ATMOSPHERIC RADIOCHEMISTRY

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Annual Progress Report

to the

United States Atomic Energy Commission

Contract No. At-(40-1)-2529

by the University of Arkansas

Department of Chemistry

Fayetteville, Arkansas 72701

April 30, 1974

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P. K. Kuroda

P. K. Kuroda, Principal Investigator

J. N. Beck

P. Y. Daniel

M. B. Gavini

R. W. Holloway

R. D. Sherrill

N. G. Sumerlin

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PART I. RESEARCH IN ATMOSPHERIC RADIOCHEMISTRY

The present Annual Report summarizes the results obtained since the last report was written in April, 1973. Four reprints, 4 preprints, and 2 data sets are attached to this report as appendices.

(1) Behavior of Radiostrontium in Rain and Air after the Ninth Chinese Atmospheric Nuclear Test, J. Geophys. Res., 78, 1419 (1973), by J. R. Noyce, D. T. Moore, J. N. Beck and P. K. Kuroda.

A reprint of this work is attached to this report as APPENDIX I.

(2) Radioactive Cerium Isotopes: The Fallout from Recent French and Chinese Nuclear Weapons Tests, Health Physics, 24, 491 (1973), by J. D. Sherwood, D. T. Moore and P. K. Kuroda

A reprint of this work is attached to this report as APPENDIX II.

(3) Fallout from Nuclear Weapons Testing and Interhemispheric Transport of Nuclear Debris, Health Physics, 25, 109 (1973), by J. R. Noyce, D. T. Moore, J. D. Sherwood, P. R. Daniel, J. N. Beck and P. K. Kuroda.

A reprint of this work is attached to this report as APPENDIX III.

(4) Radioactive Hot Particles from the Recent Chinese Nuclear Weapons Tests, J. Geophys. Res., 78, 7039 (1973), by D. T. Moore, J. N. Beck, D. K. Miller and P. K. Kuroda.

A reprint of this work is attached to this report as APPENDIX IV.

(5) Residence Time of Strontium-90 in the Atmosphere (P. Y. Daniel, A. Nevissi, J. L. Meason, J. N. Beck and P. K. Kuroda).

The concentrations of ^{89}Sr and ^{90}Sr were measured in the entire series of rain and snow samples collected at Fayetteville (36°N, 94°W), Arkansas, during the period between August 1971 and June 1973. Measurements were also made on some rain samples collected at Tokyo (36°N, 140°E), Japan, and at Ankara (48°N, 33°E), Turkey. Three nuclear tests were conducted in Lop Nor (40°N, 90°E), China, while this study was being made: the 12th (November 18, 1971), the 13th (January 7, 1972) and the 14th (March 18, 1972). The 'cycling' effect of the fresh debris was clearly observed after the 14th Chinese test: peaks in the $^{89}Sr/^{90}Sr$ ratio occurred successively at Fayetteville (94°W), Ankara (33°E), and Tokyo (140°E), in that order, indicating that the freshly injected debris travelled eastward circling the world at least twice until the distribution of the fission products in the troposphere became more or less uniform after about two months. Values of the residence time of ^{90}Sr in the troposphere were calculated from the $^{89}Sr/^{90}Sr$ ratio data. While the debris from the 12th and the 13th tests yielded rather short apparent residence times of about 8 ± 3 days and 12 ± 3 days, respectively, much longer and approximately concordant mean residence times were obtained from the $^{89}Sr/^{90}Sr$ and $^{210}Po/^{210}Pb$ ratios measured simultaneously in many of the rain samples collected after the 14th test. The results indicate that most of the ^{210}Po originated from the decay of ^{222}Rn in the troposphere. The experimental data obtained in this work are consistent with the mean residence time of ^{90}Sr in the stratosphere of approximately one year.

The results of this work written in preprint form are attached to this report as APPENDIX V.

(6) Variation of the Ratio of Cesium-137 to Strontium-90 in the Atmosphere, (R. D. Sherrill, N. G. Sumerlin, J. N. Beck and P. K. Kuroda).

Concentrations of ^{137}Cs and ^{90}Sr were measured in a total of 137 individual rain and snow samples collected at Fayetteville, Arkansas, during the period between February 1967 and June 1969. The $^{137}Cs/^{90}Sr$ ratio was found to vary quite considerably: 0.26 (12 April 1968) to 8.2 (8 August 1968). Bi-monthly average $^{137}Cs/^{90}Sr$ ratios in rain fluctuated also from 0.75 (January-February 1968) to 1.9 (July-August 1968). The variation in the $^{137}Cs/^{90}Sr$ ratio seems to be attributable to the difference in the size distributions of particulate matters in rain. ^{137}Cs and ^{90}Sr are depleted in large fallout particles, because of the fairly long half-lives of their precursors: ^{137}Xe (3.82 min) and ^{90}Kr (32.3-sec). A simple calculation, in which the half-lives of the gaseous precursors and the independent and cumulative yields of the fission products belonging to the mass 90 and 137 chains are taken into consideration, shows that extreme values of the $^{137}Cs/^{90}Sr$ ratio in large and small fallout particles expected to be observed in rain may range from about 0.2 to several times the production ratio.

The results of this work written in preprint form are attached to this report as APPENDIX VI.

(7) Mean Residence Times of the Long-Lived Radon Daughters in the Atmosphere, (M. B. Gavini, J. N. Beck and P. K. Kuroda).

The activities of ^{210}Pb, ^{210}Bi and ^{210}Po were measured in 17 rain samples which were collected at Fayetteville, Arkansas, during the months of April through June 1973, and ^{210}Pb and ^{210}Po only were measured

in 20 additional rain samples collected mostly during the months of September through December, 1973. In approximately 70 percent of the cases (12 out of 17 rains), the residence time calculated from the $^{210}Bi/^{210}Pb$ and $^{210}Po/^{210}Pb$ ratios were either concordant or near concordant. In about 30 percent of the cases (5 out of 17 rains), the $^{210}Bi/^{210}Pb$ ratios yielded mean residence time values of 3 to 6 days, while the values calculated from the $^{210}Po/^{210}Pb$ ratios varied from 16 to 40 days. The discordant residence times can be shown to result from the mixing of two different air masses: one represented by a short and the other by a much longer residence time of the radon daughters. In order to test this idea, the radionuclides ^{210}Pb and ^{210}Po were sequentially sampled from a rainstorm, which occurred at Fayetteville, Arkansas, on October 6, 1973. The $^{210}Po/^{210}Pb$ ratio was found to vary markedly within a single rainfall and the data indicated that a mixing of two air masses occurred during the rainstorm: one air mass represented by a residence time of about 30 days and the other roughly 300 days. These values are suggestive of the mean residence of aerosols for the entire troposphere (about 30 days) and for the entire stratosphere (about one year) frequently mentioned by previous investigators.

The results of this work written in preprint form are attached to this report as APPENDIX VII.

(8) The Ratios of Fission Product Pairs with Similar Half-Lives in the Atmosphere, (R. W. Holloway, N. G. Sumerlin, J. N. Beck and P. K. Kuroda).

The ratios of pairs of fission products with similar half-lives (58.5-day Y^{91} to 50.8-day Sr^{89} and 13.6-day Pr^{143} to 12.8-day Ba^{140}) were measured radiochemically in a series of rain samples collected at Fayetteville (36°N, 94°W), Arkansas, after the Chinese explosions of March 18, 1972 (the 14th) and of June 27, 1973 (the 15th). The activity ratios in rain were found to show a marked variation: The Y^{91}/Sr^{89} ratio in rain varied between the extreme values of 0.29 (March 24, 1972) and 10 (April 3, 1972), while the Pr^{143}/Ba^{140} ratio from 0.21 (March 24, 1972) to 6.3 (April 13 and 14, 1972). The observed large variations of the activity ratios appear to be attributable to the difference in size distributions of particulate matter in rain. Y^{91} and Pr^{143} become highly enriched in particles larger than a few micron size, relative to Sr^{89} and Ba^{140}, respectively, whereas the reverse appears to be the case in smaller, sub-micron size particles. A similar trend was found in the data obtained after the 7th Chinese test of December 24, 1967. The results indicate that these isotopic pairs may turn out to be extremely useful as atmospheric tracers in the studies of meteorological phenomena.

The results of this work written in preprint form are attached to this report as APPENDIX VIII.

(9) Radioactive Strontium Fallout Measured in Rain Samples Collected at Fayetteville, Arkansas; Ankara, Turkey; and Tokyo, Japan, after the 14th Chinese Nuclear Explosion, (P. Y. Daniel).

The ratiostrontium data that were measured from three different sampling stations around the world are presented as APPENDIX IX. A discussion of these results are also given in APPENDIX V.

(10) Cs^{137}, Sr^{90}, Cs^{137}/Sr^{90} Ratio and the Bimonthly Average Cs^{137}/Sr^{90} Ratio in Rains Collected at Fayetteville, Arkansas, from January 24, 1967, through June 27, 1969, (R. D. Sherrill and N. G. Sumerlin).

Radiocesium and radiostrontium data were remeasured in 137 rains during the period from January 24, 1967, through June 27, 1969, and the results are presented as APPENDIX X. A discussion of these results are also presented in APPENDIX VI.

PART II. INSTRUMENTATION

The low-level counting systems (Omnigard BLB 5020's and the Beckman and Canberras) operated very well during the current contract year. The much older counting systems (Tracerlab's CE14SL's) after undergoing extensive repair in the previous contract year also operated well during the current contract year.

A Nuclear Chicago ND 150M pulse height analyzer was used with an Ortec surface barrier detector for the counting of the Po^{210} samples. The Canberra Ge(Li) solid-state detector system also was used effectively during the year. The IBM 7040 computer at the University of Arkansas Computing Center was available for data reduction and gamma spectroscopy work.

PART III. PUBLICATIONS AND PARTICIPATION IN MEETINGS

(1) J. R. Noyce, D. T. Moore, J. N. Beck and P. K. Kuroda, Behavior of Radiostrontium in Rain and Air after the Ninth Chinese Atmospheric Test, J. Geophys. Res., 78, 1419 (1973).

(2) J. D. Sherwood, D. T. Moore and P. K. Kuroda, Radioactive Cerium Isotopes: The Fallout from Recent French and Chinese Nuclear Weapons Tests, Health Physics, 24, 491 (1973).

(3) J. R. Noyce, D. T. Moore, J. D. Sherwood, P. R. Daniel, J. N. Beck and P. K. Kuroda, Fallout from Nuclear Weapons Testing and Interhemispheric Transport of Nuclear Debris, Health Physics, 25, 109 (1973).

(4) D. T. Moore, J. N. Beck, D. K. Miller and P. K. Kuroda, Radioactive Hot Particles from the Recent Chinese Nuclear Weapons Tests, J. Geophys. Res., 78, 7039 (1973).

(5) M. B. Gavini, A. Nevissi and P. K. Kuroda, Long-Lived Alpha-Emitters in the Atmosphere: Polonium-210 and Plutonium-239, presented at the 166th ACS National Meeting, Chicago, Illinois, August 29, 1973.

(6) J. N. Beck, R. W. Holloway, N. G. Sumerlin and P. K. Kuroda, Environmental Contamination Caused by the Recent Chinese Nuclear Detonations, presented at the 9th Midwest Regional ACS Meeting, Lawrence, Kansas, October 26, 1973.

PART IV. PERSONNEL

Dr. Patsy Y. Daniel received her PhD degree in Chemistry from the University of Arkansas. The title of her PhD dissertation was "Radioactive Strontium Isotopes in the Atmosphere". She has now accepted a research position at Westinghouse Electric Corporation, Bettis Atomic Power Laboratory, West Mifflin, Pennsylvania.

Dr. J. N. Beck, B. S. Northeastern State College, Tahlequah, Oklahoma, 1963.
M. S. University of Arkansas, Fayetteville, Arkansas, 1966.
PhD University of Arkansas, Fayetteville, Arkansas, 1971.

Mr. M. B. Gavini, B.S., A. C. College, Guntur, A.P., So. India, 1970.
M. S., Andhra University, Waltair, A.P., So. India, 1972.

Mr. R. W. Holloway, B. S. Harding College, Searcy, Arkansas, 1967.

Mr. R. D. Sherrill, B. S. Kansas State College, Pittsburg, Kansas, 1968.
M. S. Kansas State College, Pittsburg, Kansas, 1969.

Mr. N. G. Sumerlin, B. S. Ouachita Baptist University, Arkadelphia, Arkansas, 1972.

Mrs. Marjorie Bright, Secretary.

Miss Judy Compton, part-time Secretary.

APPENDIX I

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APPENDIX II

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APPENDIX III

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RESIDENCE TIME OF STRONTIUM-90 IN THE ATMOSPHERE

Patsy Y. Daniel(*), A. Nevissi(**), J. L. Meason,
J. N. Beck and P. K. Kuroda

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The concentrations of ^{89}Sr and ^{90}Sr were measured in the entire series of rain and snow samples collected at Fayetteville (36°N, 94°W), Arkansas, during the period between August 1971 and June 1973. Measurements were also made on some rain samples collected at Tokyo (36°N, 140°E), Japan, and at Ankara (48°N, 33°E), Turkey. Three nuclear tests were conducted in Lop Nor (40°N, 90°E), China while this study was being made: the 12th (November 18, 1971), the 13th (January 7, 1972) and the 14th (March 18, 1972). The 'cycling' effect of the fresh debris was clearly observed after the 14th Chinese test: peaks in the $^{89}Sr/^{90}Sr$ ratio occurred successively at Fayetteville (94°W), Ankara (33°E), and Tokyo (140°E), in that order, indicating that the freshly injected debris travelled eastward circling the world at least twice until the distribution of the fission products in the troposphere became more or less uniform after about two months. Values of the residence time of ^{90}Sr in the troposphere were calculated from the $^{89}Sr/^{90}Sr$ ratio data. While the debris from the 12th and the 13th tests

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yielded rather short apparent residence times of about 8 ± 3 days and 12 ± 3 days, respectively, much longer and approximately concordant mean residence times were obtained from the $^{89}Sr/^{90}Sr$ and $^{210}Po/^{210}Pb$ ratios measured simultaneously in many of the rain samples collected after the 14th test. The results indicate that most of the ^{210}Po originated from the decay of ^{222}Rn in the troposphere. The experimental data obtained in this work are consistent with the mean residence time of ^{90}Sr in the stratosphere of approximately one year.

INTRODUCTION

We reported many years ago that the residence time of ^{90}Sr in the troposphere can be calculated from the observed $^{89}Sr/^{90}Sr$ or $^{140}Ba/^{90}Sr$ ratios in a series of rain samples collected after a nuclear explosion (Kuroda, 1958; Beck and Kuroda, 1966). This method was first applied to the debris from the November 8, 1957, British test and the December 28, 1957, Soviet test and yielded values ranging from 7 to 10 days (Kuroda, 1958). It appeared at that time that the values thus obtained were considerably shorter than the generally accepted value of 30 days (see, for example, Stewart et al, 1956; Libby, 1956a,b; Goel et al, 1959; Lal, 1959). Beck and Kuroda (1966), however, used the same method and found that the mean residence time in the troposphere of the ^{90}Sr from the first Chinese test (October 16, 1964) to be about 30 days. Thein and Kuroda (1967) used the $^{141}Ce/^{144}Ce$ ratios and calculated a mean residence time in the troposphere of ^{144}Ce injected by the second Chinese test (May 14, 1965) to be about 40 days. Swindle and Kuroda (1969) found a value of 25 days for the tropospheric residence time of ^{90}Sr from the 5th (December 28, 1966) and the 6th (June 17, 1967) Chinese tests (see also, Reynolds et al, 1967). However, the $^{89}Sr/^{90}Sr$ ratio data for the 5th test indicated the presence of an additional compartment in the upper troposphere. Using a three-compartment model of the atmosphere, Swindle and Kuroda (1969) stated that their data could be best explained if the transfer rates were such that they corresponded to the mean residence times in the stratosphere, upper troposphere, and lower troposphere of 1.4 years, 50 days, and 25 days, respectively. In their six-compartment model of the atmosphere, Krey and Krajewski (1970) adopted a value of 30 days for the mean tropospheric

residence time of ^{90}Sr. Meanwhile, Fry and Menon (1962) measured the ratios of ^{210}Bi to ^{210}Pb in a number of rain samples collected at Fayetteville, Arkansas, and reported an average value of 6 days for the mean residence time of ^{210}Pb in the troposphere.

Recently, Poet et al (1972) and Moore et al (1973a, b) reported that the results from their studies on the long-lived radon daughters in air filter and rain samples indicated a much shorter residence time for the troposphere: about 4 days from measurements in ground level air and 7 days from measurements in precipitation. The latter value was in remarkable agreement with the shorter residence times obtained by Kuroda (1958) and Fry and Menon (1962). Moreover, Moore et al (1973b) concluded from their studies on ^{90}Sr in the atmosphere that aerosols in the lower stratosphere below 15 km have a mean residence time of about 1 to 2 months. This short stratospheric residence time of 1 to 2 months was similar to the value of 50 days assigned by Swindle and Kuroda (1969) for the residence time of ^{90}Sr in the 'upper' troposphere.

Poet et al (1972) and Moore et al (1973a,b) further pointed out that most of the ^{210}Po in the tropsophere is of continental surface origin and does not originate from the decay of ^{222}Rn within the troposphere. Marenco and Fontan (1973) and Hartwig (1973), however, reported that Poet et al (1972) may not have interpreted their data properly (see also the replies given by Moore et al, 1973c,d). A similar view was expressed by Nevissi et al (1973), who measured ^{210}Pb, ^{210}Bi and ^{210}Po activities in a series of rain samples and found that the values of residence times calculated from the $^{210}Bi/^{210}Pb$ and $^{210}Po/^{210}Pb$ ratios were essentially concordant in a majority of cases.

During the period between August 1971 and June 1973, we measured the $^{89}Sr/^{90}Sr$ ratios in a total of 139 rain samples collected at Fayetteville, Arkansas. Three nuclear tests were conducted in China while this study was being made: the 12th (November 18, 1971), the 13th (January 7, 1972) and the 14th (March 18, 1972). The $^{210}Po/^{210}Pb$ ratios have been measured in many of the same rain samples and this provided us with an excellent opportunity to see whether or not the residence times calculated from the long-lived radon daughters and the fission product ratios are concordant. The experimental data obtained in this work support the view that the mean residence time of ^{90}Sr, ^{210}Po, etc. in the troposphere is about 30 days, as reported earlier by us and many other investigators.

EXPERIMENTAL

Samples of rain and snow were collected by means of a sampling system installed on the roof of the Chemistry Building of the University of Arkansas in Fayetteville (36°N, 94°W), Arkansas. The Tokyo rain samples were collected, and evaporated by one of us (P.K.K.) at the Institute of Geochemistry, Central Meteorological Agency, Tokyo (36°N, 140°E), Japan, and were brought back to our laboratories for radiochemical analysis. Radiochemical procedures for the strontium isotopes employed were essentially the same as those described by Hodges (1963). The radiochemical procedure used for ^{210}Pb and ^{210}Po were the same as those employed by Wallace (1965). Tracerlab low-level β-counting system (BLB 5020S Omniguard) was used for the radioactivity measurements. The polonium mounted carrier-free was alpha counted using an Ortec surface barrier detector connected to a Nuclear Chicago ND 150M pulse height analyzer.

RESULTS AND DISCUSSION

Fig. 1 shows the monthly and bi-monthly average ^{90}Sr concentrations ($\bar{C}$) calculated by dividing the values of the total monthly or bi-monthly ^{90}Sr fallout (ΣF expressed in the unit of picocuries per square meter) by the values of total monthly rainfall (ΣR expressed in the unit of millimeters):

$$\bar{C} = \Sigma F/\Sigma R \qquad \text{----------(1).}$$

The average ^{90}Sr concentration in rain during the early part of 1972 was about 1 picocurie per liter and that in the early part of 1973 seems to have decreased to somewhat less than a half picocurie per liter as would be expected from a mean stratospheric residence time of approximately one year (Kuroda et al, 1960, 1962; Feely, 1960; Volchok, 1967; Krey, 1968; Fabian et al, 1968; Peirson and Cambray, 1967; Cooper et al, 1970; Beck et al, 1971).

Fig. 2 shows the $^{89}Sr/^{90}Sr$ ratio data.(*) During the fall months of 1971, the $^{89}Sr/^{90}Sr$ ratio in rain was steadily decreasing after reaching a small maximum in September. This pattern of variation of the $^{89}Sr/^{90}Sr$ ratio in rain suggests that some of the fresh debris from the August 1971 French tests entered the Northern Hemisphere (see, for example Peirson et al, 1967, 1970; Palmer, 1969, Noyce et al, 1973). The $^{89}Sr/^{90}Sr$ ratios

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(*) The individual values of the ^{89}Sr and ^{90}Sr concentrations in rain are given in the PhD dissertation by Patsy Y. Daniel (1974, University of Arkansas). A complete list of the ^{89}Sr and ^{90}Sr concentrations and the $^{89}Sr/^{90}Sr$ ratios in one thousand rain and snow samples analyzed at Fayetteville, Arkansas, during the period between October 31, 1958, and November 27, 1972, has been compiled by Daniel (1973).

in the troposphere as well as the stratosphere were decreasing steadily due to radioactive decay with a half-life of about 50 days toward a value indicated by S_{12} (= about 1) in Fig. 2 shortly before the 12th Chinese test occurred on November 18, 1971. The 12th Chinese test caused a sharp increase in the $^{89}Sr/^{90}Sr$ ratio in rain suggesting a tropospheric injection of the fresh debris. The $^{89}Sr/^{90}Sr$ ratio in rain decreased rapidly and was approaching a value represented by S_{13} (= about 0.5) on January 7, 1972, when the 13th Chinese test occurred. Double peaks observed in the $^{89}Sr/^{90}Sr$ ratio in rain after the 12th test can be interpreted as due to the fact that the fresh debris travelled eastward and circled the world twice (Kuroda et al, 1965; Cooper and Kuroda, 1966; Cooper et al, 1970; Thein and Kuroda, 1967; Thein et al, 1968; Baugh et al, 1967; Chen et al, 1970; Yoshikawa et al, 1968). Only a single peak in the $^{89}Sr/^{90}Sr$ ratio in rain was observed after the 13th test, but this happened because there was a dry spell in Arkansas during the month of January 1972. Moore, Beck et al (1973) have measured the daily variation of the number of radioactive particles in the ground-level air after the 13th test and showed there were two peaks (the first on January 12, 1972, and the second on February 1, 1972) corresponding to the first and second arrivals of the fresh debris at Fayetteville.

The variation of the $^{89}Sr/^{90}Sr$ ratio observed in rain after the 14th Chinese test of March 18, 1972, is of special interest since measurements were made at three locations widely apart from each other: Tokyo (36°N, 140°E), Fayetteville (36°N, 94°W), and Ankara (40°N, 33°E). Unfortunately the rain samples collected at Tokyo and Ankara were not immediately processed for analysis and this resulted in large experimental uncertainties

in the radiostrontium isotope data for the Tokyo and Ankara rains. Nevertheless, a general pattern of global circulation of particulate matter in the northern troposphere can be seen clearly from these data. Although we were unable to obtain the data points covering the period of the first arrival at Tokyo, Fig. 2 indicates the first arrivals of the fresh debris occurred at Fayetteville and Ankara, in that order, toward the end of March 1972. While the $^{89}Sr/^{90}Sr$ ratio in rain was reaching a minimum value during the first half of the month of April at Fayetteville, a sharp increase in the $^{89}Sr/^{90}Sr$ ratio was occurring at Tokyo half-way around the world signalling the second arrival of the debris there. The debris which reached Tokyo for the second time on April 7 and 8 seems to have again passed over the Pacific Ocean and reached the United States causing a pronounced second peak in the $^{89}Sr/^{90}Sr$ ratio in rain on around April 15 to 20. The second arrival of the debris at Ankara was not seen, due probably to the fact that there were no rainfalls at that time at Ankara when the debris was arriving from the west for the second time. It is also possible that we missed the peak because the rain samples were analyzed many months after the samples were collected and there were large experimental uncertainties involved in the measurements of the $^{89}Sr/^{90}Sr$ ratios.

Fig. 2 shows that the $^{89}Sr/^{90}Sr$ ratios in the troposphere became more or less uniformly mixed after a month or two throughout the northern temperate zone and eventually decreased at a rate corresponding to the half-life of ^{89}Sr, as shown by the straightline S'_{14} S''_{14} in Fig. 2. It thus appears as if the $^{89}Sr/^{90}Sr$ ratio in the stratosphere and in the troposphere became nearly the same after a few months after the 14th Chinese test, and the line S'_{14} S''_{14} represents this ratio.

The mean tropospheric residence time of ^{90}Sr car be calculated from the $^{89}Sr/^{90}Sr$ ratio in the following manner (Kuroda, 1958; Beck and Kuroda, 1966):

$$1 - \underline{x} = \frac{-S_o + T \exp(\lambda_A - \lambda_B)t}{T_o - S_o} \qquad \text{----------(2),}$$

where S_o and T_o are the $^{89}Sr/^{90}Sr$ ratios in the stratosphere and in the troposphere, respectively, at time t = 0 (time of nuclear explosion); T is the $^{89}Sr/^{90}Sr$ ratio in rain observed $\underline{t}$ days after the nuclear explosion, λ_A and λ_B are the decay constants of ^{89}Sr and ^{90}Sr, respectively, $\underline{x}$ is the fraction of ^{90}Sr which originated in the stratosphere and hence $1 - \underline{x}$ is the fraction which originated in the troposphere. Beck and Kuroda (1966) showed that $1 - \underline{x}$ decreases exponentially with time $\underline{t}$ and hence by plotting the values of $1 - \underline{x}$ against $\underline{t}$, a value of the mean tropospheric residence time can be obtained from the slope of the line representing the decrease of $1 - \underline{x}$ with time.

If this method is applied to the observed $^{89}Sr/^{90}Sr$ ratio data after the 12th and 13th Chinese tests, relatively short residence times of about 8 ± 3 days and 12 ± 3 days, respectively, are obtained. These values are in essential agreement with the value of 7 to 10 days obtained earlier by Kuroda (1958) and probably do not represent the true mean tropospheric residence time of ^{90}Sr. We have seen in the $^{89}Sr/^{90}Sr$ ratio data obtained after the 14th Chinese test that the fresh debris injected at Lop Nor (40°N, 90°E) travelled eastward and circled the world in three to four weeks, and it takes at least two months for the troposphere to become reasonably well mixed with respect to the $^{89}Sr/^{90}Sr$ ratio. During the

first few weeks after a nuclear explosion, the fresh debris is coming and going from the west to the east, and the $^{89}Sr/^{90}Sr$ ratio in the troposphere observed at one locality of the world shows a zig-zag pattern. Meanwhile, the fresh debris are diffusing away in all directions and by the time the debris arrive at one location for the second time, the debris become much diluted with older debris. For this reason, the mean residence time calculated from the $^{89}Sr/^{90}Sr$ ratios in rain tends to be shorter than the actual value. The time intervals between the 12th, 13th and 14th Chinese tests (about 50 days and 70 days, respectively) were also perhaps too short for a reasonably accurate determination of the mean tropospheric residence time of ^{90}Sr.

On the other hand, there was no more testing by the Chinese government after the 14th test during the rest of the year 1972. Although the French government tested a few nuclear devices in the Southern Hemisphere during the months of June and July 1972, the effect on the $^{89}Sr/^{90}Sr$ ratio in rain in the northern troposphere seems to have been negligible as indicated by the fact that ^{140}Ba was not detected in rain samples collected at Fayetteville during and after the French test series.

The fact that most of the $^{89}Sr/^{90}Sr$ ratios in rain observed during the months of April, May, June and July lie above the line S'_{14} S''_{14} suggests that appreciable quantities of the strontium isotopes in the fresh debris injected into the troposphere by the 14th Chinese nuclear test were still remaining in the troposphere. If so, one should be able to calculate values of the residence time of ^{90}Sr from the observed $^{89}Sr/^{90}Sr$ ratios in rain by the use of equation (2). The values of residence time thus obtained may not be very accurate, but it seems to be of great interest

to compare them with the values of residence time calculated from the $^{210}Po/^{210}Pb$ ratios in many of the same rain samples analyzed in our laboratories. The residence time was calculated from the $^{210}Po/^{210}Pb$ ratio by the equation first used by Lehmann and Sittkus (1959):

$$\frac{N_F\lambda_F}{N_D\lambda_D} = \frac{(\tau_R)^2}{(\tau_R + \tau_F)(\tau_R + \tau_E)} \qquad \text{----------(3),}$$

where N's are the numbers of atoms, $\underline{\lambda}$'s are the decay constants, $\underline{\tau}$'s are the reciprocals of the decay constants and the removal constants ($\tau_R = 1/\lambda_R$), and the subscripts D and F refer to RaD (^{210}Pb) and RaF (^{210}Po). Table 1 compares the results.

The agreement between the residence times calculated from the $^{89}Sr/^{90}Sr$ ratio and the $^{210}Po/^{210}Pb$ ratio in the same rain appears to be unexpectedly good, as shown in Table 1. As clearly pointed out by Marenco and Fontan (1973), however, the residence time depends on the elements considered: origin, radioactive period and distribution. Thus, there is no reason why the residence times obtained from the $^{89}Sr/^{90}Sr$ ratio and the $^{210}Po/^{210}Pb$ ratio should agree perfectly. ^{210}Po and ^{210}Pb are being continuously supplied by the radon in the troposphere. Most of the ^{89}Sr and ^{90}Sr, on the other hand, were injected all at once by the 14th Chinese test conducted at Lop Nor (40°N, 90°E) on March 18.

Nevertheless, after the month of May, 1972, the troposphere became fairly uniformly mixed with respect to the $^{89}Sr/^{90}Sr$ ratio. It thus appears to be reasonable to conclude that for a few month's period after May, ^{90}Sr and ^{210}Po behaved more or less as equally good atmospheric tracers. It is interesting to note in Table 1 that the residence times calculated from

the $^{89}Sr/^{90}Sr$ ratio and the $^{210}Po/^{210}Pb$ ratio for the May 8, 1972, Fayetteville rain are both quite short and are essentially concordant: thus 9^{+5}_{-9} days and 17^{+2}_{-4} days, respectively. It is thus possible that we are dealing here with an air mass, which has picked up both the fresh bomb debris and the long-lived radon daughters a short time prior to its arrival at Fayetteville. Because of the relatively small value of the $^{89}Sr/^{90}Sr$ ratio in the May 8, 1972, Fayetteville rain, it appears as if there is an additional peak in the $^{89}Sr/^{90}Sr$ ratio in rain during the early part of May, 1972. But the peak value in the $^{89}Sr/^{90}Sr$ ratio in rain observed on May 12, 1972, perhaps should not be interpreted as an indication of the arrival of a third 'wave' of fresh debris at Fayetteville. In this connection, it is interesting to note that Moore, Beck et al (1973) found negligibly low particle concentrations in the ground level air at Fayetteville during May 10-17, 1972.

Moore et al (1973b) have recently concluded from their studies on ^{90}Sr in the atmosphere that aerosols in the lower stratosphere below 15 km have a mean residence time of about 1 to 2 months. This short stratospheric residence time is about the same as the values of tropospheric residence times calculated by us from the $^{89}Sr/^{90}Sr$ ratio and the $^{210}Po/^{210}Pb$ ratio in rain. A question thus arises that perhaps our residence time values refer to the lower stratosphere, rather than the troposphere. As reported by Moore et al (1973b), the tropopause height varied between 9.5 km and 15.9 km depending on the season and geographical locations during the period when their samples were collected. It is thus quite possible that our 'upper' troposphere corresponds to Moore et al's 'lower' stratosphere below 15 km.

As pointed out by Marenco and Fontan (1973), however, the mean tropospheric residence time of an aerosol depends mainly on its formation altitude, and ^{210}Pb and ^{210}Bi are representative of the lower troposphere, while ^{210}Po is representative of the whole troposphere (see also, Hartwig, 1973). The fact that essentially concordant residence times are obtained from the $^{89}Sr/^{90}Sr$ ratio and the $^{210}Po/^{210}Pb$ ratio in rain thus seems to rule out the possibility that there is a large excess of ^{210}Po in the troposphere, which does not originate from the decay of ^{222}Rn within the atmosphere.

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TABLE 1. Radiostrontium isotopes and long-lived radon daughters in rain and their residence times in the troposphere.

Date	^{89}Sr	^{90}Sr	^{210}Pb	^{210}Po	Mean residence time (days) from $^{89}Sr/^{90}Sr$	Mean residence time (days) from $^{210}Po/^{210}Pb$
	(10^{-12} curies/liter)					
May 1, 1972	10.6±1.5	0.44±0.10	2.52±0.21	0.35±0.04	24^{+7}_{-5}	40^{+8}_{-9}
May 7-8, 1972	15.6±1.8	1.3 ±0.1	4.10±0.30	0.22±0.02	9^{+5}_{-9}	17^{+2}_{-4}
May 12, 1972	27.7±1.9	1.1 ±0.1	4.26±0.46	0.45±0.05	34^{+5}_{-4}	31^{+6}_{-2}
May 22-23, 1972	72.6±5.4	4.6 ±0.7	12.50±0.66	1.45±0.15	27^{+8}_{-7}	34^{+7}_{-5}
May 26-27, 1972	27.5±2.2	2.1 ±0.1	7.00±0.44	0.73±0.07	25^{+4}_{-3}	29^{+5}_{-8}
May 28, 1972	25.8±2.9	1.8 ±0.4	6.48±0.72	0.81±0.08	30^{+9}_{-10}	37^{+6}_{-8}
June 14, 1972	8.2±0.8	0.5 ±0.2	1.50±0.20	0.35±0.07	55^{+10}_{-12}	68^{+36}_{-6}
June 27, 1972	7.6±0.9	0.64±0.12	8.70±0.60	0.78±0.16	53^{+12}_{-10}	38^{+14}_{-9}
June 30, 1972	8.4±0.9	0.84±0.11	4.90±0.40	0.65±0.13	49^{+4}_{-9}	37^{+12}_{-9}

TABLE 1 (Continued)

Date	^{89}Sr	^{90}Sr	^{210}Pb	^{210}Po	Mean residence time (days) from $^{89}Sr/^{90}Sr$	Mean residence time (days) from $^{210}Po/^{210}Pb$
	(10^{-12} curies/liter)					
July 1, 1972	8.1±0.8	0.82±0.15	2.50±0.30	0.28±0.06	49^{+12}_{-11}	31^{+13}_{-8}
July 3, 1972	17.1±3.1	1.0 ±0.1	4.90±0.40	0.51±0.10	98^{+32}_{-30}	30^{+9}_{-7}
July 28, 1972	5.3±0.7	0.90±0.09	7.70±0.50	0.90±0.20	51^{+11}_{-9}	33^{+10}_{-8}
July 29, 1972	5.7±0.9	0.79±0.16	5.20±0.25	0.63±0.13	66^{+21}_{-18}	34^{+10}_{-12}
Aug. 4, 1972	0.71±0.06	0.14±0.01	2.00±0.40	0.36±0.07	51^{+7}_{-9}	52^{+31}_{-18}
Aug. 14, 1972	3.3±0.6	0.67±0.13	8.60±0.70	0.69±0.14	62^{+17}_{-23}	53^{+19}_{-13}

FIGURE CAPTION

Fig. 1. Monthly and bi-monthly average ^{90}Sr concentrations in rain at Fayetteville, Arkansas, during the period between August 1971 and June 1973.

Fig. 2. Variation of the $^{89}Sr/^{90}Sr$ ratio in rain at Fayetteville (36°N, 94°W), Ankara (40°N, 33°E), and Tokyo (36°N, 140°E). Three nuclear tests were conducted in Lop Nor (40°N, 90°E), China, while this study was being made: the 12th (November 18, 1971), the 13th (January 7, 1972), and the 14th (March 18, 1972).

● : Fayetteville

⊙ : Tokyo

⊗ : Ankara

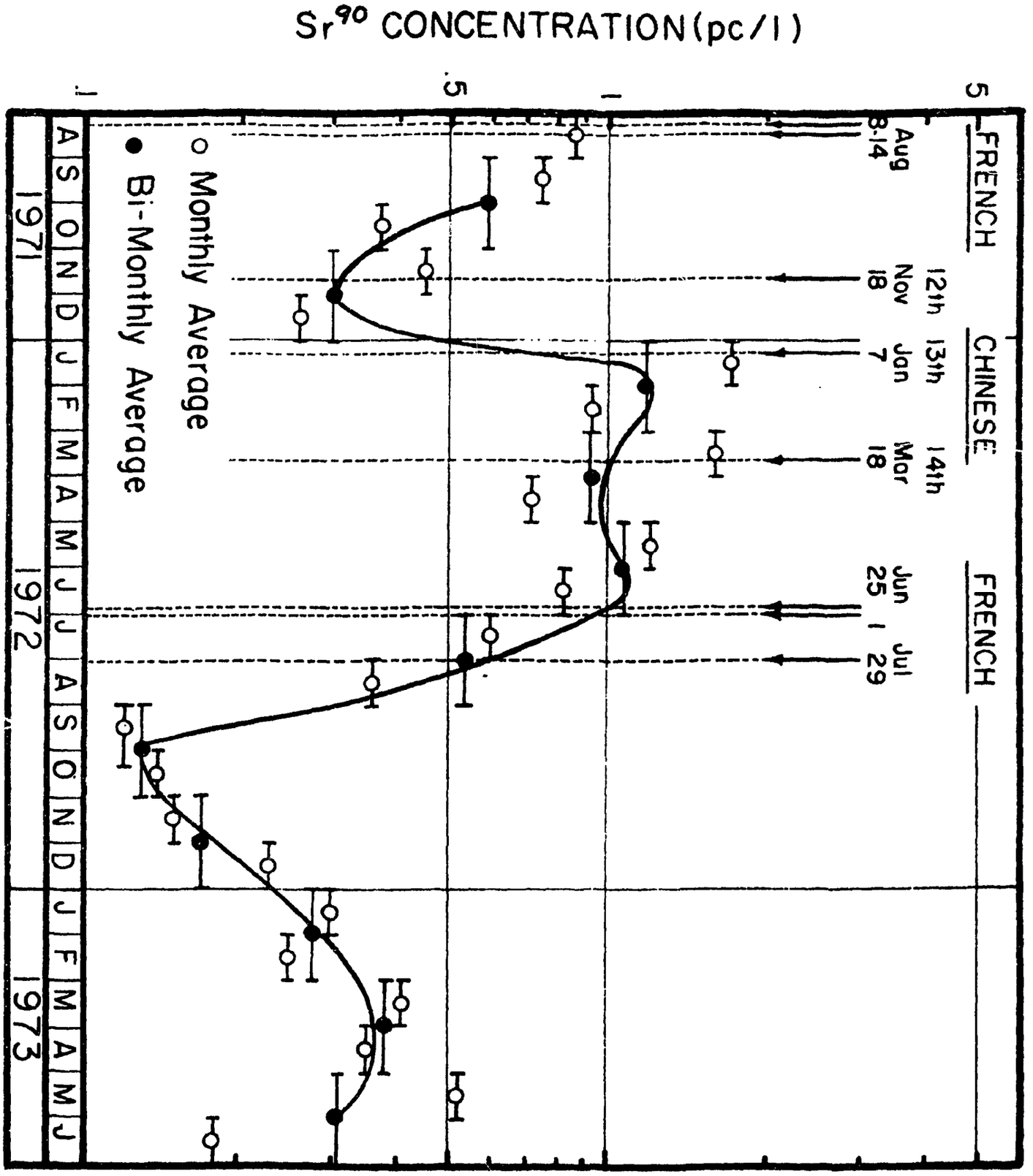
Sr90 CONCENTRATION (pc/l)
5
1
FRENCH
CHINESE
Aug 8-14
12th Nov 18
13th Jan 7
14th Mar 18
Jun 25 - Jul 29
FRENCH
o Monthly Average
• Bi-Monthly Average
A S O N D J F M A M J J A S O N D J F M A M J
1971
1972
1973

FIG. 1

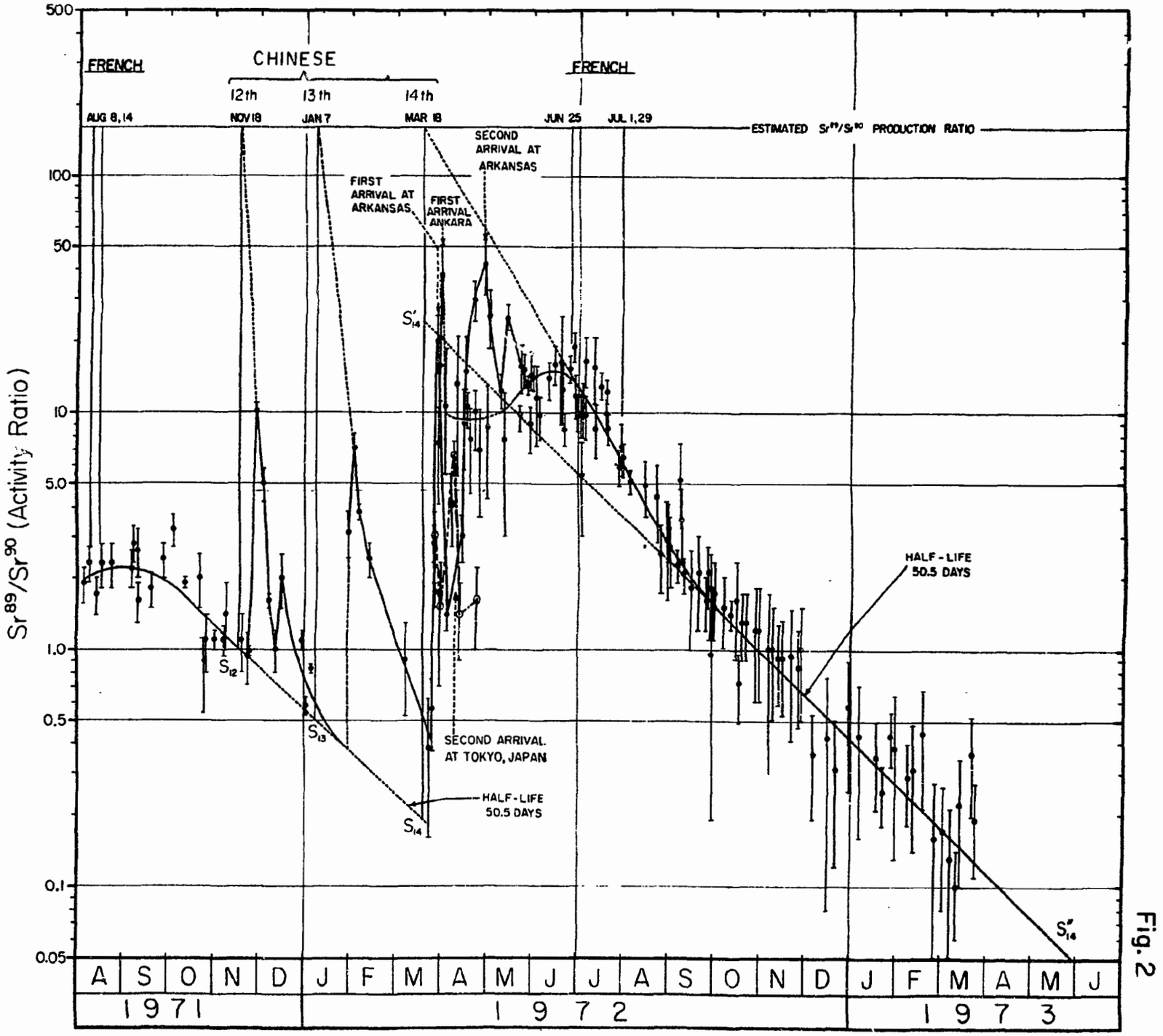
FRENCH
CHINESE
12th
13th
14th
FRENCH
AUG 8, 14
NOV 18
JAN 7
MAR 18
JUN 25
JUL 1, 29
ESTIMATED Sr89/Sr90 PRODUCTION RATIO
SECOND ARRIVAL AT ARKANSAS
FIRST ARRIVAL AT ARKANSAS
FIRST ARRIVAL ANKARA
S'14
S12
S13
S14
SECOND ARRIVAL AT TOKYO, JAPAN
HALF-LIFE 50.5 DAYS
HALF-LIFE 50.5 DAYS
S"14
Sr89/Sr90 (Activity Ratio)
500
100
50
10
5.0
1.0
0.5
0.1
0.05
A S O N D J F M A M J J A S O N D J F M A M J
1971
1972
1973

Fig. 2

APPENDIX VI

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VARIATION OF THE RATIO OF CESIUM-137 TO STRONTIUM-90 IN THE ATMOSPHERE

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Concentrations of ^{137}Cs and ^{90}Sr were measured in a total of 137 individual rain and snow samples collected at Fayetteville, Arkansas, during the period between February 1967 and June 1969. The $^{137}Cs/^{90}Sr$ ratio was found to vary quite considerably: 0.26 (12 April 1968) to 8.2 (8 August 1968). Bi-monthly average $^{137}Cs/^{90}Sr$ ratios in rain fluctuated also from 0.75 (January-February 1968) to 1.9 (July-August 1968). The variation in the $^{137}Cs/^{90}Sr$ ratio seems to be attributable to the difference in the size distributions of particulate matters in rain. ^{137}Cs and ^{90}Sr are depleted in large fallout particles, because of the fairly long half-lives of their precursors: ^{137}Xe (3.82 min) and ^{90}Kr (32.3-sec). A simple calculation, in which the half-lives of the gaseous precursors and the independent and cumulative yields of the fission products belonging to the mass 90 and 137 chains are taken into consideration, shows that extreme values of the $^{137}Cs/^{90}Sr$ ratio in large and small fallout particles expected to be observed in rain may range from about 0.2 to several times the production ratio.

The half-lives of ^{90}Sr and ^{137}Cs, 28 years and 30 years, respectively, are nearly equal and both of these isotopes are known to have high fission yields: for example, 5.77 and 6.15 percent, respectively, for the thermal neutron-induced fission of ^{235}U [Hyde[1]]. One might therefore expect to find the ratio of bomb-produced $^{137}Cs/^{90}Sr$ in the atmosphere to be more or less constant: if these isotopes are produced primarily by the fission of ^{235}U, the ratio should be approximately unity, while it is expected to be somewhat greater than two in the case of the fission of ^{239}Pu. According to Harley et al[2], the ratio of ^{137}Cs to ^{90}Sr atoms in megaton-bomb products is 1.5, and Ehhalt[3] recently used this ratio to calculate the amount of ^{137}Cs deposited from the observed value of the deposition of ^{90}Sr for the entire year 1966 in the 40°-50°N band. Thompson and Lengemann[4] carried out a feasibility study on using the urinary $^{137}Cs/^{90}Sr$ ratio as an effective estimator of ^{90}Sr when environmental levels of fallout are relatively stable.

The fact that there is a considerable scatter in the observed values of the $^{137}Cs/^{90}Sr$ ratio in atmospheric samples, however, has been known for many years. Stewart et al[5] reported that the mean ratio (total ^{137}Cs deposited/total ^{90}Sr deposited) during 1955 to 1957 at several localities of the world ranged from 1.24 to 2.32, but abnormally low values were observed at two locations: Gibraltar (0.51) and Melbourne (0.49). Miyake and his co-workers (Miyake et al[6]; Miyake and Katsuragi[7]; Katsuragi[8]; Miyake[9]) reported that the ratio calculated from the total ^{137}Cs and ^{90}Sr deposited at Tokyo since 1957 was 2.65, while the value for the entire of Japan was 2.28. Miyake[9] also pointed out that the $^{137}Cs/^{90}Sr$ ratio in the ocean was 1.1 to 1.2 and these low values probably resulted from the fact that the half-life of ^{90}Kr (33-sec) is much shorter than that of

^{137}Xe (3.9-min). According to him, a relatively large portion of the bomb-produced ^{90}Sr would be expected to be deposited locally near the test site, while most of the ^{137}Cs atoms tend to escape to the stratosphere.

Daniel et al[10] reported on the ^{137}Cs and ^{90}Sr data in rain obtained in this laboratory for the past several years. Their results indicated that, while the ^{137}Cs and ^{90}Sr concentrations in rain increased in the spring and decreased in the fall (the so-called spring peak of the radioactive fallout), the ^{137}Cs/^{90}Sr ratio also showed marked variations, which could not be explained. We therefore repeated in the present investigation the analysis of ^{137}Cs and ^{90}Sr in a total of 137 individual rain samples collected at Fayetteville, Arkansas, during the period between February 16, 1967, and June 27, 1969. The new set of data which are reported here show that the ^{137}Cs/^{90}Sr ratio in individual rain samples do vary markedly, and the variation of the ratio can not be attributed to experimental errors. An attempt is made in this report to interpret the ^{137}Cs/^{90}Sr ratio data by taking into consideration the half-lives of the gaseous precursors, and the independent and cumulative yields of the fission products belonging to the mass 90 and 137 chains.

EXPERIMENTAL

Samples of rain and snow were collected by means of a permanently installed sampling system on the roof of the Chemistry Building of the University of Arkansas, Fayetteville, Arkansas. After addition of carriers, the samples were evaporated to dryness, transferred to Teflon beakers, and treated with fuming nitric, hydrofluoric, and perchloric acids to dissolve

particulate matter. Strontium was separated from cesium as the nitrate and was radiochemically purified according to the procedure described by Hodges[11]. Carbonate metathesis was performed to insure more complete dissolution of strontium. Repeated precipitations as the carbonate and nitrate were performed. Calcium was removed with an ethanol-ether extraction. Yttrium was separated as the hydroxide after addition of ferric chloride scavenger and ammonium hydroxide. The final precipitate was strontium carbonate. Cesium was separated from impurities as described by Noyce et al[12]. Using BiI_3-HI reagent, cesium was precipitated as $Cs_3Bi_2I_9$. It was then dissolved in nitric acid, iodine was boiled off, zirconium scavenger was added, and bismuth and zirconium hydroxides were precipitated with sodium hydroxide. The supernate was acidified with hydrochloric acid and passed through a Dowex 1x8 (50-100 mesh) holdback ion exchange column. Chloroplatinic acid was added to yield the final precipitate Cs_2PtCl_6.

Samples were counted on a Tracerlab Omni-Guard system which has a background of approximately 0.7 cpm. Strontium samples were counted frequently during the early yttrium ingrowth, then less frequently until maximum activity was obtained after about two months. The cesium samples were counted several times to verify the absence of short lived contaminants.

RESULTS AND DISCUSSIONS

Fig. 1 shows the experimental results. Bi-monthly average $^{137}Cs/^{90}Sr$ ratios in rain are also plotted here. The information on the nuclear explosions which occurred during this period was taken from the

report by Zander and Araskog[20]. The line indicating the production ratio of $^{137}Cs/^{90}Sr$ = 1.5 is based on the report of Harley et al[2]. The value of 1.5 is close to an average of the production ratios which can be calculated from the yield values for thermal-induced fission of ^{235}U [Hyde[1]] and for fast neutron-induced fission of ^{238}U [Yu. A. Zysin et al[13]]: 0.995 and 1.9, respectively.

The values of bi-monthly $^{137}Cs/^{90}Sr$ ratio obtained from the experimental data are generally similar to or smaller than the production ratio 1.5, except for the period of June to August 1968, while the testing was going on in the Southern Hemisphere by the French government. Some of the high $^{137}Cs/^{90}Sr$ ratio values were observed prior to the July 1968 tests, and hence it is not clear as to whether or not the increase in the bi-monthly average $^{137}Cs/^{90}Sr$ ratio observed during this period was due to the French test series.

The high $^{137}Cs/^{90}Sr$ ratio values such as 6.42 and 8.20 observed on June 24 and August 8, 1968, respectively, are much greater than the production ratio of 2.75 for thermal neutron-induced fission of ^{239}Pu, which can be calculated from the ratio of the mass-yields [Hyde[1]]. On the other hand, bi-monthly average $^{137}Cs/^{90}Sr$ ratios smaller than 1 were observed in the early parts of 1967 and 1968, immediately following the Chinese 5th (December 28, 1966) and 7th test (December 24, 1967). In the case of the 8th Chinese test (December 27, 1968) a value of bi-monthly average $^{137}Cs/^{90}Sr$ ratio of 0.8 was observed during the period of March-April 1969. These low values of $^{137}Cs/^{90}Sr$ ratio in rain are likely to be related to the presence of radioactive "hot particles" in rain shortly after the nuclear explosions. Thein et al[14] have reported on the daily variation of the number of particles in the ground-level air

at Fayetteville, Arkansas, during the period of December 28, 1967, to January 28, 1968. They found many of the particles were larger than few-micron size and were also present in rain samples collected during this period. They reported that among the rare-earth radionuclides, ^{91}Y and ^{141}Ce were often depleted relative to other rare-earths in fallout samples containing "hot particles" and explained this as due to the existence of gaseous precursors in the mass 91 and 141 chains. Pronounced fractionation phenomena were also observed for fission products at mass numbers A = 89 (Sr), A = 131 (I), and A = 140 (Ba). Their data also indicated that ^{89}Sr and ^{140}Ba were enriched in some rain samples containing a large number of small particles (less than 1-2 micron size) [see also Kuroda *et al*[15]].

It is not a simple matter to explain the wide scatter in the values of $^{137}Cs/^{90}Sr$ ratio in individual samples of rain. The range of variation covers the extremes of $^{137}Cs/^{90}Sr$ = 0.26 (April 12, 1968) and 8.2 (August 8, 1968) and it is hard to conclude that these variations are entirely due to experimental errors. It is more reasonable to assume that these extreme values are not accidental, but that they represent values expected to result from a certain mechanism.

Moore *et al*[16] have recently reported on the processes which produce the depletion in fallout particles of fission products belonging to volatile decay chains. We shall attempt to follow the line of approach taken by these investigators. Both ^{137}Cs and ^{90}Sr have rare gas precursors ^{137}Xe (3.82-min) and ^{90}Kr (32.3-sec):

$$\text{3.82-min } {}^{137}\text{Xe} \xrightarrow[\underline{6.00\%}]{\beta^-} \text{30-yr } {}^{137}\text{Cs} \xrightarrow[\underline{6.15\%}]{\beta^-}$$

and

$$\underset{\underline{5.00\%}}{\text{32.3-sec } {}^{90}\text{Kr}} \xrightarrow{\beta^-} \text{2.7 min } {}^{90}\text{Rb} \xrightarrow{\beta^-} \underset{\underline{5.77\%}}{\text{28-yr } {}^{90}\text{Sr}} \xrightarrow{\beta^-}$$

where the percentage values are the cumulative fission yields from the thermal induced fission of ^{235}U.

A minimum $^{137}Cs/^{90}Sr$ ratio is likely to be associated with fairly large fallout particles (say, larger than 10-micron size) which condensed from the fireball within a very short time after a nuclear detonation. In such a particle, the gaseous precursors ^{90}Kr and ^{137}Xe will not be incorporated in any substantial amounts and hence the activities in the mass 90 and 137 chains observed in it would be those corresponding to the differences between the cumulative yields of ^{90}Kr (5.0 percent) and ^{90}Sr (5.77 percent), and ^{137}Xe (6.00 percent) and ^{137}Cs (6.15 percent), respectively. Hence, a minimum $^{137}Cs/^{90}Sr$ ratio expected in such a particle would be

$$(^{137}Cs/^{90}Sr)_{min} = \frac{(6.15 - 6.00)}{(5.77 - 5.0\ )} \cdot \frac{28}{30} = 0.18.$$

This value is similar to the smallest value (0.26) of the $^{137}Cs/^{90}Sr$ ratio observed in the rain of April 12, 1968.

A maximum $^{137}Cs/^{90}Sr$ ratio value is likely to be associated with very small fallout particles which condensed from the fireball after a relatively long time compared to the half-lives of the rare gas precursor isotopes. The daughters of ^{90}Kr and ^{137}Xe, ^{90}Rb and ^{137}Cs, respectively, have to be incorporated onto the surfaces of fine particles. It is difficult to calculate a maximum $^{137}Cs/^{90}Sr$ ratio value associated with very small

fallout particles, but it is quite likely that the ratio may reach a value several times the production ratio.

If the variation of the $^{137}Cs/^{90}Sr$ ratio in individual samples of rain is caused by a process such as described above and is related to the particle size distribution of the materials brought down by the atmospheric precipitation, similar variations may be observed in the case of a single rainfall. This indeed appears to be the case judging from the data obtained by Noyce et al[12], who studied temporal distributions of radioactivity and $^{89}Sr/^{90}Sr$ ratios during two rainstorms during 1968. They measured radionuclide concentrations for ^{89}Sr, ^{90}Sr, ^{144}Ce, and ^{137}Cs in sequentially sampled rainstorms of July 21 and August 14, 1974. The $^{137}Cs/^{90}Sr$ ratios were calculated from the data reported by Noyce et al[12] and are shown in Table 1.

According to Noyce et al[12], the storm of July 21, 1969, had its cloud tops penetrating above the tropopause at a height of 14.5 km. On the other hand, the storm of August 14, 1969, was less intense and of longer duration, the consequence of stratified stable-type stationary front weather. The few scattered and disorganized thunderstorms sighted in the general area were entirely tropospheric in nature, having tops at about 8.5 km. It is interesting to note here that while a relatively small variation (0.85 to 2.50) in the $^{137}Cs/^{90}Sr$ ratio was obtained in the July 21, 1969, storm, the ratio fluctuated between the extreme values of 0.54 and 6.15 in the case of the August 14, 1969, rain. This marked difference in the pattern of variations of the $^{137}Cs/^{90}Sr$ ratio in the two rainstorms may be attributed to the fact that while the storm of July 21, 1969, brought down the fission products more or less directly from the stratosphere according to the mechanism suggested by Dingle[17], the storm

of August 14, 1969, carried with it the debris from a number of stratified tropospheric air masses, which contained particulate matters with various size-distributions. The range of the extreme values of the observed $^{137}Cs/^{90}Sr$ ratio in the August 14, 1969, storm (0.54 to 6.15) is not quite as large as the range of 0.18 to 8.2 determined earlier, but it agrees fairly well with the range observed in the data shown in Fig. 1. In Table 1, it is perhaps also worthy of note that there seems to be a trend that the greater the rate of rainfall, the larger the $^{137}Cs/^{90}Sr$ ratio. This interesting phenomenon may be interpreted as due to the fact that the greater the rate of rainfall, the greater the probability that smaller particulates with high $^{137}Cs/^{90}Sr$ ratios are captured and brought down by the raindrops.

We shall now turn our attention to the variation of the $^{137}Cs/^{90}Sr$ ratio on a global scale. Unfortunately, to our knowledge, the $^{137}Cs/^{90}Sr$ ratios are being measured only for monthly average fallout samples in other laboratories of the world, and hence, we may have to be careful in making a direct comparison of the data. Table 2 compares the $^{137}Cs/^{90}Sr$ data obtained at four locations: Fayetteville, New York HASL[18], Milford Haven in England [Cambray et al[19]], and Tokyo [Miyake[9]]. One obvious conclusion which can be drawn from Table 2 is that the $^{137}Cs/^{90}Sr$ ratio observed at Fayetteville, Arkansas, tends to be much lower than the ratios observed in New York and Tokyo while the ratio at Milford Haven appears to be intermediate. If we are to apply the same principle as we have used to explain the variation of the $^{137}Cs/^{90}Sr$ ratio in rain to interpret the global differences in the observed ratios, we arrive at the interesting conclusion that perhaps large fallout particles injected into

the atmosphere at Lop Nor (40°N, 90°E), China, tend to be precipitated at Fayetteville (36°N, 94°W), which is located roughly 180 degrees away on the same 30°-40°N latitudinal band. Judging from the fact that most of the work related to the fallout of "hot particles" has been reported from our laboratories aside from those in Japan, the above conclusion may have to be considered seriously. On the other hand, it may be due to the differences in sampling and analytical procedures employed in various laboratories and we should perhaps refrain from drawing too many conclusions from a comparison such as shown in Table 2 for the time being.

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TABLE 1. Variation of the $^{137}Cs/^{90}Sr$ ratio in sequentially sampled rainstorms of July 21 and August 14, 1974, calculated from the data reported by Noyce et al[12].

Sample	Fraction No.	Collection time (start), CDT	Rainfall Rate (ℓ/min)	$^{137}Cs/^{90}Sr$ (pc/pc)
(A) Rain of July 21, 1969	(1)	1710	1.6	1.56
	(2)	1715	8.0	2.40
	(3)	1716	8.0	2.50
	(4)	1717	8.0	1.66
	(5)	1718	4.0	1.52
	(6)	1720	4.0	1.20
	(7)	1722	2.7	1.67
	(8)	1725	8.0	1.00
	(9)	1726	8.0	1.27
	(10)	1727	8.0	1.91
	(11)	1728	4.0	2.00
	(12)	1730	4.0	1.07
	(13)	1732	0.9	0.85
	(14)	1741	0.9	1.51
	Bulk Sample	----	---	1.70
(B) Rain of August 14, 1969	(1)	1000	0.33	1.37
	(2)	1006	2.0	2.16
	(3)	1007	2.0	3.36
	(4)	1008	4.0	6.15
	(5)	1010	2.0	4.13
	(6)	1014	2.7	1.89
	(7)	1017	4.0	3.66
	(8)	1019	4.0	1.54
	(9)	1021	2.7	0.97
	(10)	1024	2.7	1.51

TABLE 1 (Continued)

Sample	Fraction No.	Collection time (start), CDT	Rainfall Rate (ℓ/min)	$^{137}Cs/^{90}Sr$ (pc/pc)
	(11)	1027	1.6	0.78
	(12)	1032	0.67	1.45
	(13)	1044	0.20	0.55
	(14)	1124	0.13	0.66
	(15)	1228	0.06	0.54
	Bulk Sample	----	----	1.94

TABLE 2. Monthly average $^{137}Cs/^{90}Sr$ ratios in the fallout observed in different localities of the world.

Period	Fayetteville U.S.A.	New York U.S.A. (a)	Milford Haven, U.K. (b)	Tokyo, Japan (c)
1967				
Jan.	----	----	----	4.1
Feb.	0.37	----	----	2.1
Mar.	----	----	----	3.5
Apr.	1.49	----	----	1.9
May	1.66	----	----	2.2
June	1.56	----	----	2.8
July	----	----	----	3.3
Aug.	1.27	----	----	4.0
Sep.	1.13	----	----	2.7
Oct.	1.89	----	----	2.3
Nov.	1.61	----	----	3.0
Dec.	1.41	----	----	3.0
1968				
Jan.	0.66	----	1.6	2.5
Feb.	1.15	----	1.9	2.0
Mar.	1.01	----	1.5	2.1
Apr.	0.80	----	1.8	2.5
May	1.00	----	1.3	2.4
June	2.01	----	----	1.7
July	1.90	2.22	1.0	2.0
Aug.	1.77	2.01	1.3	2.4
Sep.	1.49	1.71	1.9	2.3
Oct.	1.60	2.30	2.1	2.4
Nov.	1.16	2.26	2.0	1.3
Dec.	1.68	1.93	1.2	2.0

TABLE 2. (Continued)

Period	Fayetteville U.S.A.	New York U.S.A. (a)	Milford Haven, U.K. (b)	Tokyo Japan (c)
1969				
Jan.	2.10	2.59	1.7	1.2
Feb.	1.15	----	2.2	3.0
Mar.	1.04	2.85	1.7	3.0
Apr.	0.64	2.14	0.98	2.7
May	1.33	2.89	1.9	1.5
June	0.89	----	1.4	2.0

a) HASL (18)
b) Cambray et al (19)
c) Miyake (9)

FIGURE CAPTION

Fig. 1. Variation of the $^{137}Cs/^{90}Sr$ ratio in rain during the period between January 1967 and June 1969 at Fayetteville, Arkansas.

● : Individual samples of rain and snow.

◎ : Bi-monthly average $^{137}Cs/^{90}Sr$ ratio.

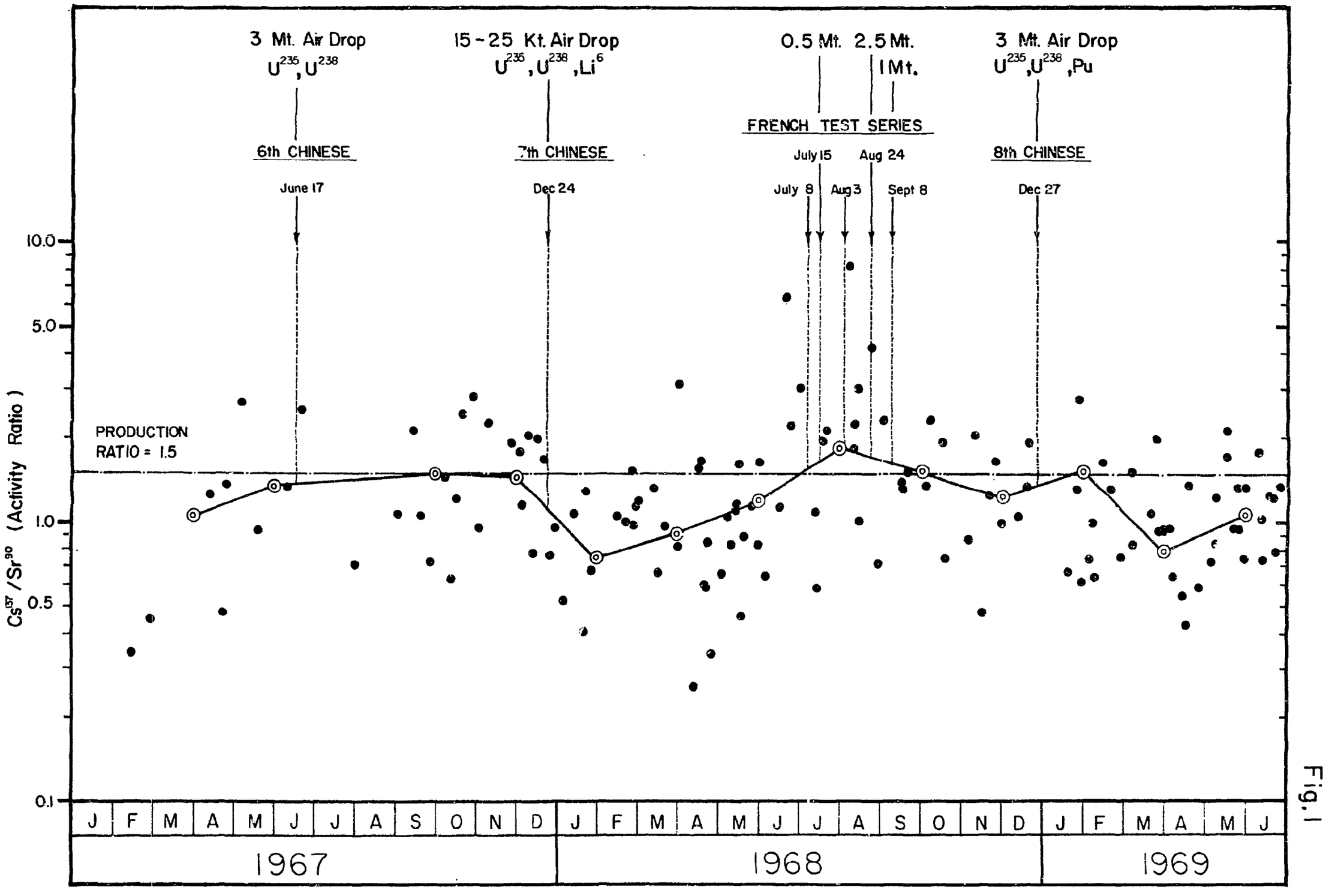
3 Mt. Air Drop
U235, U238
6th CHINESE
June 17
15 - 25 Kt. Air Drop
U236, U238, Li6
7th CHINESE
Dec 24
0.5 Mt. 2.5 Mt.
1 Mt.
FRENCH TEST SERIES
July 15
Aug 24
July 8
Aug 3
Sept 8
3 Mt. Air Drop
U235, U238, Pu
8th CHINESE
Dec 27
PRODUCTION
RATIO = 1.5
Cs137 /Sr90 (Activity Ratio)
10.0
5.0
1.0
0.5
0.1
J F M A M J J A S O N D J F M A M J J A S O N D J F M A M J
1967
1968
1969

Fig. 1

APPENDIX VII

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MEAN RESIDENCE TIMES OF THE LONG-LIVED RADON DAUGHTERS IN THE ATMOSPHERE

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The activities of ^{210}Pb, ^{210}Bi and ^{210}Po were measured in 17 rain samples which were collected at Fayetteville, Arkansas, during the months of April through June 1973, and ^{210}Pb and ^{210}Po only were measured in 20 additional rain samples collected mostly during the months of September through December 1973. In approximately 70 percent of the cases (12 out of 17 rains), the residence time calculated from the $^{210}Bi/^{210}Pb$ and $^{210}Po/^{210}Pb$ ratios were either concordant or near concordant. In about 30 percent of the cases (5 out of 17 rains), the $^{210}Bi/^{210}Pb$ ratios yielded mean residence time values of 3 to 6 days, while the values calculated from the $^{210}Po/^{210}Pb$ ratios varied from 16 to 40 days. The discordant residence times can be shown to result from the mixing of two different air masses: one represented by a short and the other by a much longer residence time of the radon daughters. In order to test this idea, the radionuclides ^{210}Pb and ^{210}Po were sequentially sampled from a rainstorm, which occurred at Fayetteville, Arkansas, on October 6, 1973. The $^{210}Po/^{210}Pb$ ratio was found to vary markedly within a single rainfall and the data indicated that a mixing of two air masses occurred during the rainstorm: one air mass represented by a residence time of about 30 days and the other roughly 300 days.

These values are suggestive of the mean residence times of aerosols for the entire troposphere (about 30 days) and for the entire stratosphere (about 1 year) frequently mentioned by previous investigators.

INTRODUCTION

While the importance of long-lived radon daughters (22-year ^{210}Pb, 5.0-day ^{210}Bi and 138-day ^{210}Po) as atmospheric tracers is now well recognized (Haxel and Schuman, 1955; Lehmann and Sittkus, 1959; Burton and Stewart, 1960; Lambert and Nezami, 1965; Pierson *et al*, 1966; Fry and Menon, 1962; Francis *et al*, 1970; Martell, 1972; Poet *et al*, 1972; Moore *et al* 1973a,b,c,d; Nevissi, 1973; Nevissi *et al*, 1973, 1974; Daniel *et al*, 1974), there remain certain interesting questions which must be dealt with in regard to the general applicability of these radionuclides in the studies of atmospheric phenomena. Poet *et al* (1972), for example, suggested that most of the ^{210}Po in the tropospheric air comes from continental surface sources, and hence $^{210}Po/^{210}Pb$ ratios are not directly related to aerosol residence times in the troposphere. Nevissi *et al* (1973, 1974), on the other hand, measured the activities of ^{210}Pb, ^{210}Bi and ^{210}Po in a large number of rain and snow samples that fell at Fayetteville, Arkansas, and found that in nearly 80 percent of the samples measured, the residence times calculated from the $^{210}Bi/^{210}Pb$ and $^{210}Po/^{210}Pb$ ratios were either concordant or near concordant. They reported that the discordant residence times can be shown to result from the mixing of two different air masses, and, by assuming a residence time of 4 days for aerosols near the surface of the earth, an average residence time of 35 days can be calculated for the troposphere as a whole. Moreover, Daniel *et al* (1974)

reported that they obtained roughly concordant residence times from the ratios of $^{89}Sr/^{90}Sr$ and $^{210}Po/^{210}Pb$ in rain samples collected at Fayetteville, Arkansas, in 1972. Meanwhile, Marenco and Fontan (1973) and Hartwig (1973) expressed the view that Poet et al (1972) may have not interpreted their data properly (see the replies to the above investigators by Moore et al, 1973c,d). In view of the differences in the opinions expressed by various investigators, we felt that it was highly desirable to continue the work initiated by Nevissi et al (1973, 1974) in our laboratories. Results presented in this report include a set of data demonstrating the variation of the $^{210}Po/^{210}Pb$ ratio within a single rainfall.

EXPERIMENTAL

Rain samples were collected by means of a sampling system located on the roof of the Chemistry Building of the University of Arkansas at Fayetteville, Arkansas (36°N, 94°W). The radiochemical procedure used for the separation of ^{210}Pb and ^{210}Po were the same as those employed by Wallace (1965), while the method presented by Poet et al (1972) was used for the separation of ^{210}Bi. The samples of lead and bismuth were each covered with aluminum absorbers to prevent the counting of the low energy beta particles of ^{210}Pb and the 5.3-MeV alpha particles of ^{210}Po. The lead and bismuth samples were counted using an Omniguard BLB 5020 low-background beta counter. The polonium mounted carrier-free was alpha counted using an Ortec surface barrier detector connected to a Nuclear Chicago ND 150M pulse height analyzer.

RESULTS AND DISCUSSION

Table 1 shows the results. Mean residence times were calculated by the use of equations first derived by Lehmann and Sittkus (1959):

$$\frac{N_E \lambda_E}{N_D \lambda_D} = \frac{\tau_R}{\tau_R + \tau_D} \qquad \text{----------(1)}$$

and

$$\frac{N_F \lambda_F}{N_D \lambda_D} = \frac{(\tau_R)^2}{(\tau_R + \tau_F)(\tau_R + \tau_E)} \qquad \text{----------(2)},$$

where N's are the number of atoms, λ's are the decay constants, τ's are the reciprocals of the decay constants and the removal constant ($\tau_R = 1/\lambda_R$), and the subscripts D, E and F refer to RaD(^{210}Pb), RaE (^{210}Bi) and RaF (^{210}Po).

The results obtained are in essential agreement with those reported by Nevissi *et al* (1973, 1974). While they reported that in about 80 percent of the samples measured, the residence times calculated from the ^{210}Bi/^{210}Pb and ^{210}Po/^{210}Pb ratios were found to be either concordant or near concordant, our results indicate that it is the case in approximately 70 percent of the samples studied (12 out of 17 rains). In about 30 percent of the cases (5 out of 17 rains), the ^{210}Bi/^{210}Pb ratios yielded mean residence time values of 3 to 6 days, while the values calculated from the ^{210}Po/^{210}Pb ratio ranged from 16 to 40 days. The average residence times for the April to June rains were calculated from the average ^{210}Pb, ^{210}Bi and ^{210}Po concentrations ($\bar{C}$) in rain:

$$\bar{C} = \Sigma F/\Sigma R \qquad \text{----------(3)},$$

where ΣF is the total amount of each of the radon daughters (in picocuries per square meter) transported by the rain during the period and ΣR is the total rainfall (in millimeters) during the same period.

The value of 17^{+11}_{-6} days for the mean residence time obtained from the average $^{210}Bi/^{210}Pb$ ratio is shorter than the value of 32^{+5}_{-3} days calculated from the average $^{210}Po/^{210}Pb$ ratio. It appears to us, however, that this can not be taken as a strong evidence for the presence of an 'excess' ^{210}Po in the troposphere (Martell, 1972; Poet et al, 1972; Moore et al, 1973a,b,c,d). As it has been pointed out by Nevissi et al (1973; 1974), the mean residence times calculated from the $^{210}Bi/^{210}Pb$ and $^{210}Po/^{210}Pb$ ratios are not expected to be concordant if the radon daughters found in a sample of rain came from two (or more than two) different air masses (see also, Marenco and Fontan, 1973).

Let us consider the case of June 13th rain, as an example of obvious discordancy: the residence times calculated from the $^{210}Bi/^{210}Pb$ and $^{210}Po/^{210}Pb$ ratios are 7^{+3}_{-2} and 26 ± 3 days. Now suppose the June 13th rain acquired α (curie) of ^{210}Pb from air mass I and 1 - α(curie) of ^{210}Pb from air mass II, so that

$$\frac{N_{E12}\,\lambda_E}{N_{D12}\,\lambda_D} = \alpha \cdot \frac{N_{E1}\,\lambda_E}{N_{D1}\,\lambda_D} + (1-\alpha)\,\frac{N_{E2}\,\lambda_E}{N_{D2}\,\lambda_D} \qquad \text{----------(4)},$$

and

$$\frac{N_{F12}\,\lambda_F}{N_{D12}\,\lambda_D} = \alpha \cdot \frac{N_{F1}\,\lambda_F}{N_{D1}\,\lambda_D} + (1-\alpha)\,\frac{N_{F2}\,\lambda_F}{N_{D2}\,\lambda_D} \qquad \text{----------(5)},$$

where $N_{E12}\ \lambda_E/N_{D12}\ \lambda_D$ and $N_{F12}\ \lambda_F/N_{D12}\ \lambda_D$ are the $^{210}Bi/^{210}Pb$ and $^{210}Po/^{210}Pb$ ratios in the radon daughters found in rain, and the subscripts 1 and 2 refer to the radon daughters which originated in the air masses I and II, respectively. Let us further suppose that the residence times calculated in both air masses I and II: τ_{R1} = 4 days and τ_{R2} = 154 days, and α = 0.79. This leads to the $^{210}Bi/^{210}Pb$ and $^{210}Po/^{210}Pb$ ratios of 0.48 and 0.092, respectively, and if we introduce these values into equations (1) and (2), we obtain the discordant residence times of 7 and 26 days. This was the way the data were interpreted by Nevissi et al (1973; 1974), while Poet et al (1972) and Moore et al (1972, 1973) made the assumption that $^{210}Bi/^{210}Pb = {}^{210}Po/^{210}Pb = 1$ or they are essentially in radioactive equilibrium in the second component found in rain.

The second component of radon daughters which we are talking about here may be dust particles from surface soils or from those aerosols captured by the surface of the leaves in the region of forests, as it was pointed out by Haxel and Schuman (1955) and also by Poet et al (1972). It is also possible that $^{210}Po/^{210}Pb > 1$, if the radon daughters are released from the process of coal burning in the factories. Obviously, additional experimental data are needed to clarify these points. Measurements on sequentially sampled samples of a single rainstorm may provide us with a set of crucial experimental data. We therefore performed an experiment, in which the ^{210}Pb and ^{210}Po activities were determined sequentially. The results from the October 6, 1973, rainstorm are shown in Table 1 and are plotted in Fig. 1.

The rain occurred in the morning of October 6 was associated with a front, which was moving from the north to the southeast. On October 3,

a low pressure center was located in Kansas and it passed over the North-West Arkansas on October 4, causing a thunderstorm with cloud tops measured at 15.2 km. As shown in Table 1, the $^{210}Po/^{210}Pb$ residence time for the October 4, 1973, rain was found to be 108 days, which appeared to be abnormally long compared with the mean tropospheric residence time of about one month for a typical tropospheric air mass. This suggested the possibility that the October 4 rain may have been associated with the process of stratospheric tapping by intense thunderstorms postulated by Dingle (1965) (see also, Noyce et al, 1971). If so, an air mass which originated from the upper troposphere or the lower stratosphere may have dominated the Northwest Arkansas region temporarily during this period. The front was located near the Gulf Coast on October 6, 1973, and a high-pressure center had developed in the central Texas region, by the time the rain fell at Fayetteville, Arkansas. The Monett Meteorological Observatory reported the passage of a thunderstorm cell having cloud tops measured at 8.6 km at about the time of collection of the fraction III. The very unstable weather conditions at Fayetteville on October 6 suggest that the storm was associated with the process postulated by Dingle (1965) or perhaps by the tropopause folding phenomenon (Danielsen, 1964, 1968; Reed, 1955; Reed and Danielsen, 1959).

Fig. 1 shows a striking contrast in the patterns of variations of the ^{210}Pb and ^{210}Po concentrations during the course of the rainstorm. While ^{210}Pb concentration decreased gradually with time, ^{210}Po concentration was found to increase steadily and reach a maximum value in the third fraction (10 A.M.). The decrease in the ^{210}Pb concentration suggested that this nuclide was being removed from the troposphere by the washout

process. After a rapid initial decrease in the ^{210}Pb concentration, the rate of decrease became somewhat slower after the second fraction was collected at about 9:20 A.M. This trend may be explained as due to the fact that a new supply of ^{210}Pb from a different air mass became available during this time period. The new air mass may have been an old one, in which the $^{210}Po/^{210}Pb$ ratio was much higher than that in the first air mass. Such a process appears to explain also the steady increase in the ^{210}Po concentration in rain observed during the initial stage of the rain. The final decrease in the ^{210}Pb and ^{210}Po concentrations in rain after the third fraction was collected may signify the return of the first air mass, in which the $^{210}Po/^{210}Pb$ ratio was typical of an average tropospheric air, with the value corresponding to a mean residence time of approximately 30 days.

It is interesting to note that the $^{210}Po/^{210}Pb$ ratio in the third fraction (10 A.M.) corresponds to a mean residence time of about 300 days. This value is in essential agreement with an 'overall' mean stratospheric residence time of about one year reported by various investigators (Kuroda et al, 1960, 1962; Feely, 1960; Volchok, 1967; Krey, 1968; Fabian et al, 1968; Cooper et al, 1970; Beck et al, 1971). The amount of ^{210}Pb transported by the third fraction amounts to about 3.4 percent of the total ^{210}Pb atoms brought down by this rainfall, and it seems to be quite reasonable to assume that this fraction contained the ^{210}Pb atoms which was associated with an old air mass from the stratosphere.

It is difficult, but perhaps not impossible, to interpret the data shown in Fig. 1 in terms of the presence of dust particles from surface soils and/or radon daughters released from the process of coal burning in the factories. According to this interpretation, the fraction III (10 A.M.)

must have contained ^{210}Pb atoms originated from the above-mentioned sources. The fraction III, however, represented a small volume of rain water collected while the rate of rainfall was small during the interval between the initial and final stages of intense rainfalls (Fractions I and IV, respectively). According to the information supplied by Local Climatological Data, weather types on the date of occurrence of October 6, 1973, rainfall were listed as fog, smoke and haze, and locally heavy thunderstorms. No duststorm was reported in this region.

The fact that ^{210}Bi concentrations could not be measured in the sequentially sampled rainfall of October 6 makes the interpretation of the ^{210}Pb and ^{210}Po data obtained in this work somewhat uncertain. It would be highly desirable to have the concentrations of some bomb-produced radionuclides measured in sequentially sampled rains together with the radon daughters. Studies along these lines are now being planned.

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Table 1. ^{210}Pb, ^{210}Bi and ^{210}Po in rain samples collected at Fayetteville, Arkansas, during 1973.

Date of Rainfall	Amount of Rainfall (mm)	^{210}Pb	^{210}Bi	^{210}Po	Mean Residence Times (Days)	
		(10^{-12} curies/liter)			$\tau(E)$	$\tau(F)$
Apr. 9	18.8	3.14±0.10	3.20±0.68	0.39±0.04	$26\rightarrow\infty$	35±5
Apr. 16	32.2	2.53±0.12	0.84±0.15	0.27±0.03	3^{+2}_{-1}	29±4
Apr. 19	10.7	4.56±0.13	---	0.01±0.007	---	2^{+1}_{-1}
Apr. 20	12.6	2.62±0.13	---	0.57±0.06	---	63^{+13}_{-10}
Apr. 25	5.6	3.85±0.18	1.69±0.42	0.54±0.06	6^{+4}_{-3}	40±6
May 1	5.6	6.42±0.13	5.97±0.21	0.64±0.07	$53\rightarrow\infty$	29^{+2}_{-3}
May 5-6	2.8	4.62±0.08	4.17±0.28	0.53±0.05	$34\rightarrow\infty$	31^{+3}_{-2}
May 11	50.0	4.55±0.13	3.78±0.42	0.79±0.08	$36^{+\infty}_{-17}$	48^{+7}_{-6}
May 21	28.5	6.26±0.15	4.63±0.33	0.62±0.07	20^{+10}_{-5}	29^{+2}_{-3}
May 23	13.7	3.99±0.14	3.15±0.42	0.73±0.08	27^{+68}_{-13}	52^{+9}_{-9}
May 24	6.9	3.31±0.09	3.08±0.36	0.33±0.03	$29\rightarrow\infty$	29^{+2}_{-3}
May 26	12.5	5.19±0.10	3.53±0.58	0.28±0.03	15^{+15}_{-6}	16^{+3}_{-3}
May 28	4.6	2.18±0.07	2.29±0.51	0.23±0.02	$28\rightarrow\infty$	31^{+6}_{-5}
May 31	33.0	3.78±0.09	1.52±0.21	0.33±0.03	5^{+1}_{-1}	26^{+3}_{-3}
June 1	4.3	2.48±0.07	2.18±0.19	0.53±0.05	53^{+240}_{-27}	61^{+11}_{-9}
June 5	22.9	2.84±0.08	1.21±0.25	0.15±0.02	6^{+2}_{-2}	16^{+2}_{-3}
June 12	6.8	4.92±0.12	5.16±0.27	0.76±0.08	$240\rightarrow\infty$	43^{+6}_{-6}
June 13	1.5	6.09±0.12	2.91±0.49	0.56±0.06	7^{+3}_{-2}	26^{+3}_{-3}
June 16	14.2	2.56±0.07	2.37±0.22	0.36±0.04	$95^{+\infty}_{-62}$	40^{+6}_{-6}
Sep. 27	54.1	0.76±0.03	---	0.15±0.02	---	58^{+10}_{-10}
Sep. 29	6.3	3.11±0.10	---	0.23±0.02	---	22^{+4}_{-3}

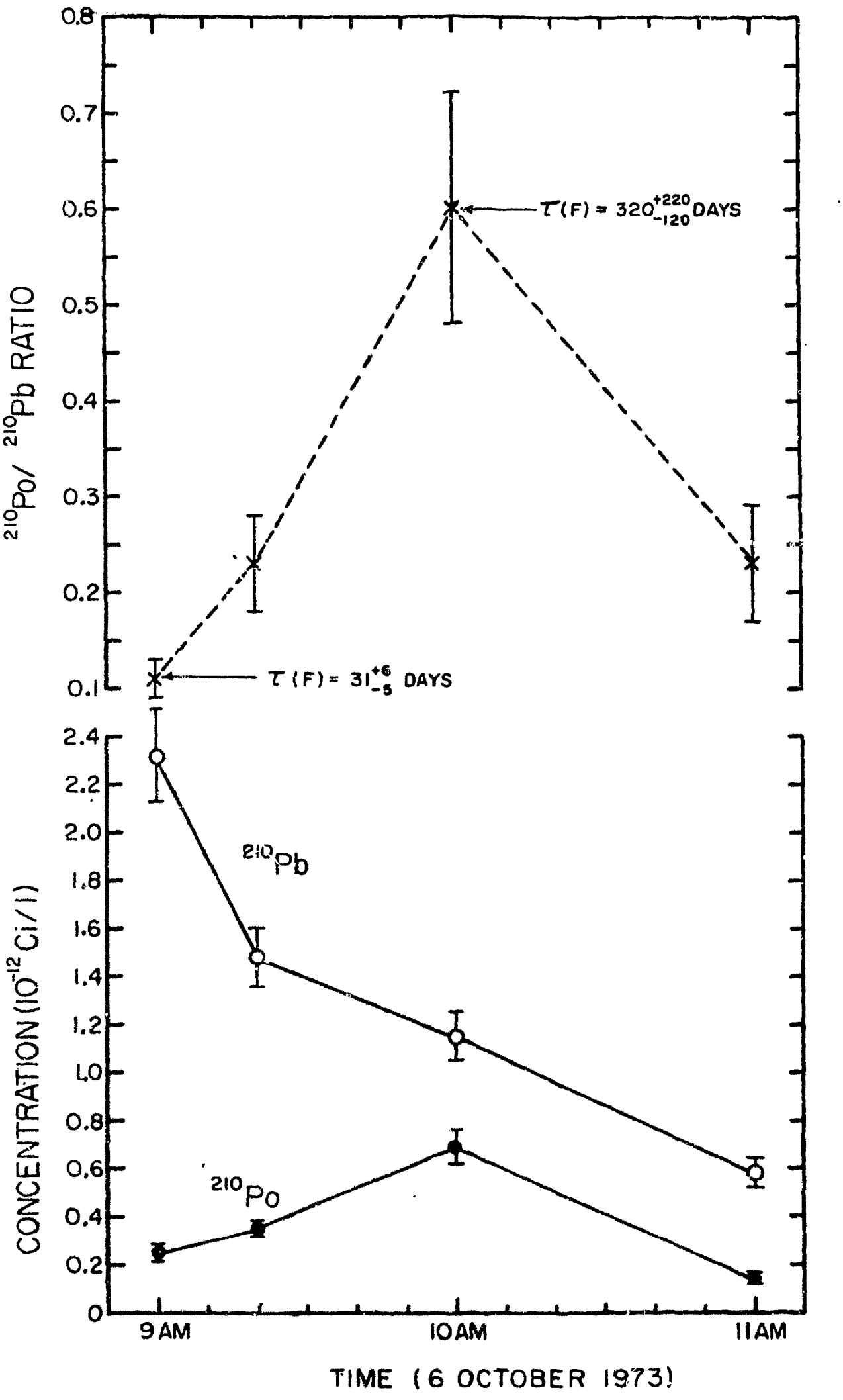
Table 1 (Continued)

Date of Rainfall	Amount of Rainfall (mm)	^{210}Pb (10^{-12} curies/liter)	^{210}Bi (10^{-12} curies/liter)	^{210}Po (10^{-12} curies/liter)	Mean Residence Times (Days) $\tau(E)$	Mean Residence Times (Days) $\tau(F)$
Oct. 4	3.2	3.14±0.12	---	1.04±0.10	---	108^{+27}_{-20}
Oct. 6						
Fraction 1 (9 AM)	8.6	2.32±0.19	---	0.25±0.03	---	31^{+6}_{-5}
Fraction 2 (9:20 AM)	0.89	1.48±0.12	---	0.35±0.03	---	68^{+16}_{-15}
Fraction 3 (10 AM)	0.89	1.15±0.10	---	0.69±0.07	---	320^{+220}_{-120}
Fraction 4 (11 AM)	6.9	0.58±0.06	---	0.14±0.01	---	68^{+19}_{-18}
Bulk Sample	17.3	1.74±0.14	---	0.25±0.02	---	40^{+9}_{-9}
Oct. 11	23.4	0.95±0.03	---	0.17±0.02	---	52^{+8}_{-8}
Oct. 12	61.0	1.70±0.06	---	0.51±0.05	---	11^{+1}_{-2}
Oct. 27	27.2	2.15±0.11	---	0.37±0.04	---	49^{+9}_{-9}
Oct. 30	19.8	1.71±0.06	---	0.19±0.02	---	31^{+5}_{-5}
Nov. 19	0.6	11.15±0.37	---	2.56±0.26	---	68^{+11}_{-10}
Nov. 20	39.9	1.20±0.05	---	0.30±0.03	---	75^{+16}_{-14}
Nov. 23	34.3	2.03±0.07	---	0.14±0.01	---	21^{+3}_{-3}
Nov. 26	45.2	1.98±0.07	---	0.11±0.01	---	17^{+3}_{-3}
Dec. 14	3.6	3.17±0.06	---	0.98±0.10	---	100^{+18}_{-17}
Dec. 19	35.5	2.69±0.05	---	0.40±0.05	---	43^{+5}_{-5}
Dec. 23	25.7	2.12±0.04	---	0.57±0.07	---	82^{+18}_{-14}
Dec. 30	6.4	6.53±0.06	---	2.35±0.25	---	123^{+20}_{-19}

FIGURE CAPTION

Fig. 1. ^{210}Pb and ^{210}Po in sequentially sampled rainstorm of October 6, 1973, and mean residence times calculated from the ratio of radon daughters.

FIG.1


0.8
0.7
0.6
0.5
0.4
0.3
0.2
0.1
$^{210}Po/\ ^{210}Pb$ RATIO
$\tau(F) = 320^{+220}_{-120}$ DAYS
$\tau(F) = 31^{+6}_{-5}$ DAYS
2.4
2.2
2.0
1.8
1.6
1.4
1.2
1.0
0.8
0.6
0.4
0.2
0
CONCENTRATION (10^{-12} Ci/l)
^{210}Pb
^{210}Po
9AM
10AM
11AM
TIME (6 OCTOBER 1973)

APPENDIX VIII

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THE RATIOS OF FISSION PRODUCT PAIRS WITH SIMILAR HALF-LIVES IN THE ATMOSPHERE

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The ratios of pairs of fission products with similar half-lives (58.5-day ^{91}Y to 50.8-day ^{89}Sr and 13.6-day ^{143}Pr to 12.8-day ^{140}Ba) were measured radiochemically in a series of rain samples collected at Fayetteville (36°N, 94°W), Arkansas, after the Chinese explosions of March 18, 1972 (the 14th) and of June 27, 1973 (the 15th). The activity ratios in rain were found to show a marked variation: the $^{91}Y/^{89}Sr$ ratio in rain varied between the extreme values of 0.29 (March 24, 1972) and 10 (April 3, 1972), while the $^{143}Pr/^{140}Ba$ ratio from 0.21 (March 24, 1972) to 6.3 (April 13 and 14, 1972). The observed large variations of the activity ratios appear to be attributable to the difference in size distributions of particulate matters in rain. ^{91}Y and ^{143}Pr become highly enriched in particles larger than a few micron-size, relative to ^{89}Sr and ^{140}Ba, respectively, whereas the reverse appears to be the case in smaller, sub-micron size particles. A similar trend was found in the data obtained after the 7th Chinese test of December 24, 1967. The results indicate that these isotopic pairs may turn out to be extremely useful as atmospheric tracers in the studies of meteorological phenomena.

INTRODUCTION

There has been a steady increase in the use of natural and artificial radioactive isotopes as atmospheric tracers in the studies of meteorological phenomena. It has been known for many years that studies on the variations of the ratios of fission products with different half-lives in air and in rain provide us with valuable information regarding the origin and the age of the atmospheric aerosols. The ratios of isotopes of the same or chemically similar elements, such as $^{89}Sr/^{90}Sr$, $^{140}Ba/^{90}Sr$, $^{141}Ce/^{144}Ce$ in air and in rain have been studied since the late 1950's (see, for example, Kuroda, 1958; Libby, 1959; Martell, 1959; Martell and Drevinsky, 1960; Menon et al, 1963). The ratios of the long-lived radon daughcers ^{210}Pb, ^{210}Bi and ^{210}Po in the atmosphere have also been studied by several investigators (Poet et al, 1972; Moore et al, 1973a,b; Nevissi et al, 1973, 1974; Gavini et al, 1974). Little attention has been paid in the past, however, to the potential importance of the use of the ratios of fission product pairs with almost identical half-lives in the atmosphere. The small advantage that decay corrections become unnecessary in these cases is nullified by the obvious disadvantage that the tracers no longer serve as a 'nuclear clock'. Moreover, one might assume that the activity ratios of fission product pairs with similar half-lives generally remain more or less constant with time.

The fact that the ratios of fission product pairs with almost identical half-lives in the atmosphere vary markedly with time has been known, however, for many years. A good example is the $^{137}Cs/^{90}Sr$ ratio in the atmosphere. The half-lives of ^{90}Sr and ^{137}Cs, 28 years and 30 years,

respectively, are nearly equal and both of these isotopes are produced at high yields from nuclear devices. Stewart et al (1957) reported that the mean ratio of $^{137}Cs/^{90}Sr$ deposited during 1955 to 1957 at several localities of the wor'd ranged from 1.24 to 2.32, but abnormally low values were observed at Gibraltar (0.51) and Melbourne (0.49). Miyake et al (1963) (see also, Miyake and Katsuragi, 1970; Katsuragi, 1971; Miyake, 1971) made a similar observation. Sherrill et al (1974) have recently measured ^{137}Cs and ^{90}Sr in a total of 137 individual rain and snow samples collected at Fayetteville, Arkansas, and found that the $^{137}Cs/^{90}Sr$ ratio varied between 0.26 to several times the production ratio, and they reported that such variations are attributable to the difference in the size distributions of particulate matter in rain.

The ratios of relatively short-lived fission product pairs with almost identical half-lives in the atmosphere have been measured occasionally in the past, but no systematic studies on the utilization of such isotope pairs as atmospheric tracers have been carried out in the past. In the present investigation, we therefore chose the following two isotopic pairs: 58-day ^{91}Y/50.8-day ^{89}Sr and 13.6-day ^{143}Pr/12.8-day ^{140}Ba, and measured the concentrations of these isotopes in a series of rain samples collected at Fayetteville (36°N, 94°W), Arkansas, after the 14th (March 18, 1972) and the 15th (June 27, 1973) Chinese nuclear tests conducted at Lop Nor (40°N, 90°E). The results indicate that these isotopic pairs may turn out to be extremely useful as atmospheric tracers.

EXPERIMENTAL

For the radiochemical separation of the Y, Sr, Ba and rare earth nuclides from rain, 8 to 20 liters of rain were used in each case. About

20 to 50 mg of stable carriers were added to the sample. The solution was then evaporated to a small volume and transferred to a teflon beaker. A 20 ml portion of HNO_3 was added to the solution and again evaporated to a small volume. The solution was treated with 20 ml of fuming HNO_3, 10 ml of conc. $HClO_4$ and 10 ml of conc. HF and was then evaporated until the perchloric acid fumed. The treatment with the three acids was repeated and the Sr and Ba isotopes were precipitated with fuming HNO_3. Sr and Ba were separated from each other and purified according to the method reported by Hodges (1963). The final carbonate precipitates were counted on a Tracerlab "Omniguard" low background beta counter. Backgrounds were about 0.75 cpm during the course of the investigation.

The supernate was transferred to a lusteroid tube and the Y and the rare earths were precipitated as the fluorides. Y and the rare earths were purified as a group from the other fission products by the method of Stevenson and Nervik (1961). Y and the rare earths were then separated by the use of 50W-X8, 200 to 400 mesh cation-exchange resin and the pH gradient-elution technique described by Wolfsberg (1962) was modified to be a concentration gradient-elution. The samples were counted on a Tracerlab CE-14SL low level beta counter. The background was about 2.5 cpm.

RESULTS AND DISCUSSION

The experimental results obtained are shown in Table 1. The $^{91}Y/^{89}Sr$ and the $^{143}Pr/^{140}Ba$ ratios were calculated from these data and are plotted in Fig. 1 and Fig. 2. The data on the number of particles in the groumd-level air, which are plotted in Fig. 1, were taken from the report of Moore, Beck *et al* (1973). Both $^{91}Y/^{89}Sr$ and $^{143}Pr/^{140}Ba$ ratios in rain were found to vary quite appreciably: after the 14th Chinese test, the

former ratio varied between the extreme values of 0.29 (March 24) and 10 (April 3), while the latter ratio from 0.21 (March 24) to 6.3 (April 13 and 14). The variations were somewhat less pronounced in the case of the 15th Chinese test (Fig. 2).

The marked variation of these ratios observed in individual samples of rain appears to be quite in line with the recent observation by Sherrill et al (1974), who found that the $^{137}Cs/^{90}Sr$ ratio in many individual rain samples collected at Fayetteville, Arkansas, showed a large fluctuation. They stated that the observed variation in the $^{137}Cs/^{90}Sr$ ratio may be attributable to the difference in size distributions of particulate matter in rain. Both ^{137}Cs and ^{90}Sr become depleted in large fallout particles, because of the fairly long half-lives of their precursors (3.82-min ^{137}Xe and 32.3-sec ^{90}Kr). Taking into consideration the half-lives of the gaseous precursors and the independent and cumulative yields of the fission products belonging to the mass 90 and 137 chains, Sherrill et al (1974) were able to show that extreme values of the $^{137}Cs/^{90}Sr$ ratio in large and small fallout particles expected to be observed in rain may range from about 0.2 to several times the production ratio.

Let us first consider the case of ^{91}Y and ^{89}Sr. The mass 89 and 91 fission decay chains both have gaseous precursors:

$$\underset{4.65\%}{\text{3.18-min } ^{89}Kr} \xrightarrow{\beta^-} \underset{4.81\%}{\text{15.2-min } ^{89}Rb} \xrightarrow{\beta^-} \underset{4.81\%}{\text{50.8-day } ^{89}Sr},$$

and

$$\underset{3.25\%}{\text{8.6-sec } ^{91}Kr} \xrightarrow{\beta^-} \underset{5.55\%}{\text{57.9-sec } ^{91}Rb} \xrightarrow{\beta^-} \underset{5.72\%}{\text{9.67-hr } ^{91}Sr} \xrightarrow{\beta^-} \underset{5.78\%}{\text{58.5-day } ^{91}Y},$$

where the cumulative yield values are for thermal neutron-induced fission of ^{235}U recommended by Meek and Rider (1972) (see also, Hyde, 1964).

The $^{91}Y/^{89}Sr$ production ratio for ^{235}U thermal neutron-induced fission is

$$\frac{50.8}{58.5} \cdot \frac{5.78}{4.81} = 1.04 \text{ (curie/curie)}$$

and in a hypothetical large fallout particle, which was formed at the time zero of a nuclear detonation and hence lost all the rare gas precursors, the $^{91}Y/^{89}Sr$ activity ratio is expected to be

$$(^{91}Y/^{89}Sr)_{max} = \frac{(5.78 - 3.25)}{(4.81 - 4.65)} \cdot \frac{50.8}{58.5} = 13.7,$$

which is similar to the value of $^{91}Y/^{89}Sr = 10$ observed for the April 3, 1972, rain. Moore, Beck et al (1973) found a 34-micron diameter single particle in rain water during the night of March 23, 1972. While they were able to measure the ^{91}Y activity, the 89 mass chain was so depleted that the ^{89}Sr activity could not be determined in this particle. Clark et al (1967) measured the ^{91}Y and ^{89}Sr activities in a 16-micron single particle collected in Osaka, Japan, after the third Chinese test of May 9, 1966. Their data correspond to a value of 33 ± 16 for the $^{91}Y/^{89}Sr$ ratio.

These results indicate that large values of $^{91}Y/^{89}Sr$ ratio observed in rain samples after the nuclear tests are due most likely to the presence of radioactive 'hot' particles. If ^{89}Sr is depleted relative to ^{91}Y in large particles, it must be enriched relative to ^{91}Y in very small particles. The half-lives of ^{89}Kr (3.18-min) and ^{89}Rb (15.2-min) are much longer than those of their counterparts belonging to the mass 91 chain. Thus it is quite likely, that the fission products belonging to the mass 89 chain may

become separated from those with mass 91, and later adsorbed by the surfaces of fine particles. In this way, $^{91}Y/^{89}Sr$ ratios may become as low as 0.2 or 0.3 in rain samples containing very small fallout particles.

In the case of the fission product pair ^{143}Pr and ^{140}Ba, the decay chains involved are:

$$\underset{\underline{3.83\%}}{\text{13.6-sec } ^{140}\text{Xe}} \xrightarrow{\beta^-} \underset{\underline{5.83\%}}{\text{63.8-sec } ^{140}\text{Cs}} \xrightarrow{\beta^-} \underset{\underline{6.30\%}}{\text{12.8-day } ^{140}\text{Ba}},$$

and

$$\underset{\underline{0.047\%}}{\text{0.96-sec } ^{143}\text{Xe}} \xrightarrow{\beta^-} \underset{\underline{1.46\%}}{\text{1.7-sec } ^{143}\text{Cs}} \xrightarrow{\beta^-} \underset{\underline{4.90\%}}{\text{12-sec } ^{143}\text{Ba}}$$

$$\xrightarrow{\beta^-} \underset{\underline{5.82\%}}{\text{14-min } ^{143}\text{La}} \xrightarrow{\beta^-} \underset{\underline{5.89\%}}{\text{33-hr } ^{143}\text{Ce}} \xrightarrow{\beta^-} \underset{\underline{5.92\%}}{\text{13.58-day } ^{143}\text{Pr}},$$

where the cumulative yields are those recommended by Meek and Rider (1972) for the ^{235}U thermal neutron-induced fission.

The $^{143}Pr/^{140}Ba$ production ratio for thermal neutron-induced fission of ^{235}U is thus

$$\frac{12.8}{13.58} \cdot \frac{5.92}{6.30} = 0.89.$$

^{140}Ba and ^{143}Pr data are available for 13-micron, 16-micron and 34-micron diameter particles (Clark et al, 1967; Moore, Beck, et al, 1973). Their analyses correspond to the $^{143}Pr/^{140}Ba$ ratios of 6.6 (13-micron particle), 8.1 (16-micron particle) and 26 (34-micron particle). The highest $^{143}Pr/^{140}Ba$ ratio values observed in rain are 6.3 (April 13 and 14, 1972) after the 14th Chinese test and 2.5 (July 14, 1973) after the 15th Chinese test. These results again suggest that the presence of at least several-micron size particles are probably responsible for the large $^{143}Pr/^{140}Ba$ ratios observed in the rain samples.

It is interesting to note that the patterns of variation of the $^{91}Y/^{89}Sr$ and $^{143}Pr/^{140}Ba$ ratios in rain observed after the 14th and the 15th Chinese tests are different from each other. While the ratios in rain seemed to have approached the production ratio in the second half of April 1972 less than one month after the 14th test, high $^{91}Y/^{89}Sr$ ratios were observed in rain samples collected after September 1973, more than three months after the 15th test of June 27, 1973, and none of the $^{91}Y/^{89}Sr$ ratios observed in rain during the months of September through November 1973 were significantly below the production ratio. This seems to indicate that perhaps several micron-size particles stayed air-borne for a period of several months. This interesting phenomenon may be interpreted in two ways: (a) the large fallout particles were injected in the stratosphere and stayed there for a long time, because of the lack of downward motion of the stratospheric air masses connected with the so-called spring peak in fallout (Brewer, 1949; Dobson, 1956; Stewart et al, 1957) or (b) they stayed in the troposphere without falling out for a long time, because the mean tropospheric residence times of aerosols during the second half of

1972 were much longer than usual. In this connection, it is interesting to note that Gavini et al (1974) have recently reported that the mean residence time values which they obtained from the $^{210}Po/^{210}Pb$ ratios in rain samples collected during the fall months of 1973 were much longer than the usual value of approximately one month. For example, they obtained a value of about 100 days from the $^{210}Po/^{210}Pb$ ratio in the October 4, 1973, rain and a value of 40 days from the ratio in the October 6, 1973, rain.

Thein et al (1968) measured ^{91}Y, ^{89}Sr, ^{143}Pr and ^{140}Ba concentrations in 6 rain samples collected after the 7th Chinese test of December 24, 1967. The $^{91}Y/^{89}Sr$ and $^{143}Pr/^{140}Ba$ ratios were calculated from their data and are plotted in Fig. 3 and Fig. 4.

Thein et al (1968)'s data seem to be in line with our recent data and can be interpreted in a similar manner. Both $^{91}Y/^{89}Sr$ and $^{143}Pr/^{140}Ba$ ratios in rain were far greater than the production ratios during the period December 30 through January 2, suggesting that the rain samples contained large fallout particles. After January 5, 1968, however, the ratios fell below the production ratios suggesting the intrusion of an air mass containing much smaller fallout particles. Fig. 5 and Fig. 6 show air trajectories at 500 mb and at 300 mb levels, respectively, prepared after the 7th Chinese test of December 24, 1967, by Chen et al (1969). The first 'wave' of the tropospheric debris from the 7th Chinese test reached the central United States by December 30, 1967, (Fig. 5). At that time, the debris that travelled at 300 mb level appears to have moved away to the south of Greenland (Fig. 6a). As reported by Thein et al (1968), there has been a sudden increase in the number of small (submicron-size) fallout particles in the ground-level air at Fayetteville, Arkansas, during January 7 to 9, which

coincides with the second arrival of the 300 mb air trajectories at the central United States (Fig. 6b).

At about the same time, the bulk of the debris injected into the troposphere appear to have just completed a circle around the world and appear to have been located near the original starting point Lop Nor (40°N, 90°E), China, on the opposite side of the world from Fayetteville (36°N, 94°W) (see Fig. 5). These results seem to suggest that the bulk of large fallout particles were travelling at 500 mb level, while the small sub-micron-size particles from the 7th Chinese test were injected at higher levels and were mostly travelling at 300 mb level (see also, Chen et al, 1969, 1970). This result is consistent with the view that the observed large variations in the activity ratios of radionuclides having similar half-lives can be attributed to the difference in size distributions of particulate matter following a nuclear explosion. This effect can be observable for many months following a nuclear explosion.

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Table 1. ^{89}Sr, ^{91}Y, ^{140}Ba and ^{143}Pr in rain at Fayetteville, Arkansas, after the Chinese nuclear explosions of March 18, 1972, (14th) and June 27, 1973, (15th).[a]

Nuclear Test	Date of Rainfall	^{89}Sr	^{91}Y	^{140}Ba	^{143}Pr
		(10^{-12} curie/l)			
(1) The 14th test (March 18, 1972).					
	March 23	1.35±0.44	1.70±0.07	12.7±0.5	17.5± 0.5
	March 24	19.8 ±3.1	6.0 ±0.2	129 ±9	27.6± 0.8
	March 26	12.8 ±1.2	21.0 ±0.7	160 ±12	276 ± 12
	April 3	3.43±0.67	33.9 ±1.5	85.8±9.1	418 ± 15
	April 13	3.73±0.63	12.9 ±0.5	12.2±1.3	62 ± 3
	April 14	6.99±0.86	11.5 ±1.1	24.6±1.7	50 ± 2
	April 15	8.93±2.9	9.1 ±0.3	18.8±2.4	27.8± 2.5
	April 19	17.3 ±2.9	24.5 ±1.9	---	---
	April 27	25.7 ±2.3	20.6 ±1.5	---	---
	May 1	13.6 ±2.6	15.3 ±0.9	---	---
	May 12	27.7 ±1.9	12.7 ±1.4	---	---
	May 28	72.6 ±6.2	27.3 ±1.5	---	---
(2) The 15th test (June 27, 1973).					
	July 10	2.9 ±0.6	1.9 ±0.1	9.6±0.7	4.9± 0.2
	July 14	0.26±0.06	1.6 ±0.1	1.7±0.2	4.2± 0.3
	July 21	1.0 ±0.3	1.1 ±0.1	3.9±0.9	1.5± 0.1
	July 23	3.1 ±1.0	1.6 ±0.2	3.6±0.5	4.2± 0.3
	July 24	1.7 ±0.8	1.5 ±0.1	2.5±0.6	4.2± 0.3
	Aug. 13	1.8 ±0.3	2.2 ±0.2	---	---
	Aug. 16	5.2 ±1.0	2.5 ±0.1	---	---
	Sep. 2	7.1 ±1.0	5.4 ±0.1	---	---
	Sep. 12	1.8 ±0.3	3.4 ±0.2	---	---
	Sep. 24	1.0 ±0.4	3.2 ±0.2	---	---
	Sep. 29	1.0 ±0.2	3.7 ±0.1	---	---
	Oct. 4	1.2 ±0.2	1.8 ±0.1	---	---
	Oct. 6	0.5 ±0.1	1.4 ±0.1	---	---
	Oct. 11	0.4 ±0.2	0.9 ±0.1	---	---

Table 1 (Concluded)

Date of Rainfall	^{89}Sr	^{91}Y	^{140}Ba	^{143}Pr
	(10^{-12} curie/l)			
Oct. 12	0.20±0.06	0.20±0.03	---	---
Oct. 27	0.74±0.10	2.3 ±0.1	---	---
Oct. 30	2.1 ±0.3	2.6 ±0.2	---	---
Nov. 20	0.4 ±0.1	0.84±0.05	---	---
Nov. 23	0.6 ±0.1	0.86±0.05	---	---
Nov. 23-25	0.9 ±0.1	1.3 ±0.1	---	---
Nov. 25-26	1.8 ±0.2	4.7 ±0.2	---	---

(a) The ^{89}Sr and ^{140}Ba data for the March to May 1972 rain samples were taken from the report by Daniel et al (1974).

FIGURE CAPTIONS

Fig. 1. Variations of the $^{91}Y/^{89}Sr$ and $^{143}Pr/^{140}Ba$ ratios in rain samples collected at Fayetteville, Arkansas, after the 14th Chinese test of March 18, 1972. The particles data were taken from Moore, Beck, et al (1973).

Fig. 2. Variations of the $^{91}Y/^{89}Sr$ and $^{143}Pr/^{140}Ba$ ratios in rain samples collected at Fayetteville, Arkansas, after the 15th Chinese test of June 27, 1973.

Fig. 3. Variation of the $^{91}Y/^{89}Sr$ ratio in rain samples collected at Fayetteville, Arkansas, after the 7th Chinese test of December 24, 1967. The ratios were calculated from the data reported by Thein et al (1968). The 16-micron particle value was taken from the report of Clark et al (1967).

Fig. 4. Variation of the $^{143}Pr/^{140}Ba$ ratio in rain samples collected at Fayetteville, Arkansas, after the 7th Chinese test of December 24, 1967. The ratios were calculated from the data reported by Thein et al (1968). The particle data were taken from the reports by Clark et al (1967) and Moore, Beck, et al (1973).

Fig. 5. Air trajectories at the height of 5670 meters (500 mb) showing the path of fresh debris from the 7th Chinese test (December 24, 1967) (Chen et al, 1969).

Fig. 6, a and b. Air trajectories at 300 mb level from the 7th Chinese test (December 24, 1967) (Chen et al, 1969).

a: the first cycle b: the second cycle

Fig.1

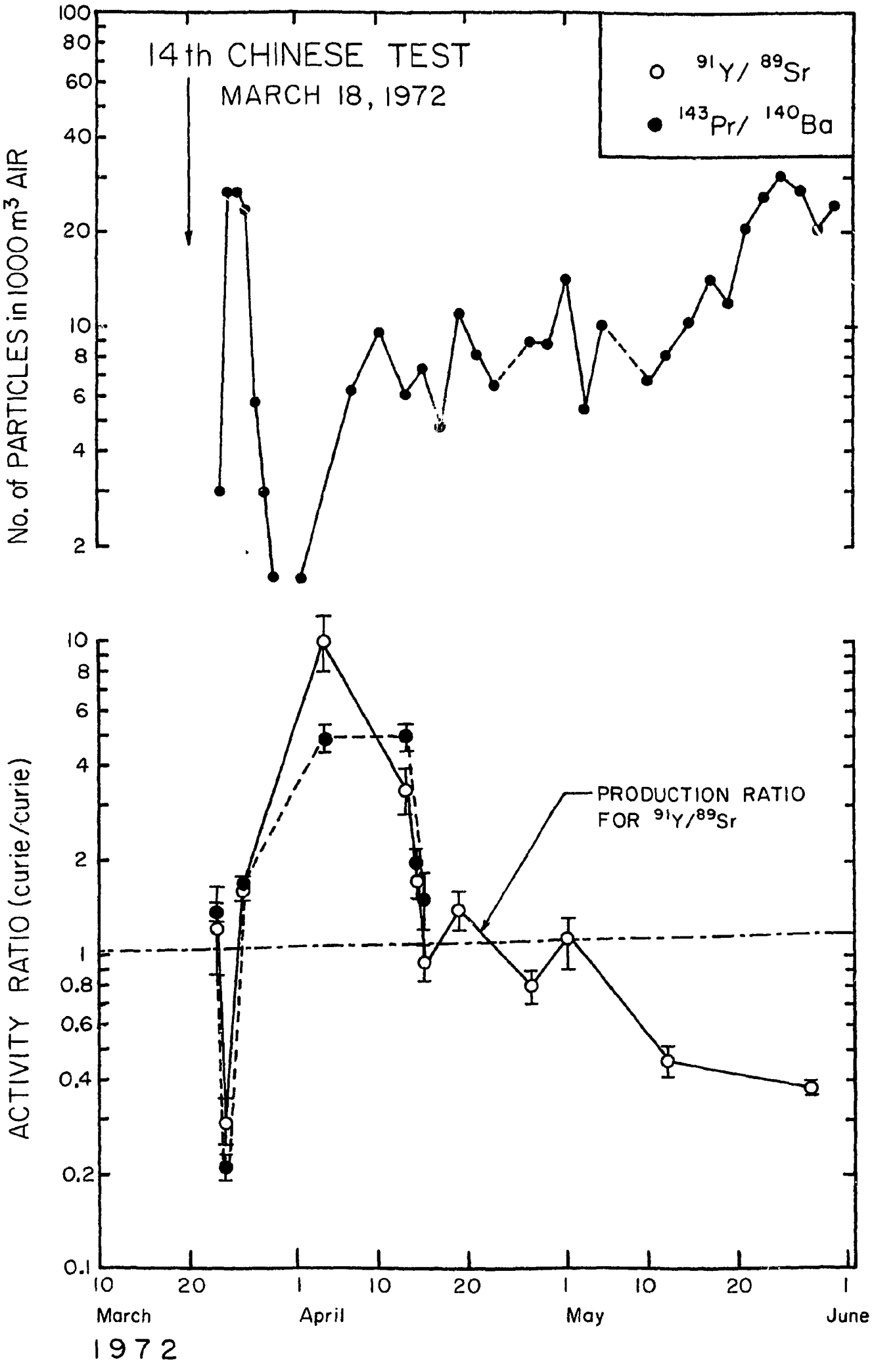

14th CHINESE TEST
MARCH 18, 1972
o $^{91}Y/^{89}Sr$
● $^{143}Pr/^{140}Ba$
No. of PARTICLES in 1000 m³ AIR
ACTIVITY RATIO (curie/curie)
PRODUCTION RATIO FOR $^{91}Y/^{89}Sr$
100
80
60
40
20
10
8
6
4
2
1
0.8
0.6
0.4
0.2
0.1
10
20
1
10
20
1
10
20
1
March
April
May
June
1972

Fig. 2

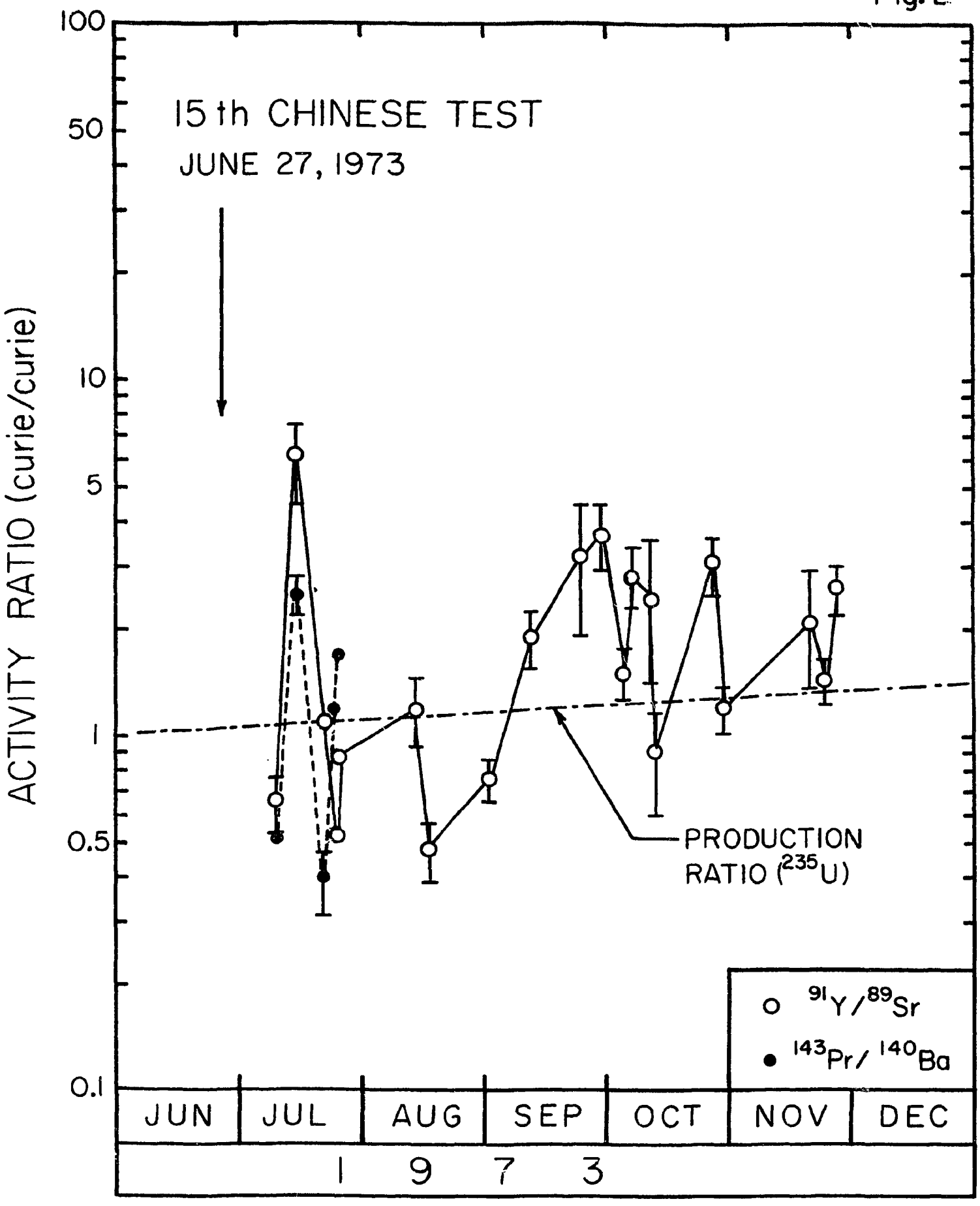

100
50
10
5
1
0.5
0.1
ACTIVITY RATIO (curie/curie)
15 th CHINESE TEST
JUNE 27, 1973
PRODUCTION
RATIO (^{235}U)
^{91}Y/^{89}Sr
^{143}Pr/^{140}Ba
JUN
JUL
AUG
SEP
OCT
NOV
DEC
1 9 7 3

Fig.3

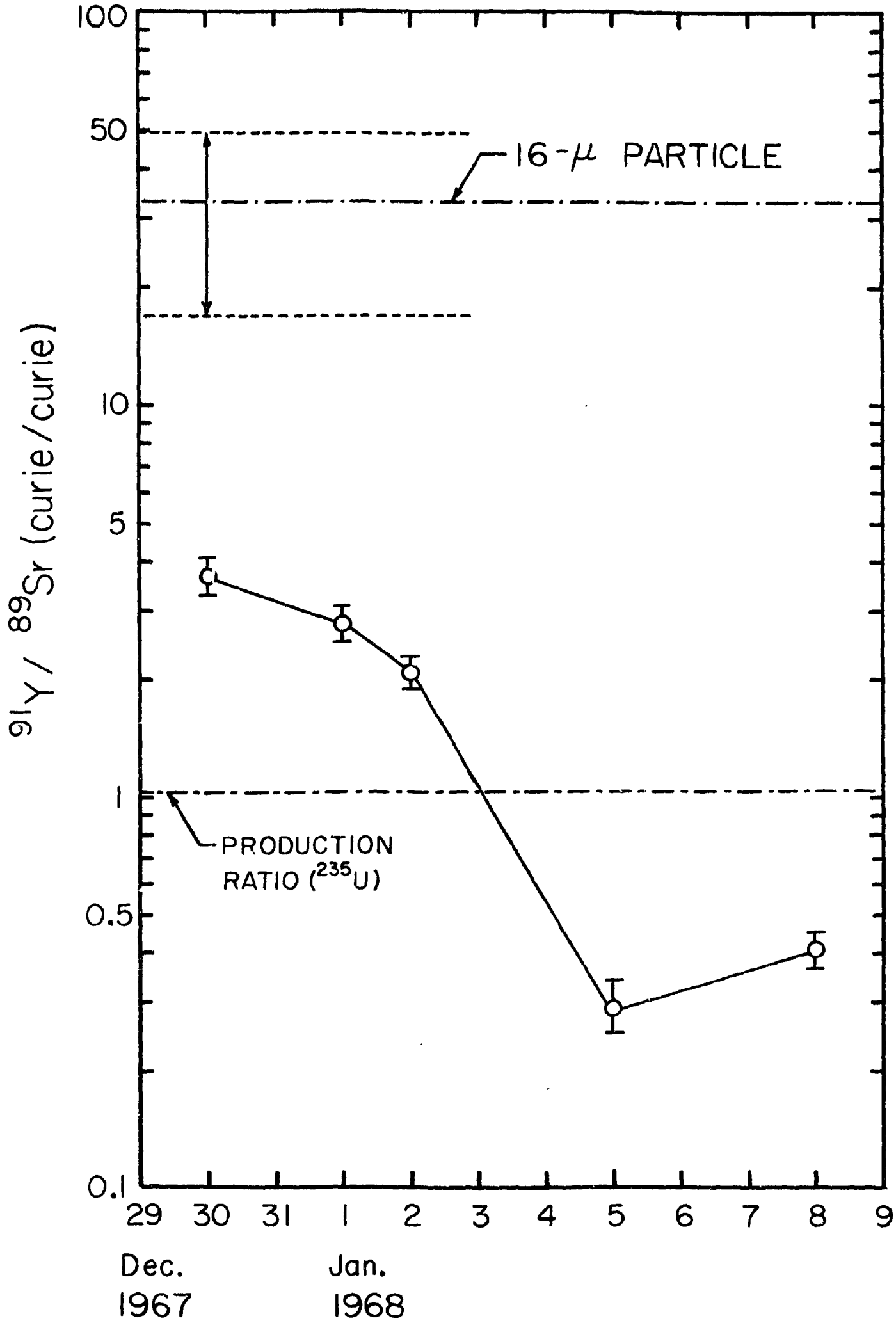

100
50
10
5
1
0.5
0.1
16-μ PARTICLE
PRODUCTION RATIO (^{235}U)
^{91}Y / ^{89}Sr (curie/curie)
29
30
31
1
2
3
4
5
6
7
8
9
Dec.
1967
Jan.
1968

Fig.4

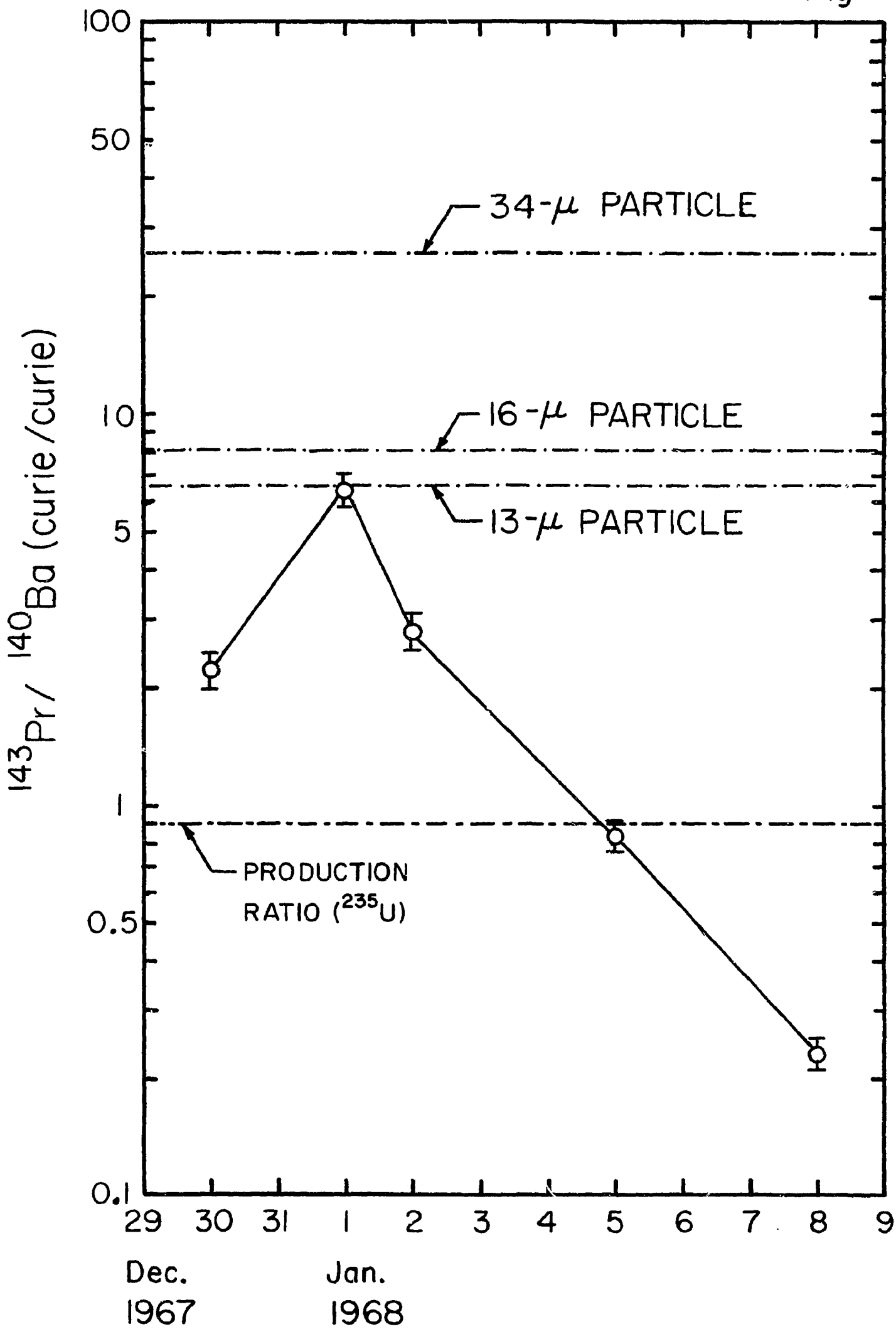

100
50
10
5
1
0.5
0.1
34-μ PARTICLE
16-μ PARTICLE
13-μ PARTICLE
PRODUCTION
RATIO (235U)
143Pr/140Ba (curie/curie)
29
30
31
1
2
3
4
5
6
7
8
9
Dec.
1967
Jan.
1968

Fig. 5

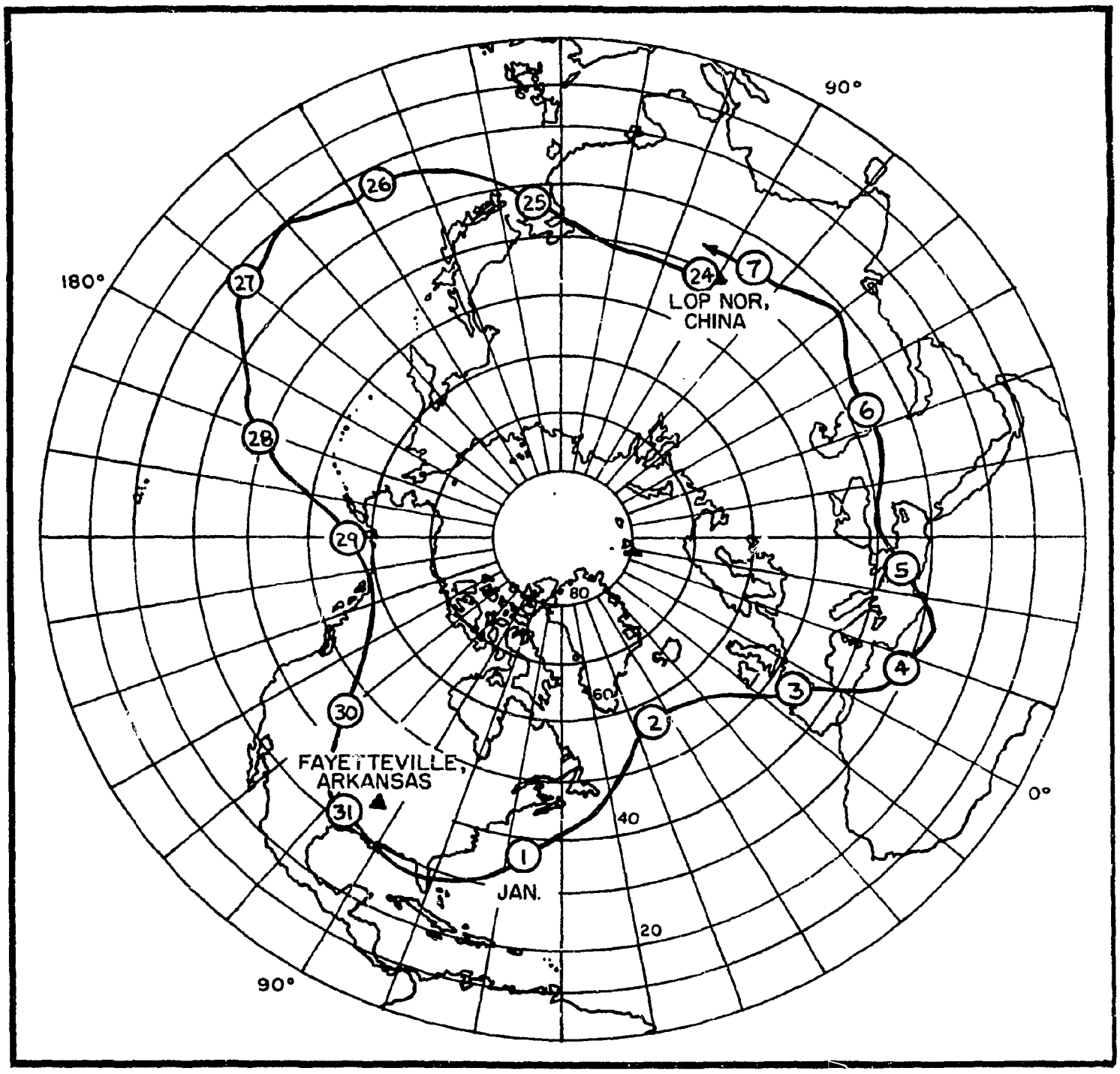

90°
180°
0°
90°
80
60
40
20
26
25
27
24
7
LOP NOR,
CHINA
6
28
29
5
4
3
30
2
FAYETTEVILLE,
ARKANSAS
31
1
JAN.

Fig. 6a

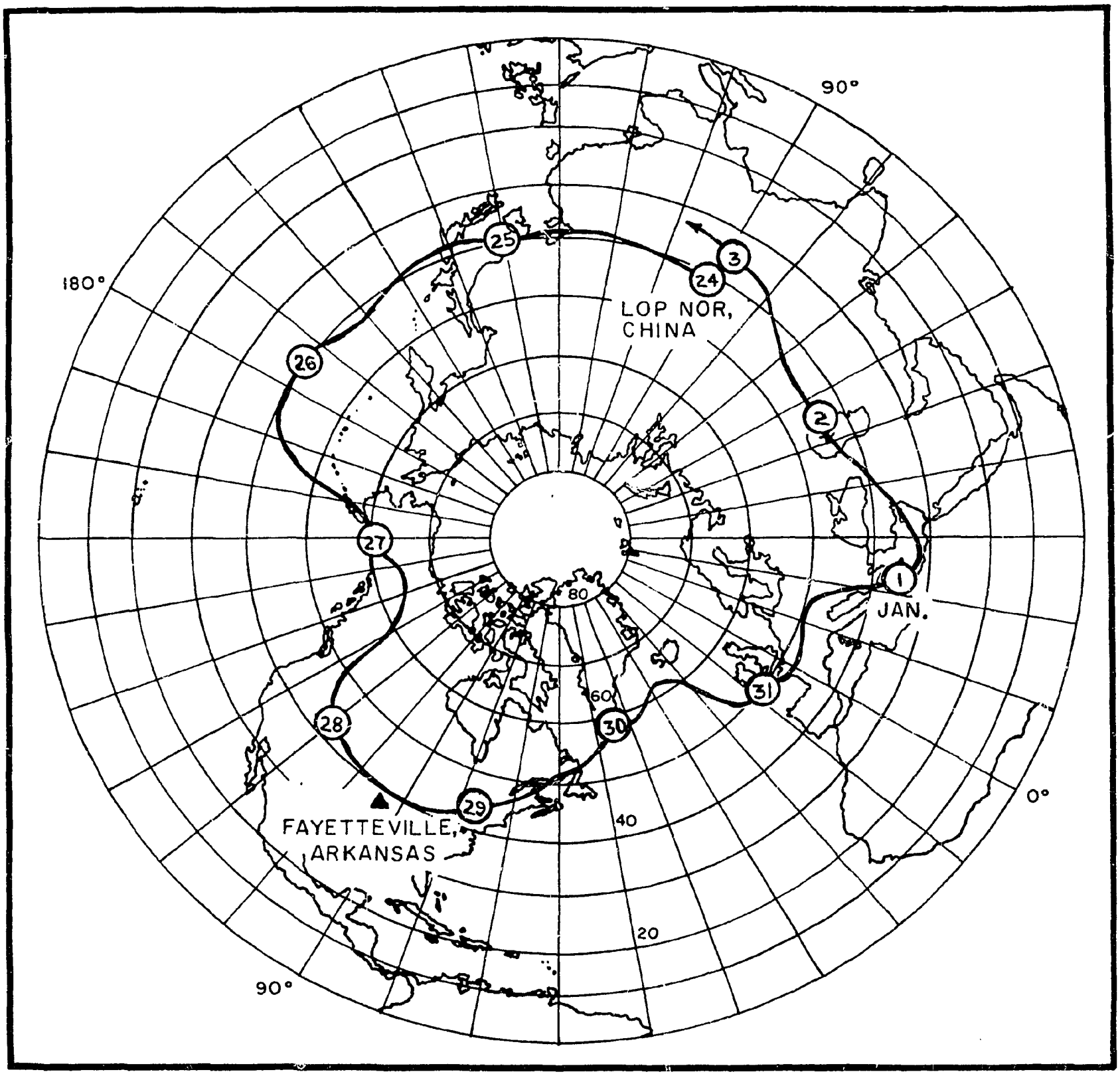
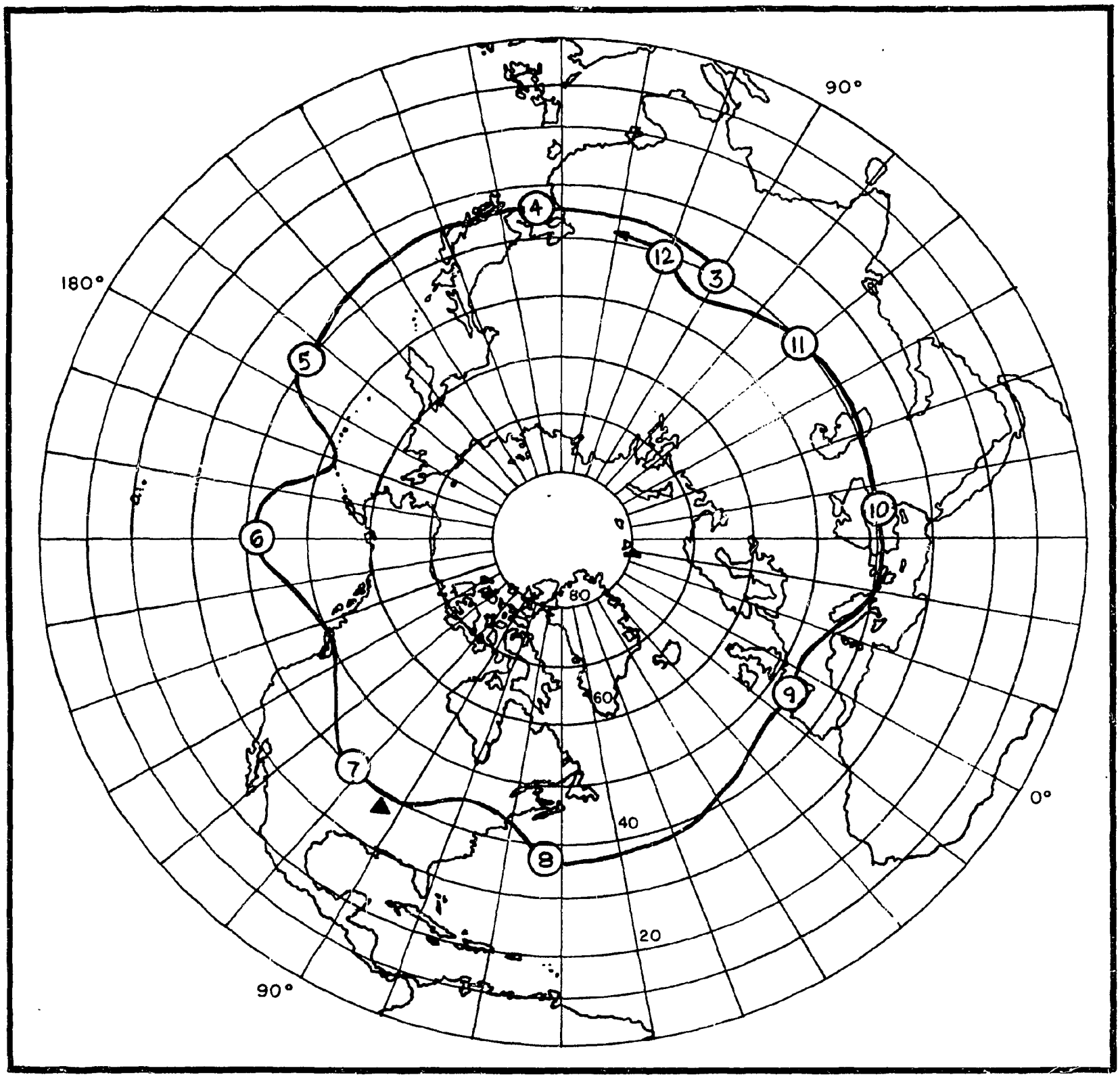

90°
180°
LOP NOR,
CHINA
25
24
3
26
2
27
1
JAN.
80
31
60
28
30
0°
29
40
FAYETTEVILLE,
ARKANSAS
20
90°

Fig. 6b



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APPENDIX IX

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Radioactive Strontium Fallout Measured in Rain Samples Collected at Fayetteville, Arkansas; Ankara, Turkey; and Tokyo, Japan, after the 14th Chinese Nuclear Explosion.

P. Y. Daniel

Table 1 gives the Sr^{89}/Sr^{90} ratios that were redetermined in individual samples of rain and snow at Fayetteville (36°N, 94°W), Arkansas, during the period from August 3, 1971, to June 29, 1973. This set of data supercedes the data given in Table I, Appendix VII in the 1973 Annual Progress Report for the period August 3, 1971, to June 29, 1973. This set of data are plotted in Fig. 2 of the paper by Daniel et al which is presented in preprint form in Appendix V of this report.

Table 2 shows the Sr^{89}, Sr^{90} and Sr^{89}/Sr^{90} ratios measured in rain samples collected at Ankara, Turkey, from March 25 through October 17, 1972. The sample collection and preliminary reduction and dissolution were completed by Professor Erfüg Edgüer and Ms. Nur Gökien at the Department of Chemistry of Hacettepe University in Ankara, Turkey. The sample collection and analysis was under the direction of Dr. J. L. Meason of the University of Arkansas. The final purification, counting and analysis of the data was done at the University of Arkansas. The Sr^{89}/Sr^{90} ratio data are plotted in Fig. 2 of the preprint by Daniel et al which appears in Appendix V of this report.

Table 3 gives the Sr^{89}, Sr^{90} and Sr^{89}/Sr^{90} ratios measured in rain samples collected in Tokyo, Japan, from March 26 through June 19, 1972. The samples were collected at the Radiation Center of Asaka Prefecture at Osaka, Japan (34°N, 135°E) by Dr. Y. Miyake and co-workers. The preliminary

reduction and dissolution of the samples were done by Professor P. K. Kuroda while on sabbatical during the summer of 1972. The final reduction of the samples was done at the University of Arkansas. The Sr^{89}/Sr^{90} ratio data are also plotted in Fig. 2 of the preprint by Daniel et al in Appendix V of this report.

The revised values of the monthly average concentration of Sr^{90} are given in Table 4 and the bimonthly average concentrations are in Table 5, for the period from September, 1971 through June, 1973. The monthly and bimonthly concentrations of Sr^{90} are plotted in Fig. 1 of the paper by Daniel et al in Appendix V of this report.

Table 1. Sr^{89} and Sr^{90} Concentrations in Rain at Fayetteville, Arkansas, from August 3, 1971, through June 29, 1973.

Date	Rainfall (mm)	Sr^{89} (pc/l)	Sr^{90} (pc/l)	Sr^{89}/Sr^{90} (pc/pc)
1971				
Aug. 3	0.3	10.6 ± 1.3	5.7 ± 0.7	1.9 ± 0.3
Aug. 6-7	32.5	1.9 ± 0.2	0.82 ± 0.09	2.3 ± 0.4
Aug. 11	0.2	5.2 ± 0.6	3.1 ± 0.4	1.7 ± 0.3
Aug. 14-15	1.7	3.6 ± 0.5	1.6 ± 0.2	2.3 ± 0.5
Aug. 22	6.6	1.6 ± 0.2	0.68 ± 0.10	2.3 ± 0.5
Sep. 5	10.6	1.3 ± 0.1	0.59 ± 0.07	2.2 ± 0.4
Sep. 6	2.2	6.6 ± 0.8	2.3 ± 0.2	2.8 ± 0.5
Sep. 8	5.6	1.5 ± 0.2	0.58 ± 0.10	2.6 ± 0.6
Sep. 9	24.0	1.6 ± 0.2	1.0 ± 0.1	1.6 ± 0.3
Sep. 15-19	34.0	1.5 ± 0.2	0.80 ± 0.09	1.8 ± 0.3
Sep. 24-25	17.6	0.60 ± 0.07	0.25 ± 0.03	2.4 ± 0.4
Oct. 2-3	41.1	0.52 ± 0.04	0.16 ± 0.02	3.2 ± 0.5
Oct. 10	0.2	3.3 ± 0.1	1.7 ± 0.1	1.9 ± 0.1
Oct. 18-21	8.9	0.53 ± 0.10	0.26 ± 0.05	2.0 ± 0.5
Oct. 23	5.6	1.4 ± 0.4	1.6 ± 0.4	0.89 ± 0.35
Oct. 27	6.9	0.79 ± 0.13	0.69 ± 0.11	1.1 ± 0.3
Nov. 1	1.3	1.1 ± 0.1	0.97 ± 0.07	1.1 ± 0.1
Nov. 6	4.3	1.6 ± 0.3	1.5 ± 0.2	1.1 ± 0.3
Nov. 7	4.9	1.0 ± 0.2	0.70 ± 0.16	1.4 ± 0.5
Nov. 17-18	28.4	0.24 ± 0.05	0.22 ± 0.05	1.1 ± 0.3
Nov. 22-23	13.5	0.17 ± 0.03	0.18 ± 0.03	0.94 ± 0.23
Nov. 25-26	1.0	3.7 ± 0.1	3.7 ± 0.1	0.98 ± 0.03
Nov. 28	0.5	16.6 ± 0.9	1.6 ± 0.1	10.2 ± 0.8
Dec. 2-3	7.6	2.8 ± 0.3	0.56 ± 0.07	5.0 ± 0.8
Dec. 5-6	8.9	0.62 ± 0.03	0.38 ± 0.02	1.6 ± 0.1
Dec. 8-10	113.5	0.21 ± 0.08	0.20 ± 0.08	1.0 ± 0.2
Dec. 13-14	29.0	0.60 ± 0.11	0.30 ± 0.05	2.0 ± 0.5
Dec. 29	14.5	0.40 ± 0.04	0.36 ± 0.03	1.1 ± 0.1
1972				
Jan. 1	1.0	3.4 ± 0.1	5.9 ± 0.4	0.58 ± 0.05
Jan. 3-4	3.8	2.6 ± 0.1	3.1 ± 0.1	0.83 ± 0.04
Jan. 27-28	9.4	2.2 ± 0.3	0.70 ± 0.11	3.1 ± 0.7
Feb. 2-3	2.5	11.7 ± 0.7	1.6 ± 0.2	7.1 ± 1.1
Feb. 5	16.3	3.3 ± 0.1	0.86 ± 0.07	3.8 ± 0.3
Feb. 11-12	12.9	2.2 ± 0.3	0.91 ± 0.10	2.4 ± 0.4

Table 1 (Continued)

Date	Rainfall (mm)	Sr^{89} (pc/l)	Sr^{90} (pc/l)	Sr^{89}/Sr^{90} (pc/pc)
Mar. 6	0.2	1.8 ± 0.7	2.0 ± 0.3	0.91 ± 0.39
Mar. 21	16.0	0.41 ± 0.22	1.0 ± 0.2	0.39 ± 0.23
Mar. 23	11.4	1.3 ± 0.4	2.4 ± 0.1	0.56 ± 0.19
Mar. 24	0.8	19.8 ± 3.1	7.0 ± 0.5	2.8 ± 0.5
Mar. 26	6.9	12.8 ± 1.2	0.64 ± 0.16	20.0 ± 5.3
Mar. 28	2.0	43.8 ± 8.0	2.8 ± 0.7	15.9 ± 4.8
Apr. 3	5.3	3.4 ± 0.4	2.4 ± 0.3	1.4 ± 0.2
Apr. 13	12.9	3.7 ± 0.5	1.2 ± 0.1	3.0 ± 0.7
Apr. 14	4.6	6.6 ± 0.7	0.72 ± 0.3	9.1 ± 3.4
Apr. 15	12.7	8.9 ± 2.9	0.60 ± 0.11	14.9 ± 5.4
Apr. 19-20	63.5	16.4 ± 2.9	0.55 ± 0.04	29.8 ± 5.6
Apr. 27	20.0	25.7 ± 2.3	0.61 ± 0.15	42.1 ±11.0
May 1	31.1	10.6 ± 1.5	0.44 ± 0.10	25.5 ± 6.9
May 8	3.0	15.6 ± 1.8	1.3 ± 0.1	12.4 ± 1.9
May 12	7.5	27.7 ± 1.9	1.1 ± 0.1	25.2 ± 2.9
May 22	4.6	72.6 ± 5.4	4.6 ± 0.7	15.6 ± 3.5
May 26	2.5	27.5 ± 2.2	2.1 ± 0.1	13.0 ± 1.1
May 28	5.3	25.8 ± 2.9	1.8 ± 0.4	14.3 ± 3.7
May 29	8.6	18.3 ± 2.1	1.3 ± 0.1	14.0 ± 1.6
June 14	9.1	8.2 ± 0.8	0.51 ± 0.2	16.1 ± 3.0
June 19	10.4	10.3 ± 1.5	0.82 ± 0.20	12.6 ± 3.6
June 24-25	10.9	20.0 ± 1.8	1.3 ± 0.1	15.4 ± 2.1
June 26	3.6	25.7 ± 2.1	1.3 ± 0.1	19.1 ± 2.6
June 26-27	18.8	7.6 ± 0.9	0.64 ± 0.12	11.9 ± 2.4
June 30	12.9	8.4 ± 0.90	0.84 ± 0.11	10.0 ± 1.6
July 1-2	4.9	8.1 ± 0.8	0.82 ± 0.15	9.8 ± 2.0
July 3-4	6.1	17.1 ± 3.1	1.0 ± 0.1	16.7 ± 3.9
July 12-13	128.3	8.8 ± 0.9	0.56 ± 0.16	15.7 ± 4.8
July 15	8.6	5.7 ± 0.6	0.44 ± 0.04	13.0 ± 1.7
July 18	0.5	7.4 ± 0.8	0.75 ± 0.07	9.9 ± 1.4
July 28	6.6	5.3 ± 0.7	0.90 ± 0.09	5.9 ± 1.0
July 29	5.6	5.7 ± 0.9	0.79 ± 0.16	7.2 ± 1.8
Aug. 4	17.6	0.71 ± 0.06	0.14 ± 0.01	5.1 ± 0.6
Aug. 14	2.8	3.3 ± 0.6	0.67 ± 0.13	4.9 ± 1.3
Aug. 22	9.1	1.8 ± 0.4	0.42 ± 0.12	4.4 ± 1.6
Aug. 24	17.3	0.98 ± 0.24	0.39 ± 0.08	2.5 ± 0.8
Aug. 31	6.6	2.1 ± 0.4	0.64 ± 0.14	3.2 ± 0.9

Table 1 (Continued)

Date	Rainfall (mm)	Sr^{89} (pc/l)	Sr^{90} (pc/l)	Sr^{89}/Sr^{90} (pc/pc)
Sep. 4-5	5.6	1.24 ± 0.14	0.55 ± 0.06	2.25 ± 0.35
Sep. 8	8.9	0.77 ± 0.16	0.22 ± 0.06	3.5 ± 1.2
Sep. 9	5.1	0.47 ± 0.05	0.23 ± 0.03	2.04 ± 0.34
Sep. 14	1.8	0.96 ± 0.28	0.53 ± 0.18	1.8 ± 0.8
Sep. 19	1.5	1.2 ± 0.3	0.55 ± 0.18	2.1 ± 0.9
Sep. 21-25	63.5	0.17 ± 0.03	0.10 ± 0.02	1.6 ± 0.4
Sep. 27	1.0	0.97 ± 0.20	0.53 ± 0.18	1.8 ± 0.7
Sep. 30	119.4	0.17 ± 0.05	0.10 ± 0.03	1.7 ± 0.6
Oct. 6	1.0	1.2 ± 0.2	0.79 ± 0.20	1.5 ± 0.5
Oct. 14	9.4	1.1 ± 0.3	0.71 ± 0.22	1.6 ± 0.7
Oct. 18	7.4	0.23 ± 0.05	0.17 ± 0.04	1.3 ± 0.4
Oct. 21	56.4	0.25 ± 0.02	0.19 ± 0.06	1.3 ± 0.4
Oct. 27	1.3	0.73 ± 0.30	0.60 ± 0.20	1.2 ± 0.6
Oct. 30	103.4	0.05 ± 0.02	0.04 ± 0.01	1.2 ± 0.6
Nov. 6	26.2	0.03 ± 0.01	0.03 ± 0.02	1.0 ± 0.7
Nov. 8-9	5.3	0.54 ± 0.19	0.52 ± 0.14	1.0 ± 0.5
Nov. 12-13	46.2	0.11 ± 0.03	0.12 ± 0.03	0.92 ± 0.34
Nov. 14-16	12.9	0.24 ± 0.09	0.26 ± 0.06	0.92 ± 0.40
Nov. 21	3.8	0.15 ± 0.08	0.16 ± 0.03	0.94 ± 0.53
Nov. 26	4.1	0.21 ± 0.07	0.25 ± 0.07	0.84 ± 0.37
Nov. 26-27	1.3	0.33 ± 0.10	0.33 ± 0.14	1.0 ± 0.5
Dec. 4-5	2.8	0.13 ± 0.06	0.36 ± 0.04	0.36 ± 0.17
Dec. 12-19	25.4	0.05 ± 0.04	0.12 ± 0.02	0.42 ± 0.34
Dec. 20	2.5	0.20 ± 0.12	0.64 ± 0.07	0.31 ± 0.19
Dec. 29	10.7	0.20 ± 0.08	0.35 ± 0.14	0.57 ± 0.32
1973				
Jan. 4-9	25.1	0.09 ± 0.05	0.21 ± 0.07	0.43 ± 0.27
Jan. 17-18	15.0	0.21 ± 0.08	0.60 ± 0.08	0.35 ± 0.14
Jan. 20-22	18.3	0.08 ± 0.02	0.32 ± 0.04	0.25 ± 0.07
Jan. 25-28	5.1	0.29 ± 0.07	0.68 ± 0.07	0.43 ± 0.11
Jan. 31	22.1	0.038± 0.02	0.10 ± 0.04	0.38 ± 0.25
Feb. 7-8	53.3	0.06 ± 0.02	0.21 ± 0.04	0.29 ± 0.11
Feb. 11-12	2.5	0.16 ± 0.08	0.52 ± 0.12	0.31 ± 0.17
Feb. 17-18	2.0	0.27 ± 0.11	0.61 ± 0.19	0.44 ± 0.23
Feb. 25-26	2.5	0.09 ± 0.06	0.57 ± 0.12	0.16 ± 0.11
Mar. 1-2	51.6	0.04 ± 0.02	0.23 ± 0.05	0.17 ± 0.09
Mar. 5-7	21.3	0.05 ± 0.03	0.39 ± 0.04	0.13 ± 0.08
Mar. 9-10	47.0	0.03 ± 0.01	0.29 ± 0.07	0.10 ± 0.04
Mar. 12	9.2	0.04 ± 0.02	0.18 ± 0.04	0.22 ± 0.12

Table 1 (Continued)

Date	Rainfall (mm)	Sr^{89} (pc/l)	Sr^{90} (pc/l)	Sr^{89}/Sr^{90} (pc/pc)
Mar. 19-20	32.5	0.33 ± 0.06	0.91 ± 0.24	0.36 ± 0.16
Mar. 22-23	10.4	0.22 ± 0.08	1.14 ± 0.28	0.19 ± 0.08
Mar. 24-26	54.6	----	0.19 ± 0.04	----
Mar. 27	3.6	----	1.1 ± 0.1	----
Mar. 30-31	14.5	----	0.62 ± 0.15	----
Apr. 2-4	26.4	----	0.85 ± 0.08	----
Apr. 6-9	18.8	----	0.80 ± 0.08	----
Apr. 15	34.0	----	0.15 ± 0.10	----
Apr. 19	10.7	----	0.31 ± 0.06	----
Apr. 19-22	84.6	----	0.19 ± 0.07	----
Apr. 23-25	39.4	----	0.33 ± 0.03	----
May 1	5.3	----	1.0 ± 0.1*	----
May 6-7	41.7	----	0.52 ± 0.12	----
May 11	49.8	----	0.38 ± 0.10	----
May 21	27.7	----	0.80 ± 0.16	----
May 22	0.4	----	1.57 ± 0.39	----
May 23	16.0	----	0.41 ± 0.10	----
May 24	7.1	----	----(a)	----
May 27	12.7	----	0.77 ± 0.09	----
May 28	4.6	----	0.15 ± 0.07	----
June 1	32.8	----	0.24 ± 0.06	----
June 3	53.8	----	0.13 ± 0.02	----
June 4-5	23.9	----	0.32 ± 0.06	----
June 12	6.6	----	0.09 ± 0.03	----
June 13-15	1.8	----	0.52 ± 0.08	----
June 16	14.2	----	0.17 ± 0.03	----
June 18-19	63.5	----	0.13 ± 0.07	----
June 19	1.8	----	0.13 ± 0.09	----
June 27	4.3	----	0.43 ± 0.21	----
June 29	18.3	----	1.1 ± 0.4	----

* Radiostrontium analysis for May-June 1973 completed by N. G. Sumerlin.

(a) Measurements not yet completed.

Table 2. Sr^{89} and Sr^{90} Concentrations in Rain at Ankara, Turkey, from March 25 through October 17, 1972.*

Date	Sr^{89} (pc/l)	Sr^{90} (pc/l)	Sr^{89}/Sr^{90} (pc/pc)
1972			
Mar. 25	1.4 ± 0.2	0.85 ± 0.03	1.7 ± 0.2
Mar. 28	50.9 ±22.3	6.9 ± 0.1	7.3 ± 3.2
Mar. 28-29	111.4 ±41.0	3.0 ± 0.2	37.0 ±13.8
Apr. 2	7.0 ± 5.7	0.68 ± 0.07	10.3 ± 8.4
Apr. 10-11	118.3 ±72.0	9.1 ± 0.4	13.0 ± 7.9
Apr. 14-17	21.4 ± 3.5	2.0 ± 0.1	10.4 ± 1.8
Apr. 17-18	37.0 ±14.8	4.9 ± 0.4	7.6 ± 3.1
Apr. 20-21	23.1 ±10.7	2.3 ± 0.1	10.1 ± 2.2
Apr. 22-24	6.4 ± 3.1	0.93 ± 0.03	6.9 ± 3.3
Apr. 28-May 2	7.2 ± 3.6	0.84 ± 0.05	8.6 ± 4.3
May 10-12	8.1 ± 4.8	1.1 ± 0.1	7.6 ± 4.6
May 18-22	15.2 ± 1.6	1.6 ± 0.1	9.5 ± 1.1
May 24	41.6 ± 6.2	2.8 ± 0.2	15.1 ± 2.5
May 28	18.1 ± 4.5	2.0 ± 0.1	8.9 ± 2.2
June 2	5.1 ± 1.9	0.45 ± 0.02	11.4 ± 4.2
June 10	96.0 ±16.5	6.9 ± 0.4	13.8 ± 2.5
July 2	10.5 ± 3.7	1.9 ± 0.1	5.5 ± 2.0
July 3	57.1 ±14.9	5.4 ± 0.1	10.6 ± 2.8
July 4	17.3 ± 3.6	1.8 ± 0.1	9.7 ± 2.0
July 11	23.8 ± 6.1	2.8 ± 0.2	8.6 ± 2.2
July 18	5.7 ± 3.1	0.35 ± 0.01	16.3 ± 8.8
July 22	5.2 ± 0.6	0.61 ± 0.07	8.6 ± 1.3
July 28	18.5 ± 4.4	3.1 ± 0.02	6.0 ± 1.5
July 30	14.9 ± 1.6	2.3 ± 0.3	6.4 ± 1.0
Aug. 31	2.2 ± 1.0	0.76 ± 0.05	2.9 ± 1.3
Sep. 1	2.5 ± 0.8	0.93 ± 0.06	2.7 ± 0.9
Sep. 8	9.4 ± 2.2	1.8 ± 0.1	5.2 ± 2.2
Sep. 26	1.1 ± 0.3	0.53 ± 0.04	2.1 ± 0.6
Sep. 28	0.42 ± 0.33	0.43 ± 0.08	0.98 ± 0.79
Sep. 29	1.6 ± 0.8	1.1 ± 0.1	1.5 ± 0.7
Oct. 11	1.3 ± 0.1	0.93 ± 0.10	1.4 ± 0.2
Oct. 17	0.13 ± 0.04	0.18 ± 0.02	0.72 ± 0.23

* Sample collection and preliminary reduction and dissolution were completed by Professor Erfüg Edgüer and Ms. Nur Gökien, Chemistry Asst., at the Department of Chemistry, Hacettepe University, Ankara, Turkey, (40°N, 33°E). The sample collection and analysis was under the direction of Dr. J. L. Meason.

Table 3. Sr^{89} and Sr^{90} Concentrations in Rain at Tokyo, Japan, from March 26 through June 19, 1972.*

Date	Rainfall (mm)	Sr^{89} (pc/l)	Sr^{90} (pc/l)	Sr^{89}/Sr^{90} (pc/pc)
1972				
Mar. 26	13.5	2.5 ± 0.7	0.83 ± 0.07	3.0 ± 0.8
Mar. 30	13.8	1.3 ± 0.7	0.92 ± 0.06	1.5 ± 0.8
Apr. 7	43.0	1.2 ± 0.4	0.29 ± 0.04	4.1 ± 1.4
Apr. 8	20.6	1.0 ± 0.1	0.16 ± 0.02	6.5 ± 1.1
Apr. 12	26.6	0.33 ± 0.11	0.24 ± 0.01	1.4 ± 0.5
Apr. 23	24.4	0.90 ± 0.31	0.55 ± 0.04	1.6 ± 0.6
June 19	30.6	5.7 ± 0.6	0.47 ± 0.03	12.2 ± 1.4

* Samples were collected at the Radiation Center of Osaka Prefecture at Osaka, Japan, (34°N, 135°E) by Dr. Y. Miyake and co-workers. Preliminary reduction and dissolution of the samples were done by Professor P. K. Kuroda while on sabbatical during the summer of 1972.

Table 4. Monthly Average Sr^{90} Concentrations in Rain at Fayetteville, Arkansas, from August 1971, through June 1973.

Month	Monthly Total Rainfall (mm)	Monthly Total Sr^{90} Fallout (pc/m^2)	Monthly Average Sr^{90} Concentration (pc/l)
1971			
Aug.	41.3	36.07	0.87
Sep.	94.0	70.19	0.75
Oct.	62.7	22.89	0.37
Nov.	53.9	24.10	0.45
Dec.	173.5	44.25	0.26
1972			
Jan.	14.2	24.39	1.72
Feb.	31.7	29.86	0.94
Mar.	37.3	60.16	1.61
Apr.	119.0	85.53	0.73
May	62.6	72.96	1.16
June	65.7	54.68	0.83
July	160.6	96.48	0.60
Aug.	53.4	19.13	0.36
Sep.	206.8	25.23	0.12
Oct.	178.9	24.34	0.14
Nov.	99.8	14.52	0.15
Dec.	41.4	9.40	0.23
1973			
Jan.	85.6	25.81	0.30
Feb.	60.3	15.12	0.25
Mar.	244.7	100.34	0.41
Apr.	213.9	74.98	0.35
May	109.87	(56.40)(*)	(0.51)(*)
June	186.42	49.62	0.26

(*) The May data are tentative, since Sr^{90} analysis for one rain sample has not yet been completed.

Table 5. Bi-Monthly Average Sr^{90} Concentrations in Rain at Fayetteville, Arkansas, from September 1971, through June 1973.

Bi-Monthly Period	Bi-Monthly Average Sr^{90} Concentration (pc/l)
1971	
Sep. - Oct.	0.59
Nov. - Dec.	0.30
1972	
Jan. - Feb.	1.18
Mar. - Apr.	0.94
May - June	0.99
July - Aug.	0.54
Sep. - Oct.	0.13
Nov. - Dec.	0.17
1973	
Jan. - Feb.	0.28
Mar. - Apr.	0.38
May - June	0.34

APPENDIX X

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Cs^{137}, Sr^{90}, and Cs^{137}/Sr^{90} Ratios and the Bimonthly Average Cs^{137}/Sr^{90} Ratios in Rains Collected at Fayetteville, Arkansas from January 24, 1967, through June 27, 1969.

R. D. Sherrill and N. G. Sumerlin

The Cs^{137} and Sr^{90} concentrations were measured in a series of rain samples collected at Fayetteville (36°N, 94°W), Arkansas, from January 24, 1967, through June 27, 1969. The variations in the Cs^{137}/Sr^{90} have been interpreted as resulting from the variation in the size distributions of particulate matter following a nuclear explosion in the atmosphere. The Cs^{137}/Sr^{90} ratio data presented in Table 1 are also plotted in Fig. 1 of the report by Sherrill *et al* which is presented in Appendix VI of this report. A further discussion of the results are also given in the report by Sherrill *et al*.

Table 1. Cs^{137}, Sr^{90}, Cs^{137}/Sr^{90} ratio and the bimonthly average Cs^{137}/Sr^{90} ratio in rains collected at Fayetteville, Arkansas, from January 24, 1967, to June 27, 1969.

Date	Amt. (mm)	Cs^{137} pc/l	Sr^{90} pc/l	$\frac{Cs^{137}}{Sr^{90}}$	$\frac{Cs^{137}}{Sr^{90}}$ Bimonthly Average
1967					
Jan. 24	.79	3.62 ± .14	----	----	
Feb. 2	28.58	.84 ± .06	----	----	.38
Feb. 16	23.11	.65 ± .08	1.89 ± .28	.34	
Feb. 27	3.18	2.46 ± .07	5.48 ± .39	.45	
Mar. 5	24.60	----	2.65 ± .16	----	
Mar. 7	2.30	----	5.23 ± .22	----	
Mar. 17	3.18	----	5.64 ± .18	----	
Apr. 9	35.26	----	.95 ± .07	----	1.05
Apr. 12	45.40	.93 ± .05	.73 ± .10	1.27	
Apr. 21	3.91	3.62 ± .17	----	----	
Apr. 22	.69	3.15 ± .11	6.51 ± .15	.48	
Apr. 25	34.93	2.18 ± .06	1.58 ± .12	1.38	
May 5	66.68	1.08 ± .05	.40 ± .02	2.70	
May 19	12.54	2.95 ± .23	3.12 ± .07	.95	
June 10	5.72	1.62 ± .13	1.22 ± .14	1.33	
June 21	6.02	6.59 ± .13	2.55 ± .22	2.58	1.34
June 28	36.0	.57 ± .07	----	----	
June 29	26.50	.71 ± .11	----	----	
June 30	1.68	2.55 ± .10	----	----	
July 1	8.40	.98 ± .07	----	----	
July 1	14.50	.45 ± .07	----	----	
July 5	.24	.46 ± .05	----	----	
July 12	19.53	.92 ± .09	----	----	
July 16	4.57	.45 ± .06	----	----	----
July 25	2.06	2.25 ± .07	----	----	
July 27	3.17	1.00 ± .06	----	----	
Aug. 2	14.28	.53 ± .10	.75 ± .21	.71	
Aug. 9	34.3	.29 ± .08	----	----	
Aug. 23	10.0	3.83 ± .15	----	----	
Sep. 2	7.77	1.02 ± .07	.95 ± .22	1.07	
Sep. 14	39.62	.26 ± .03	.12 ± .06	2.17	
Sep. 20	28.44	.18 ± .06	.17 ± .12	1.06	
Sep. 27	10.41	.72 ± .05	1.00 ± .20	.72	

Table 1 (Continued)

Date	Amt. (mm)	Cs^{137} pc/l	Sr^{90} pc/l	$\frac{Cs^{137}}{Sr^{90}}$	$\frac{Cs^{137}}{Sr^{90}}$ Bimonthly Average
1967					
Oct. 7	4.86	.54 ± .06	.37 ± .12	1.46	1.5
Oct. 11	.96	.38 ± .06	.60 ± .07	.63	
Oct. 14	88.9	.17 ± .05	.14 ± .03	1.21	
Oct. 23	4.08	2.18 ± .09	.88 ± .06	2.48	
Oct. 29	114.3	.20 ± .03	.07 ± .03	2.86	
Nov. 2	12.7	.22 ± .05	.23 ± .10	.96	
Nov. 10	.76	.76 ± .05	.33 ± .10	2.30	
Nov. 28	16.76	.62 ± .03	.32 ± .03	1.94	
Dec. 2	.30	1.71 ± .08	.89 ± .27	1.80	
Dec. 5	6.09	.42 ± .03	.36 ± .06	1.17	
Dec. 9	32.3	.37 ± .05	.18 ± .05	2.06	1.45
Dec. 13	45.75	.07 ± .05	.09 ± .03	.78	
Dec. 16	4.06	.26 ± .03	.13 ± .05	2.0	
Dec. 20	33.53	.34 ± .03	.20 ± .05	1.7	
Dec. 27	1.78	.36 ± .05	.47 ± .06	.77	
Dec. 30	5.33	1.00 ± .06	1.04 ± .06	.96	
1968					
Jan. 5	6.00	.67 ± .05	1.26 ± .07	.53	
Jan. 13	.51	2.03 ± .07	1.86 ± .10	1.09	
Jan. 18	3.30	1.06 ± .08	2.66 ± .08	.40	
Jan. 19	.38	1.55 ± .07	3.81 ± .14	.41	
Jan. 22	2.79	1.20 ± .09	.92 ± .06	1.30	.75
Jan. 27	101.6	.50 ± .03	.73 ± .06	.68	
Feb. 14	1.52	3.10 ± .09	2.90 ± .19	1.07	
Feb. 21	3.81	.90 ± .05	.89 ± .06	1.01	
Feb. 24	2.03	2.24 ± .09	1.47 ± .08	1.52	
Feb. 25	.76	1.75 ± .10	1.79 ± .09	.98	
Feb. 27	7.11	.96 ± .09	.84 ± .06	1.14	
Feb. 28	.51	2.55 ± .12	2.13 ± .08	1.20	
Mar. 12	71.12	.59 ± .06	.44 ± .05	1.34	
Mar. 15	2.29	2.80 ± .10	4.16 ± .15	.67	
Mar. 19	74.93	.85 ± .07	.86 ± .06	.99	
Mar. 31	16.00	1.77 ± .09	2.12 ± .10	.83	

Table 1 (Continued)

Date	Amt. (mm)	Cs^{137} pc/l	Sr^{90} pc/l	$\frac{Cs^{137}}{Sr^{90}}$	$\frac{Cs^{137}}{Sr^{90}}$ Bimonthly Average
1968					
Apr. 1	20.83	.95 ± .07	.30 ± .05	3.17	.91
Apr. 12	1.00	2.72 ± .11	10.21 ± .35	.26	
Apr. 15	6.35	3.70 ± .12	2.37 ± .16	1.56	
Apr. 18	49.1	.42 ± .07	.25 ± .05	1.68	
Apr. 21	4.06	1.24 ± .06	2.08 ± .08	.60	
Apr. 22	1.78	2.96 ± .09	5.04 ± .17	.59	
Apr. 22	7.62	1.32 ± .11	1.54 ± .07	.86	
Apr. 27	6.86	3.00 ± .12	8.82 ± .34	.34	
May 3	3.81	2.66 ± .09	4.03 ± .07	.66	
May 7	6.60	4.45 ± .12	4.23 ± .15	1.05	
May 9	38.10	1.32 ± .06	1.57 ± .09	.84	
May 13	20.57	.74 ± .03	.67 ± .06	1.10	
May 13	24.13	1.78 ± .10	1.52 ± .08	1.17	
May 15	8.89	2.11 ± .07	1.29 ± .06	1.64	
May 16	1.7	3.34 ± .08	7.22 ± .26	.46	
May 20	.64	5.50 ± .10	6.21 ± .21	.89	
May 25	43.69	1.73 ± .05	1.51 ± .07	1.15	1.20
May 27	7.87	2.44 ± .09	2.77 ± .11	.88	
May 31	1.52	4.24 ± .11	5.07 ± .21	.84	
June 1	7.00	2.83 ± .09	1.71 ± .07	1.65	
June 7	1.27	2.70 ± .09	4.18 ± .17	.65	
June 15	11.0	2.96 ± .10	2.80 ± .17	1.06	
June 24	36.5	1.54 ± .07	.24 ± .03	6.42	
June 25	33.0	.76 ± .05	.34 ± .05	2.24	
July 1	33.0	1.31 ± .07	.43 ± .06	3.05	
July 12	16.51	2.29 ± .08	2.08 ± .12	1.10	
July 14	2.54	1.05 ± .04	1.80 ± .10	.58	
July 18	3.24	1.25 ± .04	.63 ± .08	1.98	
July 22	2.54	1.29 ± .03	.60 ± .08	2.15	
Aug. 8	17.53	1.23 ± .04	.15 ± .07	8.2	1.84
Aug. 10	6.60	1.01 ± .06	.55 ± .06	1.84	
Aug. 11	5.59	.66 ± .04	.29 ± .06	2.28	
Aug. 14	11.43	1.27 ± .07	.42 ± .06	3.02	
Aug. 15	7.11	.57 ± .03	.56 ± .07	1.02	
Aug. 25	37.1	.47 ± .03	.11 ± .06	4.27	
Aug. 30	17.27	.56 ± .06	.78 ± .09	.72	
Sep. 3	1.02	1.11 ± .04	.47 ± .08	2.36	
Sep. 15	6.40	.66 ± .03	.47 ± .08	1.40	
Sep. 16	32.51	.21 ± .02	.16 ± .07	1.31	
Sep. 24	63.75	.80 ± .04	.53 ± .07	1.51	1.52

Table 1 (Continued)

Date	Amt. (mm)	Cs^{137} pc/l	Sr^{90} pc/l	$\frac{Cs^{137}}{Sr^{90}}$	$\frac{Cs^{137}}{Sr^{90}}$ Bimonthly Average
1968					
Oct. 5	18.03	.86 ± .04	.63 ± .06	1.37	
Oct. 8	12.45	1.04 ± .06	.44 ± .06	2.36	
Oct. 16	12.95	.72 ± .03	.37 ± .06	1.95	
Oct. 21	1.78	1.56 ± .07	2.08 ± .12	.75	
Nov. 6	.65	.99 ± .04	1.12 ± .11	.88	
Nov. 10	14.48	.81 ± .04	.39 ± .06	2.08	
Nov. 16	63.50	.21 ± .02	.44 ± .07	.48	
Nov. 23	12.79	.99 ± .03	.78 ± .07	1.27	1.24
Nov. 27	94.0	.47 ± .03	.28 ± .07	1.68	
Nov. 30	7.62	.25 ± .02	.25 ± .05	1.00	
Dec. 12	2.5	1.13 ± .06	1.06 ± .09	1.07	
Dec. 18	5.08	.59 ± .04	.43 ± .07	1.37	
Dec. 21	23.5	.53 ± .03	.27 ± .07	1.96	
1969					
Jan. 22	.25	1.10 ± .06	1.63 ± .09	.67	
Jan. 27	1.50	2.52 ± .10	1.92 ± .14	1.31	
Jan. 29	67.0	.84 ± .04	.30 ± .07	2.8	
Jan. 31	9.65	.47 ± .03	.76 ± .07	.62	
Feb. 5	4.3	1.33 ± .06	1.77 ± .13	.75	1.52
Feb. 7	4.5	1.85 ± .06	1.83 ± .17	1.01	
Feb. 8	.5	1.69 ± .06	2.62 ± .20	.65	
Feb. 14	12.9	.63 ± .04	.38 ± .08	1.66	
Feb. 20	31.8	1.02 ± .06	.77 ± .07	1.32	
Feb. 27	.94	3.58 ± .11	4.73 ± .50	.76	
Mar. 6	6.0	1.06 ± .07	.70 ± .10	1.51	
Mar. 7	8.0	2.85 ± .11	3.40 ± .17	.84	
Mar. 23	69.9	.81 ± .04	.74 ± .08	1.09	
Mar. 26	0.5	6.08 ± .16	3.04 ± .13	2.00	
Mar. 28	3.05	3.03 ± .09	3.23 ± .29	.94	
Apr. 1	6.30	1.91 ± .08	2.03 ± .17	.94	.80
Apr. 4	7.80	1.60 ± .07	1.67 ± .10	.96	
Apr. 8	0.60	1.31 ± .07	2.02 ± .10	.65	
Apr. 13	7.0	.59 ± .03	1.08 ± .07	.55	
Apr. 16	20.60	.71 ± .04	1.66 ± .14	.43	
Apr. 18	4.06	1.26 ± .07	.91 ± .28	1.38	
Apr. 27	53.0	.68 ± .04	1.16 ± .09	.59	

Table 1 (Continued)

Date	Amt. (mm)	Cs^{137} pc/l	Sr^{90} pc/l	$\frac{Cs^{137}}{Sr^{90}}$	$\frac{Cs^{137}}{Sr^{90}}$ Bimonthly Average
1969					
May 5	7.3	1.73 ± .04	2.38 ± .11	.73	
May 7	8.2	1.90 ± .08	1.54 ± .13	1.23	
May 8	12.6	1.01 ± .03	1.19 ± .10	.85	
May 16	52.5	.72 ± .03	.42 ± .07	1.71	
May 17	38.5	1.16 ± .06	.55 ± .06	2.11	
May 24	1.8	2.48 ± .09	2.57 ± .13	.96	
May 25	6.9	1.65 ± .07	1.24 ± .10	1.33	
May 26	3.0	1.21 ± .04	1.27 ± .08	.95	1.08
May 30	1.1	2.83 ± .10	3.85 ± .21	.74	
May 31	22.2	1.41 ± .08	1.06 ± .11	1.33	
June 9	19.6	.54 ± .03	.30 ± .07	1.80	
June 12	5.3	1.64 ± .09	1.60 ± .10	1.03	
June 13	24.4	2.36 ± .08	3.19 ± .18	.74	
June 17	11.5	.74 ± .04	.59 ± .06	1.25	
June 20	10.0	2.07 ± .09	1.70 ± .08	1.22	
June 24	15.0	3.38 ± .08	4.20 ± .38	.80	
June 27	2.0	5.08 ± .11	3.87 ± .25	1.31	