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

STUDY OF RAINOUT OF RADIOACTIVITY IN ILLINOIS

Sixth Progress Report
Contract Number AT(ll-l)-1199
November 1967

Sponsored by

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

Prepared by

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INTRODUCTION

Research during 1967 has been concentrated on four studies relating to radioactive rainout in rainfall. In one study, an investigation is being made of the relationship between the concentration and deposition of radioactivity in rainwater and air concentrations of radioactivity, through the use of gross beta measurements. Major objectives of this study are to determine the strength of the air-rainwater relationship and to ascertain whether air samples can be used to estimate radioactive rainout with reasonable accuracy over areas of a few square kilometers to several thousand square kilometers.

Aircraft sampling of the vertical distribution of tritium, initiated last year, was continued in 1967 and expanded to include spatial sampling and measurement of radon concentrations. Also, analyses of the 1966 data are being carried out. Results of this study should substantially increase knowledge on the vertical variation of tritium, provide some information on differences in tritium concentration between several types of synoptic weather, and contribute to better understanding of the atmospheric processes controlling rainout and fallout.

Work is progressing on several case studies involving analyses of air trajectories at levels between the surface and 200 mb. These studies are being made for selected 1963 storms in which relatively heavy concentrations of rainwater radioactivity were measured and in which spatial and time variability were large on the sampling networks. Basically, it is anticipated that the air trajectory analyses, supplemented by radar data and precipitation network data, will yield an explanation of the causes of the rainout characteristics observed in these storms and further our knowledge of the processes controlling radioactive rainout.

A pilot study was undertaken to determine the relationship between the time distributions of radioactivity and selected chemical constituents in rainwater, through use of water samples stored from network operations in previous years. The purpose of this study is to evaluate the feasibility of using concurrent measurements of radioactivity and chemical elements normally present in rainwater to acquire additional knowledge of the atmospheric processes controlling rainout and to aid in verification of certain hypotheses and observations resulting from earlier studies.

ACKNOWLEDGEMENTS

The research upon which this report is based was carried out under the general direction of G. E. Stout, Head, Atmospheric Sciences Section, Illinois State Water Survey. Professor John Pearson, University of Illinois, aided in the planning of tritium and radon sampling flights. John Wilson assisted in the aircraft sampling program. Analyses of the chemical constituents in rainwater were made possible through the cooperation and assistance of the Survey's Chemistry Section. Other staff members aided in the collection and analyses of data used in the various studies. Appreciation is expressed to Dr. Gote Ostlund of the University of Miami for the loan of a case of molecular sieves and the bake-out and tritium analysis of several water vapor samples.

1966 TRITIUM SAMPLING

Introduction

The use of tritium as a tracer for the movement of water and water vapor has enjoyed considerable success since the Castle tests in 1954. Libby (1959) used bomb-produced tritium to estimate the northern hemispheric tritium storage time and deposition rates, as well as the ground water balance of the upper Mississippi Valley. Eriksson (1958) has also studied ground water storage with the use of tritium. More, recently Smith (1966) pointed out the use of tritium in studies of atmospheric moisture transport. In addition to bomb-produced tritium, reactor-produced tritium promises to be a useful tool for the tracing of water vapor and ground water.

The increased use of tritium as a tracer has resulted in a need for further information on the present background of atmospheric tritium, especially on its vertical distribution and its variation with time and synoptic weather types. As a result of these considerations a tritium sampling program, in cooperation with Dr. Eriksson of the International Meteorological Institute in. Sweden, was initiated in the winter of 1966 in central Illinois. Periodic aircraft soundings of atmospheric water vapor were made from the surface to 4.9 kilometers (km) MSL. Usually 4 samples were taken per sounding with each sample integrated through a layer of approximately 1200 meters.

Data and Results

A list of the sounding flights and the samples taken during 1966 and 1967 is given in Table 1. On two of the earlier flights no samples were taken above approximately 4 km because of a pump failure at that altitude. It was finally determined that the failure of the carbon pump blades was the result of pumping very dry air. Special pump blades were obtained for use with dry air and no further difficulty was encountered.

During most of the soundings the concentration of tritium was constant or increasing with altitude. An example of this type of sounding taken on June 16, 1966 is shown in Figure 1. The sampling was accomplished during the. forenoon in cP air behind a cold front located in the extreme southern portion of Illinois. The front had passed through the sampling region approximately 18 hours earlier. Northwesterly winds at the surface changed to west-northwest, 40-50 knots at 500 millibars. During the sampling period, the sky was nearly overcast with stratocumulus and scattered cirrus present above 8 km. The sampling was accomplished in cloud-free air.

The temperature and humidity sounding accompanying the tritium measurements was from the USWB rawindsonde at Peoria, Illinois taken at 0600 CST. The aircraft sounding, which began 3.3 hours later, was taken 75 miles ESE of Peoria and lasted from 0915 to 1025. Temperature measurements taken on the aircraft confirm the persistence of the thermal inversion between 3.2 and 3.9 km.

The solid vertical lines TU, in Figure 1 represent the average tritium concentration within the indicated layer, A constant rate of ascent and sampling was maintained. The tritium concentration increased only slightly with increasing altitude to the bottom of the subsidence inversion where a greater increase occurred. This increase was the result of the subsidence of dry air aloft with a greater tritium concentration.

The dashed vertical lines in Figure 1 represent the tritium mixing ratio (TMR) through the layer. The TMR is the product of the average water vapor mixing ratio in grams per kilogram from the Peoria sounding, and the tritium concentration in tritium units for the layer. The TMR weights the tritium concentration with the amount of water vapor present and is thus proportional to the number of tritium atoms per kilogram of air. It is apparent from Figure 1 that although the tritium concentration increases with altitude, the total amount of tritium present (TMR) decreases.

Table 1. Tritium and Radon Sampling Flights

Date	Max. Alt. Sampled (meters MSL)	Number of Tritium Samples Collected	Number of Radon Samples Collected
1-17-66	1520	1	0
4-15-66	3660	4	0
4-21-66	5180	4	0
4-29-66	3960	3	0
6-8-66	4880	3	0
6-16-66	5180	4	0
6-23-66	4570	4	0
6-30-66	5180	4	0
7-11-66	4270	4	0
7-26-66	2440	4	0
7-26-66	2440	4	0
7-26-66	2440	4	0
10-6-66	5490	4	0
5-31-67	4880	4	0
7-5-67	5110	4	0
7-7-67	—	0	radon test flight
7-11-67	—	0	radon test flight
7-14-67	4850	5	3
7-17/20-67	3870	10	0
7-26-67	4270	0	4
8-3-67	2740	0	5
9-6-67	4880	4	4
9-20-67	4960	4	4

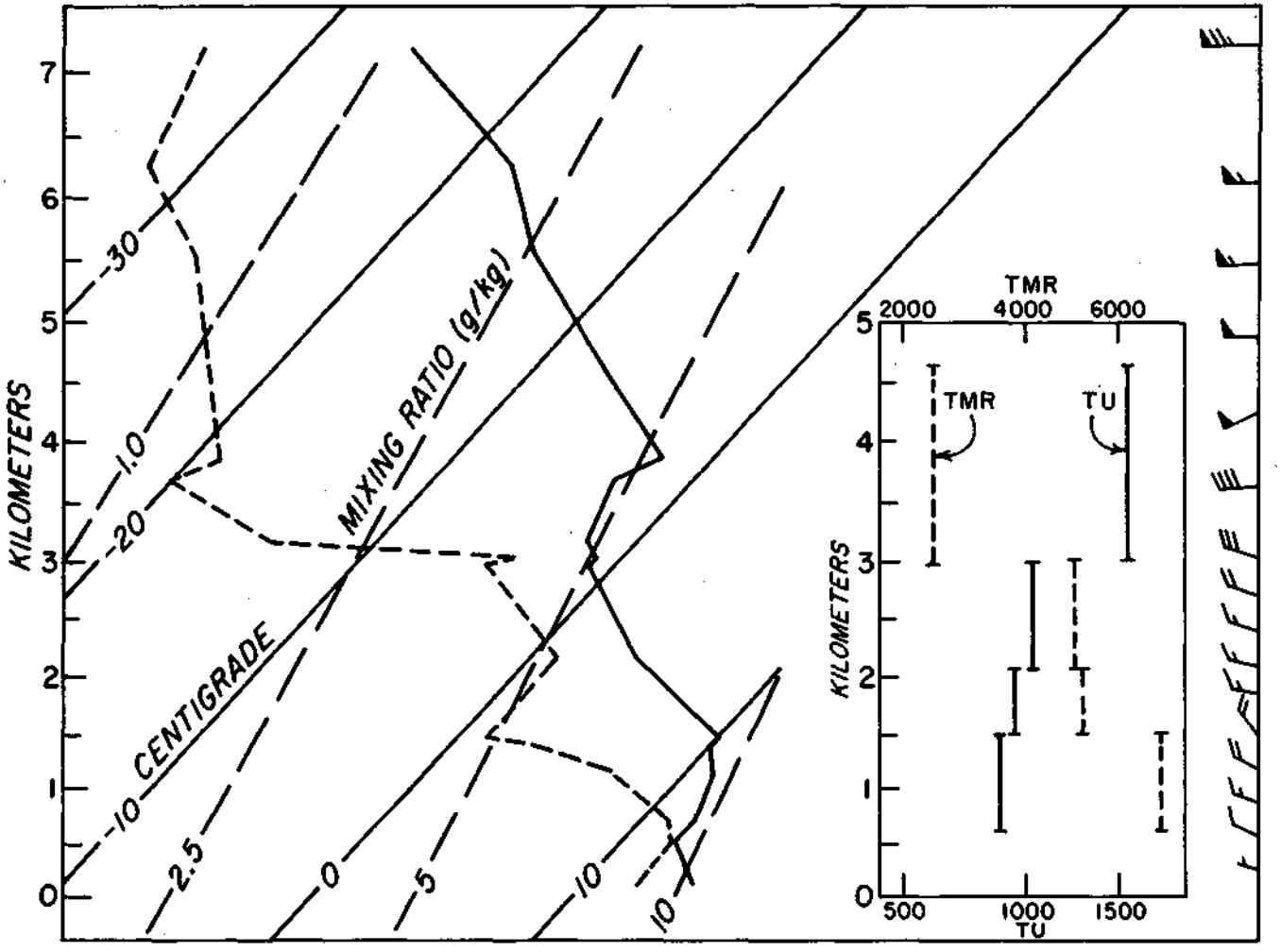

Figure 1. RAOB sounding for Peoria, Illinois, and tritium sounding for June 16, 1966

It is at first surprising to find the gradient of the TMR decreasing with increasing altitude because the primary source of tritium is the stratosphere, where it has been injected by thermonuclear detonations, and the ultimate sink is the ocean. The TMR gradient is in the direction opposite that expected from the apparent source and sink. However, the tritium measured here is associated with the hydrogen atom in the form of tritiated water, HTO. The tritium thus follows the laws of transport of water vapor, which are different from those of other gases or particulates. The normal gradient of water vapor decreased-upward and the resulting net diffusion of water vapor is therefore upward, i.e., down the gradient.

Once tritium has been transported from the stratosphere to the lower troposphere it becomes mixed with and accumulates in the large reservoir of water vapor present there. Tritium is, in time, removed from this reservoir to the surface of the earth by precipitation. The precipitation either evaporates and continues through the cycle again, seeps into the earth to add to the groundwater supply, or flows to the ocean. Very little tritium escapes from the ocean because of the low concentration of tritium in the water,

The tritium soundings were compared with the accompanying weather and air mass origin. The soundings were usually flown under fair weather conditions, but a flight on April 15, 1966 was made in cumulus congestus clouds and precipitation. The clouds were so close together that numerous penetrations could not be avoided. Cloud bases were at about 1.6 km. Three samples were taken to an altitude of 3.7 km MSL where aircraft icing forced an end to the sampling. Under these conditions there was no variation of tritium concentration with altitude to the top of the soundings and within the cloud because of the good vertical mixing.

Based upon these samples, there does not appear to be significant' correlation between the air mass type and the concentration of tritium. The vertical and time variation of the tritium concentrations are such that small differences would not necessarily be revealed but from the limited samples of the 1966 data, the mT air mass appears to have as high a tritium concentration as the cP air mass by. the time both reach the Midwest.

1967 TRITIUM AND RADON SAMPLING

The concentration of tritium found in the lower troposphere in the Caribbean by Ostlund (private communication) is usually several times lower

than that measured in the Midwest, even with southerly flow from the Gulf. Therefore, as the air moves northward there is an input of tritium from the stratosphere and from evapotranspiration. A field program was initiated to utilize this influx of tritium into the lower troposphere to make an estimate of K, the coefficient of vertical diffusivity, by means of a series of tritium soundings between the Gulf of Mexico and the Midwest.

The coefficient of vertical diffusivity (K) may be determined experimentally with the formula

$$E = -\rho K \frac{\partial (TMR)}{\partial Z}$$

where E is the vertical flux of the TMR, p the density of air, and Z the height of the level under consideration. With the aid of several soundings between the Gulf and the. Midwest, it is anticipated that E could be determined from the change in the TMR in each layer, p could be calculated from the altitude and temperature, and $\partial (TMR)/\partial Z$ obtained from the slope of the sounding. The formula could then be solved for K. Several flights were planned to make the required tritium measurements.

To increase the utility of the flights and to provide an independent means of simultaneously measuring K, a radon sampling system was also installed aboard the aircraft. The technique used to determine the radon concentration is identical to that used by Lockhart (1965). A lead shield and Gieger tube were borrowed from Mr. Charles Hosier of ESSA. Ambient air was pumped into the aircraft and passed through a 6.4-cm diameter fiberglass filter before passing into the molecular sieves and out through an orifice flow meter. Filter samples of radon daughter products were taken for a 20-minute period, followed after one minute by a 10-minute counting time on a GM tube shielded with 45 kilograms of lead. For the altitudes sampled with the aircraft the radon was assumed to be in equilibrium with its daughters and only one counting period was therefore necessary. The radon measurements were intended to be used to determine K independently with the same technique as described above for tritium.

Considerable difficulty was encountered at first with the scaler-power supply used for the radon system. Frequently, especially at higher altitudes, a series of sporadic counts was suddenly registered on the counter. The trouble was finally located by examining the equipment under operation while in an altitude simulation chamber large enough for the equipment and an engineer.

Arcing had been occurring between two components resulting from the lesser insulating qualities of the rarefied air found at higher elevation, and was easily rectified.

Several radon soundings were taken during 1967, some with simultaneous tritium soundings. The radon sounding taken on July 26 is shown in Figure 2. The measurements were taken from 0938 to 1218 CST near Champaign, Illinois. The first sample may have been taken within the exchange layer but the higher samples were definitely taken in nonturbulent air The sample from 0.4 km had a radon concentration of 139 picocuries per meter^3 (pc/m^3) and the sample from 4.3 km was 2 pc/m^3. The concentrations differed by a factor of 70. The low concentration of radon at 4.3 km was due to decay, dilution, and possible washout.

It had been proposed to make three to five flights to the Gulf of Mexico to measure the vertical distribution of tritium and radon. The meteorological situation necessary for the experiment was southerly flow from the Gulf moving northward into the Midwest. With this type of flow, air with low tritium concentration could be followed in the direction of the mean flow at 1.5 km with soundings being taken every 12 hours. This type of flow accompanies Bermuda highs which exist periodically off the southeast coast of the U. S. Usually summers in the Midwest are accompanied by a number of Bermuda highs bringing warm, moist air northward to the Midwest. However, the summer of 1967 was an unfortunate exception. From the time the field program was operational in July, there was not a satisfactory sampling situation throughout the summer and early fall. During one week, there appeared to be a possibility of favorable weather and a flight was made to the Gulf. However, the weather became unfavorable with light and variable wind with no definite northward trajectories, and the intended sampling could not be accomplished.

Several local tritium and radon soundings were made during the summer to serve as comparison data for the Gulf flights (assuming that they would be made), to test the radon equipment and to determine the relationship between the radon and tritium concentration in the vertical. The tritium samples are at present being analyzed by Isotopes Inc., Westwood, N. J. These data and the 1966 tritium data will be discussed in detail in a separate report.

In addition to the aircraft tritium samples, a number of surface water vapor and precipitation samples were collected during the summer as listed in Table 2. These were taken primarily for use as background data to serve as before and after references for the Gulf flights. The water vapor samples were

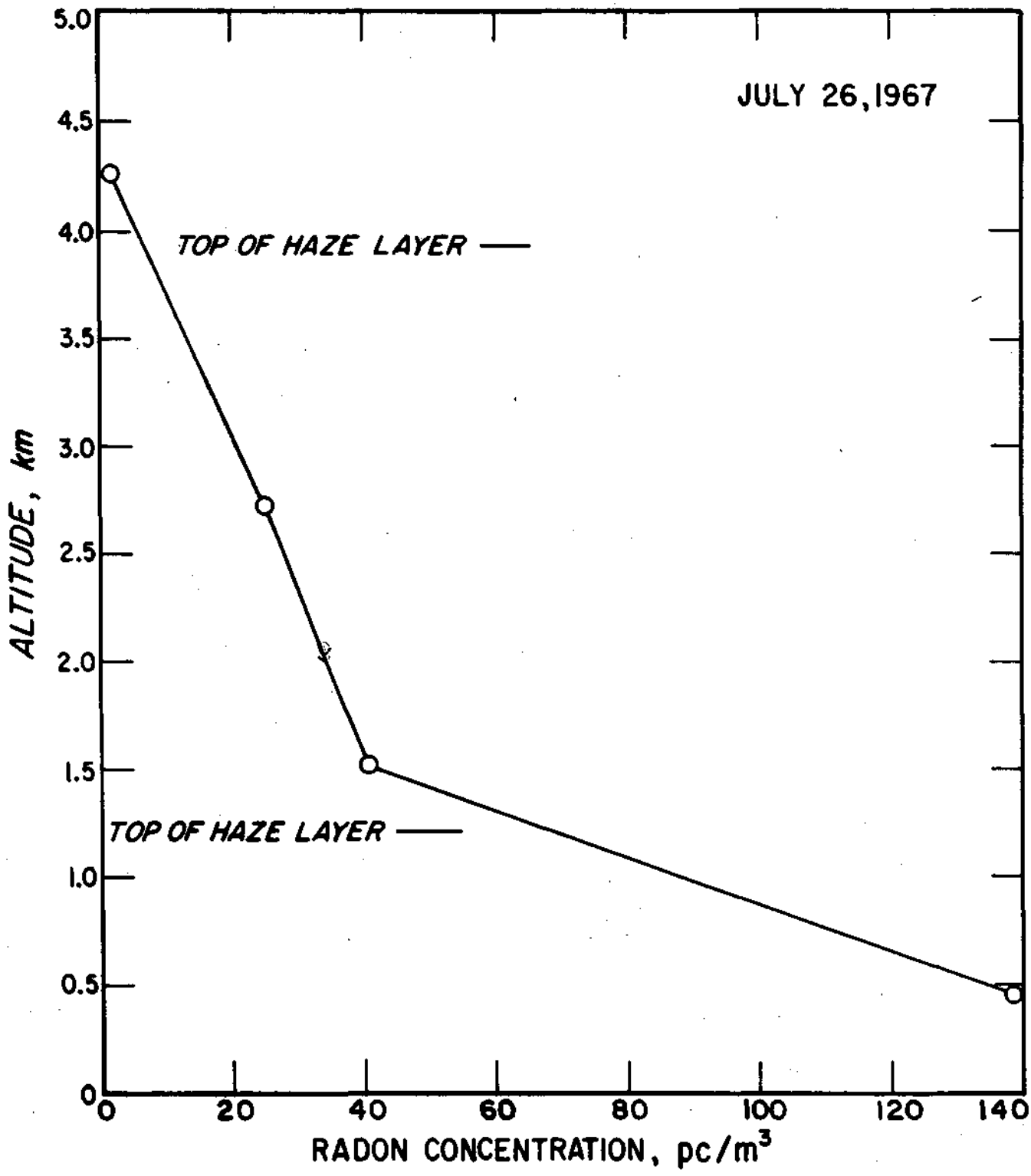

Figure 2. Radon profile for July 26, 1967

collected by pumping air slowly through a trap cooled with dry ice.; A few of these samples relating to interesting meteorological conditions will be analyzed and discussed in the next report.

Table 2. Surface Water Vapor and Precipitation Samples

Date	Vapor	Rain	Date	Vapor	Rain	Date	Vapor	Rain
6-15-67	1		7-3-67	1		7-23/24-67		1
6-22-67	1		7-10-67	1		7-24-67	1	
6-23-67	1		7-13-67	1		7-26-67	1	1
6-24-67	3		7-14-67	1		7-27-67		1
6-25-67	1		7-17-67	1		8-3-67		1
6-26-67	2		7-18-67	1	1	8-7-67	1	
6-27-67	2		7-19-67	1		8-8-67.	1	2
6-28-67	1		7-20-67	1		8-15-67		1
6-29-67	1		7-21-67	1		8-26-67		2

RELATION BETWEEN AIR AND RAINWATER BETA ACTIVITY

During the summers of 1962 through 1965 the Illinois Water Survey operated several rainwater sampling networks of various sizes and sampling densities. These networks were used to measure the areal and time distribution of the rainout of radioactivity. Several thousand water samples were collected and analyzed for gross beta radioactivity and reported on by Huff (1963, 1964, 1965a, 1965b, 1965c), Huff and Bradley (1965) and by Bradley and Feteris (1966). An analysis is currently in progress to study the correlation between the gross beta activity of precipitation samples and the ambient air. The primary purpose is to determine how well the radioactivity concentration and total deposition in precipitation over areas of various sizes are related to and can be predicted by the 24-hour air samples collected by the existing air sampling network of the U. S. Department of Public Health.

From the 4-year period, 2961 water samples were rated suitable for the analysis. The beta activity of the samples, along with other pertinent data, were punched on cards and at present are being computer processed in several ways. Maximum, mean, and minimum activities for individual stations and for the networks are being compared with the corresponding air activity to determine the correlation coefficients and regression equations. A similar study is being done with station and network total depositions.

Figure 3 is a plot of the average 24-hour beta concentration of air at Springfield, Illinois against the average beta concentration of station precipitation for each storm period. All four years of data are included in the plot. The air sample filters were changed at 0730 CST each day. The station precipitation average used was the arithmetic mean of all precipitation samples taken during the 24 hours preceding the corresponding air filter. The volumes of water collected at any particular station varied to some degree, but in this portion of the study the averages were not weighted according to the amount of water in the sample. In the deposition studies to be reported on later, the amount of water in each sample was necessarily included in the calculation.

There appears to be a slight trend for a curvilinear relationship in Figure 3 and this trend was found also in similar plots of the station maxima and minima. A regression equation is being computer calculated and the statistical significance of the curvilinear relationship will be determined. A curvilinear relationship could be explained by a particle size distribution changing with time. If the radioactive particulates reaching the troposphere in 1962 and 1963 were of greater mean diameter than those in 1964 and 1965, the larger particles would have been scavenged more efficiently by precipitation, producing a greater slope in the regression line. The significance of this relationship is currently being investigated in more detail.

Scatter plots of the data for any individual year show no relationship between the two variables. The data from 1963 through 1965 have very little overlap of the plotted points, but the 1962 data fall almost midway between 1963 and 1964. The 1963 activity is higher than the 1962 because of the continued bomb tests in summer and fall of 1962. If Figure 3 is used to estimate the average concentration at any station from a given air sample, the estimate would be within a factor of approximately 5 at the 95% confidence level. The relationship between the beta activity in the air in a portion of a cloud and the precipitation from that portion of the cloud undoubtedly would

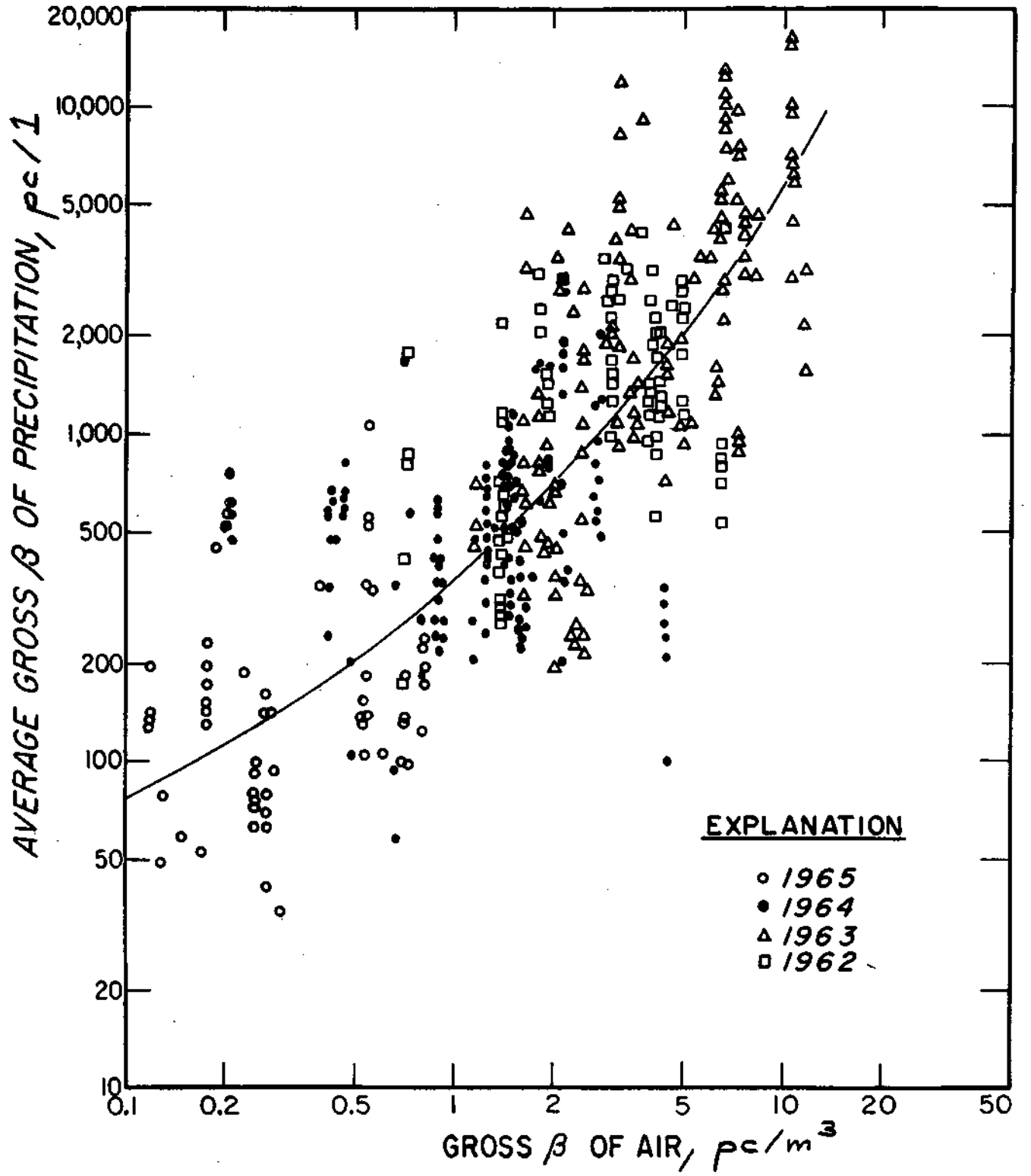

Figure 3. Relationship between radioactivity in air and rainwater for 1962-1965

have a much stronger correlation than that exhibited in Figure 3. The spatial and time separation between the air and water samples used in this study and other variables involved in the relationship preclude strong correlations between the two variables.

RELATION BETWEEN AIR MOTIONS AND RADIOACTIVE RAINOUT IN CONVECTIVE STORMS

Seven case studies of convective storms in 1963 have been presented in Research Report No. 1 by Huff (1965). Detailed data on the time and space distribution of radioactive rainout from these storms was studied in relation to the location, stage of development, and movement of storm cells, and it was concluded that most of the samples in the seven storms were obtained from cells that did not penetrate the tropopause level as defined by the U. S. Weather Bureau (1957). The purpose of the investigation presently in progress is to trace the air which moved into the storms at levels between the surface and the , coded tropopause back to its position ,24 to 48 hours previous to the rainout. This should provide a more precise answer to whether stratospheric air was involved in the rainout process. A negative answer would mean that the locally heavy concentrations observed in some places were of tropospheric origin and caused by a particularly favorable combination of exposure of the precipitation embryo to radioactive debris and microphysical processes (condensation or capture) as discussed by Hicks (1966).

Case studies are being made of the air trajectories at levels between the surface and 200 mb with respect to the radar echo agglomerates which were associated with the rainfall over the rainwater sampling network. This is a synoptic-scale study dealing with rather large masses of air, since the spacing of the radiosonde and upper wind data does not allow analyses at a scale less than 50 miles in the horizontal and a few thousand feet in the, vertical (Feteris, 1965). The synoptic-scale computations should reveal the approximate locations at which air from different sources met during the life history of the storm cell agglomerations and whether this can be related to the age of the debris (Ce^{144}/Sr^{90} ratio) and other characteristics of the radioactivity found in the rainwater samples. The 1963 storms of June 10, July 1 and July 13 were considered most suitable for the initial study.

The upper air data are first computer-processed by Danielsen's isentropic analysis program (Duquet, 1964) to obtain potential temperatures, mixing ratios, relative humidities and winds for standard levels and special points. The program also provides pressures, temperatures, relative humidities, and mixing ratios for isentropic levels at 5°K intervals between 295°K and 400°K. Since the input is on cards, the computer program first had to be modified to account for a different input format, missing data, etc. From the computer output, maps of streamfunction, pressure, and relative humidity are hand-analyzed for the levels of 300, 305, 310, 315, 320, 325, 330 and 335°K at 00.00 GMT and 12.00 GMT.

The pressure and streamfunction maps are subsequently digitized from a JNWP grid overlay and the values at the appropriate grid points are fed into a computer program that computer-balances winds, isentropic vorticities, average vertical displacements, and quasi-horizontal isentropic trajectories for 12-hour intervals, starting at each grid point (Danielsen, 1966). The distribution of potential vorticity'(the product of isentropic vorticity and stability) then provides information about the presence of air of stratospheric origin at different levels in the troposphere.

To date, all maps for June 9 and 10, 1963 have been analyzed for pressure, streamfunction, and relative humidity up to the level of 335°K. These maps have been digitized and the computer output received. Both isentropic programs work satisfactorily.

As analysis progresses, isentropic trajectories will be evaluated for all levels up to 335°K; these should indicate the sources of the air that entered the storm of June 10 over central Illinois. Similar analyses are planned for the storms of July 1 and July 13. The results of these analyses will provide an answer to the question whether there was direct quasi-horizontal transport of air from the stratosphere to mid-tropospheric levels through stratospheric extrusions (Danielsen, 1964). At the lower levels, smaller-scale convective circulations could then have mixed it with surface air. If the answer is negative, a search must be made for smaller-scale processes that cause rapid downward diffusion of radioactive matter that has entered the, troposphere at an earlier time in a different region.

TIME DISTRIBUTIONS OF RADIOACTIVITY AND CHEMICAL CONSTITUENTS IN RAINFALL

Introduction

A study has been undertaken recently to determine the relationship between the time distributions of radioactivity and several metallic aerosols in the rainwater from convective storms. Several investigators have presented experimental results which indicate that the lower troposphere is a major source of radioactivity input to convective storms, as opposed to direct input from the stratospheric reservoir of radioactive debris through extrusion processes or from stratospheric penetration by convective clouds. Also, previous studies have shown a strong trend for a relatively high concentration of radioactivity at the forward edge of storms, and the causes of this anomaly have been debated. The present study has been undertaken in an effort to obtain further information on the foregoing subjects. Since this study is, described in considerable detail in the forthcoming Proceedings of the Chalk River AEC Micrometeorological Meeting of September 11-14,. 1957, it will only be discussed briefly here.

In the time distribution comparisons, analyses have been made of the concentration of gross beta, Na, K, Ca, and Mg in the rainwater from convective storms. Soil material and man-made atmospheric pollution are the primary sources of the four metallic ions in continental regions such as the midwestern U. S. A. Therefore, similar time distributions would indicate similar source regions for the radioactive debris and chemical aerosols, and, thereby, imply a low-level source of direct input of radioactivity in the rainstorm. Conversely, consistently poor correlation between the two types of time distributions would be indirect evidence that the lower troposphere is not the major contributor to radioactive rainout in convective storms.

Data and Analysis

Time distribution samples were selected for 6 storms from available data collected with the network of automatic rainwater samplers operated during 1964 and 1965. The selected samples are considered representative of midwestern warm-season storms which result primarily from thunderstorms and rainshowers. These samples also contain storms of various intensity associated with several distinct types of synoptic weather. Gross beta counting was accomplished with standard equipment. The metallic ion concentrations were determined with a Beckman atomic absorption spectrophotometer.

Results of this pilot study are illustrated in Figures 4 to 7. Individual time distributions are shown in Figure 4 for gross beta and each of the four metallic elements at one of the sampling stations during the passage of a squall-line thunderstorm with heavy rainfall rates on May 26, 1965 Except for a modest reversal of trend near the time of minimum concentration, this graph shows similar trends throughout the storm for radioactivity and each of the chemical elements, with the maxima and minima nearly coinciding.

In Figures 5 to 7 normalized distributions in selected storms are shown. In normalizing, the concentration in each individual sample collected within a storm's passage is expressed as a ratio to the average concentration for the entire storm at the sampling point. This procedure facilitates comparisons of measured concentrations within and between storms of different magnitude, storm type, etc. Median values of the 4 chemical-element ratios have been employed in the illustrations to eliminate minor fluctuations not typical of the general storm trend. Normalized rainfall rate curves are also shown for comparison purposes.

Figure 5 shows normalized curves for the same storm and station presented in Figure 4. The trend curves for beta and the metallic elements are strikingly similar in shape, although the chemical-element curve has higher initial ratios and lower final ratios than beta. The rainfall rate curve indicates a single-burst storm.

In Figure 6, relations are shown in an air mass storm on May 25, 1965. In this 2-burst storm, the beta and chemical-element curves are again very similar in shape and in the location of maxima and minima in the time distribution. Also, similar to the previous figure, both curves show a relatively high concentration at the leading edge of the storm.

This time distribution in the storm of April 20, 1964 shows strikingly similar behavior between the beta and chemical-element curves throughout the storm and both again show relatively high concentrations at the start of the storm.

Results obtained for 3 other storms not illustrated were similar to those shown here. . Thus, the results lend support to the findings of previous investigations that the lower troposphere is a region of major input of radio-activity into convective storms, and that the relatively high concentrations frequently observed at the leading edge of rainstorms is related to the entrainment of particulates and evaporation processes in the lower atmosphere. Also, the relatively strong relationship observed between the time distributions

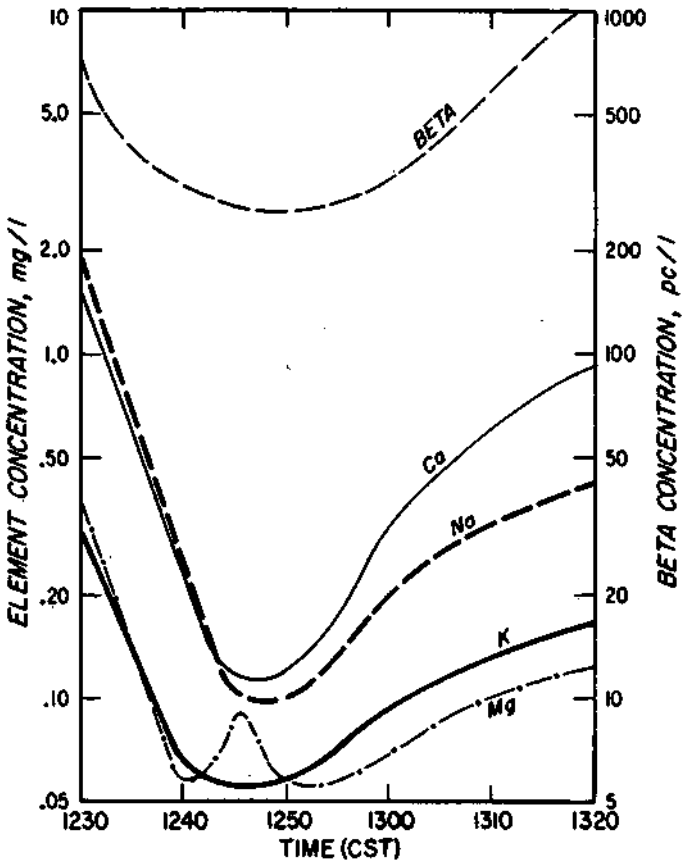

Figure 4. Individual Distributions on May 26, 1965

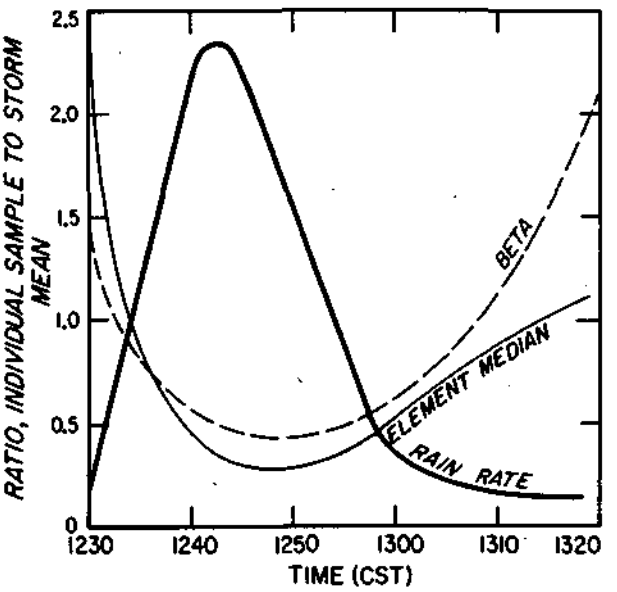

Figure 5. Normalized Distributions on May 26, 1965

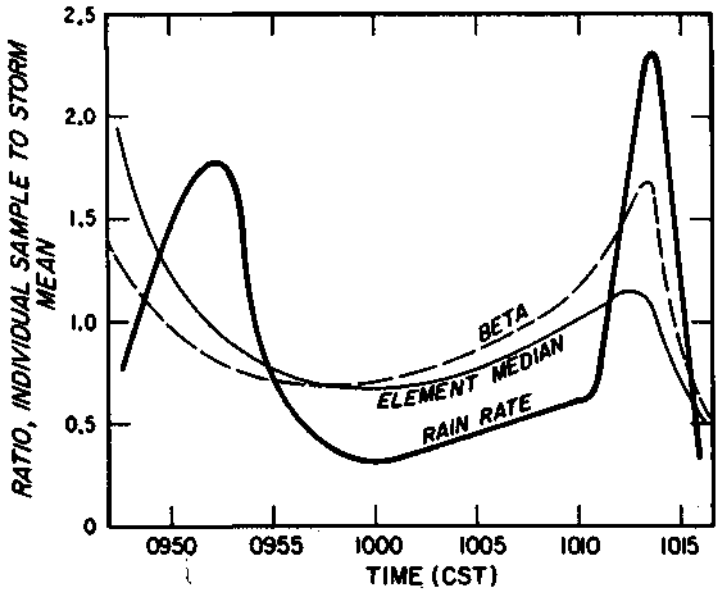

Figure 6. Normalized Distributions on May 25, 1965

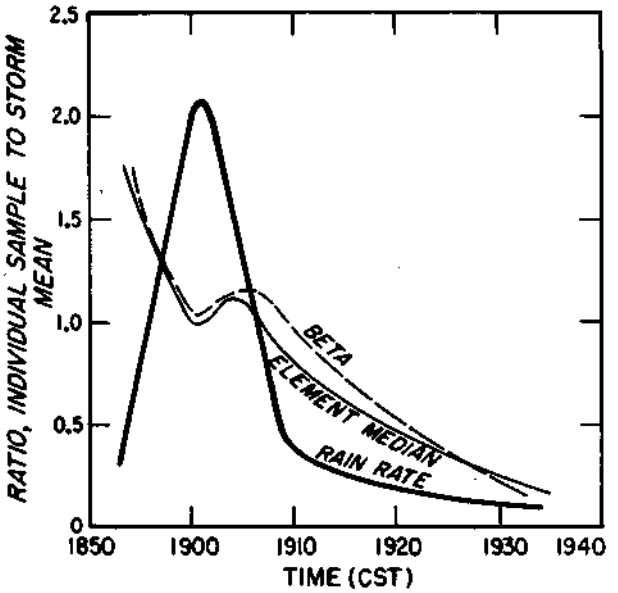

Figure 7. Normalized Distributions on April 20, 1964

of radioactivity and the chemical constituents is in agreement with the earlier investigations of Georgii and Weber (1961) who concluded from European data that "the accumulation of radioactive fission products in rainwater and of natural tract substances obey the same law."

To aid in evaluating how well the Illinois storm samples represented general atmospheric conditions, comparisons were made with average relationships provided by Junge (1963) in his well-known text on air chemistry and radioactivity. The average concentrations of Na and K in the storms, 0.26 and 0.08 mg/1, respectively, compared satisfactorily with Junge's values of 0.25 and 0,12 mg/1 in the sampling area. Furthermore, the average ratio of Na/K in the Illinois storms, 3/1, compared closely with results from a previous analysis of 62 Illinois rainwater samples (Larson and Hettick, 1956). However, the Ca average was much lower than Junge's average, and we have no satisfactory explanation of it.

Comparison of the ratio, K/Na, in the Illinois sample with Junge's averages for the United States showed the best correlation with areas SE, S, and SSW of the sampling network, such as southern Illinois, the western parts of Kentucky and Tennessee, and Missouri. These correlations are reasonable since the low-level, moisture-bearing winds were from the above directions in the storms analyzed. The ratio varied considerably between storms, ranging from 0.17 to 0.44. This range is not unexpected considering (1) the localized differences which may result from variations in wind strength and convergence in and near the storm systems, and (2) variations that may occur in the water transport into a storm system depending upon the circulation and stability in the lower troposphere.

Conclusions

The amount of data analyzed to date is insufficient to reach firm conclusions on the radioactive input processes in convective storms. However, this abbreviated study does lend additional support to previous findings which indicate the lower troposphere is a major region of input into such storms, and perhaps more important than direct stratospheric input. Also, results indicate that the concurrent analysis of radioactivity and chemical constituents in rainwater may provide useful information related to the atmospheric processes controlling radioactive rainout.

WORKSHOP ON TRITIUM APPLICATIONS IN THE ATMOSPHERIC SCIENCES

On June 5-6, 1967, a conference was held at the Illinois Water Survey to discuss the status, present operations, and future" plans in tritium research in the United States and elsewhere. Another major purpose of the meeting was to obtain recommendations from the invited group of scientists on the Water Survey's tritium sampling program for 1967. The conference was attended by: Dr. Gote Ostlund, University of Miami, Florida; Dr. D. H. Ehhalt, NCAR; R. M. Smith, International Meteorological Institute, Stockholm; Dr. R. J. Engelmann, AEC Fallout Studies Branch; Eugene Bierly, NSF; G. E. Stout, F. A. Huff, W. E. Bradley, and P. J. Feteris of the Illinois Water Survey.

The first part of the conference was devoted to a discussion of the present activities of each participating organization in tritium research and an interchange of ideas on existing research. For example, Ostlund discussed his hurricane research. Ehhalt described his tritium-deuterium studies of hailstones. Smith spoke on his current plans and proposals to use tritium to define tropospheric-stratospheric exchange relations and recycling of tritium resulting from surface evapotranspiration. Engelmann discussed a project underway at Hanford to use isotopes produced by cosmic rays and found in rainwater to study scavenging effects in and below clouds. Bradley outlined the planned 1967 program of the Water Survey for sampling flights from the Gulf of Mexico to the Midwest, and a number of helpful suggestions was received from the participants relative to this program.

The latter part of the 2-day conference was spent in summarizing the history of tritium research, outlining the advantages and the disadvantages of tritium as an atmospheric tracer, and discussing recommendations for future studies. Ostlund summarized the history briefly. He pointed out that it became apparent in the late 1950's that the fission bombs were causing the tritium level to increase considerably, and that tritium samples should be secured for future use as a tracer in atmospheric processes. In 1959 a continental network of rainwater stations was established by WMO and IAEA in Vienna. This system has since then been expanded to all parts of the world except continental Asia and open ocean areas. The tritium samples collected are sent to various laboratories for analysis. Their data are available to virtually anyone who needs them from IAEA in Vienna.

The use of tritium as a tool in meteorology has been pursued only by a limited number of scientists and the. applications that have been tried fall into four problem categories: global circulation, synoptic-scale systems, mesometeorology, and micrometeorology.

It was pointed out that the primary, advantage of tritium as a tracer is that it is in a water state and can be used effectively to trace the movement of water vapor in the atmosphere. Other advantages include the fact that it is already present in the atmosphere, it is not particularly biologically dangerous as a tracer, it is conservative since it has a relatively long half-life, it is more accurately measured than fission products, and several techniques have been developed and tested for collecting and analyzing both the liquid and vapor states.

The primary disadvantage listed by the participants is that the atmospheric concentrations have decreased to levels which will make it unsatisfactory for tracer studies in some latitudes soon, and this condition will worsen with time unless more fission products are released into the atmosphere. For example, Ostlund estimated that the last possible time that tritium can be used as a tracer in tropical latitudes and marine air in conjunction with his hurricane studies will be Summer 1969. However, Ehhalt expressed the opinion that at continental stations it may be possible to detect tritium concentrations above the background for another 10. years with reasonable accuracy.

It was pointed out that with generally decreasing levels of tritium concentration, the atmosphere will be subject to contamination from local sources, such as reactors. Also, contamination may occur locally from the industrial application of tritium, such as in the manufacture of luminescent paint.

In discussing recommendations for future studies, Ostlund indicated the situation is still reasonably favorable to carrying out experiments with durations not exceeding two years, provided the capital outlay is not too large. On the other hand, it is not scientifically or economically feasible to start large-scale projects requiring extensive design, development of facilities and equipment, and long-term sampling.

It was pointed out that it is highly desirable at the present time to utilize other tracers simultaneously with tritium. This will: 1) allow work underway to be extended, if necessary, when the tritium concentrations reach levels too low for tracing, 2) aid in detecting contamination of samples, and 3) provide data of greater value for atmospheric studies, The group indicated

that research pertaining to improvements in samplers and techniques should be supported, so that it will be possible to exploit tritium's power as a tracer if there are future influxes into the atmosphere. Bierly stressed that there should be greater interplay between researchers in this field, since there appears to have been a communication problem in the past.

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APPENDIX A

Reports prepared under the
Contract Number AT(11-1)-1199
U. S. Atomic Energy Commission

COO-1199-1 = First Progress Report - January 31, 1963 - F. A. Huff
"Study of Rainout of Radioactivity in Illinois"

COO-1199^2 = Second Progress Report - January 31, 1964 - F. A. Huff
"Study of Rainout of Radioactivity in Illinois"

COO-1199-3 = SWS Reprint Series No. 46 - F. A. Huff
"Radioactive Rainout Relations on Densely Gaged Sampling Networks"

COO-1199-4 = SWS Reprint Series No. 45 - F. A. Huff and G. E. Stout
"Distribution of Radioactive Rainout in Convective Rainfall"

COO-1199-5 = Third Progress Report - January 31, 1965 - F. A. Huff
"Study of Rainout of Radioactivity in Illinois"

COO-1199-6 = Research Report No. 1 - March 1965 - F. A. Huff
"Radioactive Rainout Relations in Convective Rainstorms"

COO-1199-7 = Research Report No. 2 - October 1965 - P. J. Feteris
"1964 Project Springfield Studies"

COO-1199-8 = Fourth Progress Report - October 1965 - F. A. Huff
"Study of Rainout of Radioactivity in Illinois"

COO-1199-9 = Reprint - Vienna paper - Symposium on the Use of Isotopes in Hydrology - G. E. Stout and F. A. Huff - November 14-18, 1966
"Rainout Characteristics for Hydrologic Studies"

COO-1199-10 = Fifth Progress Report - December 1966 - W. E. Bradley and P. J. Feteris
"Study of Rainout, of Radioactivity in Illinois"

COO-1199-11 = Reprint - February 10, 1967 - W. E, Bradley and Gordon E. Martin
"An Airborne Precipitation Collector"

COO-1199-12 = Reprint - TELLUS - October 1967 - F. A. Huff and G. E. Stout
"Relation between Ce^{144} and Sr^{90} Rainout in Convective Rainstorms"

COO-1199-13 = Conference at Chalk River Laboratories, Canada - September 11-14, 1967 - F. A. Huff and G. E. Stout
"Time Distributions of Radioactivity and Chemical Constituents in Rainfall"

COO-1199-14 = Sixth Progress Report- November 1967 - F. A. Huff, W. E. Bradley and P. J. Feteris.
"Study of Rainout of Radioactivity in Illinois"