COO-1199-8

Illinois State Water Survey
at the
University of Illinois
Urbana, Illinois

STUDY OP RAINOUT OF RADIOACTIVITY IN ILLINOIS

Fourth Progress Report
Contract Number AT(11-1)-1199
October 1965

Sponsored by

United States Atomic Energy Commission
Fallout Studies Branch
Division of Biology and Medicine
Washington, D. C.

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CONTENTS

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INTRODUCTION

On June 1, 1962, the Illinois State Water Survey entered into a 12-month contract with the Atomic Energy Commission to conduct research on the rainout of radioactivity, with primary emphasis on convective storm studies. As a result of periodic extensions of the initial contract, this research has continued through 1965.

During the 1962-1965 period, particular emphasis has been placed upon evaluation of the time and space variability of radioactive rainout and its relationship to precipitation characteristics, through use of dense raingage networks, radar observations, radiochemical analyses of gross beta and selected isotopes in storm rainwater, and synoptic weather data. Surface rainwater sampling has been conducted on dense networks with areas ranging from 10 to 6000 square miles. In 1964, aircraft sampling of selected rainstorms was initiated to supplement the surface data. In 1965, air sampling at the surface and aloft was initiated in an effort to define further relationships between radioactive rainout and meteorological factors indicated by earlier studies.

Starting in spring 1963, the Water Survey engaged in a cooperative program with the AEC, U. S. Weather Bureau, Pennsylvania State University, and other interested groups in an aircraft sampling program aimed toward determining the conditions under which radioactive debris is transferred from the stratosphere to the troposphere and then to the surface. This program, known as Project Springfield, was initiated by Professor Edwin Danielsen of Penn State. The sampling program was continued in 1964 and 1965, and during these two years it was directed by G. E. Stout, Head, Atmospheric Sciences Section, Illinois State Water Survey.

In this report, Illinois studies in the 1962-1964 period will be described briefly, and some of the more important results of the research program briefly reviewed and summarized. Detailed discussion of all phases of the 1962-1964 research are contained in Progress Reports 1, 2, and 3 and Research Report 1 under this contract. The 1965 program will be described and preliminary analytical results discussed briefly. However, detailed discussion of results and conclusions from the 1965 program must await further data analyses. Results of analyses of data collected on Project Springfield are presented in Research Report 2, recently distributed, and will not be repeated here.

ACKNOWLEDGMENTS

The research on which this report is based was carried out under the general direction of G. E. Stout, Head, Atmospheric Sciences Section, Illinois State Water Survey. Douglas Jones supervised the radar operations and assisted in the analyses of data. Rena Gans supervised the rainwater analyses for gross beta activity and assisted in data analyses.

Analyses of Sr^{89}, Sr^{90}, and Ce^{144} in rainwater samples were made by Isotopes, Inc., Westwood, New Jersey, under a subcontract. Similarly, radiochemical analyses of air filter samples in 1965 were made under a subcontract with TRACERLAB, Waltham, Massachusetts.

Design and development of the automatic time samplers were accomplished primarily through the efforts and ingenuity of G. R. Boyd and Ronald Tibbetts. Substantial contributions to the design and development of the aircraft rainwater sampler were made in 1964 by Professor Gordon Martin, University of Illinois.

Pieter Feteris joined the staff in September 1964 and has been engaged primarily in the analyses of Project Springfield data. Credit is due many other staff members who aided in the collection and analysis of field data. Also, the assistance of cooperative observers in the collection of field data in 1962-1963 is gratefully acknowledged.

1962 PROGRAM

The 1962 program is discussed in detail in the First Progress Report. Under this program, rainwater samples of total storm rainfall were collected within three concentrated networks of raingages on areas of 10, 12, and 400 square miles in east central Illinois during spring and summer. Radar observations of most storms were made with horizontal and vertical scanning sets. Data analyses were undertaken to evaluates the spatial variability of radioactivity; the areal representativeness of point radioactivity samples; relations between storm rainfall volume, intensity, and duration and radioactivity concentration and deposition; and radioactivity-tropopause relations. Several case studies of individual storms were completed also in efforts to accumulate more knowledge of the relationship between rainout and meteorological factors.

In the areal variability study, a slight trend was found for the relative variability of radioactive rainout to decrease with increasing rainfall volume and storm duration. Radioactive rainout was found to have greater areal variability than the storm rainfall that brought it to the ground. Thus, in a 15-storm sample on two networks of 10 and 12 square miles, the average relative variability was 25 percent for gross beta rainout compared with 14 percent for storm rainfall. With respect to areal representativeness of point radioactivity samples, the same 15-storm sample indicated an average sampling error of 23 percent for beta concentration, 22 percent for beta deposition, and 9 percent for storm rainfall when the most central sampling point In each network was assumed to represent the average rainout over the network.

Analyses of areal patterns of the concentration and deposition of radioactive rainout and storm rainfall on the sampling networks revealed that in the majority of the storms a pattern relationship existed. In general, an inverse relationship was noted between storm rainfall and concentration of radioactivity, whereas a direct relationship was Indicated between radioactive deposition and amount of rainfall in the 1962 storms. However, exceptions to this trend were observed.

As a result of experiences in 1962, it was deemed highly desirable to obtain samples of the time distribution of radioactive rainout in storms as they passed over a given location, in addition to the areal distribution obtained from the station networks. Such information is pertinent to understanding of the rainout process in the atmosphere. Consequently, in fall 1962 design of an automatic time sampler was undertaken to obtain a series of rainwater samples through a storm as it passed a station. The experimental model was constructed and tested during the winter and early spring of 1962-1963.

1963 PROGRAM

Upon completion of tests on the experimental model of the automatic rainwater sampler in early spring 1963, construction of 15 samplers was undertaken. These were completed in time for inclusion in the Project Springfield program starting in mid-April. Each sampler was capable of collecting twelve 4-liter samples in a storm, without human attention. For Project Springfield, these samplers were installed over an area of 6000 square miles in central Illinois. Later the samplers were rearranged to obtain mesoscale measurements on an area of 400 square miles.

A 12-month extension of the original contract was granted on June 1, 1963, to continue the Illinois research program. Under this supplemental agreement, research was to be performed to: determine the time distribution of radioactivity in storms on an area basis; establish relations between radioactive rainout and various rainfall factors; ascertain characteristics of the radioactive rainout as revealed by radar observations and analyses; and develop a technique for aircraft sampling of rainwater below cloud bases.

Prom rainwater samples collected in spring-summer 1963, a total of 1029 samples were analyzed for gross beta and 400 samples for Sr^{89} and Sr^{90}. Analyses of station time distributions in 1963 storms indicated that the distribution pattern of gross beta and isotope concentrations through storms could be grouped into seven types, of which four types accounted for most of the cases. It was found that more than one type of time distribution may occur at various stations in a relatively small area within a convective storm system, although a particular type usually dominated in a particular storm. Investigation indicated that rainfall volume, rate, and duration did not exert a strong control over the characteristics of the rainout time distribution. Also, no unique association was found between distribution type and synoptic weather types.

The most outstanding feature of the time distributions of radioactive rainout revealed in the 1963 study was the strong trend for a relatively high concentration at the start of rainstorms, followed by a lesser trend for an increase in concentration at the end of storms. Further analyses indicated that the high initial concentration could be related to the partial evaporation of falling raindrops and/or contamination of the storm fringes by low-level or surface particulates. Another possibility is that the precipitation removes some of the activity in the air between the cloud base and the surface, and that this air is more highly contaminated at the leading edge of the storm where it has not been cleansed as much by rainfall as it is farther in the storm system.

The areal variability and point representativeness studies conducted with 1962 data were continued with the 1963 networks, and resulted in verification of the 1962 conclusions. Similarly, comparison of area patterns supported the 1962 conclusions.

Extensive case studies were conducted for seven storms during 1963 in which detailed data on the time and space distributions of radioactive rainout were available (Research Report 1). These studies were undertaken to combine the information from the rainwater samplers, raingages, and radar with synoptic weather data in search of greater knowledge of the radioactive rainout processes and the relationship of the rainout to various storm characteristics.

Analyses were made to evaluate the effects of developmental stage of convective cells and location of cells with respect to sampling stations on the concentration of radioactive rainout. Results indicated that these two factors may be partial causes of the large spatial and temporal variability in rainout concentrations observed in convective rainstorms, but other major causes must be involved also.

Evidence was found that the initially high concentration of radioactivity at the forward edge of rainstorms was related to evaporation of falling raindrops in some cases. However, contrary evidence was also found, and the general conclusion was that evaporation of falling raindrops is only one of several contributors to the initially high concentrations observed in Illinois storms.

Most of the samples in the seven storms were obtained from clouds which did not penetrate the tropopause level. Wide variance in time and space was found between the rainout concentrations from these tropospheric storm clouds, and, consequently, the great variability observed in Illinois storms cannot be attributed to stratospheric penetration of clouds. Furthermore, very heavy concentrations were found in some cases from clouds which definitely did not reach close to the tropopause level during their approach and passage of sampling stations. Indirect evidence was found of a stratospheric extrusion of radioactive debris in several storms.

A trend was found for the ratio of Ce^{144}/Sr^{90}, a debris age indicator, to be appreciably lower at the end of storms than at or near the beginning of rainfall. This implies a tendency for older debris in the latter part of rainstorms. The cuase of this trend has not been ascertained. In four of the seven storms, a trend was found for the ratio of Ce^{144}/Sr^{90} to decrease with increasing rainfall; that is, a tendency existed for older debris to occur in the more intense rainshowers or thunderstorms in a convective storm system. In the other three storms, no significant relationship was indicated between storm rainfall volume and Ce^{144}/Sr^{90}.

A trend was found for an inverse relationship between the concentration of radioactive rainout and storm rainfall volume. However, the relationship was sometimes weak, and pattern comparisons showed that reversal from an inverse to direct relationship may-take place within relatively short distances in the same storm system. From the study of these seven storms and other storms in the 1962-1964 period, it was found through correlation analyses that rainfall volume, although it frequently explains 50 percent or less of the variance in rainout concentration among stations in a storm, is superior to rainfall rate, rainfall duration, cloud heights, jet stream proximity, and tropopause level as a predictor of rainout concentration and deposition in convective storms.

In the 7-storm study, maxima in rainfall rate profiles through convective storms were frequently associated with minima in the rainout concentration profiles, but in a minority of cases concentration maxima were associated with rainfall rate maxima. This finding is in agreement with statistical analyses of a large number of 1963-1964 storms (Third Progress Report). The above reversal in profile relationships may occur within the same storm system, and contributes to the reversal from direct to inverse relationships between rainfall volume and rainout concentration observed in pattern comparisons in storms.

More detailed discussions of various studies made with 1963 data are included in the Second Progress Report and Research Report 1,

1964 PROGRAM

As described in the Third Progress Report, the Illinois research continued in 1964 through granting of a second extension to the original contract. In addition to continuation of the surface rainwater sampling, aircraft sampling was initiated with a sampler designed at the State Water Survey. Aircraft sampling is necessary in order to evaluate the changes that may occur in radioactivity concentrations between cloud level and ground, and, consequently, to define more accurately the various factors influencing the rainout process. Limited sampling in the summer of 1964 indicated the sampling technique was satisfactory, and plans were then made for more comprehensive sampling in 1965.

The types of studies made in 1962 and 1963 were continued in 1964 through use of time samplers and total storm samplers installed within a network of 49 recording gages in an area of 400 square miles. The final conclusion of the 3-year study of areal patterns of rainout and rainfall is that an inverse relation occurs most frequently between rainout concentration and storm rainfall, whereas a direct relation usually occurs between rainout deposition and storm rainfall. The relationship between rainout deposition and storm rainfall is stronger and more consistent than the relation between rainout concentration and rainfall. A reversal from an indirect to a direct relation between rainout concentration and storm rainfall is not uncommon, and this reversal occurs within storms as well as between storms. Reversal from a direct to inverse relation between patterns of rainout deposition and storm rainfall occurs also, but the reversal occurs far less frequently than with concentration patterns.

Earlier conclusions with respect to areal variability, time variability, and time distribution characteristics in convective storms noted in 1962-1963 studies were further verified by the 1964 data.

1965 PROGRAM

As indicated earlier, one of the more important characteristics of the radioactive rainout revealed by the 1963-1964 studies is the frequent occurrence of a relatively high concentration of radioactivity at the leading edge of storms. Knowledge of the mechanisms responsible for this phenomenon is important both for the prediction of the rainout of radioactive debris and for a better understanding of the microphysics of clouds. Another question not answered by previous studies is the extent to which the concentration of radioactivity in rainfall is related to the existing level of radioactivity in the surface and low-level air; that is, does the storm rainout concentration correlate strongly, weakly, or not at all with the ambient air?

In an effort to enhance existing knowledge on the above problems, a field program was designed to make simultaneous surface and airborne measurements of the radioactivity in precipitation and air. Instrumentation was accomplished and sampling carried out in spring 1965, as planned.

Instrumentation

A network of five instrument shelters was installed in the East Central Illinois Raingage Network 30 miles west of Champaign, Illinois, as shown in figure 1. Another was located at the State Water Survey Meteorology Laboratory, 20 miles southeast of the 5-station network. Each station was equipped with air and water sampling devices, a wind speed and direction set, and a recording raingage. A standard Weather Bureau shelter was located at the central station. Figure 2 shows a schematic diagram of a sampling station. Air enters through a 6-foot long, 4-inch duct; passes through an 8- x 10-inch filter; and is exhausted through a Gelman Hurricane blower out the side of the shelter. Air flow is monitored with a pressure-drop orifice gage at the exhaust. The air filters are changed manually, but the duration of the sample is controlled by a timer. The time and duration of the sample is recorded on a strip chart.

The 1965 automatic rainwater sampler, discussed in the Third Progress Report, is shown also in figure 2. During precipitation, runoff from a portion of the roof is collected by a gutter and drained into a large tipping bucket on top of the collector. When one side of the bucket is full, it tips and allows the water to run into the first bottle in the top rack and advances the lower circular rack of bottles. When the bucket fills and tips again, the water flows to the lower rack of bottles while the top rack is advanced. Rotation of the bottle racks is actuated by a relay on the tipping bucket and pulled by a spring.

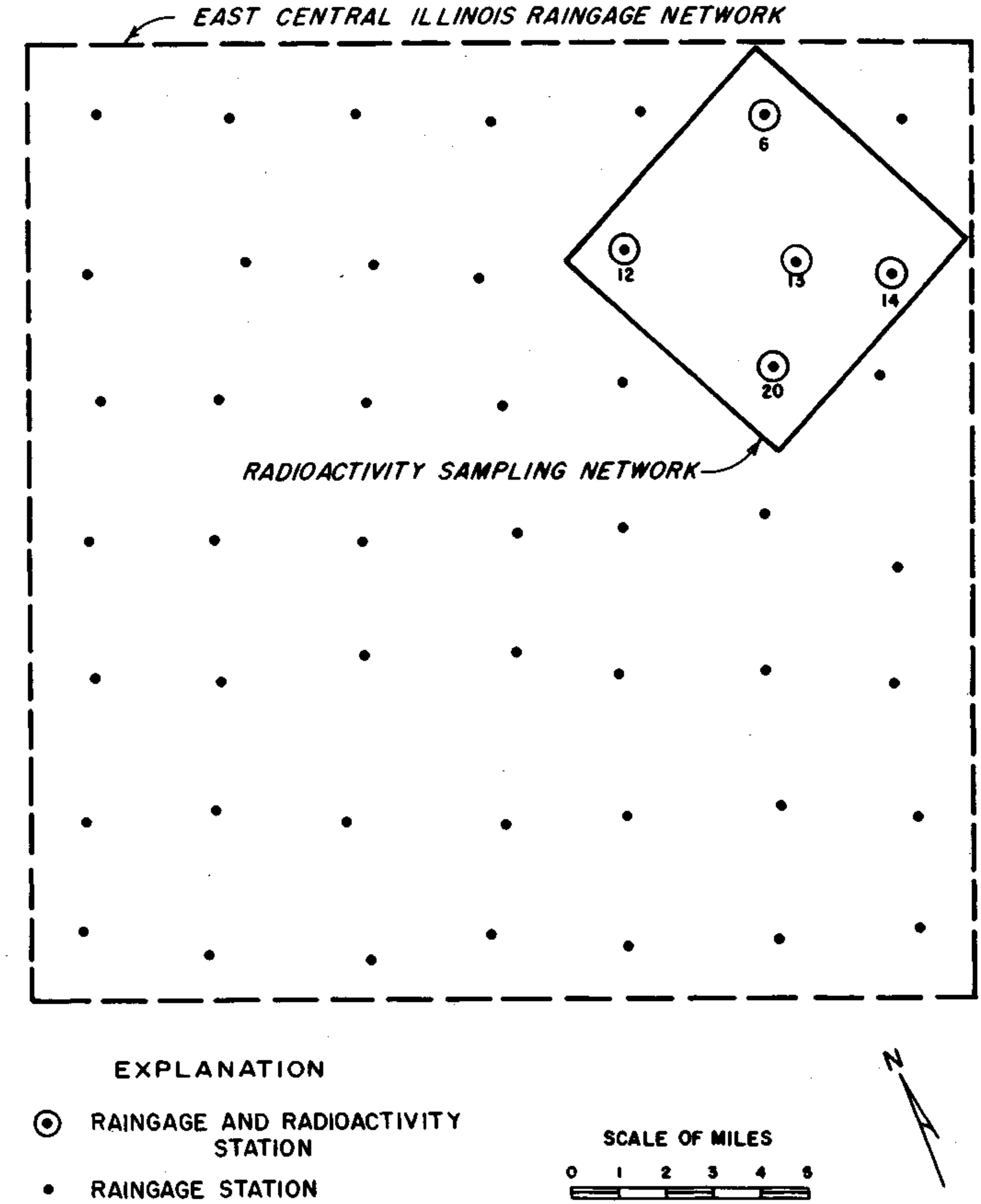

Fig. 1 1965 SAMPLING NETWORK

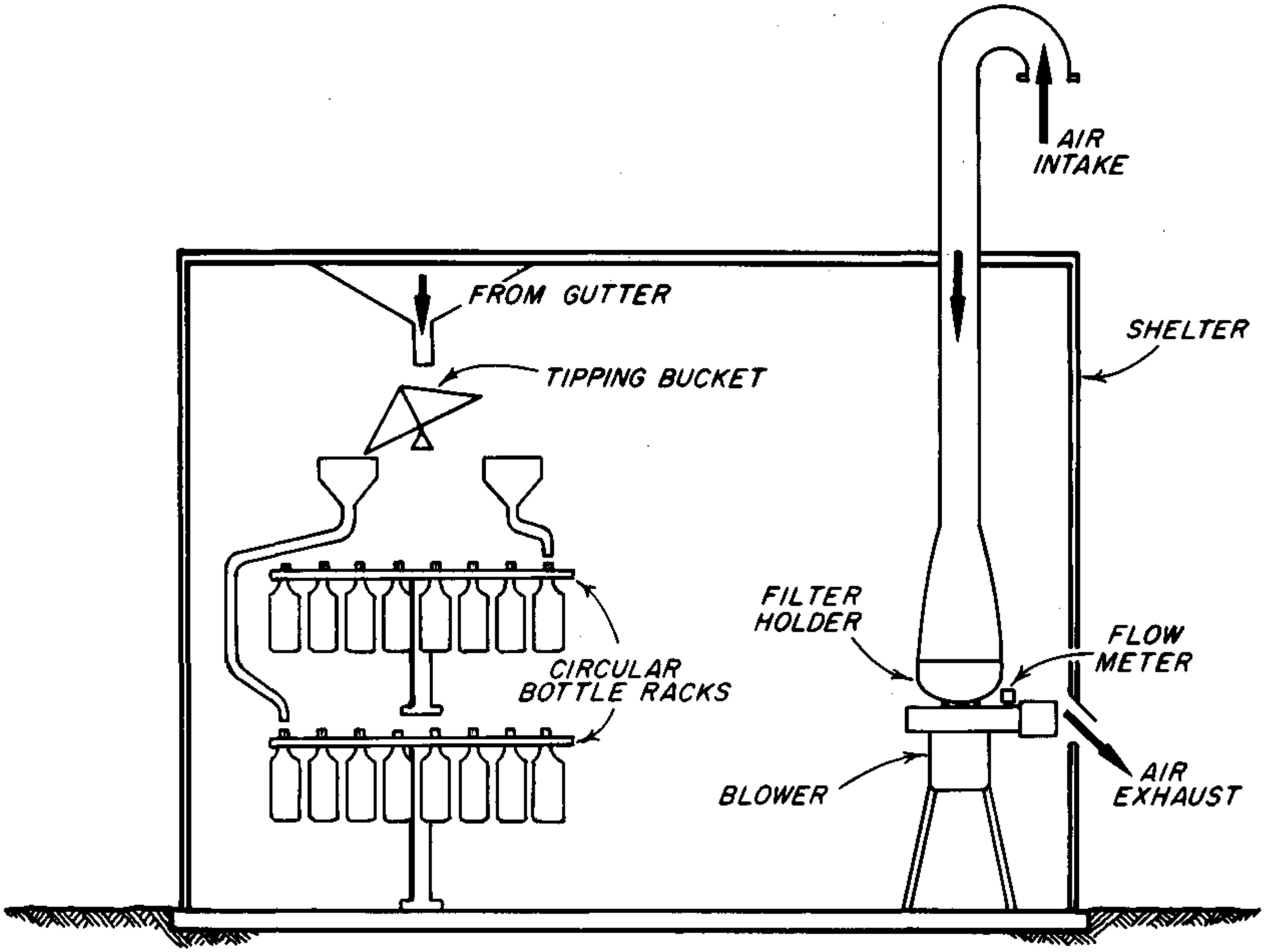

Fig. 2 SCHEMATIC DIAGRAM OF A SURFACE AIR AND PRECIPITATION SAMPLING STATIONS

The aircraft instrumentation was installed in a C-45-H twin-engine Beechcraft. New radio equipment was installed in this aircraft to permit instrument flying under adverse conditions. The precipitation collector, described in detail in the Third Progress Report, consists of a 12-inch diameter tube extending ahead of the aircraft nose. Both air and water enter the tube where the water is then removed by centrifugal force. The air-borne air sampling installation is shown in figure 3. Air enters the system through a 1.4-inch nozzle and a 2.5-inch diameter tube. It passes through an 8- x 10-inch filter and out the Gelman air blower, as in the surface samplers. The air intake nozzle size is matched to the sampler air flow and aircraft air speed so that the air flow at the nozzle approximates isokinetic sampling.

Radar coverage of the radioactivity sampling network was accomplished with a CPS-9, 3-cm PPI radar; an M-33, 10-cm, PPI radar; and, occasionally, with a TPS-10, 10-cm, RHI radar. The radars operated through several gain reductions, and the scope presentation was recorded on 35-mm film.

The sampling network was operational from April 1 to June 30. Crews were sent to operate the sampling stations when a significant precipitation was forecast during the daylight hours. Before sampling began, the air intake duct as well as the roof and gutter of each station were thoroughly scrubbed with water to remove contamination. Whenever possible, air filter samples were taken from an hour or more before the precipitation began to an hour or so after it stopped. Air was sampled at a rate of approximately 65 cfm for

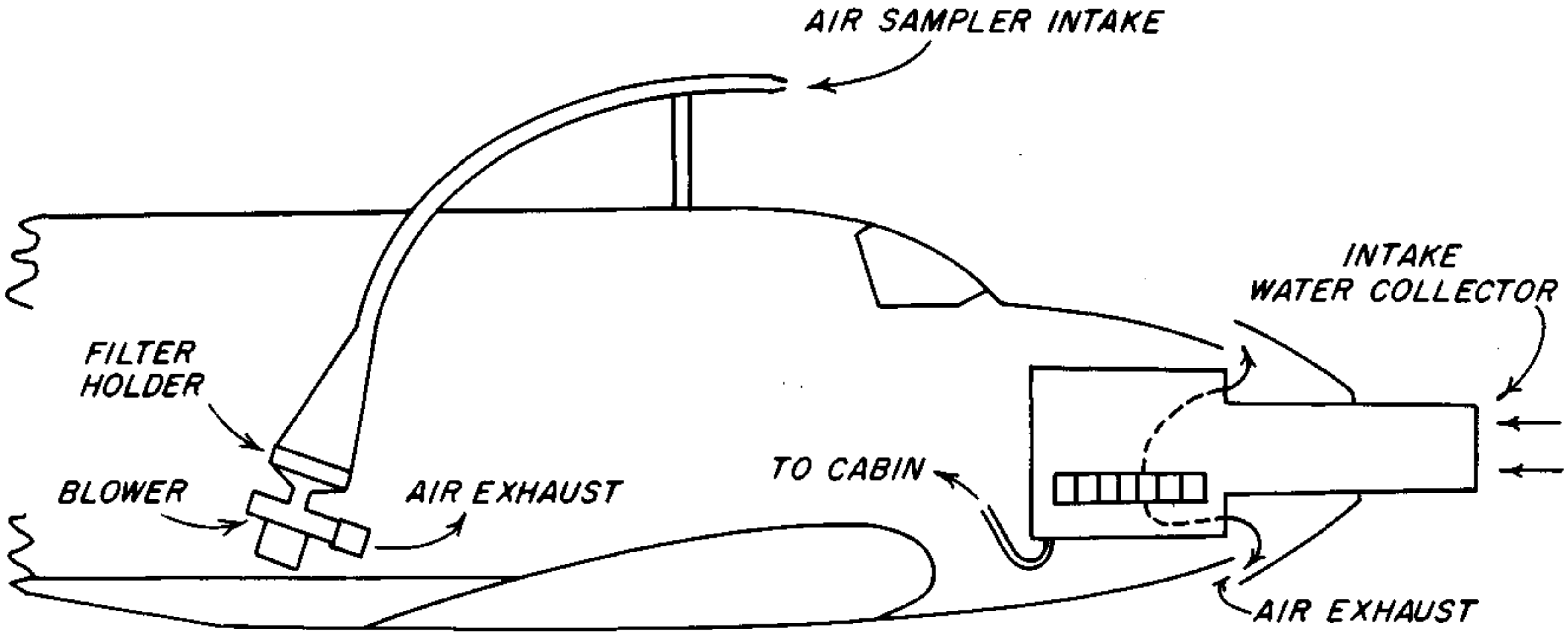

Fig. 3 SCHEMATIC DIAGRAM OF AIRPLANE PRECIPITATION AND AIR SAMPLING SYSTEMS

a duration of 10-20 minutes per filter, depending upon the amount of activity present in the air. Filters were changed simultaneously over the whole network.

The Struer filters used are reported to have a collection efficiency of 100 percent for airborne fission products at the flow rate used, as reported by Lockhart (1964). After a filter was exposed to the airflow, it was individually wrapped in a polyethylene bag and sent to TRACERLAB, Inc., for gross beta analysis.

Separate water samples of 800 ml were taken in polyethylene bottles with approximately every 0.01 inch of rain. The water samples were subjected to a gross beta analysis at Water Survey laboratory facilities.

The aircraft was dispatched to take water or air samples over the network whenever the network was in operation, and, before June 15, whenever a pilot was available for charter. Unfortunately, pilots were not always available until after June 15 when a full-time pilot was added to the staff. Water collection flights were limited to non-thunderstorm precipitation.

Observations

During the sampling season, 970 air filters were taken. Of these, 583 were obtained at the surface on days with precipitation, whereas 322 were taken on days of no rain. The remaining 65 samples were made by the aircraft; 15 were made on days of precipitation. A total of 936 water samples was collected, with 911 taken at the surface and 25 taken aloft with the aircraft. Fourteen storms were sampled on eight different days.

A complete analysis and comparison of the data with the accompanying weather has not yet been performed, but a preliminary inspection has revealed several interesting points. No evidence has been found of a consistent increase in the radioactivity of the air immediately preceding the commencement of precipitation, even though the first sample or samples of rain usually have an activity considerably higher than those immediately following. Prom 24 station time distributions on the eight storm days sampled, nine cases were found in which the surface air concentration was increasing prior to the start of rain. There were also nine cases of decreasing air concentration with the approach of rainstorms and six cases of no appreciable change in the time distributions. Cases were included in this analysis only if one or more air samples were collected prior to the start of rain. Furthermore, analysis was restricted to the beginning of the first shower on a given storm day. This finding indicates that the capture of radioactive particulates from entrainment of surface and/or low-level contaminants at the leading edge of a storm is not a necessary condition for the production of the initially high rainwater concentrations.

The data also show that often there is not a direct correlation between the radioactive content of the surface air and the accompanying precipitation. Some showers are concurrent with relatively clean air, indicating that perhaps the associated air was cleansed by the rain prior to arrival at the sampling network. Others occur with above average radioactive concentration. Still other showers show no apparent relationship between the air activity, the rain activity, and the amount or intensity of rainfall.

Figure 4 is an example of how the beta radioactivity of the air varies with time at an individual station and from station to station. There was no precipitation on the network during this sampling period.

Figure 5 is a plot of the air and precipitation beta radioactivity at station 12 on May 26. On this date both the air and water radioactivity at all of the stations showed the presence of unusually high radioactivity following the second rain. The radioactivity of the first shower and accompanying air samples are of the approximate magnitude normally measured throughout the spring and summer. During the second rain, however, the air concentrations began to increase and reached a value of 15.3 pc/m^3, one of the highest concentrations measured this season. In the third rainfall, the radioactivity of the water sample also became unusually high. It is believed that high activities were the result of the Chinese bomb test 12 days earlier. Later analysis of the rainwater showed a considerable decay in radioactivity indicating that much of the debris was indeed new debris from the recent test. A study of the synoptic, radar, and micrometeorological data for this storm, and the other storms, will be included in a future research report.

As mentioned before, only 25 water samples were collected aloft on three different days. This number was limited by several

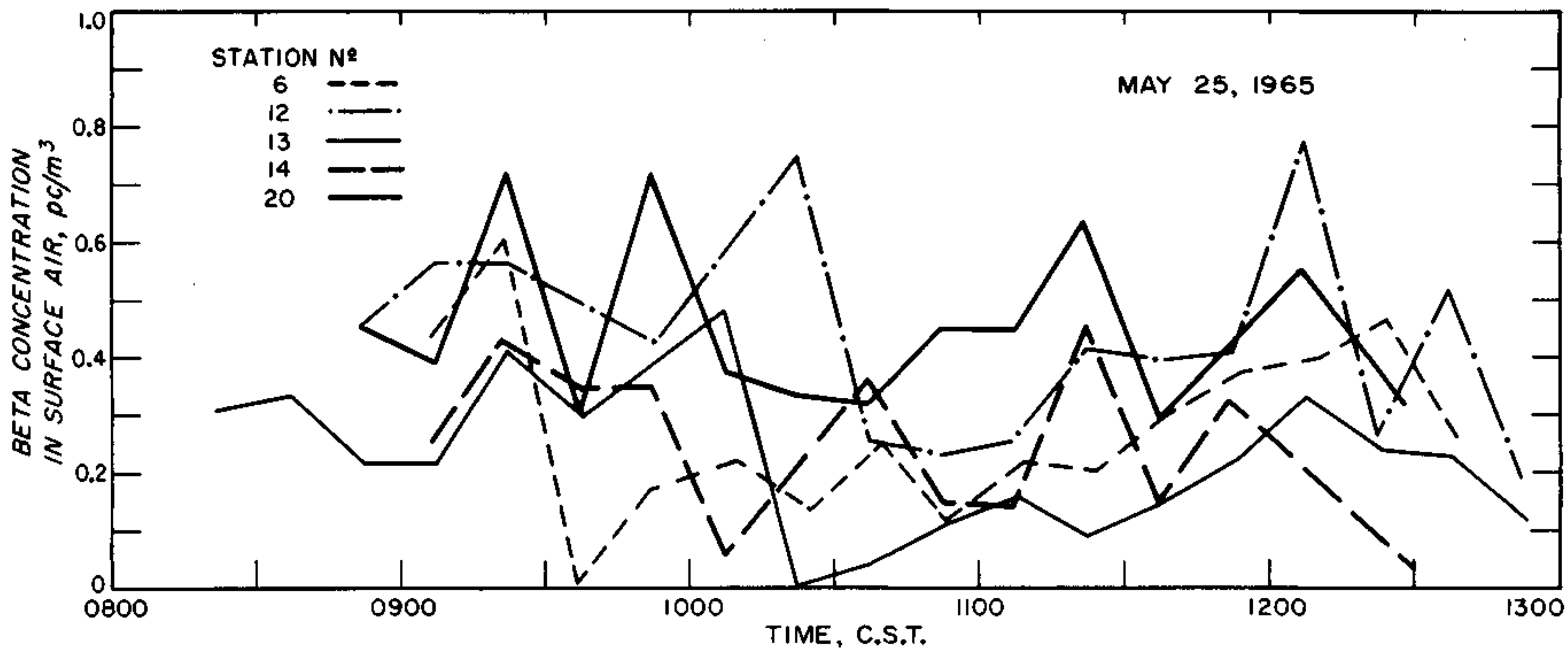

Fig. 4 CONCENTRATION OF BETA RADIOACTIVITY IN THE AIR AT THE FIVE NETWORK SAMPLERS ON MAY 25, 1965

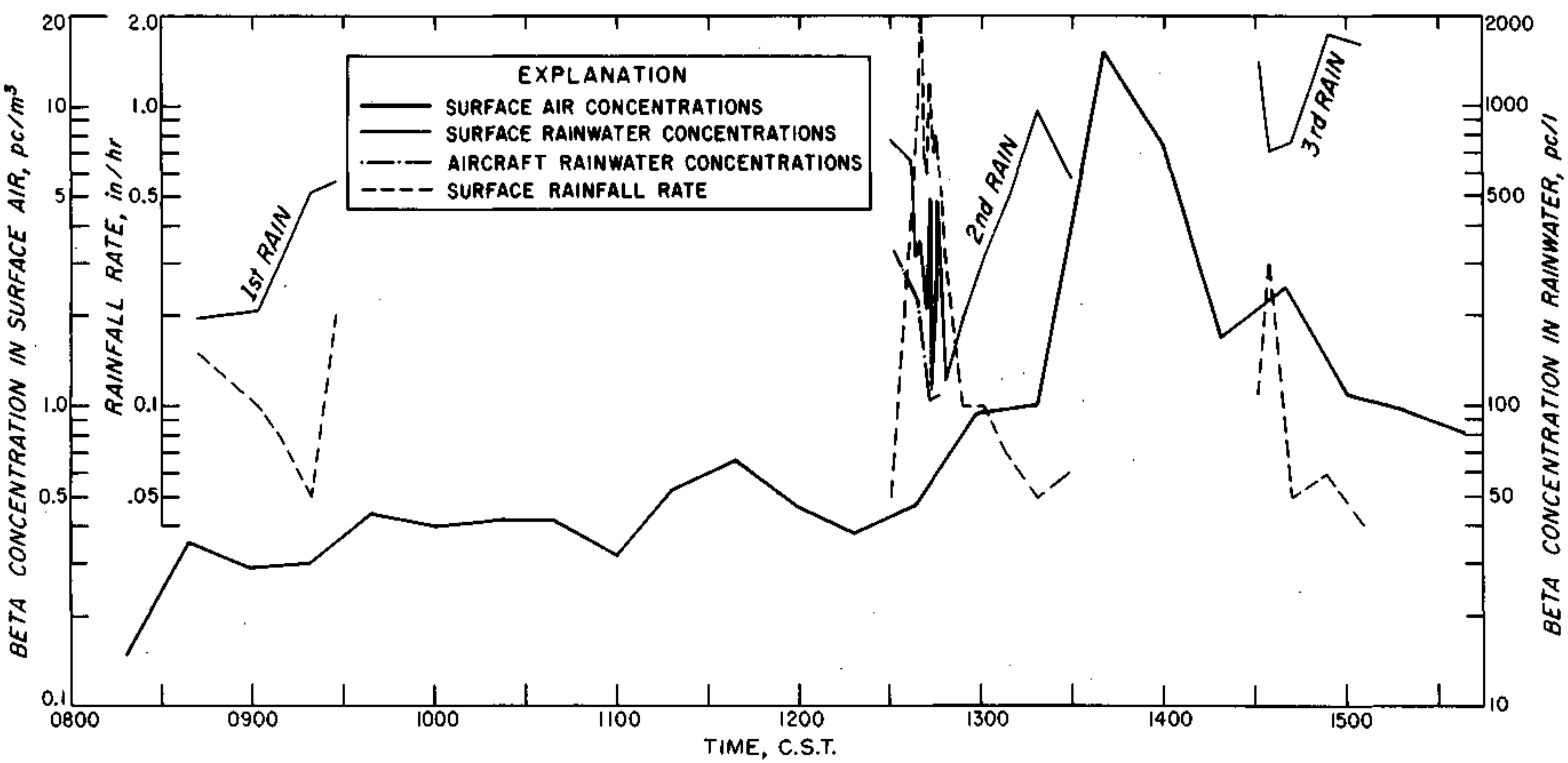

Fig. 5 DISTRIBUTION OF RADIOACTIVITY CONCENTRATIONS AND RAINFALL RATE AT STATION 12 ON MAY 26 , 1965

factors. A pilot was not always available on short notice prior to June 15. Plights were not made in thunderstorm precipitation because visual contact with the surface was necessary for the aircraft to circle directly above an individual collector,, Also, a longer period of time is necessary to collect an airborne sample than a surface sample.

Several of the airborne water samples were taken 2200 feet above sampling station 12 on May 26 during the second rain, as shown in figure 5. The airborne precipitation beta activity was compared with measurements taken at the surface after a time correction was applied to account for the time of fall. In all cases, the beta activity in the water collected aloft was considerably less than in the surface water. The increase in activity was greater than could be accounted for by assuming a reasonable rate of evaporation of the drops. The possibility therefore exists, despite theoretical calculations to the contrary (Greenfield, 1957), that some radioactive debris was collected between the level at which the airborne sample was taken and the surface. A more detailed discussion of this data will be included in the next report.

The use of 1-liter rainwater samples in 1965 compared with 3-liter to 4-liter samples in 1963-1964 provided an opportunity to define further the location of the leading edge radioactivity maximum with respect to the rest of the rainstorm. Combining all 57 station samples of separate showers in 1965 storms showed that the relatively high concentrations were contained most frequently in the first two samples, or in the first 0.02 inch of rainfall.

That is, the rate of decrease in rainout concentration tended to slow rapidly after the first 0.02 inch of rain was recorded.

Other statistical findings presented in the Second and Third Progress Reports with respect to rainout distribution types and to the relationship between the time distribution of gross beta concentration and rainfall rate distributions in storms were supported by the 1965 data. Thus, the Type A distribution continued to be the most frequent distribution associated with radioactive rainout, and the beta minimum in the Type A distribution was usually associated with a peak in the rainfall rate distribution through the storm. Similarly, the beta maximum in the Type C distribution usually occurred with a peak in the rain rate distribution, and the secondary peak in beta concentration in the Type D distribution was frequently associated with a rainfall rate peak, as shown in the Third Progress Report.

Summary

Surface air and precipitation samples were collected during 14 rains and analyzed for gross beta radioactivity. Preliminary inspection of the data indicates relatively weak correlation between radioactivity concentrations in surface air and precipitation. An exception occurred on May 26 when high air and water concentrations occurred close together in the second and third showers.

No consistent increase in radioactivity concentrations has been found immediately preceding the onset of rainstorms to correspond with the initially high concentrations of rainwater radioactivity usually found at the leading edge of storms.

Airborne measurements of precipitation radioactivity during one storm indicated an increase in gross beta activity between flight altitude and the surface. The increase in concentration is more than can be accounted for by evaporation,,

However, further analysis of the radioactivity data in conjunction with associated meteorological conditions must be made before firm conclusions can be established with regard to relations between surface air and rainwater concentrations and between concentrations at cloud base and the ground.

CONTRACT PUBLICATIONS

As listed below, three progress reports and two research reports have been prepared and distributed under this contract. Two technical papers have been published in national journals. Two other papers have been submitted to technical journals for publication.

Huff, F„ A. 1963. Study of Rainout of Radioactivity in Illinois. First Progress Report to AEC under Contract AT(ll-l)-1199.

Huff, F. A. 1964. Study of Rainout of Radioactivity in Illinois. Second Progress Report to AEC under Contract AT(ll-l)-1199.

Huff, F. A. and G. E. Stout. 1964. Distribution of Radioactive Rainout in Convectlve Rainfall. Journal of Applied Meteorology, Vol. 3, No. 6.

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