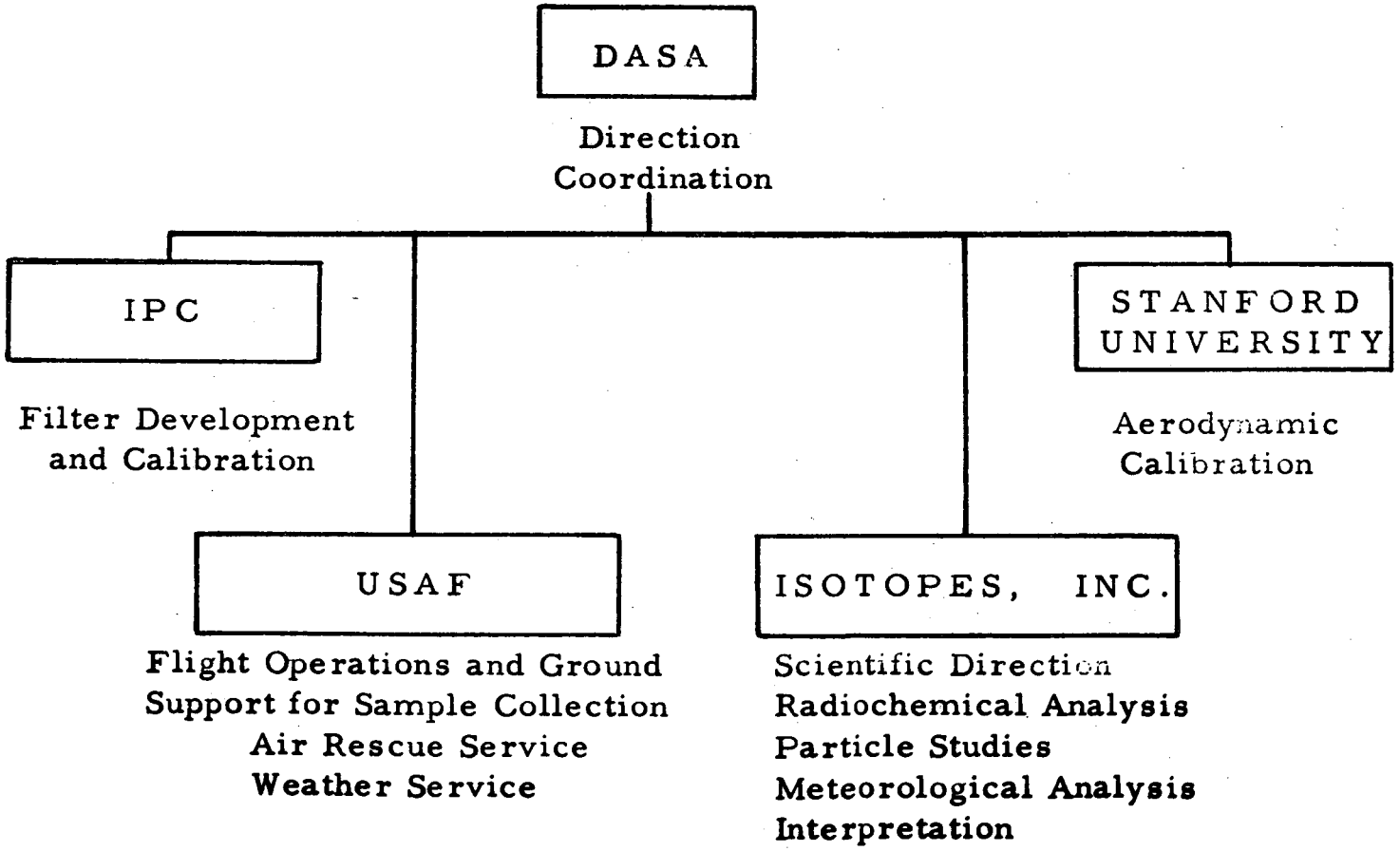


FIGURE 1

determined as a reference for comparison with activities of individual radionuclides. The sequential analysis of the sample is then carried out with separation of the various fractions and purification and measurement of certain nuclides. These nuclides include strontium-90 and cesium-137, which are considered potentially hazardous, strontium-89, yttrium-91, cerium-144 and zirconium-95, which are useful for estimating the age of debris, and tungsten-185, a tracer introduced into the stratosphere during HARDTACK. Separate aliquots of some samples have been used for the analysis of plutonium, a potentially hazardous element, of rhodium-102, a tracer introduced by a megaton weapon injected at a great height during HARDTACK, of beryllium-7 or phosphorus-32, nuclides produced by cosmic ray interaction with the atmosphere, or of sodium-22, an activation product produced by water surface bursts in the Pacific. A few nuclides are measured by gamma ray spectroscopy of the untreated filter paper. These include barium-140 and zirconium-95, useful for estimating the age of debris, and tungsten-181, a second activation product produced by the weapons which produced tungsten-185.

Radiochemical data for HASP filters are compiled and interpreted in accordance with theories of stratospheric meteorology. These data and the conclusions derived from them are compared with results from other studies of world-wide fallout being carried on in the United States and abroad. Through the synthesis of all available data, conclusions are reached on the potential biological hazard existing as a result of

world-wide fallout from past nuclear bomb testing and future hazards to be expected from any further testing.

Collateral Programs

A contract was let in September 1956 with the Institute of Paper Chemistry (IPC), Appleton, Wisconsin, for further development and calibration of a filtering medium. The filter medium being used in the HASP is IPC filter paper No. 1478, consisting of cotton fibers with gauze backing. The fibers are impregnated with di-butoxyethyl phthalate, a sticky, oily substance which assists the retention of particulate matter.

Further studies of the retention of IPC filter paper No. 1478 for small particles, carried out under the direction of Dr. J. A. Van den Akker at the Institute of Paper Chemistry, lend support to the belief that the filter medium has approximately 100 percent retention efficiency for stratospheric particulates under flight conditions.

Prof. Elliott G. Reid of Stanford University, one of this country's leading authorities on the application of aerodynamic theory to atmospheric sampling mechanisms, was consulted regarding the design and application of a filter-type sampler for the HASP. Based on Prof. Reid's theoretical work in this field, Lockheed Aircraft Corporation designed and built a sampler having a capability of exposing four filter papers, consecutively. This sampler was installed in the nose position as an integral part of each of the six U-2 aircraft assigned to DASA for carrying out the HASP. During the summer of 1959, hatch samplers,

located beneath the body of the aircraft and capable of exposing six filter samples in succession, were also installed.

An experimental flight test calibration program, under the direction of Prof. Reid, was conducted at Laughlin Air Force Base, Texas, during September 1958 and was supplemented with wind tunnel tests at the NASA Ames Laboratory. Calibration curves prepared by Professor Reid are used in conjunction with flight data recorded by the aircraft pilot and obtained from 16 mm films of an auxillary control panel which indicated time, altitude and filter exposure. These ducts will be recalibrated in July 1960 in order to eliminate an apparent discrepancy in the duct flow rate data. (See Chapter II.)

Throughout the course of HASP, personnel at Isotopes, Inc., have carried out an analysis of the radiochemical data for HASP samples and of information on the distribution of radioactive particles through the filters together with theoretical analysis of data from calibration tests of the samples and of the filter medium to assess the accuracy of the calculations of the quantity of air sampled by HASP filters.

A number of experiments have been performed on HASP filter samples and on dust samples collected by impaction to measure the size distribution and physical characteristics of stratospheric particles. Autoradiography of whole filters, of individual fibers and of microtomed sections from filters have been employed together with optical and electron microscopy.

Preliminary results from studies of the particulate matter in HASP samples indicate that the radioactivity is carried almost entirely by particles between 0.01 and 0.5 microns in diameter.

A mission was flown in which a HASP aircraft collected four samples while orbiting a balloon which was collecting a sample for Project ASHCAN. Intercomparison of the radiochemical data permits calibration of the ASHCAN sampler at one altitude. A recalibration flight is planned sometime during the summer of 1960.

Additional intercalibration flights between the U-2 and B-57 aircraft conducting sampling for the U. S. Weather Bureau have been performed at Minot AFB, North Dakota. Future operations in South America and elsewhere contemplate the coordinated use of U-2 and B-57 sampling by the U. S. Air Force to continue spot checking in the HASP network.

Operational Program

The operational responsibility for carrying out Project HASP was assigned to the Strategic Air Command (SAC).

The Project got underway when six Lockheed U-2 aircraft were made available to SAC for assignment to the 4080th Strategic Reconnaissance Wing (4080th SRW) in August and September 1957. The U-2 is an unarmed, single engine jet aircraft with a crew of one. It has tandem landing gear and outrigger wheeled struts, sometimes referred to as "pogo sticks." These struts drop off when the aircraft becomes airborne. Landing is accomplished with the aid of a drag chute and, after the landing roll,

wing tip skids prevent damage to the aircraft. Additional characteristics are given in Chapter II.

Following an initial training and testing phase in August-October, 1957, at Laughlin Air Force Base, Texas, the sampling aircraft were based at Plattsburg Air Force Base, New York, and Ramey Air Force Base, Puerto Rico. From these two bases a north-south sampling corridor along 70° West Longitude, and extending from 66° North to 6° South Latitude, was monitored systematically from November, 1957 through May, 1958. During June and July, 1958, sampling was conducted only in the vicinity of Puerto Rico. Between September, 1958, and August 1959, the aircraft operated from Ramey Air Force Base and Ezeiza Airport, Buenos Aires, Argentina, in order to sample the Southern Hemisphere more completely. A corridor along 63° West Longitude, and extending from 38° North to 57° South Latitude was monitored. The Ramey aircraft made occasional deployments to Plattsburg to check the northern reaches of the sampling corridor. Since September 1959 the aircraft have operated from Laughlin AFB, Texas, and Minot AFB, North Dakota. Sampling between 10° N and 71° N roughly along 100° West Longitude. The Laughlin aircraft have made monthly deployments to Ramey to continue trans-equatorial measurements. During May and June 1960 these aircraft will conduct a spot-check out of Ezeiza along with four B-57 aircraft from the Air Research and Development Command (ARDC) of the U. S. Air Force. Since these sampling missions are flown over terrain which includes sub-arctic - water - and tropical jungle areas having a minimum of emergency landing fields and

navigation aids, Air Rescue support is provided for each mission by the 64th Air Rescue Squadron. SC-54 type aircraft are used for this support.

It is believed that the air sampled along a meridional corridor will be representative of the entire atmosphere as long as sampling is carried out over a sufficiently long period of time. Eventually the whole atmosphere will be carried through the corridor by normal zonal circulation.

Two sampling missions, each of which collects 40 samples from the corridor, are scheduled per week. To date, some 4000 samples have been collected. These samples, with all accompanying data, have been forwarded to Isotopes, Incorporated for radiochemical analysis and interpretation. In addition samples have been supplied to the Air Force Cambridge Research Center (AFCRC) in support of their Rhodium-102 tracer experiment.[1]

Since January 1960 two aircraft have had installed dust impactor probes supplied by AFCRC for electron microscopic investigation of stratospheric particulate matter.

CHAPTER II

SAMPLE COLLECTION

Introduction

This chapter will cover a detailed description of the meteorological experiment encompassed by the HASP program. The discussion will include a description of the stratosphere, the sampling network, the sampling vehicles, and extrapolation techniques.

Review of Atmospheric Structure

The troposphere is characterized by a decrease of temperature with altitude. (See Fig. 2.) This lowest layer of the atmosphere extends from the ground up to altitudes which vary with season and with latitude. In the tropics the depth of the troposphere is approximately 55,000 feet, while in the polar regions it decreases to 30,000 feet or less. It is deeper in summer than in winter, and, except for a seasonal oscillation, is roughly symmetrical about the equator.

Vertical and horizontal mixing in the troposphere by turbulent and convective processes tends to produce a homogeneous distribution of airborne material. (The atmospheric gaseous constituents themselves are found in almost the same proportions everywhere.) The concentration of gaseous and small particulate radioactive debris is therefore quickly diluted by turbulent diffusion following its injection into the troposphere. Particulate material in the troposphere becomes

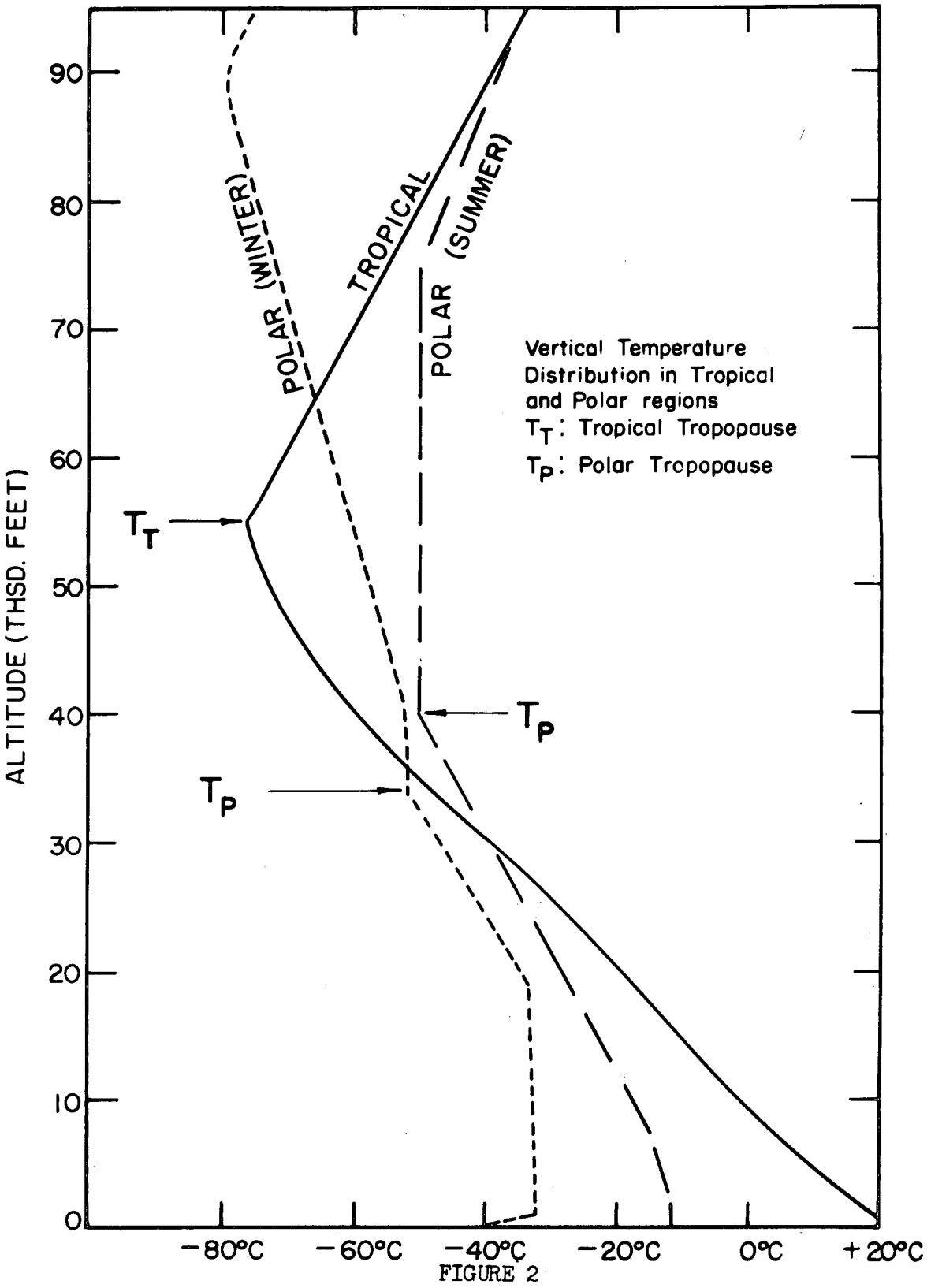

ALTITUDE (THSD. FEET)
90
80
70
60
50
40
30
20
10
0
POLAR (WINTER)
TROPICAL
POLAR (SUMMER)
Vertical Temperature
Distribution in Tropical
and Polar regions
T_T: Tropical Tropopause
T_P: Polar Tropopause
T_T
T_P
T_P
−80°C
−60°C
−40°C
−20°C
0°C
+20°C

FIGURE 2

attached to cloud droplets, ice crystals, snow flakes, and raindrops, and is removed from the atmosphere by precipitation. Thus the troposphere is a region in which the concentration of radioactive material is low, due to turbulent mixing, and from which radioactive debris is constantly being removed by precipitation. (It has been estimated that the particulate radioactivity in the troposphere is reduced to half its initial value in about one month by precipitation scavenging.[2,3,4]) The background level of Sr^{90} in the upper troposphere, within a few months after weapons testing, is about 1 dpm/1000 SCF* or less, while in the lower troposphere the level is about 0.3 dpm/1000 SCF. It is easily shown that the entire tropospheric reservoir of Sr^{90} is less than 3 percent of the total amount of bomb-produced radioactivity in the atmosphere. (The remainder resides in the stratosphere.)

In most, but not all, of the stratosphere the temperature does not decrease with altitude. (The decrease of temperature with height is called the lapse rate.) Where the temperature does decrease with altitude in the stratosphere, the decrease is much smaller than in the troposphere. The intensity of vertical mixing is, other things being equal, closely related to the variation of temperature with altitude, and is greater where the decrease of temperature with height is large. Thus vertical mixing is generally strong in the troposphere and weak,

* One decay per minute per one thousand standard cubic feet of air.

although not entirely absent, in the stratosphere. The relative weakness of vertical mixing in the stratosphere accounts for the inhomogeneous distribution of radioactivity in the stratosphere compared with the troposphere, and also for the relatively long time of residence of radioactive debris in the stratosphere.

The stability of the stratosphere (i.e., its resistance to vertical mixing) is not uniform. At all seasons of the year the tropical stratosphere, between about 25°N and 25°S, exhibits an inversion, or increase of temperature with altitude. This represents a very stable stratification, which strongly inhibits vertical mixing. On the other hand, in arctic and sub-arctic latitudes the stratosphere tends to be more nearly isothermal. This represents a less stable stratification than that of the tropical stratosphere, although it is still far more stable than the troposphere. In winter the arctic stratosphere becomes quite cold at upper levels, with the result that the temperature decreases with altitude. Thus the stratosphere is least stable, and therefore most easily mixed in the vertical, in winter in the arctic regions.

The boundary between the troposphere and the stratosphere is a layer known as the tropopause (Fig. 3.) Often the transition from troposphere to stratosphere is so abrupt that the tropopause can be identified as a surface, with positive lapse rate below and an inversion above. But just as frequently the transition is gradual, so that one may only be able to identify as the tropopause a layer several thousand feet thick above which the lapse rate is clearly stratospheric and

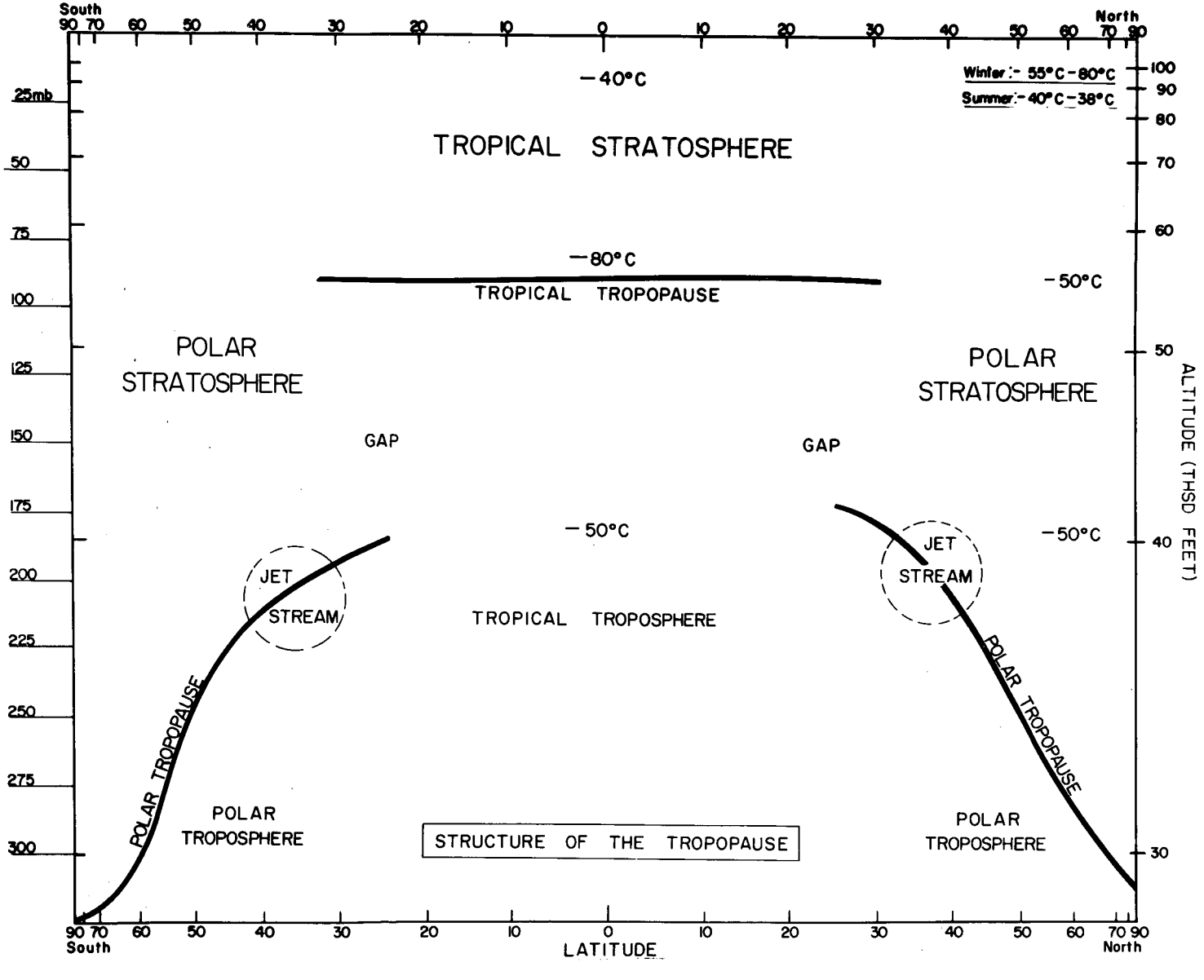

South
90 70 60 50 40 30 20 10 0 10 20 30 40 50 60 70 90
North
−40°C
Winter :- 55°C - 80°C
Summer:- 40°C - 38°C
TROPICAL STRATOSPHERE
25mb
50
75
100
125
150
175
200
225
250
275
300
−80°C
TROPICAL TROPOPAUSE
−50°C
POLAR STRATOSPHERE
POLAR STRATOSPHERE
GAP
GAP
−50°C
−50°C
JET STREAM
JET STREAM
TROPICAL TROPOSPHERE
POLAR TROPOPAUSE
POLAR TROPOPAUSE
POLAR TROPOSPHERE
POLAR TROPOSPHERE
STRUCTURE OF THE TROPOPAUSE
100
90
80
70
60
50
40
30
ALTITUDE (THSD FEET)
90 70 60 50 40 30 20 10 0 10 20 30 40 50 60 70 90
South
LATITUDE
North

FIGURE 3

below which it is clearly tropospheric.

The tropopause is not a continuous surface, but rather consists of several partially overlapping layers. The tropical tropopause is a nearly horizontal layer at about 55,000 feet above sea level extending from about 35°N to 35°S. It extends farther from the equator and is higher in summer than in winter. (It is also higher over warm air masses in the troposphere than over cold air masses. But rarely, if ever, is it found lower than 40,000 feet.) The pressure at the mean altitude of the tropical tropopause is about 10 percent of sea level pressure, indicating that only 10 percent of the mass of the atmosphere lies above the tropopause in tropical latitudes. The polar tropopauses, on the other hand, slope upward from an altitude less than 30,000 feet in the polar regions of the two hemispheres, to about 40,000 feet in the vicinity of 30°N and 30°S. The polar tropopauses are much more variable, exhibiting not only large seasonal variations (high in summer, low in winter), but also large fluctuations with the weather systems from day to day. The mean pressure at the polar tropopause is about 25 percent of sea level pressure, so that about 25 percent of the mass of the atmosphere poleward of latitude 30° is stratospheric.

In the gap between the two principal tropopause layers an intermediate tropopause is frequently observed. The gap is a transition zone between tropospheric air on the equatorial side and stratospheric air on the poleward side. Particularly in winter, a narrow band of strong westerly winds, the jet stream, is found meandering in a wave-like

fashion around each hemisphere. On a north-south (meridional) cross-section the jet stream appears as a relatively small area of high velocity penetrating the vertical plane near the lower part of the gap. Turbulence is usually severe in the jet stream region, and indeed throughout the gap, despite the fact that the lapse rate in the gap is not large. The turbulence in this region is a consequence of the excessively large wind shear associated with the velocity maximum. The gap is a region of strong mixing, and undoubtedly represents a region of active mass exchange between the polar stratosphere and the tropical troposphere. The latitude of the gap varies with season, being found closer to the pole in summer and closer to the equator in winter. But there is an equally large variation from day to day associated with the migratory pressure systems.

The sampling conducted by Project HASP has been restricted mainly to a vertical, north-south cross-section of the atmosphere along approximately the 70° West Meridian and the 100° West Meridian. The meterorological analysis has also (with few exceptions) been restricted to the examination of the structure of the atmosphere in this narrow corridor, and is displayed in the form of meridional cross-sections. The sampling program was designed on the assumption that east-west (zonal) variations of atmospheric structure and radioactivity are transient in nature, and are fully compensated for by sampling over a period of time long enough to permit the essentially zonal winds of the earth to transport all the air through the sampling net. Figure 4

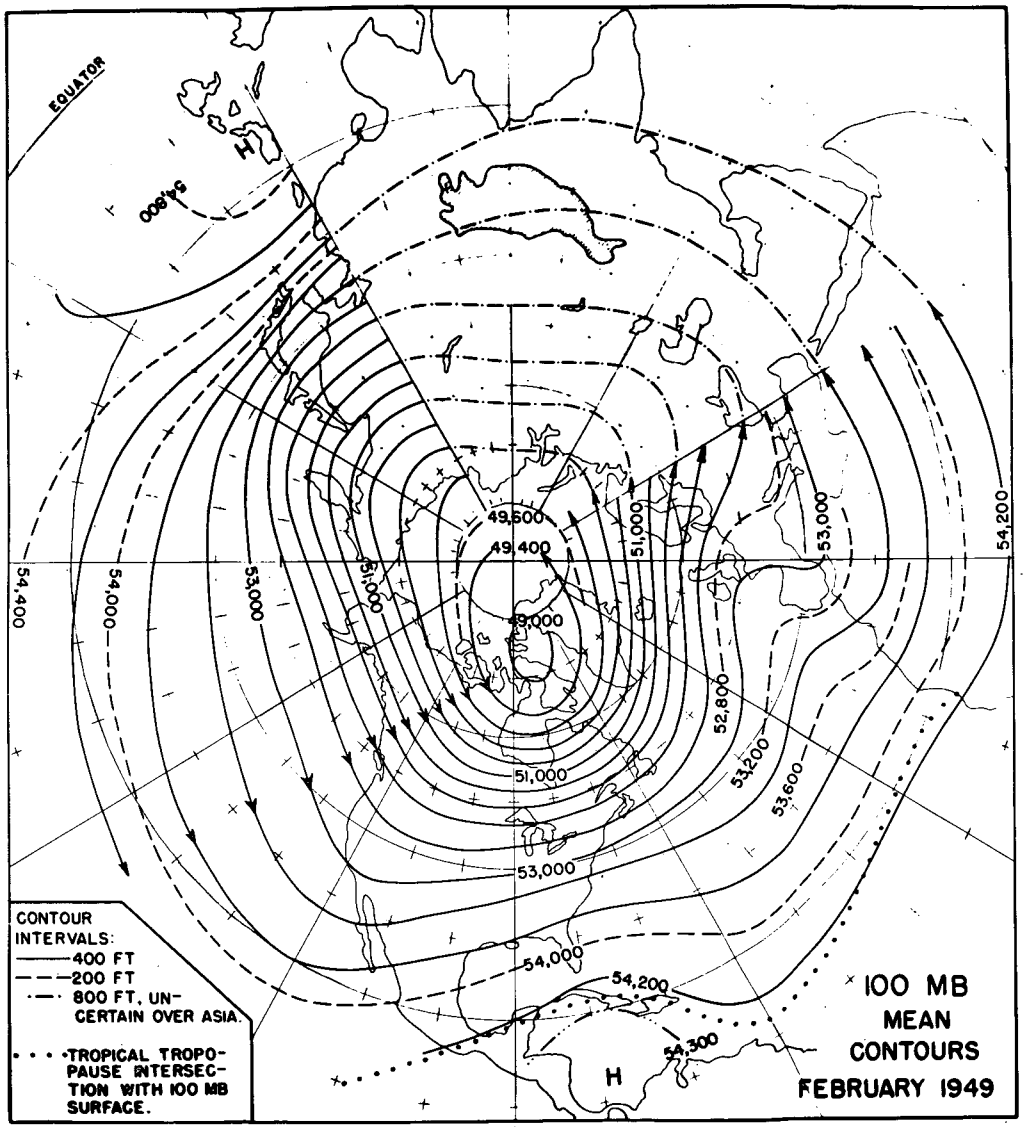

EQUATOR
H
49,500
49,400
49,000
51,000
53,000
54,200
54,400
54,000
53,000
51,000
52,800
53,200
53,600
51,000
53,000
54,000
54,200
54,300
H
CONTOUR INTERVALS:
400 FT
200 FT
800 FT, UNCERTAIN OVER ASIA.
TROPICAL TROPOPAUSE INTERSECTION WITH 100 MB SURFACE.
100 MB MEAN CONTOURS
FEBRUARY 1949

FIGURE 4

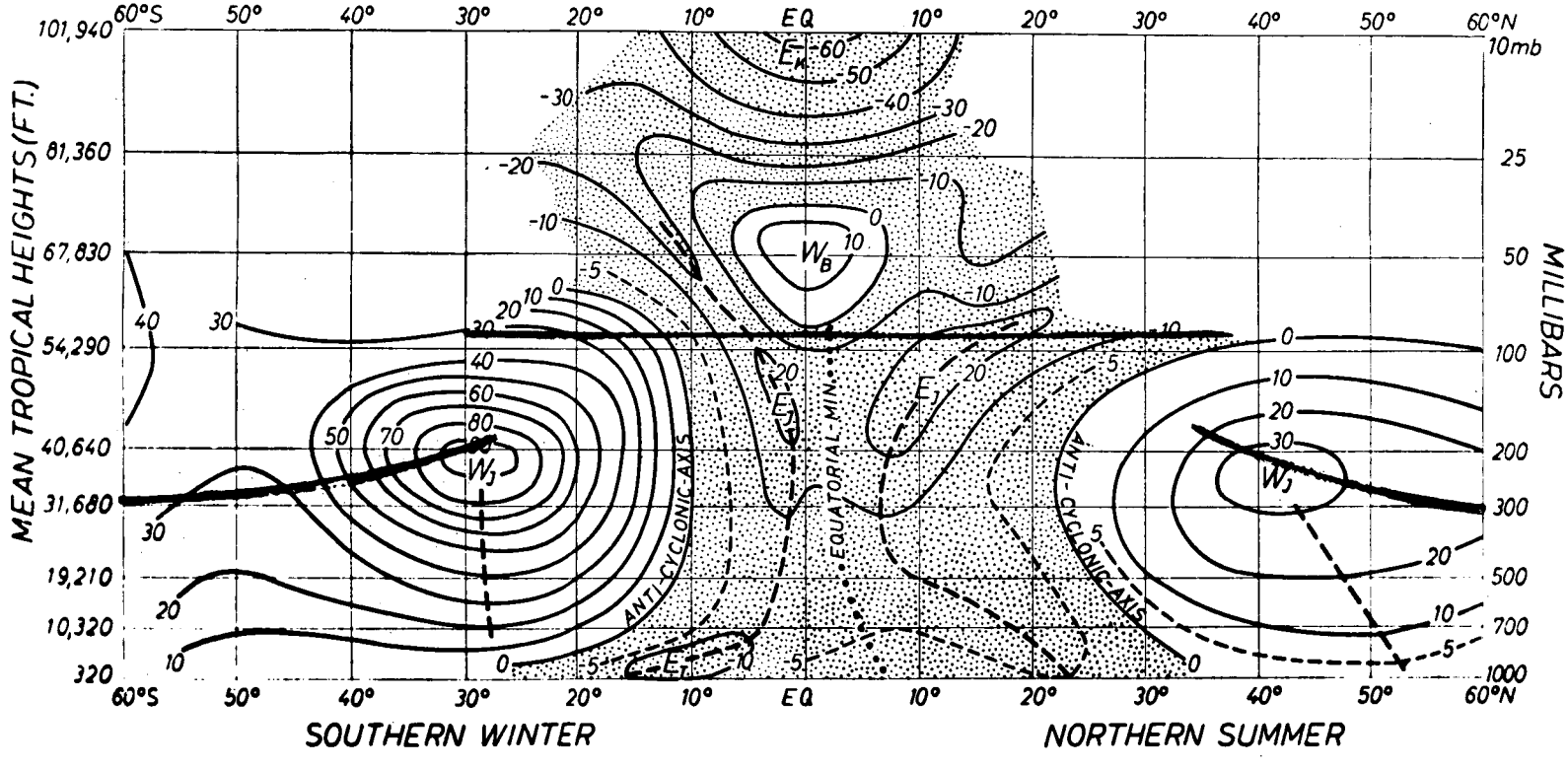


FIGURE 5 Zonal Flow

illustrates a typical flow pattern showing westerly winds in the northern stratosphere. Figure 5 shows typical isotachs or lines of constant speed in the atmosphere. The strong winter jet is shown as well as the light stratospheric winds. The stippled areas on the chart represent easterly flow.

The Sampling Program

Crow Flight, the sampling operation of the High Altitude Sampling Program, has been divided into five phases. The bases of operation and the approximate limits of the flight tracks followed are given in Table 1 and in Figure 6.

Table 1

Flight Tracks for the High Altitude Sampling Program

			Range Limit	
Phase	Base	Direction	Latitude	Longitude
1	Laughlin, Texas	North	54°31'N	100°30'W
		South	14°00'N	100°30'W
		East	31°30'N	76°00'W
2	Plattsburg, N. Y.	North	67°00'N	71°00'W
		South	21°00'N	71°00'W
	Ramey, Puerto Rico	North	39°00'N	69°00'W
		South	07°00'S	69°00'W
3	Plattsburg, N. Y.	North	68°00'N	71°00'W
	Ramey, Puerto Rico	North	38°00'N	64°00'W
		South	07°30'S	64°00'W
	Ezeiza, Argentina	North	09°30'S	64°00'W
		South	57°00'S	64°00'W
4	Minot, N. D.	North	71°00'N	132°30'W
	Laughlin, Texas	North	49°30'N	98°00'W
		South	12°30'N	81°30'W
	Ramey, Puerto Rico	South	08°00'S	67°00'W

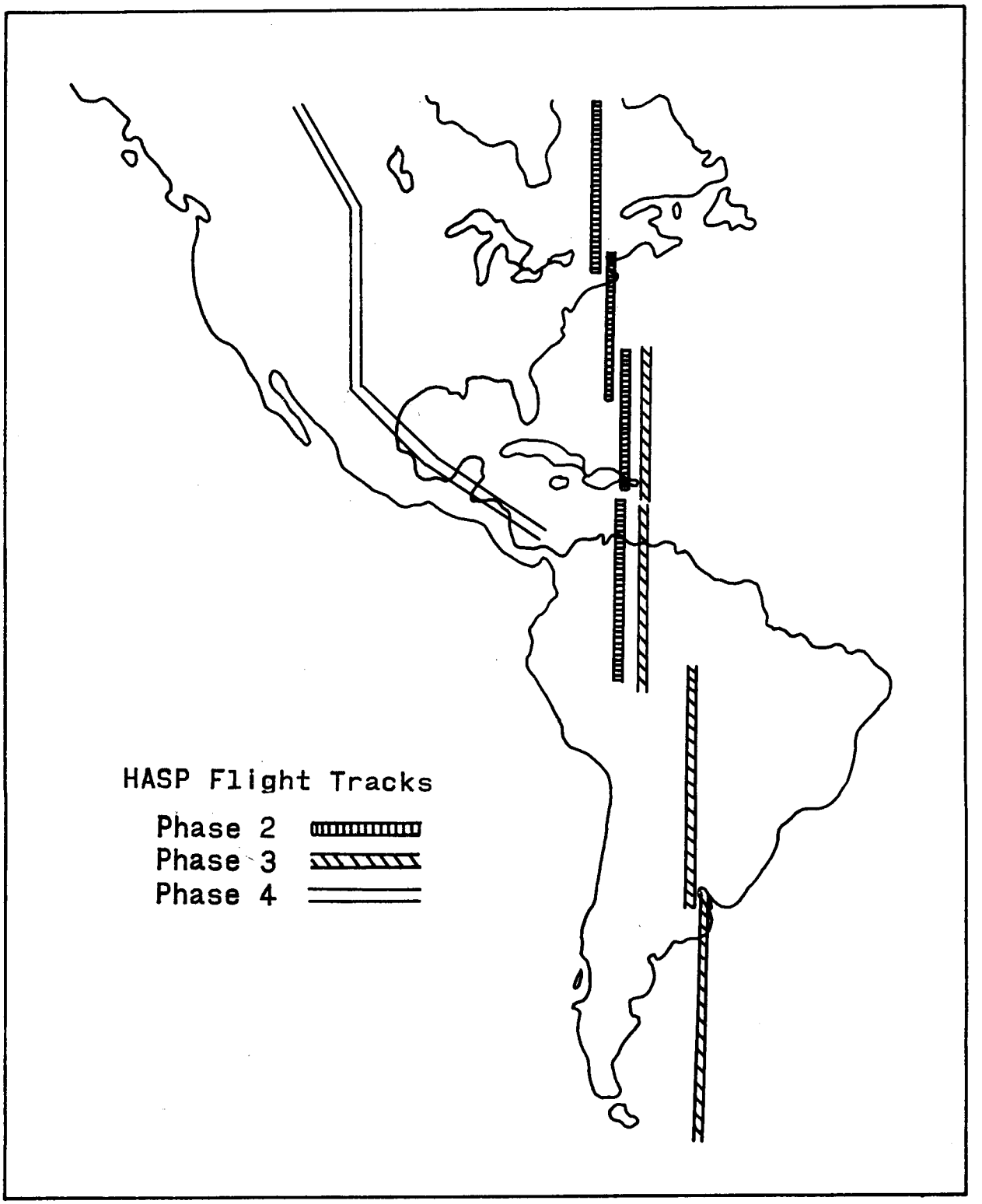


FIGURE 6

Phase 1 was devoted to training of personnel and testing of equipment. During Phase 2 the stratosphere of the Northern Hemisphere was sampled During Phase 3 the sampling corridor was extended southward into the Southern Polar Stratosphere. During Phase 4 a more thorough sampling of the stratosphere of the Northern Hemisphere is being carried out. The number of samples collected during each month of HASP is indicated in Table 2.

Table 2

Rate of Collection of Samples During HASP

Phase 1		Phase 2		Phase 3		Phase 4 & 5	
Month	Samples	Month	Samples	Month	Samples	Month	Samples
Aug 57	8						
Sep 57	4			Sep 58	41	Sep 59	118
Oct 57	8			Oct 58	114	Oct 59	303
		Nov 57	79	Nov 58	99	Nov 59	208
		Dec 57	33	Dec 58	94	Dec 59	238
		Jan 58	56	Jan 59	82	Jan 60	225
		Feb 58	56	Feb 59	108	Feb 60	225
		Mar 58	99	Mar 59	124	Mar 60	225
		Apr 58	79	Apr 59	120	Apr 60	225
		May 58	62	May 59	106	May 60	225
		Jun 58	48	Jun 59	201		
		Jul 58	24	Jul 59	274		
		Aug 58	0	Aug 59	75		
total:	20	total:	536	total:	1438	total:	1992

Throughout Phase 1 of HASP, during August through October 1957, the sampling aircraft were based at Laughlin Air Force Base, Texas, for training and testing operations. A total twenty samples were obtained from the five missions flown during this phase. Two of these missions were vertical soundings, one was a northward, one a southward and one an eastward flight.

During Phase 2, from November 1957 through July 1958, the aircraft were based at Plattsburg Air Force Base, New York, and Ramey Air Force Base, Puerto Rico. The sampling corridor, extending from 67° North to 7° South latitude, was approximately along the 70° West Meridian. Sampling from Plattsburg was terminated in May 1958 and missions flown from Ramey during June and July 1958 were mostly vertical soundings. A total of 536 samples were collected during Phase 2.

Phase 3 began with the deployment of three HASP aircraft to Ezeiza Airport, Buenos Aires, Argentina, in September 1958 and ended with their return to the United States in August 1959. During this phase, which was designed to permit sampling of the stratosphere of the Southern Hemisphere, three aircraft continued to operate from Ramey. Once a month, during October through December 1958 and March through July 1959, the aircraft stationed at Ramey were deployed to Plattsburg to sample the northern reaches of the sampling corridor, which otherwise extended from 38° North to 57° South latitude along the 64° West meridian. A total of 1438 samples were collected during Phase 3.

Since the beginning of Phase 4, in September 1959, the aircraft have been stationed at Laughlin Air Force Base and at Minot Air Force Base, North Dakota. The sampling corridor extends from 71° North to 12°30' North latitude and is more or less centered on the 100° West meridian. Once a month the aircraft stationed at Laughlin are deployed to Ramey to extend the sampling into the Southern Hemisphere. Information gained during this phase should reveal the seasonal variations which

occur in the distribution of debris through the stratosphere of the Northern Hemisphere. Figure 6 shows routes during this phase which have slight east-west components. This move was necessary to improve operational reliability. It is not felt that this introduces any significant deterioration of the data since there are at present no large scale day-to-day variations in concentration within the stratosphere. By 10 May 1960 a total of 1792 samples had been collected during Phase 4.

Phase 5 will consist of spot checks in both the Northern and Southern Hemisphere. Samples are being taken during May and June 1960 operating fromEzeiza Airport at Buenos Aires. Concurrent samples will be obtained using four B-57 aircraft assigned to ARDC.

In addition to samples collected during HASP missions a number of other samples have been made available to HASP by the Air Force. These have included some collected north of Point Barrow, Alaska, 8 collected during September-October 1958 and 6 collected during April 1958. Also, to provide information on stratospheric concentrations between the northern limits of HASP sampling and the North Pole, three sampling missions were flown between 60° North and 90° North latitude, one in April 1959 yielding 14 samples, one in August 1959 yielding 15 samples and one in October 1959 yielding 15 samples, by B-52 aircraft equipped with hatch samplers.

Several types of missions have been flown during the course of HASP sampling. During Phase 1, Phase 2, and most of Phase 3 the aircraft were capable of collecting only 4 samples apiece during a single mission.

Since considerable importance was attached to the measurement of horizontal concentration gradients in the stratosphere, most missions were scheduled for collection of all four samples at a single altitude during either the outbound or inbound leg of the flight. Frequently all four aircraft flying a mission flew at the same altitude, one north and one south from each base. Occasionally the two aircraft from a single base flew in the same direction but at different altitudes. At other times missions were flown with two samples collected outbound and two inbound, especially at "maximum" altitude with the aircraft climbing throughout the entire flight. Vertical soundings or orbits were also flown from time to time. These involved collection of a series of samples by one or two aircraft at several different altitudes while circling a selected spot, usually near the base. To insure the detection of temporal changes in stratospheric concentrations, most HASP missions during the first months of Phase 3 were flown at a single altitude. By December 1958, however, the flight schedule was changed to provide periodic sampling at a series of altitudes between 40,000 feet and maximum. Flights south from Ezeiza were through the polar stratosphere and were scheduled for 40,000, 48,000, and 55,000. During the deployment of aircraft to or from Plattsburg each aircraft flew at a different altitude. Flights north from Plattsburg were scheduled for a series of altitudes between 45,000 feet and maximum altitude.

With the addition of hatch samplers to the aircraft based at Ramey in June 1959 the sampling capability became more flexible. Sampling was

scheduled for one altitude outbound and a second inbound. Since only four samples could be collected on one leg of the mission if adequate activity was to be insured, numerous simultaneous collections by both nose and hatch samplers were carried out. These were desired also for inter-calibration of the samplers and for reproducibility checks of the radiochemical analyses.

The sampling schedule for Phase 4 is similar to that for the last months of Phase 3. All aircraft are equipped with both nose and hatch samplers. Sampling altitudes range from the high tropical troposphere and low polar stratosphere at 40,000 feet to the maximum altitude of the aircraft.

During Phase 2 the least satisfactorily sampled areas were the higher altitudes in both the Tropical and Polar Stratosphere, though, except for 55,000 feet, no altitude in the Polar Stratosphere was well sampled. During Phase 3 the Northern Polar Stratosphere was not well sampled, primarily because winter weather at Plattsburg led to the curtailment of the missions originally scheduled to be flown from that base.

In Table 3 is shown the distribution with latitude of samples used to calculate the mean distribution of strontium-90 through the stratosphere in November 1957-December 1958 and January 1959-August 1959, respectively.

Table 3

Latitude Distribution of Sample Collections

Latitude	Nov 1957-Dec 1958	Jan-Aug 1959
70°-80°N	8	6
60°-70°N	29	22
50°-60°N	51	38
40°-50°N	99	59
30°-40°N	141	95
20°-30°N	149	173
10°-20°N	140	125
0°-10°N	73	123
0°-10°S	49	64
10°-20°S	26	60
20°-30°S	14	35
30°-40°S	61	169
40°-50°S	46	66
50°-60°S	14	24
Total	900	1056

The validity of conclusions reached in the High Altitude Sampling depends on the extent to which HASP samples are representative of nuclear debris in the stratosphere. As discussed below in this chapter, the calibration of the samplers and the measurements of filter efficiency permit an accurate and precise estimate to be made of the concentration of nuclear debris in those regions of the stratosphere which have been sampled by HASP. All evidence pertaining to the variations in the relative abundances of the various radionuclides from place to place within the stratosphere indicates that these variations are explicable in terms of the time and place or origin of the debris found at each place. The only factor which could seriously affect the calculation of

the stratospheric burden and stratospheric residence time of strontium-90 and which cannot be obtained through the analyses of the HASP samples themselves is the concentration of debris in regions of the stratosphere which have not been sampled by HASP. Such concentrations must be inferred from a consideration of HASP data and data from other programs of investigation of stratospheric fallout, such as Project ASHCAN and Air Force stratospheric carbon-14 measurements. The three polar flights by B-52 aircraft confirmed the validity of the assumption that no major variations in concentration of nuclear debris occur as a function of latitude at a given altitude between 60° North and 90° North latitude. There is no evidence that such variations occur in the Southern Polar Stratosphere either, and there is no reason to believe they should occur. Some uncertainty remains as to the concentrations of debris which occur above the maximum altitude of the aircraft, though ASHCAN data provide the basis for a fairly good estimate. The use of stratospheric carbon-14 data instead of ASHCAN data to make the estimate leads to a slightly different result but does not significantly change the conclusions. Moreover, measurements of strontium-90 in HASP samples collected at high altitudes in both the North and South Polar Stratospheres indicate that the maximum concentrations of debris occur in the lower stratosphere. If the meridional transfer processes which produced the observed stratospheric distribution of tungsten-185 apply also to layers in the stratosphere higher than those which contain the maximum concentrations of tungsten-185, the concentrations of strontium-90 in the lower Polar

Stratosphere may be used to judge the concentrations which occur in the Tropical Stratosphere. This comparison confirms the accuracy of the concentrations deduced from the ASHCAN data. It is concluded, therefore, that the data on stratospheric concentrations provided by the High Altitude Sampling Program are a sufficient basis for the accurate calculation of the quantity of debris present in the entire stratosphere.

Sampling Equipment

The basic sampling vehicle for HASP is the Lockheed U-2 aircraft. Six of these aircraft are fitted with a duct in the nose of the aircraft. Sampling starts when the pilot opens the nose door allowing ram air to enter the duct. The duct widens gradually to a 10 inch diameter at the paper location. The filters are secured in a ringholder with wire mesh fore and aft to provide rigidity and to prevent tearing of the paper. Four sample holders are placed in a circular rack by which the filters may be rotated sequentially into the duct by the pilot. Inflatable neoprene seals are secured against the filter holders to prevent leakage around the paper. Position lights indicate when the paper is positioned properly. The duct terminates behind the paper at a point on the bottom of the aircraft near a pressure minimum.

In a space behind the pilot a panel containing an altimeter, clock, temperature gauge and filter position indicator lights is photographed once every fifteen seconds after sampling starts. Since January 1960 this panel has had installed a pressure gauge to compare pitot pressure with duct inlet pressure. When calibrated, this gauge will indicate

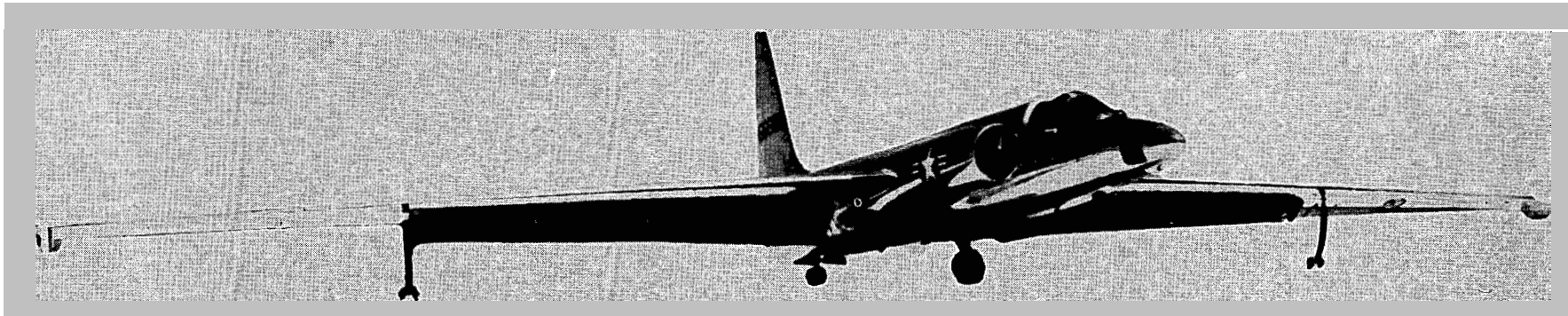
instantaneous flow rate and will be useful in detecting variations between various samples. At present the assumption is made that flow rates through all the filters is the same as that through the filter used in the original duct calibration. IPC work shows that there is very little varyability between individual samples of IPC 1478. (See Chapter III.)

In June 1959 an independent detachable hatch duct was installed on the aircraft. This duct is similar in design to the nose duct and provides a capacity of six additional sixteen-inch filter papers.

Besides the particulate filter ducts described above, two aircraft have been fitted with a dust impaction probe since January 1960. This probe is described more fully in Chapter IV.

Figure 7 shows some of the characteristics of the U-2. It is essentially a soaring plane powered by a Pratt and Whitney J-57 turbojet engine capable of up to 10,000 pounds thrust. The aircraft is operated by one man and has very limited communication and navigation equipment aboard. Its 3000 mile range at a speed of 475 mph operating up to 70,000 feet make it an ideal vehicle to visit the points in the HASP network. One peculiar feature is its tandem landing gear and removable outriggers. Upon take-off, these outriggers drop from the aircraft. Wing tip skids prevent damage when landing. Since every landing is a potential ground loop, cross-winds on the runway must be practically nil.

LOCKHEED U-2 AIRCRAFT



FEATURES

Wing Tip Skids — Pogo Sticks

Rear Gear - Dual Hard Rubber - Approx 8", Steerable through 6°

Forward Gear - Dual Pneumatic - Approx 20", Non-steerable

Thin Skin — Lightly Stressed

Length - 49' 8" — Wing Span - 80' 2" — One Man Crew

Speed - .73 to .8 Mach — Flight Duration - 7½ Hours

Altitude - 70,000 Ft

FIGURE 7

Duct Calibration Program

In September 1958, flight tests were carried out at Laughlin Air Force Base by Professor E. G. Reid of Stanford University to establish the flow rate characteristics of the Nose and Hatch air samplers of the U-2 airplane and to determine their dependence upon filter properties and meteorological conditions. Typical flow rate characteristics which typify the operation of both samplers when equipped with the normally-used Knowlton filters were developed.

The particulate samplers incorporated in the U-2 are filter-obstructed, internal, fuselage ducts through which ambient air is forced by flight-induced pressure differences. To determine the quantities of air sampled per unit time at various altitudes and speeds of flight, tests of two kinds were made. In one, flow rates were determined with the normally used Knowlton filters in the ducts; in the other, the filters were replaced by variously perforated metal plates which served as flow-metering devices and, by the variety of their perforation, enabled arbitrary fixation of the flow rates. Analysis of the results of the latter tests are expected to enable reliable prediction of the effects of meteorological conditions and filter properties upon such typical flow rate characteristics as those presented herein.

The determination of duct flow rates was accomplished by the use of precalibrated filters. The availability of suitable calibration data necessitated the measurement of only the pressure difference across the filter and the air temperature and pressure at its upstream face.

This method utilizes the analysis of Prof. Reid[6] wherein it is shown that the pressure loss coefficient, k, which characterizes the flow of air through a particular filter depends only upon the (pseudo) Reynolds number $\sigma V/\omega$, i.e.

$$k = f\ (\sigma V/\omega) \qquad \text{.....(1)}$$

and that this relationship may be alternatively stated in the form

$$\sigma\Delta p/\omega^2 = f\ (\sigma V/\omega) \qquad \text{.....(2)}$$

In these equations

k = pressure loss coefficient $(= \frac{\Delta p}{q})$

Δp = pressure difference across filter

q = dynamic pressure at filter face $(= \sigma\rho_o V^2/2)$

ρ = mass density of air

V = velocity of air at filter face

σ = relative density of air at filter face $(= \rho/\rho_o)$

ω = relative viscosity of air at filter face $(= \mu/\mu_o)$

and the subscript "o" is used to denote standard sea-level values. By inspection of (2), it will be seen that, when filter calibration data are available in the form of a curve of

$$\sigma\Delta p/\omega^2 \text{ vs } \sigma V/\omega$$

knowledge of the values of σ, Δp and ω suffices for determination of the corresponding values of $\sigma V/\omega$ - and V, if desired.

In this flight test, the pressure difference across the filter (Δp) and the absolute pressure at the filter face (p) were determined by direct measurement. The temperature at the filter face was assumed to

be equal to the total, or stagnation, temperature, T_t - which was also measured directly.* Since the value of σ depends only upon temperature and pressure, while that of ω is fixed by temperature alone, the recording of Δp, T_t and p sufficed for the determination of $\sigma V/\omega$ by use of the filter calibration curve.

To enable the correlation of flow rate determinations with flight conditions, the values of pressure altitude, indicated air speed, (total) air temperature and duct pressures were recorded simultaneously.

The flow rate characteristics of the U-2 samplers are qualitatively consistent with the results of previous wind tunnel calibrations of tip tank samplers and with analyses of the effects of altitude variation upon the flow rates which characterize fixed Mach number operation. Such quantitative peculiarities as are evident in the present results appear to be largely, if not entirely, explainable as consequences of:

a. The incorporation of samplers in an airplane fuselage - which results in some sacrifice of flow-inducing pressure difference,

b. Differences between the characteristics of currently used Knowlton filters and those of the previously used IPC type (although these appear to be small. See Chapter III),

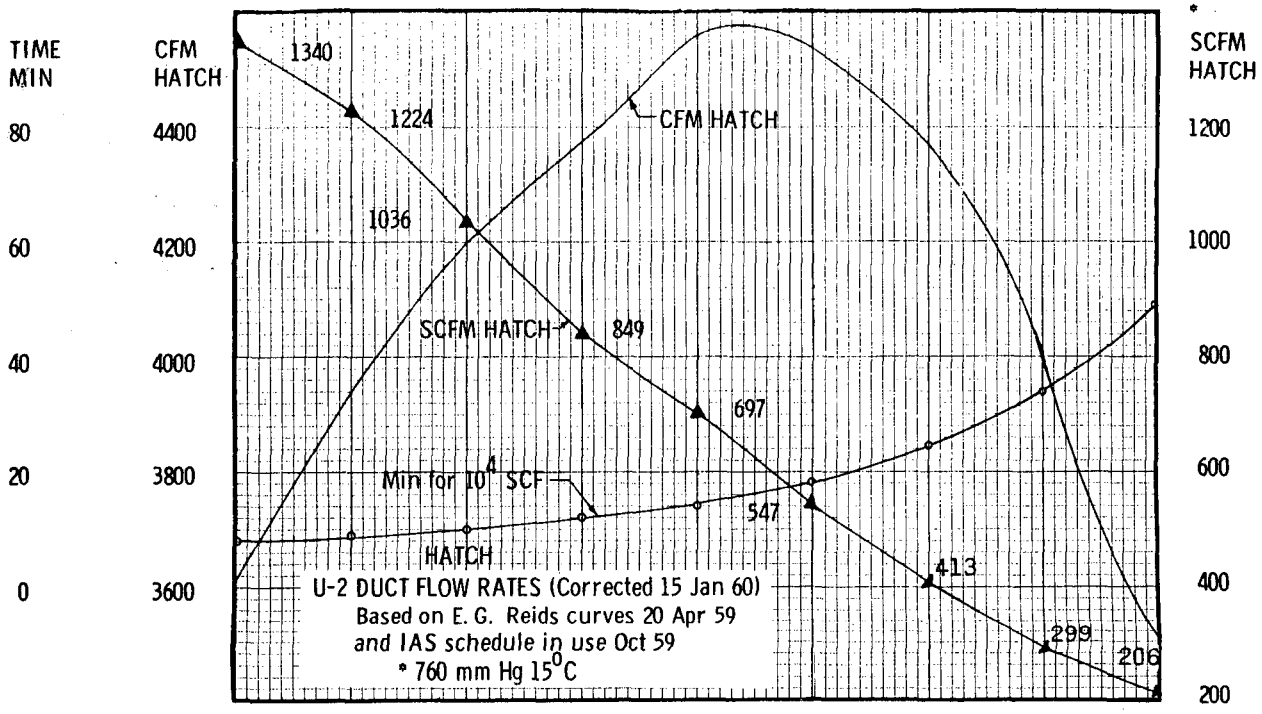
c. The prevalence during the flight of typically tropical upper air characteristics, and

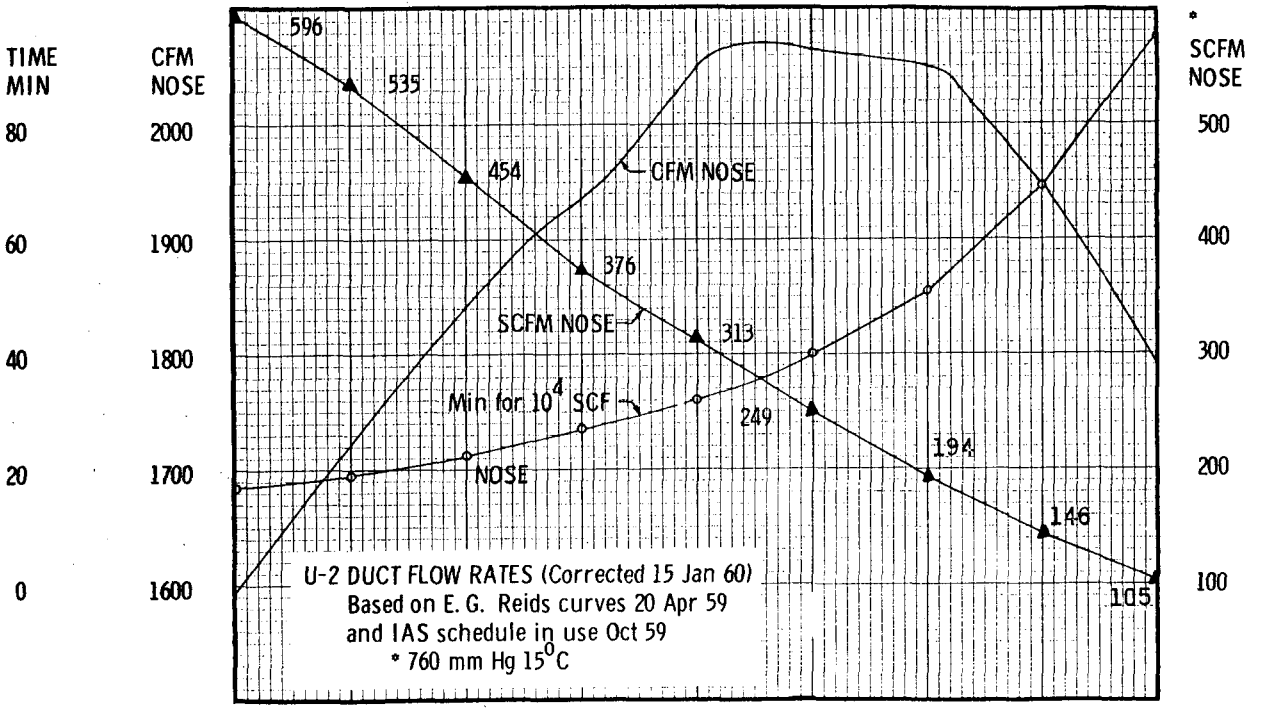
d. The relatively far aft and asymmetric location of the Hatch sampler.

* The maximum Mach number attained at the filter face was less than 0.1; when $M < 0.1$, $T/T_t > 0.998$.

Wind tunnel calibration of the pressure drop-flow rate characteristics of the filter paper was performed at the NASA, Ames Laboratory, Moffett Naval Air Station, California. The overall error of flow rate is felt to be no more than 20%. Figure 8 shows flow rates based on Professor Reid's work. Multiplication of the appropriate value of [illegible] by the exposure time of the filter gives the volume of ambient air sampled over the flight path. This volume is converted to the equivalent volume at standard conditions by correction for air density. Some recent simultaneous exposures of hatch and nose papers have yielded data which tend to show that the hatch is slightly more efficient than indicated by Professor Reid's flow rate curves especially at higher altitudes. Figure 9 is a typical vertical sounding showing the apparent increased sampling rate of the hatch at higher altitudes.

The ratio of activities collected by these filter pairs may be compared with the ratio of volumes of air sampled by the filters as calculated from Reid's data on flow rates for the two samplers. As long as the filter papers have identical pressure drop-flow rate characteristics the ratio of activities will be strictly equal to the ratio of the flow rates for level flights. The calculated, or "predicted", ratios of flow rates were compared with the strontium-90 ratios measured in 100 selected pairs of simultaneously exposed hatch and nose filters (Table 4).





ACT. ALT.	30,000	35,000	40,000	45,000	50,000	55,000	60,000	65,000	70,000
IAS CORR.	7%	7%	7%	7%	7%	8%	9%	10%	11%
IAS	175	173	170	165	160	150	135	121	108
TAS	276	305	334	362	392	410	415	419	420
IND. TEMP.	-40	-40	-40	-38	-36	-34	-34	-33	-33
MACH NO.	.48	.53	.58	.63	.68	.71	.72	.73	.73

FOR EVERY 10 KNOTS IAS HIGH, ADD IAS CORR. % TO CFM AND SCFM
FOR EVERY 10°COLDER IND. TEMP., ADD 4.75 % TO SCFM ONLY

FIGURE 8

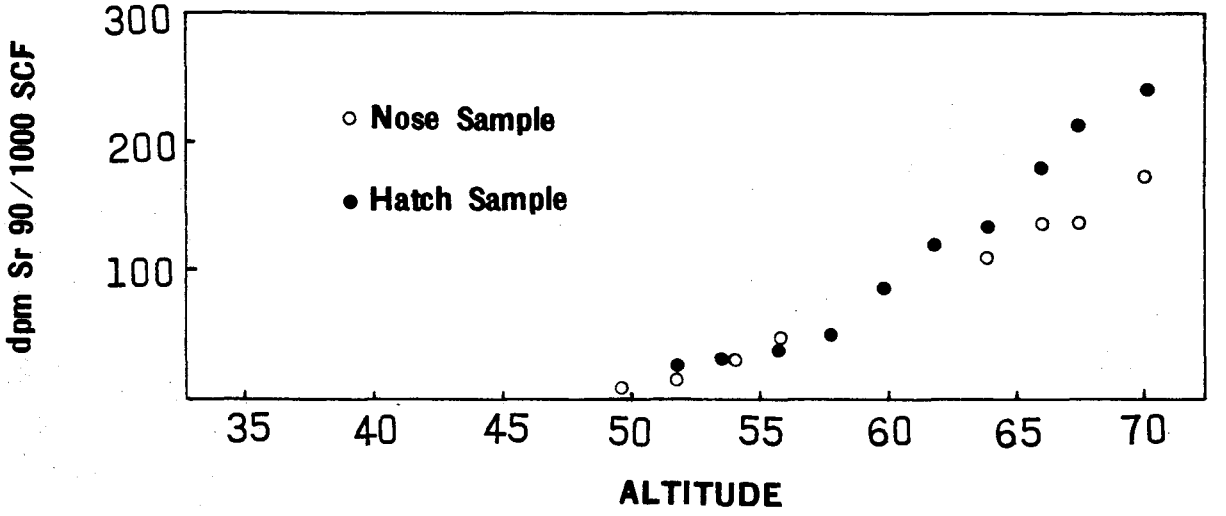


FIGURE 9 Hatch and Nose Samples in a Vertical Sounding

Table 4

Comparison of Nose and Hatch Activities

$\frac{\text{dpm } Sr^{90} \text{ Hatch}}{\text{dpm } Sr^{90} \text{ Nose}}$	No. of Sample Pairs
2.10	7
2.10-2.20	11
2.20-2.30	9
2.30-2.40	13
2.40-2.50	17
2.50-2.60	16
2.60-2.70	6
2.70-2.80	10
2.80-2.90	5
2.90	6
Total	100

Measured Average 2.46 ± 0.26

Predicted Average 2.10 ± 0.05

Of the 100 measured ratios of hatch sample activity to nose sample activity only fourteen were equal to or less than the corresponding calculated ratios of air flow through the hatch sample to air flow through the nose sample. The calculated air flow ratios for the selected samples averaged 2.10 ± 0.05. The average of the 100 measured activity ratios was 2.46 ± 0.26. The ratio of the areas of the exposed portions of the hatch and nose filter papers is 2.54. The conclusions which are statistically valid are:

1. The hypothesis that the measured ratio is equal to the predicted ratio is rejected.

2. The hypothesis that the measured ratio is equal to the ratio of the exposed areas of the filter paper cannot be rejected on the basis of the data analyzed.

3. There appears to be a systematic discrepancy between the predicted air flow ratio and the measured activity ratio (which is considered to be the actual air flow ratio). The predicted ratio is too low by about 20 percent.

It is believed that the discrepancy between the calculated and measured ratios of hatch sampler to nose sampler flow rates is probably due to the fact that, during the calibration flight, the measurement of the pressure at the upstream face of the nose filter was affected by adverse changes in instrument temperature. These, in turn, could have given rise to a faulty calculation of the flow rate through the sampler. It is hoped that this discrepancy will be resolved in the recalibration effort to be made during Phase 5. It should be noted that no weighting has been given the concentrations in the hatch papers, consequently the inventory since 1 June 1959 may be as much as 10% too high relative to the pre-1 June 1959 inventories.

CHAPTER III

CHARACTERISTICS OF IPC PAPER 1478

Introduction

Two aspects of IPC Paper 1478 that are of greatest interest to the HASP experimenters are the flow rate characteristics and the retention characteristics. Dr. J. A. Van den Akker of The Institute of Paper Chemistry has reported on both characteristics.[5] In addition Dr. James P. Friend of Isotopes, Incorporated has investigated the retention characteristics of stratospheric particulates. These investigations will be described here.

The IPC Experiment

This work was conducted to study the filtration and pressure drop - flow rate characteristics of IPC 1478 filter paper over wide ranges in pressure and temperature simulating as well as possible the HASP sampling conditions. The project was done in two phases: In the first, a wind tunnel was designed and constructed for operation at a pressure of 1/20 atm., a temperature of 7°F. (simulating a stagnation temperature) and a flow rate of 5000 ft./min. at 1/20 atm. The aerosol was monodisperse spherical particles of SiO_2 whose size distribution function (mass basis) was narrow, peaking at about 0.02 micron. The retention of IPC 1478 paper for these particles was found to be essentially 100%. The study revealed an unusually high diffusion of these particles (and also of a boron oxide aerosol) in air. In the second phase, the wind

tunnel was modified for operation at temperatures down to -40°F., a generator for the production of very small (about 40 A.) particles of aluminum oxide was developed, and a General Electric Continuous Condensation Nuclei Detector was employed for particle counting and retention determination. Retention (generally in the range 30-75%) for the aluminum oxide particles and pressure drop were determined over wide ranges in pressure, temperature, and flow rate. Data reduction theory was developed, and applied to the pressure drop-flow rate observations. Modern kinetic theory was applied to the diffusion of very small particles in a gas, and the theory of diffusion of particles from a streaming gas to the tube was evolved and applied to the estimation of particle size.

The development of a filter paper having good retention for small airborne particles, high air permeability, minimum ash content and low radioactivity was undertaken in 1947. The filter paper known as "IPC 1478" evolved from this work in 1949. The paper was made from second-cut cotton linters, Grade 46, obtained from the Hercules Powder Company. The fibers were lightly beaten and the sheets were formed on large, specially made, handsheet molds. Cotton scrim was used as a backing material to add strength to the sheet. The sheets were impregnated with "Kronisol" (di-butoxyethyl phthalate) for the purpose of improving the retention. Tests performed at atmospheric pressure with a 2000-ft./min. air velocity, at room temperature and -40°F., employing a carbonyl-iron aerosol, showed that the retention was improved markedly after

impregnation with the "Kronisol." The retention, for the conditions of testing, was essentially 100% at both room temperature and -40°F. The filter paper was produced on the handsheet mold until 1956, when paper machine production was initiated by Knowlton Brothers, Watertown, New York.

Wind Tunnel

The closed-system, recirculating wind tunnel was given first consideration. Calculation showed that an unusually powerful turbine-type compressor would be required for the range of pressure differentials when operating at an air density of only 5% of normal. On turning to positive-displacement compressors and vacuum pumps it was found that a two-stage Nash "Hytor" vacuum pump would meet the requirements. Calculations and reference to the characteristics of this pump showed that it could be used on an open-ended wind tunnel. Several advantages then became apparent in favor of this type of wind tunnel. Probably the greatest advantage is that particles which pass through the test filter are simply discharged, and no special means for their removal is necessary as in the operation of a recirculating tunnel.

Tunnel, Construction Details

The open-ended wind tunnel constructed for this work consists of a linear array of components, shown in Figs 10 and 11, which are connected with 2-inch copper tubing of suitable wall thickness. The air from the room enters the tunnel through a filter, consisting of 3 plies of IPC

The Modified IPC Wind Tunnel

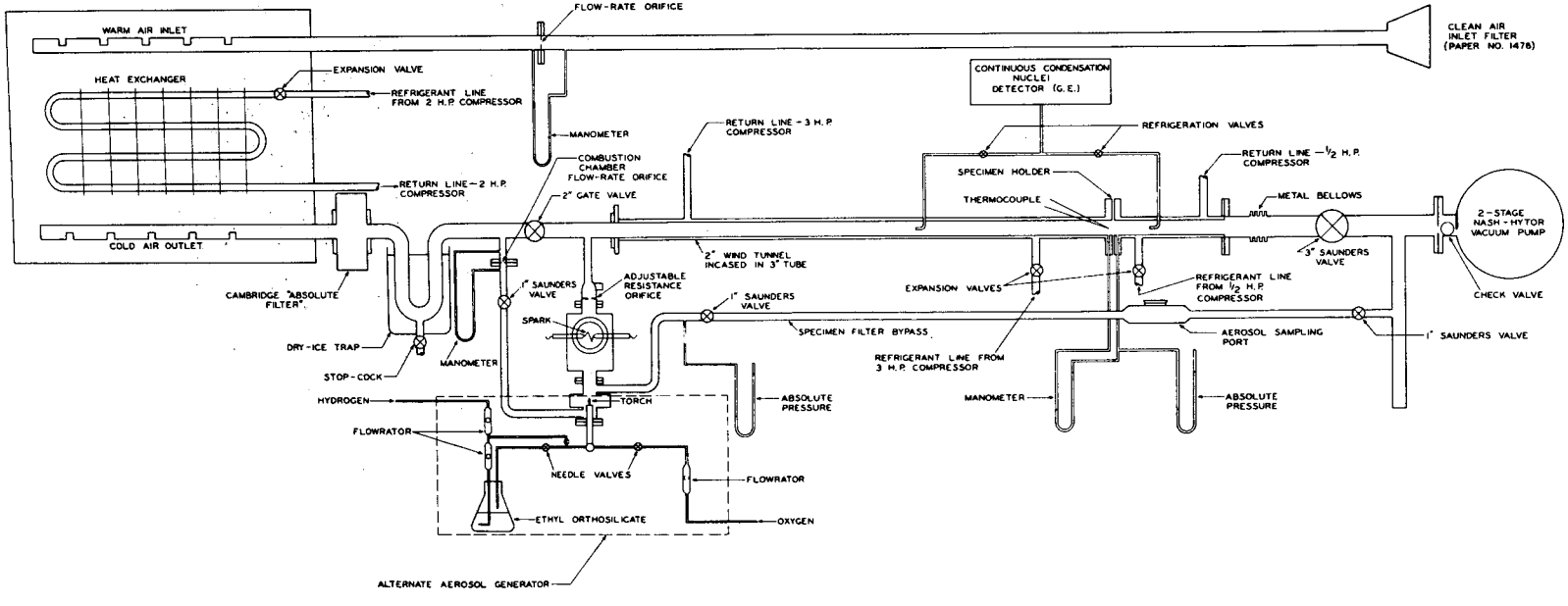


FIGURE 10

The Modified IPC Wind Tunnel

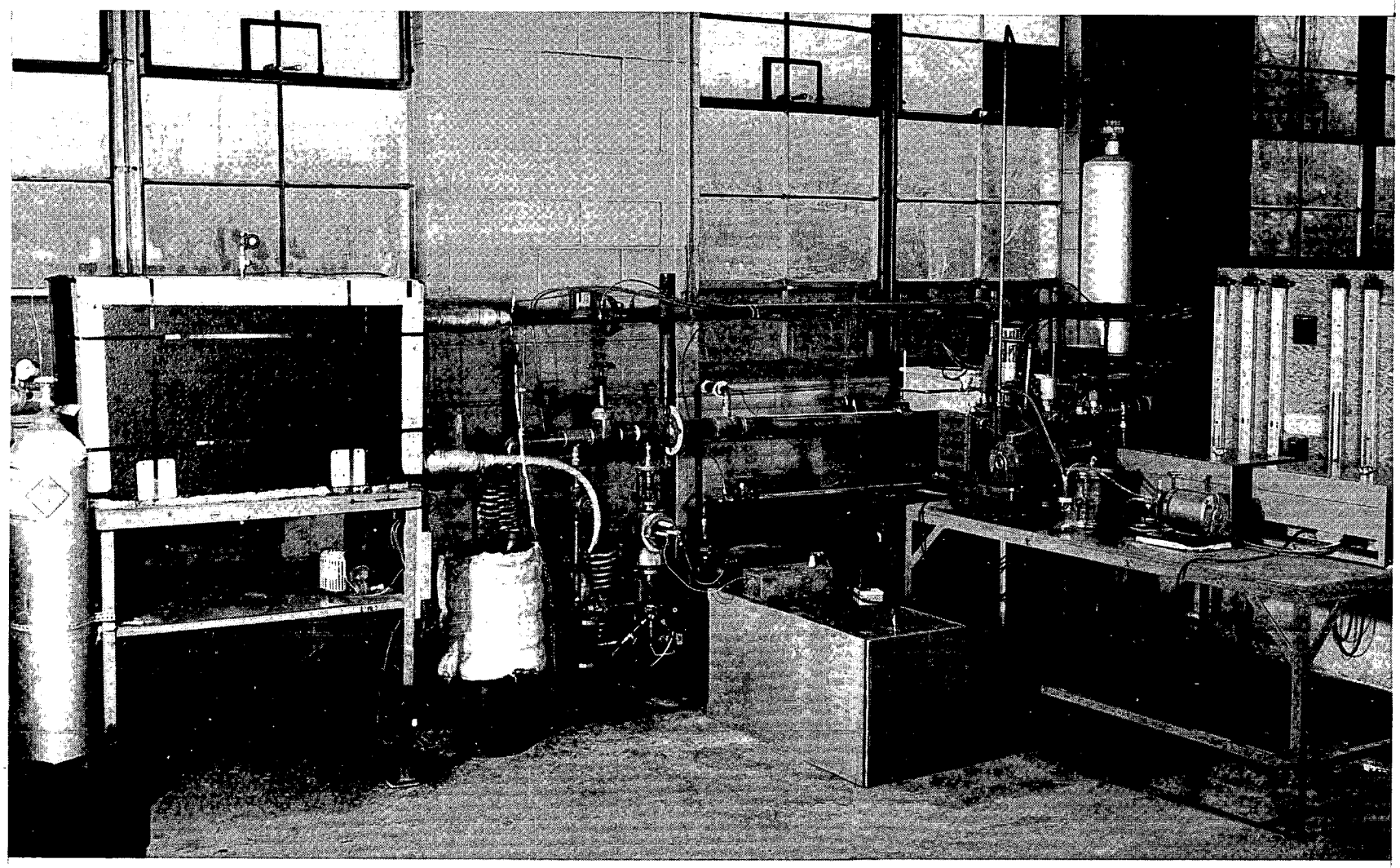


FIGURE 11

1478 paper, which removes most of the particulate matter from the air stream. (The area of the filter is about 20 times that of the test specimen.) The air then passes through an orifice-type flowmeter, calibrated to give the mass rate of flow in the tunnel. To substantiate the accuracy of the orifice design and calculations, volumetric checks of the two orifices used in the study were made. The air next moves through a heat exchanger which reduces the temperature to about -30°F.; most of the atmospheric moisture is removed at this point. The cross-sectional area of the heat exchanger is so large that the air at essentially atmospheric pressure moves through it at very low velocity.

A 2-inch Saunders valve located at the outlet of the heat exchanger together with a second valve at the vacuum pump serve to regulate the flow rate and pressure in the tunnel. A portion of the air is by-passed around the Saunders valve through the aerosol generating chamber. Two 1-inch Saunders valves located up and downstream from the aerosol chamber serve to regulate the absolute pressure and air flow in the chamber. An orifice flowmeter located upstream from the chamber indicates the mass rate of flow of air through the chamber. A mercury manometer serves to indicate the absolute pressure in the chamber.

Modifications were eventually made in that portion of the wind tunnel between the heat exchanger and the Nash "Hytor" vacuum pump. A Cambridge "Absolute Filter" was installed in the exit line from the heat exchanger to remove any particles from the air stream; a very low "clean air" or aerosol-free count was desired in the work with the CN

Meter. A moisture trap consisting of a large "U" tube immersed in a bath of alcohol and "dry ice" was installed just downstream from the Cambridge filter. The heat exchanger drops the air temperature to about -30°F. and removes most of the moisture. The "dry ice" bath further reduces the air temperature to about -50°F. and removes more moisture so that the relative humidity of the air at -40°F. is well below saturation. The 2-inch Saunders valve was replaced with a 2-inch gate valve as the rubber diaphragm in the former failed to function at the lower temperatures. A 6-foot section of the 2-inch tunnel (upstream from the filter specimen) was enclosed in a 3-inch copper tube. Freon-22 was circulated in the annular space between the two tubes by a 3-horsepower refrigeration unit. An 18-inch section of the 2-inch tunnel (downstream from the filter specimen) was similarly enclosed within a 3-inch copper tube with Freon-22 circulated in the annular space between the two tubes by a 1/2 horsepower refrigeration unit.

The aerosol generator was modified in that the volume of the torch chamber was reduced and a second and new chamber was added which permitted the introduction of an aerosol generated by means of a high-voltage, low-current spark employing electrodes of elements of interest.

Freon-12 was circulated through copper tubing wrapped around the 2-inch tunnel to cool the air to about 7°F. The tunnel was insulated with fiberglass and was vapor sealed with a polyethylene film. The temperature of the air stream at the specimen was measured with a thermocouple consisting of 0.004-inch Chromel, Alumel wires. The junction

was located centrally in the 2-inch tunnel about 1/4 inch upstream from the filter specimen. The small diameter wire minimizes the effects of radiation and conduction, thus providing a true measure of air temperature. The termocouple was calibrated through the temperature range of interest using a brine solution and several mercury thermometers.

The 2-stage Nash "Hytor" vacuum pump driven at 1100 r.p.m. by a 15-horsepower electric motor moves the required volume of air at 3.8 cm. Hg absolute pressure in the tunnel. The volume rate of flow corresponding to a velocity of 5000 ft/min. in the 2-inch tunnel is 109 c.f.m.

The aerosol generator consists of a 4-inch diameter cylinder about 20 inches long, standing in an upright position, with an oxyhydrogen torch located at the bottom. The chamber is connected to the tunnel, as stated earlier, so that a portion of the air stream is drawn from the tunnel and then is returned to the tunnel after admixture with the particles from the flame. The air enters the bottom of the cylinder and rises past the torch. The torch barrel is insulated from the cold air so that vapors carried through the barrel will not condense on the inner surface of the latter. The oxygen and hydrogen supply lines are equipped with regulators, rotameters, and needle valves to permit control of the flow rates. A small portion of the hydrogen bubbles through an organic liquid (ethyl orthosilicate) containing silicon atoms. The vapor from the ethyl orthosilicate is carried to the oxyhydrogen flame, where the reactions result in the production of silicon dioxide. Particles generated in this manner are carried up through the chamber

and into the tunnel where the stream is mixed with and carried down the tunnel by the main air stream.

Electrostatic Precipitators

Electrostatic precipitators were employed in attempts to capture particles on grids for viewing with the electron micorscope. The precipitator consists of a 1-inch diameter copper tube about 3 inches long with a 1/8-inch polished brass rod located centrally in the tube. The supporting 3/8-inch diameter copper rod and the 1/8-inch brass rod project through a 1/2-inch thick by 3-inch square piece of Glyptal-painted transite which forms the seal over the 2-inch opening in the aerosol sampling port. The electron microscope grids are placed in slight recesses on a thin, flat brass plate which was positioned on the inside of the 1-inch diameter copper tube. The recesses are just slightly deeper than the thickness of the grid so that the top surfaces of the grids are essentially on the same plane as the surrounding surface.

One electrostatic precipitator was placed upstream and another downstream from the filter specimen; thus, the retention of the specimen could be determined by counting the particles collected on the grids in the two precipitators. It was necessary to warm the precipitators to approximately room temperature before removing them from the cold tunnel to avoid condensation of moisture on the grids. This was done by placing a soldering iron (tip removed) over the 3/8-inch copper rod supporting the precipitator for several minutes before removing the precipitator.

Positive and negative potentials from 800 to 2000 volts were applied to the cylinders of the precipitators, and positive and negative potentials of 7000 volts were applied to the torch flame. At times the electrostatic field seemed to influence the collection efficiency but, after performing many experiments, the conclusion was that the electrostatic field had very little influence on the collection efficiency for the very small particles involved in the study. Whatever the mechanism of collection, very satisfactory samples were obtained for electron microscope examination.

Electron micrographs permitted determination of the dimensions and shape of the particles. The earliest tests showed serious agglomeration. Through reduction of aerosol concentration, which was made possible after improvement in techniques, the agglomerates were eliminated and the particles produced were sperical and of uniform dimension. Figure 12 shows the particle size distribution after eliminating the agglomeration. For Fig. 12 the concentration was 10 to 20 micrograms of silicon dioxide per cubic meter. This concentration was determined by analyzing the filter specimen spectrographically for silicon after passing a known volume of aerosol-laden air through the filter. Calibration curves for spectrographic analysis were prepared using filter specimens containing known added amounts of silicon. The quantity of silicon contained in the original filter paper was accounted for in the calibration.

Continuous Condensation Nuclei Detector

In view of the lengthy procedure involved in sampling the aerosol

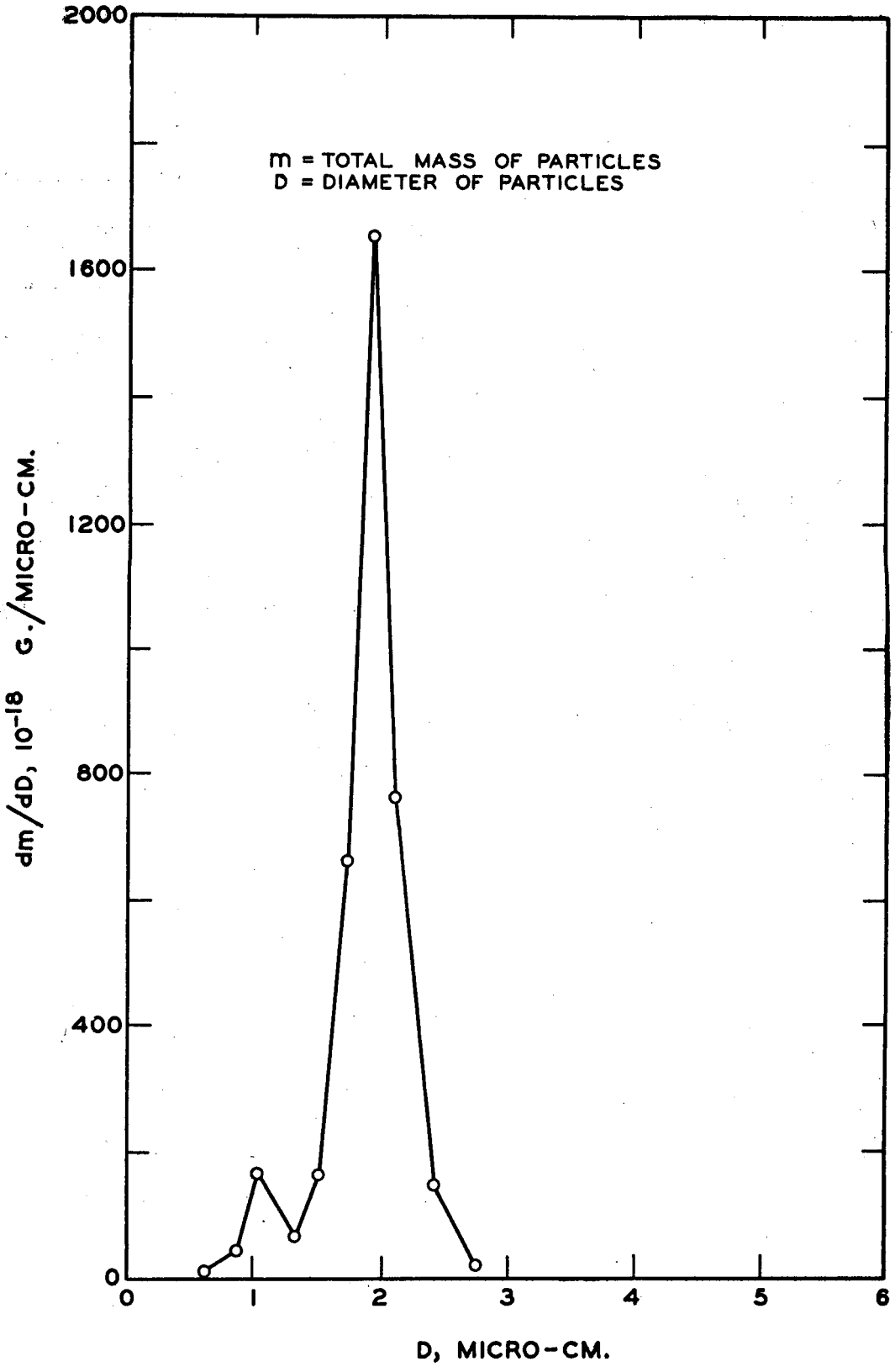


Figure 12 Particle Size Distribution for Silicon Dioxide Aerosol After Eliminating Agglomeration

for viewing with the electron microscope, and the difficulties in maintaining the necessary resolving power of microscope, other means of detecting the aerosol were sought.

The CN Meter was received from the General Electric Company early in July, 1958. This device provides a very sensitive* means for counting extremely small particles. The air sample input to the CN Meter should be essentially at atmospheric pressure and, hence, it was necessary for the General Electric Company to design and build a sampling device which would remove air from the wind tunnel at 3.8 cm. Hg and pressurize it to atmospheric pressure before introduction to the CN Meter.

Wind Tunnel Conditions for Retention Tests

Numerous retention tests were made with IPC 1478 paper employing the silicon dioxide aerosol. Tests were also made with a 0.003-inch thick perforated brass plate (in place of the filter specimen) which had a sufficient number of 1-millimeter holes to give a pressure drop very nearly equal to that for the filter paper for the same flow conditions. The micrographs taken up and downstream at the velocity and pressure of interest indicated that the number of particles collected downstream was very nearly equal to the number collected upstream. This was done to assure that the collection efficiencies of the grids in the up and downstream positions were very nearly equal for the conditions of the test. It was presumed that very few particles would be stopped

*An illustrative range of operation: 1×10^{-10} to 30×10^{-10} gram per cubic meter of air for 40-angstrom particles having a density of 4.0 g./cc.

by the perforated brass sheet.

The conditions for one illustrative determination for IPC 1478 paper, with reproductions of the electron micrographs, is presented in Fig. 13.(a) and (b) designate, respectively, up and downstream positions.

During the experimental phases of this work, more than four hundred grids were exposed, "shadowed" and examined with the electron microscope. After perfecting all the techniques involved, sixteen determinations were very carefully conducted; excluding runs in which agglomeration was obviously a factor (high concentrations), retentions of essentially 100% were found for the IPC 1478 paper. The high collection effeciency appears to be due to diffusion at an unusually high rate.

Aluminum Oxide Aerosol

An electric spark, employing aluminum wire electrodes, was set up with a 15,000-volt, 60-cycle transformer. A 10.5 megohm resistor connected in series with the spark limited the current through the spark to 1.4 ma. The number of particles generated per unit time could be controlled by adjusting the current-limiting resistor. The diameter of the wire electrodes was about 2mm. and the spark gap was set to about 1.5 mm. After a short "conditioning period" the rate of generation of particles became fairly constant. The particles generated in this manner undoubtedly consist of aluminum oxide with a density of 3.5 to 4.0. These particles could be transported through the sampling device to the CN Meter in sufficient numbers to permit making a reliable retention measurement.

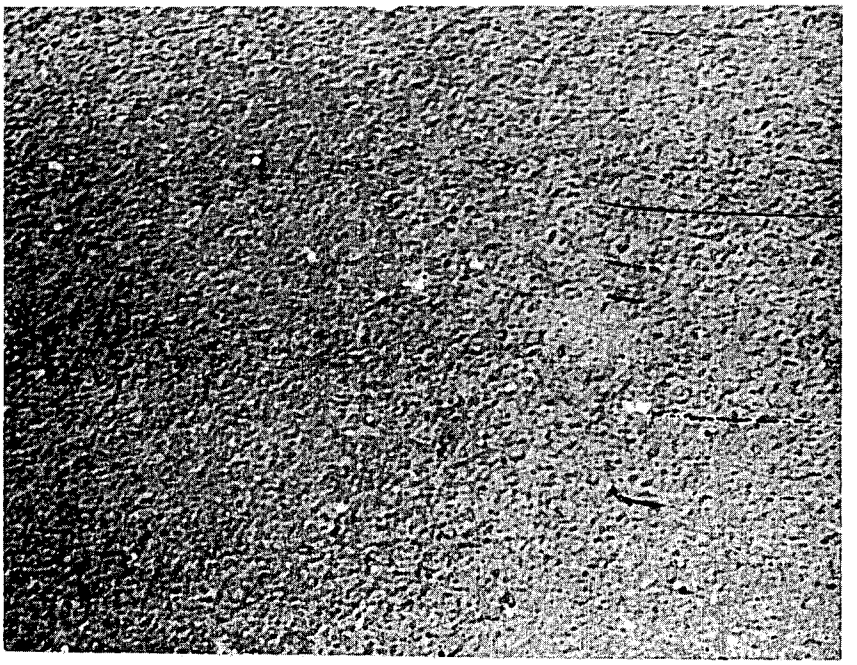


Figure 13b

Electron Micrograph of Silicon Dioxide Aerosol Collected Downstream from IPC 1478 Filter Specimen - Magnification 50,000X

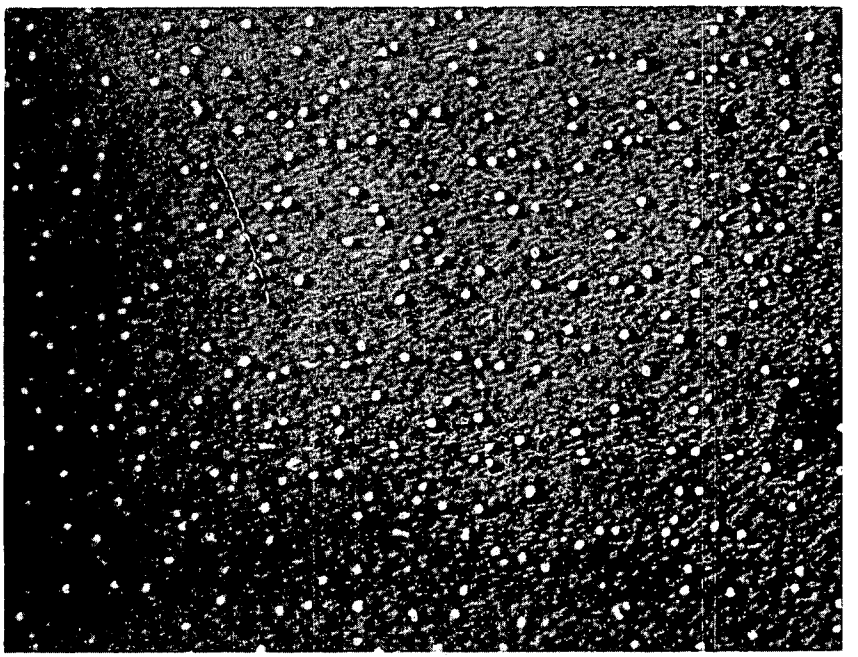


Figure 13a

Electron Micrograph of Silicon Dioxide Aerosol Collected Upstream from IPC 1478 Filter Specimen- Magnification 50,000X

Upstream grid	287-1
Downstream grid	288-1
Magnification (this reproduction)	50,000 X
Downstream velocity	4000 ft./min.
Downstream absolute pressure	4.7 cm. Hg
Absolute pressure in torch chamber	28.8 cm. Hg
Voltage on electrostatic precipitator cylinder (-)	1000 volts
Collection time	24 min.
Voltage on torch (+)	7000 volts
Pressure drop across filter specimen	9.8 inches H_2O
Air temperature at filter specimen	$0^oF.$
Filter specimen	IPC 1478
Retention, mass basis	98%

Attempts were made to determine the particle size and shape through use of the electron microscope, but without success. Apparently the particles were either too small to be visible or they were not being deposited on the grids. The collection time for these experiments was lengthened to several hours with no evidence of the particles showing on the grids.

The dimension of the particles was determined by measuring the rate of diffusion loss upon passing through tubing of known dimension and at a known air velocity. This determination led to an estimate of 40 angstroms.

The aerosol concentration for the retention tests employing the aluminum oxide aerosol ranged from a minium of about 1×10^{-10} gram per cubic meter of air to a maximum of about 30×10^{-10} gram per cubic meter of air. These concentrations are based on a particle density of 4.0 and an average particle diameter of 40 angstroms. This estimate of concentration in the wind tunnel does not consider the loss of particles in the sampling device. It is essential that the concentration of aerosol be sufficiently low that retention measurements will not be influenced by particle agglomeration or by loading of the filter.

Retention of Aluminum Oxide Aerosol

The retention of the aluminum oxide aerosol depends upon the conditions, but is generally in the range, 30-75%. It should be recalled that these particles, too small to be seem on the electron micrographs (if, indeed, a sufficient number were collected on the

grids for observation), were estimated to be extremely small (about 40 A.). The dependence of retention on velocity and pressure appears to be in good accord with filtration theory.

Pressure Drop-Flow Rate Relationship

When a filter is employed under widely different conditions covering substantial ranges in temperature, absolute pressure, flow rate, and pressure drop, it is, of course, desirable to have a general relationship between the frictional pressure drop across the filter and the flow rate. Reid[6,7] saw the need for this and in 1949 carried out an analysis for the reduction of data. His theory is based on the analogy between the pressure loss coefficient of a duct obstruction and the drag coefficient of a solid body. Accordingly, the velocity term appearing in Reid's theory was taken by him to be the velocity of the air at a point immediately upstream from the filter; consistent with this, the density and viscosity of the air were taken to be these quantities at an upstream point.

A modification of Professor Reid's treatment has been evolved by Dr. Van den Akker.

On the basis of his theory, a relationship should exist between $\bar{\sigma}\Delta p/\omega^2$ and $\bar{\sigma}\bar{V}/\omega$ over wide ranges of temperature and pressure, where $\bar{\sigma}$ is the arithmetic mean of the upstream and downstream densities of the air, Δp is the pressure drop across the filter, ω is the viscosity of the air, and $\bar{V}$ is the harmonic mean of the upstream and downstream velocities, i.e., $\bar{V} = 2V_1V_2(V_1 + V_2)$, in which V_1 and V_2 are, respectively,

the up and downstream velocities. In the interest of maintaining consistency with past work in the presentation of variables, reduced quantities, $\bar{\sigma}\Delta p/\omega^2$ and $\bar{\sigma}\bar{V}/\omega$, have been employed; these are analogous, but not equal, to Reid's reduced pressure drop, $\sigma\Delta p/\omega^2$, and "pseudo Reynolds' number," $\sigma V/\omega$. In these numbers, $\sigma = \rho/\rho_o$ and $\omega = \mu/\mu_o$, where the subscript zero refers to a reference condition of temperature and pressure (59°F., 2116.2 lb./sq. ft.--the NACA standard atmosphere at sea level). In the present work, $\bar{\sigma}$ is calculated as of the temperature of the air stream and the arithmetic mean of the up and downstream pressures, whereas in Reid's work the calculation is made for a point immediately upstream from the filter.

Using 1450 experimental points and fitting the best quadratic using an IBM 610 computer a relationship was evolved. This relationship, with $x = \bar{\sigma}\bar{V}/\omega$, and $y = \bar{\sigma}\Delta p/\omega^2$, is:

$$\log y = -0.0388 + 1.0717 \log x + 0.1577 (\log x)^2.$$

The machine computation indicated a standard deviation in log y of 0.0254, which corresponds approximately to 6% in y (for the machine-made papers). Figure 14 shows the fit to the plotted points.

The Isotopes, Incorporated Experiment

During the course of the HASP work at Isotopes, Inc., a few attempts have been made to gain a knowledge of the physical properties of the stratospheric particulate material which contains the fission products. One set of experiments which we have performed has consisted of locating

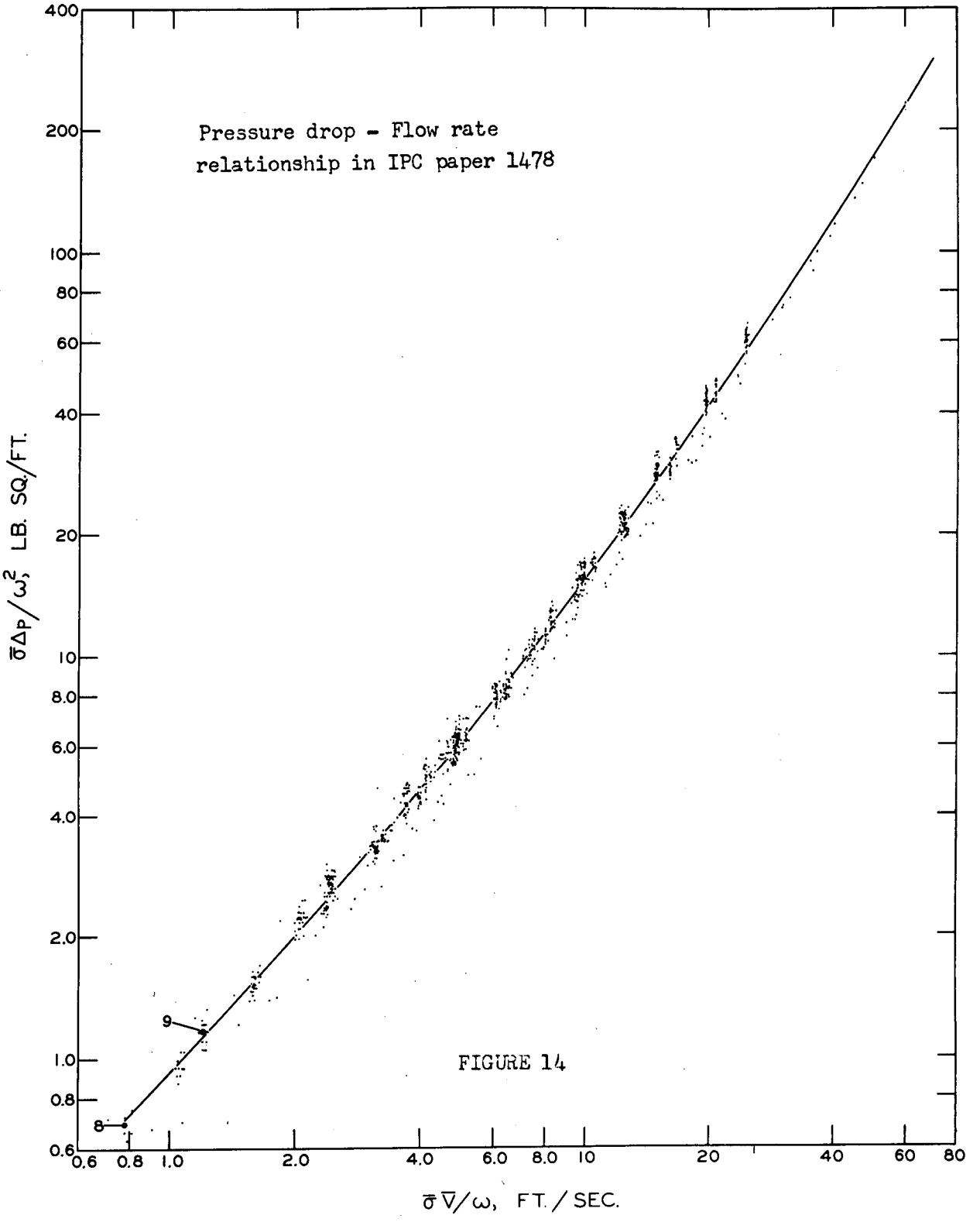

Pressure drop - Flow rate
relationship in IPC paper 1478
400
200
100
80
60
40
20
10
8.0
6.0
4.0
2.0
1.0
0.8
0.6
9
8
0.6
0.8
1.0
2.0
4.0
6.0
8.0
10
20
40
60
80
$\bar{\sigma}\Delta p/\omega^2$, LB. SQ./FT.
$\bar{\sigma}\bar{V}/\omega$, FT./SEC.

FIGURE 14

"hot spots" on the filter samples by autoradiography, cutting out those sections of the filter, embedding them in plastic, microtoming the embedded filter into thin sections, and then taking an autoradiograph of the thin sections. In the few studies which have been completed, it has been found that very few radioactive particles penetrate more than 90% of the way through the paper. Figure 15 shows the general relationship between the activity per thin section of the filter paper and the distance of the thin section from the upstream face of the filter. In view of the importance of experiments of this type, in confirming the 100% retention of radioactive particles by the paper, several samples are currently being studied by this technique. From these it is expected that quantitative estimates will be made of the extent to which the radioactive particulate material penetrates the filter paper.

An experiment was performed in which several coated electron microscope grids, attached to the upstream face of a filter paper in a Lockheed U-2 nose sampler, were exposed to stratospheric air during a HASP sampling mission. The purpose of the experiment was to collect stratospheric particles in such a manner as to allow examination of the particles by electron microscopy. Subsequently, electron micrographs of these grids were obtained which showed many small spherical particles in the range of 0.01 to 0.06 microns. These are shown in Figure 16. These particles were classified according to diameter. Upon the assumption of a log normal distribution it was calculated

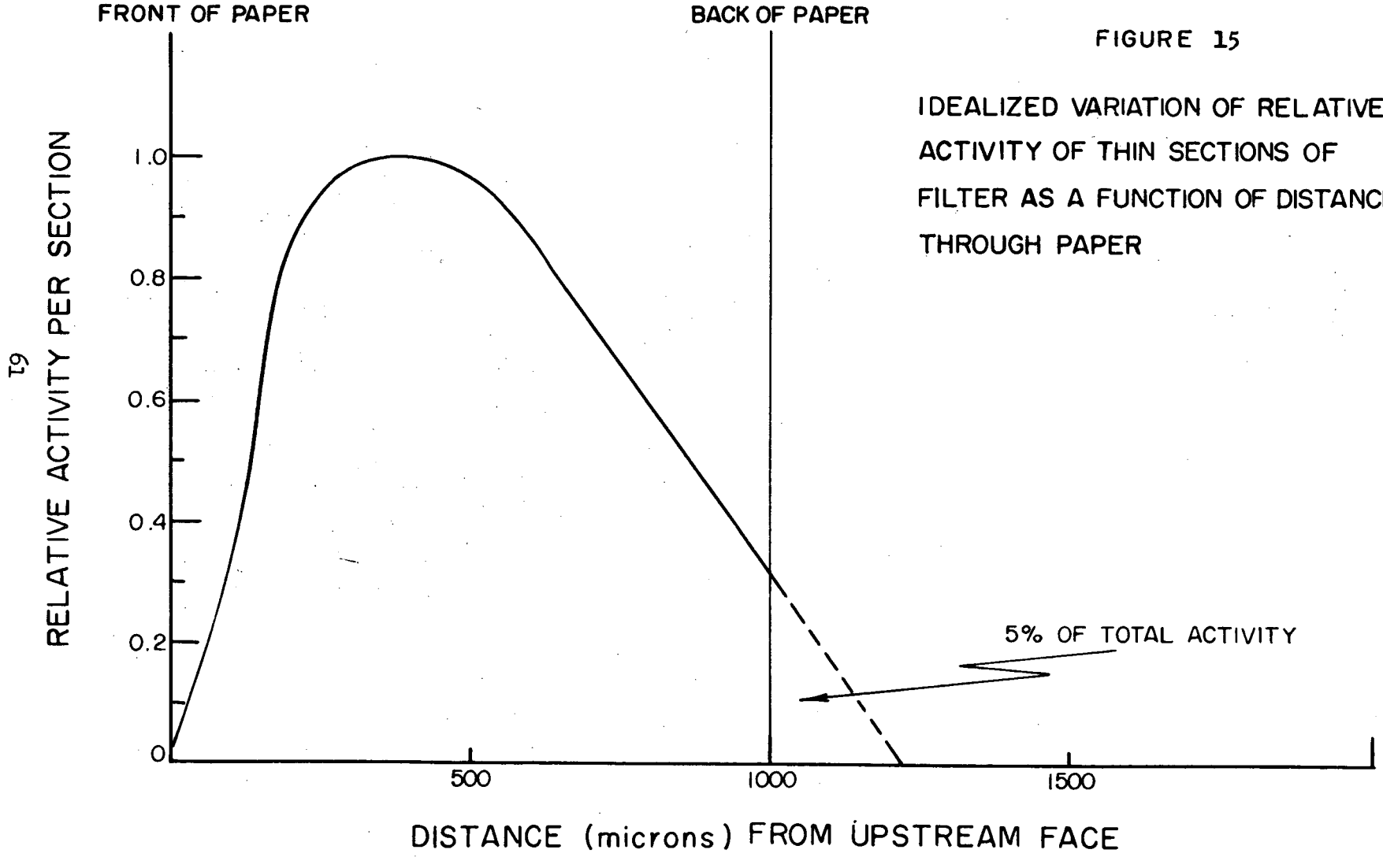
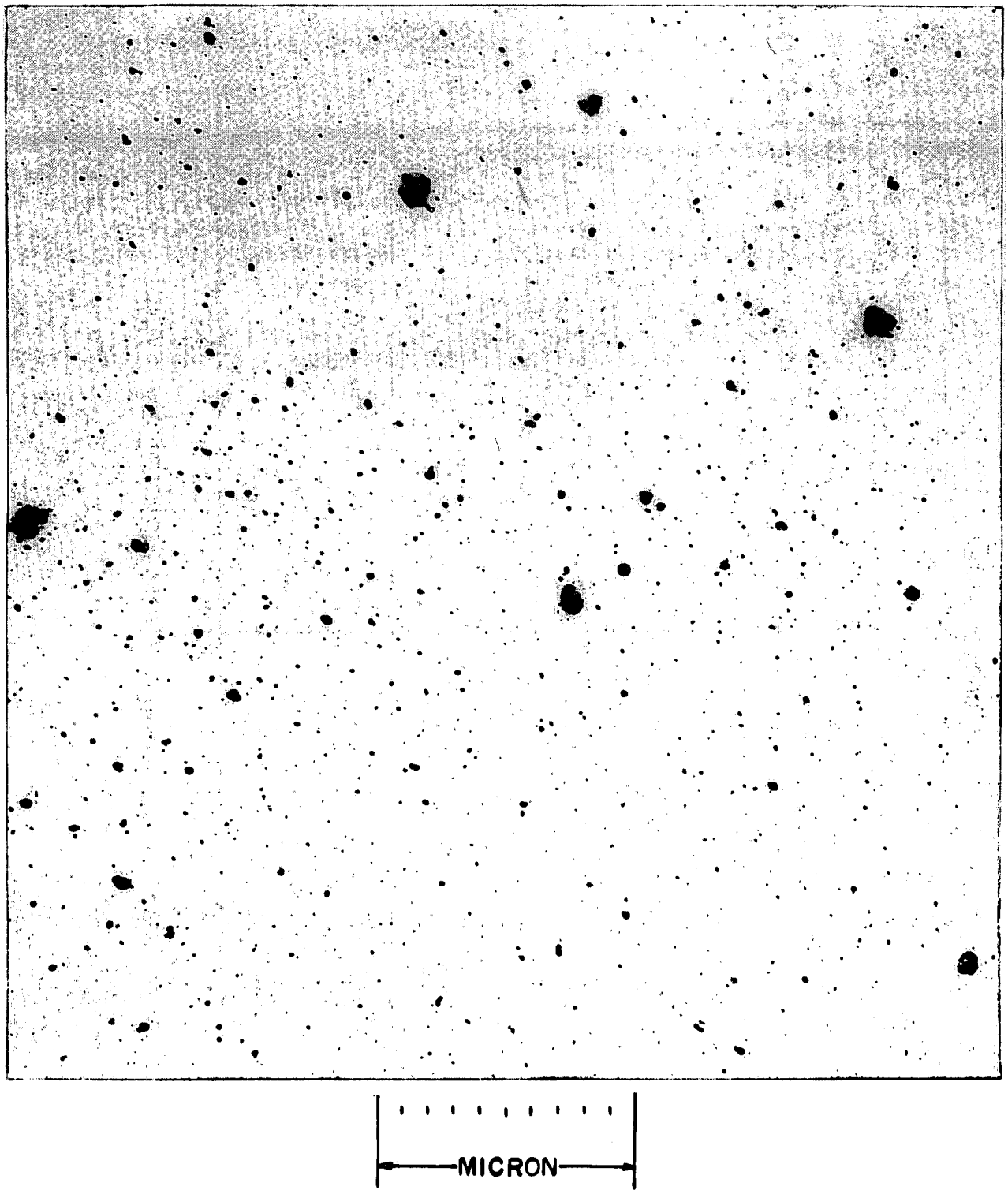


FIGURE 15

IDEALIZED VARIATION OF RELATIVE ACTIVITY OF THIN SECTIONS OF FILTER AS A FUNCTION OF DISTANCE THROUGH PAPER



Sample #1. Thick Formvar. 40,000x.

FIGURE 16 ELECTRON MICROGRAPH OF IMPACTED STRATOSPHERIC PARTICLES

that 95% of the mass of the material was contained in particles with diameters greater than 0.01 microns. If these spherical particles are representative of the particles which contain the radioactive material in the stratosphere, and if the radioactivity is distributed with the mass of the particles, an assumption given weight in recent Swedish studies[8], it is reasonable to believe that nearly 100 percent of the radioactiv ity is filtered by the HASP samplers. The experiments on particles are more fully discussed in Chapter IV of this report.

The Tungsten Experiment

A third approach to the problem of determination of retention efficiency is contained in the Tungsten experiment described in more detail in Chapters VIII and IX. Briefly, it is found that the stratospheric Tungsten inventory shortly after injection is in good agreement with the amount that was calculated to have been injected. The inventory is made on the assumption that the filter is 100% efficient; consequently the results tend to show the assumption is warranted. Certainly an efficiency as low as 75% would be reflected in the Tungsten inventory.

It is concluded that, while the quantitative determination of the efficiency of HASP filters is still incomplete, the weight of evidence from the several independent approaches to the problem indicates that the efficiency is nearly 100 percent for stratospheric particulate matter. Additional evidence is discussed in Chapter IV.

CHAPTER IV

STRATOSPHERIC PARTICULATE MATTER

Introduction

At the beginning of the High Altitude Sampling Program it was believed that much could be learned of the physical characteristics of stratospheric particulate matter through the study of particles collected in HASP filter samples. Several approaches have been made to the study of these particles including autoradiography, microtomy, optical microscopy and electron microscopy. It has become clear, however, that dust contamination of the filters is so enormous and detection of submicron particles within the fiber matrix is so difficult that the only techniques which hold promise of yielding useful information are those which use the radioactivity of the particles themselves to provide identification. Fortunately a method of particle collection has become available which avoids many of the difficulties introduced by the filter matrix. Two experiments were performed on collection of stratospheric particulates by impaction on electron microscope grids attached to HASP filters and exposed at flight altitudes during HASP missions. In September 1959 particle collection probes, provided by the Geophysics Research Directorate of the Air Force Cambridge Research Center were installed on two HASP aircraft.

Investigations of the distribution of radioactive particles through the fibrous mat of the filter paper and on or in individual fibers have been continued since the results obtained should shed some light on

filter efficiency. Electron microscope studies have been completed on impaction grids exposed on HASP filters. These grids are being autoradiographed in an attempt to measure the radioactivity of the various particle types identified on the grids.

Filter Paper Studies

To study the distribution of radioactive particles through HASP filters, areas of certain papers have been selected and microtomed to provide a set of serial sections through the paper. Two such sets, cut parallel to the face of a filter, are shown, together with autoradiographs of the sets, in Figures 17 and 18. These sets were cut from the areas of two papers which had contained "hot spots" detected by the original autoradiographs of the papers. Sample 299 was collected in the Northern Polar Stratosphere on 18 March 1958 and contained debris from a Soviet test conducted in late February 1958. Sample 535 was collected in the Tropical Stratosphere on 10 June 1958 and contained some debris, including a high tungsten-185 concentration, from 2 United States tests conducted in May 1958. The sections displayed in serial order, reading the first row from left to right, the second row from right to left, the third from left to right, etc. The sections containing fibers from the front and back faces of the filter paper are marked in the photographs. The corresponding autoradiographs indicate that the radioactive particles which caused the original "hot spot" were located well within the body of the fiber mat and few if any radioactive particles penetrated to the back face of the filter paper.

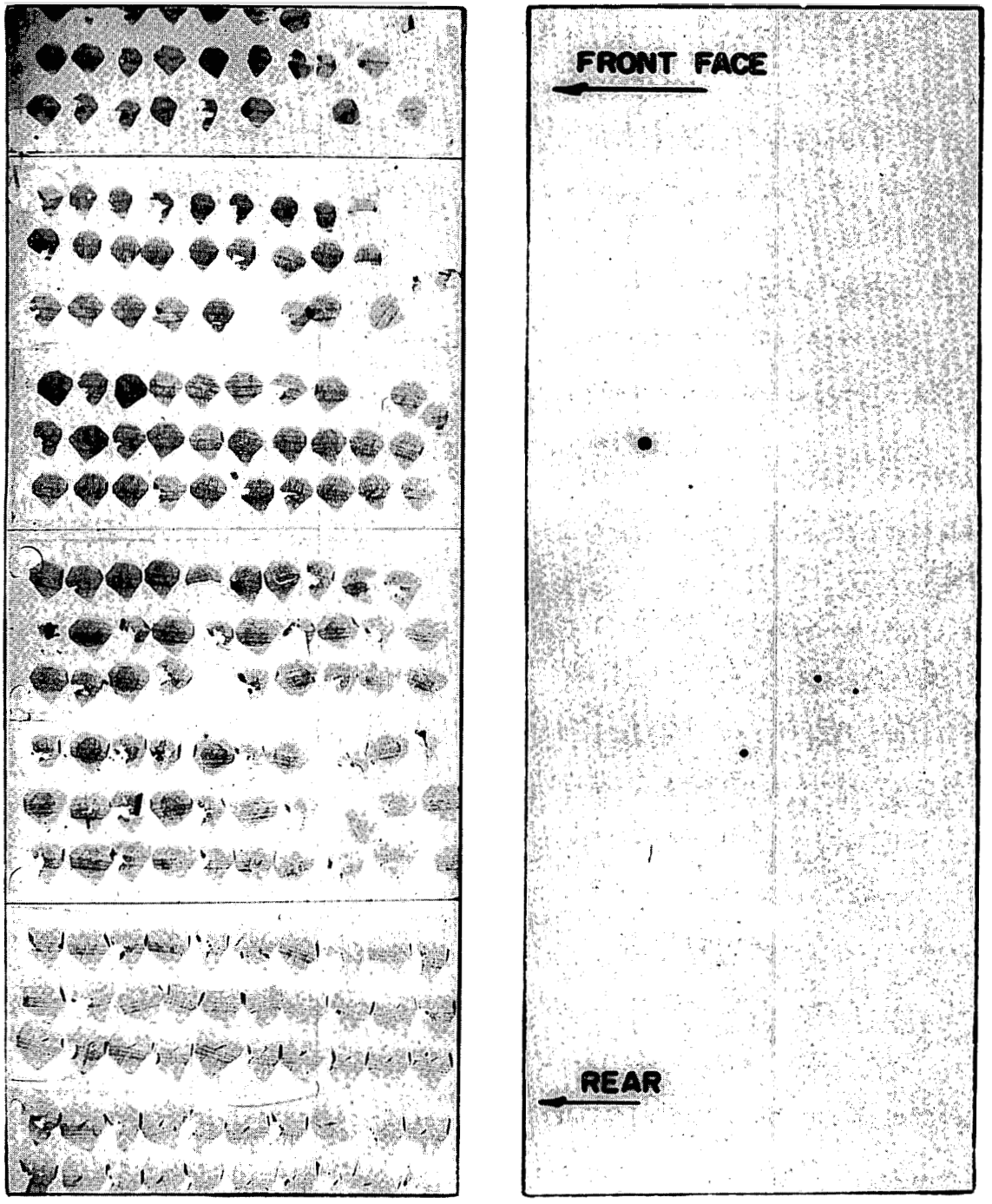


FIGURE 17 PARALLEL MICROTOME SECTIONS 10 MICRONS THICK OF METHACRYLATE EMBEDDED DISK OF SAMPLE No. 299 AND CORRESPONDING X-RAY FILM AUTORADIOGRAM (1 Month)

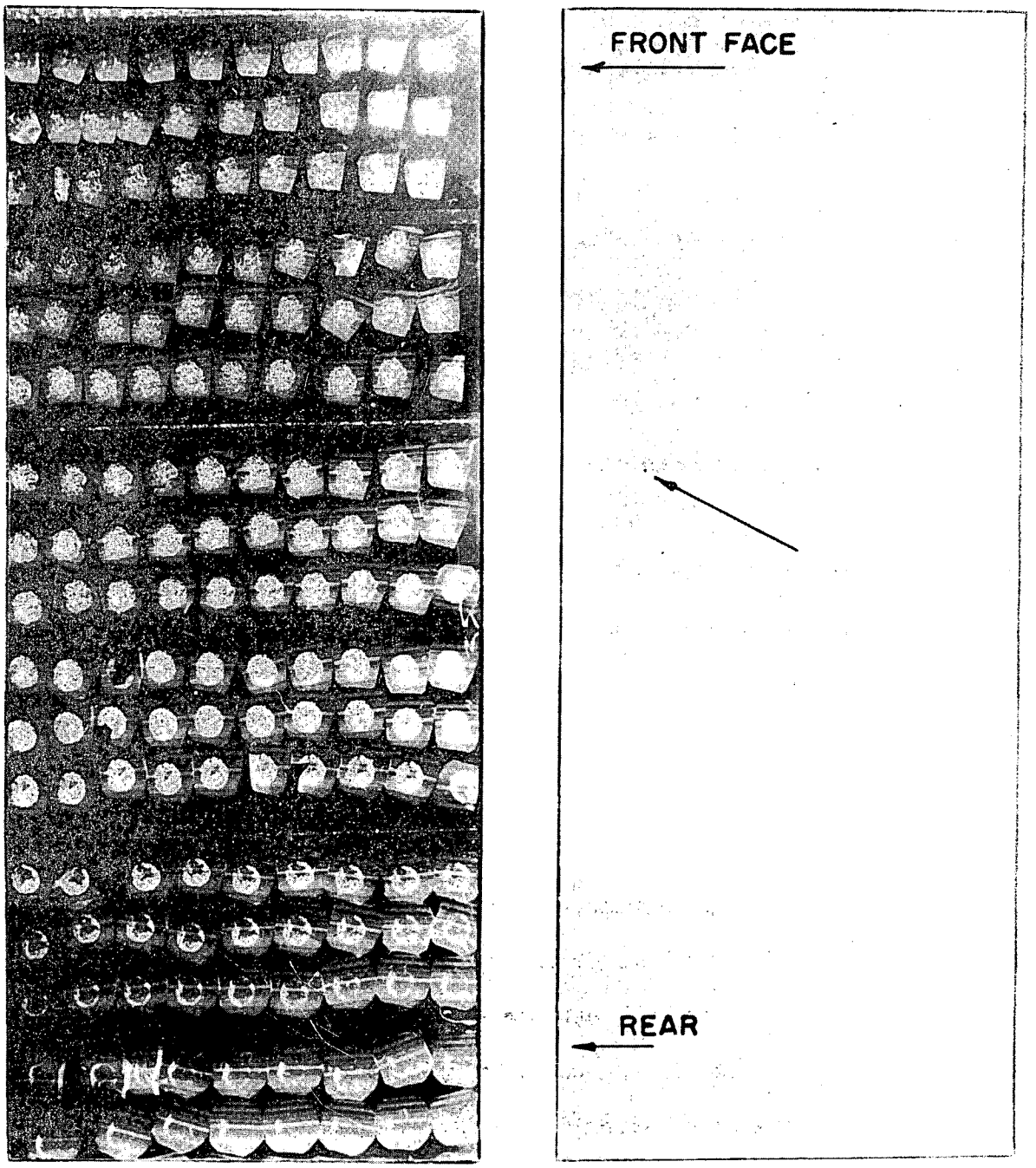


FIGURE 18 PARALLEL MICROTOME SECTIONS 10 MICRONS THICK OF METHACRYLATE EMBEDDED DISK OF SAMPLE No. 535 AND CORRESPONDING X-RAY FILM AUTORADIOGRAM (1 Month)

Figure 19 shows a microtomed section cut perpendicular to an imbedded piece of filter containing a "hot spot" and the corresponding autoradiograph. Sample 732 was collected in the Northern Polar Stratosphere on 22 October 1959 and contains debris from the Fall 1958 Soviet tests. The photograph of the section shows the ends of the fibers as black dots. The large dot located at the back of the filter paper is a fiber of the gauze backing. In this case the autoradiograph shows a noteworthy concentration of radioactivity at the front of that portion of the filter paper wherein the fibers are most closely spaced. Again, this indicates that few of the total number of radioactive particles find their way to the back of the filter paper.

It is interesting to note here that the exposed filter paper shows a non-uniform spacing of fibers in vertical cross section. The spacing toward the rear of the filter paper was imbedded and microtomed vertically and did not show this non-uniform spacing of fibers. It is suspected, since the evidence is only based on this one set of observations, that the filter paper becomes compacted during exposure thus giving the non-uniform spacing of fibers observed.

Two microtomed sections, cut perpendicular to a filter paper (sample 732), were covered with liquid emulsion and autoradiographed. The sections and superimposed autoradiographs are shown in Figure 20. In these sections, as a result of the high resolution obtained by use of liquid emulsion, localized spots of high optical density can be found in the emulsion amongst the fibers of the filter. Figure 21 shows a

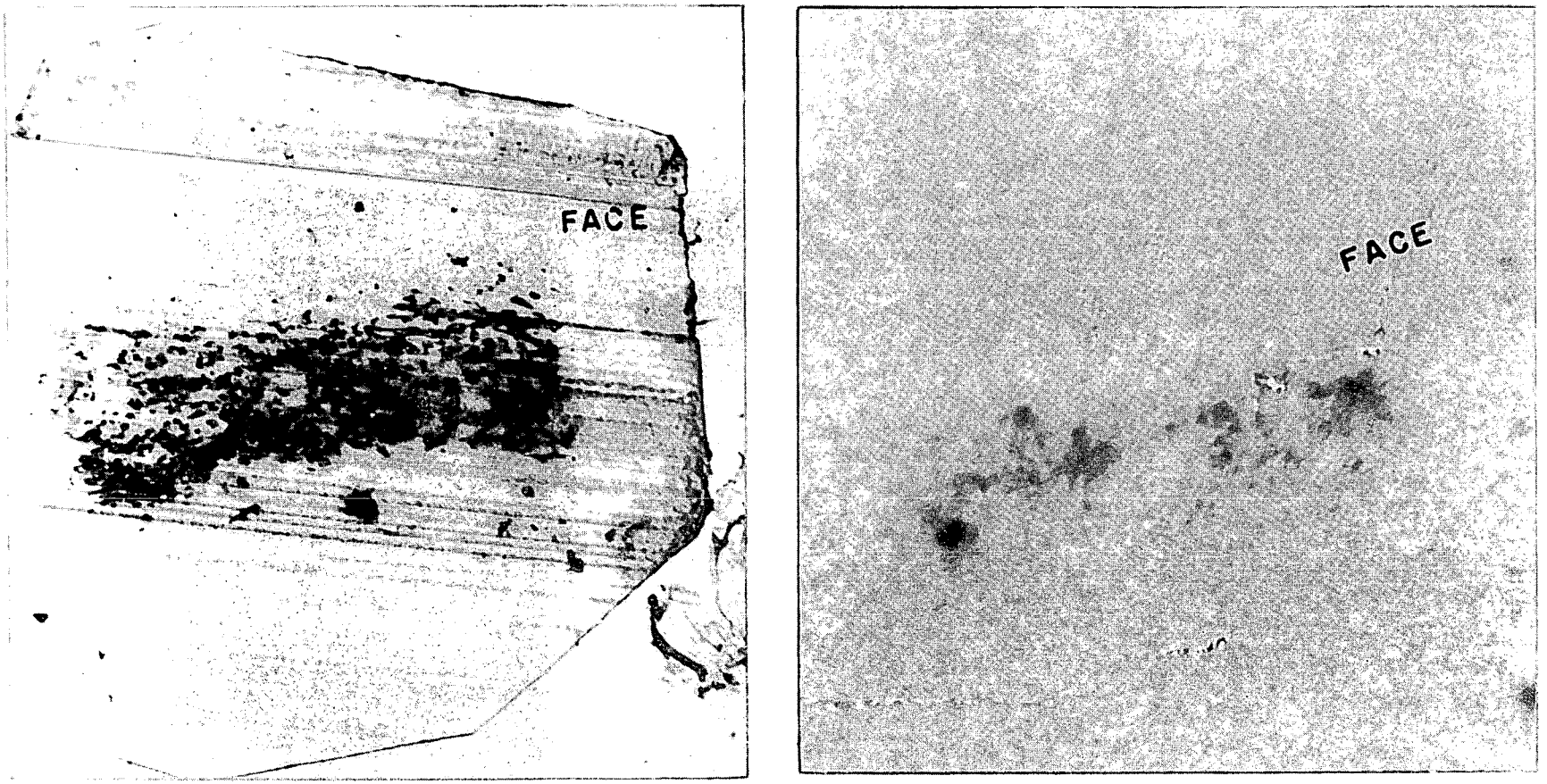


FIGURE 19 VERTICAL MICROTOME SECTION 5 MICRONS THICK OF METHACRYLATE EMBEDDED STRIP OF SAMPLE No. 732 AND CORRESPONDING X-RAY FILM AUTORADIOGRAM (1 Month)

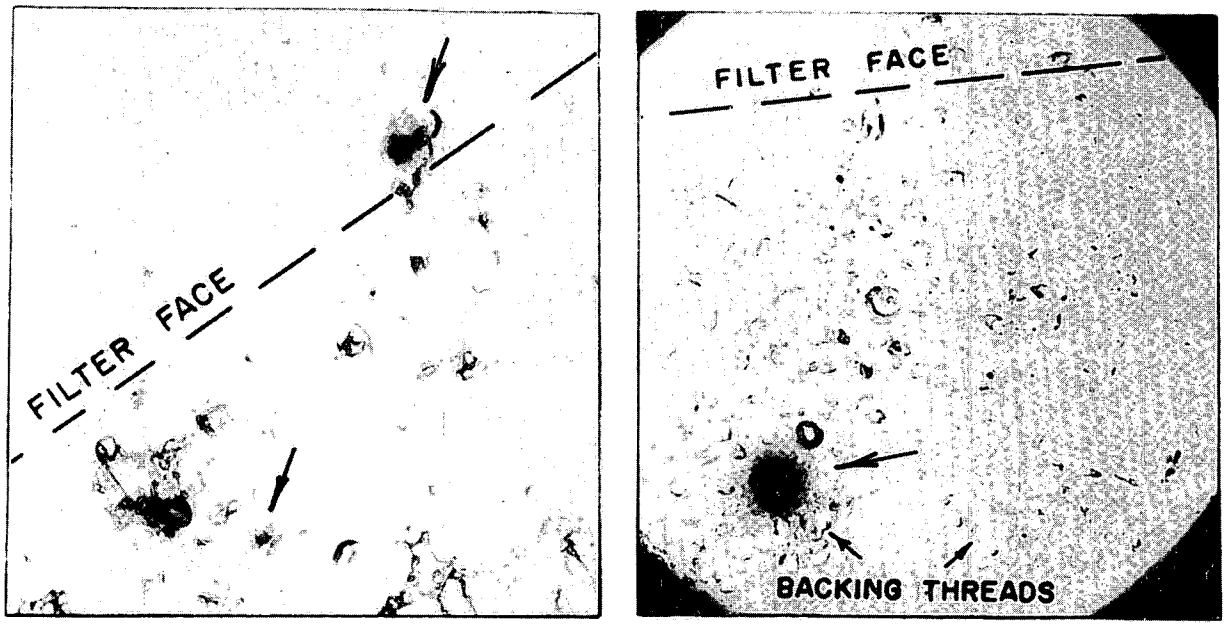


FIGURE 20 AUTORADIOGRAM IN LIQUID EMULSION COATING ON 5 MICRON THICK VERTICAL MICROTOME SECTIONS OF SAMPLE No. 732

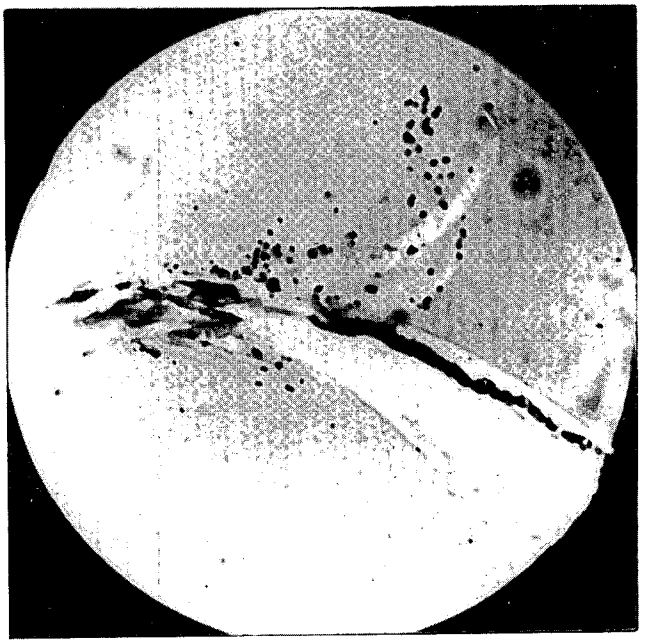


FIGURE 21 AUTORADIOGRAM IN LIQUID EMULSION OF A PORTION OF A SINGLE FIBER CONTAINED IN A MICROTOME SECTION OF SAMPLE No. 732

portion of a single fiber in a perpendicular microtomed section with liquid emulsion autoradiograph. In this case the radioactivity is not concentrated in single "hot spots," but appears to be generally distributed along the length of the fiber.

In an effort to observe the particles containing the radioactivity, some individual fibers were extracted from a region of a filter paper which contained a "hot spot" as indicated by x-ray plate autoradiography. The fibers were coated with liquid emulsion and exposed for about 30 days. The developed autoradiographic images are shown in Figures 22 and 23 on the left hand sides. These photographs also show portions of the fibers. The silver grains of the images were dissolved away with a solution of potassium ferricyanide and sodium thiosulfate, described later in this chapter, and the reaction was observed with the light microscope. In both cases, after the silver had been dissolved, a particle was discerned in the locations which had been the center of the silver images. Figure 22 (right hand side) shows a 5 micron diameter particle and Figure 23 shows a 1.5 micron diameter particle, which remained after removal of the silver. Under microscopic examination, the particles appear to be crystalline and possible agglomerates of particles smaller than 0.5 micron in diameter.

Collection of Stratospheric Particles by Impaction

As described in Chapter II several coated electron microscope grids of 1/8 inch diameter were attached to the upstream face of a regular HASP filter installed in the U-2 nose sampler. The mission was flown northward

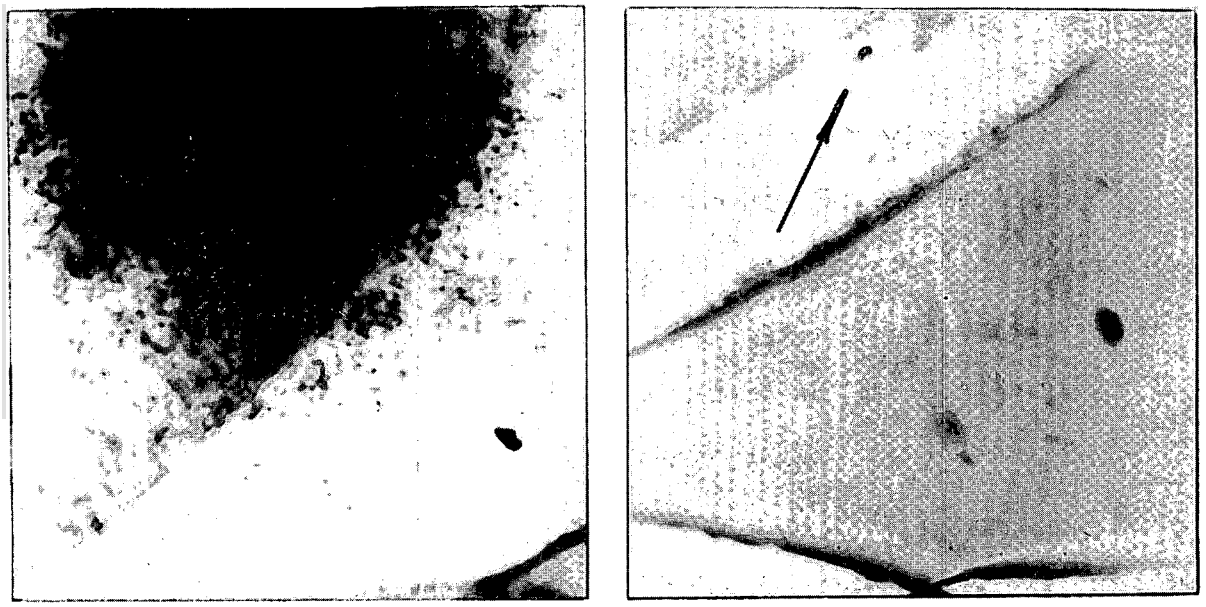


FIGURE 22 AUTORADIOGRAM IN LIQUID EMULSION COATING ON SINGLE FIBERS REMOVED FROM SAMPLE No. 732, AND 5 MICRON PARTICLE REMAINING AFTER DISSOLVING THE SILVER IMAGE

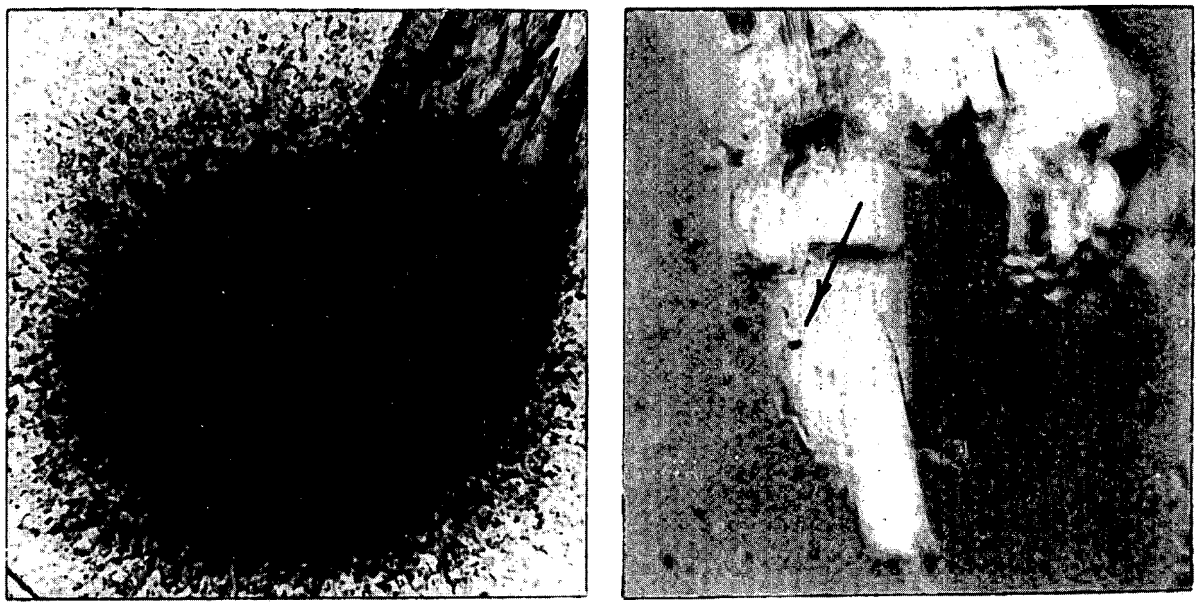


FIGURE 23 AUTORADIOGRAM IN LIQUID EMULSION COATING ON SINGLE FIBER REMOVED FROM SAMPLE No. 732, AND 1.5 MICRON PARTICLE REMAINING AFTER DISSOLVING THE SILVER IMAGE

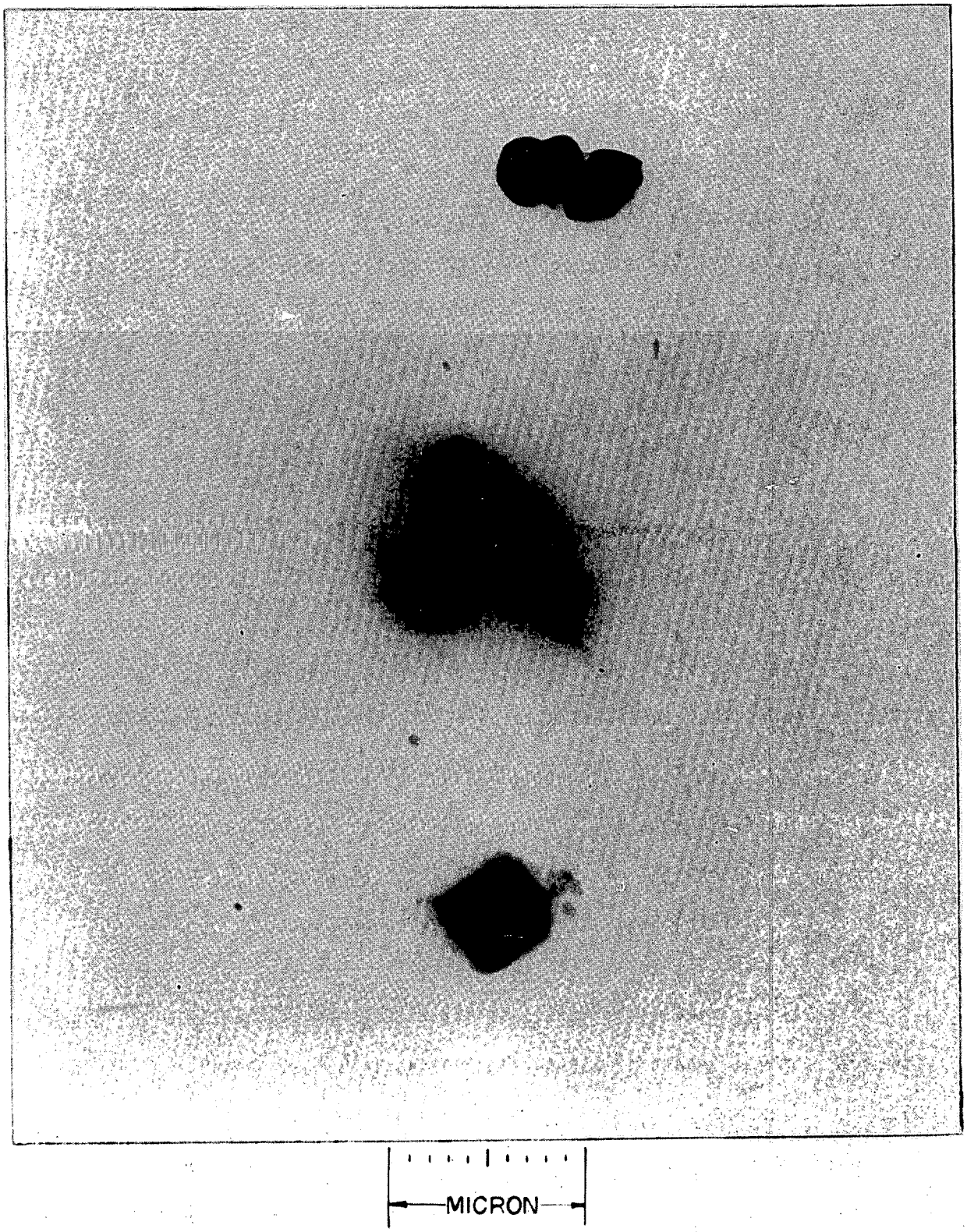
at maximum altitude from Plattsburg Air Force Base. The grids were coated with thin films of formvar and nitrocellulose. The samples were prepared in the laboratory of Ernest F. Fullam, Inc. in a "dust free" atmosphere. After the samples were recovered from the exposed filter paper, they were returned to Fullam's laboratory for electron microscopic examination.

Figures 24, 25, and 26 show some typical results of these electron microscopic studies. All of the particles shown have a very high electron optical density. This leads to the belief that these particles were collected from the stratosphere, since blank samples and samples of ground level contaimination show only particles which have low electron optical density. The fact that the particles shown in Figure 25 and 26 are spherical, or nearly spherical, also tends to confirm that the particles do not represent contamination, since most contaminating particles have irregular, crystalline structures.

The particles in Figure 24 appear to be cubic in structure. There can be little doubt as to their crystallinity in view of the regular outlines and straight edges. The individual particles are about 0.5 micron in their large dimensions. The agglomerate is about 1.5 microns.

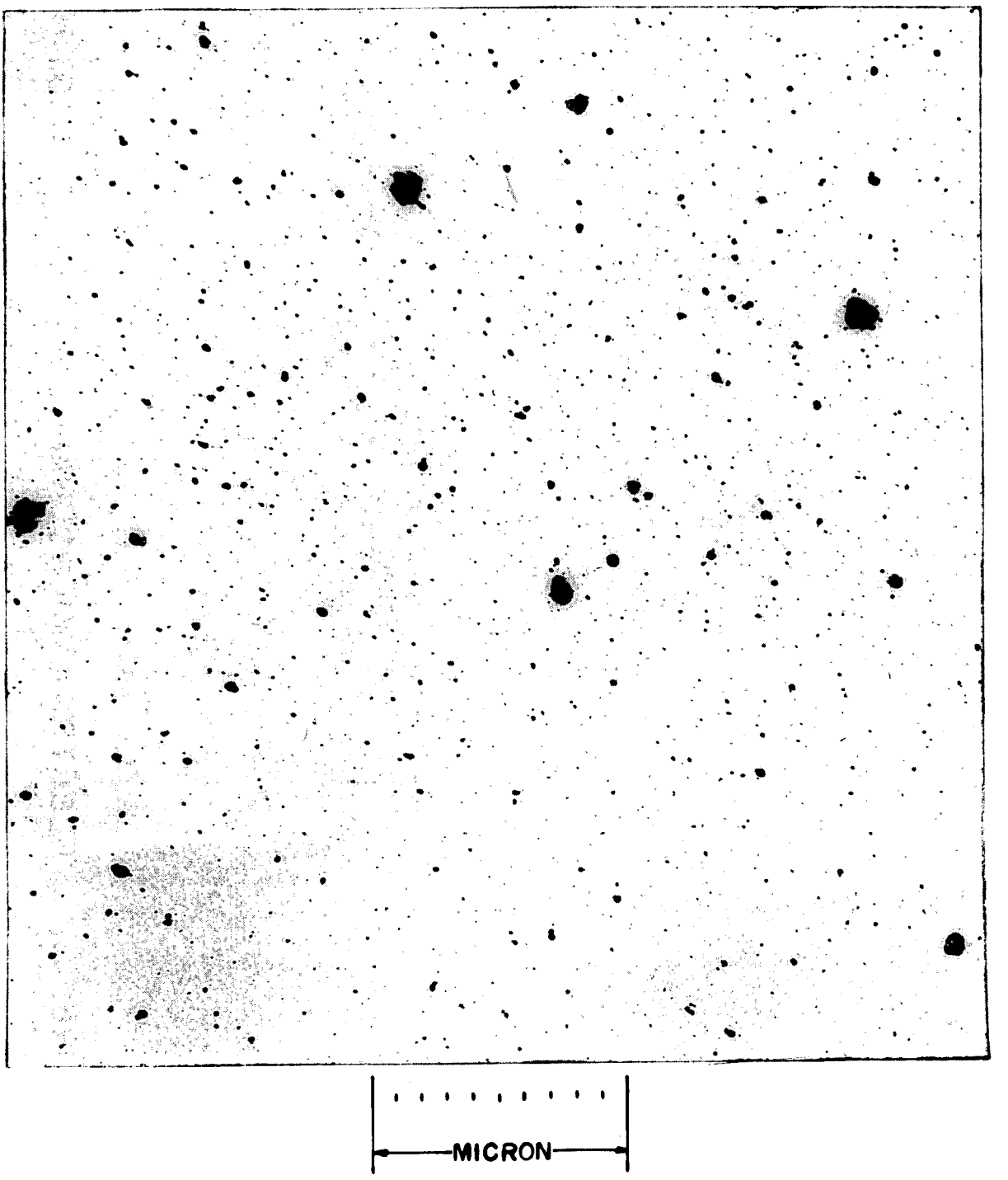
The spherical particles shown in Figures 25 and 26 range in size from about 0.01 micron to about 0.05 micron, with agglomerates about 0.1 micron to 0.4 micron.

A size-frequency plot was made of particles in the sample illustrated in Figure 24. All of the particles were of the same general nature as the particles shown therein. A count of 239 particles ranging in diameter



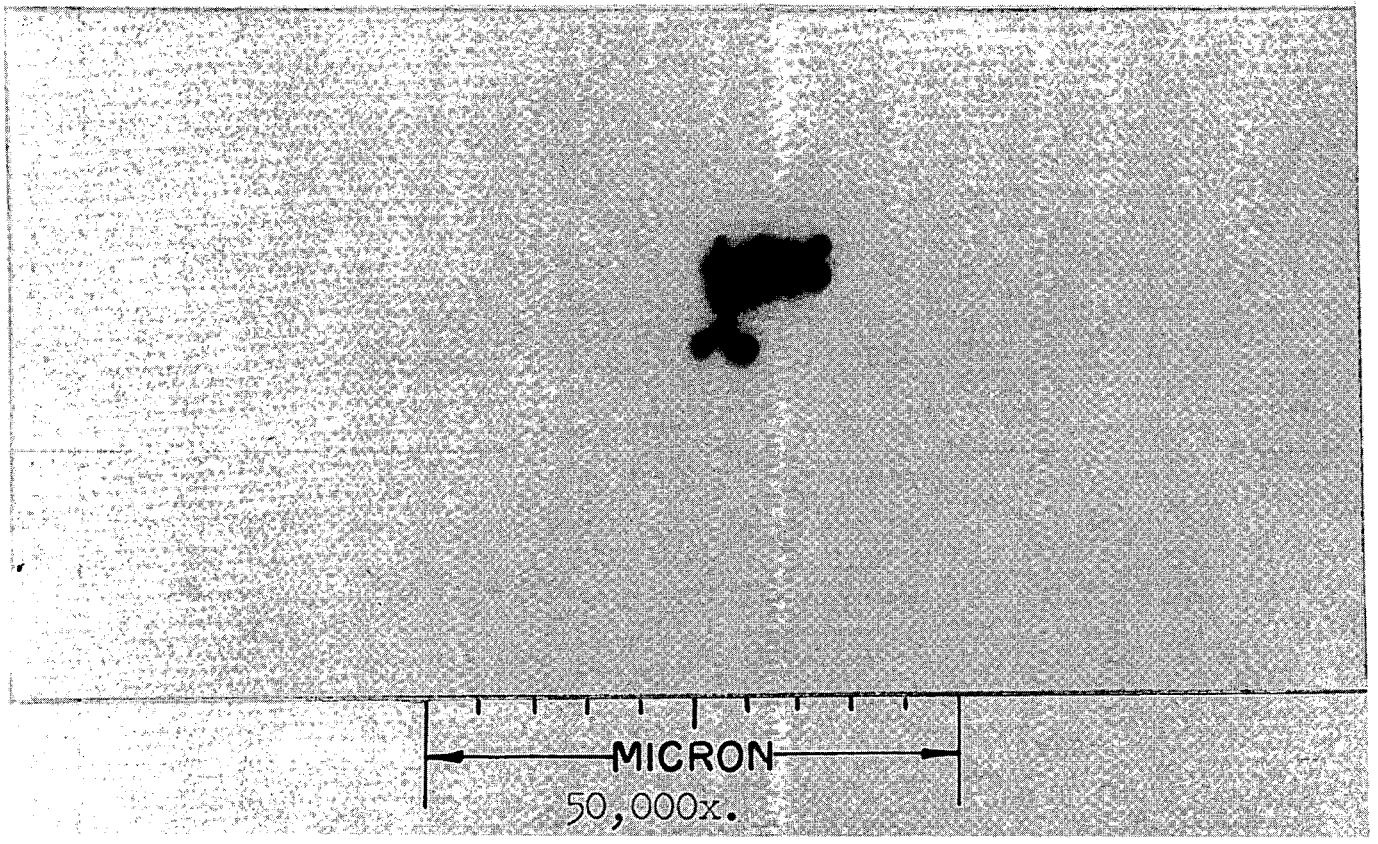
Sample #6. Nitrocellulose plus Carbon. 30,000x.

FIGURE 24 ELECTRON MICROGRAPH OF IMPACTED STRATOSPHERIC PARTICLES



Sample #1. Thick Formvar. 40,000x.

FIGURE 25 ELECTRON MICROGRAPH OF IMPACTED STRATOSPHERIC PARTICLES



Sample #4. Thin Formvar.

FIGURE 26 ELECTRON MICROGRAPH OF IMPACTED STRATOSPHERIC PARTICLES

from 0.05 micron to 1.2 microns gave a size-frequency distribution which was log-normal with a geometric mean diameter of 0.26 micron and a geometric standard deviation of 2.0.

An attempt was made to determine a size-frequency curve for the particles shown in Figure 25. This was difficult to do in an accurate manner since 60 percent of the particles shown have diameters less than 0.01 micron, which was the smallest particle size which can be measured in the photograph. However, in order to determine the order of magnitude of the distribution parameters, three measured points on a curve of diameter versus percent of particles having diameters less than the stated diameter were plotted on log probability paper and a straight line was drawn which was most nearly represented by the three points. The resulting log-normal distribution had a geometric mean diameter of 0.009 micron and a geometric standard deviation of about 2. It should be emphasized that the preceding parameters are based on a very crude fit of the data to a log-normal distribution, and were only found by this method in order to give an order of magnitude estimate to the true parameters of the size-frequency distribution.

Attempts have been made to autoradiograph several of the electron microscope grids with x-ray film. However, the results have been inconclusive so far.

Discussion

The study both of microtomed sections cut parallel and sections cut perpendicular to the faces of HASP filter papers thus far supports the

arguments regarding the nearly 100 percent efficiency of sampling. This conclusion stems from the fact that no activity has been observed toward the rear face of the filter in parallel sections and, indeed, the activity per section appears to reach a maximum at a location well within the bulk of the fibrous mat and thence taper off toward the rear face. From the perpendicular sections of the filter paper it appears that the maximum activity occurs in the portion where the fibers become compacted.

The work so far accomplished in the study of microtomed sections of the HASP filter papers and on individual fibers extracted from the filter papers points strongly to the conclusion that there are basically two types of collected particulate material. One type gives rise to the highly localized "hot spots" while the other type is responsible for the diffuse distribution of radioactivity in the filter paper fibers. The fact that two particles greater than 1 micron in diameter have been observed in the same location as two "hot spots" on individual fibers, and that no particles have been observed (in the light microscope) which could be said to have caused the diffuse radioactivity leads to the belief that the "hot spots" may have been caused by particles larger than 0.5 microns in diameter or by agglomerates of smaller particles, and that the diffuse radioactivity patterns may have been caused by individual, sub-microscopic particles smaller than 0.5 microns in diameter.

The results of the electron microscopy study of the one set of impaction samples reveal, also, two different types of particles. The large crystalline particles of 0.1 to 1 micron in diameter seem to occur

often in agglomerates of size greater than 1 micron. The small spherical particles, as found in the samples are, for the most part, discreet single particles, although agglomerates of the order of 0.1 micron to 0.5 micron were observed. Although there is no direct evidence, and the indirect evidence is admittedly meager, there exists the possibility that the larger crystalline particles observed by electron microscopy are the particles which cause the "hot spots" found on the HASP filter papers and the smaller spherical particles are primarily responsible for the diffuse radioactivity in the filter paper.

Besides the high optical density particles shown in Figures 24 through 26, other particles of low optical density were found in the electron micrographs.

These particles were volatile under the electron beam giving rise to images which appeared as oily, rounded blobs on the electron micrographs. These particles are similar to those found by C. E. Junge at Air Force Cambridge Research Center[9], in samples collected during balloon flights. It is thought that these low density particles are probably of tropospheric origin and were brought into the stratosphere and subsequently mixed upward from the tropopause either by eddy diffusion or by Brownian motion or by both[10].

The sphericity of some of the particles observed by electron microscopy suggests a "thermal origin" such as a nuclear detonation. These particles have also been found in samples from balloon flights by Air Force Cambridge. The high electron optical density (with the inferred

relatively high molecular weight) of the larger crystalline particles does not necessarily preclude their existence in normal tropospheric air. However, it seems more likely that particles of this type were thrown into the atmosphere by nuclear detonations which carried large amounts of terrestrial material into the lower stratosphere.

The few results obtained from the one set of observations made with the electron microscope certainly leave the picture of stratospheric particulate material incomplete. The problems regarding the relationships of particle size to radioisotope content have not been solved, but a few possible answers to some of the problems have been suggested.

Future Work

A program for routine collection of particles on impaction surfaces has been initiated and is currently in operation. A sampling probe, which contains coated electron microscope grids and/or small glass slides, has been installed on two HASP U-2 aircraft. The probe is loaded and unloaded in the laboratory and is designed for minimal contamination during times when it is not exposed to stratospheric air. The exposure is made during the normal HASP missions.

To date no samples have been completely analysed. It is estimated that this program will yield about 20 to 30 exposures of probe samples through June 1960 and then 4 per month indefinitely. The samples will be examined by electron microscopy and autoradiography. The particles observed will be classified as to size, shape, crystallinity and optical density. Size-frequency distributions will be obtained when possible.

Laboratory Technique

In the impaction experiment carried out on 21 July 1959, dust proof electron microscope substrate sample holders, in the form of 1" flanges were sewn to the face of a HASP filter. Each flange contained a central hole 5/16" in diameter surrounded by a 1/16" high stamped lip. Brass screen or copper mesh electron microscope grid material was inserted into the inner side of the lip. A dust cap 5/16" in diameter and 3/8" long fitted over the outer side of the lip. Two small holes were punched in the 0.005" thick brass flange to allow sewing to the filter. The steps taken to obtain inflight exposed electron microscope substrates are described in sequence.

1.) The prepared flanges were cleaned in acid and coated electron microscope grids were tacked to the brass screen or thin films were floated onto the copper mesh. Ten samples, plus controls, were coated. These included substrates of Formvar, nitrocellulose, carbon coated nitrocellulose, and Millipore filter.

2.) A gummed paper was added to the flat back side of the flange and the dust cap added to the lipped front side. Steps 1 and 2 were performed in a "dust free" laboratory.

3.) The samples were transported to Plattsburg Air Force Base, New York and were sewn to a filter (HASP Sample 1900) so that the dust caps protruded through the 1/2" wire mesh in front of the filter.

4.) Fifteen minutes before take-off the dust covers were removed, the filter was rotated immediately into the nose tunnel and the nose access door was bolted down.

5.) Ten minutes after the aircraft returned, the access door was opened and the dust cover was immediately replaced.

6.) The electron grids and portions of the filter surrounding them were removed and returned to the dust free laboratory for electron microscopy.

Farmers Reducer (1 percent potassium ferricyanide in 10 percent sodium thiosulphate) has been used to remove extremely dense autoradiographic images to allow viewing of any particles which could have caused the silver density. Successive drops are added while the progress of reduction is studied under the light microscope. When the reduction is completed the remaining particles are studied by light microscopy.

The chemistry of the reduction involves oxidation of silver by the ferricyanide to form silver ferrocyanide, which is dissolved by the sodium thiosulphate solution.

Since Fe $(CN)_6^{-4}$ reacts with ferric iron to give a dark blue color (Prussian blue), the reaction serves also as a test for iron. Further work is being carried out with this technique.

CHAPTER V

RADIOCHEMICAL SEPARATION AND RADIOMETRIC ASSAY

Introduction

The success of the HASP program is largely dependent upon the accuracy and reliability of the radiochemical analyses. This analytical requirement is an imposing one since thousands of samples must be processed for three or more radionuclides at intermediate and low levels of activity. To satisfy this requirement a sequential method of analysis was developed by modification of existing procedures which gave an over-all reproducibility to within a few percent. Quality performance is maintained by systematic analyses of standards, duplicates and blanks. Absolute calibration is based upon standards from the National Bureau of Standards and other qualified laboratories, primary calibrations by 4π counting and other techniques, and intercalibrations with laboratories that have the most experience in the analysis of radionuclides at similar levels.

When the filter paper arrives at the laboratory, the collection data are recorded, a code number is assigned and a 1 inch diameter disk removed for total beta assay. Then either $\frac{1}{4}$ or $\frac{1}{2}$ of the paper is ashed under conditions which yield quantitative retention even of volatile nuclides such as Ru^{106} and Cs^{137}. The ash is dissolved in a mixture of perchloric and hydrofluoric acids, the hydrofluoric acid is then distilled off, and the solution diluted to volume for the sequential analysis.

Radiochemical Separation

Figure 27 shows the general outline of the sequential analysis. After the addition of cesium, barium, strontium, zirconium, and cesium carriers, the cesium is separated by precipitating the hydroxy-carbonates of the other elements. The alkaline earths are separated by precipitating zirconium and ceric hydroxides. Zirconium is isolated as the phenylarsonate leaving the cerium in solution. Barium is separated as the chromate yielding strontium in solution for subsequent Sr^{89} and Sr^{90} determinations. Each fraction obtained from this sequential scheme is further purified to remove all traces of radioactive contamination prior to radiometric assay.

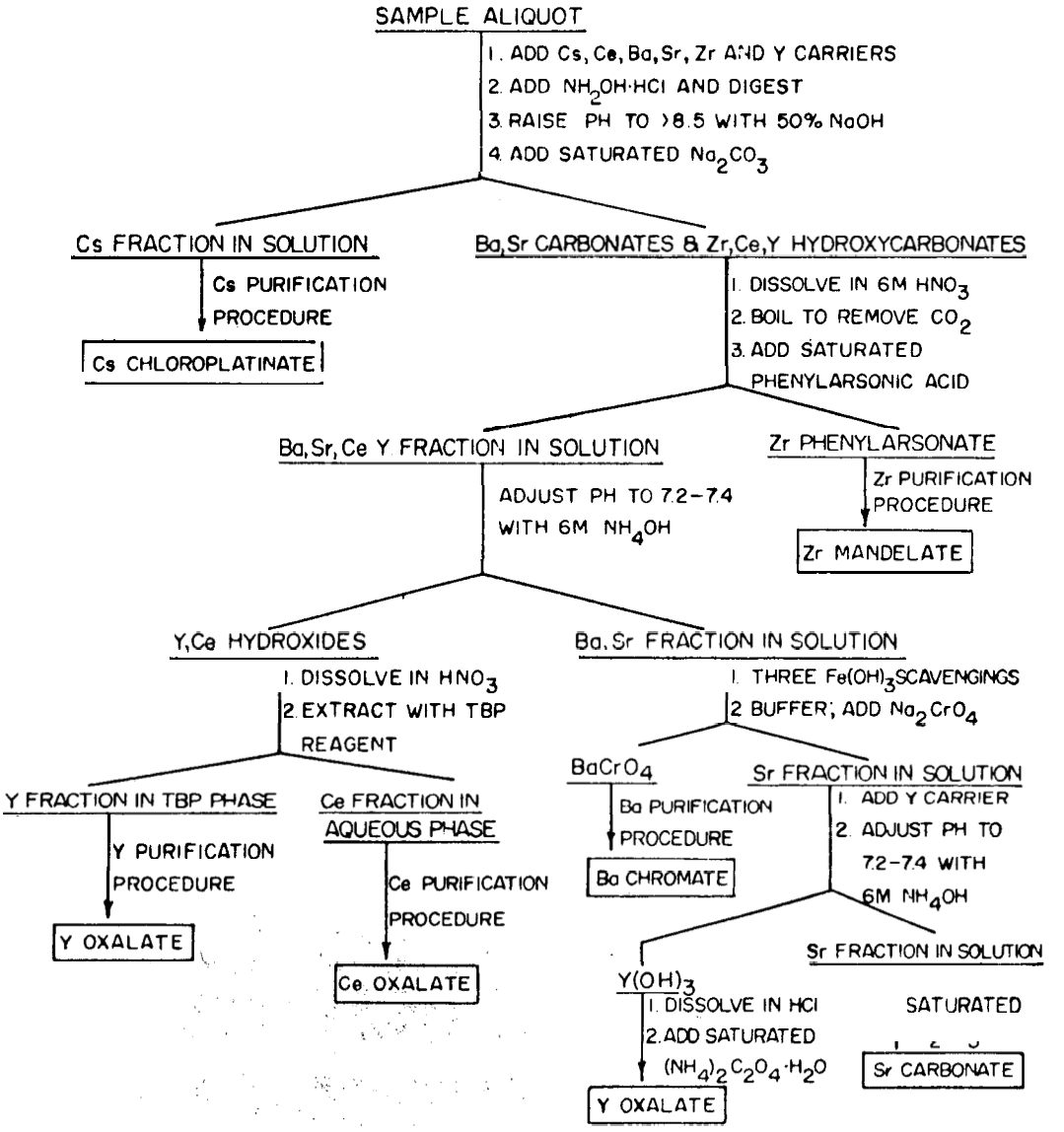
Plutonium, tungsten and rhodium activities are currently determined on separate aliquots of the original sample solution. However, with some modification of the group separations procedure, these latter two analyses could also be incorporated into the sequential scheme. Procedures have been developed for the analysis of Be^{7}, Y^{91}, Mo^{99}, and I^{131}.

Radiometric Assay

In general three methods of radiometric assay are used in the analyses of the HASP samples, namely, alpha and beta counting and gamma ray spectroscopy. Plutonium, for example, is determined by alpha scintillation counting. For moderate levels of beta activity, i.e., greater than 15 cpm which have been commonly encountered in the HASP

FIGURE 27

OUTLINE OF SEQUENTIAL ANALYSIS



samples, an end window, halogen filled Geiger tube is employed.

An increasing number of samples have required low level counting techniques. Figure 28 shows a unit of 4 low-level beta counters which was designed and constructed especially for HASP. The basic unit consists of a central end-window Geiger tube surrounded by and in anti-coincidence with a ring of cosmic ray counting tubes. A unique and highly economical feature of this unit is that 4 independent Geiger tubes are placed within and utilize the one anti-coincidence ring.

Figure 29 illustrates a bank of 4 of these multiple counting units in operation with their associated electronics. There are actually 16 low-level beta counters in this array. With a 2 inch lead shield, the background is on the order of 2-3 cpm permitting identification at a level of 1 dpm per sample.

The gamma ray spectrometer employed in the HASP program is shown in Figure 30. The activity is detected by a 1-3/4" diameter by 2" high NaI well crystal surrounded by 4" of Pb shielding. The electrical pulses are then amplified and sorted by a Penco 100 channel pulse height analyzer. The gamma spectrum is recorded in the magnetic cores of the analyzer and then printed out by a Victor recorder.

The counting equipment is calibrated with standard solutions of the various nuclides obtained from NBS, Nuclear-Chicago, Health and Safety Laboratory of the NYOO, Los Alamos Scientific Laboratory and Brookhaven National Laboratory. These standards are cross checked whenever possible at Isotopes, Inc., by 4π counting and defined

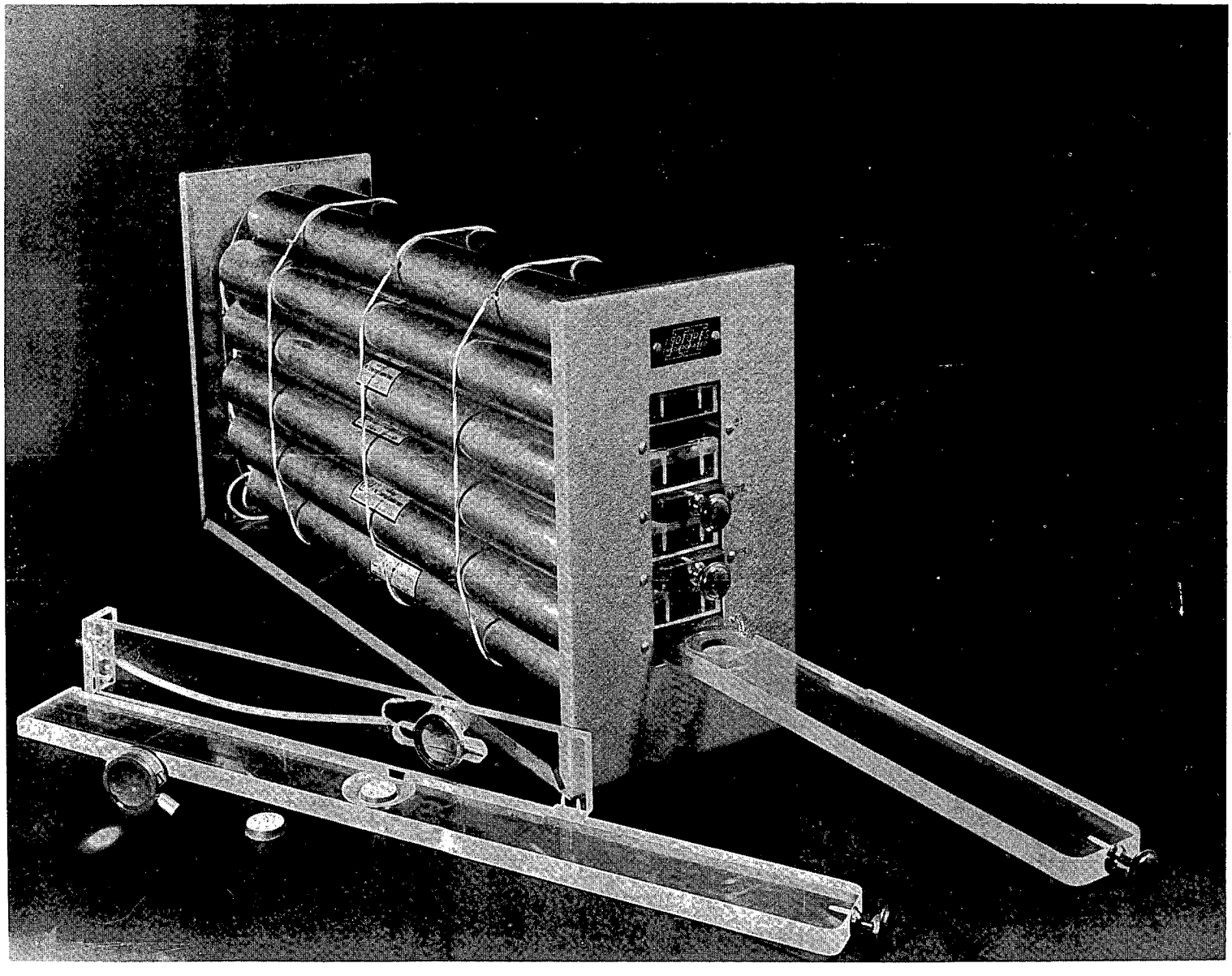


Figure 28 Low Level Beta Counter

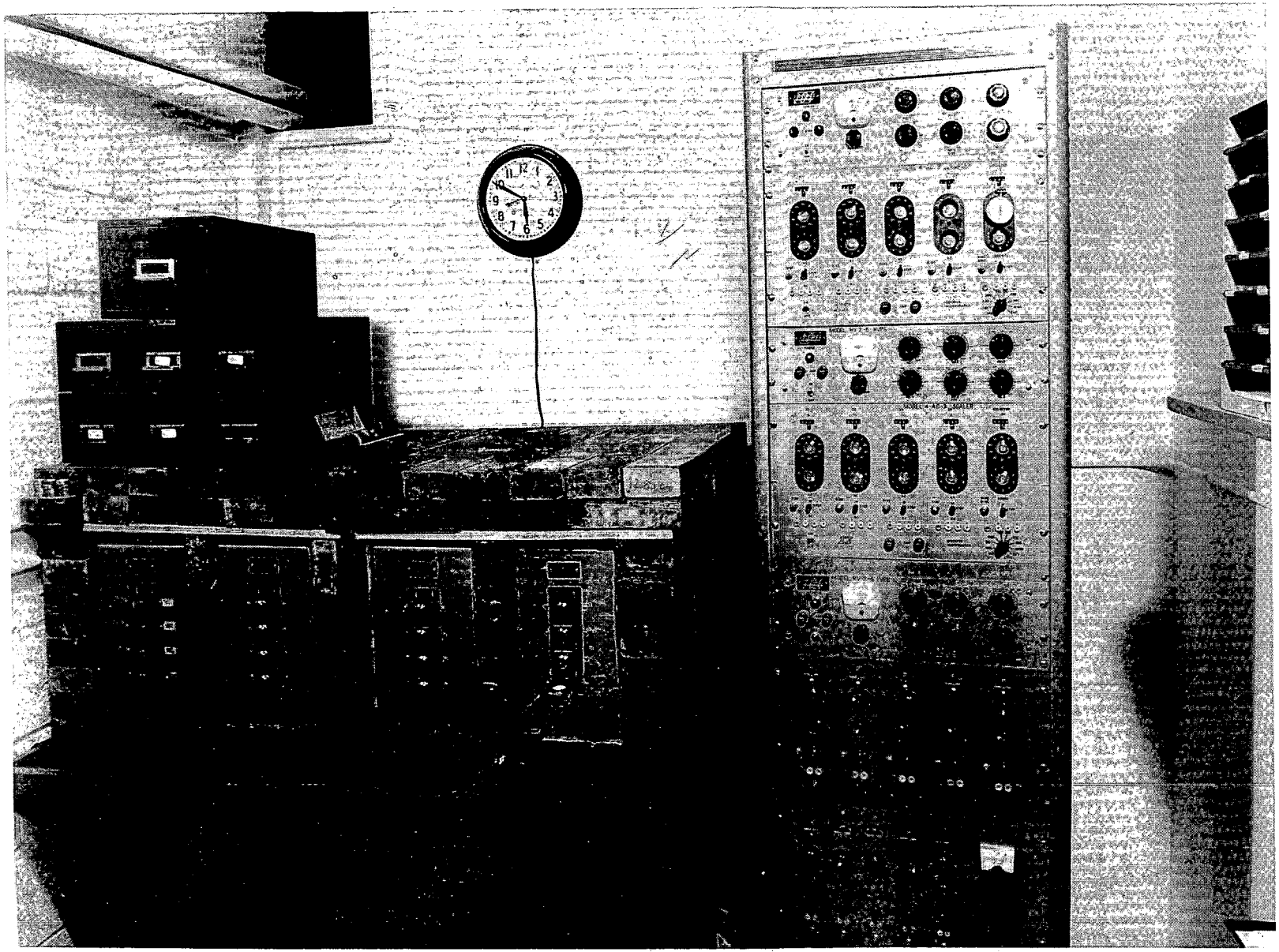


Figure 29 Three Units of Four Low Level Beta Counters

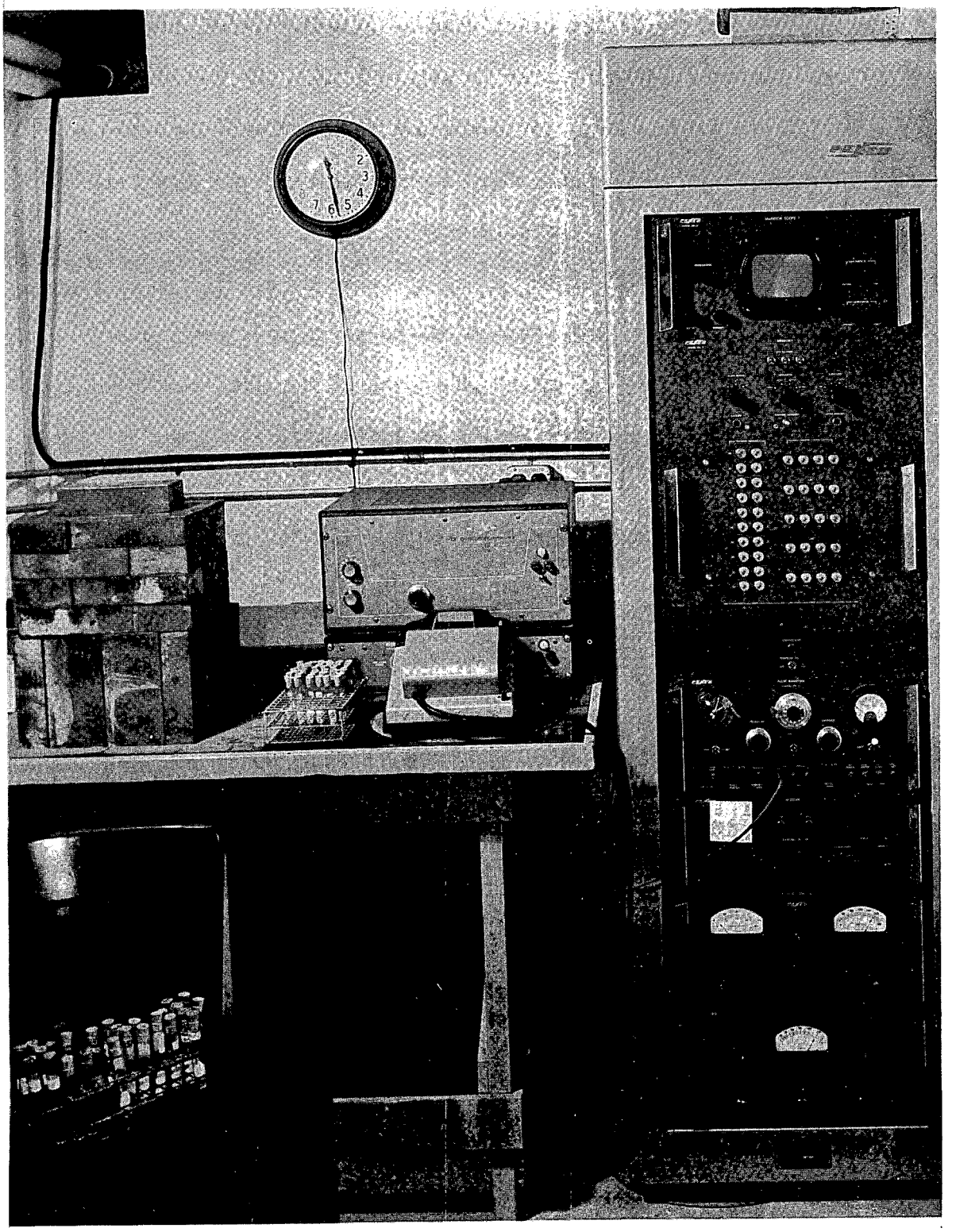


FIGURE 30

geometry measurements before being used as primary standards. Such solutions are generally certified to within ± 3%. The counters are checked daily with secondary standards and are recalibrated every 4 months with primary standards. The counter efficiencies have been remarkably stable; some of the original beta counters have retained their initial calibration of Y^{90} after 1½ years of continuous operation.

Figure 31 shows the calibration of the gamma ray spectrometer. The percent counting efficiency in the photopeak of the spectrum is plotted against gamma ray energy in Mev. The nuclides from which the specific gamma rays were derived are shown in the figure. The excellent fit of the smooth curve to the experimental points attests to the accuracy of the various standards which were used to obtain the data. The curve is presently being extended to lower energies for more complete coverage. The spectrometer can thus function as a primary standard for any nuclide which emits a gamma ray within the calibrated energy limits provided that the decay scheme of the nuclide is known.

Precision of Analyses

To document and maintain a satisfactory level of precision in the analytical methods, duplicates and blanks are processed each week by different analysis. A summary of the replication data is presented in Table 5.

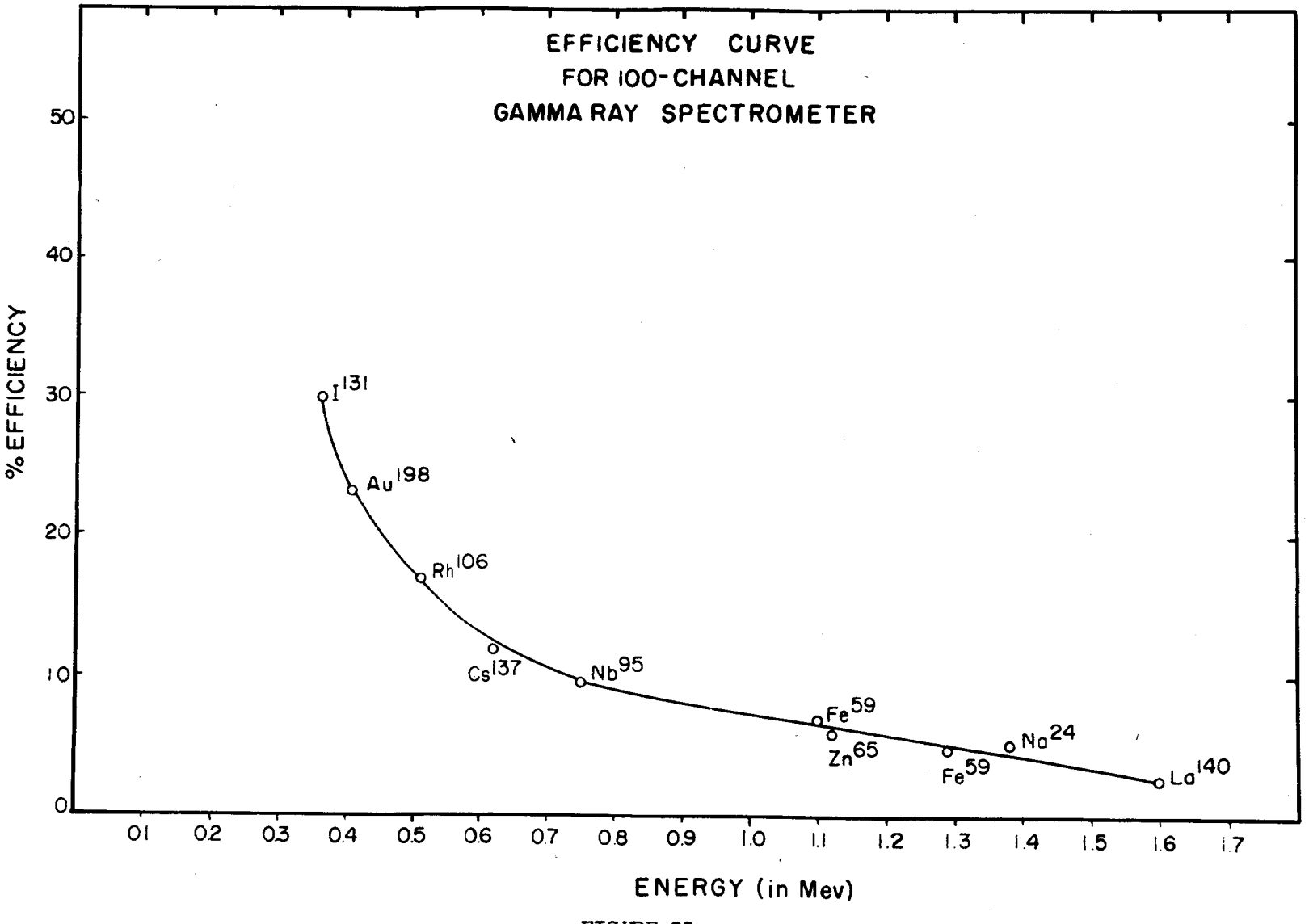

EFFICIENCY CURVE
FOR 100-CHANNEL
GAMMA RAY SPECTROMETER
% EFFICIENCY
0
10
20
30
40
50
I131
Au198
Rh106
Cs137
Nb95
Fe59
Zn65
Fe59
Na24
La140
0.1
0.2
0.3
0.4
0.5
0.6
0.7
0.8
0.9
1.0
1.1
1.2
1.3
1.4
1.5
1.6
1.7
ENERGY (in Mev)

FIGURE 31

Table 5

Reproducibility of Radiochemical Analysis

Nuclide	No. of Duplicate Analysis	Average % Standard Deviation
Sr^{90}	42	5.2
Sr^{89}	23	6.6
Zr^{95}	20	4.0
Cs^{137}	16	5.0
Ba^{140}	19	4.5
Ce^{144}	5	2.3
W^{185}	22	3.6
Pu^{239}	11	3.0

The average percent standard deviation is in the neighborhood of 5% or less. When nuclear testing was in progress and high stratospheric concentrations were encountered, most of the blank samples processed through the laboratory gave background counts and none ever exceeded 1 cpm over background. This level was generally less than 1% of the activities being measured. Now that the activity in the HASP samples has markedly decreased, all of the blank samples are below the limits of detection of the radiometric assay methods.

As a further indication of accuracy and precision, interlaboratory cross calibrations have been conducted with the Health and Safety Laboratory of the New York Operations Office of the AEC, Los Alamos Scientific Laboratory and the Air Force Radiochemical Laboratory with satisfactory agreement.

Summary

In summary, the analytical requirements of the HASP program have been successfully met. A radiochemical facility has been developed for the routine measurement of a large number of samples containing moderate and low levels of radioactivity with a high degree of precision. At the present time, the HASP analytical staff includes 15 radiochemists and 7 technicians with appropriate supervision who perform approximately 500 analyses each week with an accuracy and precision of about $\pm$ 5%. A summary of radiometric assay techniques presently being employed is presented in Figure 32.

FIGURE 32 Summary of Radiometric Assay Techniques

Nuclide	Method of Calibration	Counting Method	Purity Check	Counting Efficiency
Cs^{137}-Ba^{137}	Nuclear-Chicago Standard	End-window GM tube	Absorber Ratio	20%
Zr^{95}	Gamma ray spectrometry	Gamma ray spectrometry	Nb^{95} growth	Source on top of NaI crystal: 4.2% Source in well of NaI crystal: 9.5%
Y^{91}	Defined geometry	End-window GM tube	Absorber Ratio	20%
$Pr^{144}(Ce^{144})$	Harwell, England Standard	End-window GM tube with 200 mg/cm^{2}Al absorber	Absorber Ratio	10%
Ba^{140}	Defined geometry	Gamma ray spectrometry End-window GM tube	Spectrum La^{140} growth	3% 25%
$Sr^{90}(Y^{90})$	NBS standard	End-window GM tube	Y^{90} decay	25%
Sr^{89}	Defined geometry	End-window GM tube	Sr^{89} decay	30%
W^{185}	LASL standard	End-window GM tube	W^{185}	17%
Rh^{102}	LASL standard	End-window GM tube	Absorber Ratio	8%
Pu^{239}	BNL standard	ZnS scintillation counting	----------	42%
Be^{7}	Gamma ray spectrometry	Gamma ray spectrometry	Spectrum	2%
I^{131}	Nuclear-Chicago standard	End-window GM tube	I^{131} decay	19%

CHAPTER VI

ANALYSIS METHODS

The Calculation of Specific Activities of Stratospheric Air

The data needed in order to calculate the specific activities of stratospheric air from the radiochemical analyses of HASP filters include the altitude, latitude and velocity of the aircraft during the collection of the sample. This information is recorded by the pilot and is reported on flight data cards supplied with the samples. The flight cards list the Air Force Base from which the mission was flown, the aircraft number, the mission number, the flight date, the name of the pilot, the filter numbers and the sequence in which they were exposed, the altitude, time, latitude and longitude at which exposure of each filter was begun and at which it was terminated, the monitored activity of the filter, the forecast and measured air temperature, the forecast wind directions and velocities, the forecast tropopause heights, any report of turbulence by the pilot and the indicated and true aid speeds of the aircraft. In addition a 16 mm film which provides a photographic record of an auxillary instrument panel is supplied for each aircraft for each mission. The auxillary panel contains an altimeter, clock and temperature gauge together with a series of lights to indicate which filters are being exposed. The 16 mm films are used to confirm the altitude and exposure time data given on the flight data cards. The mach number for each sample is calculated from the true air speed and forecast temperature and is used, together with the flight altitude and Professor Reid's sampler calibration

curves, to determine the air flow rate for the sample. The flow rate is multiplied by the exposure time to obtain the volume of ambient air sampled and, through the application of a density correction, the volume of standard air.

Construction of Flight Cross-Sections

A meteorological analysis is prepared for each sampling mission as an aid in the interpretation of the radioactivity data. This analysis is based almost entirely on the radiosonde observations (RAOBS) from 44 weather stations located in the vicinity of the sampling corridor.

Each RAOB employed in the analysis is plotted on a meteorological sounding diagram (tephigram), which shows the temperature as a function of altitude (pressure). For most of the stations upper wind data, obtained by electronic tracking of the balloon, are also available. (The combined radiosonde and upper wind measuring system is known as "rawinsonde," and the wind observation is referred to as a RAWIN report.) The RAOB and RAWIN data corresponding to each sampling mission are coordinated by plotting them on a vertical (latitude-altitude) cross section representing the sampling plane. The meteorological analyses shown in the flight cross sections are based on these data, and on the lapse rate (temperature-altitude) curves on the tephigrams. The analysis is restricted to the representation of the jet stream (where sufficient upper wind data were available), delineation of the tropopause, and a general indication of stability in the stratosphere.

The jet stream, a narrow core of high wind speed found near the polar tropopause and generally at the bottom of the gap between the polar and tropical tropopauses, is shown by means of isotachs (lines of equal wind speed).

The tropopause is the boundary layer separating the troposphere below, with its large lapse rate of temperature, from the stratosphere above, in which the lapse rate is small or even negative. A negative lapse rate signifies temperature increasing with altitude, and is also known as an inversion. In many cases the tropopause can be identified unambiguously by the discontinuity of lapse rate. But often the transition from troposphere is diffuse. Since there is no universally accepted strict definition of the tropopause, there is some element of subjectivity in the tropopause analysis. The tropopause is shown on the cross sections as a thick black line, the width of the line indicating the fact that the tropopause is a layer rather than a mathematical surface. Almost invariably two (or more) distinct tropopauses, the low polar and high tropical tropopauses, can be found, with the two overlapping in the gap region.

The lapse rate is a useful measure of the intensity of convection (overturning, vertical mixing) that can develop in a layer of air. This tendency to convection is referred to as instability, while the inhibition of convection is called stability. The greater the lapse rate, the smaller the stability. An inversion layer is one of great stability. The stratosphere is generally stable, but not uniformly so. Some regions

of the stratosphere, e.g., the tropical stratosphere, are almost invariably characterized by inversions. But others, e.g., the arctic stratosphere in winter, exhibit positive lapse rates and are less stable. To indicate the spatial variation of stratospheric stability in the vertical cross sections, the inversion layers are hatched. The hatched areas thus represents definitely stable regions of the stratosphere.

The tracks and sampling segments of the aircraft are shown by arrows on the cross sections. The strontium-90 activity and the Tungsten-185 activity, expressed in dpm 1000 SCF, are plotted respectively above and below the appropriate arrow. Also shown on the cross sections are the reports of atmospheric turbulence provided by the pilots of the U-2 aircraft. While these reports are necessarily subjective estimates, they are of considerable interest. There is convincing evidence of turbulence not only in the jet stream and gap regions, where it was expected, but in the stratosphere itself as well. These unique HASP observations of stratospheric clear-air turbulence suggest a mixing process the mechanism of which has not yet been fully explained. (The fact that moderate and severe turbulence has been reported in inversion layers of the stratosphere suggests that the source of this bumpiness may be internal wave motions rather than convection. Such wave motions, it should be noted, would not necessarily be effective for the transport of debris.)

Figures 33 and 34 show typical cross sections of **missions** conducted in February 1959 and October 1958. Figure 33 shows the tendency of

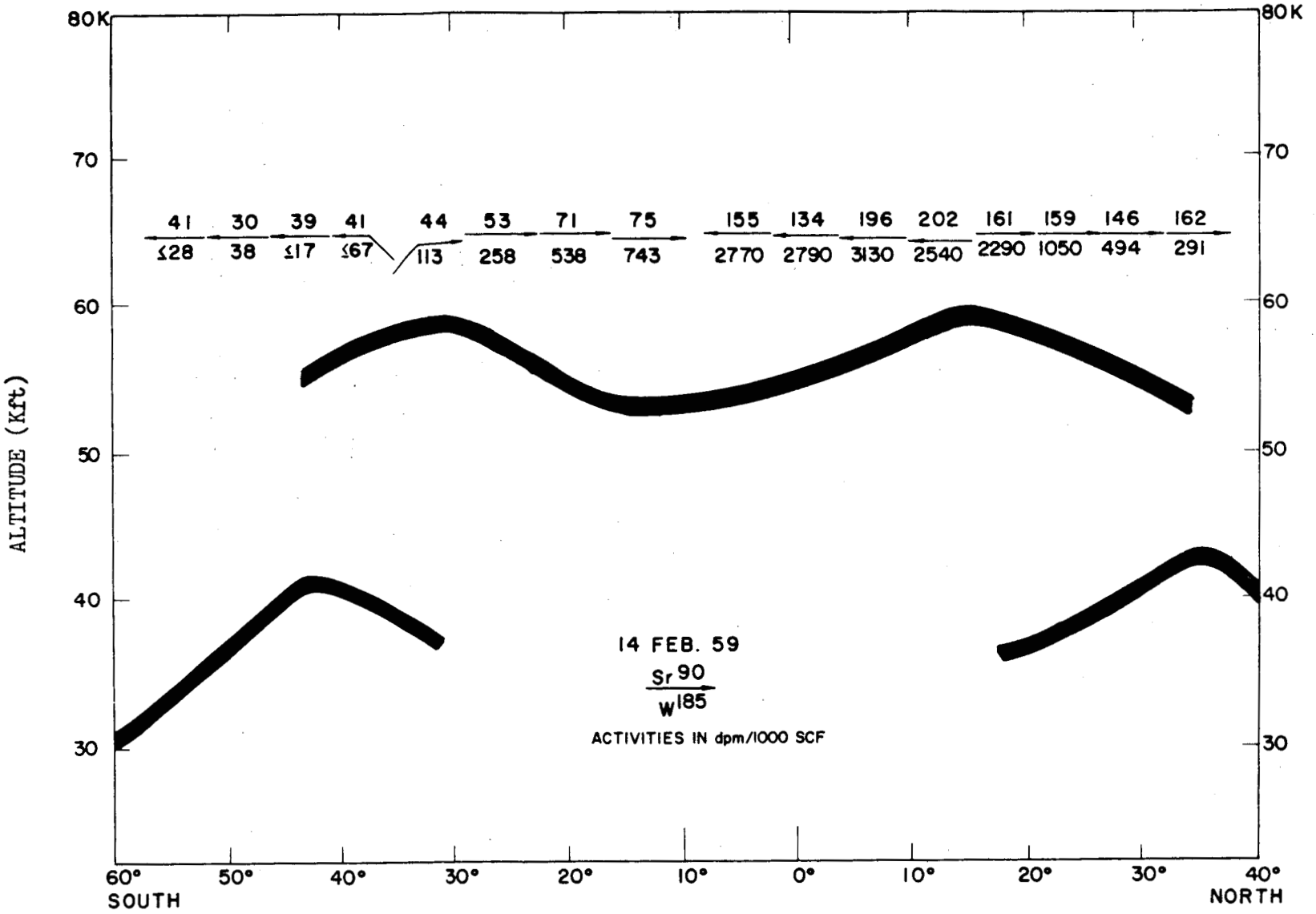

80K
70
60
50
40
30
ALTITUDE (Kft)
41 30 39 41 44 53 71 75 155 134 196 202 161 159 146 162
≤28 38 ≤17 ≤67 113 258 538 743 2770 2790 3130 2540 2290 1050 494 291
14 FEB. 59
Sr 90
W 185
ACTIVITIES IN dpm/1000 SCF
60° SOUTH
50°
40°
30°
20°
10°
0°
10°
20°
30°
40° NORTH

FIGURE 33

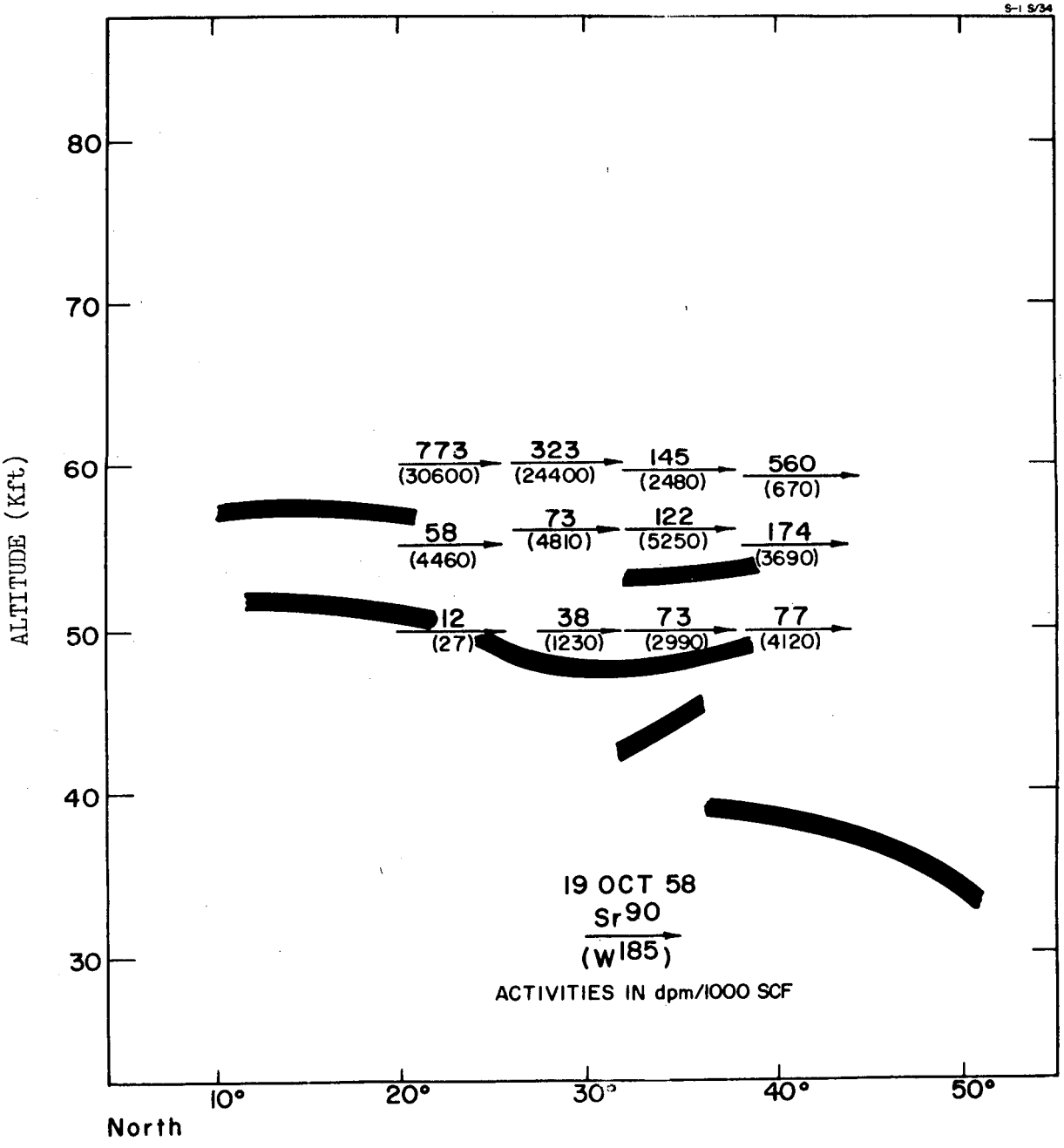

S-1 5/34
ALTITUDE (Kft)
80
70
60
50
40
30
773
(30600)
323
(24400)
145
(2480)
560
(670)
58
(4460)
73
(4810)
122
(5250)
174
(3690)
12
(27)
38
(1230)
73
(2990)
77
(4120)
19 OCT 58
Sr90
(W185)
ACTIVITIES IN dpm/1000 SCF
10°
20°
30°
40°
50°
North

FIGURE 34

material injected in the tropical stratosphere to move toward the Northern Hemisphere. A rather regular gradient is displayed.

Figure 34 shows interceptions of hot clouds both from Pacific and Siberian tests. Note the Tungsten drops off toward the north showing the lack of this material in the Russian cloud. This figure also shows the increasing gradient above the tropopause and through the tropopause gap. Further discussions of these phenomena are contained in Chapters VII and VIII.

Analytical Data for Cesium-137 and Plutonium

Since cesium-137 and plutonium are potentially hazardous a number of HASP samples have been analyzed for these nuclides. It has been expected that the relative production rates of cesium-137 and strontium-90 and to some extent those of plutonium-239 and strontium-90 should have been fairly constant from one weapon to another. Since all three of these nuclides have long half lives it should then follow that there are fairly constant cesium-137/strontium-90 and plutonium/strontium-90 ratios in stratospheric debris and in surface fallout from stratospheric debris. The HASP measurements have been carried out in hopes of determining these ratios.

Analytical Data for Other Fission Products

A number of fission products which are not generally believed to be hazardous are measured in HASP filter samples. These nuclides all have half lives of less than a year and their relative concentrations in debris are a measure of its apparent age.

Cerium-144 is one of these. This nuclide has a half life of 285 days and besides being useful for estimating the age of old debris can be used in studies of fractionation of debris during the cooling of the fireball; cerium-144, unlike strontium-90 and cesium-137, has no rare gas precursor.

Barium-140 (half life = 12.8 days) is useful for distinguishing and dating debris produced only a month or two before sample collection. Zirconium-95 (half life = 65 days) and yttrium-91 (half life = 58 days) are, with strontium-89 (half life = 50 days) useful for determining the apparent age of debris which is several months old.

Analytical Data for Tungsten-185 and Rhodium-102

Two types of tracer nuclides were introduced into stratospheric debris during HARDTACK. These were tungsten-185 and other isotopes of tungsten (wolfram), produced by certain weapons detonated on barges containing silica sand, and rhodium-102 produced in small amounts by several weapons during HARDTACK. One high stratospheric injection of rhodium-102 was made during this period.

Analyses of the distribution of tungsten-185 through the stratosphere have proven quite useful in the calculation of the mixing rates of stratospheric air. Since the short half life of that nuclide (about 74 days) has led to increasing imprecision in the measurements, analyses of tungsten-181 (half life = about 125 days) have been performed on many samples in place of those on tungsten-185 since the summer of 1959.

Extrapolation Beyond the Range of HASP Sampling

In order to determine the strontium-90 burden of the stratosphere it is necessary to supplement data from the High Altitude Sampling Program with data on the distribution of nuclear debris through the upper stratosphere. For this purpose use has been made of measurements of stratospheric concentrations of fission products performed as part of Project ASHCAN, sponsored by the Atomic Energy Commission. Samples have been collected by balloon-borne filter samples at altitudes up to about 90,000 feet. The reliability of the data from this project has been questioned[11, 12] because of uncertainties in the sampler efficiency, especially at the higher altitudes, and because of the poor reproducibility of some of the measurements. In spite of the objections which have been raised against the ASHCAN data, however, the vertical distribution of debris through the stratosphere which they indicate appears generally to be reasonable and should provide a usable basis for extrapolation of HASP data to the upper stratosphere.

Calibration Program

Calibration of the ASHCAN samplers has been carried out using laboratory tests run at simulated flight conditions and using in-flight calibration with aircraft samplers. Holland[13] has summarized the data on filter calibration and has concluded that results from the in-flight tests correspond with laboratory results for particles with diameters between 0.02 and 0.2 micron. It would appear that the curve, shown by Holland, for a diameter of 0.026 micron best fits the results of the

intercalibration flight of 6 March 1958, flown with a HASP aircraft. It would also appear that curves for particles with diameters of 0.14 micron and about 0.015 micron include between them all in-flight data and that the efficiencies represented by these curves may be taken as upper and lower limits.

Table 6 shows a comparison of ASHCAN and HASP strontium-90 data.

TABLE 6

Comparison of ASHCAN and HASP Strontium-90 Data

Altitude (feet)	45°N Minneapolis		32°N San Angelo		9°N Canal Zone		23°S Sao Paulo	
	HASP	ASHCAN	HASP	ASHCAN	HASP	ASHCAN	HASP	ASHCAN
				November 1957 - December 1958				
90,000	--	16	--	19	--	73	--	30
80,000	--	31	--	33	--	181	--	54
70,000	83	--	110	--	292	--	--	--
65,000	229	119	152	156	326	419	89	87
60,000	361	--	184	--	437	--	69	--
55,000	159	--	94	--	3	--	18	--
50,000	83	95	63	60	1	--	12	5
				January 1959 - August 1959				
90,000	--	66	--	90	--	--	--	74
80,000	--	160	--	235	--	--	--	119
70,000	--	--	--	--	251	--	100	--
65,000	194	272	212	175	167	--	65	81
60,000	148	--	152	--	56	--	50	--
55,000	137	--	106	--	13	--	9	--
50,000	94	110	--	95	2	--	6	--

Since the sampler efficiencies for the ASHCAN system are not well known and the accuracy of the radiochemical analyses has been questioned, an independent check on the concentrations of nuclear debris in the upper stratosphere is desired. This is supplied by measurements of stratospheric concentrations of excess carbon-14 published by Hagemann, et al[14].

Although there may be some differences in behavior between gaseous and particulate debris there should be at least a general correspondence.

There is some question as to the amount of correlation which may be expected between vertical concentration profiles of gaseous nuclear debris, such as excess carbon-14, and those of particulate debris, such as strontium-90. A difference in vertical concentration could be caused by a difference in total yield between the weapons which produced the bulk of the carbon-14 now in the stratosphere and the weapons which produced the bulk of the strontium-90.

A second mechanism for the separation of carbon-14 from the particulate debris produced during a surface burst is suggested by the observation that clouds from air and water surface bursts stabilize at higher altitude than do clouds from land surface or sandloaded barge shots. Evidently the presence of the sand or soil in the fireball inhibits its ascent. It is possible that the air around the weapon when it is detonated, in which the carbon-14 is produced by neutron activation, rises faster than the center of the fireball which contains the fission products and vaporized soil. Thus there might be separation of carbon-14 from fission products in ground surface shots and some barge shots even if there were no separation in air shots. If CASTLE tests produced a high concentration of carbon-14 at very high altitudes, the residence time of this debris would have been so much longer than that of the fission products injected at lower altitudes that the maximum could have persisted for many years. Later injections of carbon-14, from bombs of

lower yield, would not have reached these high altitudes and would have been removed more rapidly than the CASTLE debris. Thus only temporary modification of the original distribution would have resulted from later tests, until 1958 at least.

According to the interpretation of the carbon-14 data by Hagemann, et al[14] the concentrations increase upward to at least 90,000 feet at all sampling sites, the concentrations are higher in the Northern than in the Southern Hemisphere. The concentration at each altitude at the Canal Zone is lower than that at the same altitude at San Angelo or Minneapolis, and there is a marked variability in the distribution with time at the higher altitudes. They conclude that the "Brewer-Dobson Theory" best fits these phenomena and that the low concentrations at the Canal Zone result from large scale stratospheric circulation with the penetration of the equatorial tropopause by rising tropospheric air accompanied by poleward drift of air from the tropical stratosphere. The very poor representation of either CASTLE, REDWING, or British debris in samples collected at the Canal Zone seems to imply that this circulation proceeds at a rapid rate, i.e., that the air in the equatorial stratosphere is replaced almost completely in less than a year. No meteorological data have been cited to substantiate such a rapid replacement and it is contradicted completely by HASP strontium-90 and tungsten-185 data.

According to the HASP interpretation of the stratospheric distribution of nuclear debris there is a maximum in the vertical concentration profile in the lower stratosphere. The carbon-14 data appear to

be consistent with the existence of such a maximum at 80,000 to 90,000 feet at most or all of the four sampling sites. It is possible that such a maximum exists at altitudes above 90,000 at 10° North latitude. HASP data indicate the highest stratospheric concentrations of fission products occur at about 10° North latitude. If the highest carbon-14 concentrations also occur there, they must be at altitudes above 90,000 feet. The carbon-14 measurements seem to confirm the evidence from HASP that Soviet injections prior to October 1958 were all at low altitudes in the polar stratosphere and that they made no long range contribution to the stratospheric burden of nuclear debris. Virtually all of the "hot" carbon-14 samples are attributable to debris from United States Tests in the Pacific. Carbon-14 from Soviet tests was either removed from the stratosphere rapidly or was fairly well diluted before it reached the sampling sites. In confirmation of HASP observations, the carbon-14 measurements indicate that poleward transfer of debris occurs preferentially during the winter season in each hemisphere. Increases in carbon-14 concentrations observed at San Angelo and Minneapolis occurred almost entirely during October through April while those observed at Sao Paulo occurred during May through September. Except for REDWING (?) debris encountered at Sao Paulo in October 1956 - February 1957 and Soviet (?) debris encountered at Minneapolis in July - August 1957 the seasonal dependence of increases in activity is quite evident. It is evident from the "hot clouds" encountered throughout the carbon-14 program that carbon-14 from CASTLE rose to higher altitude - probably much of it rose

beyond 90,000 feet - than did that from REDWING or HARDTACK, for which the most active samples obtained at the Canal Zone came from 65,000 to 85,000 feet.

Table 7 shows relative ASHCAN and Carbon-14 values using 65,000 feet as a reference and the HASP profile extrapolated using carbon-14 data.

TABLE 7

Comparison of ASHCAN and Carbon-14 Data

Altitude (feet)	ASHCAN 1958	ASHCAN 1959	Carbon-14 Apr 58 - Sep 58	HASP data extrapolated by Carbon-14 Profile 1958	HASP data extrapolated by Carbon-14 Profile 1959
				(dpm Sr^{90}/1000 SCF)	
	Minneapolis, Minnesota				
90,000	0.13	0.24	1.20	275	232
80,000	0.26	0.59	1.21	277	234
70,000	--	--	1.08	247 (83)	210
65,000	1.00	1.00	1.00	229 (229)	194 (194)
60,000	--	--	0.91	208 (361)	176 (148)
55,000	--	--	--	- (159)	- (137)
50,000	0.80	0.40	0.38	87 (83)	74 (94)
	San Angelo, Texas				
90,000	0.12	0.51	1.24	188	263
80,000	0.21	1.34	1.47	223	312
70,000	--	--	1.23	187 (110)	260
65,000	1.00	1.00	1.00	152 (152)	212 (212)
60,000	--	--	0.76	115 (184)	161 (152)
55,000	--	--	--	- (94)	- (106)
50,000	0.38	0.54	0.24	36 (63)	51

Altitude (feet)	ASHCAN 1958	ASHCAN 1959	Carbon-14 Apr 58 - Sep 58	HASP data extrapolated by Carbon-14 Profile 1958	HASP data extrapolated by Carbon-14 Profile 1959
	Canal Zone, Panama				
90,000	0.17	--	1.07	349	179
80,000	0.43	--	1.30	424	217
70,000	--	--	1.16	378 (292)	194 (251)
65,000	1.00	--	1.00	326 (326)	167 (167)
60,000	--	--	0.83	270 (437)	138 (56)
55,000	--	--	--	- (3)	- (13)
50,000	--	--	0.07	23 (1)	12 (2)
	Sao Paulo, Brazil				
90,000	0.34	0.91	1.33	118	86
80,000	0.62	1.47	1.66	148	118
70,000	--	--	1.33	118	86 (100)
65,000	1.00	1.00	1.00	89 (89)	65 (65)
60,000	--	--	0.67	60 (69)	44 (50)
55,000	--	--	--	- (18)	- (9)
50,000	0.06	--	0.17	15 (12)	11 (6)

Actual HASP mean values are listed in parentheses with these calculated values for comparison. The correspondence is rather poor. It appears that ASHCAN data are in better agreement with HASP data than are the carbon-14 measurements. Even if the carbon-14 concentration profiles were accepted for calculation of strontium-90 concentrations above the maximum altitude of the aircraft, however, the stratospheric burden obtained from HASP data would be raised by only 10 percent or so.

Extrapolation to the poles beyond the range of HASP aircraft has been made through the use of B-52 flights to the North Pole. The assumption that concentrations from 70°N to 90°N are similar is borne out. There is no reason to suspect that a similar situation does not prevail

in the South Polar regions south of 60°S. The HASP inventory is made on this assumption.

PART B

RESULTS

CHAPTER VII

DISTRIBUTION OF STRONTIUM-90 IN THE STRATOSPHERE

DISCUSSION

Preliminary results of the HASP Program have been reported in various places [15, 16, 17, 18].

During the course of the High Altitude Sampling Program the primary emphasis has been placed upon the determination of the distribution of strontium-90 through the atmosphere. Aspects of this distribution may be deduced from the individual missions and from compilations of data from many missions. Some compilations have been made which exhibit the mean distribution over a selected time interval and others which exhibit the changes which occur in the distribution with time. Certain characteristics of the processes which bring about mixing and transfer of nuclear debris through stratospheric air may be deduced from the observed distributions. The tungsten-185 data for HASP samples have also proven to be of great worth in delineating rates and mechanisms of stratospheric transfer. These processes are discussed in some detail in Chapters VIII and IX.

The stratospheric concentrations of cesium-137 and plutonium are calculated from the mean values of the cesium-137/strontium-90 ratio and plutonium/strontium-90 ratio and the stratospheric strontium-90 concentrations. Observed

variations in these ratios are often due to analytical error but some are clearly attributable to differences in the composition of debris from different weapons.

The ratios between strontium-90 and various fission products which have half lives of less than a year have been tabulated and interpreted in terms of the apparent origin of debris encountered in various parts of the stratosphere at various times.

Observed stratospheric concentrations of the tracer nuclides tungsten-185, tungsten-181 and rhodium-102, injected into the stratosphere during HARDTACK, have been tabulated according to latitude, altitude and time of sample collection to provide information on the mechanism and rate of stratospheric mixing.

The Distribution of Strontium-90 in the Stratosphere

The study of data for samples collected in various parts of the stratosphere sheds light upon the distribution of strontium-90 through the stratosphere. During the time that nuclear weapons tests were being carried out the concentrations of debris encountered in the various parts of the stratosphere depended upon the location of the test sites, the types of detonation and the yields of the weapons. More than six months had to elapse after the detonation of some weapons before the debris from them became mixed more or less uniformly through the stratosphere parallel to the direction of zonal circulation. However, the vertical concentration gradient, which was normally quite steep in the region just above the tropopause during times of weapon testing, diminished in slope rather

rapidly. Within six months after the cessation of testing, strontium-90 concentrations at about 5000 feet above the mean tropopause had decreased to one fourth of the values which were present shortly after the passage of the last "hot cloud" of debris. Although the strontium-90 concentration of air increases upward from the tropopause through several thousand feet, it should eventually pass through a maximum and then decrease toward higher altitudes. This maximum concentration should occur at a greater height in the tropical stratosphere than it does in the polar stratosphere. In the tropics it should occur well above the limit of HASP sampling, probably between 70,000 and 85,000 feet, but in the polar regions, at latitudes above 50^{o}, it may occur between about 60,000 and 70,000 feet. There are abundant data from HASP samples which demonstrate a pronounced difference in stratospheric concentrations of debris between the Northern and Southern Hemispheres.

Calculations have been made of the mean distribution of strontium-90 through the stratosphere during several time intervals. In order to achieve these summations from data for the individual HASP missions, each sample, according to the midpoint of its collection track, has been assigned to a 5^{o} latitude band and to a 5000 feet altitude layer. All samples collected in each layer of each band during a chosen time interval have been averaged to obtain the mean concentration for that stratospheric region. The intervals chosen were November

1957 through December 1958, January 1959 through August 1959, and September 1959 through November 1959.

The Distribution During November 1957-December 1958

During the first year or so of HASP sampling, a number of nuclear weapons test series, carried out by the Soviets, the British and the United States were injecting debris into the stratosphere. Debris from recent tests was often encountered during HASP missions and the resultant "hot" samples usually had strontium-90 concentrations several times as high as the "background" concentrations displayed by most samples. Such "hot" samples were collected in the tropical stratosphere following British test in November 1957 and September 1958 and United States tests in May through July 1958 and in the Northern polar stratosphere following Soviet tests in the fall of 1957, the spring of 1958 and the fall of 1958. December 31, 1958 was chosen as the end of the first interval for which the mean distribution of strontium-90 should be calculated since it not only ended a calendar year but also ended the last month during which fairly distinct "clouds" of debris from recent tests were encountered.

Meridional mixing should be sufficiently rapid to insure a fairly uniform concentration throughout the polar stratosphere at any one altitude. Although some variations might be expected in the Northern polar stratosphere, if meridional mixing there is actually quite slow and injections of debris from Soviet tests produce long lived "hot clouds," there is no evident reason for the

accumulation of large quantities of debris above the South Pole. Three missions were flown to the North Pole by a B-52 aircraft equipped with hatch samplers. The data from these missions show no significant variations in strontium-90 concentrations as a function of latitude between 60^{o} and 90^{o} North, in the lower polar stratosphere at least.

Extrapolation of HASP data to the top of the stratosphere may be done in several ways. Meteorological data indicate that, in the tropical stratosphere at least, vertical mixing should be comparatively slow. Observed heights of stabilization of clouds from nuclear weapons lead to the conclusion that little debris has actually been injected at altitudes above 100,000 feet in the Tropical Stratosphere. None has been injected above or even at that altitude in the Polar Stratosphere. It is natural, therefore, to expect a maximum in the vertical concentration gradient somewhere below 100,000 feet. It would appear most likely that this maximum should occur below 90,000 feet since few radioactive clouds, especially among those from tests carried out since 1954, have injected debris at even these altitudes. Vertical concentration gradients extending up to 100,000 feet may be calculated from Project ASHCAN filter sample data or from stratospheric carbon-14 data (see Chapter VI). The ASHCAN data generally indicate a maximum concentration between 65,000 feet and 80,000 feet. The carbon-14 data generally indicate such a maximum somewhere between 80,000 and 90,000 feet. The ASHCAN data have been used for the extrapolation of the HASP

data because they represent analyses of filter samples. The accuracy of the assumed ASHCAN filtering efficiency is not well established but, since the concentration profiles agree in general with those from the carbon-14 sampling program and with distributions predicted on the basis of heights of stabilized clouds, their use appears to be justified.

The calculation of the stratospheric burden of strontium-90 from extrapolated HASP data is shown in Table 8, Parts A and B. In Part A are listed the mean strontium-90 activities for each 25 millibar layer in each 10 degree latitude zone. In Part B these mean activities, X_i, have been used to calculate the total millicuries per square mile and the total megacuries in each 10 degree latitude zone. The equation used is:

$$A = (\sum X_i)\,(8.55 \times 10^{-3}\ \frac{mc}{dpm}\quad \frac{1000\ SCF}{mi^2})$$

where A is the mean activity for the 10 degree latitude band, expressed in millicuries per square mile, and X_i is the mean activity for a 25 millibar layer, expressed in disintegrations per minute per 1000 standard cubic feet of air. The stratospheric strontium-90 burdens during November 1957 through December 1958 were 0.66 megacuries in the Northern Hemisphere and 0.29 megacuries in the Southern Hemisphere, for a total of 0.95 megacuries. It must be pointed out that this estimate does not adequately reflect the contribution from the Fall 1958 Soviet tests. Only a few samples representing debris from these tests were included in calculating this burden. We have used this calculation,

Table 8. THE DISTRIBUTION OF STRONTIUM-90 IN THE STRATOSPHERE - NOVEMBER 1957 - DECEMBER 1958

A. Mean concentrations (in dpm Sr^{90}/1000 SCF) for each 25 millibar layer, Estimated from extrapolated HASP data.

	Latitude (South)								
Millibar Layer	90-80	80-70	70-60	60-50	50-40	40-30	30-20	20-10	10-0
0- 25	---	---	---	---	5	5	5	15	50
25- 50	5	8	10	15	55	50	60	140	300
50- 75	20	25	30	50	80	75	90	150	250
75-100	50	50	55	55	65	60	30	40	110
100-125	50	50	45	35	35	40	15	---	---
125-150	30	30	30	25	30	25	10	---	---
150-175	25	25	20	20	20	15	5	---	---
175-200	20	20	15	15	15	10	---	---	---
200-225	15	14	12	8	5	---	---	---	---
225-250	11	9	5	3	---	---	---	---	---
250-275	5	3	---	---	---	---	---	---	---

	Latitude (North)								
Millibar Layer	0-10	10-20	20-30	30=40	40-50	50-60	60-70	70-80	80-90
0- 25	100	75	30	15	5	3	---	---	---
25- 50	250	175	105	90	60	35	25	20	15
50- 75	300	180	190	200	150	160	130	90	60
75-100	170	80	100	160	200	190	220	210	205
100-125	---	10	30	75	140	230	240	220	210
125-150	---	---	10	70	150	190	170	175	180
150-175	---	---	5	50	120	125	115	115	120
175-200	---	---	---	25	70	80	80	85	90
200-225	---	---	---	8	30	40	60	70	75
225-250	---	---	---	---	9	20	30	50	55
250-275	---	---	---	---	---	8	12	15	18
275-300	---	---	---	---	---	---	5	10	10

Table 8: (Continued)

B. Calculation of stratospheric strontium-90 burden.

Latitude Band	Σ Xi	$\frac{\text{mc } Sr^{90}}{mi^2}$	Surface Area (10^6 mi^2)	Stratospheric Burden (MC)
80-90N	1038	8.87	1.50	0.013
70-80N	1060	9.06	4.44	0.040
60-70N	1087	9.29	7.25	0.067
50-60N	1081	9.24	9.85	0.091
40-50N	1034	8.84	12.2	0.108
30-40N	693	5.92	14.1	0.083
20-30N	470	4.02	15.6	0.063
10-20N	520	4.45	16.6	0.074
0-10N	820	7.01	17.1	0.120
0-90N				0.66 megacuries
0-10S	710	6.07	17.1	0.104
10-20S	345	2.95	16.6	0.049
20-30S	215	1.84	15.5	0.029
30-40S	280	2.39	14.1	0.034
40-50S	310	2.65	12.2	0.032
50-60S	226	1.93	9.85	0.019
60-70S	222	1.90	7.25	0.014
70-80S	234	2.00	4.44	0.009
80-90S	231	1.98	1.50	0.003
0-90S				0.29 megacuries

therefore, to represent the "mid-1958" burden.

The Distribution During January 1959 - August 1959

Since January 1959 no "hot clouds" of debris have been sampled by HASP. Mission to mission variations in stratospheric strontium-90 concentrations, for the most part, have been small. Trends have been discernible in the data for several regions but pronounced fluctuations in concentration have been evident only in regions adjacent to the tropopause gap where stratospheric and tropospheric conditions alternate.

The general distribution is the same as that for 1958, with higher concentrations in the Northern than the Southern Hemisphere. Concentrations have fallen, especially in the Northern Hemisphere and especially at altitudes near the tropopause. In the high polar stratosphere, however, concentrations have increased. The increase in the Northern polar stratosphere probably resulted from injections of debris at high altitudes by high yield Soviet weapons detonated in October 1958. The increase in the Southern Hemisphere probably resulted from southward migration of debris from the tropical stratosphere during May through July 1959.

In general it may be stated that relatively large amounts of debris were lost from the stratosphere between mid-1958 and mid-1959. The loss would be even more evident except for a pronounced disparity between 1959 and pre-1959 ASHCAN data. ASHCAN collection efficiency seems to have increased markedly

at the end of 1958 with a corresponding increase in the reported stratospheric concentrations.

In Table 9, Parts A and B, the stratospheric burden for this interval is calculated, using the same techniques described for Table 8. The results are 0.57 megacurie for the Northern and 0.24 megacurie for the Southern Hemisphere, for a total of 0.8 megacurie.

Trends in the Distribution of Stratospheric Strontium-90

Between 10^{o} and 15^{o} North the strontium-90 concentrations were raised substantially by the 1958 United States and British tests. After November 1958, however, these concentrations fell rather rapidly until about April 1959. They then remained more or less constant through at least November 1959. It may be hypothesized that the rapid decrease in the strontium-90 concentrations in this latitude was accomplished through a northward movement of debris into and through the Northern polar stratosphere. This debris contributed to the "spring high" in surface fallout in the Northern Hemisphere during February - May 1959. This surface fallout displayed high tungsten-185 concentrations and thus must have contained large contributions from the tropical stratosphere. It will be instructive to see if there is another pronounced loss of debris from this region of the stratosphere during November 1959 - April 1960. It is noteworthy that "hot" debris from the November 8, 1957 British test at Christmas Island was encountered in this latitude band (as well as farther south) in February

Table 9. THE DISTRIBUTION OF STRONTIUM-90 IN THE STRATOSPHERE -- JANUARY - AUGUST 1959

A. Mean concentrations (in dpm Sr^{90}/1000 SCF) for each 25 millibar zone, estimated from extrapolated HASP data.

Millibar Layer	Latitude (South) 90-80	80-70	70-60	60-50	50-40	40-30	30-20	20-10	10-0
0- 25	8	12	18	30	48	50	50	50	58
25- 50	50	50	50	52	100	105	113	115	195
50- 75	50	50	50	55	57	55	70	70	108
75-100	43	45	50	52	48	33	15	8	7
100-125	32	35	35	38	28	20	---	---	---
125-150	25	28	30	32	17	12	---	---	---
150-175	12	13	15	20	10	10	---	---	---
175-200	5	5	5	5	10	10	---	---	---
200-225	---	---	---	---	2	---	---	---	---
225-250	---	---	---	---	---	---	---	---	---
250-275	---	---	---	---	---	---	---	---	---

Millibar Layer	Latitude (North) 0-10	10-20	20-30	30-40	40-50	50-60	60-70	70-80	80-90
0- 25	75	88	88	55	48	42	40	32	32
25-50	233	258	248	238	205	160	140	128	128
50- 75	132	132	150	188	200	208	188	190	190
75-100	13	25	58	118	142	153	180	185	185
100-125	---	---	22	107	98	100	125	128	128
125-150	---	---	12	85	78	75	75	78	78
150-175	---	---	---	27	57	60	55	58	58
175-200	---	---	---	2	20	35	37	37	37
200-225	---	---	---	---	5	15	20	20	20
225-250	---	---	---	---	---	5	10	12	12
250-275	---	---	---	---	---	---	---	5	5

Table 9: (Continued)

B. Calculation of stratospheric strontium-90 burden.

Latitude Band	Σ Xi	$\frac{\text{mc Sr}^{90}}{\text{mi}^2}$	Surface Area (10^6 mi^2)	Stratospheric Burden (MC)
80-90N	873	7.47	1.50	0.011
70-80N	873	7.47	4.44	0.033
60-70N	870	7.44	7.25	0.054
50-60N	853	7.30	9.85	0.072
40-50N	853	7.30	12.2	0.089
30-40N	820	7.01	14.1	0.099
20-30N	578	4.94	15.6	0.077
10-20N	503	4.30	16.6	0.071
0-10N	453	3.88	17.1	0.066
0-90N				0.57 megacuries
0-10S	368	3.15	17.1	0.054
10-20S	243	2.08	16.6	0.035
20-30S	248	2.12	15.5	0.033
30-40S	295	2.52	14.1	0.036
40-50S	320	2.74	12.2	0.033
50-60S	284	2.43	9.85	0.024
60-70S	253	2.16	7.25	0.016
70-80S	238	2.04	4.44	0.009
80-90S	225	1.92	1.50	0.003
0-90S				0.24 megacuries

1958. The pronounced layering of air in the stratosphere at this latitude causes the sharp concentration gradient evident in the data for 1959.

It appears that several points will merit further study. The first of these concerns the poleward migration of debris from the tropical stratosphere. This migration, rather than occurring at uniform rates throughout the year, appears to proceed most rapidly toward the winter pole: toward the North Pole in November - April, and toward the South Pole in May - October. It will be interesting to look for verification of this hypothesis in data from HASP samples collected in the Northern Hemisphere during the winter of 1959-1960. This preferential transfer of debris toward the winter pole, combined with an increase in the rate of loss of debris from the stratosphere during the spring, could produce a "spring high" in surface fallout rate in the Northern Hemisphere even in the absence of fall or spring Soviet testing, though such testing would naturally increase the size of the effect.

A second important point concerns the actual path followed by debris from the tropical stratosphere as it diffuses toward the poles. The best evidence on this path stems from the analyses of samples for tungsten-185. A zone of maximum tungsten-185 concentration, shown diagrammatically in Figure 35, lies near the limit of HASP sampling in the tropical latitudes and dips down to between 50,000 and 55,000 feet in the polar stratosphere. Evidently, this zone exists because poleward transfer of debris from the tropics occurs along such

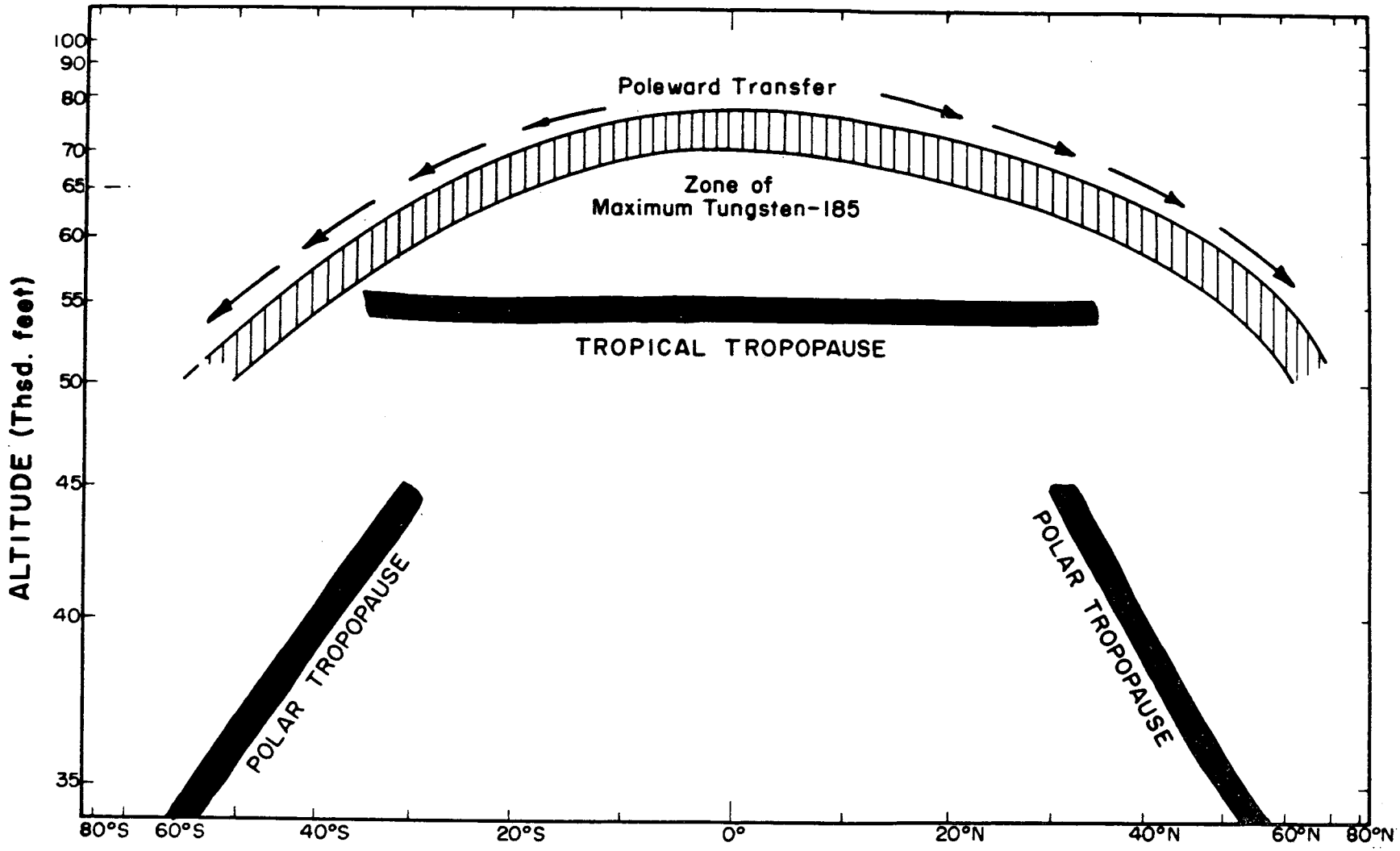


FIGURE 35 THE CONFIGURATION OF SURFACES OF POLEWARD TRANSFER DEDUCED FROM THE SHAPE OF THE ZONE OF TUNGSTEN-185 CONCENTRATION

curved surfaces with meridional mixing rates through several hundred miles exceeding rates of vertical mixing through 5000 feet. Since the weapons which injected most of the tungsten-185 were of moderate yield, the zone of maximum tungsten-185 concentrations occurs at relatively low altitudes. The corresponding zone of maximum strontium-90 concentration should be at a higher altitude. Debris from high yield Soviet weapons has penetrated the stratosphere only as high as this zone, probably because the rising of radioactive clouds from nuclear weapons through the polar stratosphere is impeded by the stratospheric structure. Transfer of debris from the tropical stratosphere into the Southern polar stratosphere during May - October 1958 resulted in high concentrations at all altitudes in that region during late 1958 because United States and British tests were injecting debris at all altitudes in the tropical stratosphere during the transfer. The transfer of debris into the Southern polar stratosphere during May - August 1959 resulted in high concentrations only at high altitudes in that region during mid-1959; however, since little debris remained in the lowermost tropical stratosphere by mid-1959, it had been lost through the tropopause or had been transferred into the Northern polar stratosphere during November 1958 - April 1959.

A third important point concerns the stratospheric residence time of United States-British and Soviet debris. This is discussed at length in Chapter IX, but a few remarks will not be out of place here. It is obvious that United

States-British debris injected into the lower stratosphere during 1958 exhibited a short residence time. As confirmed by the tungsten-185 data (see Chapter VIII and Chapter IX), more than half had escaped from the stratosphere within a year. Soviet debris injected into the stratosphere in September - October 1958 exhibited an even shorter residence time; more than one half had escaped within six months.

As a consequence of these short residence times there was a high rate of surface fallout in the Northern Hemisphere in the spring of 1959. If the behavior of stratospheric debris could be described strictly by first order kinetics there should be no "spring highs." If mixing within the stratosphere could be described by first order kinetics with modifications in the rate of escape into the troposphere to account for "spring highs," it should be predicted that there will be another "spring high" in 1960. HASP data suggest that the "spring high" is, fundamentally, a consequence of increased rates of poleward transfer in the winter hemisphere and increased rates of loss to the troposphere in the spring. However, the size of the spring high is primarily a function of the amount of debris present in the lower stratosphere -- tropical as well as polar -- as a result of testing during the preceeding year. Thus, there should be a "spring high" in 1960, but it should be small in magnitude. How pronounced it will be depends upon how much vertical mixing occurs in the Polar stratosphere during the winter. If this mixing is sufficient to bring debris from 65,000 feet down to 40,000 feet,

the "spring high" should be clearly discernible; if it is not most of debris which will contribute to the "spring high" will be that transferred to the polar stratosphere from the lower tropical stratosphere during the winter of 1959-1960. This may not be enough to cause a significant increase in surface fallout rates.

The Distribution During September 1959 - November 1959

Over 500 samples collected during September 1959 - November 1959 have been analyzed for strontium-90 and the mean concentrations for each latitude and altitude are tabulated in Table 8. Sampling during this interval was limited almost exclusively to the Northern Hemisphere. No ASHCAN data are available for this time interval as of this writing, so ASHCAN data for early 1959 have been used to extrapolate these HASP data to the top of the stratosphere. In the mean the distribution is the same as that for January 1959 - August 1959, but the concentrations have continued to decrease in almost all stratospheric regions sampled. The strontium-90 burden for the stratosphere of the Northern Hemisphere has been calculated in Table 10, Parts A and B. The value of 0.51 megacurie can be compared with 0.66 megacurie for November 1957 - December 1958 (Table 8), and 0.57 megacurie for January 1959 - August 1959 (Table 9).

Table 10. THE DISTRIBUTION OF STRONTIUM-90 IN THE STRATOSPHERE -- SEPTEMBER - NOVEMBER 1959

A. Mean concentrations (in dpm Sr^{90}/1000 SCF) for each 25 millibar layer, estimated from extrapolated HASP data.

Millibar Layer	Latitude (North) 0-10	10-20	20-30	30-40	40-50	50-60	60-70	70-80	80-90
0- 25	150	150	125	75	65	45	30	25	20
25- 50	250	235	215	200	180	155	120	100	90
50- 75	120	110	130	140	165	170	175	165	160
75-100	15	35	50	70	105	140	150	150	150
100-125	---	---	5	20	55	90	100	90	90
125-150	---	---	---	8	50	60	70	70	70
150-175	---	---	---	5	30	35	40	40	40
175-200	---	---	---	3	25	30	30	25	25
200-225	---	---	---	2	10	20	20	15	15
225-250	---	---	---	---	8	10	15	10	10
250-275	---	---	---	---	5	8	10	8	8
275-300	---	---	---	---	3	5	8	6	6
300-325	---	---	---	---	---	---	5	5	5

B. Calculation of stratospheric strontium-90 burden.

Latitude Band	Σ Xi	$\frac{\text{mc } Sr^{90}}{mi^2}$	Surface Area (10^6 mi^2)	Stratospheric Burden (MC)
80-90N	689	5.89	1.50	0.009
70-80N	709	6.06	4.44	0.027
60-70N	773	6.61	7.25	0.048
50-60N	768	6.57	9.85	0.065
40-50N	701	5.99	12.2	0.073
30-40N	523	4.47	14.1	0.063
20-30N	525	4.49	15.6	0.070
10-20N	530	4.53	16.6	0.075
0-10N	535	4.57	17.1	0.078
0-90N				0.51 megacuries
0-90S			(Estimated)	0.13 - 0.21 megacuries

The tabulated Northern Hemisphere values may seem higher than would be expected during September - November 1959 for three reasons. First, the concentrations seem to be rather constant in the Autumn Polar Stratosphere with no marked departure noted. Second, some activity may have began to appear in the higher Northern Polar Stratosphere from one of the extremely high altitude shots of HARDTACK. Finally, the inventory has a proportionately larger number of hatch samples represented and this would tend to increase the inventory as much as 10%. The Southern Hemisphere value is estimated from past experience and cannot be confirmed until the results of the May-June 1960 South America spot-check are analyzed.

CHAPTER VIII

DISTRIBUTION OF SELECTED ADDITIONAL RADIONUCLIDES IN THE STRATOSPHERE

Stratospheric Concentrations of Cesium-137

If there is no fractionation between cesium-137 and strontium-90 or between plutonium and strontium-90 during the condensation of fallout particles in the fireball of nuclear explosions and if there is no process which actively separates these nuclides in debris stabilized in the stratosphere, the stratospheric strontium-90 burden combined with the measured cesium-137/strontium-90 and plutonium/strontium-90 ratios in debris will permit the calculation of stratospheric burdens of cesium-137 and plutonium. Consequently, cesium-137/strontium-90 and plutonium/strontium-90 ratios have been measured in a number of samples.

The number of HASP samples which have yielded each cesium-137/strontium-90 ratio is indicated in Figure 36. The variation from the mean appears to be normal and is doubtless due to real variations and to analytical errors. Samples which gave ratios lower than 1.0 or higher than 3.0 probably suffered principally from analytical errors. The cesium-137 measurement especially is subject to error. In general the diagram suggests that a mean value, calculated to be 1.8 ± 0.5 when only the two samples with the most extreme values of the ratio are omitted, is applicable to stratospheric debris. In Table 11 this ratio is

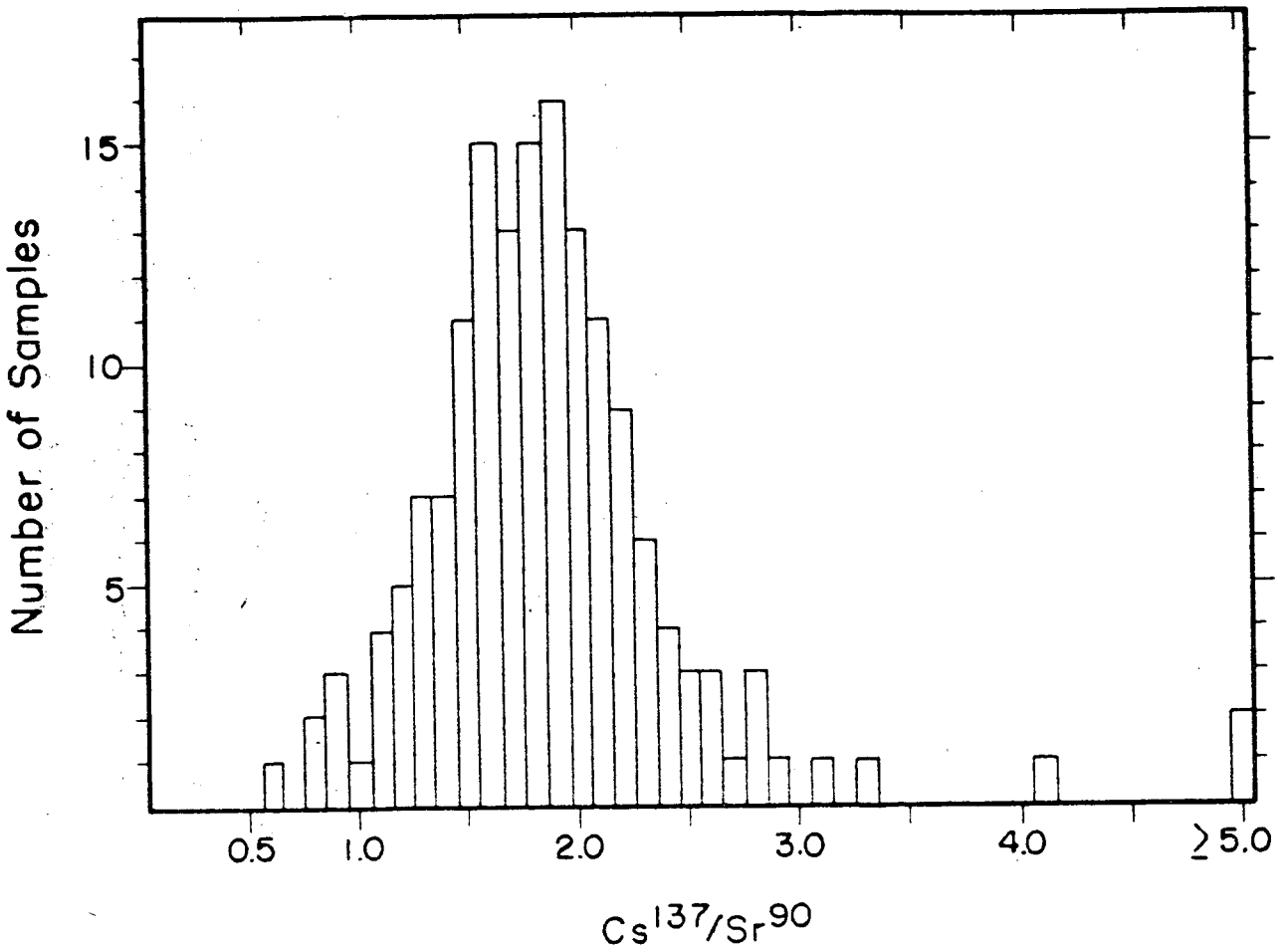


FIGURE 36 The Distribution of Cs^{137}/Sr^{90} Ratios in HASP Samples

tabulated for HASP samples containing debris attributable to specific bomb tests or characteristics of a stratospheric region at a particular time. Debris sampled in the Northern polar stratosphere appears to have a lower cesium-137/strontium-90 ratio than that sampled in either the Southern polar stratosphere or in the tropical stratosphere. Perhaps the debris in these latter regions has suffered some fractionation as a result of cesium-137 enrichment relative to strontium-90 in debris stabilized aloft. This would be more likely in United States ground or barge shots than in Soviet air shots so that a lower ratio could be found in the Northern polar stratosphere due to the presence of Soviet debris. The xenon-137 gaseous precursor of cesium-137 has a longer half life (3.8 minutes) than the krypton-90 precursor (33 seconds) of strontium-90. This might lead to fractionation in large yield weapons.

Stratospheric Concentrations of Other Fission Products

The relative concentrations of fission products in nuclear debris depend upon several factors, of which the most important should be the structure and yield of the weapon, the amount of fractionation which occurs during condensation and the age of the debris. Evidently, however, most weapons which have had yields high enough to inject debris into the stratosphere have had uranium-238 as a component and the greatest part of the fission products in the debris injected into the stratosphere has come from fast neutron fission or uranium-238. Consequently, there should be little variation in the composition of

Table 11. Cesium-137/Strontium-90 Ratios in HASP Filters

Northern Polar Stratosphere		Cs^{137}/Sr^{90}
Debris from fall 1957 Soviet tests	(1 sample)	1.7
Background debris, February-March 1958	(14 samples)	1.8 ± 0.4
Background debris, September-October 1958	(6 samples)	1.5 ± 0.4
Debris from fall 1958 Soviet tests	(6 samples)	1.6 ± 0.4
Background debris, April 1959	(4 samples)	1.6 ± 0.1
Background debris, May-June 1959	(12 samples)	1.8 ± 0.4
Background debris, July-August 1959	(3 samples)	1.4 ± 0.5
Background debris, September-October 1959	(4 samples)	1.8 ± 0.2
Tropical Stratosphere		
Debris from fall 1957 British tests:		
1. Intercepted in November 1957	(2 samples)	1.7 ± 0.1
2. Intercepted in February 1958	(3 samples)	1.6 ± 0.1
Debris from Spring 1958 Soviet tests	(4 samples)	2.0 ± 0.6
Background debris, May-June 1958	(8 samples)	2.4 ± 0.4
Debris from Fall 1958 British tests	(15 samples)	1.4 ± 0.5
Background debris, November-December 1958	(9 samples)	1.6 ± 0.2
Background debris, January-February 1959	(4 samples)	2.3 ± 0.6
Background debris, March-April 1959	(7 samples)	1.9 ± 0.1
Background debris, May-June 1959	(9 samples)	1.9 ± 0.9
Background debris, July-August 1959	(2 samples)	1.8 ± 0.1
Southern Polar Stratosphere		
October-November 1958	(5 samples)	2.2 ± 0.3
January-February 1959	(9 samples)	2.1 ± 0.5
March-April 1959	(10 samples)	1.8 ± 0.3
May, June and July 1959	(11 samples)	1.9 ± 0.5

stratospheric debris as a result of the structure or yield of the original weapon, except for the effect of these factors upon fractionation. Fractionation of nuclides occurs during the condensation of debris in the fireball of a weapon, presumably through the exclusion of volatile nuclides from the first formed particles and from adsorption onto solid material swept through the cooling fireball or subsequent cloud. Thus nuclides such as cerium-144 and zirconium-95 should condense early but nuclides such as xenon-140 and krypton-90 should not. Daughter products of gaseous nuclides, such as barium-140 and strontium-90 may become incorporated into smaller particles which form later. As a result stratospheric debris from some weapons, especially ground shots, may contain a higher percentage of volatile nuclides and nuclides with volatile or gaseous precursors than does debris from other weapons, especially air shots. But in spite of the initial composition of debris, which may vary somewhat as a result of weapon construction and fractionation effects, the composition at any specified time will depend primarily upon the age of the debris, i.e., the time which has elapsed since detonation.

Because samples collected during HASP have always contained debris from many weapons, detonated at quite different lengths of time before sample collection, few have had a composition suitable for identification of a particular weapon as a source. Occasionally, however, "hot" debris was sampled following the initiation of a new test series. Under these circumstances the "background"

of short-lived nuclides was often low enough to permit the use of the rate of decay of beta activity (according to the $t^{-1.2}$ law) or, better yet, the barium-140/strontium-90 ratio in the debris to pinpoint the detonation date of the weapon which contributed the fresh debris.

The youngest component of debris sampled in the tropical stratosphere before 30 October 1958 had been produced during HARDTACK in June and July 1958. By 30 October 1958, however, debris from the 3 September 1958 British test had reached the sampling corridor. This British debris continued to be sampled by all missions flown through the tropical stratosphere during October. The half life of the total beta activity suggests that debris from the 11 September 1958 shot was sampled by 10 October 1958, but the barium-140/strontium-90 ratios indicate that only debris from the 3 September 1958 shot was sampled. A sample collected at 38^{o} South latitude contained no detectable debris younger than that from HARDTACK, as might have been expected. Various samples show that the British debris had spread as far North as 20^{o} North and as far South as 19^{o} South, but had not reached 26^{o} South, by 17 October 1958. Actually, by 3 October 1958 enough British debris had reached 20^{o} North and 19^{o} South to affect the half life of the total beta activity and, at 19^{o} South, the barium-140/strontium-90 ratio of samples collected at those latitudes.

With the first interception of debris from the Fall 1958 Soviet test series, the identification of specific sources of debris became much more difficult. The

most southerly penetration of Soviet debris which can be identified with any degree of confidence occurred at 7° North on 28 November 1958.

It is especially interesting, since these Soviet tests were all carried out North of the Arctic Circle, that samples collected between 40° and 45° North on 19 and 22 October 1958 contained high concentrations of debris from these tests while samples collected at 50° North on 22 and 23 October contained little. Evidently zonal circulation was symmetrical about Northern Canada rather than about the North Pole at this time so that the main clouds of debris from Soviet tests were carried to more southerly latitudes as they moved about the earth in the stratosphere.

The absence of Soviet debris from a sample collected at 48° North on 20 November 1958, indicates that radioactive clouds retain at least a small measure of their original integrity for a number of weeks at least, so that some parcels of uncontaminated air remain even in a zone receiving large injections of debris over the course of a month.

We may conclude, then, that barium-140/strontium-90 ratios are useful for dating debris in HASP samples for the first eight to twelve weeks following weapon detonation and that these data may be used to measure diffusion rates of such debris during this time period. The half-life of decay of total beta activity may be useful for an even longer period, especially if a single test series follows a long suspension of testing, but the data are less precise. Actual information

on mixing of debris from the Fall 1958 British and Soviet tests was limited. However, no evidence was found for spread of the British debris beyond 20° South or 20° North during the first two months after detonation, and it probably spread almost that far within the first month. **On the other hand, Soviet debris,** injected North of the Arctic Circle, reached 20° North (and perhaps small quantities later reached 5° North) along the 64° West Meridian within a month after detonation, but this more likely resulted from symmetry of zonal circulation about Northern Canada rather than from extremely fast meridional diffusion rates. It is also noteworthy that while debris from a single British test required a month to complete its passage across the HASP sampling corridor, debris from a whole succession of Soviet tests, carried out over the course of several weeks, was swept through the corridor in less than a month with few interceptions of any one cloud by more than one HASP mission.

Other fission products, such as cerium-144, strontium-89, zirconium-95 and yttrium-91, which have half lives longer than that of barium-140 have proved less suitable for determining the age of individual samples, but may still yield useful information about stratospheric debris. None of the cerium-144/strontium-90 ratios are as high as would be expected on the basis of fission yield data. Newly formed debris, such as that which was abundant in both the tropical and North polar stratosphere in September-December 1958, should have a cerium-144/strontium-90 ratio of about 50. Actually this value was

seldom approached by any HASP sample, even by those containing high concentrations of recent debris. If the fission yield data are accurate this discrepancy has probably resulted from fractionation, to which the cerium-144/strontium-90 ratio should be rather sensitive. The only generalization of undoubted validity which can be drawn from cerium data appears to be the statement that debris in the Southern polar stratosphere is of a slightly greater average age than debris in the tropical or **Northern polar stratosphere.**

Strontium-89 Data

Strontium-89/strontium-90 ratios observed in HASP samples collected in each of four 10 degree latitude band are shown in Figure 37. Changes in the ratio as a result of the injection of fresh debris are evident, as is the decrease in the ratio due to radioactive decay. Since November 1958 this ratio has been fairly uniform, throughout the Northern Hemisphere. The ratio has been significantly lower in the Southern Hemisphere, however, though the difference in ratio corresponds to a difference in age of only two and a half months; in early 1959 at least. This difference in apparent age is less than might be expected on the basis of rates of southerward transport of strontium-90 and other nuclides observable in HASP data for samples collected during December 1958 - April 1959. It is significant, however, that the rate of southward transport from the tropical stratosphere increased in May - August 1959. If we hypothesize a similar rapid rate of southward transport during May - August 1958, we can explain

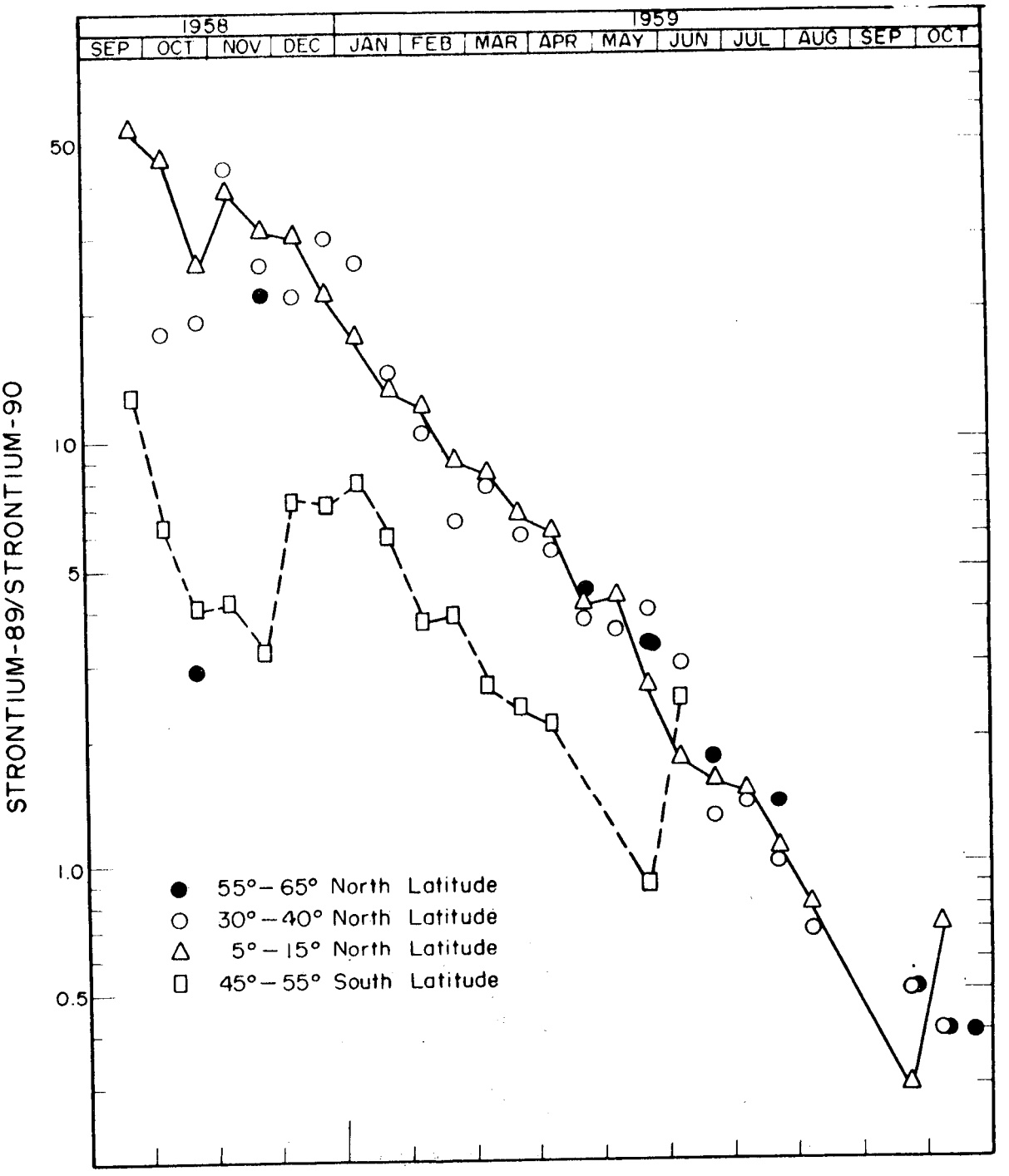


FIGURE 37 STRONTIUM-89/STRONTIUM-90 RATIOS IN STRATOSPHERIC DEBRIS

why the strontium-89/strontium-90 ratio in the Southern polar stratosphere was not much less than that in the tropical stratosphere during early 1959.

Stratospheric Concentrations of Tungsten Tracer Nuclides

Tungsten-185 and tungsten-181 were injected into the stratosphere in large quantities during HARDTACK. The distribution of these nuclides through the stratosphere during subsequent months may be used as evidence concerning the fate of debris injected into the stratosphere at about 11^{o} North latitude by weapons of moderate yield. Although the tungsten-185 data are discussed in Chapter IX, a few comments concerning them are appropriate here.

Bimonthly distributions of tungsten-185 have been computed and are shown in Figures 45 and 46, Chapter IX. The shape of the zone of maximum tungsten-185 concentration has been discussed above (see Figure 35) and the configuration of poleward diffusion surfaces has been deduced from it. The relative efficiency of vertical mixing in the polar stratosphere may also be estimated from the persistence of this zone in the polar stratosphere through many months. Obviously vertical mixing through 10,000 feet in this region is slow compared to the rate of meridional transfer from the Equator to 50^{o} North or South Latitude.

A second relationship concerns the ratio of peak tungsten-185 concentrations in the Northern polar stratosphere to those in the Southern polar stratosphere. This ratio was about 2 during September - October 1958, indicating a fairly rapid Southward transfer of debris from the tropical stratosphere during

May through August 1958, since the initial injection of this debris was at 10^{o} North. Peak tungsten-185 concentrations in the Northern polar stratosphere remained fairly constant through December 1958 - February 1959, but those in the Southern polar stratosphere decreased so that the ratio between these concentrations was about 8 by March - April 1959. This suggests rapid Northward diffusion of debris from the tropical stratosphere during the winter of the Northern Hemisphere. During April 1959 - July 1959 the peak tungsten-185 concentrations in the Northern polar stratosphere decreased, but those in the Southern polar stratosphere remained the same or actually increased, so that by July - August 1959 the ratio of these concentrations between Northern and Southern polar stratospheres had again dropped to 2. This suggests rapid southward diffusion of debris from the tropics during the winter of the Southern Hemisphere.

Tungsten-181 measurements have been made on a number of HASP samples in an effort to augment tungsten-185 data. Since tungsten-181 may be analyzed by gamma spectrometry of untreated filter paper, it presents a less expensive method of obtaining the same information as is supplied by tungsten-185 analyses. Unfortunately, correlation of tungsten-181 data with those for tungsten-185 is hampered by the lack of available standards for tungsten-181 and uncertainties in determinations of the half lives of both isotopes. An intercalibration of radiometric measurements of these isotopes was performed by comparison of their relative

counting rates in HASP samples collected in May through July 1958 with their relative activities predicted from tungsten isotope yields for the shot which produced the tungsten isotopes in these HASP samples. The tungsten-185/tungsten-181 ratio for HASP samples collected through August 1959 is sufficiently constant and sufficiently similar to the predicted ratio to justify confidence in the use of tungsten-181 data to augment tungsten-185 data. A sudden increase in the ratio in September 1959 suggests that the activity levels had become too low by then to yield reliable data.

Stratospheric Concentrations of Rhodium-102

An additional tracer nuclide, rhodium-102, was injected into the stratosphere in the amount of about 0.8 megacuries (based on beta decay). It was hoped that this nuclide would provide a unique tracer for debris injected at high altitudes. Unfortunately, rhodium-102 was also produced, in lower concentrations, by several surface bursts during HARDTACK. Table 12 indicates that the highest rhodium-102 concentrations are found in the tropical stratosphere with lower concentrations in the Northern and still lower in the Southern polar stratosphere. This distribution is that which would be expected for rhodium-102 injected into the stratosphere by HARDTACK surface bursts, but the concentrations are higher than would be expected. This distribution is not that which would be expected from high stratospheric debris if vertical mixing is more rapid in the polar than in the tropical stratosphere since, under these conditions, debris

from the high stratosphere would be returned to the lower stratosphere most rapidly in the polar regions.

Table 12. Stratospheric Rhodium-102 Concentrations

Measured concentrations in the stratosphere:

1. Southern Polar Stratosphere, July-August 1959:	10 dpm/1000 SCF
2. Tropical Stratosphere, September 1958-January 1959:	47 dpm/1000 SCF
Tropical Stratosphere, August 1959:	45 dpm/1000 SCF
3. Northern Polar Stratosphere, June 1959:	27 dpm/1000 SCF
Northern Polar Stratosphere, July 1959:	18 dpm/1000 SCF

More recent data indicates that higher North polar concentrations run as much as sixty times more than Equatorial stratospheric concentrations. This may be an indication that high stratospheric debris is beginning to descend in the polar regions. Similar data has been obtained by Martell.[20] Further rhodium-102 measurements will be required to verify the observed concentrations and to provide a better basis for interpretation of these data.

Characteristics of the Transfer Mechanism

Some of the characteristics of transfer and mixing of nuclear debris within the stratosphere may be deduced from a study of HASP data:

1. Meridional transport of debris occurs through diffusion along the concentration gradient rather than through the poleward drift of bodies of air con-

taining the bulk of the injected debris, as suggested by Hagemann, et al.[14] This is indicated by the stratospheric distribution of strontium-90 and tungsten-185 revealed by HASP samples.

2. Merirional transport of debris occurs along surfaces which slope downward from the Equator toward the poles. This is quite evident from the stratospheric distribution of tungsten-185 and is also suggested by the stratospheric distribution of strontium-90.

3. Poleward transport in each hemisphere is much more rapid during the winter season of that hemisphere than during the summer season. This is suggested by:

a. The occurrence of relatively high concentrations of strontium-90 in some regions of the Southern polar stratosphere during the Spring (September, October) of 1958 and Winter (June, July) of 1959;

b. The rapid decrease in strontium-90 activities at several altitudes between 10^{o} and 15^{o} North Latitude during the Winter (December through January) of 1958-1959;

c. Fluctuations in the ratio between the peak concentrations of tungsten-185 observed in the Northern and in the Southern polar stratospheres; Northern/Southern equalled about 2 following the Southern Hemisphere winter of 1958, about 8 following the Northern Hemisphere winter of 1958-1959, and about 2 during the Southern Hemisphere winter of 1959.

d. The similarity in apparent age between debris in the Southern polar stratosphere and tropical stratosphere during early 1959, as indicated by strontium-89/strontium-90, yttrium-91/strontium-90, and cerium-144/strontium-90 ratios. This similarity can be explained most readily by extensive Southward transport of HARDTACK debris during the Southern Hemisphere winter of 1958, during which HARDTACK was carried out.

This hypothesis explains, in part, the "spring high" in surface fallout. It also suggests that the almost equal partition of HARDTACK debris between the Northern and Southern Hemispheres during May through July 1958, observed by Lockhart, et al[21] was a result of stratospheric rather than tropospheric transport. It also explains the "spring high" in tungsten-185 fallout observed in Westwood rain in 1959.

CHAPTER IX

INJECTION, MIXING, AND TRANSFER OF STRATOSPHERIC DEBRIS

Stratospheric Injection - Depletion Model

During the course of nuclear weapons testing between 1945 and 1959, approximately 92 megatons of fission yield have been introduced into the atmosphere (Table 13).

Table 13a. United States and United Kingdom Nuclear Events
(Yield in Kilotons)

Year	Total Fission Yield	Total Fission Yield from Events, the Total Yield of Which was 1MT or Greater
1945	60	
1946	40	
1948	100	
1951	500	
1952-1954	37,000	36,000
1955	200	
1956	9,000	8,000
1957-1958	19,000	14,000

Table 13b. Soviet Nuclear Events
(Yield in Kilotons)

Inclusive Years	Total Fission Yield*
1945-1951	60
1952-1954	500
1955-1956	4,000
1956-1958	21,000

* A value of 50% has been arbitrarily selected for the fission-to-total yield ratio for all Soviet thermonuclear tests. As indicated in Table 13c, 50% is about the average fission-to-total yield ratio for all US/UK thermonuclear tests.

Table 13c. United States, United Kingdom, and Soviet Nuclear Events
(Yield in Kilotons)

	Fission Yield			Total Yield*	
Inclusive Years	Air Burst	Ground Surface Burst	Water Surface Burst	Air Burst	Surface Burst
1945 - 1951	190	550	20	190	570
1952 - 1954	1,000	15,000	22,000	1,000	59,000
1955 - 1956	5,600	1,500	6,000	11,000	17,000
1957 - 1958	31,000	4,400	4,600	57,000	28,000

These weapons have been fired under varying environmental conditions (air bursts, land surface bursts, and water surface bursts, in both Tropic and non-Tropic atmospheres) and varying yields. Cloud height and meteorological observations allow one to measure the fraction of the visible cloud which stabilizes in the stratosphere. Figure 38a shows the precent of the visible cloud which stabilizes above the tropopause as a function of yield under varying conditions of burst. This curve is based on the assumption that the mass of the cloud is distributed exponentially from its bottom to its top at a rate equal to the drop in pressure from the bottom to the top of the cloud. If the further assumption is made that the radioactive particles are distributed in the same manner as the

FIGURE 38a

STRATOSPHERIC CLOUD INJECTION (UNSCAVENGED)

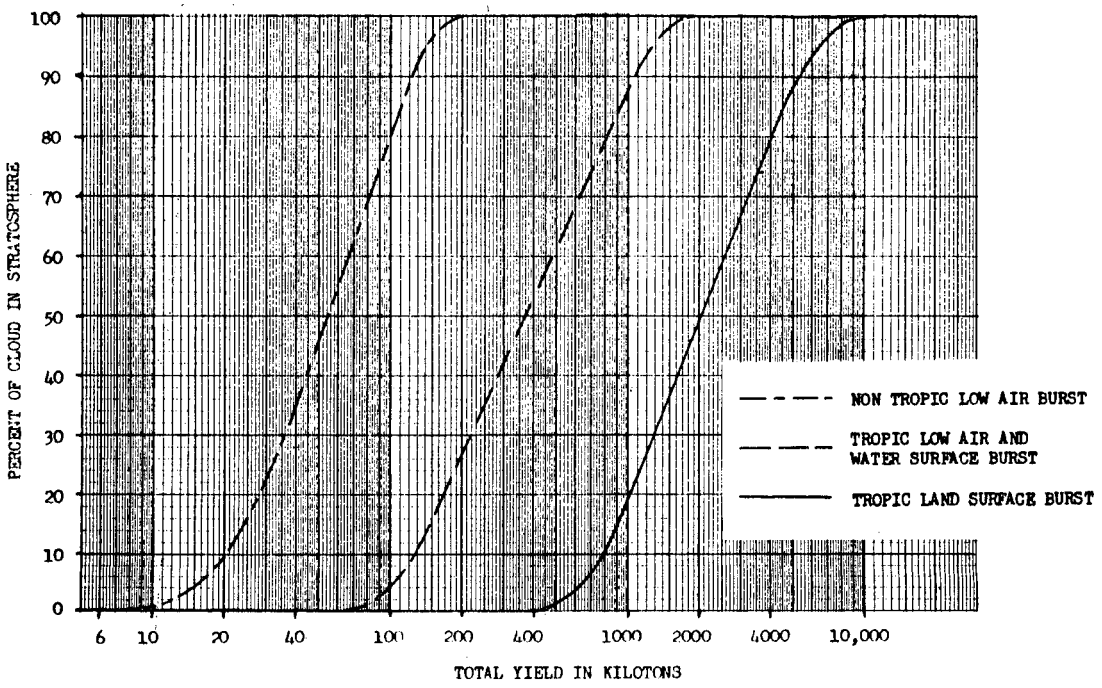
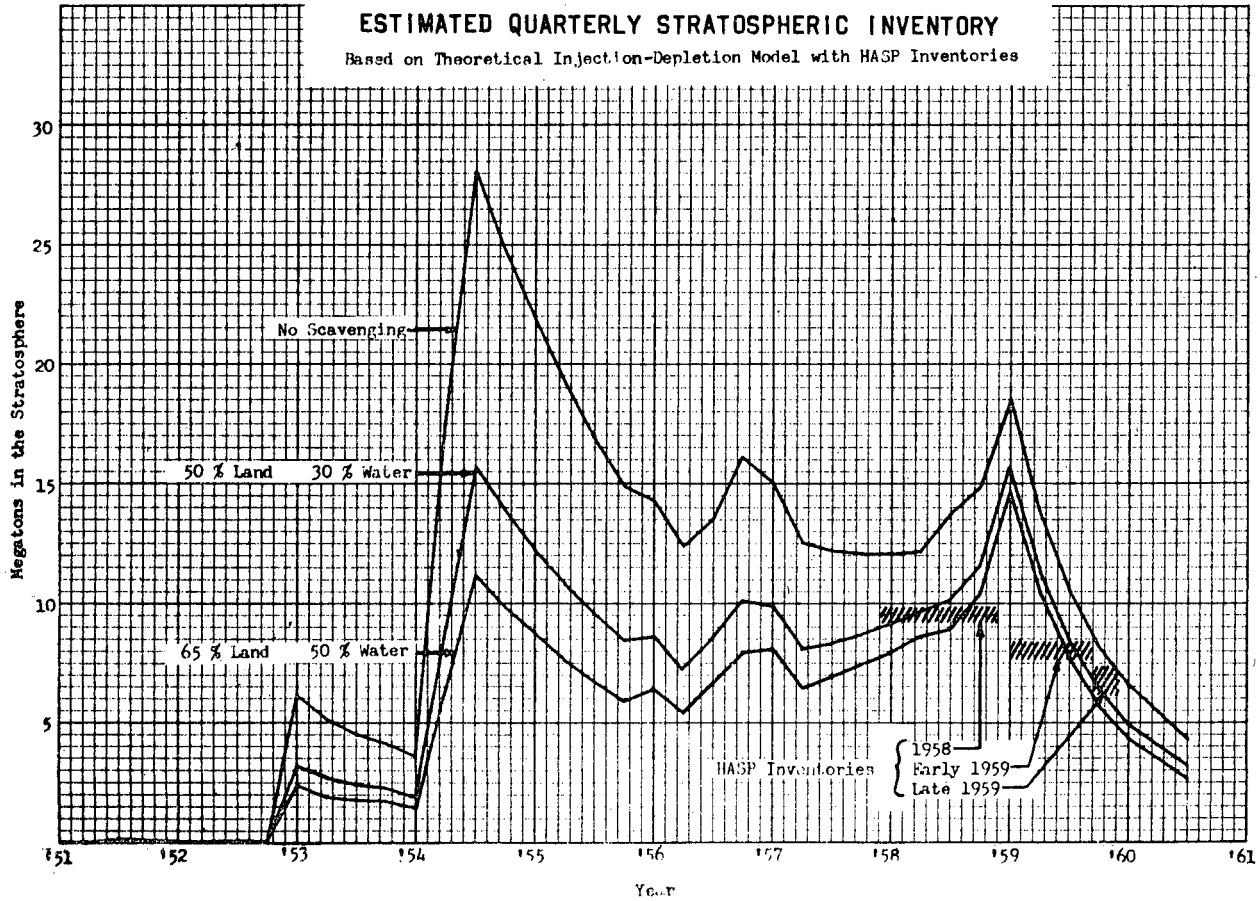


FIGURE 38b

ESTIMATED QUARTERLY STRATOSPHERIC INVENTORY

Based on Theoretical Injection-Depletion Model with HASP Inventories



air in the cloud, calculations of fission product injection can be made. Scavenging by water, tower materials, or earth sucked up into the fireball would have to be accounted for. It is noted in Figure 38a that almost all of a 200 KT low air burst fired in temperate or arctic regions would enter the stratosphere while an equatorial burst would not completely penetrate the tropopause. One reason is that the rising cloud has further to go to reach the stratosphere, another reason is that the tropical inversion is more pronounced. In the case of the 5 MT air burst, all of the material from both tropical and non-tropical weapons would enter the stratosphere but the tropical cloud would probably stabilize at a higher altitude because it would have less distance to travel in the stratosphere.

As the height of burst of a weapon is lowered to the surface of the earth, particles of dirt or water will be sucked up into the fireball. This material will reduce the amount of radioactivity, placed in the stratosphere, in two ways. First, the mass of material will tend to depress the height to which the cloud rises. This is especially noticeable in shots fired on land surfaces or on sand filled barges. Shots fired on water surfaces do not differ markedly from low air bursts in this regard, however. The second mechanism reducing the stratospheric inventory is that of scavenging. Apparently the large mass of material raised into the fireball has a tendency to condense into relatively large particles which drop out of the cloud to the ground within 100 miles or so of the detonation. A significant portion of the radioactivity from the cloud is removed by these

larger particles. (It is this material which constitutes the so-called "local" fallout which can produce lethal acute doses of radiation to unprotected individuals many miles away from the site of burst and beyond the radius of primary effects of the weapon, namely, blast, heat, and prompt radiation.) The net effect of these two mechanisms has been estimated to reduce stratospheric injections by as much as 80% for land surface bursts **and 70% to 20% for water surface** bursts.[11, 15] An estimate of reasonable numbers for the scavenging effect alone may be made from a knowledge of test yields, stratospheric residence times, and stratospheric inventory.

While the notion of a single half-residence time to characterize the rate of departure of material injected into the stratosphere from all tests to date has fallen into complete disrepute,[22] the idea of half-residence times for various latitudes and altitudes of injection may be a useful tool for calculation of inventories. The existence of seasonal effects detracts from the procedure but, if they are not too pronounced, probably will not invalidate the results. Figure 38b has been constructed on the assumption that material injected into the lower tropical stratosphere had a half-residence time of 10 months, higher tropical stratosphere 20 months, and non-tropical stratosphere 5 months. The evidence to be discussed later in this chapter that removal rate decreases with time is ignored in this calculation. The material was "decayed" with the half-life of strontium-90 in order that HASP inventories could be used for comparison. The

assumption was made in this chart that 0.1 megacurie equals 1.0 megatons of fission yield. The cloud percentage chart shown in **Figure 38a was used in the** construction.

A comparison of the upper "no scavenging" curve and the HASP inventories suggests that there has been a significant amount of scavenging. Since water surface burst cloud heights do not differ markedly from low air bursts it is felt that this type of burst has less scavenging than a land surface burst. Two sets of scavenging factors which give reasonable agreement with HASP Sr-90 values are indicated in the figure and have been used to construct the two lower curves. The net injection appears to be about 25 MT from Siberian tests and from 25 to 30 MT from Pacific tests. Injections from Teak and Orange, the two high altitude shots of HARDTACK, are not included.

Introduction to Spar's Mixing Theory

When a high yield nuclear weapon is detonated, the radioactive cloud which enters the stratosphere undergoes modifications due to several processes. The largest particles in the cloud fall to the earth within a few hours in the vicinity of the test site, constituting the so-called local fallout. Subsequently, smaller particles continue to settle out of the stratosphere under the influence of gravity and are distributed downwind of the test site for days or weeks as intermediate fallout. The particles smaller than about one micron (1 μ) in diameter, however,

have negligible gravitational settling velocities. Because of the viscous drag of the air on these particles, they remain airborne for long periods of time and behave like the air itself. This fine radioactive dust is redistributed in the stratosphere by large scale quasi-horizontal air motions and by vertical and horizontal turbulent mixing processes. Simultaneously, the exchange of air between the stratosphere and the troposphere results in a net transfer of radioactive debris out of the stratosphere. The radioactive particulates which enter the troposphere are rapidly mixed in this turbulent part of the atmosphere, and are brought down into the lower troposphere by the mixing process. As a result of this dilution, the tropospheric levels of radioactivity remain low compared with those of the stratosphere, and the mass exchange results in a net depletion of the stratospheric reservoir. However, any increase in the tropospheric inventory associated with the depletion of the stratospheric reservoir appears to be transitory. The stratospheric fallout reaches the earth, largely through the scavenging action of precipitation, within a few months or even weeks after the particles enter the troposphere.

Gravitational Settling Rate

The terminal velocity of a small sphere falling through a viscous medium, such as air, is given by the modified form of Stokes' Law,

$$V = \frac{g}{18\,\eta}\, d^2 \,(e - e_a)\, C \qquad (1)$$

where v is the terminal velocity relative to the air, e is the density of the sphere, e_a is the density of the medium (e.g., air), g is the acceleration of gravity, d is the diameter of the sphere, and η is the coefficient of molecular viscosity of the medium. At this velocity the viscous drag just balances the weight of the sphere, and the latter is falling at constant speed.

The factor C, known as Cunningham's correction, is given by

$$C = 1 + k\frac{\lambda}{d} \qquad (2)$$

where k is a constant, for which the value 0.8 is widely accepted, and λ is the mean free path of the medium.

The density of air may be neglected in (1) for the calculation of the settling speed of natural or bomb produced particles. The densities of these particles are of the order of 1 gm cm^{-3} or more (specific gravity equal to or greater than one). The molecular viscosity, η, of air, which depends almost entirely on the temperature alone, is approximately 1.6×10^{-4} gm cm^{-1} sec^{-1} in the lower stratosphere, and varies only slightly with altitude. The mean free path, λ, of air, however, is strongly dependent on altitude, increasing from about 0.3×10^{-4} to 2.2×10^{-4} cm between 40 and 80 thousand feet above sea level. An average value of 1μ for λ may be adopted for the lower stratosphere for the computation of the settling velocity of stratospheric particles. Table 14 shows the computed gravitational settling (terminal) velocities for particle

diameters between 10 and 0.001 microns in kilometers per year for particles with specific gravity equal to one. (For heavier particles, multiply these values by the particle's specific gravity.)

Table 14. Terminal velocity of a sphere of specific gravity equal to one in the lower stratosphere as a function of particle diameter.

Diameter		Terminal Velocity	
10	microns	100	km/year
1		2	
0.1		0.1	
0.01		0.01	
0.001		0.001	

It is clear from the table above that the sub-micron particles to which most of the stratospheric radioactivity is attached fall so slowly that, except for particles very close to the tropopause, gravitational settling out of radioactive stratospheric particulates is of negligible importance.

Horizontal Transport and Wind Shear

The airborne particles are transported horizontally by the large scale, meandering, **circumpolar** circulation in the upper atmosphere. Although this circulation is predominantly zonal (west - east), the wave-like meanders in the wind field produce a significant meridional (north - south) transport of the radioactive debris. However, the variation of the atmospheric circulation in space and time makes the precise prediction of the trajectory of a radioactive cloud extremely unreliable beyond a few days.

Because of the effects of turbulent mixing, due to eddies in the wind field, and vertical wind shear, i.e., the variation of wind with altitude, the radioactive cloud soon ceases to be an identifiable entity. The effect of vertical wind shear is to cause the cloud to tilt with time so that it becomes elongated along the direction of the wind. Within a few months the radioactive cloud is extended as a "streak" around the earth roughly parallel to the latitude circles. Subsequently the cloud streak expands laterally and vertically, principally through the mechanism of turbulent mixing. Since the HASP observations are confined to a vertical-meridional plane, they may be used to evaluate the intensity of the mixing processes in this plane, i.e., in the vertical and meridional directions, after the zonal cloud streak has been established. On the other hand, the data shed relatively little light on the transport of the cloud along the wind (except for young clouds) or on the intensity of mixing along the wind.

Turbulent Mixing in the Stratosphere

We may immediately dispense with ordinary molecular diffusion as a significant exchange process in the atmosphere, for it is well known that this process is negligible compared with turbulent mixing. The latter process is sometimes referred to as turbulent diffusion, eddy diffusion, or Austausch.

The laws governing turbulent mixing in the atmosphere are still not well established, and most of the work that has been done on the problem has been

carried out in the air near the ground. The results of these experiments are not directly applicable to the free atmosphere.

While considerable progress has been made in recent years in the development of statistical theories of turbulence, this theoretical work is also not easily or directly applicable to the stratospheric problem. On the other hand, it is possible to gain some insight into the nature and magnitude of the turbulent exchange process through the use of the older, and less satisfactory, simple mixing theory which is derived by analogy with the molecular diffusion theory.

In the simple mixing theory the turbulent exchange process is considered to be analogous to ordinary molecular diffusion, so that the heat conduction or diffusion equation is applicable. Thus, if Q represents the concentration of radioactivity, the differential equation governing the exchange process is,

$$\frac{\partial Q}{\partial t} = \frac{\partial}{\partial z}\left(K_z \frac{\partial Q}{\partial z}\right) + \frac{\partial}{\partial y}\left(K_y \frac{\partial Q}{\partial y}\right) \qquad (3)$$

where t is time, and K_z and K_y are coefficient of turbulent mixing (or "effective" diffusion coefficients) for the vertical (z) and meridional (y) directions. The effect of large scale transport is not included in (3), so that $\frac{\partial Q}{\partial t}$ is the local rate of change of activity following the air motion. Also, turbulent mixing along the wind direction is ignored on the assumption that a homogeneous zonal cloud streak has been established.

The solution of the problem is greatly simplified by assuming that each mixing coefficient is a constant, so that we may write:

$$\frac{\partial Q}{\partial t} = K_z \frac{\partial^2 Q}{\partial z^2} + K_y \frac{\partial^2 Q}{\partial y^2} \qquad (4)$$

This is a crude, and generally unacceptable, assumption, for a major part of the research efforts in atmospheric turbulence is devoted to determining how K varies with meteorological parameters, scale, etc. However, it will be adopted for the purpose of making some gross calculations of mixing rates.

If K is the same for all directions the mixing is said to be isotropic. However, atmospheric mixing is not isotropic, and it is necessary to employ different values for the vertical and horizontal exchange coefficients.

For isotropic diffusion from a point source, to which the bomb may be likened as a first approximation, the distribution of activity in the cloud is given by the solution to the diffusion equation:

$$Q(r,t) = Q(o,t)\, e^{\frac{-r^2}{4Kt}}, \qquad (5)$$

where $Q(r,t)$ is the activity at time t after injection at a distance r from the center of the cloud, $Q(o,t)$ is the activity at the center of the cloud, e is the base of natural logarithms, 2.718....., and K is the mixing coefficient. The activity profile of the cloud is thus represented by a normal probability distribution (error function) with central (maximum) value equal to $Q(o,t)$ and a standard

deviation S, given by:

$$S = \sqrt{2Kt} \qquad (6)$$

This "Gaussion" cloud profile is shown in Figure 39. The "width" of the cloud may be expressed in terms of the standard deviation or in terms of the half-width of the cloud. The latter is defined here as the distance from the center of the cloud to the point where the activity is half the central value. (The half-width is sometimes defined as twice this distance.) For a normal distribution the half-width, h, is given by:

$$h = S\sqrt{2 \log_e 2} = 1.18\, S = 1.67\sqrt{Kt} \qquad (7)$$

Lateral Spreading of the Radioactive Cloud

The simple mixing theory above may be used to calculate the rate at which the radioactioactive cloud streak expands in the north - south direction in the stratosphere. The meridional half-width, h_y, is given by:

$$h_y = 1.67\sqrt{K_y t} \qquad (8)$$

To compute, and predict, the growth rate, it is necessary to determine the magnitude of the meridional mixing coefficient, K_y, in the stratosphere. Information about K_y from meteorological sources is generally meager, and it is likely that the best estimate of K_y can be obtained from the HASP data.

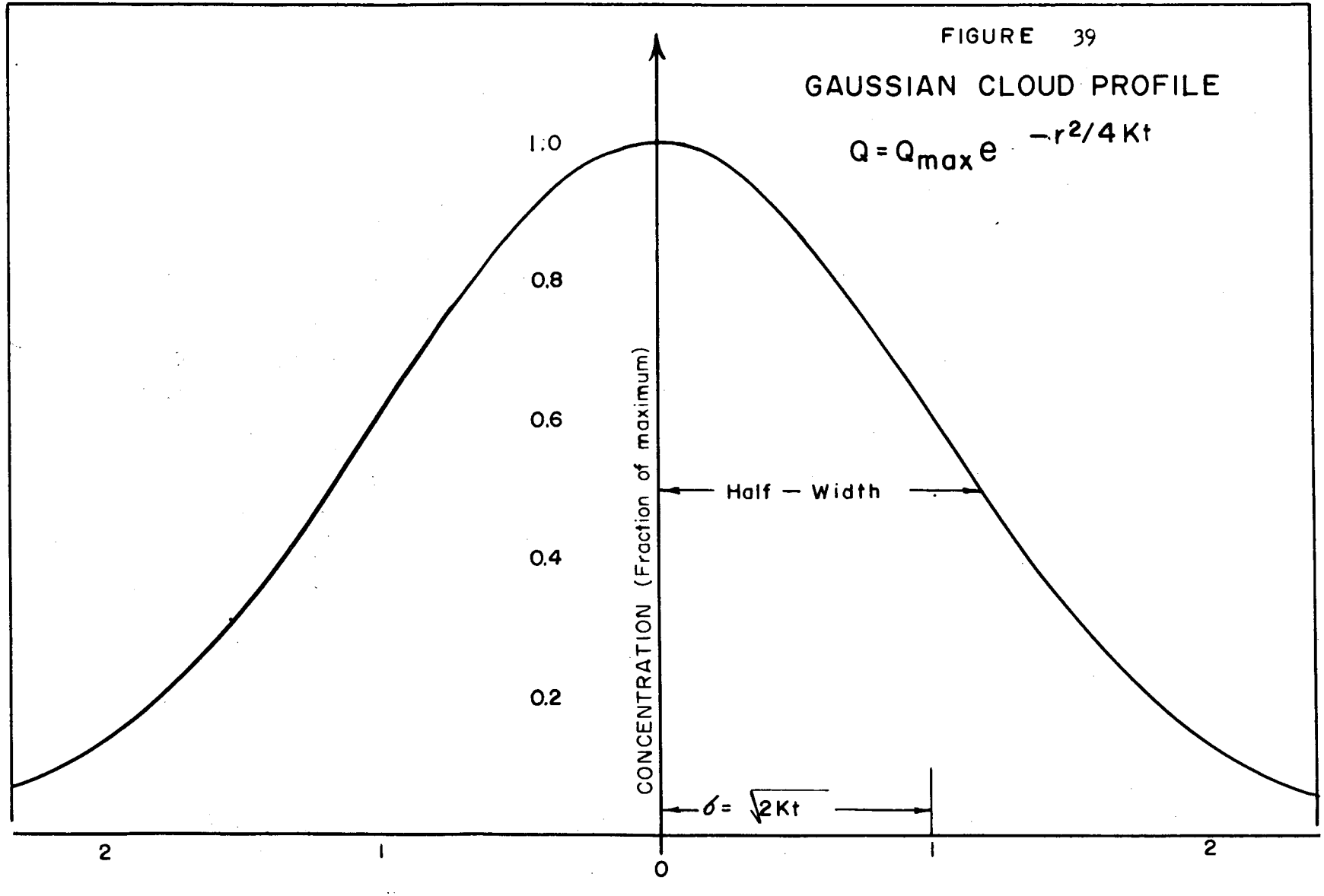


FIGURE 39
GAUSSIAN CLOUD PROFILE
$Q = Q_{max} e^{-r^2/4Kt}$
1.0
0.8
0.6
0.4
0.2
CONCENTRATION (Fraction of maximum)
Half — Width
$\sigma = \sqrt{2Kt}$
2
1
0
1
2
DISTANCE FROM THE CENTER OF THE CLOUD (r/σ)

From (8) it follows that:

$$K_y = 0.36\ h_y^2/t \qquad (9)$$

Observations of the spreading of young clouds of strontium-90 and more recent measurements of tungsten-185 indicate that the half-width of a radioactive cloud after a period of the order of a month is of the order of 10 degrees in latitude (approximately 10^8cm). Thus, the order of magnitude of the effective mixing coefficient is $10^9 cm^2\ sec^{-1}$.

The meridional spreading of the radioactive cloud in the stratosphere has been computed from the simple mixing theory for K_y equal to $10^9\ cm^2\ sec^{-1}$, and also for values of 10^8 and $10^{10}\ cm^2\ sec^{-1}$. The results of the calculation are shown in Figure 40 in which the horizontal half-width is shown as a function of time. According to the simple theory the cloud spreads rapidly at the beginning, then more slowly after it has aged. For K equal to 10^9, the half-width increases to 40 degrees of latitude after 2 years and to 55 degrees of latitude after 4 years. However, the calculation is quite sensitive to the value of K selected. The points plotted on the diagram are observations of half-widths of identifiable clouds of strontium-90 and tungsten-185 for which the shot dates were known. These data suggest a value of K_y between 5×10^8 and 5×10^9 $cm^2\ sec^{-1}$.

Vertical Mixing of the Radioactive Cloud in the Stratosphere

To calculate the vertical spreading of the cloud of radioactive debris in the

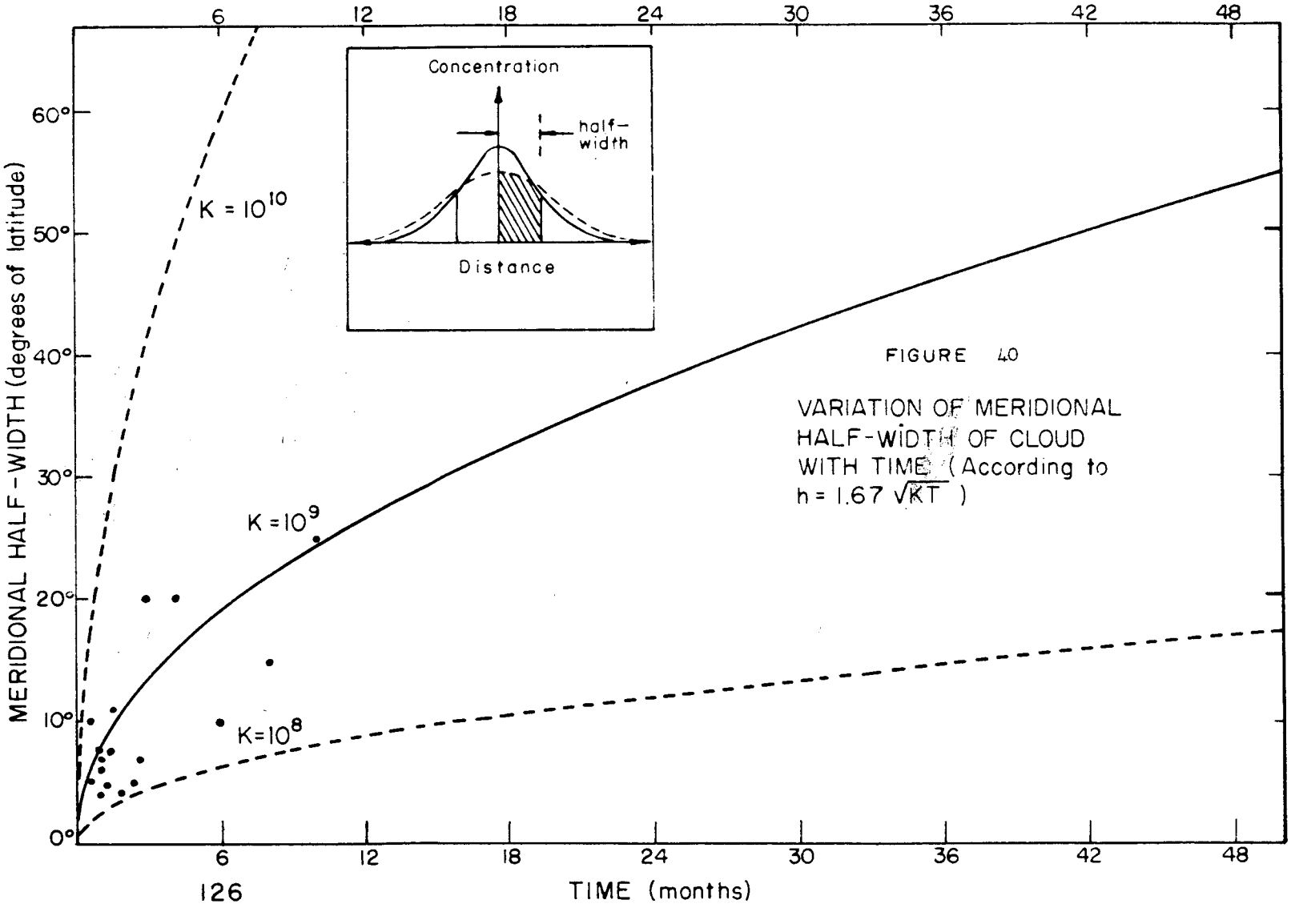


FIGURE 40

VARIATION OF MERIDIONAL HALF-WIDTH OF CLOUD WITH TIME (According to $h = 1.67\sqrt{KT}$)

stratosphere, the simple mixing theory is again employed to compute the vertical half-width, h_z, of the cloud from:

$$h_z = 1.67 \sqrt{K_z t} \qquad (10)$$

The vertical mixing coefficient, K_z, is known to be dependent on the lapse rate of temperature in the stratosphere. The characteristic inversion of the tropical stratosphere (negative lapse rate) is associated with strong stability and weak vertical mixing. On the other hand, the greater lapse rate (especially in winter) in the polar stratosphere suggests the possibility of stronger vertical mixing there than in lower latitudes. Thus it is to be expected that K_z should be greater in high latitudes in the stratosphere than in low latitudes.

From observations of tungsten-185 in the tropical stratosphere the vertical half-width of the cloud appears to be about 2 to 3 km after about 2 months, suggesting a value of K_z of the order $10^3 cm^2\ sec^{-1}$ for that region of the atmosphere. A similar calculation based on the more questionable identification of Soviet debris in high latitudes as well as on the observed behavior of tungsten-185 after it had drifted into the polar stratosphere indicates a value of h_z about twice as large in the same time. The value of K_z for the polar stratosphere is estimated to be of the order of $10^4 cm^2\ sec^{-1}$.

The calculation of the vertical spreading of the cloud in the polar and tropical stratospheres is illustrated in Figure 41. After 2 years the tropical cloud

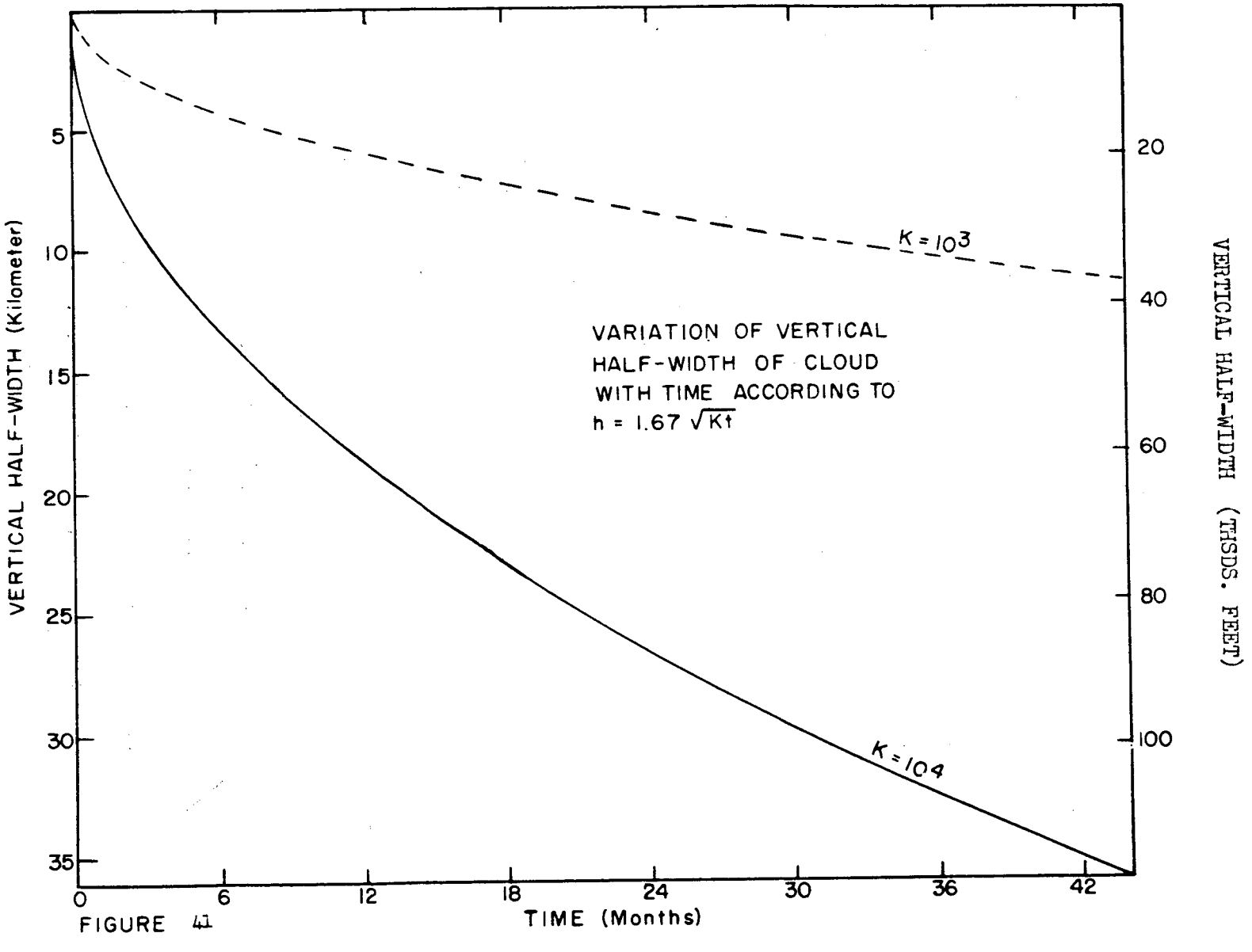

VERTICAL HALF-WIDTH (Kilometer)
VERTICAL HALF-WIDTH (THSDS. FEET)
TIME (Months)
K = 10³
K = 10⁴
VARIATION OF VERTICAL HALF-WIDTH OF CLOUD WITH TIME ACCORDING TO h = 1.67 √Kt
0
6
12
18
24
30
36
42
5
10
15
20
25
30
35
20
40
60
80
100

FIGURE 41

will have reached a half-width of 8.4 kilometers (27,500 feet) while the polar cloud will have grown to 26.5 kilometers (87,000 feet). Of course, since the stratosphere is bounded this process obviously cannot be extended in time without limit. As shown below, the vertical spreading of the cloud must result in removal of some of the debris through the tropopause.

It must be emphasized that the calculations above are based on the assumption that the Gaussian (normal) profiles are maintained in the vertical plane, whereas in nature this is not the case. The irregular motions of the atmosphere distort the cloud profiles, and they are clearly not Gaussian. The calculations from diffusion theory can only provide rough estimates of the behavior of the cloud.

Transfer Through the Tropopause

A basic meteorological problem of fallout is the mechanism of transfer of airborne radioactive dust from the stratosphere to the troposphere. Several modes of egress from the stratosphere have been suggested, and it seems reasonable that all of them can contribute in some measure to the stratospheric fallout. Unfortunately, precise quantitative information on the relative magnitudes of these various processes is lacking.

The tropopause is neither a substantial surface nor a barrier to air motion. It is merely a more-or-less thin layer of air above which vertical mixing is

relatively weak, and below which vertical mixing is relatively strong. It is also probably a level of maximum horizontal velocity divergence and minimum vertical motion more often than not. Thus vertical exchange of air and downward transport of radioactive debris is slower at the tropopause than below it, but not appreciably slower than in the stratosphere.

As a result of radiative processes the tropopause may disappear at one level and reform at a higher (or lower) level. (This is indicated by the fact that the potential temperature at the tropopause is not necessarily conserved from one day to the next.) In this way formerly stratospheric air may be "transferred" into the troposphere without any vertical motion at all. However, it must be stressed that this can only "peel away" the radioactivity in the lower stratosphere. Vertical motions (steady or turbulent) in the stratosphere are needed to transport the debris from the levels of high concentration down to the vicinity of the tropopause.

The transfer of debris from the stratosphere to the troposphere by air motions may be accomplished by (a) large scale, organized vertical circulation (subsidence) in the stratosphere and through the tropopause, (b) turbulent vertical exchange in the stratosphere and through the tropopause, (c) large scale, organized, meandering horizontal circulation through the tropopause gap, or (d) turbulent horizontal exchange through the tropopause gap. There is as yet no direct evidence of the organized meridional circulation cell required for

the first of these mechanisms, and there are some meteorological arguments against the existence of such a circulation. The existence of the other transfer mechanisms can hardly be doubted, but there still remains the problem of calculating the magnitudes of these processes. Some rough calculations of the intensity of the turbulent transfer process are described below.

The turbulent vertical transfer of radioactive debris across the tropopause may be calculated on the assumption that the vertical profile of the activity concentration is similar to the form shown schematically in Figure 42. The dashed curve in the figure represents the model vertical distribution of activity per unit mass of air (e.g., dpm Sr^{90}/1000 scf). The scale along the abscissa is provided to illustrate typical current values of the activity, but the absolute numbers are not relevant to the calculation. The solid curve illustrates schematically the profile of activity per unit volume of air. (Because of the decrease of air density with altitude, the peak in the volume concentration is found below the peak in the mass concentration.) For the calculation a normal (Gaussian) distribution of activity per unit volume will be assumed along the vertical. The tropopause is shown in the figure at an altitude Z_T below the level of maximum volume concentration.

The transfer of radioactive debris out of the stratosphere is calculated on the assumption that the vertical activity profile remains normal, but spreads out with time about the level of maximum activity according to the simple

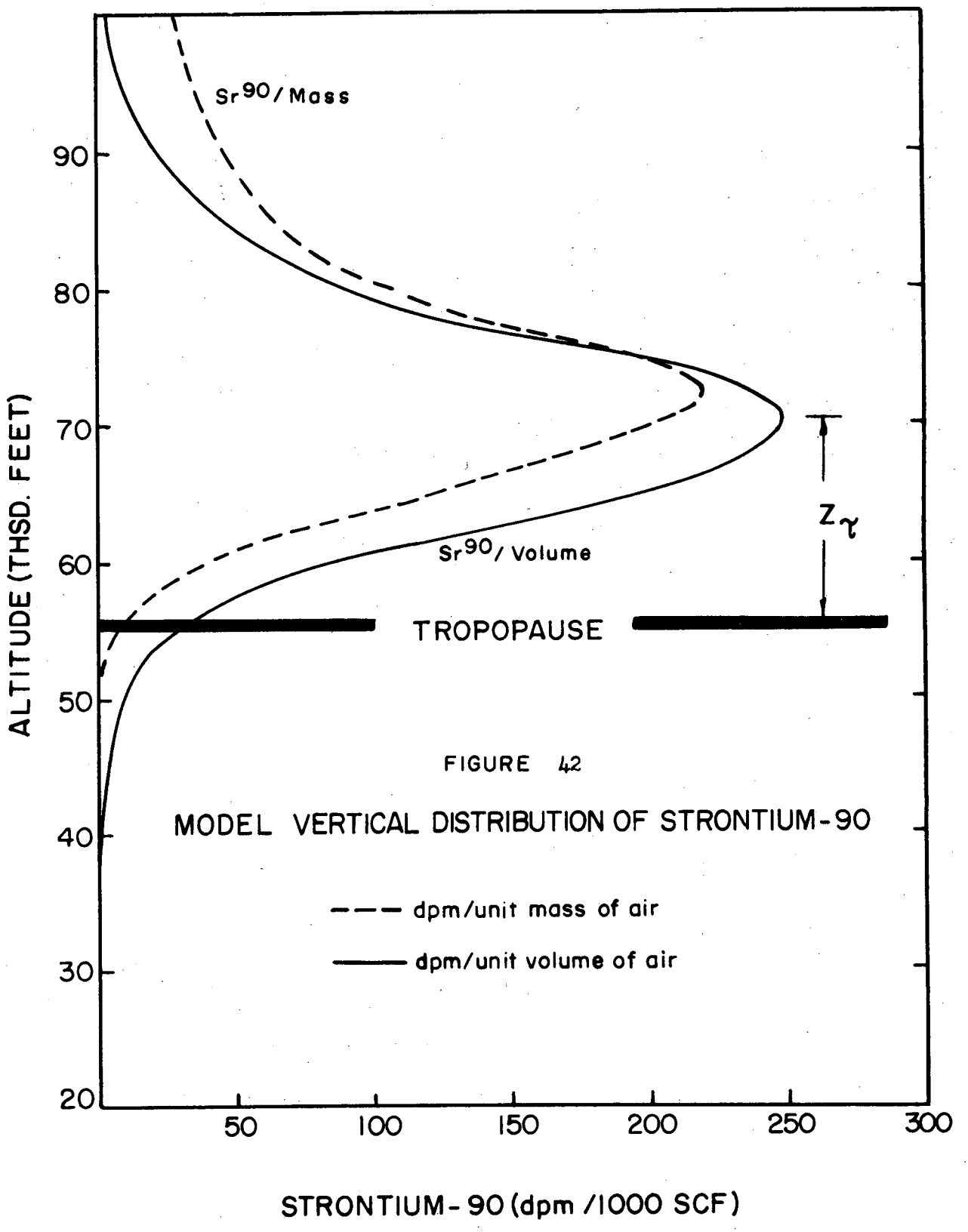


FIGURE 42

MODEL VERTICAL DISTRIBUTION OF STRONTIUM-90

turbulent diffusion theory. As the peak concentration diminishes, the standard deviation of the distribution increases according to (6). This process places an increasing fraction of the total activity below the tropopause. It is this fraction of the total that is considered to be the cumulative stratospheric fallout. (One of the serious shortcomings of this simple model is the inherent assumption that the activity increases with time above the level of the maximum as well as below, as the distribution becomes "flatter." Another obvious deficiency is the assumption that the distribution follows the normal law below the tropopause, whereas in fact the level of activity there remains low and nearly constant due to the turbulent mixing in the troposphere.)

The fraction of the total activity below the tropopause may be found from tables of the normal probability integral for values of the standard deviate corresponding to $Z_T / \sqrt{2K_z t}$. The results of the calculations are shown in Figure 43. Curves A and B show the computed cumulative fallout as a function of time, expressed in percent of the initial total atmospheric burden, for the tropical and polar regions respectively. For the tropics a mixing coefficient of 4×10^3 cm^2 sec^{-1} has been used, and the level of maximum activity is assumed to be 5.2 km above the tropopause. In the polar regions the values 2×10^4 cm^2 sec^{-1} and 8 km were assumed for these quantities. The fallout curves show initially rapid removal followed by a diminution of the fallout rate. After one year 15 percent of the debris will have been transferred out of

the tropical troposphere, and 24 percent will have fallen from the polar stratosphere. After two years, however, the cumulative fallout for the tropics and polar regions respectively will have risen to only 23 percent and 31 percent respectively. Since this rate is considerably smaller than the observed fallout rate (discussed below), it may be concluded that direct vertical transfer through the tropopause by turbulent mixing appears to be too small to account for the stratospheric fallout.

A Note on Residence Time

It has become customary to discuss the removal of radioactive material from the stratosphere in terms of residence time, and it may be of interest to discuss the computations of fallout from this viewpoint.

The differential equation of "first order kinetics" is:

$$\frac{dq}{dt} = -\lambda q \qquad (11)$$

where q represents the total stratospheric burden of a nuclide (e.g., strontium-90) at time t, and λ is a constant known as the "removal rate." Equation (11) assumes that the rate of depletion of the reservoir is proportional to the magnitude of the reservoir. A corollary of this assumption is the assumption that a constant fraction of the existing reservoir is removed per unit time.

The integral of (11) is:

$$q = q_o \, e^{-\lambda t} \qquad (12)$$

where q_o is the initial total burden and e is the base of natural logarithms, 2.718..... From (12)

$$q = \frac{q_o}{e} \qquad \text{at} \qquad t = \frac{1}{\lambda} = \tau \qquad (13)$$

There τ is the time required for the total burden to drop to $1/e$ of the initial burden. This is known as the "mean residence time," or sometimes simply as "residence time."

The "residence half-time," T, is the time required for the total burden to decrease to one-half the initial burden.

Thus:

$$e^{-\lambda T} = 1/2$$

$$\text{or} \qquad T = (\log_e 2)\tau = (0.693)\tau \qquad (14)$$

Thus, if the residence half-time is one year, the "mean residence time" is 1.44 years, and the "removal rate" is 69% per year. This last number is misleading because of the choice of units, for in fact after one year only 50% of the stratospheric inventory will have been removed in this instance. If the "removal rate" is 50% per year, the mean resi dence time is 2 years, and the residence half-time is 1.39 years.

The cumulative fallout from the stratosphere computed on the assumption of constant residence time is shown in Figure 43 for comparative purposes. Curves C, D, and E are drawn for residence half-times of 4, 5, and 1 year respectively. Clearly constant residence time fallout does not resemble "Gaussian" fallout, except perhaps for short periods. Thus the turbulent vertical transfer in the polar regions appears to correspond to an average residence half-time of 1-4 years in the first year, whereas that of the tropics appears to be consistent with an average half-time of 4-5 years in the first year. However, the turbulent transfer does not correspond to a constant residence time over longer periods.

It is observed (see discussion below) that half the stratospheric debris falls out in less than the first year after input, so that the effective residence half-time for the first year is less than one year. This again indicates that direct turbulent transfer through the tropopause does not appear to account for the fallout rate. There is also observational evidence (see below) that the concept of a constant residence time is untenable.

Horizontal Transfer Through the Tropopause Gap

It is estimated that approximately 40% of the strontium-90 in the atmosphere (in 1959) resides in the tropical stratosphere, between latitudes 30N and 30S and above the 100 millibar (53,000 foot) level. The remainder is found in the polar stratosphere. Of this 60% which lies in the polar stratosphere, a

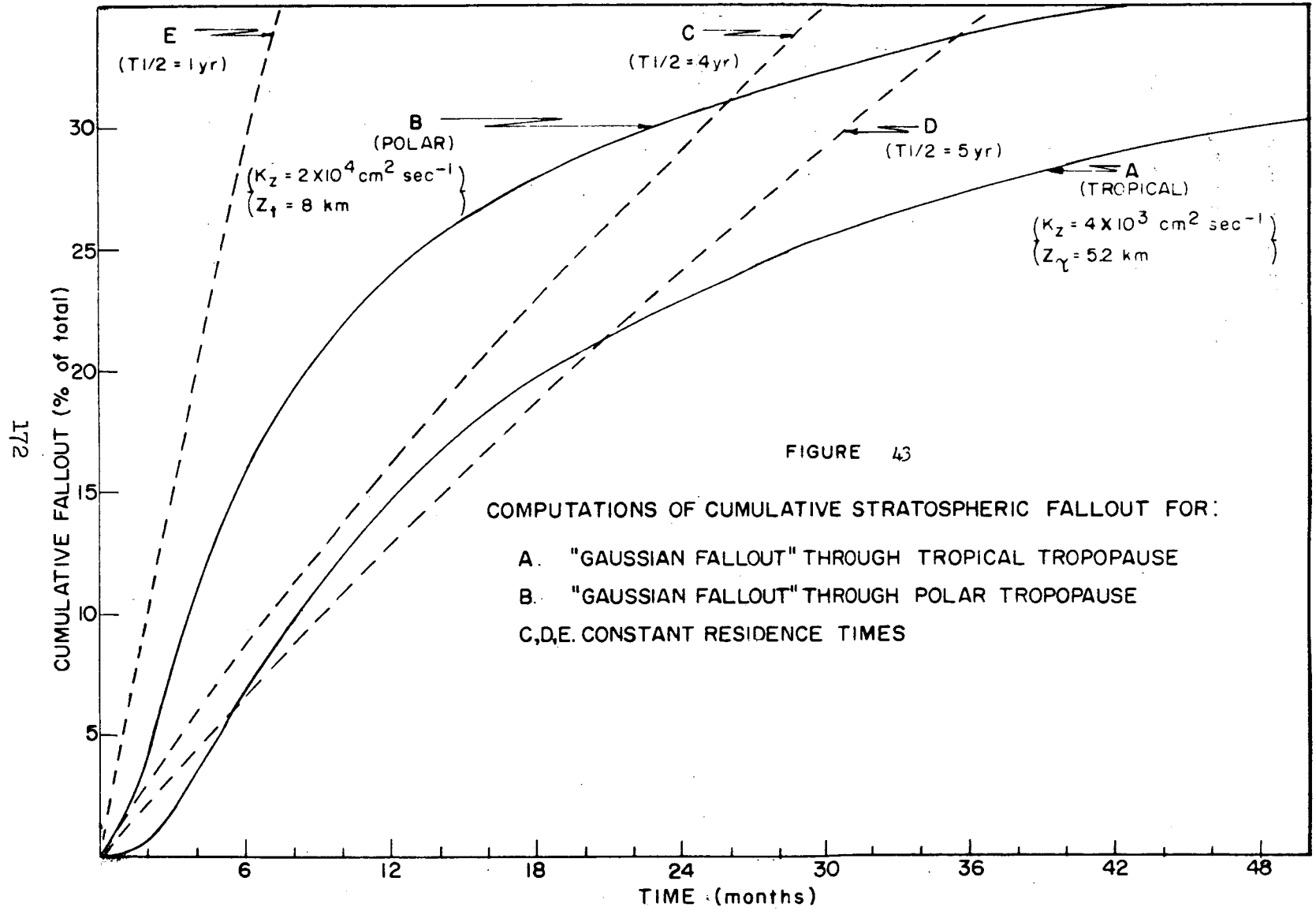


FIGURE 43

COMPUTATIONS OF CUMULATIVE STRATOSPHERIC FALLOUT FOR:

A. "GAUSSIAN FALLOUT" THROUGH TROPICAL TROPOPAUSE

B. "GAUSSIAN FALLOUT" THROUGH POLAR TROPOPAUSE

C,D,E. CONSTANT RESIDENCE TIMES

significant fraction is found in the layer containing the tropopause gap, i.e., the layer between about 100 and 200 millibars in which about 30% of the mass of the stratosphere is included. Roughly 25% of the total strontium-90 in the Northern Hemisphere lies in this gap layer. The state of affairs is indicated schematically in Figure 44, in which the idealized tropopause structure is shown on a cross-section with pressure as ordinate and latitude on a cosine scale as abscissa, with the pole at the center. (Equal areas on this diagram correspond to equal masses of air.) The diagram represents a vertical cross-section through an axially symmetrical hemisphere. The tropopauses are indicated by heavy lines, and the gap layer is shown by cross-hatching.

The gap layer may be thought of as a reservoir which is supplied by turbulent and organized transport of debris from the tropical stratosphere (as shown by the arrows in Figure 44), as well as by direct injections (e.g., Soviet tests). The debris is removed from the gap layer by organized and turbulent downward transport through the tropopause, and by organized and turbulent horizontal transport through the gap.

Computations of the transport of debris through the gap cannot be made with any confidence because the mixing coefficients for this region are unknown. It is known that the gap, particularly in winter, contains the jet stream, and that this is a region of strong turbulence. Therefore, it is to be expected that the gap is a region of active lateral, as well as vertical, mixing. A horizontal

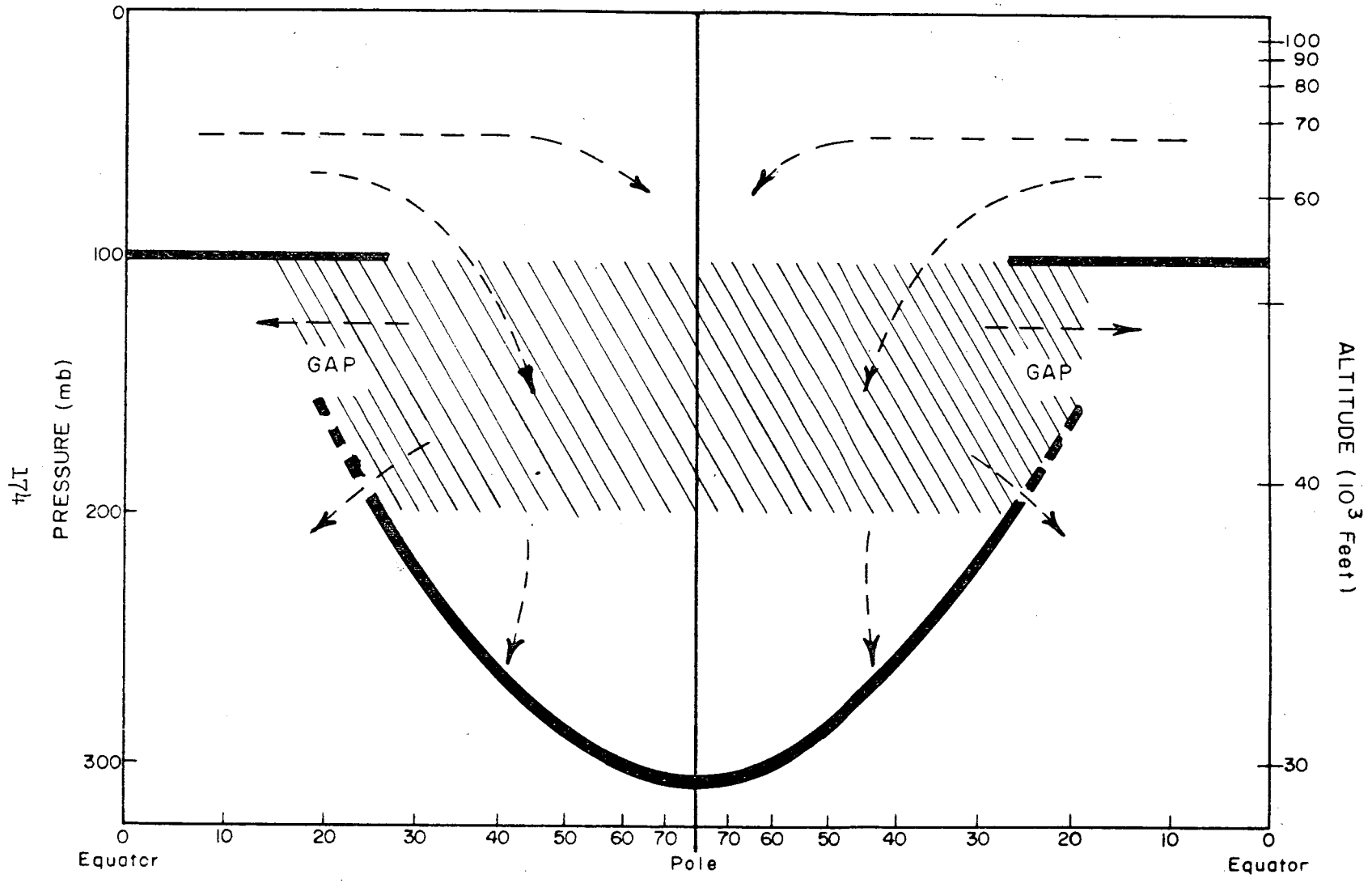


FIGURE 44 SCHEMATIC MODEL OF TROPOPAUSE STRUCTURE CENTERED ON THE POLE

mixing coefficient in the gap region of 10^{10} and even 10^{11} cm^2 sec^{-1} does not appear to be unreasonable. However, the problem is further complicated by the fact that the mixing coefficient is undoubtedly not constant.

If the mixing coefficient were known it would be necessary to assume some simple form for the lateral distribution of activity in the gap in order to calculate the removal rate. The most convenient assumption is of an axially symmetric, two-dimensional normal distribution with the maximum at the North Pole. The problem then is reduced to the calculation of the fraction of the total activity which lies outside the circle corresponding to the latitude of the gap. For a normal circular distribution this fraction is equal to:

$$e^{-L^2/2S^2} \qquad (15)$$

where L is the distance from the pole to the gap, and the standard deviation, S, is computed from (6). For example, if we assume a distance of 6000 km (approximately 55 degrees of latitude) for L and let K_y equal 10^{10} cm^2 sec^{-1} in the gap region, we find that after one year 75% of the debris in the gap layer has been removed. If we place the gap at 45^{o}N, so that L is equal to 5000 km, and let K_y equal 10^{11} cm^2 sec^{-1}, the annual depletion of the gap layer increases to 98%. To calculate the rate at which turbulent transfer through the gap depletes the <u>total</u> stratospheric reservoir, these numbers must be multiplied by about 25%, i.e., the fraction of the total stratospheric burden of

strontium-90 that is contained in the gap layer. However, this simple model underestimates the depletion rate because it does not take account of the replenishment of the gap reservoir (and re-establishment of the profile) by transfer from the rest of the stratosphere.

Although the computations are extremely crude, they indicate that turbulent transfer through the gap, together with direct turbulent transfer through the tropopause, may almost account for the observed fallout rate. The additional factor of horizontal transfer through the gap by the large scale, meandering streamflow also undoubtedly contributes significantly to the removal of debris from the stratosphere, so that the fast fallout is not unreasonable. However, this process is even less amenable to direct calculation than the turbulent process.

We must conclude this section with the caution that the calculations of meteorological transfer processes are largely speculative, and can give little more than some physical insight into the problem. The direct calculation of the removal rate from inventory measurements, supported by material balance checks, is still the only reliable method. This is discussed below.

Calculation of Removal Rate from Strontium-90 Inventories

The material balance calculation should provide a check on the estimates of removal rate. Thus the world-wide fallout (exclusive of local and intermediate fallout) should equal the difference between the estimated stratospheric input

and the stratospheric inventory. It is estimated that the total stratospheric input from the beginning of testing of megaton weapons through the fall of 1958 was 5 MC of strontium-90, exclusive of very high altitude shots. The 1959 stratospheric inventory is estimated to be between 0.8 and 1.1 MC. Thus, the cumulative world-wide fallout in mid-1959, rounded off to the nearest digit, should be 4 MC of strontium-90. This is in good agreement with the world fallout estimates of Walton[23] and Machta[24] (see Chapter X).

The calculations above are not intended to imply a constant residence time or removal rate. Indeed, it has already been suggested in the discussion of the turbulent transport process that the removal rate may decrease with time. In order to study this problem an effort has been made to compute the removal rate during the period of the test moratorium from overlapping short period strontium-90 inventories. This procedure has the advantage of being free of any assumptions regarding the magnitude of the stratospheric input. On the other hand, it suffers from the defects of small sample size, incomplete spatial distribution of the samples, and possible non-representativeness of the cross-sections.

Stratospheric strontium-90 inventories were computed for overlapping four-month periods as shown in Table 15. Between the first two intervals (November 1958 - February 1959) (January - April 1959) the inventory decreased about 17 percent, corresponding to an effective residence half-time of 8 months. However,

in the next interval the inventory declined only 11 percent, corresponding to a residence half-time of 12 months. In the last interval the removal amounted to 3 percent with an effective half-time of 46 months. The median dates of the intervals are, of course, two months apart.

Table 15. Stratospheric Strontium-90 Inventories for Successive Four-Month Periods.

Period	Sr^{90} Inventory (MC)	Percent Removed	Effective Residence Half-Time (Months
November 1958-February 1959	1.00		
		17	8
January-April 1959	0.83		
		11	12
March-June 1959	0.74		
		3	46
May-August 1959	0.72		

Although the short period inventories are far from reliable, they do appear to support the suggestion that the removal rate decreases with time. The newly injected debris apparently falls out rather quickly (more than half of it comes from the stratosphere in a half-year, after which the fallout rate diminishes with time. However, one cannot attach much quantitative significance to the numbers in Table 15. For example, the tabulated value for the first **interval** is probably too low as a result of inadequate sampling in high latitudes of the Northern Hemisphere.).

An additional factor which must be considered in the interpretation of Table 15 is the seasonal variation of fallout. The fallout data for the Northern Hemisphere, especially in 1959, clearly show a spring maximum in the rate of transfer of debris from the stratosphere to the troposphere. An early spring (or late winter) maximum could be explained on meteorological grounds as a consequence of the enhancement of turbulent mass exchange between the stratosphere and the troposphere in late winter. The time intervals used in Table 15 are, of necessity, too coarse to expose a seasonal effect. However, it is quite possible that part of the decline in the removal rate can be attributed to the seasonal factor.

Meteorological Analysis of Tungsten-185 Data

The injection of tungsten-185 into the stratosphere during the HARDTACK series of 1958 provided a unique tracer for the study of the dispersion and fallout of equatorial debris. It is estimated that 94.7 megacuries of tungsten-185 were injected into the stratosphere during the entire series. The amount of tungsten-185 present in the stratosphere on 15 August 1958 was computed by correcting the estimated stratospheric tungsten-185 yields of the individual weapons for radioactive decay at a half-life of 74 days. If no adjustment is made for fallout, the stratospheric input of tungsten-185 on 15 August 1958 is found to be 58 MC. Allowing for a residence half-time of one year in the stratosphere, and

assigning a weighted mean date of injection of 1 July 1958 for tungsten-185, we obtain a value of 53 MC for the "initial" tungsten-185 input by 15 August 1958. All tungsten-185 data shown are corrected for radioactive decay back to this date.

The tungsten-185 data provide a unique opportunity to calculate the removal rate of debris injected into the equatorial stratosphere. For this computation we require vertical cross-sections of the tungsten-185 distribution. Fortunately most of the tungsten-185 is found in the tropical stratosphere where the sampling was most satisfactory. Consequently, the short-period tungsten-185 inventories are somewhat more reliable than those for strontium-90, and it was deemed feasible to calculate bi-monthly totals for tungsten-185.

The bi-monthly tungsten-185 totals , corrected to 15 August 1958, are shown in Figure 45 for the periods September-October 1958, November-December 1958, and January-February, March-April, May-June, and July-August 1959.

The cross-section for July-August 1959* provides the first reliable evidence that the maximum tungsten-185 activity does not lie above the maximum altitude sampled. The altitude of maximum tungsten-185 activity has clearly lowered in the last two months. This behavior is not consistent with the theory of an organized meridional circulation with an ascending current at the Equator, but rather supports our concept of lateral diffusion from an Equatorial line source.

* not shown here

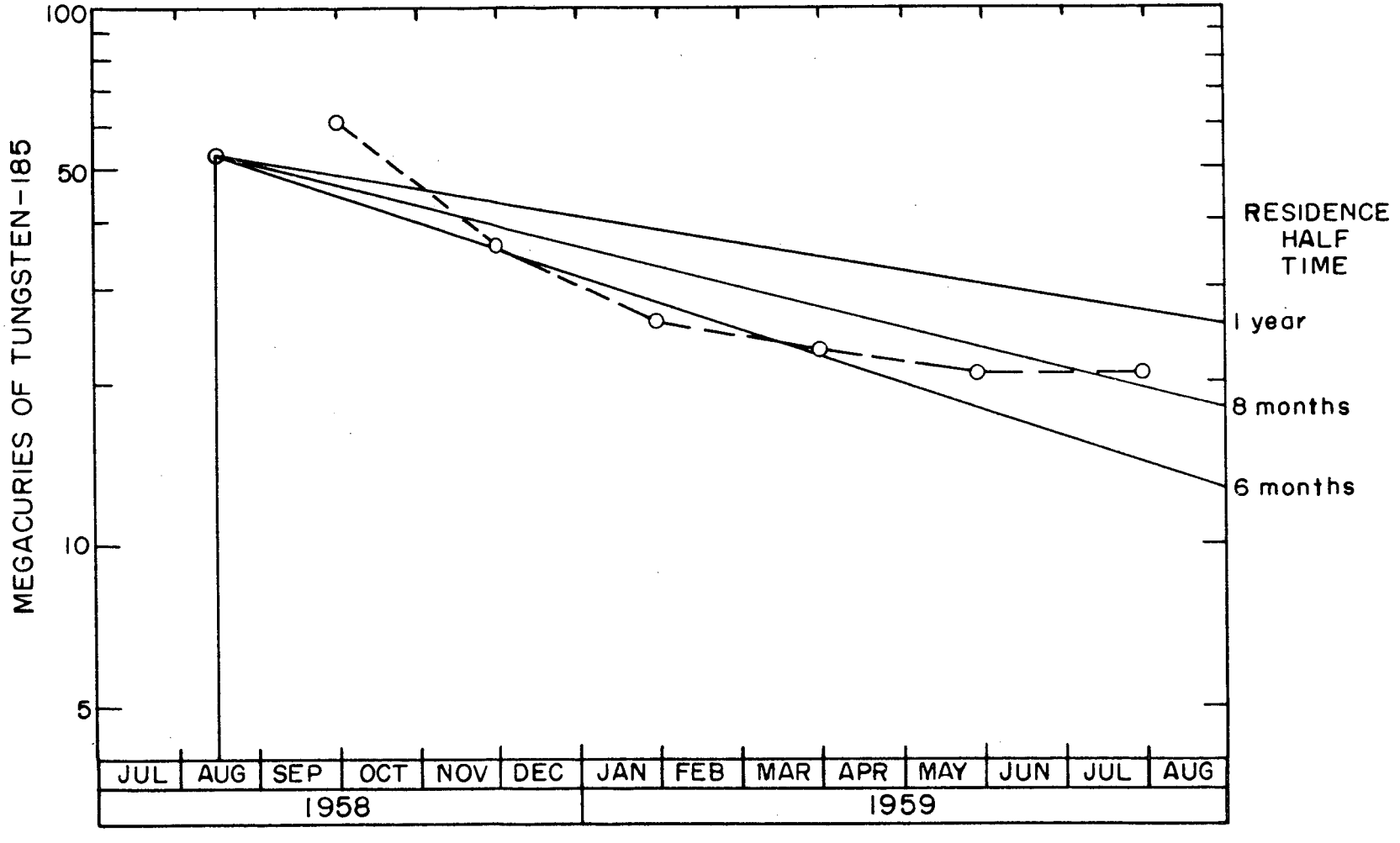


FIGURE 45 BIMONTHLY STRATOSPHERIC BURDENS OF TUNGSTEN-185 WITH CONSTANT RESIDENCE TIME CURVES

The cross-section also shows a general decline in the concentration of tungsten-185 in the stratosphere, with a decrease of about 10 percent in the last two months. The total stratospheric reservoir of tungsten-185 is distributed asymmetrically, with most of the activity in the Northern Hemisphere.

The variation of the tungsten-185 distribution with time in the stratosphere is summarized in Figure 46. In this figure the meridional bi-monthly tungsten-185 profiles at one altitude only are shown for the period November 1958 - August 1959. The profiles have been shifted relative to each other so that their maxima coincide, and the abscissa of the diagram is degrees of latitude North or South of the latitude of the peak. The ordinate is tungsten-185 activity in dpm/scf, corrected for radioactive decay to 15 August 1958.

The profiles show clearly the decline in the tungsten-185 concentrations from the beginning to the end of the period, especially in the zone lying within 20 degrees of latitude of the peak. At the same time, the cloud has spread laterally to the North and South so that the activity on the "wings" of the distribution has actually increased in places, following the initial decline. The major decline in the concentrations of tungsten-185 apparently occurred in the period up to the spring of 1959, after which the cloud exhibited a more stable behavior.

The distribution of tungsten-185 shown in Figure 46 is certainly not precisely Gaussian (e.g., a normal probability distribution); but on the other hand

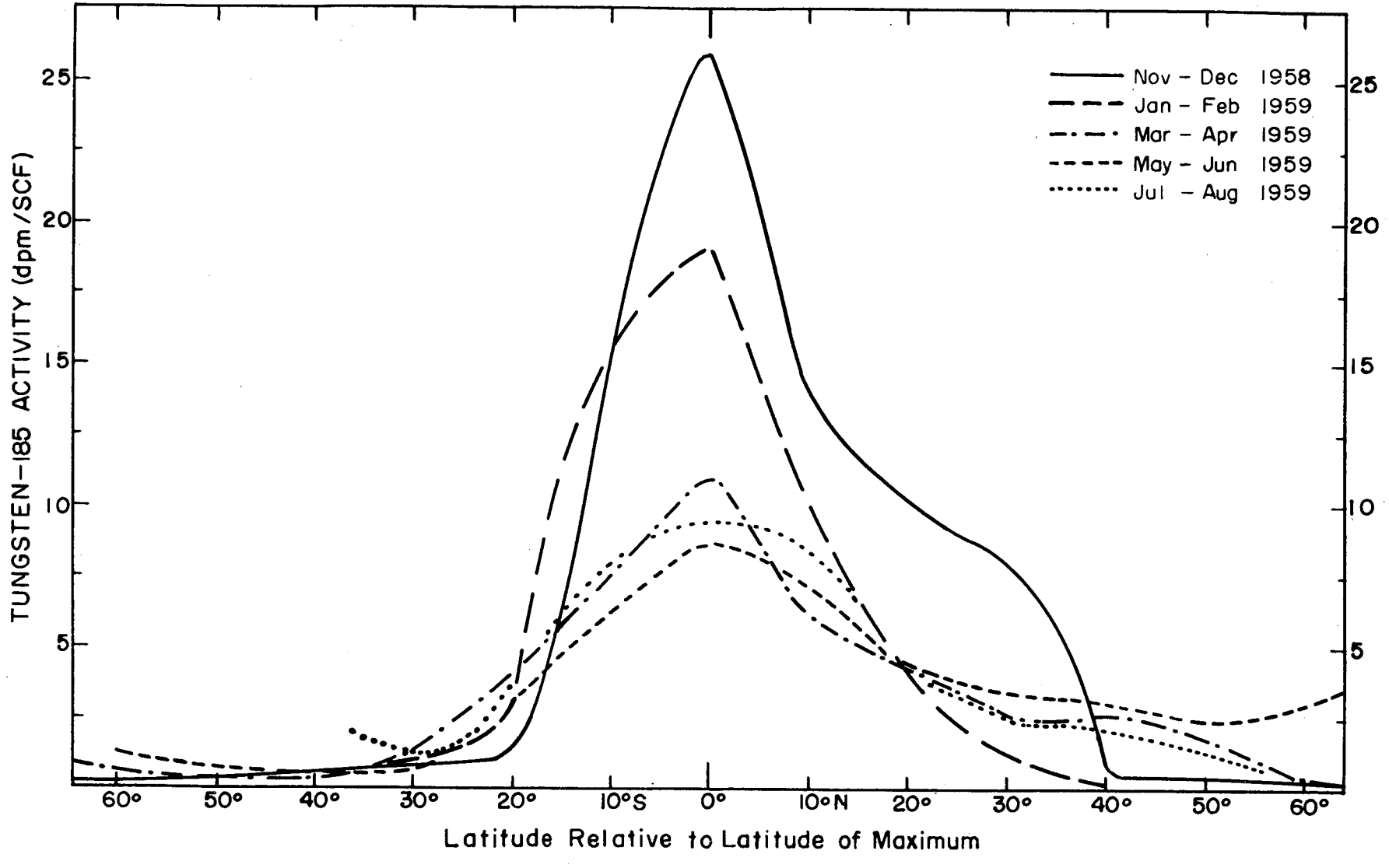


FIGURE 46 BIMONTHLY MERIDIONAL PROFILES OF TUNGSTEN — 185
(dpm/SCF Corrected to 15 August 1958)

it cannot be said that it departs radically from such a distribution, particularly toward the end of the period. If the form of the distribution is approximately Gaussian, it is possible to estimate the lateral mixing coefficient in the lower tropical stratosphere from the observed time-variation of the half-width of the tungsten cloud. According to equation (9), if the lateral mixing follows the law of simple diffusion with a constant mixing coefficient, the square of the half-width should be directly proportional to time. In Figure 47, the half-width, h, of the tungsten-185 cloud is plotted on a scale of h^2 against time. Three lines have been drawn on the diagram to estimate the range of values of the mixing coefficient, K_y, that are consistent with the observed spreading of the tungsten-185 cloud. The lines drawn to fit (a) the three values for January through June 1959, (b) all five values, and (c) the end-point values for November-December 1958 and July-August 1959 yield values of Ky, from eq. (9), of 8.3, 5.8, and 4.4×10^8 cm^2 sec^{-1} respectively, in agreement with the estimates shown in Figure 40. It should be noted that this experiment constitutes the first use of unique radioactive tracers for the calculation of the large scale mixing coefficients in the stratosphere.

The total stratospheric inventory of tungsten-185 was computed by the same technique used for the computation of the strontium-90 inventories. The removal rate of tungsten-185 from the stratosphere was calculated from the successive bi-monthly inventories as shown in Table 16 and in Figure 45. The ordinate in

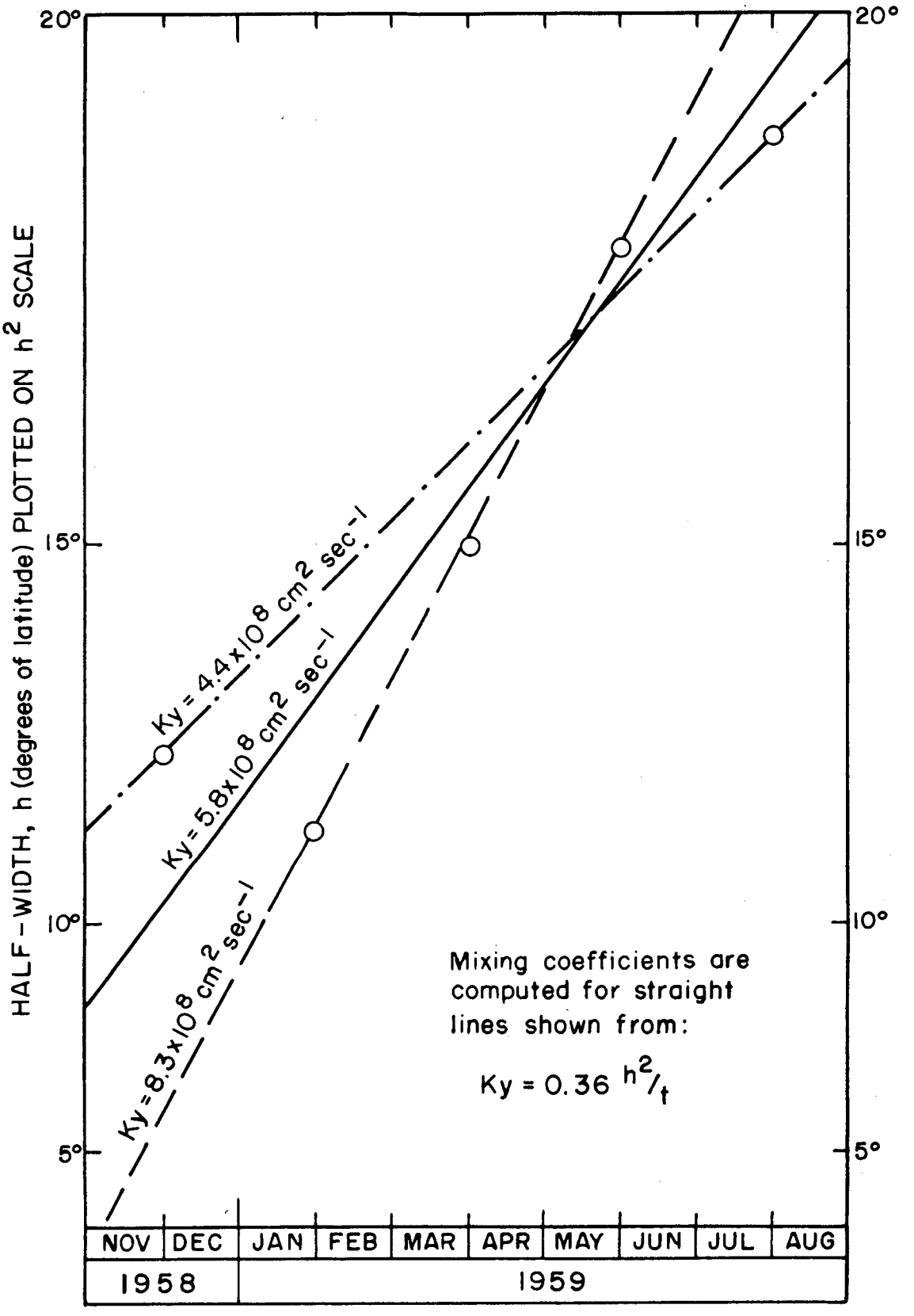


FIGURE 47 VARIATION OF MERIDIONAL HALF—WIDTH OF TUNGSTEN-185 CLOUD

Table 16. Bi-Monthly Stratospheric Inventories of Tungsten-185
(Corrected for Radioactive Decay to 15 August 1958)

Sampling Interval	Mos. After July-Aug 58	Megacuries W^{185}	Percent * Removed per Month	Cumulative: * Percent of Initial Reservoir Removed	Cumulative: ** Percent of Initial Reservoir Removed if T-1/2 = 6 Months
Sept-Oct. 58	2	(61)			
Nov-Dec. 58	4	36	38	32	37
Jan-Feb. 59	6	26	32	51	50
Mar-Apr. 59	8	23	12	57	60
May-Jun. 59	10	21	9	60	68
Jul-Aug. 59	12	21	0	60	75

* The removal rates are computed from an estimated "initial" value in July-August 1958 of 53 MC, and the September-October 1958 inventory is disregarded in this calculation.

** The cumulative removal at a residence half-time of 6 months is included here for comparison.

Figure 45 is megacuries of tungsten-185, corrected to 15 August 1958, on a logarithmic scale, and the abscissa is time. The effective input on 15 August 1958 is shown as 53 MC, and the bi-monthly inventories are plotted at the mid-points of the bi-monthly sampling intervals. Also shown on the diagram as sloping straight lines emanating from the input point are the residence half-time curves corresponding to 6, 8, and 12 months.

The value for September-October 1958 is apparently not representative of the world total, for this inventory value exceeds the estimated input. In this period the aircraft were apparently sampling relatively "hot" clouds of debris which were not yet "smeared out" around the hemisphere. Therefore this inventory value has been ignored in the calculations of removal rates.

As shown in Table 16, the depletion of the stratospheric reservoir has not proceeded at a constant removal rate. At the end of the first six months after injection approximately 50 percent of the debris had been removed from the stratosphere. However, the removal rate, which was initially rapid, decreased markedly with time as shown in the column marked "percent removed per month." In the spring-summer period (May-August), effectively none of the tungsten fell out of the stratosphere (i.e., the removal was less than half of a megacurie). The column labelled "cumulative percent of initial reservoir removed" also shows the decline in the fallout after the winter of 1959. A comparison of this column with the last column on the right which shows the cumulative percent

removal corresponding to a residence half-time of 6 months indicates that the "effective" half-time was about 6 months for the first 6 or 8 months after injection. However, the "effective" half-time clearly increases with time, as the fallout rate decreases during the spring-summer period.

The variable removal rate is also illustrated in Figure 45. The "effective" half-time increases from about 6 months for the first six months, to about 8 months for the first year.

The tungsten-185 data clearly show that the removal rate of Equatorial debris is somewhat less rapid than that of polar debris, but still much faster than earlier estimates. The removal rate is also clearly variable and does not correspond to a constant residence time.

CHAPTER X

SURFACE DISTRIBUTION OF STRONTIUM-90 FALLOUT

Introduction

To utilize fully the HASP information, it is necessary to determine the quantity of strontium-90 which has been deposited on the surface of the earth as world-wide fallout. The knowledge gained from these investigations can be used in the evaluation of the hazard resulting from world-wide fallout and to predict future hazards which may result from renewed nuclear testing, a nuclear exchange, or from the operation of atomic installations being used for peaceful purposes.

Previous attempts to estimate the surface inventory and surface distribution of strontium-90 as a function of time have been described in the Report of the United Nations Scientific Committee on the Effects of Atomic Radiation (1958) and the congressional hearings before the Joint Committee on Atomic Energy[25]. This chapter will describe the distribution and will estimate the total concentration of strontium-90 on the earth's surface for various time intervals between July 1954 and January 1960. Consideration has been given to the assumptions used in the various calculations, to sources of uncertainty, to the desirability of additional information, and to the implications of these results as applied to the fallout problem.

Computation of Surface Inventory of Strontium-90

The method of computation of total fallout is based on the following assumptions:

1. Strontium-90 analyses of suitable soil samples provide a good estimate of the total strontium-90 fallout which has occurred at a given site.

2. The computation of total world fallout from point observations of strontium-90 (in soil or in precipitation) is based on the integration formula:

$$F = \int_0^{\pi} \int_0^{2\pi} f\, p\, r^2 \cos\phi \, d\lambda \, d\phi \qquad (1)$$

where f is the specific isotopic activity per unit area per unit of precipitation, p is the amount of precipitation (expressed as the depth of the liquid water equivalent in the case of snow) measured over the same time interval, r is the mean radius of the earth, and F denotes the total isotopic activity deposited on the area A. The world fallout may be determined by integrating over latitude ϕ, and longitude, λ.

We may evaluate F by means of the approximation formula:

$$F = \sum_{i=1}^{N} A_i \overline{(fp)}_i \qquad (2)$$

where A_i represents the area of the latitude belt, i, around the earth, N denotes the number of such latitude belts on the earth, and $\overline{(fp)}_i$ represents the mean value of fp in the latitude belt.

The mean values, $\overline{(fp)}_i$, may be computed from the approximation formula:

$$\overline{(fp)}_i \simeq \frac{1}{A_i} \sum_{j=1}^{n} f_j p_j \Delta A_j \qquad (3)$$

Where the subscript j denotes a point observation, n is the number of observations in the latitude belt, and Δ Aj denotes the area over which the observations f_j and p_j are assumed to be representative (i.e., over which they may be treated as appropriate mean values). If all observations of f and p are assumed to be representative of areas of equal size within the latitude belt, and if there are enough observations to cover the entire latitude belt, the formula for the arithmetic mean may be used, i.e.:

$$\overline{(fp)}_i \simeq \frac{1}{n} \sum_{j=1}^{n} f_j p_j \qquad (4)$$

In general, however, the distribution of observations is neither uniform nor sufficiently dense over most of the earth, so that (4) cannot be applied directly. For the calculation of the world fallout, it is therefore necessary to adopt certain working hypotheses.

The mean precipitation and mean specific activity in each latitude belt will be denoted by $\bar{P}_i$ and $\bar{f}_i$ respectively [where each mean is computed in the sense of (3)], and deviations from the zonal mean will be represented by p_j' and f_j' respectively. The zonal mean fallout is given by:

$$\overline{(fp)}_i = \bar{f}_i \bar{P}_i + \overline{f'_j P'_j} \qquad (5)$$

where the bars denote space means over the latitude belt. If f and p are not correlated, the second term in **(5)** is zero, and the mean fallout may be computed by multiplying the mean specific activity in the latitude belt by the mean precipitation. Regions with quite low rainfall appear to exhibit anomalously high specific activity and there seems to be some evidence of a slight correlation between precipitation and specific activity but it is not too marked.[4] The desert phenomenon may be due to the dry fallout, and an appropriate adjustment can be made for that effect. It may not be too unreasonable to calculate the zonal mean fallout from the first term in (4) alone, and the only remaining questions concern the reliability of the stimates of f_i and p_i.

3. World-wide fallout is assumed to be almost entirely of stratospheric origin, i.e., delayed fallout. Other sources of strontium-90 now on the surface of the earth are:

(a) Local fallout adjacent to test sites (within 30 miles of the Nevada Proving Grounds and other sites where only small weapons have been tested and within several hundred miles of sites where megaton-weapons have been detonated).

(b) Intermediate fallout downwind from test sites and as much as 1000 miles distant, e.g., Des Moines, Iowa and Mandan, North Dakota). This includes debris from kiloton shots and from the "stem" of the megaton shots.

(c) The world-wide deposition from clouds from kiloton shots and from the stems of megaton shots. This deposition is generally restricted to a 10-20 degree latitude band containing the test sites.

In this computation of the total world surface inventory of strontium-90 of stratospheric origin the contributions from (a) and (b) have been avoided by omitting the results of soil samples collected in these areas. The contribution from (c) is difficult to estimate, but it is generally agreed to make up less than 5% of the surface inventory and for this reason no attempt has been made to estimate this contribution separately.

General Contributions Concerning Strontium-90 in Soils

The most important point to be considered in the sampling of soils which are to be used to estimate the total amount of strontium-90 which has fallen on a given region is the choice of a suitable site. A basic requirement for a site is that it should have retained all of the strontium-90 which was deposited. There should have been no addition of strontium-90 from latent movement on the earth's surface (e.g., by wind or run-off) and the sample should be taken to sufficient depth to ensure that all deposited strontium-90 is included. In theory these requirements appear reasonably easy to fulfill, but in practice many subtle problems are encountered. If the area is large in extent, level, and has a substantial grass cover, it should retain virtually all of the strontium-90 which falls

on it. Unless the soil is highly impermeable, appreciable run-off will not occur except in the case of the heavy showers. Fortunately, as will be shown below, the strontium-90 is retained in the upper few inches of the soil so that with reasonable effort essentially all of the strontium-90 in the sampling area can be taken in the sample. The area to be sampled should not have been plowed or otherwise disturbed since 1952, and it must be relatively distant from forests or buildings that might systematically shield it from rain in one direction. These criteria have been met in most of the samples which have been collected to date. There are, in addition, several other problems to consider.

1. In the plains areas and in the most northerly latitudes a smooth unprotected area may be swept clean of much of the deposited snow and hence would lose this fraction of the annual fallout. The sites at **Ellesmere** Island and Cornwallis Island (70^{o} - 80^{o} N) will suffer from the drawback that they were sampled in areas of slight vegetation. As a result of drifting, snow might be expected to accumulate in these areas but when the low growth becomes uniformly covered with snow there will be no further preferential accumulation. Without detailed investigation at such sites it must be assumed that there is considerable uncertainty in the data compared with those from lower latitudes. On the other hand, the site at Bardufoss (Norway) is probably one of the best, since it is situated in an open forest where the snowfall is certainly more uniform. A second unanswered question here involves the estimation of the amount of precipitation

(snow) for the northerly latitudes. It appears at present that no reliable routine method exists for the estimation of quantities of snowfall.

2) When lawns of institutions or level meadow land are sampled, as has often been done, a possible error is introduced if a fraction of the strontium-90 has been removed by (a) cutting and raking grass (b) cutting and removal for hay or (c) grazing. Factor (a) may remove as much as 10% of the total deposited strontium-90. In the 1958 U. S. D. A. collection two sites were sampled at Salisbury, Southern Rhodesia. One of these sites was on the lawn of an institution and should be subject to the renewal factor, while the other was taken from a tract of virgin bunch grass. The cumulative strontium-90 values were the same within the experimental error. Similar agreement was obtained on a pair of samples from Durban, South Africa. About 20% of the sample sites reported to date were not from lawn areas and a comparison of these values with those from lawn samples indicates similar average values. While the possibility that this effect is significant in certain areas cannot be excluded, these results suggest that the error introduced into the calculation of the total fallout at the site is less than 10 percent.

3) Biological activity may be a factor in rare instances. For example, a high population of worms may cause extensive cracking of the surface, resulting in the strontium-90 being moved rapidly downward. Actually no such cases have been reported in the sampling to date.

The best and most reliable procedure for soil sampling is that developed by Dr. L. T. Alexander and used in obtaining the 1958 U. S. D. A. collection. At each site 20 cores of 3 1/2 inches diameter are taken to 0-2 inches and 2-6 inches depth at ~3 foot centers. The samples are then carefully mixed and pulverized prior to an aliquot being analyzed for strontium-90. This technique provides samples with minimum surface contamination, accurately known depth and cross section and tends to average out the very local variations caused by rocks, stones, etc. The validity of this procedure is shown by the reproducibility of duplicate samples taken within twenty five miles of each other in areas where there is no obvious topographic reason for differences in rainfall. Thus in the 1958 set where the analytical reproducibility gave a standard deviation of about 5 percent the standard deviation among such paired samples from the same locality was only 7 percent. It should be noted that all except two of these pairs of samples were taken within five miles of each other.

A second method, used by the United Kingdom workers[26] for the 1956-1957 collection, consists of taking several samples down to a total depth of four inches in soil. Ten plots of one square yard in area are sampled over a total area of half an acre. Samples of grass from each site are obtained and pooled followed by removal of the grass "mat" and finally a core of soil to 4 inches depth is taken at the same spot from which the mat was removed.

The third method which was used extensively during the earlier work at

Lamont Geological Observatory, consists of taking a single square foot sample to 2 inch depth with a one foot square metal form and a hand trowel. A standard deviation of about 20 percent was obtained on the results from many pairs of samples collected within ten to twenty miles of each other.

Both the second and third methods suffer in precision because the soil is not sampled to sufficient depth. The United Kingdom method is extremely useful, however, because it gives us knowledge of the distribution of strontium-90 not only with depth but also within the various solid phases which are present. In future work it would be advisable for these refinements to be added to the Alexander method and for the sampling to be carried to greater depth.

Variation with Depth of Strontium-90 in Soil

It has been evident from the time the first measurements of strontium-90 in soil were made at Lamont in the summer of 1953, that most of this isotope is retained in the uppermost inches of the soil. By 1955, a fairly large number of analyses had been performed on samples from various depths. Most of this sampling was relatively crude, the common intervals being 0-2 inches and 2-6 inches, although some 6-10 inch and 6-12 inch samples were taken. The early work at Lamont in 1953 (Lamont Geological Observatory, 1954) suggested that about 80 percent of the strontium-90 remains in the upper 2 inches with a negligible amount penetrating below 6 inches.

Adequacy of Sampling

A most critical question to be asked about any computation of the world surface inventory of strontium-90 concerns the adequacy of the sampling network. In Table 17 is listed the number of sampling stations on land for each 10^{o} latitude band covered by the U. S. D. A. world-wide collection. This list does not indicate of course, the extensive coverage of the United States by the U. S. A. E. C. or of the United Kingdom by the U. K. A. E. C. It should be remembered that 70% of the earth's surface is covered by water and the number of stations in this extensive section is small. With the exception of the four weather ships which are operating in the Atlantic Ocean we have to rely on data from island stations and measurements of strontium-90 in ocean water for oceanic fallout estimates. Although this represents a meager coverage of the earth's surface it is felt that it is adequate to estimate the surface burden of strontium-90 to a first approximation. As will be discussed below the large uncertainty in the estimate stems from the uncertainties in the value of $\overline{p}_i$ the average rainfall for a given latitude belt.

Results of Strontium-90 Surface Inventory Calculations

The computation of the total world inventory of strontium-90 has been performed for various dates from 1954 to 1960. These results were calculated according to the following steps:

1. All of the reported soil activities measured in a given year have been

TABLE 17--Distribution of U.S.D.A. 1958 Worldwide Soil Collection Sites

Latitude band	Percent land	No. of stations
70-80 N	29	3
60-70 N	70	7
50-60 N	57	2
40-50 N	52	8
30-40 N	43	6
20-30 N	38	4
10-20 N	27	7
0-10 N	23	4
0-10 S	24	4
10-20 S	22	4
20-30 S	23	6
30-40 S	11	5
40-50 S	3	3
50-60 S	1	2

corrected to July 1 of the same year. The increment to be added or subtracted from the measured strontium-90 concentration (expressed in $mC/mile^2$) can be found from the specific activity of precipitation for the period of correction and the rainfall for the period. The rainfall for the period under question is estimated from the data published with the soil analyses, as for example in the U. S. D. A. suite (Alexander, 1959), or from rainfall distribution maps (U. S. Department of Agriculture, 1941). Specific activity values for precipitation are taken from Table 18, which has been computed from measured rainfall specific activities. The data for the third and fourth quarters of 1959 have been interpolated on the basis of the current moratorium in nuclear testing and an assumed residence time for nuclear debris in the troposphere of about one month. In general this correction is small for the majority of stations.

2. If the sample were taken to 6 inches depth it is assumed that 100% of the strontium-90 was obtained, if taken to 4 inches-90% and if to 2 inches, 70% was assumed to have been obtained.

3. The cumulative $mC/mile^2$ concentration at the selected date is divided by the mean annual rainfall.

4. An average specific activity of soil per inch of mean annual rainfall is calculated for each 10^o latitude band and the total amount of strontium-90 in each band is obtained by multiplying this figure by the area of the band and the mean annual rainfall for the band[27] (taken from Möller's data, 1951). The

TABLE 18--Specific Activity Values of Precipitation

Used for Extrapolation of Soil Data (mC/mile^2/in.)

	South Latitude				North Latitude			
Quarter	90-30	30-20	20-10	10-0	0-10	10-20	20-30	30-90
1954								
1	0.03	0.01	0.01	0.01	0.01	0.02	0.06	0.11
2	0.03	0.01	0.01	0.01	0.01	0.03	0.11	0.16
3	0.05	0.03	0.01	0.01	0.01	0.03	0.06	0.09
4	0.05	0.03	0.02	0.02	0.02	0.03	0.08	0.10
1955								
1	0.06	0.02	0.02	0.02	0.02	0.04	0.13	0.23
2	0.06	0.06	0.02	0.02	0.03	0.09	0.24	0.36
3	0.07	0.12	0.03	0.03	0.03	0.07	0.16	0.24
4	0.07	0.12	0.01	0.01	0.02	0.04	0.08	0.10
1956								
1	0.07	0.03	0.02	0.02	0.02	0.06	0.19	0.33
2	0.07	0.05	0.02	0.02	0.03	0.12	0.32	0.47
3	0.09	0.06	0.02	0.02	0.03	0.05	0.12	0.18
4	0.09	0.05	0.02	0.02	0.03	0.06	0.14	0.19
1957								
1	0.09	0.02	0.02	0.02	0.02	0.05	0.14	0.24
2	0.09	0.04	0.03	0.03	0.03	0.08	0.20	0.30
3	0.06	0.06	0.03	0.03	0.02	0.05	0.13	0.20
4	0.09	0.06	0.03	0.03	0.02	0.05	0.12	0.16
1958								
1	0.07	0.02	0.02	0.02	0.02	0.05	0.18	0.25
2	0.04	0.04	0.03	0.03	0.03	0.09	0.28	0.58
3	0.07	0.10	0.03	0.03	0.03	0.07	0.16	0.30
4	0.11	0.10	0.03	0.03	0.03	0.07	0.16	0.35
1959								
1	0.08	0.02	0.02	0.02	0.05	0.13	0.48	0.80
2	0.05	0.04	0.03	0.03	0.08	0.21	0.72	1.24
3	0.08	0.10	0.03	0.03	0.01	0.03	0.09	0.18
4	0.13	0.10	0.03	0.03	0.01	0.01	0.02	0.02

total world inventory is then calculated from the summation of the latitude band results.

In Table 19 the specific activity values are given for each 10^{o} latitude band. Since there are virtually no sampling stations north of 70^{o} N or south of the 60^{o} S certain assumptions have to be made about the strontium-90 deposition in these areas. It has been assumed that the specific activity in rain is constant from 30^{o} N to 90^{o} N and 30^{o} S to 90^{o} S. Such a conclusion receives support from other theoretical considerations. It is now apparent that the mechanism of entry of the dominant fraction of nuclear debris into the troposphere from the stratosphere is through the "break" in the tropopause in the temperate latitudes. Furthermore since there is very little precipitation in the polar regions it might be expected that the troposphere in the region 70^{o} - 90^{o} north and south would be essentially in equilibrium with the 30^{o} - 60^{o} belts. Hence the specific activity of strontium-90 in air at ground level should be the same in the higher latitudes as in the 30^{o} - 60^{o} bands in each hemisphere. In this table, therefore, the specific activity values observed in the 30^{o} - 60^{o} N and 30^{o} - 90^{o} S region have been averaged according to the number of observations and these averages applied to the whole region 30^{o} - 90^{o} N and 30^{o} - 90^{o} S respectively. Some of the values reported for rainfall and snow in these northerly latitudes suggest that a tapering off in activity might be more in keeping with the true situation. The overall change, however, which this assumption incurs in the total

TABLE 19--Values of mC/mile^2/in. Mean Annual Rainfall In Soil for Each 10° Latitude Band at Various Times

	South Latitude				North Latitude			
Date	90-30	30-20	20-10	10-0	0-10	10-20	20-30	30-90
1960 1/1	0.43	0.31	0.18	0.22	0.20	0.38	0.94	1.71
1959 7/1	0.38	0.26	0.16	0.21	0.19	0.36	0.88	1.61
1958 7/1	0.30	0.20	0.13	0.18	0.14	0.24	0.50	0.94
1957 7/1	0.21	0.13	0.10	0.07	0.10	0.13	0.34	0.53
1956 7/1	0.12	0.09	0.07	0.08	0.07	0.15	(0.21)	0.33
1955 7/1	0.06	0.06	0.02	0.02	0.04	0.07	0.17	0.21
1954 7/1	----	----	----	----	----	----	----	0.08

inventory is certainly less than 5%.

The results in Table 19 have been used to calculate the total world inventory of strontium-90 at these dates and the final tabulation is shown in Table 20. It must be pointed out that this inventory is for stratospheric fallout only and that stations receiving intermediate and local fallout have been omitted for most of the sites in the continental United States. Results from sites such as Antofagasta, Chile (rainfall less than 5 inches per annum) have been omitted also in calculating the specific activity reaching the soil per inch of rainfall because the extremely low rainfall in these areas leads to anomalously high specific activity values. The contribution from dry fallout is probably primarily responsible in these cases because the specific activity of fission products in air over these areas in not anomalous. Extrapolations to 1959 and 1960 were based on the specific activity of rainfall shown in Table 18.

Discussion

One of the major parameters in this calculation which merits discussion is the choice of the mean annual rainfall for each latitude band. The values of **Möller** (1951) have been adopted. Rates of depletion from the stratosphere are such that it appears the maximum level of strontium-90 on the ground will be reached in 1961. After that time the rate of decay of strontium-90 will be greater from the rate of deposition. The estimated maximum values shown in Table 20 were computed by adding 10% to the Southern Hemisphere 1 January 1960

TABLE 20--Summary of Surface Inventory of Strontium-90 (in megacuries)

Latitude	Area $x10^{-6}$ $mile^2$	Mean Rain-fall	Est. 1961 Max.	1/1/60	7/1/59	7/1/58	7/1/57	7/1/56	7/1/55
North									
90-80	1.6	4	0.014	0.011	0.010	0.006	0.003	0.002	0.001
80-70	4.3	6	0.057	0.044	0.041	0.024	0.014	0.009	0.005
70-60	7.3	14	0.228	0.175	0.165	0.096	0.054	0.034	0.021
60-50	9.8	26	0.567	0.436	0.410	0.240	0.135	0.084	0.054
50-40	12.2	31	0.876	0.647	0.609	0.356	0.200	0.125	0.079
40-30	14.0	28	0.871	0.670	0.631	0.368	0.208	0.129	0.082
30-20	15.5	25	0.473	0.364	0.341	0.194	0.132	0.081	0.066
20-10	16.5	34	0.277	0.213	0.202	0.135	0.073	0.084	0.039
10-0	17.1	61	0.272	0.209	0.198	0.146	0.104	0.073	0.042
South									
0-10	17.1	46	0.190	0.173	0.165	0.142	0.055	0.063	0.016
10-20	16.5	37	0.121	0.110	0.098	0.079	0.061	0.043	0.012
20-30	15.5	26	0.138	0.125	0.105	0.081	0.052	0.036	0.024
30-40	14.0	31	0.206	0.187	0.165	0.130	0.091	0.052	0.026
40-50	12.2	44	0.254	0.231	0.204	0.161	0.113	0.064	0.032
50-60	9.8	38	0.176	0.160	0.142	0.112	0.078	0.045	0.022
60-70	7.3	16	0.055	0.050	0.044	0.035	0.025	0.014	0.007
70-80	4.3	3	0.007	0.006	0.005	0.004	0.003	0.002	0.001
80-90	1.6	1	0.001	0.001	0.001	-----	-----	-----	-----
			4.78	3.81	3.54	2.31	1.40	0.94	0.53

figure and 30% to the Northern Hemisphere 1 January 1960 figure. The total lies near the amount injected into the stratosphere. Figure 48 shows the expected distribution of Sr-90 averaged over each latitude band when the maximum is reached. The figure also shows that by May 1960 90% of the amount of material originally placed in the stratosphere from bursts below 100,000 feet had reached the ground.

Material Balance

Table 21 shows an attempt to strike material balances using the sum of stratospheric and surface inventories against estimated stratospheric injections for mid-1958 and mid-1959.

Considering the uncertainties involved in the measurements, there is remarkably good agreement. It appears that about one third of all the strontium-90 produced by all nations to date has been deposited locally in the Pacific Ocean.

FIGURE 48

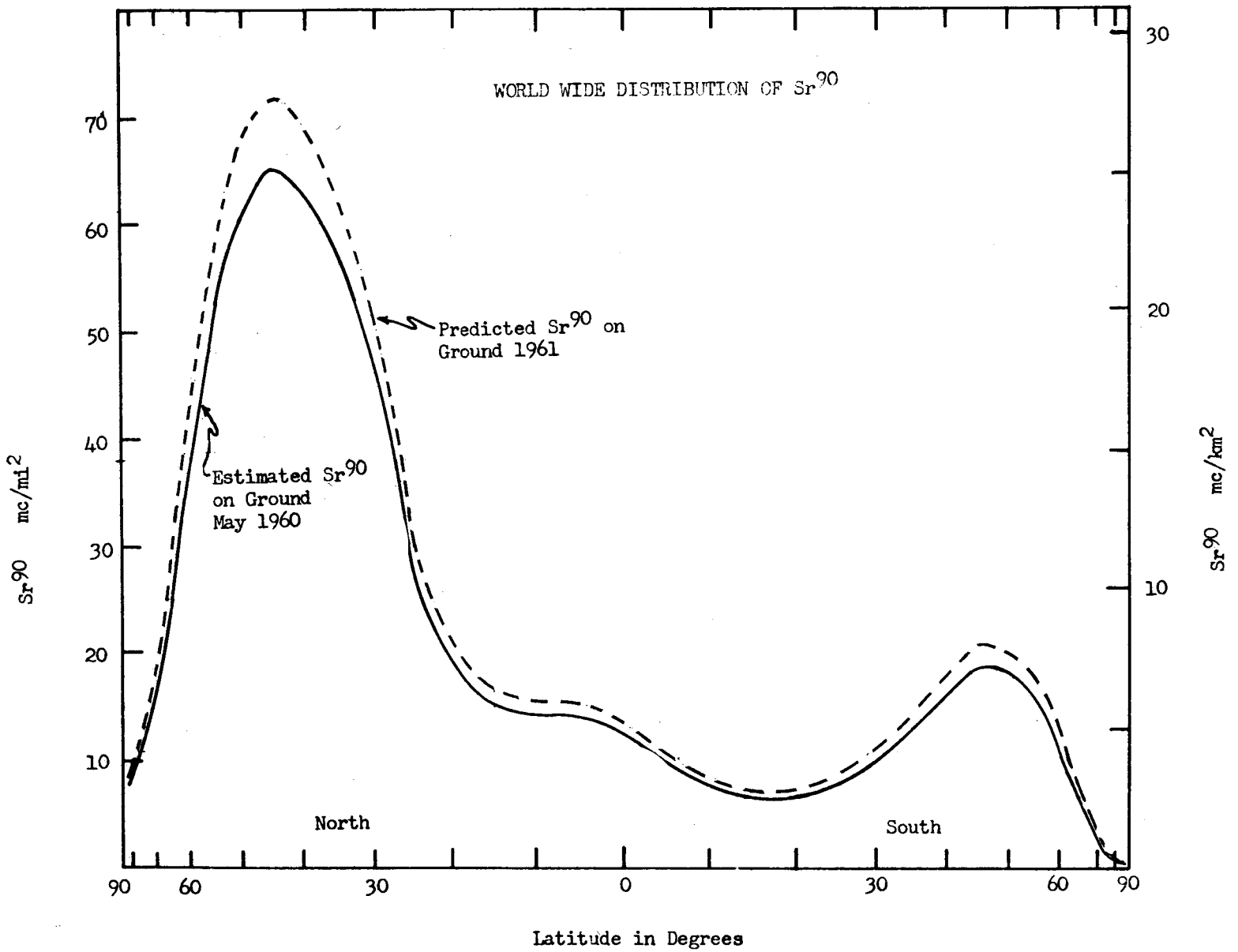

WORLD WIDE DISTRIBUTION OF Sr90
Predicted Sr90 on Ground 1961
Estimated Sr90 on Ground May 1960
Sr90 mc/mi2
Sr90 mc/km2
North
South
90
60
30
0
30
60
90
10
20
30
40
50
60
70
10
20
30
Latitude in Degrees

Table 21

Distribution of Stratospheric Strontium-90

Fallout (Megacuries)

	July 1958	Mid-1959
Stratosphere:		
"ASHCAN" Extrapolation	0.9 Mc	0.8 Mc
"Uniform" Extrapolation	(1.2)	(1.1)
"Carbon-14" Extrapolation	(1.1)	(1.0)
Surface:		
Alexander (Adjusted)	(2.8)	
Kulp and Walton	2.3 Mc	3.6 Mc
TOTAL	3.2 Mc	4.4 Mc
Stratospheric Injection:		
Before Fall 1958 Soviet Tests	3.6 Mc	
Including Fall 1958 Soviet tests		4.8 Mc

CHAPTER XI

RADIATION HAZARD FROM FALLOUT

Introduction

Several factors must be considered in evaluating the potential hazard to the human population from the radioactive components of world-wide fallout. Basically, however, the extent of the hazard from a nuclide depends upon (1) the amount of radiation received from that nuclide by the human body and (2) the effect of that radiation on the body. The High Altitude Sampling Program is contributing information which permits a better estimation of the first of these.

The amount of radiation that a human will receive from a particular radioactive nuclide that enters his environment in world-wide fallout depends upon (1) the amount of fallout deposited in the region in which he lives, (2) the concentration of that nuclide in this fallout, (3) the amount and strength of the radiation emitted by that nuclide, (4) the amount of that nuclide which he ingests or inhales into his body, (5) the length of time during which the nuclide remains in his body, (6) any concentration of the nuclide in a critical tissue. The fourth factor, the amount of the nuclide taken into the body, is in itself a function of several variables. For nuclides such as Sr^{90} the concentration of the nuclide in soil, the discrimination in favor of or against the nuclide during its incorporation into plants or animals which will serve as human food and the relative amounts of such foods eaten by an individual will influence the danger of the nuclide to him.

Because of the complexity of the processes by which fallout affects man, the hazard must be evaluated in terms of entire populations to make allowance for fluctuations in concentrations and susceptibilities.

Uptake from Lower Atmosphere

Since the various fission products which reach the ground have different half-lives and chemical properties, their effect upon man will vary greatly. Dosages to the body can consist of either whole body irradiation or localized irradiation such as to the bone, to the thyroid, or to the skin. Either of these types of dosages may be received from radioactive isotopes residing outside the body or taken into the body.

We are forced to decide which isotopes are of greatest potential hazard and then study each in detail. Strontium-90 and Cesium-137 are the most obvious elements to consider in the case of dosage received from internal sources. They are produced in relatively large abundance, have chemical properties which promote their retention in the body, and have half-lives on the order of 28 years. This latter characteristic is important since a very short half-life would mean that most of the material would decay away before it had a chance to enter the body. Obviously we receive no dose from this material. On the other hand, if the half-life were long the victim would die of other causes before he received much of a dose.

Strontium-90 behaves chemically much like calcium and consequently if taken into the body will be deposited in the bone structure of an

individual. Therefore, Strontium-90 is probably the most important single isotope to consider from a health standpoint since its radiations may contribute to the development of bone cancer or leukemia. Cesium-137 and carbon-14 may possibly contribute to the genetic mutation rate in man. Other possibly important materials produced are unfissioned plutonium and iodine-131.

The avenues along which these products can get into the human body are varied. They can fall directly upon vegetables whose leaves can be eaten, or they can be gradually incorporated into plants from the soil through their roots. If these plants are eaten by animals, such as sheep and cattle, we will find these products in our meat and dairy produce.

Among western people, milk is the greatest source of calcium in the diet and consequently is the major source of Strontium-90. However, the cow acts as a natural filtration plant in that the strontium to calcium ratio in its milk is only about 15% of the strontium to calcium ratio in its fodder. There is further discrimination in the human body in that the level deposited in bone will be less than that of the diet by a factor which appears to range from two to four.

Dietary Levels

Future Dietary levels of strontium-90 in the U. S. for testing to date are shown in Table 22.

Table 22

Predicted Sr^{90} in U. S. Diet

Mid-Year	Dependent on Cumulative Deposit	Dependent on Rate of Fallout
1959	(15.0) μμc/gr Ca	(15.0) μμc/gr Ca
1960	16.6	5.0
1961	17.0	2.5
1962	17.0	1.2
1963	16.6	.6
1964	16.2	.3
1965	15.8	.1
1970	13.8	0

There are two limiting cases indicated by the two columns. Both cases assume Walton's estimate of the surface deposition, the HASP stratospheric inventory, a half residence time of about one year for the residual debris in the stratospheric reservoir and no more tests. The difference lies in whether the strontium-90 level in diet is proportional to the cumulative surface deposit or the rate of fallout. A considerable body of evidence is accumulating that the actual case is somewhere in between.

The dietary level in 1959 is fairly well known from the various milk networks in the U. S. It is taken as 15 μμc Sr^{90}/g Ca in the summer of 1959. If the strontium-90 content of the average diet is exclusively related to the cumulative deposit, the level will rise to about 17 units in 1961 and then begin to decay with the strontium-90 half life of 28 years. If it were purely related to the rate of fallout, the concentration in food will drop essentially to zero in a few years. The most probable estimate at present is that the cumulative factor represents 25 to 50%.

It appears that the maximum equilibrium bone levels which will be reached in the United States population from material produced by tests to date will average between 5 and 6 micromicrocuries of strontium-90 per gram of calcium. Among Eastern people, where rice is a greater source of calcium, the levels may rise as high as 10 or 12 micromicrocuries of strontium-90 per gram of calcium.

Much publicity has been given to some of the high levels of strontium-90 in certain foods in the northern part of the U. S. These levels have been generally of rather transient nature, and it is felt that these hot spots in locality and time are important only to the extent that they add to the overall national average. It should be pointed out that levels of activity in food are higher in the United States than anywhere else in the world.

Effects of Fallout on Man

Although there have been many advances in measurement of the contribution of fallout to the radiation hazard to man, there still remain a number of problems in determining the ultimate effects of such foreign material on humans. Broadly speaking these effects fall into two categories, namely, genetic and somatic. Genetic effects are those effects we may hand down to our offspring through disrupted genes in our reproductive cells. Somatic effects are those effects we may feel in our own body such as diseases or accelerated ageing.

When approaching this problem we are forced to consider past experience in a number of other fields. The direct determination of the

ultimate human effects of long term, low level fallout would require many years, since a number of generations would need to be followed. Since we cannot mark time for several human generations, we must use other methods. One approach is to utilize laboratory animals which will produce many generations in a much shorter period. However, this approach presents the problem of interpreting the data in the light of species difference between man and other animals.

Another approach is to relate fallout exposures to other similar human radiation exposure, which occurred long enough ago to allow some analysis of the ultimate results. One such type of exposure which has been occurring since the evolution of man is that of the natural background radiation. This is similar to that fallout radiation which gives whole body radiations, whether from external or internal sources.

The natural background radiation has always existed and actually was greater in the past than it is today. In fact it may be that this radiation was a major contributor to the evolution of the species we find on this earth. This background consists of two parts, that of terrestrial origin and that of extra-terrestrial origin. The latter, known as cosmic radiation, varies with latitude and altitude being greater at higher latitudes and altitudes. The background radiation of terrestrial origin comes from those naturally radioactive elements found in rocks and minerals.

At the present time the chief known sources of natural background radiation on earth consist of:

a. The uranium and thorium ores, including pitchblends, carnalite, and monazite sands. Uranium is also spread widely through rocks and oceans.

b. Potassium-40, a radioactive relative of the stable potassium. Both are normally found in all animal cells and wherever salts are found.

c. Cosmic radiation.

d. Carbon-14, occurring in all organic material as a result of cosmic ray bombardment of atmospheric nitrogen.

In general, the natural background radiation varies roughly between 100 and 150 milliroentgens per year to the world's population.

The variations result mainly from the following factors:

a. Cosmic ray doses are higher with increasing altitude.

b. The component from uranium is lower over the ocean than over the land.

c. Wood used in construction produces less dose than some bricks used in construction.

Whether or not the present background radiation is injurious has not been established. It provides, however, a partial basis for comparison of long term genetic effects from fallout. During the thirty year period starting with 1952, the total dose to human reproductive tissue from both internal and external sources in fallout, which has reached

the ground or will reach the ground, will be no more than 0.05 roentgen. This figure has been calculated for the north temperate regions where the fallout is greatest. The natural background, on the other hand, will give a dose of at least 3 roentgens during the same period. In other words, nuclear weapons testing will increase the genetically important background by no more than 2%.

Table 23 shows the relative contributions of the various potentially hazardous isotopes for tests to date based on the inventory and residence time as determined by HASP.

TABLE 23

Radiation Dose from Existing Nuclear Debris

Isotope	Time	Situation	Per cent of MPC
Sr^{90}	end of 1960	0-5 yr. bone, Prop. to deposit	4-6
	1965	0-5 yr. bone, Prop. to rate	1
	2000	0-5 yr. bone, Prop. to rate	0
Cs^{137}	1960	Internal, Prop. to deposit	0.4
	1965	Internal, Prop. to rate	0
	1965	External	0.08
Zr^{95}, etc.	early 1959	External	2.0
	1965	External	0
C^{14}	1960		0.1
	1967		0.05
	2000		0.002
Pu^{239} (From inhaled resuspended dust)			
	1960	lungs	0.05-0.3
	1960	gonads	0.7
	1960	pulmonary lymph nodes	0.3-1.9
Pu^{239} (From fresh fallout in inhaled air)			
	1965	all organs	0

Considering strontium-90 first, the average concentration in the skeleton of young people may reach 4 to 6% of the MPC by the end of 1960 if the level in bone is strictly proportional to the cumulative deposit. If proportional to rate, by 1965 the average bone level for this group would have dropped to 1% and would be negligible by the year 2000.

The gamma dose due to ingested cesium-137 should be about 0.4% of MPC in 1960 if proportional to the deposit but if, as seems more likely the case, it is largely dependent on surface uptake and thus, the rate of fallout, it should be negligible by 1965. The external gamma field due to cesium-137 will decrease only with the radioactive decay. In 1965 it is estimated to be .08% of MPC assuming a ten fold attenuation due to geometry.

The intermediate lived isotopes such as Zr^{95}, Y^{91}, Ru^{103}, Ce^{144}, etc., made greater contributions than Cs^{137} in early 1959 but as tests stop this quickly becomes negligible. Under conditions of continuous testing the contributions from these isotopes would dominate the internal or external Cs^{137} gamma dose rate.

The carbon-14 concentration in the living biosphere has increased by about 30% so that at present it is at a level of about 0.1% of MPC or about one-fourth of the cesium-137 dose rate. This will decay away into the oceans with a 7 year half time so that by the year 2000 it will be only .002%.

A recent survey of human tissue for plutonium-239 has shown levels such as those given in the right-hand column. If these human burdens are due to inhaling resuspended surface dust, they may represent an essentially permanent level from existing fallout. If they are due to inhalation of fresh fallout, the levels will drop to negligible proportions by 1965. In the first case, plutonium-239 could become the most important source of radiation from nuclear tests. The large variation in the potential resuspension at different geographical sites indicates further investigation is desirable.

Summary

In a real sense local fallout is of greater significance than world-wide fallout. In the event of a nuclear war, local fallout has the capacity of producing a large number of early casualties at distances hundreds of miles away from the point of detonation. In the two weapons utilized in Japan in 1945, there was essentially no local fallout because high air bursts and relatively low yields were used. However, there is no guarantee that any countries would confine themselves to air bursting their weapons in the future because to do this would require a greater expenditure of weapons to knock out the desired targets.

Local fallout with its high levels of gamma radiation can produce incapacitating injuries and fatalities within a few days. It does this by giving an acute whole body dose of ionizing radiation which causes changes throughout the body. Now the body can tolerate relatively large acute doses in a localized area without producing immediate deleterious

effects. However, a whole body dose of 1000 roentgens given in a short time would probably kill any human exposed to it. Whether or not an individual would receive a lethal dose in any particular situation depends upon many factors, such as his distance from the burst, the period of time he is exposed, and the use he makes of various shielding materials.

Table 24 summarizes the genetic dose and strontium-90 dose we have received from tests to date. Comparison is made between the levels we receive from natural radiation background, the amount we might receive from world-wide fallout in a nuclear war and the amount which the ICRP recommends as a maximum dose. It is apparent that the contribution from fallout to date is quite low, and that world-wide fallout in a nuclear war would be an insignificant factor to consider when compared to fallout in localities near the target areas.[28,29]

TABLE 24

Radiation Dosage in the United States

	Natural Background	Tests to Date	Possible War	ICRP MPD
GENETIC (whole body dose 30 yrs)	3 rem	0.05 rem	0.9 rem	15 rem
SOMATIC (Sr-90 dose to bone 70 yrs)	7 rem	0.4 rem	8.4 rem	24.5 rem

CHAPTER XII

CONCLUSIONS

General

The High Altitude Sampling Program has to date provided the most detailed and extensive study of radioactive material in the stratosphere carried on **anywhere** in the world. Over 100 million standard cubic feet of stratospheric air have been sampled. The program has fulfilled the task it set out to do, namely, to determine the role played by the stratosphere in the world-wide distribution of radioactive debris from low atmospheric weapons testing. The basic concept of the meridional network has been shown to be sound. Sampling in this network has provided inventories and distributions of material which help explain the major features of surface fallout noted by other programs both here and abroad. The U-2 can sample a major fraction of the stratosphere and good extrapolation may be made from balloon data up to 100,000 feet. The fate of injections made above 100,000 feet is still uncertain and can only be decided by continued sampling.

Sampler Efficiency

The most recent results of the studies of sampler efficiency and filter paper retentivity of stratospheric particles have tended to confirm the original conclusions that the volume of air sampled by each filter may be determined with an error of less than 20 percent and that virtually 100 percent of the radioactive debris is removed from the

sampled air by the filter. Stratospheric particles in one sample collected by impaction ranged in diameter from 0.01 to 0.05 microns while those in a second ranged from 0.05 to 1.2 microns with a geometric mean diameter of 0.26 microns.

Strontium-90

The distribution of strontium-90 in the stratosphere has been studied as a function of latitude, altitude and time. The concentrations in the Southern Stratosphere have been found to be between one-third and one-half those at equivalent latitudes and altitudes in the Northern Stratosphere. The concentrations increase upward from the tropopause but pass through a maximum, still within the lower stratosphere, and then decrease toward higher altitudes. The Tropical Stratosphere appears to be strongly stratified and to exhibit steep vertical concentration gradients. Changes in the distribution of strontium-90 in the stratosphere appear to result from diffusion phenomena which operate throughout the year but at rates which vary in response to processes of a seasonal character.

Tungsten-185

The specific activity of tungsten-185 in stratospheric air increases with altitude above the tropopause, passes through a maximum and then decreases toward higher altitudes in much the same way as do strontium-90 concentrations. However, the maximum concentration of tungsten-185 occurs 10,000 feet or so below the maximum strontium-90

concentration. The configuration of the zone of maximum tungsten-185 concentration suggests that meridional mixing occurs along surfaces which slope downward toward the poles and that vertical mixing is too slow to obliterate this zone even in the polar stratosphere. If this is so, the stratospheric residence time of debris should be strongly dependent upon the altitude of stabilization and therefore upon the total yield of the shot which produced it. Debris sampled at high altitudes in the Southern Polar Stratosphere should represent material from high yield shots almost exclusively and much of it should have been produced during CASTLE and REDWING. Debris sampled at high altitudes in the Northern Polar Stratosphere should represent a mixture of material from high yield United States shots and high yield Soviet shots.

Inventory

The stratospheric burden of strontium-90 during two time intervals has been calculated from HASP data, extrapolated upward to the top of the stratosphere through the use of data from Project ASHCAN. The mean burden in June 1958 was about 0.9 megacurie and in May 1959 it was about 0.8 megacurie. A preliminary calculation of the burden in the Northern Stratosphere during September through November 1959 gives 0.51 megacurie, compared with 0.66 megacurie for the same region in June 1958 and 0.57 megacurie in May 1959. The stratospheric burden of the Northern Hemisphere was more than twice that of the Southern Hemisphere during 1957. Comparison of the mid-1958 burden, which preceded the Soviet injection of about 1.2 megacuries of strontium-90 into the stratosphere in the

fall of 1958, with the early 1959 burden indicates a loss from the stratosphere of at least half the Soviet injection within 6 months. Calculations were made of the stratospheric tungsten-185 burden for two month intervals beginning with September 1958. They suggest a loss from the stratosphere of one-half the original tungsten-185 injection within 9 months. The results confirm the short stratospheric residence time of debris injected into the Polar Stratosphere or into the lower Tropical Stratosphere. A mathematical analysis of rates of diffusion through the tropopause indicates that of all the suggested mechanisms of loss of debris from the stratosphere only mass movement of stratospheric air through the tropopause gap into the troposphere can explain the short residence time of debris in the polar stratosphere. Surface concentrations in the Southern Hemisphere are about one-third those at comparable latitudes in the Northern Hemisphere.

Dose to Man

On the basis of the predicted future surface concentrations of strontium-90, it is concluded that the concentration of strontium-90 in newly formed bone will pass through a maximum in 1961 if the concentration in bone is primarily a function of cumulative fallout and has probably already passed if it is largely a function of the rate of fallout. All information thus far accumulated indicates that fallout constitutes a hazard to the human population which is small compared to the hazard from natural radiation.

Future Work

While the HASP program is moving toward the end of its weekly sampling schedule, spot checks will continue. There are several tasks upon which there will be continued work. The most pressing item is to resolve the Nose-Hatch flow rate discrepancies. It is hoped that this can be done by another intercalibration flight with instrumented ducts. Additional work will be undertaken at the Institute of Paper Chemistry in paper flow. It is hoped that absolute flow rates will be known to within 10% after this effort. In addition work will be carried on to improve ASHCAN and B-57 intercalibration.

Isotopes, Incorporated will continue radiochemical analysis. Neutron activation studies of 10 HASP samples will provide data on the chemical composition of debris.

Samples collected by means of impactor probes mounted on two HASP aircraft will be studied by electron microscopy as well as by light microscopy and autoradiography. These samples should provide a good picture of stratospheric particulates and may allow the quantitative estimation of the relative abundances of particles of various sizes and of the size distribution of radioactive particles. Investigations of the distribution of radioactive particles through HASP filters will also be continued to substantiate estimates of filter retentivity for stratospheric particulates.

A small number of soil samples will be collected at several sites in about three regions of contrasting climate and soil conditions for

analyses for the variation of concentrations of fission products and induced activities as a function of depth below the surface. The nuclides sought would be strontium-90, cesium-137, cerium-144, ruthenium-106 and plutonium. Such measurements would supply basic data for the interpretation of future hazards from fallout, the extent to which the various nuclides migrate downward through uptake by plant root systems, external gamma radiation of the human population and through inhalation of resuspended dust.

A small program of analysis of rainwater for tritium activity and of the carbon dioxide of surface air for carbon-14 activity will be carried out. At least two samples of tritium and one of carbon-14 will be analyzed each month. These data will indicate whether there is an increase in the rate at which gaseous nuclear debris reaches the ground during the spring of 1960. The correlation of the behaviour of gaseous debris with that of particulate debris will be instructive. Since tritium may be a good tracer for the debris introduced into the high stratosphere by the megaton weapons detonated over the Pacific in August 1958, any appreciable fallout of this debris during the spring of 1960 may cause a detectable increase in rainwater tritium concentrations in the Northern Hemisphere. This increase will be detected only if there is a regular monitoring of tritium levels in precipitation through the spring and summer of 1960. Similarly, a spring high in the fallout of particulate debris should be accompanied by an increase in tropospheric carbon-14 concentrations. A periodic sampling and measurement of tropospheric

carbon-14 will be required to detect such a rise. It is believed that the proposed measurements will contribute valuable information on the behaviour of this gaseous debris which will otherwise be lost.

A final study will be made of all HASP data. They will be tabulated in several ways to facilitate their use by others studying fallout. They will be analyzed in the light of all available theories on the behaviour of stratospheric fallout and the validity of these theories will be evaluated.

The analysis, which is being performed as part of the HASP, of data from other programs of fallout study will be completed. Comparison of HASP data will be made with data from Project ASHCAN studies of stratospheric fallout, from the measurements of stratospheric carbon-14 concentrations and from studies of surface fallout. Estimates of future biological hazards from fallout will be revised in the light of the most recent available data.

A final report will be prepared containing all data obtained during the HASP and all conclusions reached on the strength of these data.

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