

NRL Report 6104

**Summary Report on Fission Product Radioactivity
in the Air Along the 80th Meridian (West)
1957 - 1962**

L. B. LOCKHART, JR., R. L. PATTERSON, JR.,
A. W. SAUNDERS, JR., AND R. W. BLACK

*Physical Chemistry Branch
Chemistry Division*

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**U.S. NAVAL RESEARCH LABORATORY
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CONTENTS

Abstract	1
Problem Status	1
Authorization	1
INTRODUCTION	1
EXPERIMENTAL PROCEDURE	2
RESULTS	4
GROSS FISSION PRODUCT CONCENTRATIONS IN THE AIR	4
Radioactivity Profiles During 1962	4
Temporal Changes in Airborne Radioactivity, 1957-1962	7
RADIOCHEMICAL ANALYSES	9
Sr ⁹⁰ IN THE AIR AT GROUND LEVEL	9
SOURCES OF Sr ⁹⁰ OBSERVED IN THE GROUND-LEVEL AIR DURING 1962	15
ACTIVITY RATIOS	20
CONCLUSIONS	25
REFERENCES	26
APPENDIX A — Radioactivity Blanks	27
APPENDIX B — Estimation of Production Dates of Nuclear Debris and Their Sr ⁹⁰ Contributions to the Radioactivity of the Atmosphere	29

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Operation of the NRL 80th Meridian Air Sampling Program, 1956-1962, took place during an interesting period of the atomic age and the development of nuclear weaponry and covered the nearly three-year moratorium on nuclear testing as well as the periods of intensive testing of high yield devices both before and after the moratorium. Data collected during this program helped substantiate a number of concepts regarding atmospheric mixing processes and residence times of radioactive particulate matter in the atmosphere.

The current report summarizes measurements of both gross β activity and Sr^{90} activity in the air at ground level along the 80th meridian during the period 1957 through 1962. More detailed results and interpretations are given for the collections of radioactivity made in 1962.

Practically all of the gross β activity encountered in the air of the Northern Hemisphere during the early part of 1962 and 80-90% of the Sr^{90} activity resulted from debris produced in the USSR series of tests held in the fall of 1961. Beginning in the late summer a second large increment of fresh activity, primarily from the USSR tests (August-December 1962), intruded into the air at most of these sites.

In the Southern Hemisphere the source of the Sr^{90} activity was primarily pre-moratorium bomb debris from a stratospheric source until the initiation of testing by the United States at Christmas Island. From May throughout the remainder of 1962 these tests contributed the bulk of the Sr^{90} except in the southernmost areas of Chile, where there was a gradual rise in the contribution of the fresh component.

INTRODUCTION

The U.S. Naval Research Laboratory initiated a study of the radioactivity of the atmosphere as a result of its interest in the problem associated with the long-range detection of nuclear explosions. This early work, dating back to 1948, involved the development of methods for identifying and measuring radioactivity collected by rainfall and by air filters, and the operation of a network of radioactivity monitoring stations in the Northern Hemisphere which supplied information on both the fission products and natural radioisotopes in the atmosphere (1).

With the increasing interest in the use of airborne radioactivity as a tracer for atmospheric mixing processes, generated in part by the forthcoming International Geophysical Year, representatives of the U.S. Naval Research Laboratory, the U.S. Atomic Energy Commission, and the U.S. Weather Bureau in early 1956 considered the

feasibility and desirability of establishing a network of stations along the 80th Meridian (West) for making measurements of atmospheric radioactivity. At that time little data was available on radioactivity in the Southern Hemisphere and it was impossible to compare activity levels in the two hemispheres or to determine the extent of migration of fission products southward from Northern Hemisphere sources. It was decided to go ahead with the establishment of this network with the expectation that it would later become incorporated into the IGY program.

The final program was made possible by the technical assistance of the U.S. Weather Bureau, the financial support supplied by the Division of Biology and Medicine, U.S. Atomic Energy Commission, and the cooperation of the various agencies and organizations located in countries on or near the 80th meridian. During the IGY (1957-58) and IGC (1959), this network consisted of 21 stations extending from Punta Arenas, Chile to Thule, Greenland making filter collections of airborne radioactivity by the same procedure. During recent years the network was reduced to 13 stations covering the same range of latitude.

NRL Problem A02-13; Projects RR 004-02-42-5151 and AEC AT(49-7)-13. This is a final report on AEC Contract AT(49-7)-13; work on other phases of the problem is continuing. Manuscript submitted April 2, 1964.

A complete list of sites at which radioactivity collections were undertaken as part of this study together with the cooperating organization, the geographical location of the site and the period of operation is shown in Table 1.

In December 1962, the Naval Research Laboratory voluntarily terminated its association with this network and transferred direction to the Health and Safety Laboratory of the U.S. Atomic Energy Commission. The program is being continued under their auspices. Information obtained from the NRL program is contained in Refs. 2-9 and is briefly reviewed in this report, which also presents the radioactivity measurements made on the collections of 1962.

EXPERIMENTAL PROCEDURE

During the initial year of operation of the 80th-meridian network, parallel collections of airborne radioactivity were made by three different methods, namely, air filtration, fallout collection by gummed films, and collection on cloth screens. On the basis of this study it was decided that air filtration was the only technique which gave results that could be interpreted reasonably in terms of atmospheric mixing processes. Consequently, this method of sample collection was used exclusively during the IGY and later. During the IGY and IGC years, collections were made daily at each site, while in 1960 they were made on a three-per-week schedule, and in 1961 and 1962 they were made on a weekly schedule. The collection and analytical procedures were otherwise identical throughout the period July 1957-December 1962.

The sampling procedure involved drawing air continuously at a known rate (approximately 1200 cubic meters per day) through high-efficiency cellulose-asbestos filters 8 inches in diameter by the use of positive-displacement blowers. At the end of the sampling period, the filters, together with a notation as to the sampling schedule, were placed in envelopes and mailed to NRL for assay for gross β activity two weeks after collection. On receipt at NRL the samples were ashed at 650°C, and the ash from each sample compacted in an hydraulic press to a disk 1-1/8 inches in diameter, which was then counted under a thin-window flow-type GM tube in an automatic sample changer. The standard employed was $\text{Pb}^{210}(\text{Bi}^{210})$ similarly mounted with varying amounts of ashed filter

paper. The filter samples were weighed and compared with the corresponding standard.

The interpretation of gross β counts is complicated by the changing spectrum of β energy of fission products with age with a resulting continuous change in counting efficiency; moreover, the calibration standards employed introduce further uncertainties since none of them has a β spectrum which is comparable to that of a fission product mixture. Some of the complications involved have been considered in other studies (10,11). To enable this work to be interpreted by others in terms of their counting techniques, an attempt has been made to characterize the response of the NRL counting setup to β particles of different energy. Figure 1 shows graphically the experimentally determined efficiency corrections for various β emitters (Cs^{137} , $\text{Sr}^{90}(\text{Y}^{90})$, $\text{Pb}^{210}(\text{Bi}^{210})$, and KCl) incorporated into disks of compressed filter ash and makes possible a rough estimation of the changing efficiency with fission product mixtures of different ages.

Radiochemical analyses were performed at a later date on composited collections from each site through use of the procedure described in an earlier report (12). Basically, the radioisotopes included in the analytical scheme were Sr^{89} , $\text{Sr}^{90}(\text{Y}^{90})$, Y^{91} , Cs^{137} , Ce^{141} , $\text{Ce}^{144}(\text{Pr}^{144})$, Pm^{147} , and $\text{Pb}^{210}(\text{Bi}^{210})$; during part of 1958 and 1959, W^{185} was included in the scheme in place of $\text{Pb}^{210}(\text{Bi}^{210})$. The counting equipment was standardized by counting a variety of standards of known disintegration rates under the same conditions of geometry, backscattering, and self-absorption as were employed to measure the radiochemically separated isotopes.

Since, by the nature of the sample and the quantity of radioactivity present, analysis of aliquots or of duplicate samples was not possible special efforts were taken to see that the analytical results were as free as possible of controllable errors. This involved counting to a statistical accuracy of better than $\pm 1\%$ (standard deviation) where practicable and rechecking the count on different equipment, or on the same equipment after decay of several half-lives has occurred. In so far as possible all discontinuities in activity concentrations or activity ratios were resolved by recounting and reevaluation of the results.

TABLE 1
Collecting Sites Associated with NRL in the Study of Airborne
Fission Products Along the 80th Meridian

Station	Latitude	Longitude	Elevation (m)	Period of Operation	Cooperating Organization
Thule, Greenland	76°35'N	68°35'W	259	Apr 1958-present	U.S. Air Force
Coral Harbour, NWT, Canada	64°12'N	83°22'W	59	Mar 1958-Dec 1959	Meteorological Branch, Department of Transport, Canada
Moosonee, Ontario, Canada	51°16'N	80°39'W	10	Nov 1957-present	Meteorological Branch, Department of Transport, Canada
Bedford, Mass., USA	42°27'N	71°22'W	80	Jan 1958-Dec 1959	USAF Cambridge Research Center
Washington, D.C., USA*	38°59'N	77°29'W	82	May 1956-present	U.S. Weather Bureau
Columbia, S.C., USA	33°57'N	81°07'W	69	Jan 1958-Oct 1959	U.S. Weather Bureau
Miami, Fla., USA	25°49'N	80°17'W	4	June 1957-present	U.S. Weather Bureau
San Juan, Puerto Rico	18°26'N	66°00'W	10	June 1957-present	U.S. Weather Bureau
Miraflores, Panama Canal Zone	9°00'N	79°35'W	10	June 1956-present	Canal Zone Corrosion Laboratory of the U.S. Naval Research Laboratory
Bogota, Columbia	4°37'N	74°04'W	2640	July 1957-Dec 1959	Instituto Geofísico de los Andes Colombianos
Quito, Ecuador	0°08'S	78°26'W	2818	Apr 1957-Dec 1959	Observatorio Astronómico de Quito
Guayaquil, Ecuador	2°10'S	79°52'W	7	May 1956-present	Meteorological Office, Dirección General de Aviación Civil
Iquitos, Peru	3°45'S	73°11'W	117	Oct 1957-Dec 1959	Corporación Peruana de Aeropuertos y Aviación Comercial (CORPAC)
Lima, Peru	12°01'S	77°07'W	30	May 1956-present	Corporación Peruana de Aeropuertos y Aviación Comercial (CORPAC)
Huancayo, Peru	12°07'S	75°20'W	3353	Oct 1957-Dec 1959	Instituto Geofísico de Huancayo
Chacaltaya, Bolivia	17°10'S	68°15'W	5220	Sept 1957-present	Universidad Mayor de San Andrés, Laboratorio de Física Cosmica de Chacaltaya
Antofagasta, Chile	23°37'S	70°16'W	519	Nov 1957-present	NASA Satellite Tracking Station
Porto Alegre, Brazil	30°02'S	51°13'W	24	Dec 1957-Oct 1959	Servico de Meteorologia (Brazil)
Santiago, Chile	33°27'S	70°42'W	520	July 1956-present	Oficina Meteorologica de Chile
Puerto Montt, Chile	41°27'S	72°57'W	5	Dec 1957-present	Oficina Meteorologica de Chile
Punta Arenas, Chile	53°08'S	70°53'W	3	Oct 1956-present	Oficina Meteorologica de Chile

*Present location of station at Dulles International Airport.

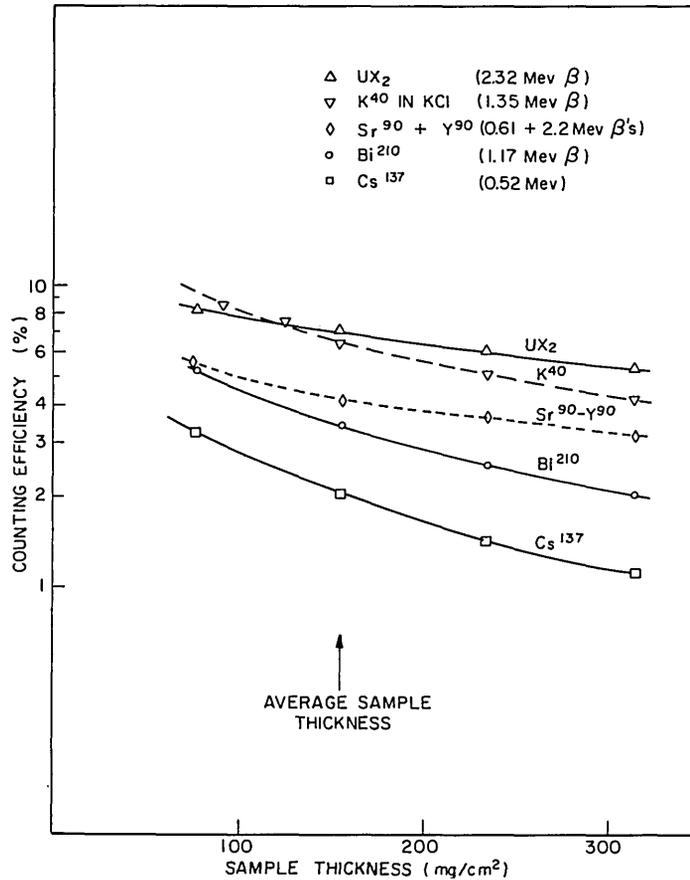


Fig. 1 - Counter efficiency as a function of β energy and sample thickness

RESULTS

The gross β activity and the concentrations of individual radioactive fission products in the ground-level air at the various sites along the 80th meridian prior to 1962 have been reported in earlier citations (2-9); the 1962 results are reported in detail here. In the graphical presentations the results cover the entire period of measurement so that the influence of bomb test schedules and of the seasonal variation in stratospheric deposition is more evident.

GROSS FISSION PRODUCT CONCENTRATIONS IN THE AIR

The monthly averages of the gross fission product concentrations in the ground-level air at the various sites along the 80th Meridian (West) during 1962 are shown in Table 2. Also included

with the results from these sites are measurements made at Mauna Loa, Hawaii, which is a high altitude site in the Northern Hemisphere roughly corresponding to Chacaltaya, Bolivia, in the Southern Hemisphere. These results may be compared with those of the U.S. Public Health Service Radiation Surveillance Network or those of the Air Sampling Program of the Radiation Protection Division, Canadian Department of National Health and Welfare by multiplying the 80th meridian results by the factors 0.58 and 0.87 respectively (11).

Radioactivity Profiles During 1962

Plots of the monthly average concentrations of fission products in the air during 1962 as a function of latitude are shown in Fig. 2. These profiles are similar to those observed in the past in that there are well-defined maxima in the midlatitudes

TABLE 2
Gross Fission Products in the Air at Ground Level Along the 80th Meridian (West) During 1962

Station	Activity (dis/min/m ³ of air)*											
	Jan	Feb	Mar	Apr	May	June	July	Aug	Sept	Oct	Nov	Dec
Thule, Greenland	11.1	9.4	16.0	10.7	5.6	7.4	5.2	2.8	2.3	4.7	6.8	9.3
Moosonee, Ontario, Canada	8.7	8.8	12.8	13.7	11.3	13.6	9.0	4.4	3.7	5.2	7.4	8.1
Washington, D.C.	15.4	14.4	14.2	17.4	14.0	15.4	12.6	8.4	9.0	11.0	15.1	14.3
Miami, Florida	17.9	16.3	20.0	28.5	16.9	4.8	4.7	2.8	3.0	11.4	31.0	18.4
Mauna Loa, Hawaii†	7.3	11.2	13.2	10.0	11.1	8.1	6.5	4.0	2.5	4.2	10.3	7.7
San Juan, Puerto Rico	11.2	15.3	17.6	—	8.2	5.3	5.1	3.4	2.4	1.43	9.8	11.6
Miraflores, Panama Canal Zone	8.2	5.6	6.3	6.9	4.1	0.69	1.32	1.04	0.34	0.37	4.0	3.7
Guayaquil, Ecuador	0.79	0.96	0.88	—	—	—	(0.78)	0.79	0.63	0.72	0.76	—
Lima, Peru	0.09	0.20	0.28	0.20	6.9	4.8	1.84	2.1	1.93	1.67	1.83	1.73
Chacaltaya, Bolivia	0.07	0.15	0.05	0.05	10.9	4.4	6.3	3.2	1.47	1.42	1.52	0.37
Antofagasta, Chile	0.21	0.17	0.22	0.12	3.7	4.0	3.2	2.5	1.66	1.09	1.17	1.27
Santiago, Chile	0.15	0.15	0.08	0.07	0.52	1.24	—	0.96	0.73	0.62	0.81	0.78
Puerto Montt, Chile	0.08	0.06	0.06	0.05	0.08	0.12	0.44	0.18	0.14	0.23	0.35	0.50
Punta Arenas, Chile	0.06	0.06	0.03	0.03	0.04	0.08	0.18	0.14	0.19	0.16	0.15	0.20

*Activity not corrected for decay; volume measured at station conditions.

†Station at 155°36'W longitude but operated in conjunction with 80th-meridian network.

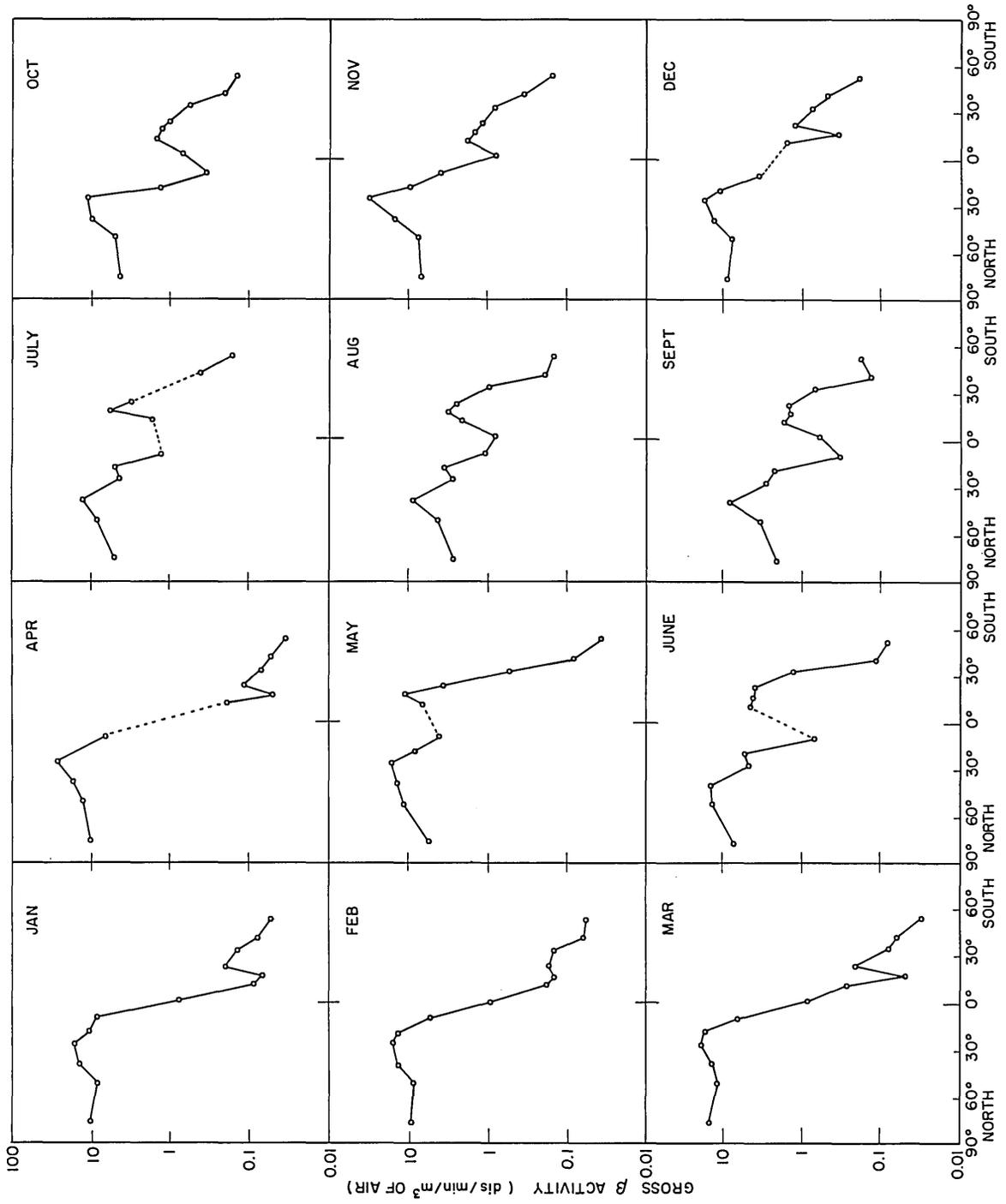


Fig. 2 — Profiles of the monthly average gross fission product concentrations in the air at ground level along the 80th Meridian (West) during 1962

of each hemisphere and a minimum near the Equator. Though there is some evidence of trans-equatorial transfer of radioactivity between hemispheres, both in the upper troposphere and in the stratosphere, there is no indication that a ground-level path exists. The quite large difference that developed in the air concentrations of fission products in the two hemispheres following resumption of nuclear testing by the Soviet Union in September 1961 with extremely large activity gradients just north of the Equator (a factor of 100 in about 30 degrees of latitude) existed until the U.S. nuclear tests in the Christmas Island area introduced debris more directly into the Southern Hemisphere. The high background of activity in the Northern Hemisphere masked the arrival of debris from Christmas Island; actually, as indicated in the radiochemical analyses presented in a later section, there is little evidence that any large quantity of radioactivity from the U.S. tests appeared in this area at ground level during this time. The Soviet tests of late 1962 further increased the activity in the Northern Hemisphere, while normal tropospheric processes of dispersal and deposition together with radioactive decay were serving to reduce the concentrations in the Southern Hemisphere. Here too, there was little evidence of debris from the latter part of the U.S. Christmas Island series of tests.

The introduction of several extensive sources of fresh radioactivity into the atmosphere during 1962 prevented observation of the usual seasonal migrations of the equatorial minimum toward the summer (and fall) hemisphere and of the mid-latitude maxima poleward in the late spring and summer.

Temporal Changes in Airborne Radioactivity, 1957-1962

The monthly average concentrations of gross fission products in the air at a number of the 80th-meridian sites during the period 1957 through 1962 are shown in Figs. 3 and 4. The major peaks of activity corresponding to various episodes of nuclear testing are evident; unfortunately, the linear scale does not have the sensitivity to permit the observation of a number of interesting effects that took place between tests programs. These latter effects, which are the direct result of seasonal changes in the radioactivity distribution, can be seen more clearly in Fig. 5

where air concentrations of fission products at Miami and Antofagasta are displayed on a logarithmic scale, and in a later section where the results of radiochemical analyses are presented.

In 1957, the Northern Hemisphere sites were intercepting radioactivity from U.S. tests in Nevada (PLUMBBOB, May through October) and from tests in the Soviet Union (January through April, and August through December). Below the Equator, there was a little fission product radioactivity in the atmosphere, most of which was accounted for by the U.K. tests in Australia (September and October) and at Christmas Island (May, June, and November). There was a background of older debris here which was relatively rich in the longer lived fission products and which may be attributed to the delayed appearance (stratospheric fallout) of material from the U.S. 1954 Castle or 1956 Redwing series at the Pacific Proving Ground (4).

As the pace of nuclear testing, particularly of high-yield devices, became intensified in 1958 with major series of tests by the United States, the United Kingdom and the USSR (two series), the concentration of artificial radioactivity in the atmosphere reached record levels in both hemispheres. While the bulk of the activity in the Southern Hemisphere could be attributed to the U.S. HARDTACK tests (and the U.K. GRAPPLE tests), the Northern Hemisphere loadings were principally due to the USSR tests.

The rapid decrease in the concentration of fission products in the ground-level air in the year following the 1958 moratorium on nuclear testing is readily apparent. The peak of activity was actually reached in the spring of 1959, nearly six months following the last nuclear test in 1958, after which time the air concentration decreased with an apparent half-life of less than 30 days for a period of several months. The tropospheric burdens of activity in the Northern Hemisphere (obtained by integrating the air concentrations over the hemisphere and assuming a uniform volume concentration up to the tropopause) decreased from a maximum of over 22 megacuries (MCi) in April 1959 to about 0.3 MCi in October 1959; a further, slower decrease took place to a minimum of less than 0.10 MCi in August 1961, just prior to the renewal of large scale atmospheric testing.

Radioactivity from the first French atomic test (February 13, 1960) was clearly detectable at

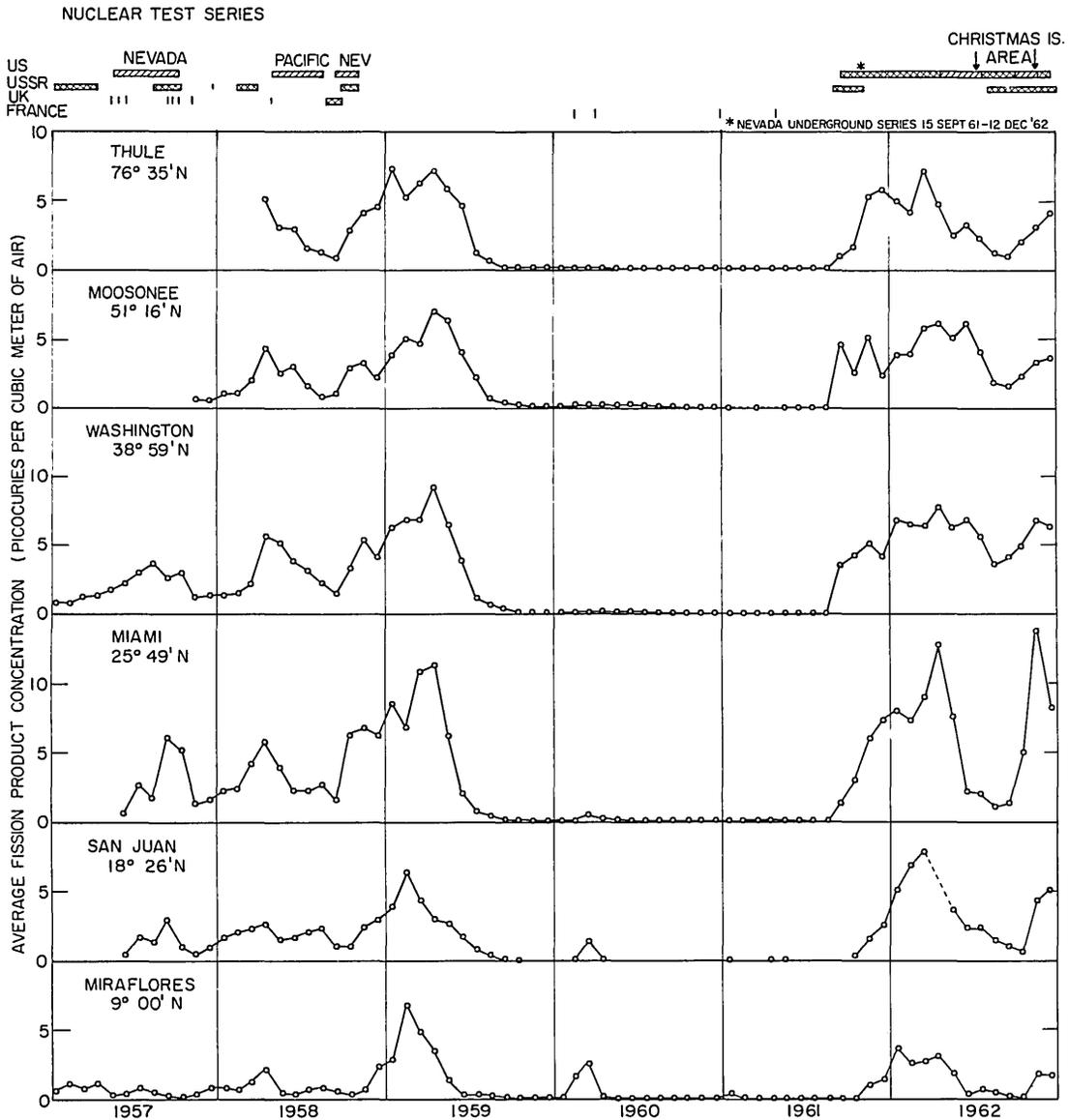


Fig. 3 — Fission product radioactivity in the air of the Northern Hemisphere, 1957-1962

several sites along the 80th meridian (Miami to Guayaquil) as indicated in Figs. 3 and 4. Radiochemical results previously reported (8) indicated this fresh debris eventually extended from Thule, Greenland to Antofagasta, Chile; however, changes produced in the gross activity of the air by this test were generally masked by the background of old debris in the Northern Hemisphere which was at that time experiencing its normal spring increase. The third French test (December 27, 1960) showed up only as a small spike

in the gross activity concentrations at Miraflores (9°N) in January 1961. None of the other reported French tests were identified.

With the resumption of atmospheric testing of large yield nuclear devices by the Soviet Union in September 1961, followed by U.S. tests in the Christmas Island area of the Pacific (April 25-November 4, 1962), and further testing by the USSR (August 5-December 24, 1962), the pattern of activity in both hemispheres returned to that existing in the 1958-1959 period. As before, the

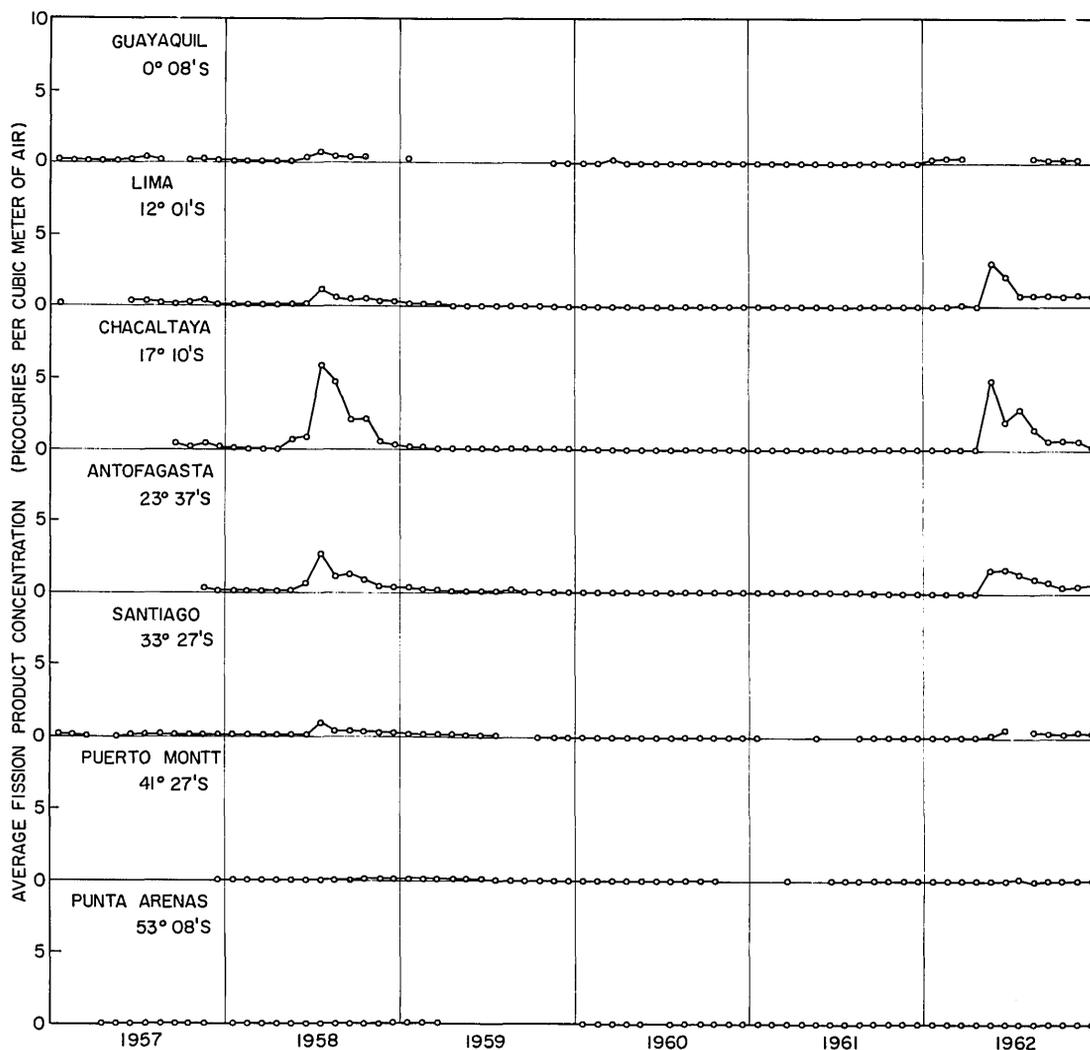


Fig. 4 — Fission product radioactivity in the air of the Southern Hemisphere, 1957-1962

gross activity levels in the Northern Hemisphere were directly related to the rapid appearance at ground level of debris introduced into the atmosphere by the Soviet Union, while those in the Southern Hemisphere were due to the testing taking place in the tropics (U.S. tests at Christmas Island). The smaller extent of this latter contamination suggests that the tropospheric component of the debris from the U.S. tests was not large and that the season and site of injection has resulted in the delayed appearance of stratospheric radioactivity at ground level.

RADIOCHEMICAL ANALYSES

The results of the radiochemical analyses of composited bimonthly samples from each

of the 80th-meridian stations (except San Juan) in operation during 1962 are presented in Table 3 in units of disintegrations/minute per 100 standard cubic meters of air. Results from the Mauna Loa station are also included. All radioactivity concentrations have been corrected for decay to the midpoint of the collection period. Some activity ratios of current interest are also presented. The background radioactivity of the filter materials was determined as described in Appendix A and found to be generally insignificant.

Sr^{90} IN THE AIR AT GROUND LEVEL

As a result of the number of large yield nuclear tests carried out by the Soviet Union in

TABLE 3
Radiochemical Analyses of Composite Bimonthly Air-Filter Collections During 1962

Month	Days§ Sampled	Gross β	Activity (dis/min per 100 std. cubic meters of air)†								Activity Ratios				
			Ce ¹⁴¹	Sr ⁸⁹	Y ⁹¹	Ce ¹⁴⁴	Pm ¹⁴⁷	Sr ⁹⁰	Cs ¹³⁷	Pb ²¹⁰	Ce ¹⁴¹ / Ce ¹⁴⁴	Sr ⁸⁹ / Sr ⁹⁰	Ce ¹⁴⁴ / Pm ¹⁴⁷	Ce ¹⁴⁴ / Sr ⁹⁰	Cs ¹³⁷ / Sr ⁹⁰
Thule, Greenland - Lat. 76°35'N Long. 68°35'W Elev. 259 m															
Jan-Feb	49	1022	(65)	145	239	129	21.0	3.71	5.58	0.249	(0.5)	39.1	6.1	34.8	1.50
Mar-Apr	63	1335	*	120	205	195	30.0	6.61	10.6	1.74	-	18.2	6.5	29.5	1.60
May-June	56	651	*	42.2	69.2	115	22.3	5.02	8.44	1.02	-	8.4	5.2	22.9	1.68
July-Aug	63	399	*	15.7	26.6	73.6	18.7	3.32	6.09	0.764	-	4.7	3.9	22.2	1.83
Sept-Oct	63	349	(57)	23.9	40.3	45.1	15.2	1.71	2.79	0.645	(1.3)	14.0	3.0	26.4	1.63
Nov-Dec	60	806	(79)	94.1	125	85.5	23.2	2.88	4.23	0.950	(0.9)	32.7	3.7	29.7	1.47
Moosonee, Ontario, Canada - Lat. 51°16'N Long. 80°39'W Elev. 10 m															
Jan-Feb	57	873	(61)	111	201	105	14.8	2.97	3.64	1.55	(0.6)	37.4	7.1	35.4	1.23
Mar-Apr	63	1325	*	113	164	173	26.9	5.86	6.65	1.91	-	19.3	6.4	29.5	1.13
May-June	56	1245	*	66.0	107	168	32.1	7.26	10.4	1.30	-	9.1	5.2	23.1	1.43
July-Aug	63	670	(7.7)	28.7	53.8	113	27.7	5.13	8.99	0.366	(0.07)	5.6	4.1	22.0	1.75
Sept-Oct	63	443	(40)	28.8	46.5	50.9	14.0	2.01	3.51	1.11	(0.8)	14.3	3.6	25.3	1.75
Nov-Dec	63	775	145‡	83.2	114	66.0	14.0	2.09	3.52	2.08	2.2‡	39.8	4.7	31.6	1.68
Washington, D.C. - Lat. 38°59'N Long. 77°29'W Elev. 82 m															
Jan-Feb	63	1490	(255)	186	342	162	24.7	4.43	8.70	0.605	(1.6)	42.0	6.6	36.6	1.96
Mar-Apr	63	1580	*	145	221	225	34.2	7.83	-	1.81	-	18.5	6.6	28.7	-
May-June	56	1470	(134)	89.6	141	231	61.9	8.86	14.9	3.23	(0.6)	10.1	3.7	26.1	1.68
July-Aug	56	1050	(52)	48.9	86.9	187	49.2	9.01	14.3	1.15	(0.3)	5.4	3.8	20.8	1.59
Sept-Oct	63	999	(32)	84.8	102	101	26.1	3.58	6.23	3.64	(0.3)	23.7	3.9	28.2	1.74
Nov-Dec	63	1470	(107)	154	201	111	27.0	3.45	5.69	4.39	(1.0)	44.6	4.1	32.2	1.65
Miami, Florida - Lat. 25°49'N Long. 80°17'W Elev. 4 m															
Jan-Feb	63	1710	(234)	196	409	181	27.7	5.22	3.94	0.491	(1.3)	37.5	6.5	34.7	0.75
Mar-Apr	63	2425	*	209	401	288	54.0	11.0	11.2	0.592	-	19.0	5.3	26.2	1.02
May-June	56	1084	*	68.8	122	169	38.4	7.31	6.39	1.21	-	9.4	4.4	23.1	0.87
July-Aug	63	373	*	19.0	34.0	62.3	16.2	2.47	2.94	0.965	-	7.7	3.8	25.2	1.19
Sept-Oct	63	722	(42)	67.8	118	72.7	15.1	2.43	2.10	1.05	(0.6)	27.9	4.8	29.9	0.86
Nov-Dec	63	2470	425‡	244	326	170	40.1	5.71	5.66	0.760	2.5‡	42.7	4.2	29.8	0.99
Mauna Loa, Hawaii - Lat. 19°28'N Long. 155°36'W Elev. 3394 m															
Jan-Feb	63	1290	(170)	201	254	125	23.7	4.88	7.93	0.573	(1.4)	41.2	5.3	25.6	1.63
Mar-Apr	63	1620	*	174	294	249	42.7	8.73	14.8	1.88	-	19.9	5.8	28.5	1.70
May-June	56	1340	*	98.1	142	197	47.4	8.14	14.5	1.59	-	12.1	4.2	24.2	1.78
July-Aug	53	734	*	54.2	86.9	114	30.5	4.33	8.22	1.68	-	12.5	3.7	26.3	1.90
Sept-Oct	56	470	*	44.9	54.0	45.4	10.9	2.04	2.90	0.908	-	22.0	4.2	22.3	1.42
Nov-Dec	54	1260	(226)	164	198	106	26.1	3.90	5.70	1.34	(2.1)	42.1	4.1	27.2	1.46
Miraflores, Panama Canal Zone - Lat. 9°00'N Long. 79°35'W Elev. 10 m															
Jan-Feb	63	690	*	91.9	148	83.5	13.8	2.33	2.63	0.089‡	-	39.4	6.1	35.8	1.13
Mar-Apr	63	662	*	64.7	102	101	16.3	3.42	4.07	0.266	-	18.9	6.2	29.5	1.19
May-June	56	238	*	21.8	26.1	33.8	8.63	1.37	2.10	0.600	-	15.9	3.9	24.7	1.53
July-Aug	62	118	*	7.4‡	12.8	19.8	4.00	0.684	1.33	0.648	-	10.8‡	5.0	28.9	1.94
Sept-Oct	60	35	(2.1)	4.22	5.12	5.09	1.47	0.182	0.313	0.692	(0.4)	23.2	3.5	28.0	1.72
Nov-Dec	63	383	(30)	49.7	60.8	35.2	10.9	1.05	1.57	0.182	(0.9)	47.3	3.2	33.5	1.50
Guayaquil, Ecuador - Lat. 2°10'S Long. 79°52'W Elev. 7 m															
Jan-Feb	56	87	13.6‡	9.24	11.6	7.62	2.32	0.326	0.799	2.03	1.8‡	28.3	3.3	23.4	2.45
Mar	35	88	(5.6)	7.52	9.30	8.88	2.93	0.418	1.006	2.36	(0.6)	18.0	3.0	21.2	2.41
Aug	39	79	(14)	6.26	11.4	8.70	2.77	0.212	0.425	2.89	(1.6)	29.5	3.1	41.0	2.00
Sept-Oct	56	68	(6.3)	5.52	8.64	9.07	2.74	0.410	0.669	2.42	(0.7)	13.5	3.3	22.1	1.63
Nov	21	76	(22.3)	5.37	8.21	8.83	2.70	0.447	0.806	1.79	(2.5)	12.0	3.3	19.8	1.80

*Activity not detectable at time of analysis.

†All activities (except gross β) corrected for decay to midpoint of collection period. Counting error less than $\pm 2\%$ (standard deviation) unless otherwise indicated.

‡Counting error estimated to be less than 10% (standard deviation).

§Samples collected weekly but grouped by months.

()Value uncertain.

TABLE 3 (Continued)
Radiochemical Analyses of Composite Bimonthly Air-Filter Collections During 1962

Month	Days§ Sampled	Gross β	Activity (dis/min per 100 std. cubic meters of air)†								Activity Ratios				
			Ce ¹⁴¹	Sr ⁸⁹	Y ⁹¹	Ce ¹⁴⁴	Pm ¹⁴⁷	Sr ⁹⁰	Cs ¹³⁷	Pb ²¹⁰	Ce ¹⁴¹ / Ce ¹⁴⁴	Sr ⁸⁹ / Sr ⁹⁰	Ce ¹⁴⁴ / Pm ¹⁴⁷	Ce ¹⁴⁴ / Sr ⁹⁰	Cs ¹³⁷ / Sr ⁹⁰
Lima, Peru - Lat. 12°01'S Long. 77°07'W Elev. 30 m															
Jan-Feb	55	15	(0.6)	0.79	0.965	0.963	0.604	0.226	0.557	0.646	(0.6)	3.5	1.6	4.3	2.46
Mar-Apr	63	24	(1.1)	1.35	1.60	2.30	1.46	0.505	1.095	0.902	(0.5)	2.7	1.6	4.6	2.17
May-June	63	585	134 ‡	49.5	64.5	26.9	4.65	0.706	1.226	1.00	5.0 ‡	70.1	5.8	38.1	1.74
July-Aug	63	195	42 ‡	13.5	27.5	17.6	3.39	0.554	0.852	0.626	2.4 ‡	24.4	5.2	31.8	1.54
Sept-Oct	56	180	(13.8)	12.4	22.8	24.9	6.53	1.050	1.50	0.391	(0.6)	11.8	3.8	23.7	1.43
Nov-Dec	49	178	(16.9)	14.2	22.2	27.5	8.36	1.51	2.22	0.599	(0.6)	9.4	3.3	18.2	1.47
Chacaltaya, Bolivia - Lat. 17°10'S, Long. 68°15'W Elev. 5220 m															
Jan-Feb	56	19	(1.6)	2.15	2.28	1.57	0.673	0.194	0.470	1.66	(1.0)	11.1	2.3	8.1	2.42
Mar-Apr	59	9	(0.31)	-	0.589	0.852	0.520	-	0.374	1.76	(0.4)	-	1.6	-	-
May-June	63	1310	299 ‡	119	138	48.5	9.67	1.245	2.03	2.62	6.2 ‡	95.6	5.0	39.0	1.63
July-Aug	42	808	(125)	54.7	105	72.2	14.1	1.38	1.99	2.94	(1.7)	39.6	5.1	52.3	1.44
Sept-Oct	50	247	(21)	26.5	41.9	37.8	10.2	1.162	1.75	3.08	(0.6)	22.8	3.7	32.5	1.51
Nov-Dec	63	161	(15)	13.2	18.8	23.3	7.73	1.022	1.61	1.97	(0.6)	12.9	3.0	22.8	1.58
Antofagasta, Chile - Lat. 23°37'S Long. 70°16'W Elev. 519 m															
Jan-Feb	49	20	1.13 ‡	1.00 ‡	1.19	1.59	1.31	0.544	1.092	1.28	0.7 ‡	1.8 ‡	1.2	2.9	2.01
Mar-Apr	60	18	(0.8)	0.82 ‡	1.04	1.54	1.09	0.422	1.022	1.55	(0.5)	1.9 ‡	1.4	3.6	2.42
May-June	56	411	107 ‡	45.9	49.9	15.8	3.94	0.656	1.14	2.60	6.8 ‡	70.0	4.0	24.1	1.74
July-Aug	63	299	(43)	21.2	44.1	27.9	6.31	0.945	1.52	2.09	(1.5)	22.4	4.4	29.5	1.61
Sept-Oct	56	146	(11)	8.96	16.8	18.4	6.17	0.847	1.157	0.510	(0.6)	10.6	3.0	21.7	1.37
Nov-Dec	63	129	(11)	9.24	15.5	20.8	7.11	1.110	1.75	0.588	(0.5)	8.3	2.9	18.7	1.58
Santiago, Chile - Lat. 33°27'S Long. 70°42'W Elev. 520 m															
Jan-Feb	48	16	(0.6)	(0.58)	(0.33)	1.28	1.59	0.713	1.44	1.55	(0.5)	(0.8)	0.8	1.8	2.02
Mar-Apr	63	8	*	(0.16)	(0.23)	0.707	0.856	0.361	0.856	1.64	-	(0.4)	0.8	2.0	2.37
May-June	42	93	32.5 ‡	11.6	15.6	6.36	2.28	0.351	0.886	3.83	5.1 ‡	33.0	2.8	18.1	2.52
July-Aug	Insufficient samples for analysis														
Sept-Oct	49	72	(9.1)	4.54	7.87	8.63	3.07	0.543	1.046	1.36	(1.1)	8.4	2.8	15.9	1.93
Nov-Dec	56	84	(10)	4.83	8.57	12.0	3.65	0.676	1.41	1.14	(0.8)	7.1	3.3	17.8	2.09
Puerto Montt, Chile - Lat. 41°27'S Long. 72°57'W Elev. 5 m															
Jan-Feb	56	7	(0.8)	1.58 ‡	0.27 ‡	0.878	1.04	0.477	0.812	0.187	(0.9)	3.3 ‡	0.8	1.8	1.70
Mar-Apr	56	6	(0.13)	(0.19)	(0.14)	0.735	0.923	0.477	0.831	0.368	(0.2)	(0.4)	0.8	1.5	1.74
May-June	63	10	2.5 ‡	1.79	1.73	0.919	0.711	0.235	0.351	0.644	2.7 ‡	7.6	1.3	3.9	1.49
July-Aug	63	31	7.5 ‡	4.07	5.76	3.59	1.79	0.307	0.391	0.654	2.1 ‡	13.3	2.0	11.7	1.27
Sept-Oct	56	19	2.9 ‡	2.33	3.82	3.79	1.33	0.326	0.383	0.410	0.8 ‡	7.1	2.8	11.6	1.17
Nov-Dec	49	42	(2.5)	2.90	5.16	7.41	2.12	0.516	0.735	0.239	(0.3)	5.6	3.5	14.4	1.42
Punta Arenas, Chile - Lat. 53°08'S Long. 70°53'W Elev. 3 m															
Jan-Feb	48	6	(0.51)	(0.05)	(0.06)	0.485	0.710	0.313	0.683	0.289	(1.1)	(0.2)	0.7	1.5	2.18
Mar-Apr	63	3	(0.08)	(0.04)	(0.06)	0.334	0.450	0.228	0.550	0.183	(0.2)	(0.2)	0.7	1.5	2.41
May-June	56	6	1.39 ‡	0.85 ‡	1.09	0.597	0.520	0.147	0.414	0.359	2.3 ‡	5.8 ‡	1.1	4.1	2.82
July-Aug	63	16	2.47 ‡	1.33 ‡	2.30	1.65	0.855	0.145	0.363	0.516	1.5 ‡	9.2 ‡	1.9	11.4	2.50
Sept-Oct	49	17	(1.8)	1.35 ‡	2.53	2.70	0.873	0.190	0.388	0.249	(0.7)	7.1 ‡	3.1	14.2	2.04
Nov-Dec	56	17	(0.6)	1.27 ‡	1.63	2.55	0.914	0.194	0.426	0.130	(0.2)	6.5 ‡	2.8	13.1	2.20

*Activity not detectable at time of analysis.

†All activities (except gross β) corrected for decay to midpoint of collection period. Counting error less than $\pm 2\%$ (standard deviation) unless otherwise indicated.

‡Counting error estimated to be less than 10% (standard deviation).

§Samples collected weekly but grouped by months.

()Value uncertain.

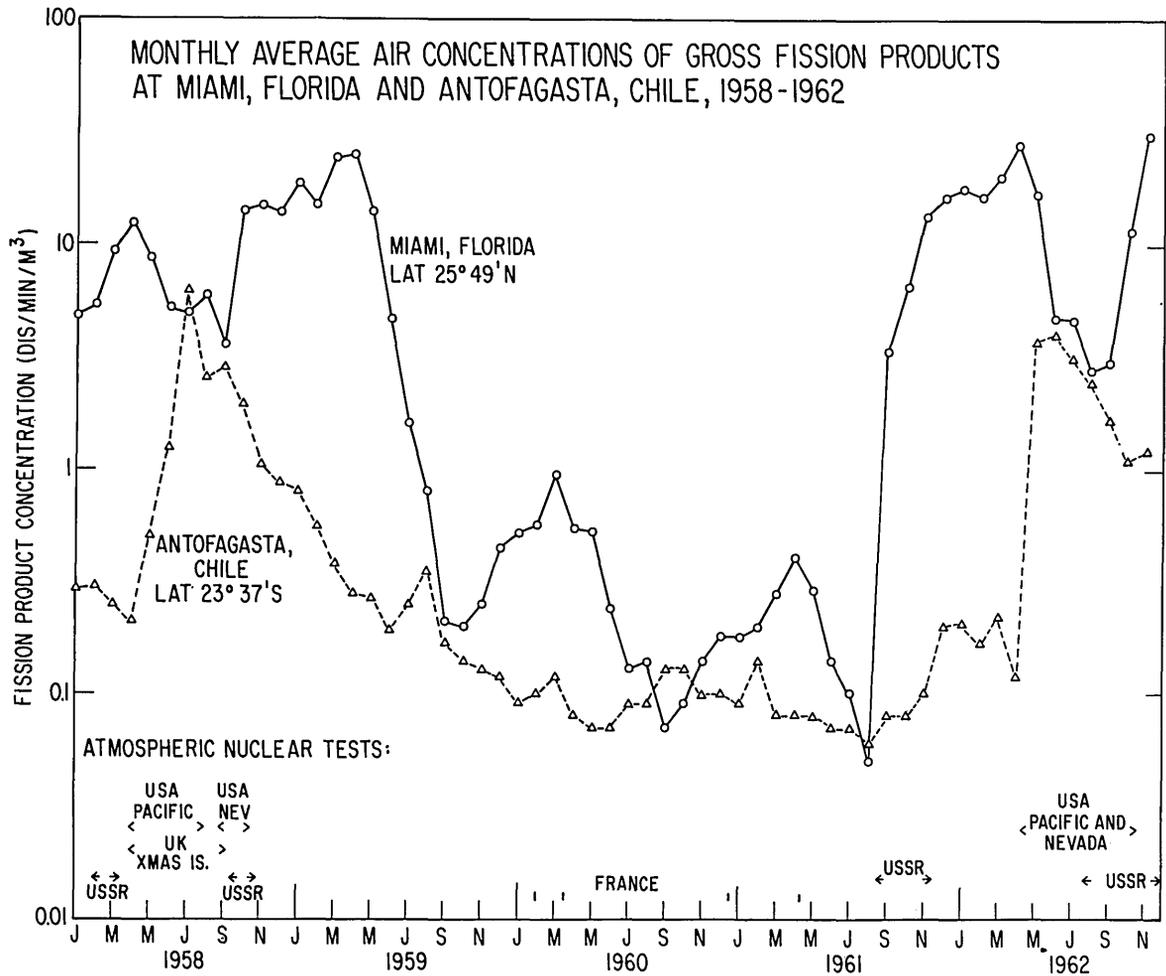


Fig. 5 — Monthly average air concentrations of gross fission products at Miami, Florida, and Antofagasta, Chile, 1958-1962

1961, the concentration of Sr^{90} in the ground-level air throughout the Northern Hemisphere during the spring of 1962 approached levels approximating those reached in 1959 following the extensive series of tests held in 1958. As will be shown later, this 1962 spring maximum contained only a minor contribution of activity from the U.S. Christmas Island tests which began in late April 1962. On the basis of total yield alone, the 1962 maximum should have been higher than 1958; however, debris from the largest of the Soviet explosions, estimated to have had total yields of 25 and 55-60 megatons (13), evidently rose sufficiently high that they were not appreciably involved in the 1962 winter-spring subsidence of stratospheric air.

The concentrations of Sr^{90} that have been encountered in the ground-level air at a number of sites in the Northern and Southern Hemispheres during the course of this study are shown graphically on a logarithmic scale in Figs. 6 and 7, respectively. Blank areas in the graphs represent periods when no analyses were performed on samples from that site; the dotted portions indicate either lack of receipt of samples or an analytical disaster which prevented the desired data being obtained. Starting in 1957, monthly samples from each site were analyzed; this schedule, however, overtaxed the available facilities and it became evident by July 1958 that this schedule could not be maintained. Accordingly, only a few analyses were performed on samples

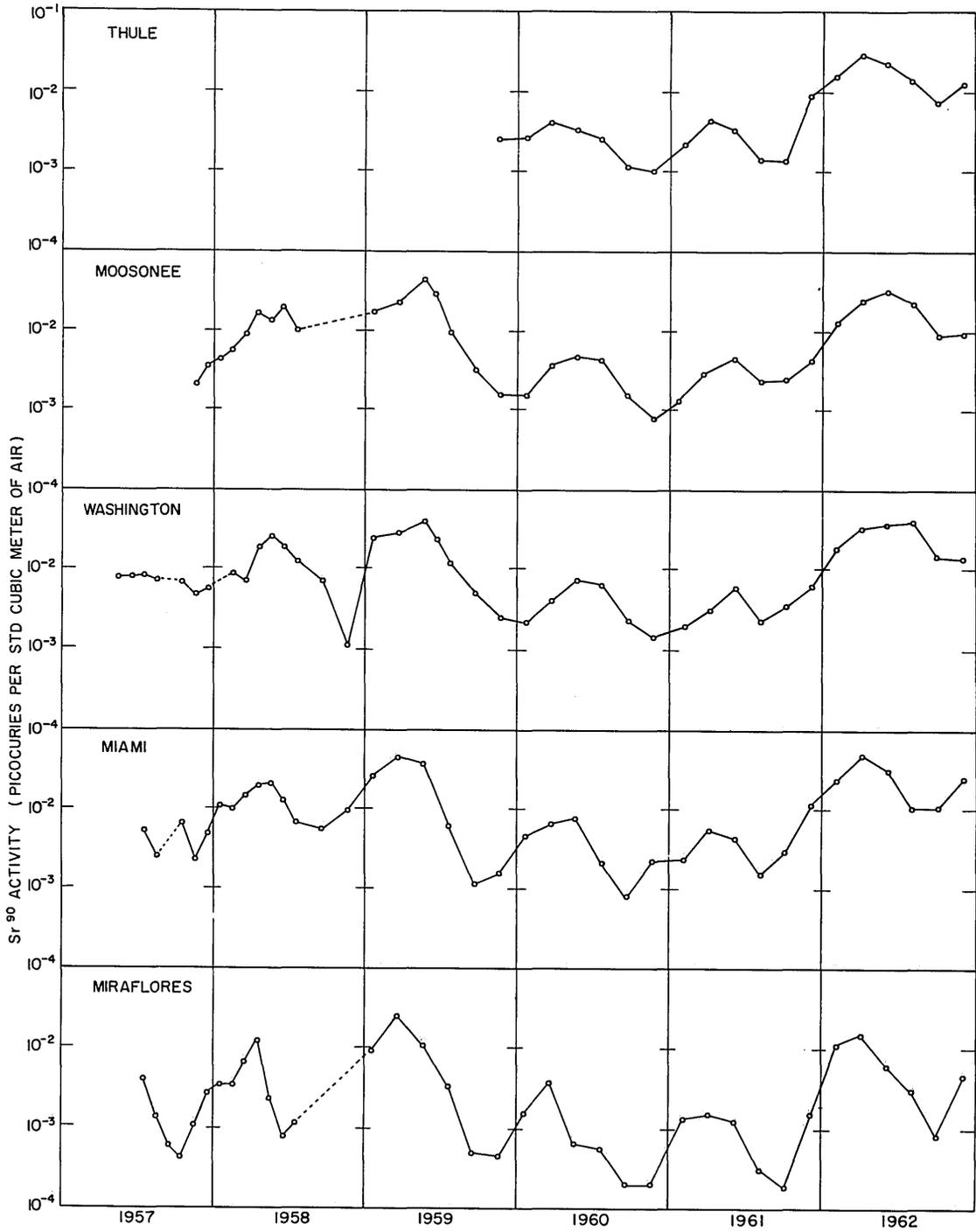


Fig. 6 — Concentrations of Sr⁹⁰ in the air at selected sites in the Northern Hemisphere along the 80th Meridian (West), 1957-1962

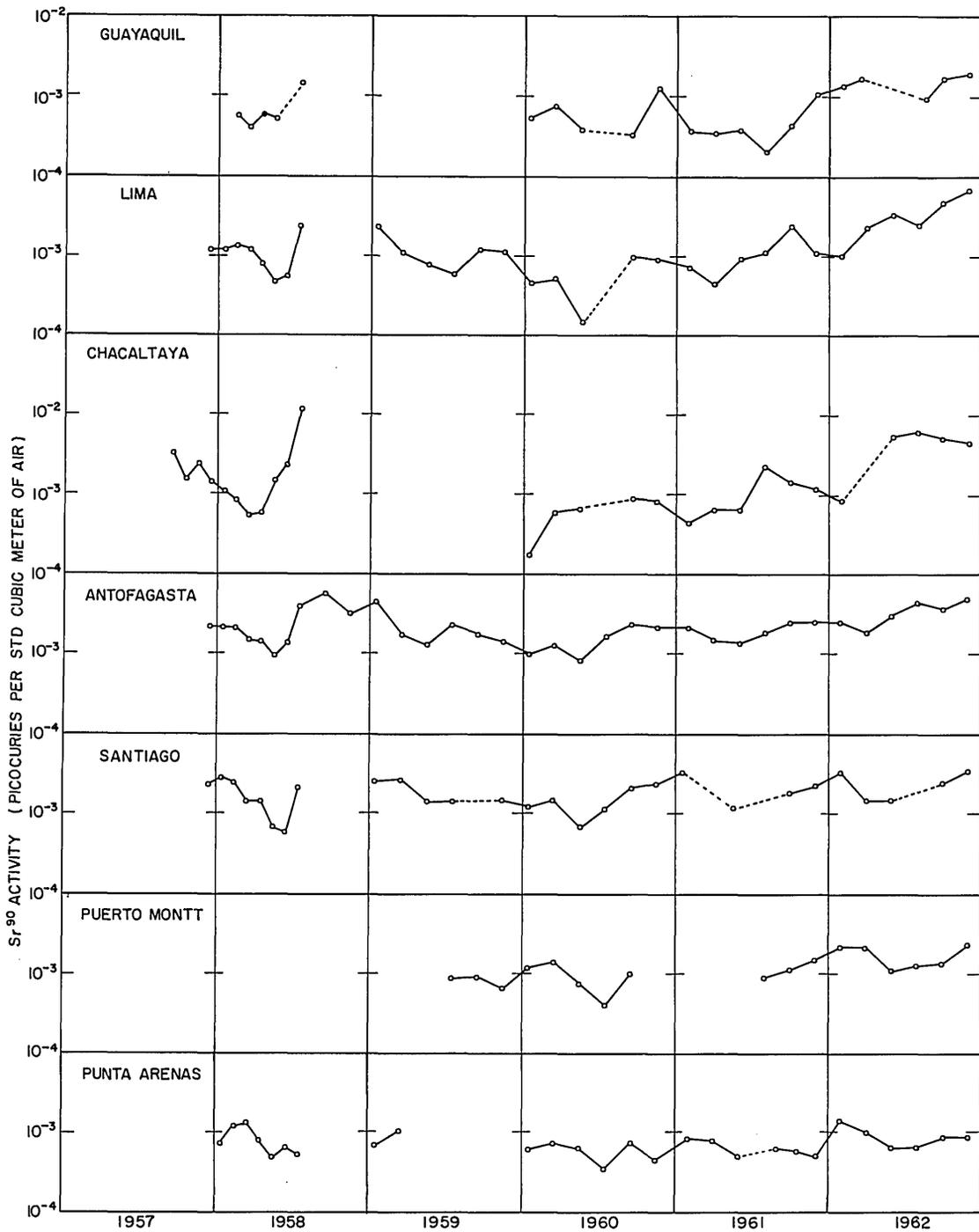


Fig. 7 - Concentrations of Sr⁹⁰ in the air at selected sites in the Southern Hemisphere along the 80th Meridian (West), 1957-1962

collected in the last half of 1958 and, beginning in January 1959, composited samples from alternate months were analyzed. During 1961 and 1962, composited bimonthly samples from the various sites were analyzed radiochemically.

In the Northern Hemisphere, a definite seasonal effect with a maximum each spring or early summer is evident at all sites regardless of the previous schedule of nuclear testing. Moreover, the time of the arrival of the radioactivity maximum varies with the latitude, arriving latest in the midlatitudes. During the 1959-1961 period when the major source of radioactivity was the stratosphere, the amplitude of the cycle ranged from about 3 (ratio of maximum to minimum) at Thule to about 8 at Miami; it was even greater at Miraflores, Panama, where the dry season coincided with the period of highest air concentrations of radioactivity. If the rate of removal of Sr^{90} by rainfall can be considered to be a constant regardless of season, it is evident that the rate of replenishment of the radioactivity in the surface air must approximate the deposition rate (50% in 4 to 5 weeks) at the time of the spring peak and, for several months prior to this peak, the replenishment rate must be doubling about every 3 weeks.

In the Southern Hemisphere, the data is less complete but it is obvious that the seasonal variation is much less pronounced. Transequatorial leakage of some of the radioactivity from the Northern Hemisphere conceivably could cause a reduction in the magnitude of the observed yearly cycle; information presented later (Table 4) would indicate that the tropospheric component of the radioactivity crossing the equator is not sufficient to account for all the depression in the amplitude of the cycle. While it is possible that the meteorology of the two hemispheres is sufficiently distinct to account for the observed differences in the amplitudes, it seems more plausible that the cause is related to the unsymmetrical insertion of activity into the upper atmosphere. Less material is available to the antarctic region to undergo downward mixing in its spring season so that the nonseasonal mixing processes cause an apparent damping of the amplitude of the cycle. The lack of symmetry of the stratospheric source in 1961 after some 2-1/2 years of moratorium on nuclear testing is suggested by the much higher activity concentrations at Moosonee and Washington than at comparable

stations in southern Chile, and by the much greater decrease in activity with increasing latitude in the Southern Hemisphere.

The imbalance in the stratospheric inventories of radioactivity over the two hemispheres at the end of atmospheric nuclear testing in December 1962 was even greater than existed at the end of 1958. The pattern of Sr^{90} concentrations in the air to be expected during the coming years is for the high levels observed in the Northern Hemisphere in 1963 (14,15) to be followed by a large decrease in 1964 and by smaller changes thereafter. In the Southern Hemisphere, transequatorial mixing should predominate over deposition for several years so that the air concentrations observed should actually increase slightly before going into a slow decline. Moreover, there should be an increasing amplitude to the seasonal cycle here as stratospherical-held debris migrate from the tropical to the midlatitude or polar regions when spring subsidence is more intense. In time, after depletion of the excess radioactivity in the arctic stratosphere, the seasonal variations in the two hemispheres should become similar in amplitude but out of phase, unless the meteorological regimes north and south of the equator are fundamentally different.

SOURCES OF Sr^{90} OBSERVED IN THE GROUND-LEVEL AIR DURING 1962

The bimonthly Y^{91} , Ce^{144} , and Sr^{90} radiochemical data has been analyzed, as described in Appendix B, to permit assignment of reasonable proportions of the Sr^{90} content of the ground-level air at the various sites to the different nuclear test series. Since the available data is suitable only for the evaluation of two component mixtures, some rather arbitrary assumptions have been made: (a) that only inconsequential equatorial crossover of debris from the USSR tests of 1961 and 1962 occurred; (b) that all fresh debris in the Northern Hemisphere was due to the USSR tests; and (c) that the residual pre-moratorium Sr^{90} in the Northern Hemisphere during the period September through December 1962 could be reasonably represented by extrapolation of 1960 and 1961 levels.

The results of these assignments and the indicated production dates of the fresher debris are listed in Table 4. If an average production

TABLE 4
Sr⁹⁰ Contributions of Various Sources to the Radioactivity of the Atmosphere and the Apparent Production Date of Debris from the Youngest Source

	Jan-Feb		Mar-Apr		May-June		July-Aug		Sept-Oct		Nov-Dec	
	(%)	(dis/min/100 scm)	(%)	(dis/min/100 scm)								
Thule												
Apparent Age* (days)	78		136		194		246		131		82	
Production date	Nov. 13, 1961		Nov. 15, 1961		Nov. 18, 1961		Nov. 27, 1961		May 22, 1962		Sept. 9, 1962	
Sr ⁹⁰ assignment:												
Pre-1961	13	0.48	15	0.99	25	1.25	17	0.56	12	(0.20)	7	(0.20)
USSR 1961	87	3.23	85	5.62	75	3.77	83	2.76	29	0.50	32	0.92
U.S. 1962	0	0	0	0	0	0	0	0	0	0	0	0
USSR 1962	0	0	0	0	0	0	0	0	59	1.01	61	1.76
Moosonee												
Apparent Age* (days)	75		147		188		218		128		74	
Production date	Nov. 16, 1961		Nov. 4, 1961		Nov. 24, 1961		Dec. 25, 1961		May 25, 1962		Sept. 17, 1962	
Sr ⁹⁰ assignment:												
Pre-1961	12	0.36	13	0.76	25	1.81	23	1.18	15	(0.30)	10	(0.20)
USSR 1961	88	2.61	87	5.10	75	5.45	77	3.95	30	0.60	21	0.44
U.S. 1962	0	0	0	0	0	0	0	0	0	0	0	0
USSR 1962	0	0	0	0	0	0	0	0	55	1.11	69	1.44
Washington												
Apparent Age* (days)	64		143		193		221		128		68	
Production date	Nov. 27, 1961		Nov. 8, 1961		Nov. 19, 1961		Dec. 22, 1961		May 25, 1962		Sept. 23, 1962	
Sr ⁹⁰ assignment:												
Pre-1961	11	0.49	16	1.25	14	1.24	27	2.43	11	(0.40)	9	(0.30)
USSR 1961	89	3.94	84	6.58	86	7.62	73	6.58	20	0.71	21	0.72
U.S. 1962	0	0	0	0	0	0	0	0	0	0	0	0
USSR 1962	0	0	0	0	0	0	0	0	69	2.47	70	2.42
Miami												
Apparent Age* (days)	57		106		175		204		63		48	
Production date	Dec. 4, 1961		Dec. 15, 1961		Dec. 7, 1961		Jan 9, 1962		July 29, 1962		Oct. 13, 1962	
Sr ⁹⁰ assignment:												
Pre-1961	17	0.89	30	3.30	28	2.05	15	0.37	6	(0.15)	7	(0.40)
USSR 1961	83	4.33	70	7.70	72	5.26	85	2.10	41	0.99	40	2.28
U.S. 1962	0	0	0	0	0	0	0	0	0	0	0	0
USSR 1962	0	0	0	0	0	0	0	0	53	1.29	53	3.03

*Apparent age of youngest component.

TABLE 4 (Continued)
 Sr⁹⁰ Contributions of Various Sources to the Radioactivity of the Atmosphere and the
 Apparent Production Date of Debris from the Youngest Source

	Jan-Feb		Mar-Apr		May-June		July-Aug		Sept-Oct		Nov-Dec	
	(%)	(dis/min/ 100 scm)	(%)	(dis/min/ 100 scm)								
Mauna Loa												
Apparent Age* (days)	67		124		175		170		50		36	
Production date	Nov. 24, 1961		Nov. 27, 1961		Dec. 7, 1961		Feb. 12, 1962		Aug. 11, 1962		Oct. 25, 1962	
Sr ⁹⁰ assignment:												
Pre-1961	38	1.85	20	1.75	24	1.95	18	0.78	15	(0.30)	8	(0.30)
USSR 1961	62	3.03	80	6.98	76	6.19	82	3.55	61	1.25	51	2.00
U.S. 1962	0	0	0	0	0	0	0	0	0	0	0	0
USSR 1962	0	0	0	0	0	0	0	0	24	0.49	41	1.60
Miraflores												
Apparent Age* (days)	82		141		168		187		138		72	
Production date	Nov. 9, 1961		Nov. 10, 1961		Dec. 14, 1961		Jan. 26, 1962		May 15, 1962		Sept. 19, 1962	
Sr ⁹⁰ assignment:												
Pre-1961	9	0.21	14	0.48	12	0.16	6	0.041	16	(0.030)	3	(0.030)
USSR 1961	91	2.12	86	2.94	88	1.21	94	0.643	5	0.009	25	0.263
U.S. 1962	0	0	0	0	0	0	0	0	0	0	0	0
USSR 1962	0	0	0	0	0	0	0	0	79	0.143	72	0.757
Guayaquil												
Apparent age* (days)	96		135		-		114		146		148	
Production date	Oct. 26, 1961		Nov. 16, 1961		-		Apr. 8, 1962		May 7, 1962		July 5, 1962	
Sr ⁹⁰ assignment:												
Pre-1961	40	0.130	40	0.167	-	-	0	0	35	0.144	42	0.188
USSR 1961	60	0.196	60	0.251	-	-	0	0	0	0	0	0
U.S. 1962	0	0	0	0	-	-	(113)	0.212	65	0.266	58	0.259
USSR 1962	0	0	0	0	-	-	0	0	0	0	0	0
Lima												
Apparent age* (days)	100		148		51		94		150		162	
Production date	Oct. 22, 1961		Nov. 3, 1961		Apr. 10, 1962		Apr. 28, 1962		May 3, 1962		June 21, 1962	
Sr ⁹⁰ assignment:												
Pre-1961	92	0.208	90	0.454	6	0.042	17	0.094	30	0.315	45	0.680
USSR 1961	8	0.018	10	0.051	0	0	0	0	0	0	0	0
U.S. 1962	0	0	0	0	94	0.664	83	0.460	70	0.735	55	0.830
USSR 1962	0	0	0	0	0	0	0	0	0	0	0	0

*Apparent age of youngest component.

TABLE 4 (Continued)
 ^{90}Sr Contributions of Various Sources to the Radioactivity of the Atmosphere and the
 Apparent Production Date of Debris from the Youngest Source

	Jan-Feb		Mar-Apr		May-June		July-Aug		Sept-Oct		Nov-Dec	
	(%)	(dis/min/ 100 scm)	(%)	(dis/min/ 100 scm)	(%)	(dis/min/ 100 scm)	(%)	(dis/min/ 100 scm)	(%)	(dis/min/ 100 scm)	(%)	(dis/min/ 100 scm)
Chacalaya												
Apparent age* (days)		85				34		104		131		163
Production date		Nov. 6, 1961				Apr. 27, 1962		Apr. 18, 1962		May 22, 1962		June 20, 1962
^{90}Sr assignment:												
Pre-1961	82	0.159			11	0.137	0	0	7	0.082	30	0.307
USSR 1961	18	0.085			0	0	0	0	0	0	0	0
U.S. 1962	0	0			89	1.108	(142)	1.38	93	1.08	70	0.715
USSR 1962	0	0			0	0	0	0	0	0	0	0
Antofagasta												
Apparent age* (days)		105		140		22		93		150		171
Production date		Oct. 17, 1961		Nov. 11, 1961		May 9, 1962		Apr. 29, 1962		May 3, 1962		June 12, 1962
^{90}Sr assignment:												
Pre-1961	96	0.522	93	0.393	48	0.315	23	0.217	36	0.305	42	0.466
USSR 1961	4	0.022	7	0.029	0	0	0	0	0	0	0	0
U.S. 1962	0	0	0	0	52	0.341	77	0.728	64	0.542	58	0.644
USSR 1962	0	0	0	0	0	0	0	0	0	0	0	0
Santiago												
Apparent age* (days)		91		144		45		-		149		176
Production date		Oct. 31, 1961		Nov. 7, 1961		Apr. 16, 1962		-		May 4, 1962		June 7, 1962
^{90}Sr assignment:												
Pre-1961	99	0.704	98	0.354	59	0.207	-	-	54	0.293	44	0.297
USSR 1961	0.6	0.004	2.0	0.007	0	0	-	-	0	0	0	0
U.S. 1962	0	0	0	0	41	0.144	-	-	46	0.250	56	0.379
USSR 1962	0	0	0	0	0	0	-	-	0	0	0	0
Puerto Montt												
Apparent age* (days)		83		111		46		86		136		177
Production date		Nov. 8, 1961		Dec. 10, 1961		Apr. 15, 1962		May 6, 1962		May 17, 1962		June 6, 1962
^{90}Sr assignment:												
Pre-1961	99+	0.474	99+	0.475	93	0.219	72	0.221	68	0.222	55	0.284
USSR 1961	0.7	0.003	0.5	0.002	0	0	0	0	0	0	0	0
U.S. 1962	0	0	0	0	7	0.016	28	0.086	32	0.104	45	0.232
USSR 1962	0	0	0	0	0	0	0	0	0	0	0	0
Punta Arenas												
Apparent age* (days)		-		76		50		100		145		156
Production date		-		Jan. 15, 1962		Apr. 11, 1962		Apr. 22, 1962		May 8, 1962		June 27, 1962
^{90}Sr assignment:												
Pre-1961	100	0.313	99+	0.227	93	0.137	72	0.104	59	0.112	62	0.120
USSR 1961	0	0	0.4	0.001	0	0	0	0	0	0	0	0
U.S. 1962	0	0	0	0	7	0.010	28	0.041	41	0.078	38	0.074
USSR 1962	0	0	0	0	0	0	0	0	0	0	0	0

*Apparent age of youngest component

date of October 15, 1961 for 1961 USSR debris had been taken instead of those actually calculated, even larger assignment of Sr^{90} to this series would have resulted. The assignments appear reasonable on the basis of the past distribution of debris from the various test sites with the exception of the September-October collections in the Northern Hemisphere. Even though the indicated dates for this set of collections correspond to the period of U.S. OPERATION DOMINIC at Christmas Island, the Ce^{141} data, though incomplete, indicated fresher debris were present. In the absence of any direct evidence of debris from the U.S. tests in the Northern Hemisphere, the arbitrary assignment of these debris to the 1962 USSR series has been made. In partial confirmation of this assignment is the report that no significant amounts of the short-lived Ba^{140} were found over the United Kingdom between April and mid-August 1962, when fresh debris attributed to the new USSR series were encountered (16). Moreover, only small amounts of Ba^{140} were contained in precipitation collections made at Westwood, New Jersey in May and June 1962 indicating a minor contribution of U.S. debris to the mid-latitudes of the Northern Hemisphere during this time (17).

Profiles of the Sr^{90} concentrations from each of the assigned sources as a function of the latitudes of the collecting sites for each of the bi-monthly collection periods are shown in Fig. 8.

In the January-February collection period, the fission product radioactivity encountered at all sites could reasonably be considered to come from only two sources: (a) pre-moratorium debris with an effective production date of January 1958 (based on measured $\text{Ce}^{144}/\text{Sr}^{90}$ activity ratios prior to resumption of testing in September 1961) and (b) debris from the 1961 tests in the Soviet Union with an effective date of mid-October 1961. The profile of the pre-moratorium Sr^{90} exhibits the normal distribution of radioactivity at ground level expected from a stratospheric source, with peaks in the midlatitudes of each hemisphere and an equatorial minimum. At this period, the Sr^{90} levels from the old stratospheric source are roughly of equal magnitude above and below the Equator. Radioactivity from the 1961 USSR series is concentrated primarily in the Northern Hemisphere with a sharp gradient at the Equator. However, there is definite evidence of penetration of

this fresh debris into the Southern Hemisphere where it appears in greatest concentration at the high altitude collection site at Chacaltaya. The quantity of Sr^{90} transported to this region is of minor importance both in regard to its Northern Hemisphere source and in relation to the residual Sr^{90} from the 1958 and earlier test series. The quite different shapes of the Sr^{90} profiles from the two sources in the Southern Hemisphere, and particularly the existence of an out-of-phase relationship at Chacaltaya, would indicate that a purely tropospheric transequatorial transport mechanism for this newer material collected at ground level was operating.

In the March-April collections the pattern of the distribution of the Sr^{90} in the ground-level air along the 80th meridian was relatively unchanged from the preceding period; even at this date negligible penetration of the fresher debris to southern Chile had occurred. The general shape of the latitudinal profile of the older debris, too, underwent little change except for a general increase in Sr^{90} concentrations in the Northern Hemisphere and a northward movement of the position of the maximum, both associated with the advent of spring. An estimated 85% of the Sr^{90} activity in the air at the three northernmost sites can be attributed to the 1961 USSR tests; this is in agreement with reports that 85-90% of the Sr^{90} concentration in rainfall at Abingdon and Milford Haven in the United Kingdom and at Westwood, New Jersey during the spring of 1962 could be ascribed to the 1961 Soviet Test Series (17,18).

The May-June profile of the pre-1961 Sr^{90} continued to exhibit the expected shape with a Northern Hemisphere spring maximum and a Southern Hemisphere fall minimum. The Northern Hemisphere portion of the profile of 1961 USSR debris was also relatively unchanged; this curve could not be extended below the Equator because of the influx there of fresh debris from the USA 1962 DOMINIC tests which started April 25, 1962. The activity from the Christmas Island area arrived in highest concentration at Chacaltaya, as had occurred in past tests in the Pacific area (5,6), and was confined principally to relatively low latitudes of the Southern Hemisphere. The rather consistent production dates obtained for the fresher component of the Northern Hemisphere debris, falling between mid-November and mid-December 1961 (Table 4),

indicated no significant contribution of radioactivity from this new source to the collections made north of the Equator.

In July and August there evidently was a small but unresolvable contribution of fresh material to several sites in the Northern Hemisphere, as shown by dates pointing to apparent production times in January and February 1962; however, it is assumed that this source made no significant contribution to the total Sr^{90} observed at these sites. The size and shape of the Sr^{90} profile attributed to the 1961 USSR debris and its close resemblance to the distribution of the pre-1961 Sr^{90} , would indicate the mixture collected came from a reasonably well-mixed source, presumably the stratosphere. The Sr^{90} from the 1962 U.S. tests continued to appear principally in the subtropical region of the Southern Hemisphere with a peak concentration at Chacaltaya; migration of this material to more southerly regions was also evident. The pre-1961 Sr^{90} distribution was not significantly changed, though decreases in the concentrations of Sr^{90} in the air in the Northern Hemisphere indicated that the annual peak in stratospheric fallout or subsidence had passed.

In September and October and also in November and December 1962, the air of the Northern Hemisphere could have contained debris from four possible sources, two of which (1962 USSR and 1962 U.S.) overlapped in time. To make possible an analysis of the data, it was necessary to make simplifications by assuming that the Sr^{90} from pre-moratorium tests would follow the trends established in prior years and that the principal contributions to the remainder were stratospheric debris from the 1961 USSR tests, with an effective generation date of October 15, 1961, and a fresh component from the 1962 USSR or the 1962 U.S. test series. Assignment of this fresh component to the USSR source has been made, as discussed earlier.

The profiles of pre-moratorium Sr^{90} in both the September-October and November-December (1962) periods appear reasonable in the light of past experience, which indicated the approaching equilibrium of the stratospheric burdens over the two hemispheres, with the corollary result that the activity during the Southern Hemisphere spring maximum would exceed that of the Northern Hemisphere fall minimum.

The fresher component of the Sr^{90} in the air of the Southern Hemisphere, which has been assigned to the U.S. Christmas Island tests, though collections at such sites as Guayaquil and Chacaltaya undoubtedly contained some contribution of USSR debris, continued the pattern of past months with some broadening of the peak as activity migrated away from its primary path or descended from higher altitudes. The lack of evidence of fresh debris (Table 4) in these collections, particularly at Chacaltaya, in spite of continued U.S. testing during October and November 1962 at Christmas Island is noteworthy. It may suggest a seasonal shift of direction of the upper tropospheric air currents from the Christmas Island area to a northeastward rather than a southeastward direction. The lack of a tropospheric component of fresh activity in the Southern Hemisphere collections made during November and December may account for the greater similarity of the 1962 U.S. and pre-1961 Sr^{90} profiles at this time.

In the Northern Hemisphere, the fresh Sr^{90} component which has been assigned to the USSR test series (August 5-December 24, 1962) has in each case the distribution which might be expected for the tropospheric component of nuclear tests carried out in the arctic area. If the U.S. Christmas Island tests were a large contributor to the fresh component, the profile would have been expected to have peaked more strongly in the tropical or subtropical region.

ACTIVITY RATIOS

Several activity ratios plotted as a function of time are shown in Figs. 9 and 10, and are included to show the well-mixed condition of the Northern Hemisphere air at ground level during early 1962 and to document the fact that there was little contribution of fresh long-lived activity to this area prior to resumption of testing in the USSR in August 1962.

The $\text{Y}^{91}/\text{Ce}^{144}$, $\text{Sr}^{89}/\text{Ce}^{144}$, and $\text{Sr}^{89}/\text{Sr}^{90}$ ratios are in excellent agreement among the four northernmost stations (Thule, Moosonee, Washington, Miami) during each of the first three bimonthly collection periods of 1962. For each ratio there is a general trend toward decreasing age (relative increase in the younger component) with time that

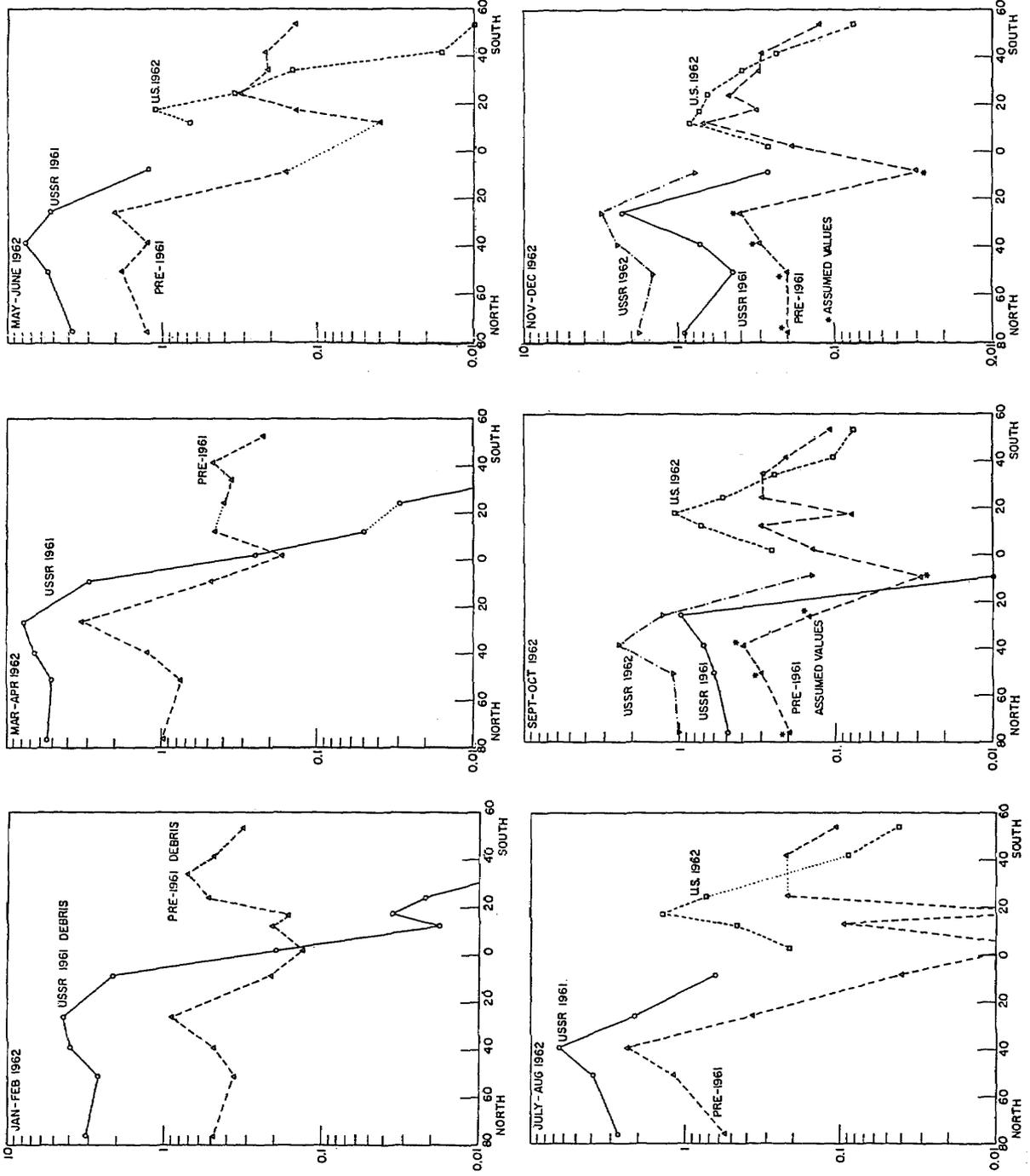
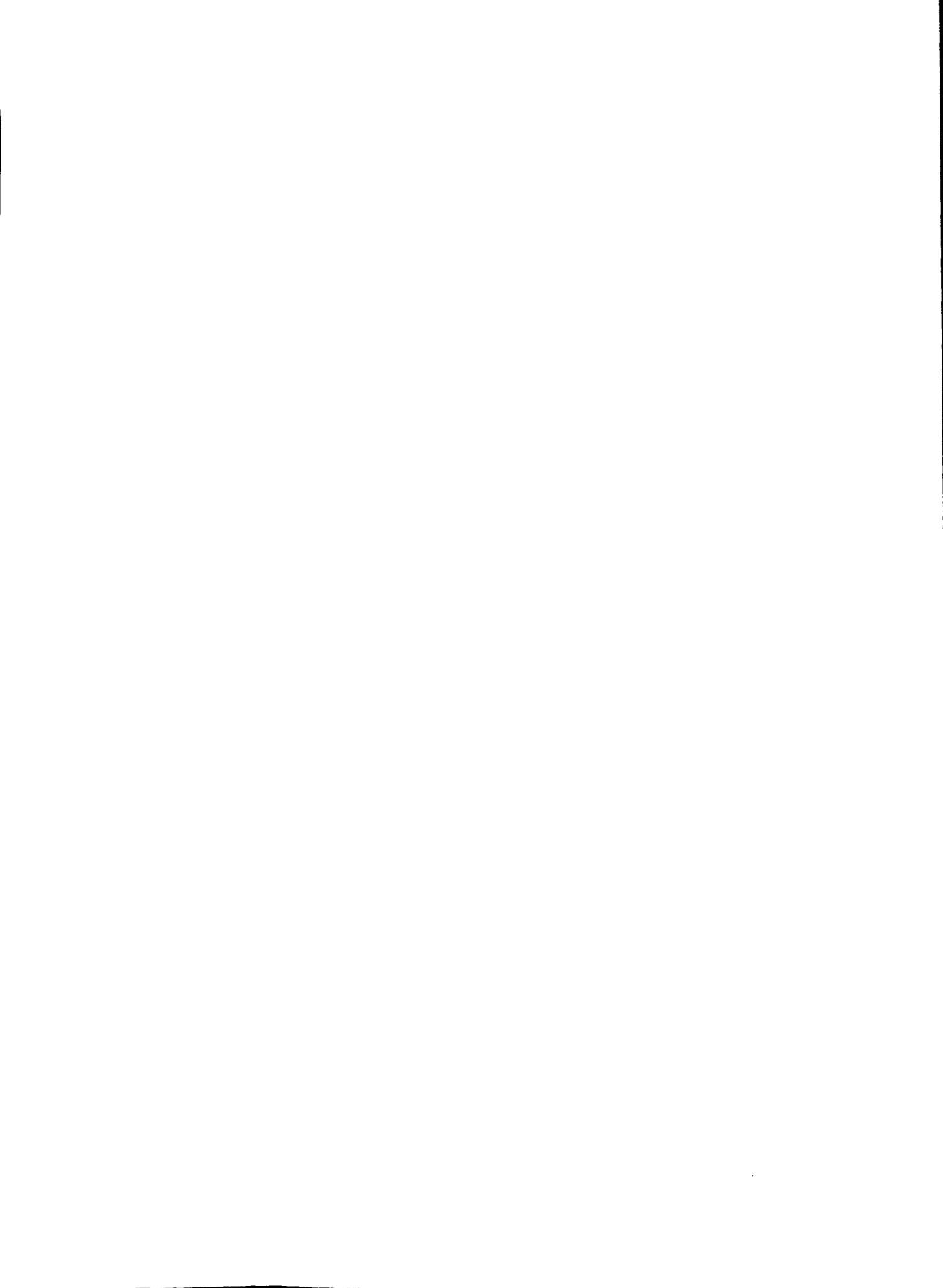
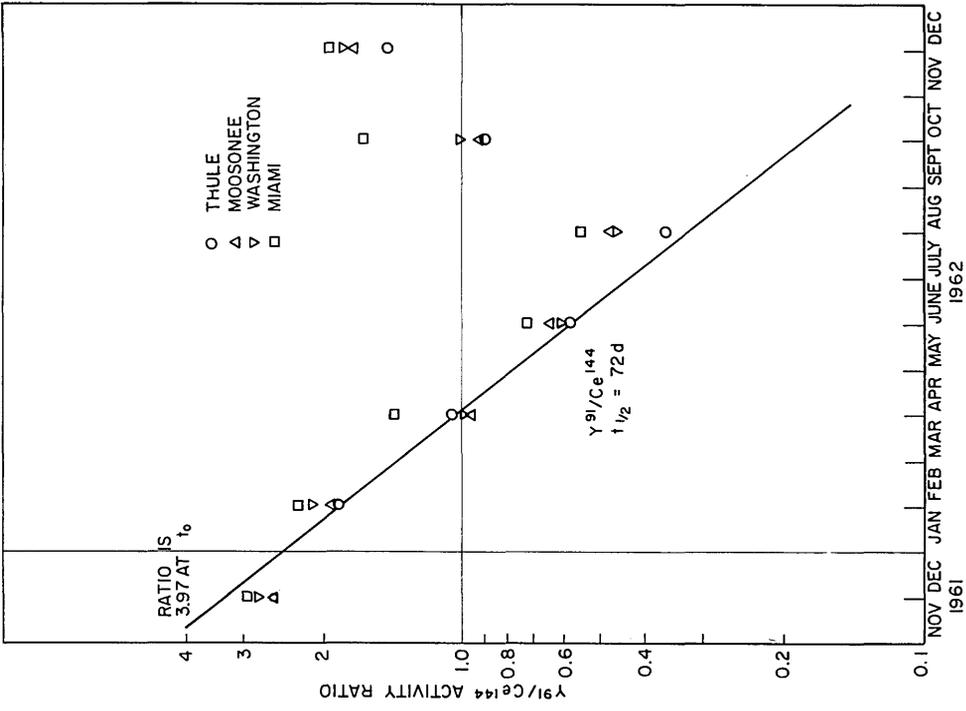
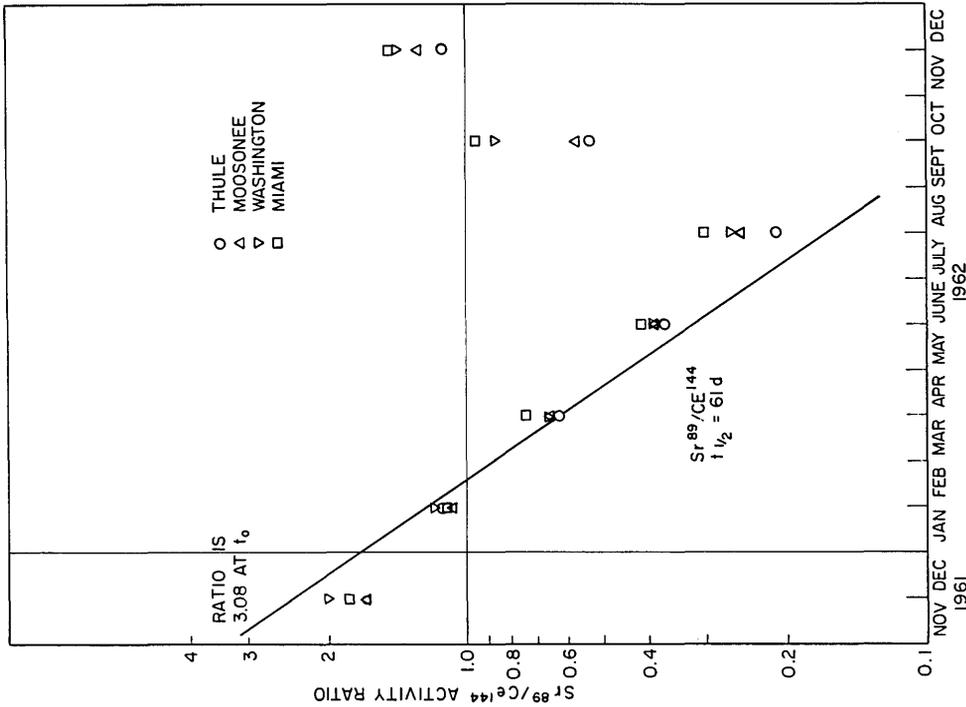


Fig. 8 - Profiles of the Sr^{90} concentrations in the air along the 80th Meridian (West) during 1962 contributed by the various nuclear test series





(a)



(b)

Fig. 9 — Ratios of Sr^{89} (Fig. 9a) and Y^{91} (Fig. 9b) to the Ce^{144} activity in the ground-level air at several sites in the Northern Hemisphere during 1962

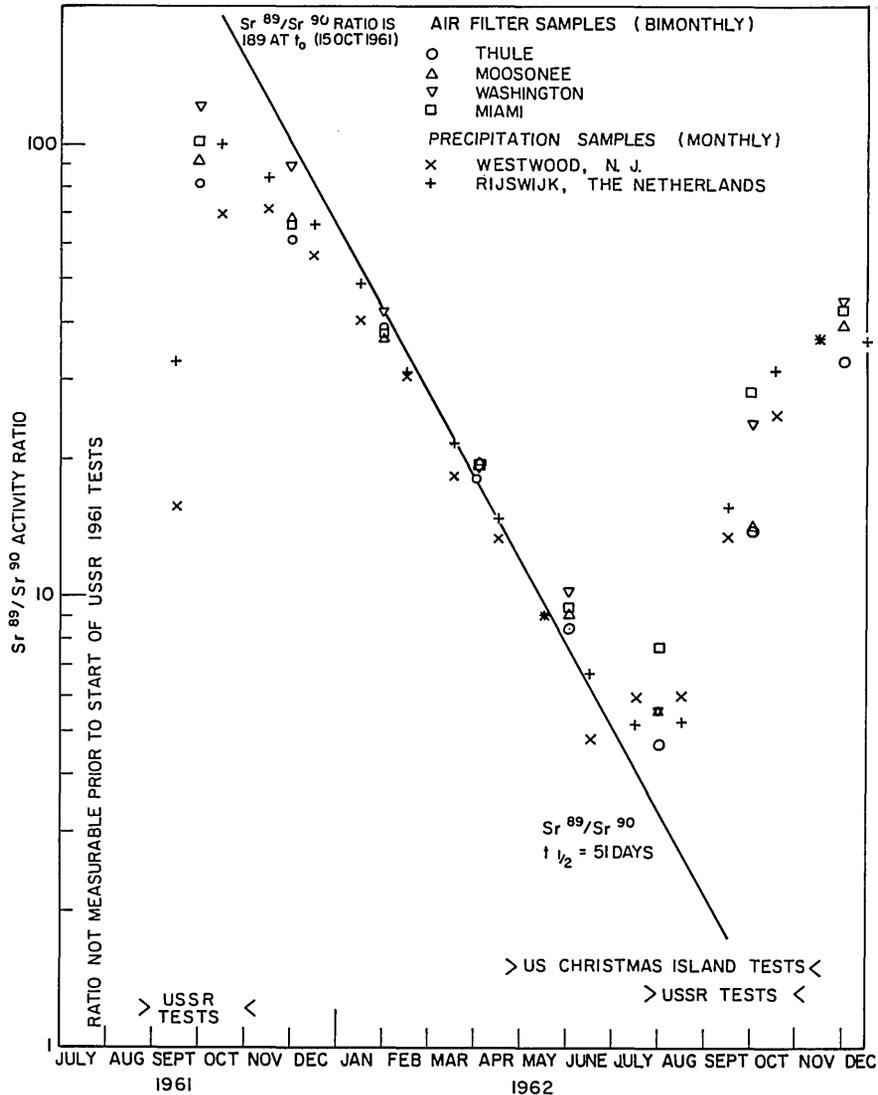


Fig. 10 — Ratios of Sr^{89} to Sr^{90} activity in the air and in precipitation at various Northern Hemisphere sites during 1962

is suggestive of a gradually increasing influx of debris from a later test of the 1961 USSR series. This may possibly be due to an increasing contribution from the large yield USSR tests of October 23, and October 30 or, after the end of April, it may be due to some contamination from the U.S. Christmas Island tests.

In Fig. 10, in addition to the Sr^{89}/Sr^{90} activity ratios obtained from air collections made at several 80th-meridian sites, the Sr^{89}/Sr^{90} ratios for monthly precipitation samples collected at Westwood, New Jersey (19), and Rijswijk, the Netherlands (20),

during 1962 are presented. The agreement among the various sites is remarkable considering the different collection and analytical techniques employed. The Sr^{89}/Sr^{90} ratios reported in precipitation collections of individual rains at Westwood exhibit the same trends as observed in the air at Washington, D.C. with about a 10-15% increase during May and June, at which time some Ba^{140} activity was also observed (17). The increased Sr^{89} content is roughly equivalent to a 1% increase in the total Sr^{90} which is the most that could be attributed to the U.S. Christmas

Island tests. The $\text{Sr}^{89}/\text{Sr}^{90}$ activity ratios in monthly rainwater collections at Rijswijk, The Netherlands (20), were very close to those obtained from air samples collected at Thule, Greeland, and did not show any deviation from that due to decay until July, when about a 15% increase in the younger component was observed.

In the activity ratios presented in Figs. 9 and 10, there is a larger deviation in July and August than in the previous months from the values expected through the normal radioactive decay process. Moreover, this increase in the younger component of each pair of isotopes shows up strongest at Miami and least at Thule, which is suggestive of the presence of fresh activity from the Christmas Island area. However, in past years it has been commonplace for fresh debris from the arctic region to show up in this same area; since a new USSR series started in early August 1962, no certain determination can be made as to the source of this small component. Examination of the weekly gross activity records at Miami and the more southerly sites in the Northern Hemisphere most likely to be in the path of debris from the mid-Pacific gave no indication of the influx of highly active air masses until late October.

In the September-October and November-December collections there was a substantial increase in the shorter lived component of each activity pair that has, on the basis of purely circumstantial evidence, been assigned to the presence of debris from the 1962 USSR series. The scatter of values of the activity ratios is typical of the pattern encountered during and immediately after nuclear tests when tropospheric transport of bomb debris is important. As was evident in the earlier collections made during the spring and early summer, there is more uniformity at the various sites in the composition of fission products which have been able to undergo mixing during their stratospheric residence period. The timing and yield of these USSR tests and of the somewhat earlier U.S. tests was such that a large proportion of the radioactive products from both test series should have entered the stratospheric reservoir. The great increase in the $\text{Sr}^{89}/\text{Sr}^{90}$ ratios during September and October was not due so much to the influx of large quantities of fresh activity but rather to the decreased contribution of the stratospheric source during this period.

As has been observed in the past (8,9), the $\text{Cs}^{137}/\text{Sr}^{90}$ activity ratios in the air during 1962 were highly variable with quite obviously lower ratios at Miami and at Puerto Montt. During 1962, the $\text{Cs}^{137}/\text{Sr}^{90}$ ratios were generally lower in the Northern Hemisphere than in the Southern Hemisphere and, moreover, were lower than had been observed in prior years. To determine if the β counting techniques employed for evaluating the Cs^{137} were at fault, a Cs^{137} standard (prepared from NBS standardized solution) and two Cs^{137} samples representing collections having normal and abnormal $\text{Cs}^{137}/\text{Sr}^{90}$ activity ratios (Washington, January-February 1962; Miami, January-February 1962) were taken to the Health and Safety Laboratory, U.S.A.E.C., New York, for γ -spectral analysis (21). All gave spectra which indicated pure Cs^{137} activity; furthermore, the gross γ counts, when compared to the β disintegration rates determined at NRL, gave constant ratios (less than 2% deviation) which effectively eliminated counting error as a source of the extremely low activity ratios. The reproducibility of Sr^{90} activity measurements, both by direct counting on different units and by separation and evaluation of its Y^{90} daughter, and the consistent interrelations of Sr^{90} with isotopes other than Cs^{137} , had already discounted the possibility of errors in its determination as a source of the observed variations. Again, the indication is that some natural process of fractionation has occurred which operates more strongly in certain areas than in others.

CONCLUSIONS

This study of the fission product content of the atmosphere has covered an interesting period of the atomic age and the development of nuclear weaponry. At its initial stage the concept of a stratospheric residence time for radioactive debris was highly speculative, and seasonal variations in the deposition from this reservoir were not apparent, primarily due to the disadvantageous scheduling of nuclear tests. The temporary moratorium on nuclear testing following several extensive series of high yield nuclear tests permitted the unambiguous verification by many investigators of seasonal changes in the deposition rate of stratospherically stored debris over the entire Northern Hemisphere. It also permitted determination of the effect of the latitude of injection

on the residence time of radioactivity in the stratosphere with shorter residence periods observed in the arctic stratosphere. During this period and also after the renewed testing had begun in the arctic region, the lack of an effective mechanism by which particulate matter could be transferred across the equator in the lower atmosphere was again noted.

With the conduct of full scale testing of large yield nuclear devices in both the arctic and tropic regions in 1962, followed by what is presumed will be a more permanent moratorium on testing, the condition of the stratospheric inventory and prospects for observations during the coming years are equivalent to those existing in the period 1959 to 1961. The multiplicity of sources will again cause difficulty in making quantitative interpretations of atmospheric mixing or transport processes, though the ample quantity of radioactivity available will simplify qualitative studies.

REFERENCES

- Lockhart, L.B., Jr., Baus, R.A., King, P., and Blifford, I.H., Jr., "Atmospheric Radioactivity Studies at the U.S. Naval Research Laboratory," NRL Report 5249, Dec. 1958; *J. Chem. Ed.* **36**:291-5 (1959)
- Lockhart, L.B., Jr., Baus, R.A., and Blifford, I.H., Jr., "Atmospheric Radioactivity Along the 80th Meridian, 1956," NRL Report 4965, July 1957
- Lockhart, L.B., Jr., Baus, R.A., and Blifford, I.H., Jr., "Fission Product Radioactivity in the Air Along the 80th Meridian, January-June 1957," NRL Report 5041, Nov. 1957; *Tellus XI*:83-90 (1959)
- Baus, R.A., Patterson, R.L., Jr., Saunders, A.W., Jr., and Lockhart, L.B., Jr., "Radiochemical Analyses of Air Filter Samples Collected During 1957," NRL Report 5239, Dec. 1958
- Lockhart, L.B., Jr., Patterson, R.L., Jr., and Anderson, W.L., "Measurements of the Air Concentration of Gross Fission Product Radioactivity During the IGY, July 1957-December 1958," NRL Report 5359, Sept. 1959; *Tellus XII*:298-307 (1960)
- Lockhart, L.B., Jr., Baus, R.A., Patterson, R.L., Jr., and Saunders, A.W., Jr., "Radiochemical Analyses of Air Filter Samples Collected During 1958," NRL Report 5390, October 1959; *J. Geophys. Res.* **65**:1711-22 (1960)
- Lockhart, L.B., Jr., Patterson, R.L., Jr., Saunders, A.W., Jr., and Black, R.W., "Fission Product Radioactivity in the Air Along the 80th Meridian (West) During 1959," NRL Report 5528, Aug. 1960; *J. Geophys. Res.* **65**:3987-97 (1960)
- Lockhart, L.B., Jr., Patterson, R.L., Jr., Saunders, A.W., Jr., and Black, R.W., "Fission Product Radioactivity in the Air Along the 80th Meridian (West) During 1960," NRL Report 5692, Oct. 1961
- Lockhart, L.B., Jr., Patterson, R.L., Jr., Saunders, A.W., Jr., and Black, R.W., "Fission Product Radioactivity in the Air Along the 80th Meridian (West) During 1961," NRL Report 5869, Jan. 1963
- Lockhart, L.B., Jr., and Patterson, R.L., Jr. "Critical Analysis of Measurements of the Gross Fission Product Activity in the Air at Ground Level," NRL Report 5440, Feb. 1960
- Lockhart, L.B., Jr., and Patterson, R.L., Jr., "Intercalibration of Some Systems Employed in Monitoring Fission Products in the Atmosphere," NRL Report 5850, Nov. 1962; *Rad. Health Data* **3**:466-70 (1962)
- Baus, R.A., Gustafson, P.R., Patterson, R.L., Jr., and Saunders, A.W., Jr., "Procedure for the Sequential Radiochemical Analysis of Strontium, Yttrium, Cesium, Cerium and Bismuth in Air-Filter Collections," NRL Memorandum Report 758, Nov. 1957
- "Announced Nuclear Detonations, 1945-1962, United States, United Kingdom, Republic of France, Union of Soviet Socialist Republics," compiled by the Atmospheric Radioactivity Research Project, Meteorological Research Projects, U.S. Weather Bureau, in Health and Safety Laboratory Fallout Program Quarterly Summary Report HASL-142, Jan. 1, 1964
- Lockhart, L.B., Jr., Patterson, R.L., Jr., and Saunders, A.W., Jr., "Filter Pack Technique for Classifying Radioactive Aerosols by Particle Size. Part 2 - Isotopic Fractionation with Particle Size," NRL Report in progress.
- Cambray, R.S., Fisher, E.M.R., Spicer, G.S., Wallace, C.G., and Webber, T.J., "Radioactive Fallout in Air and Rain - Results to the Middle of 1963," United Kingdom Atomic Energy Authority Research Group Report AERE-R 4392, Nov. 1963
- Cambray, R.S., and Fisher, E.M.R., "Radioactive Fallout: Short-Lived Fission Products in Air and Rain, August 1962-April 1963," United Kingdom Atomic Energy Authority Research Group Report AERE-R 4384, July 1963
- Walton, A., and Leo, M.W.M., "Studies of Nuclear Debris in Precipitation," Isotopes, Inc., report on AEC Contract Number AT(30-1)-2415, NYO-9533, Nov. 15, 1962
- Cambray, R.S., Fisher, E.M.R., Spicer, G.S., Wallace, C.G., and Webber, T.J., "Radioactive Fallout in Air and Rain - Results to the Middle of 1962," United Kingdom Atomic Energy Authority Research Group Report AERE-R 4094, Nov. 1962
- Walton A., and Leo, M.W.M., "Studies of Nuclear Debris in Precipitation," Isotopes, Inc., report on AEC Contract Number AT(30-1)-2415, NYO-9534, Mar. 31, 1963
- Blok, J., and Bleichrodt, J.F., "Summary of Radioactive Fallout Measurements. III.," Netherlands Medisch Biologisch Laboratorium RVO-TNO Report MBL/1963/13, Aug. 1963
- Collins, W.R., Health and Safety Laboratory, U.S. Atomic Energy Commission, New York, N.Y., personal communication

APPENDIX A RADIOACTIVITY BLANKS

The gross β activity of the ignited ash of a group of seventeen 8-inch filters selected at random from the available stock was determined on the automatic sample changer and β counter used to measure the radioactivity of exposed filters. An average activity of 0.54 ± 0.10 counts per minute above background was found; this is roughly equivalent to 0.0025 dis/min per cubic meter of air in a normal one week collection. In practice this background count was included in the equipment blank and was subtracted from the measured counts prior to calculation of the activity concentrations.

The combined ash from these 17 unexposed filters was subjected to radiochemical analysis for long-lived activities to determine if the contaminants adversely affected the determination of the concentration of airborne radioisotopes. The results indicated in Table A1 represent the overall effect of extraneous activities including both the reagent blank and the filter blank.

The major long-lived activity identified was Pb^{210} (Bi^{210}) as might be expected; no attempt was made to determine other natural radioisotopes which might have been present as they were not considered in the evaluation of exposed filters. The contribution of the Pb^{210} background to the calculated air concentration of Pb^{210} was generally small (<10%) though in a few cases, *e.g.* at Punta Arenas where the Pb^{210} content of the atmosphere

TABLE A1
Determination of the Radioactive Background
of Type 6 Cellulose-Asbestos Filter Paper

Species Measured	Radioactivity* (dis/min/filter)	Equivalent Activity Concentration † (dis/min/m ³ of air)
Gross β	21.1	2.5×10^{-3}
Pb^{210} (or Bi^{210})	1.9	2.3×10^{-4}
Cs^{137}	0.58	6.9×10^{-5}
Pm^{147}	0.35	4.2×10^{-5}
Sr^{90} (or Y^{90})	0.21	2.5×10^{-5}
Ce^{144}	0.013	1.5×10^{-6}

*Average of 17 filters (8-inch diameter); radiochemical analysis performed on combined filter ash.

†Based on a 7-day exposure at 1200 m³ per day.

was extremely low, it was as high as 20-25%. On the other hand the Sr^{90} and Cs^{137} backgrounds usually contributed less than 1% (maximum approximately 2%) of the calculated atmospheric content of these isotopes. The contribution of contaminant Pm^{147} (<1.0%) and Ce^{144} (<0.1%) was negligible in all cases. No corrections were made to the air concentrations of any of the separated radioisotopes for the presence of these backgrounds of radioactivity.

APPENDIX B

ESTIMATION OF PRODUCTION DATES OF NUCLEAR DEBRIS AND THEIR Sr^{90} CONTRIBUTIONS TO THE RADIOACTIVITY OF THE ATMOSPHERE

A mathematical analysis of the radiochemical data can be employed to make assignments of the relative Sr^{90} contributions due to each of two components and to assign an apparent shot date for the more recent component. The limited quantity of data on hand, and particularly, the lack of reliable and complete data on a short-lived radioisotope, limits the analysis to simple two-component mixtures; the older component of which can be preassigned a reasonable date of origin. The following series of equations is based

on the utilization of the measured concentrations of the radioisotopes Y^{91} (57.5 day half-life), Ce^{144} (285 day half-life), and Sr^{90} (27.7 year half-life). Other equations involving Sr^{89} in place of Y^{91} , Pm^{147} in place of Ce^{144} , or Cs^{137} in place of Sr^{90} could have been derived and would have given essentially the same results. The above isotopes were selected on the basis of their ease of isolation by radiochemical techniques and the greater reliability of their evaluation by β counting techniques.

For two-component mixtures, or for more complicated mixtures which can be effectively reduced to the above, the following relationships between the various components exist:

$$Sr_t^{90} = Sr_1^{90} + Sr_2^{90}$$

$$Ce_t^{144} = Ce_1^{144} + Ce_2^{144}$$

$$Y_t^{91} = Y_1^{91} + Y_2^{91}$$

where the t refers to the total activity and 1 and 2 refer to the contributions of the components of the mixture at the midpoint of the collection period.

Since the following identity exists:

$$Ce_1^{144} = Ce_0^{144} e^{-\lambda_c t_1} = Ce_0^{144} e^{-\lambda_c t_1} \cdot \frac{Sr_1^{90}}{Sr_0^{90} e^{-\lambda_s t_1}}$$

$$= \left(\frac{Ce^{144}}{Sr^{90}} \right)_0 Sr_1^{90} e^{(-\lambda_c + \lambda_s) t_1}$$

the second equation may be rewritten to give:

$$Ce_t^{144} = \left(\frac{Ce^{144}}{Sr^{90}} \right)_0 Sr_1^{90} e^{(-\lambda_c + \lambda_s) t_1}$$

$$+ \left(\frac{Ce^{144}}{Sr^{90}} \right)_0 Sr_2^{90} e^{(-\lambda_c + \lambda_s) t_2}$$

where

$$\left(\frac{Ce^{144}}{Sr^{90}} \right)_0 = 47.6$$

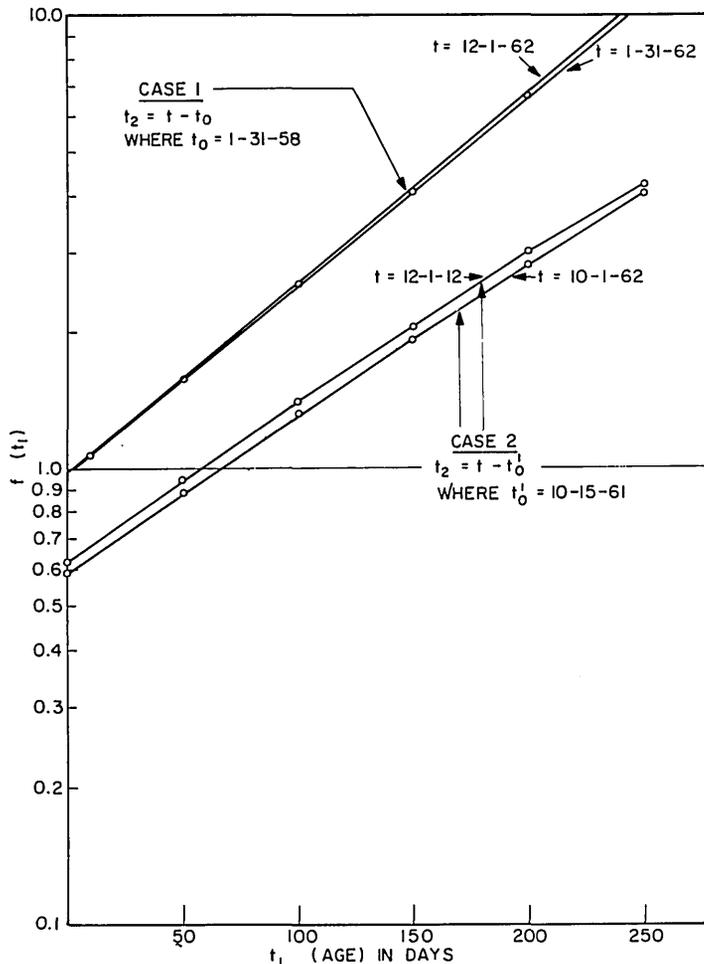


Fig. B1 – Graphical solution of the exponential function $f(t_1)$

for megaton weapons at the time of detonation,* λ_c and λ_s are the decay constants of Ce^{144} and Sr^{90} respectively, and t_1 and t_2 are the elapsed times between formation and collection of the two components.

Similarly,

$$Y_t^{91} = \left(\frac{Y^{91}}{Sr^{90}}\right)_0 Sr_1^{90} e^{(-\lambda_y + \lambda_s)t_1} + \left(\frac{Y^{91}}{Sr^{90}}\right)_0 Sr_2^{90} e^{(-\lambda_y + \lambda_s)t_2}$$

where

$$\left(\frac{Y^{91}}{Sr^{90}}\right)_0 = 189$$

for megaton weapons at the time of detonation* and λ_y is the decay constant of Y^{91} .

The three working equations thus become

$$Sr_t^{90} = Sr_1^{90} + Sr_2^{90} \quad (B1)$$

$$Ce_t^{144} = 47.6 Sr_1^{90} e^{-0.00236t_1} + 47.6 Sr_2^{90} e^{-0.00236t_2} \quad (B2)$$

$$Y_t^{91} = 189 Sr_1^{90} e^{-0.0120t_1} + 189 Sr_2^{90} e^{-0.0120t_2} \quad (B3)$$

Substitution of $Sr_t^{90} - Sr_1^{90}$ for Sr_2^{90} in Eqs. (B2) and (B3) and elimination of Sr_1^{90} , gives the following expression:

$$\frac{\frac{Ce_t^{144}}{47.6} - Sr_t^{90} (e^{-0.00236t_2})}{\frac{Y_t^{91}}{189} - Sr_t^{90} (e^{-0.0120t_2})} = f(t_1)$$

where

$$f(t_1) = \frac{e^{-0.00236t_1} - e^{-0.00236t_2}}{e^{-0.0120t_1} - e^{-0.0120t_2}}$$

When t_2 is fixed by assigning a formation time for the older debris, a solution of $f(t_1)$ can be found from analytical values for Sr_t^{90} , Ce_t^{144} , and Y_t^{91} . The age t_1 of the younger component can then be conveniently determined from plots of $f(t_1)$ vs t_1 as indicated in Fig. B1.

An expression for determining the contribution of Sr^{90} from the younger component to the total Sr^{90} can be obtained by eliminating Sr_2^{90} between Eqs. (B1) and (B2) and dividing by Sr_1^{90} to give:

$$\frac{Sr_1^{90}}{Sr_t^{90}} = \frac{\left(\frac{Ce_t^{144}}{Sr_t^{90}}\right)\left(\frac{1}{47.6}\right) - e^{-0.00236t_2}}{e^{-0.00236t_1} - e^{-0.00236t_2}}$$

*N.A. Hallden, I.M. Fisenne, L.D.Y. Ong, and J.H. Hartley, "Radioactive Decay of Weapons Debris," Fallout Program Quarterly Report, USAEC, HASL-117, pp. 194-199, Dec. 1961.