

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

STUDY OF RAINOUT OF RADIOACTIVITY IN ILLINOIS

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INTRODUCTION

The natural variability of radioactivity in precipitation has been carefully studied in recent years. Study has progressed to a laboratory investigation of the aerosol impact collection efficiencies of raindrops, and a search for new tracers of atmospheric moisture processes in the atmosphere.

During 1969, efforts have been channeled into two areas, one performed in the laboratory and the other in the field. The two efforts complement each other in the attempt to define the physical processes responsible for the distribution of trace materials in the atmosphere.

ACKNOWLEDGMENTS

The work during the past year was extensive, and hence involved many people. It did not seem proper, however, to name any single person or group of persons as being authors of the work. Several individuals organized these projects, and spent much time supervising their operations G. E. Stout, R. G. Semonin, J. W. Wilson, and W. E. Bradley. Since so many people played integral parts in both the field work and the laboratory experiments discussed on the following pages, the names of these personnel are listed with no reference to their status at the Water Survey or on the project T. C. Brandner, R. C. Buchhass, R. Cataneo, M. C. Clevenger, C. W. Furrey, R. H. Hoffman, D. K. Larson, P. M. Meyer, S. D. Rogers, J. A. Spencer, C. J. Sturek, and D. D. Watson.

Other individuals also contributed their time and efforts to the work. Mr. S. A. Changnon performed the rainfall analyses useful for the 29 May tracer case, his help is appreciated. Mr. A. H. Bodenschatz of the Chemistry Section of the Water Survey conducted analyses of water samples for trace metals. The DeWitt County, Illinois, 4-H Chapter, represented by Mr. H. D. Cruthis, kindly consented to the construction of the field laboratory on the 4-H Fair Grounds at Clinton. Many volunteer observers, too numerous to mention, agreed to the placement of instruments and samplers in their front yards. Their patience and helpfulness are appreciated; we particularly thank those who undertook the task of caring for and servicing these instruments. The National Severe Storms Laboratory of ESSA kindly loaned 9 microbarographs for use on the raingage network. Time spent by the Severe Local Storms Center of ESSA and the Military Weather Warning Center of the U. S. Air Force is also appreciated. Their advice and forecasts proved valuable to the project. The CPS-9 radar used in the field work was provided by the U. S. Army Atmospheric Sciences Laboratory at Ft. Monmouth, New Jersey. Personnel from Chanute AFB should also be thanked for providing radiosonde data to the project. Radiosonde data supplied by the Peoria and Salem, Illinois Weather Bureaus of ESSA was also used, the cooperation of Mr. R. Fennel and Mr. M. D. Shore of these offices is appreciated.

PROJECT ITREX FIELD STUDY

Introduction

The problem of the scavenging efficiency of precipitating storms has been examined from the theoretical approach as well as from certain limited laboratory and field experiments. However, it is necessary to confront the problem in the laboratory of the atmosphere using actual storms. This type of experiment requires knowledge of the thunderstorm structure, the placement of a unique chemical tracer into a prescribed part of the cloud, and the subsequent analysis of the tracer in the storm rainwater.

During 1969, such an experiment was undertaken by the Illinois State Water Survey, in cooperation with the University of Michigan and the Argonne National Laboratory. The name chosen for this project was ITREX, an acronym for Illinois TRacer Experiment. The primary mission of ITREX was to release a chemical tracer into a specific portion of a convective cloud and measure the areal deposition of the chemical at the surface. Determining the scavenging efficiency of a thunderstorm requires a complete budget of the tracer material. A description of the tracer budget requires the chemical analysis of the rainwater, a meteorological analysis of the synoptic situation, and knowledge of the micro-physical parameters of the convective system.

A complete assessment of the movement of particulates through a thunderstorm system necessitates knowledge of the identity, size, and concentration of the particles entering the storm. The tracer can be material artificially injected into a cloud, or particles occurring naturally in the atmosphere. In addition to the primary mission using artificial tracers, effort has been directed toward determining the feasibility of using naturally occurring aerosols as tracers. This part of the study involved the collection of a large number of ramout and dry fallout samples to determine the areal and temporal variability of certain trace metals.

Surface air samples also were taken to determine the normal concentrations of these metals in surface air. These analyses are being carried out at the Argonne National Laboratory by Dr. S. S. Brar.

Project ITREX was conducted over the Water Survey's Central Illinois Network of 196 recording raingages in a 1600 square mile area. Field activities were performed during two different periods, and neither was during normal peak periods of thunderstorm frequency. However, conflicts in the scheduling of personnel and aircraft dictated the dates of each period.

The first phase of investigation was from 15 May through 14 June. The University of Michigan participated actively during this time. The operational plan was to release an artificial tracer by burning pyrotechnic flares into low-level updraft ahead of a thunderstorm and to measure the tracer which fell in precipitation. The flight program for this phase was sub-contracted to Weather Science, Inc.

The tasks of the Weather Science aircraft were to deliver chemical tracers into selected storms and to measure meteorological parameters pertinent to these storms. Cloud physics measurements of cumulus clouds were also desired, but were to be made on days when no thunderstorms were present. Complete details of the Weather Science operations have been given by Sutherland (1969).

The second phase of field study was from 23 July through 8 August. The main effort shifted from particulate tracer studies to air sampling using gas as a tracer and releasing it into the upwind side of a storm. Argonne National Laboratory was responsible for the majority of work conducted during this period, and Michigan personnel did not participate. An aircraft was supplied by the National Center for Atmospheric Research (NCAR) with duties similar to those of Weather Science, Inc.

During both field periods, the Water Survey conducted its investigations of the areal variability of the rainout of natural tracers, although greater effort was expended during the 15 May to 14 June period.

The report is mainly concerned with the Illinois State Water Survey's operations during Project ITREX. Activities of the other participating organizations are discussed, but specific details have been left to those organizations.

Network Instrumentation

As previously mentioned, the Central Illinois Network (CIN) of 196 recording raingages was used as the study area for Project ITREX. All gages were of the standard weighing bucket type, and had either 8- or 12-inch diameter tops. Twenty-four hour gears and charts allowed measurement of 0.01 inch per 5- to 10-minute period, which gave good definition of the intensity and time distribution of most rains. Figure 1 shows the location of the network.

The rainwater collected from the raingages could not be subjected to chemical analysis because of the metallic composition of the buckets, so polyethylene baskets were placed at each raingage site. The baskets were chosen for water collectors because of their large size, lack of metallic content, and economical price. Each basket was 20 inches in diameter, and was to be lined with a disposable polyethylene bag to eliminate collector cleansing and provide a clean surface for the rainwater deposition. The large basket top was useful for collecting large amounts of water in light rain, (59.5 cc per 0.1 inch of rain), and the bag could be removed and used as a temporary storage container for the rainwater. Altogether, 242 baskets were installed on the CIN, with 176 placed by the Water Survey at the 3-mile spacing of the raingages (figure 1) and 66 placed by the University of Michigan at a tighter (1-1.5 mile) spacing in the Clinton area. The liners for the baskets were changed as needed.

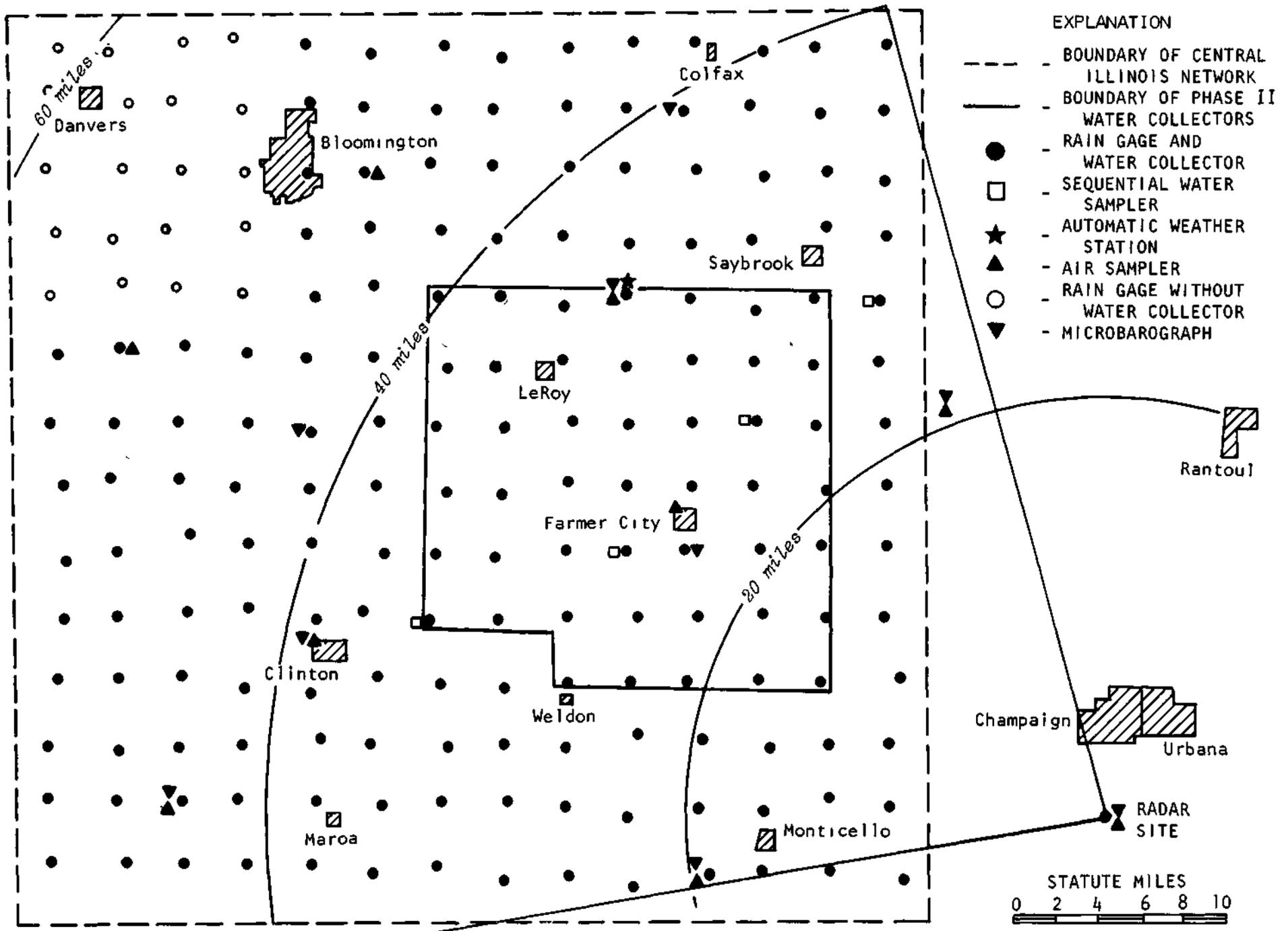


Figure 1 Instrumentation on the Central Illinois Network during Project ITREX in 1969

In addition to measuring the areal deposition of artificial and natural tracers, it was desirable to measure the time variability of the tracer rainout to facilitate modeling the storm washout. For this purpose, four sequential water samplers (Huff and Bradley, 1965) were modified and installed at the locations shown in figure 1. Runoff from the roof of each sampler was dispersed sequentially into 36 sample bottles on two turntables as shown in figure 2. A chart recorder logged the time of each sample collection. These four sequential samplers, plus the Clinton field laboratory, constituted a 32-mile line along which sequential samples could be taken.

A field laboratory was constructed at Clinton as headquarters for the University of Michigan scientists. The 18- by 24-ft building was a receiving point for rainwater collected on the CIN. Three large Fiberglas funnels (6-ft 10-mch diameter) were placed beside the building for rainwater collection. These yielded approximately a gallon of water for each 0.01 inch of precipitation. Rough filtering of water was also done by the field laboratory staff. Also at the laboratory were a raindrop size spectrometer (Dingle and Schulte, 1962), a tipping bucket raingage, and a weighing raingage. Communications with both the aircraft and ground-based mobile units from the laboratory used an FM frequency of 151.625 MHz.

The raindrop camera (Jones and Dean, 1953) was operated at Clinton by the Water Survey. Data reduced from photographs of raindrops can provide a comparison with the Michigan raindrop size spectrometer's output, and should yield basic knowledge of the rain structure in the storms chosen for study.

An automatic weather station (Meteorology Research, Inc., Model 1071) was placed on the network (figure 1). Continuous recording of wind speed, wind direction, and temperature provided additional data for meteorological analysis on a scale smaller than synoptic.

Atmospheric pressure at 9 points within and to the east of the network was recorded on microbarographs (figure 1), loaned to the Water Survey by the National Severe Storms Laboratory of ESSA.

Two 3.2 cm radars were used on Project ITREX. Both were operated by the Water Survey, and were located east of the raingage network. A CPS-9 was the primary radar used for surveillance and echo tracking when a storm of interest moved into a chosen target area. Automatic gain stepping and continuous scope photography were used to document echo activity. Polaroid pictures were taken at regular intervals, and were used for de-briefing purposes.

During the second phase of the project (23 July - 8 August), a SPA-8 remote scope became operational for use with the CPS-9. This scope has an offsetting capability and a variable range, which allowed particular storms to be studied closely regardless of their placements on the scope. The SPA-8 was especially valuable when vectoring an aircraft to a particular cell.

Since the Champaign VORTAC and the radar site are in nearby locations, the pilots could use the radar position of the cell to set the heading (VOR) and distance measuring equipment (DME) without transformation. If the scope is off-centered to appropriate coordinates, the vector from any other VORTAC to a cell can be read and relayed to the pilot. Since the CIN lies within



Figure 2. Sequential precipitation sampler

the ranges of three VORTACs, this feature of the Water Survey radar facility was very useful.

The second radar used on ITREX was a modified TPS-10. This RHI set yielded gain-reduction profiles of storms over the network, and therefore permitted quantitative measurements of the stage of development of individual storms.

As mentioned, the second phase of the field program was primarily concerned with air sampling and secondarily with precipitation sampling. For this reason, the number of polyethylene baskets was reduced from 242 to 47, as shown in figure 1. Air samplers, supplied by Argonne, consisting of polyethylene bags in large metal enclosures were stationed at 9 locations within and to the east of the CIN. Eight additional air samplers were used as mobile collectors, and their locations on any given day depended upon the particular storm situation. The bags, initially deflated, were filled by a small motorized pump. Sampling times could be either 15 minutes or 1 hour, depending on the bag size. Full bags were taken to Argonne for analysis.

Experiment Design

A typical day for Project ITREX began at approximately 0730 CDT, when the project forecaster began preparing the precipitation forecast. His local forecast was further confirmed by telephone discussions with the Military Weather Warning Center (MWWC) and/or the civilian Severe Local Storms Center (SELS), both located in Kansas City, Missouri. Their continuous watch for thunderstorms and their in-depth analyses of synoptic weather were helpful when determining areas and times of thunderstorm formation. If conditions indicated that thunderstorms might occur during the ensuing 12 hours, the Project Field Supervisor procured vehicles necessary for the day's operations.

The Weather Bureau at Peoria, Illinois, was contacted at about 0930 to obtain the 0700 CDT radiosonde data for local analysis. Arrangements had been made with ESSA before the beginning of Project ITREX for special radiosonde releases from Peoria at the request of the project. These special observations could give radiosonde coverage every 3 hours for 12 hours, if desired. Thus, it was conceivable to release a balloon both before and after a thunderstorm passed Peoria and reached the ITREX target area, providing the storm moved from the northwest. The times of these special releases were determined and given to Peoria during the morning if possible. During the second phase, radiosonde data from Salem, Illinois, also were available.

Under a prior agreement with the U. S. Air Force, data from student-conducted radiosondes released from Chanute AFB at Rantoul (figure 1) were made available to the Water Survey.

At 1000 CDT each day, a briefing of all ITREX personnel, except the Clinton field observers, was held at the Water Survey headquarters. Weather expected during the day was discussed, and plans for the day's activities

were made. If thunderstorms were in the forecast (a "go" day), the field crews were given instructions for deployment of polyethylene liners for the rainwater collectors. The particulars of the briefing were then relayed by telephone to the Michigan scientists at Clinton.

If storms were expected before mid-afternoon, radar surveillance was begun immediately after the briefing; otherwise it was postponed until a more appropriate time. Hourly surface maps were plotted from teletype data if necessary to track significant synoptic conditions. Facsimile maps were also watched for upper air data, surface prognostic maps, and radar depictions throughout the Midwest.

During the first phase of the project, the field crews spent most of the "go" days replacing or installing polyethylene bags in baskets on the CIN. The crews then went to the Clinton laboratory to await further instructions.

Normally the aircraft did not take off until either thunderstorms were observed and had been tracked by radar to within approximately 75 miles of Clinton, or conditions indicated that storms would form within a 75-mile distance upwind of Clinton. If storms had not yet begun to form, the aircraft was sent to a 5-mile-square "target box" upwind of Clinton. This target area was so positioned that thunderstorms would pass over Clinton approximately 20-30 minutes after inoculation with tracer material. This time period is the estimated residence time of air in a mature thunderstorm (A. N. Dingle, personal communication), and would allow tracer-laden rainwater to fall near Clinton.

Once a storm had been encountered in the target area by the aircraft, and radar indicated it was moving toward the Clinton area at the correct speed, the aircraft crew began measuring the horizontal extent and the strength of the updraft. If it appeared to the flight meteorologist that the cell was not in the dissipating stage, or would not reach this stage before it had moved across the CIN, tracer flares of indium tri-chloride were burned in the updraft ahead of the rain curtain. Traverses were flown parallel to the rain curtain, and the aircraft was kept in the updraft as much as possible.

Communication was maintained between the radar operator and the aircraft crew until the tracer release time. After the flares had been burned, the pilot instructed the Clinton field laboratory crew regarding which raingages were receiving rain from the storm, and where to distribute additional water samplers with the mobile units. During this time the aircraft could either continue to take updraft measurements, or fly through the rain to measure rainfall rate with a special instrument on board. An alternative plan considered required that the airplane fly at a very low altitude in the precipitation over the Clinton laboratory. This would result in quasi-simultaneous data for comparison of the aircraft ram-rate sensor, the Michigan raindrop size spectrometer, and the Illinois raindrop camera. A modified tipping bucket raingage was to be used as the control instrument in the rain rate comparisons.

In previous work, Gatz et al. (1969) noted that metallic ions in solution precipitated to the walls of the polyethylene bags and were difficult to remove.

In order to prevent this, hydrochloric acid was added to the collected samples to keep the ions in solution. Rather than add acid to the bags prior to a rain, which would give varying concentrations of acid depending upon the amount of rain, acid was added as quickly as possible after the rain.

The operational plan called for the collection of the polyethylene bags immediately after the tracer-impregnated storm had passed. The collected samples were brought to the laboratory at Clinton, where 100 ml of water was transferred from each bag to a 120 ml bottle pre-inoculated with 1 ml of 12 N HCl. This produced a 0.12 N HCl solution which was adequate to keep metals from adhering to the walls of the bottle. The remaining rainwater was analyzed by the Michigan scientists for indium. This procedure has been described by Gatz et al. (1969).

The main purpose of flights conducted during the second phase of Project ITREX was to release sulfur hexafluoride gas (SF_6) into the upwind side of a storm, in a region where air is thought to enter the cloud and eventually descend to the surface. This procedure assumes a thunderstorm model with mid-level inflow as discussed by Browning and Ludlam (1962). This mid-level inflow region is characterized by having the minimum wet-bulb potential temperature of the environment. The Peoria and Salem, Illinois, radiosondes were analyzed to determine a first approximation of where the SF_6 might be released.

The general operational procedure was much the same as for the first phase of the project. When a line of thunderstorms was observed on radar to be several hours from the raingage network, the NCAR airplane was sent to an airport 75-100 miles upwind of Champaign. This facilitated access to the upwind side of the squall line, and avoided penetrations through the line. When the line had passed the upwind airport, the aircraft could easily fly to the desired position. If non-squall line storms were expected, this procedure was unnecessary, as circumnavigation was no problem.

The radar operator vectored the aircraft to a particular storm that would track across the network and fulfill qualitative developmental requirements. The final selection of the storm and the location for the gas release was at the discretion of the flight meteorologist. When all conditions were satisfied, the valves on the SF_6 storage cylinders were opened while flying as close to the storm cloud as possible. This released 100 cubic feet of gas in 20 minutes. Once the gas had been released the airplane returned to the airport at Champaign.

The procedure for air sampling was different from that for water sampling. On a day when thunderstorms were likely, CIN residents who had volunteered to operate the air samplers were alerted by telephone or to the predicted storm period. A second call telling them to activate their samplers was made when the tracer gas was being released. The activation times were different for every location and depended upon wind speeds aloft and the altitude of the tracer release. After each sampler had operated for its specified length of time, it was turned off, and the full bag was sealed. Eight other air samplers were used as portable units, and were placed in the path of an approaching storm by Water Survey personnel. The full sampling bags were taken to Argonne National Laboratory for analysis.

Water-sampling operations were the same as during the first phase of the project, but were conducted only in the reduced network area of 47 raingages. The Water Survey made all analyses of these water samples.

During the interim period between the two phases (15 June - 22 July), the Water Survey continued to deploy and collect polyethylene liners in 176 water collectors on the CIN. Rain which fell during this time was analyzed for the trace metals.

Weather During Field Periods

The weather during the 15 May to 14 June period (first phase) was a typical for this time of year, yielding very little precipitation in central Illinois. The precipitation that fell came as steady rain or showers rather than as thunderstorms, making the goals for ITREX difficult to achieve.

Seven cold fronts passed through the area, and one of them became a warm front and moved north several days later. Several of the cold fronts became stationary in southern Illinois for approximately a day, then moved southward and out of the area. One cold front stalled in central Illinois on 5 June and presented a thunderstorm threat for four consecutive days before moving south. Thunderstorms did develop, but did not occur in an area suitable for the ITREX sampling.

Precipitation varied considerably over Illinois during the 31-day study period. Figure 3 shows the percent of normal rainfall as computed from U. S. Weather Bureau climatological station records. The lowest values are for stations in the central portion of the state, with some stations receiving less than 25% of their normal amount. Precipitation increased in almost every direction from the CIN, and northern Illinois received more than twice the normal amount of rain.

Clinton, located within the CIN boundaries, received 1.14 inches of rain, 25% of the normal amount. No thunderstorms were reported during the period, and all of the rain fell on five days, four less than normal. Urbana had 24% of the normal rainfall with four thunderstorm days. On two of these days the total precipitation was a trace, and on the others the thundershowers developed over Urbana and moved east.

Nevertheless, there were 10 "go" days forecast during this phase of ITREX, but tracer material was released on only one of these days. On the remaining 9 "go" days the storms either failed to develop in a suitable location, or did not move into the CIN. Several times the aircraft was sent to intersect a squall line upwind of Clinton, only to find that the line had dissipated some distance northwest. This is evident in figure 3, as the area only 60 miles northwest of the CIN received normal rainfall for the 31-day period. Appendix A summarizes the daily weather and operations during the 15 May - 14 June period.

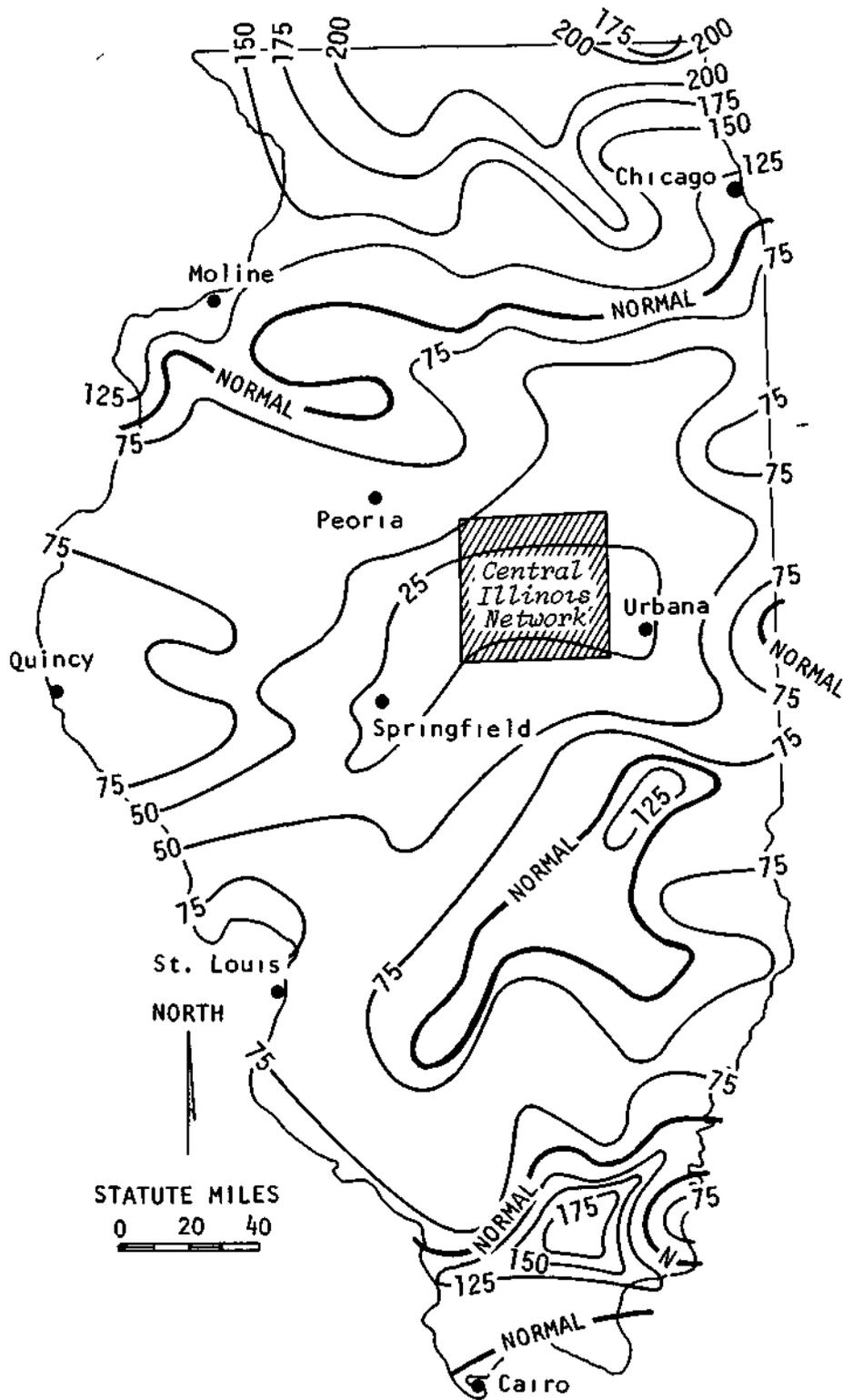


Figure 3. Percent of normal precipitation, 15 May - 14 June 1969

The weather during the 23 July to 8 August phase of ITREX was not much more conducive for tracer studies than that during the first phase. Very dry weather with fewer thunderstorms than normal characterized this period.

During the entire 16 days, a large ridge of high pressure over the western United States dominated 500-mb flow patterns. Winds at that level over Illinois were generally west to northwest, resulting in a cool dry period. Occasionally a small closed low moved across the top of the ridge and formed a trough east of the Mississippi River, but the only effect in Illinois was to increase the northerly component of the winds aloft, bringing in more cold air.

Only four cold fronts passed through Illinois during the period; three of these were at sunrise and the fourth was before noon. Thunderstorms associated with two of the cold fronts were reported at Urbana, but since the storms occurred near midnight on the CIN, they were of little use for the tracer operations which required visual conditions.

Only two "go" days were declared during the second phase, and on both days pre-cold frontal storms were expected. The storms did develop, but too far west to pass through Illinois before sunset. It is of interest that no air-mass storms were even forecast, as the cool dry air was too stable for their formation.

Precipitation in Illinois was more variable during the second phase than during the first. Figure 4 shows many localized maxima, most of which were the result of one or two heavy storms. Rainfall on the network varied considerably, from 26% of normal in the northwest corner to 124% of normal in the southeast corner.

Appendix B summarizes the ITREX daily operations and weather during this second period.

Tracer Case of 29 May 1969

During the two phases of Project ITREX, the aircraft several times searched in vain for a thunderstorm into which tracer could be released. On only one day, 29 May 1969, was such a storm found.

Figure 5 shows a series of surface synoptic maps on that date, and figure 6 shows the 500 mb charts for the same time period. Thunderstorms were present prior to 0700 CDT from Wisconsin to the Texas panhandle, but most dissipated during mid-morning. The strongest storms were associated with the short-wave trough dipping into Nebraska. This frontal activity was forecast to re-form during the afternoon. In anticipation that a squall line would form in central Illinois, preparations for a mission were begun at the 1000 CDT briefing.

From 0700 to 1000 CDT, the cold front advanced into Illinois at approximately 30 knots. During the next three hours, however, surface southerly flow increased west of Indiana, decelerating that portion of the front to 10 knots. Figure 5b shows the cold front extending through Chicago and north of Peoria, Illinois. A

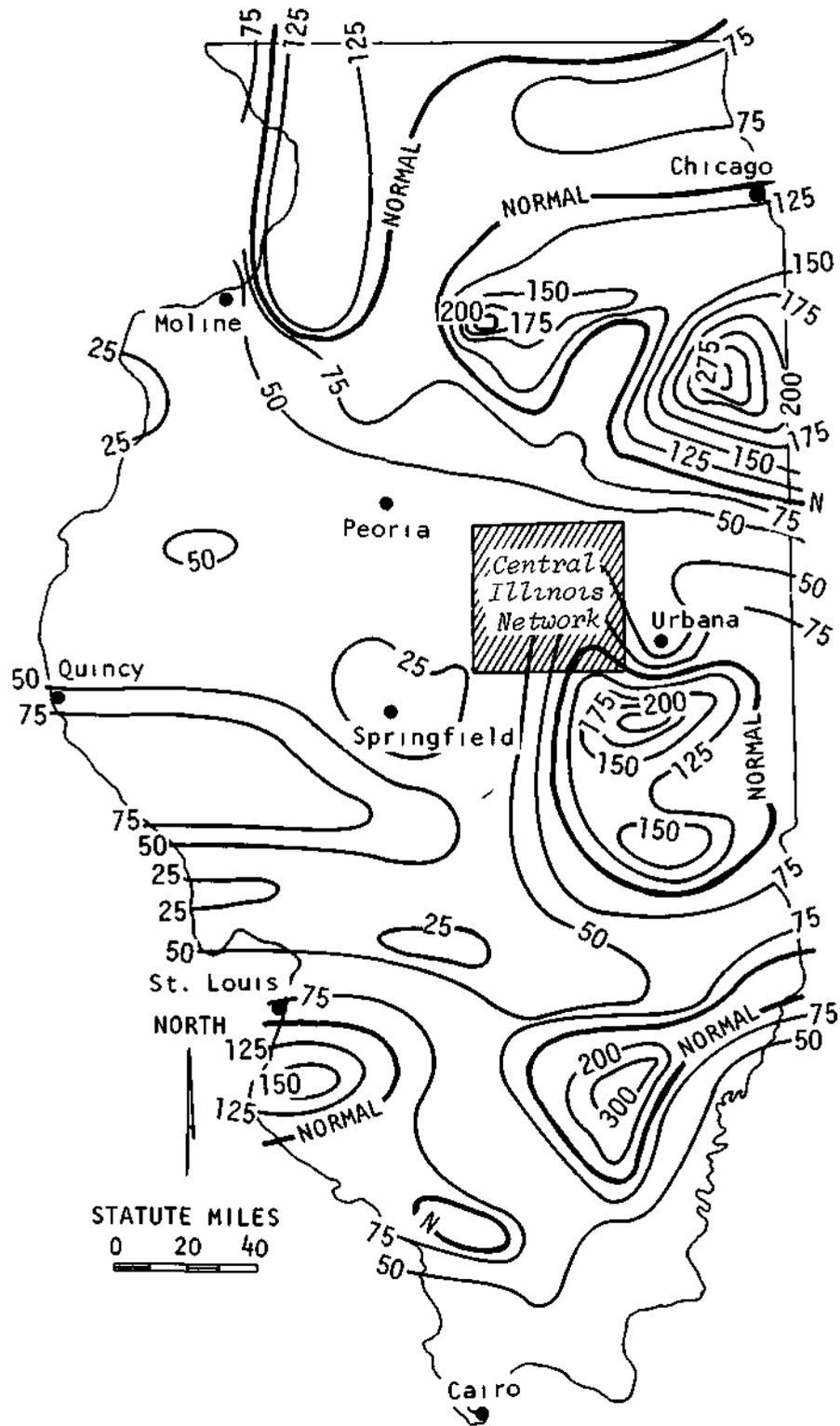


Figure 4 Percent of normal precipitation, 23 July - 8 August 1969

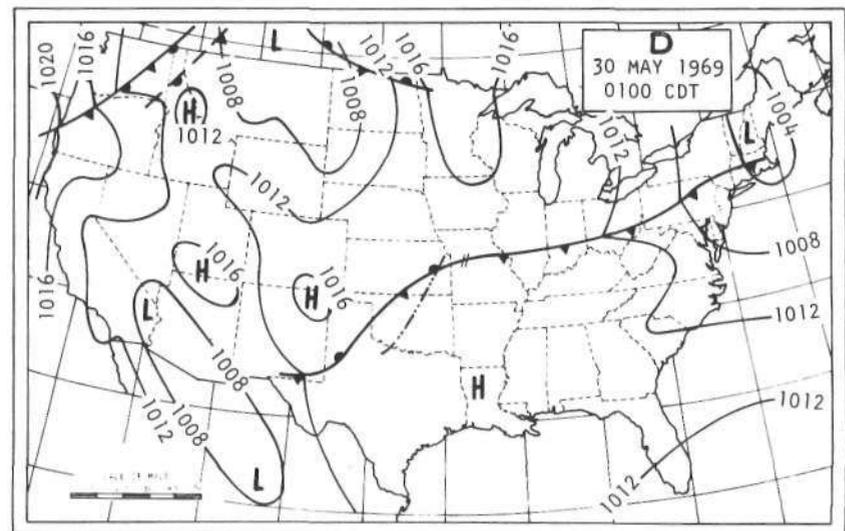
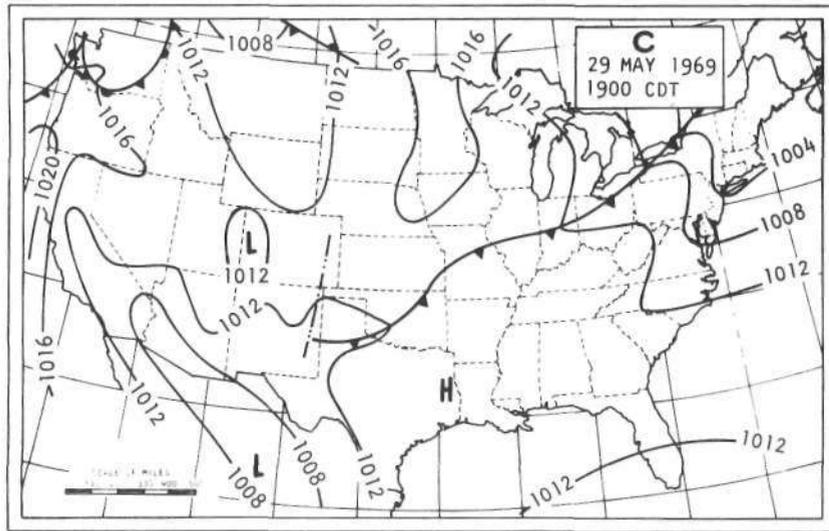
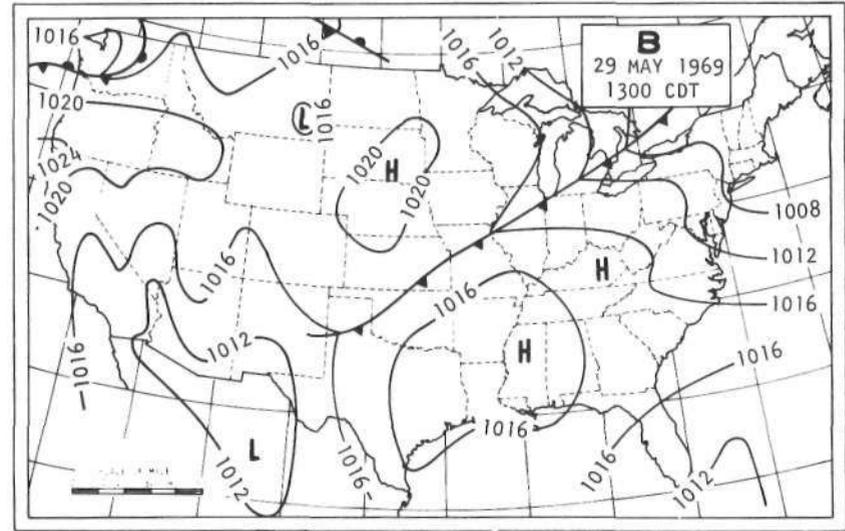
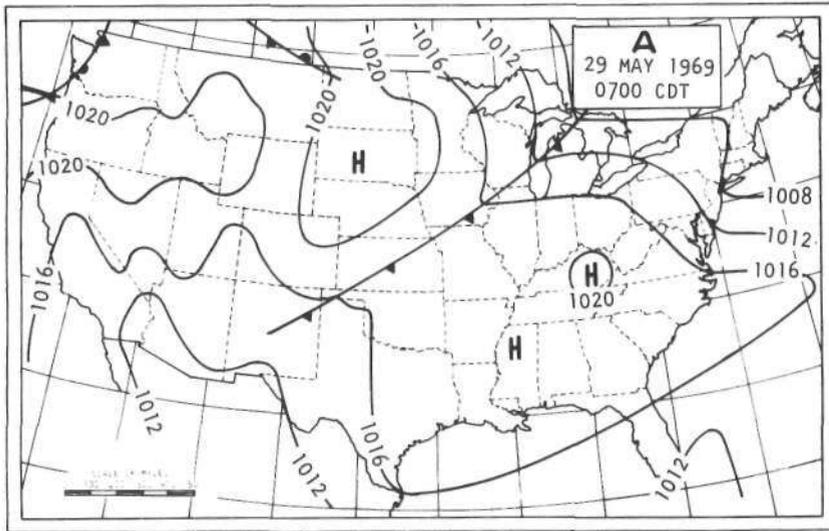


Figure 5. Surface synoptic weather maps pertinent to the 29 May 1969 tracer release

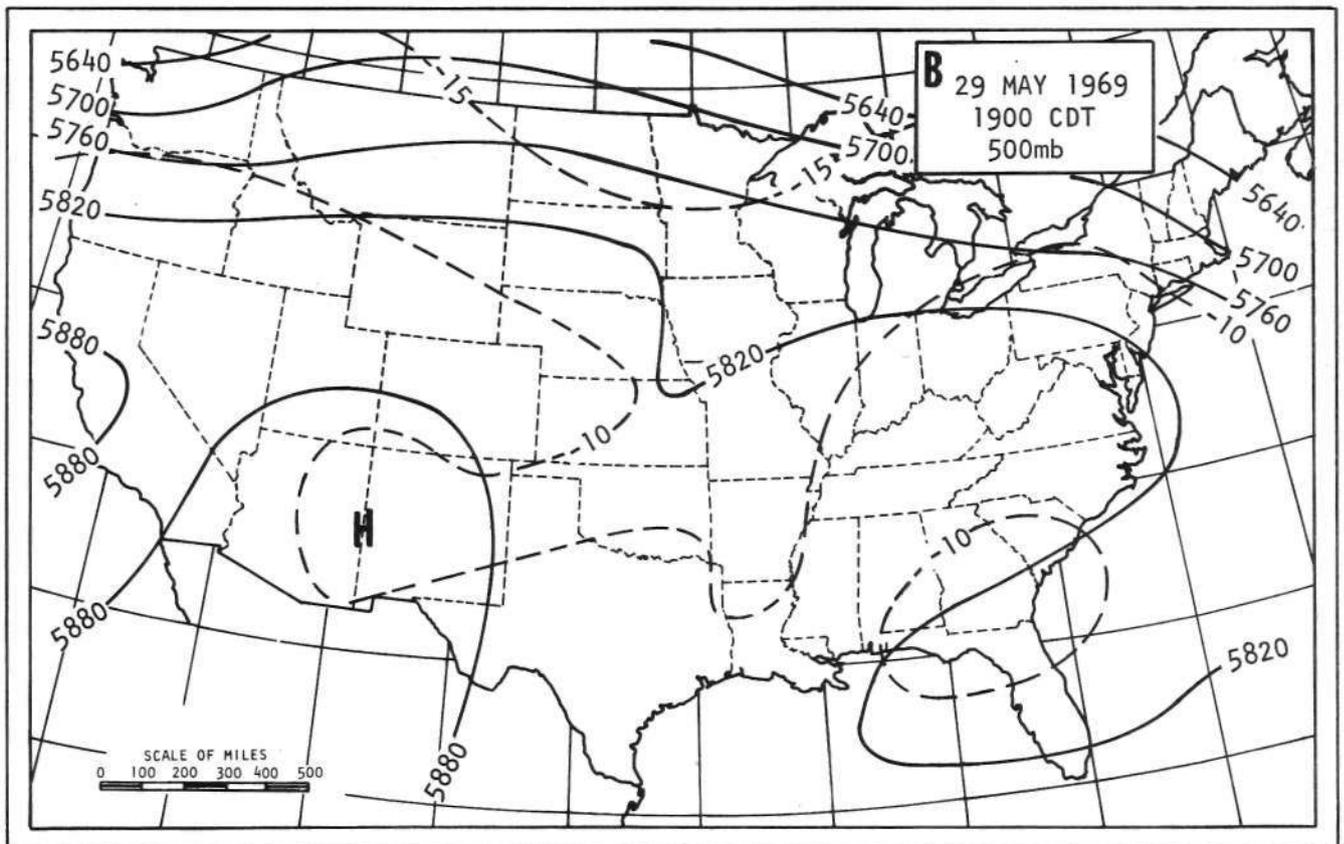
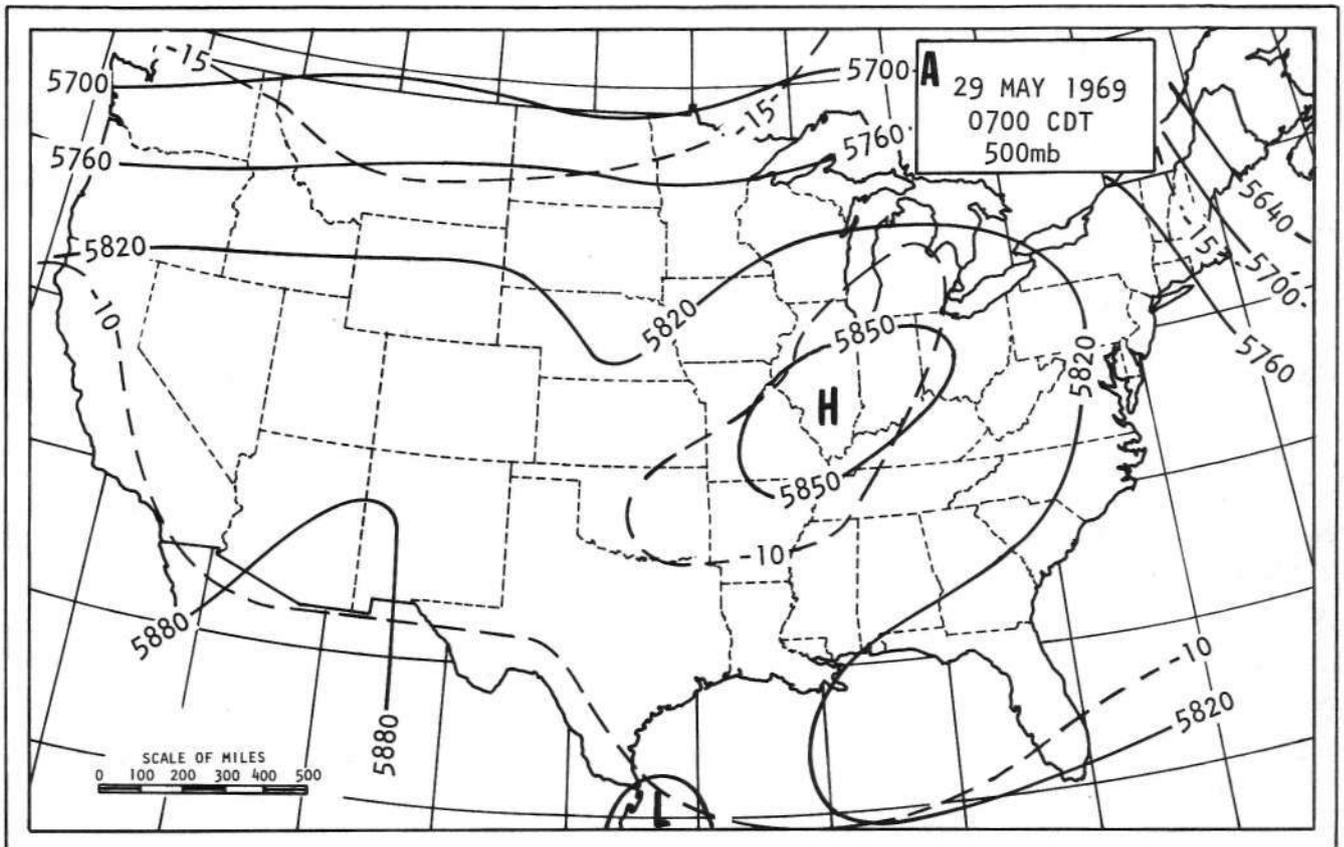


Figure 6. Upper air maps at 500 mb pertinent to the 29 May 1969 tracer release. Isohypsies (solid lines) are in meters, and isotherms (dashed lines) are in degrees C.

few radar echoes were evident along the front at 1300 CDT, but all were isolated and with little motion.

During this time 87 polyethylene liners were placed in baskets on the CIN, and 14 indium chloride flares were loaded onto the aircraft. Radar surveillance was continued, but the aircraft remained on the ground as the echo activity had not yet become organized.

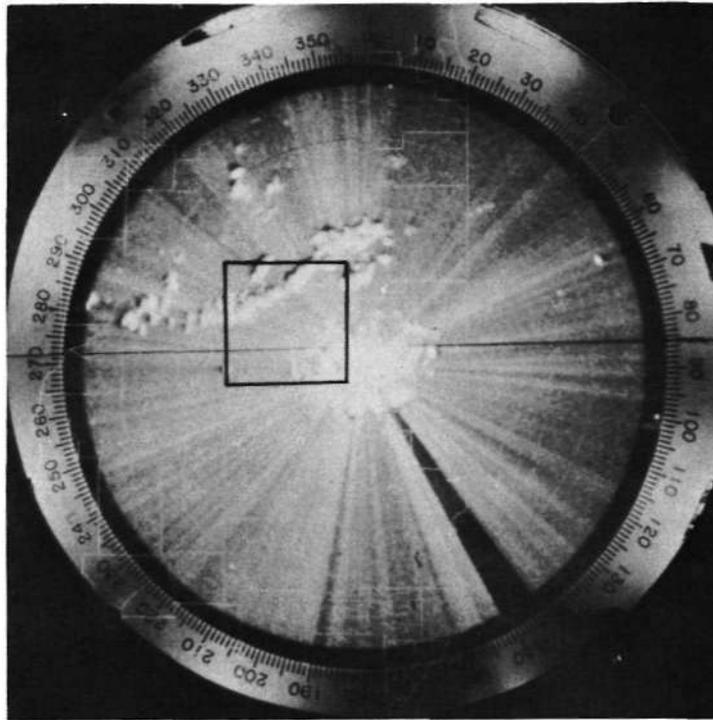
By 1800 CDT the front had moved to 15 miles north of Champaign, and was just south of Bloomington. A broken squall line formed very rapidly along the front. A Polaroid photograph of the CPS-9 scope taken at 1749 CDT is shown in figure 7. At 1800 CDT the aircraft took off from Champaign and flew to the pre-designated target area west of Clinton. A crew left from Champaign and drove to the sequential water samplers, since the samplers needed to be manually activated prior to the rain.

The aircraft arrived in the Clinton area at approximately 1820, and observed the thunderstorm tops to the north. Since no development appeared west of Clinton, the pilot flew north to search for a suitable storm in the squall line. The initial line of thunderstorms moved very little during the period shown in figure 7. Heavy rain and lightning were present along the leading edge of the line, and updrafts of several hundred feet per minute (fpm) were encountered.

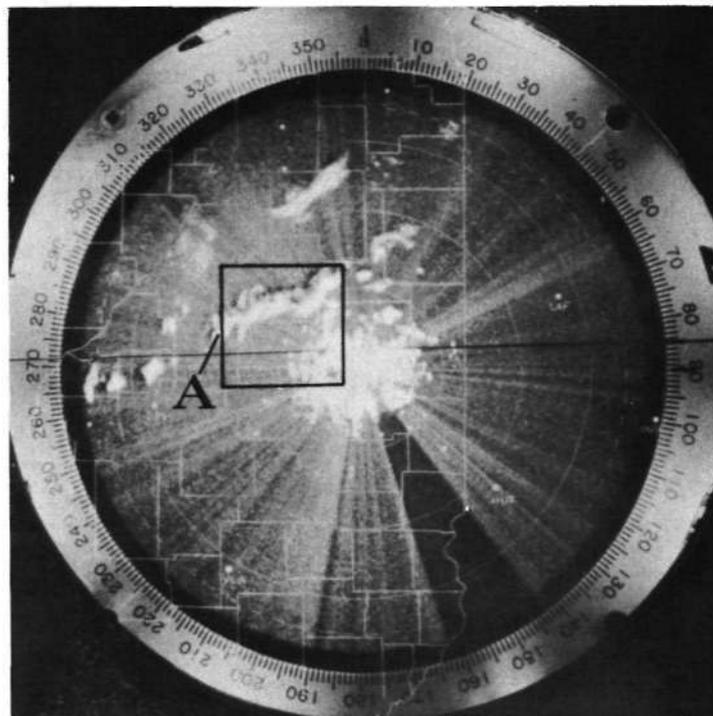
The cold front had slowed to about 5 knots, and as a result the line of thunderstorms remained quasi-stationary, and unlikely to arrive at the target area around Clinton. A small cell on the western edge of the CIN appeared on radar to be increasing (cell A at 1834 CDT, figure 7) and the aircraft was vectored to it. Updrafts of 1000 fpm were present beneath the cell at 5000 ft MSL, and a rain curtain was evident west of the region of maximum updrafts. This cell moved northeast (even though the line of storms was almost stationary), and since it would not pass over the Clinton laboratory no tracer was released into it.

At 1853 CDT another line of echoes formed very rapidly south of the main squall line. The TPS-10 operator noted echo tops of 28,000 ft, and since the echoes were still growing, the aircraft was sent to investigate. The strongest cell had a maximum updraft of 500 fpm, and was in a position southeast of Clinton that would allow precipitation to fall into the dense sub-network of collectors set out by the Michigan personnel. This echo is labeled as B in the 1859 CDT photograph of figure 7. Although not particularly strong, it was the most promising cell for tracer injection in the vicinity. The likelihood was becoming small that cells of the main squall line would move into a suitable area on the CIN, since the line was decreasing in intensity.

At 1859 CDT the decision to release tracer into this cloud was made, and at 1900 CDT 14 indium flares were ignited simultaneously and burned for 6 minutes. Most of the 560 grams of tracer went into the updraft, but some was unavoidably released into precipitation falling from the cloud. The maximum updrafts in this storm were still 500 fpm beneath the shower ahead of the rain. The TPS-10 measured the echo top at 32,000 ft at 1914 CDT. During burning of the flares, the storm drifted slowly eastward and was still southeast of Clinton. Figure 8 shows the indium flares before, during, and after the burn.

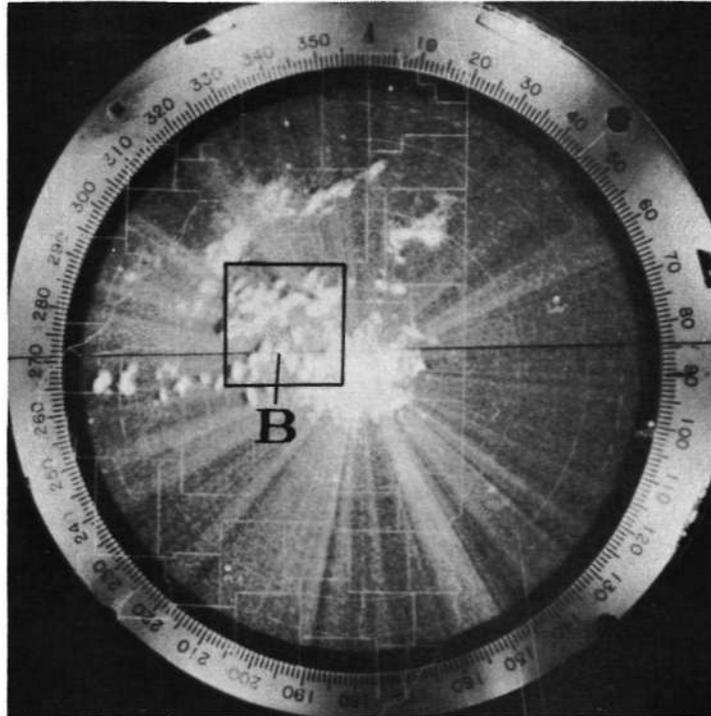


Time: 1749 CDT

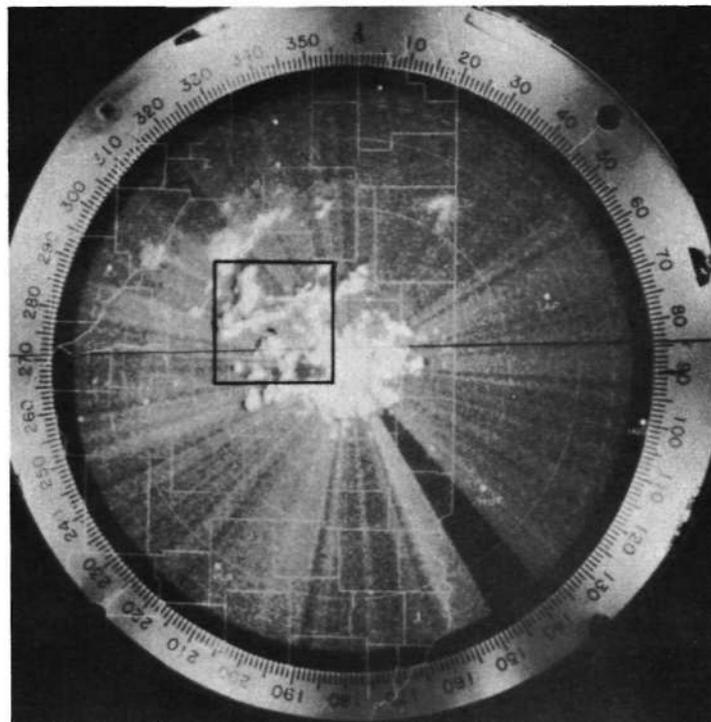


Time: 1834 CDT

Figure 7. Polaroid photographs taken on 29 May 1969 of CPS-9 radar scope, with radar at 0° elevation and full gain. Square is outline of raingage network



Time: 1859 CDT



Time: 1920 CDT

Figure 7. (continued)

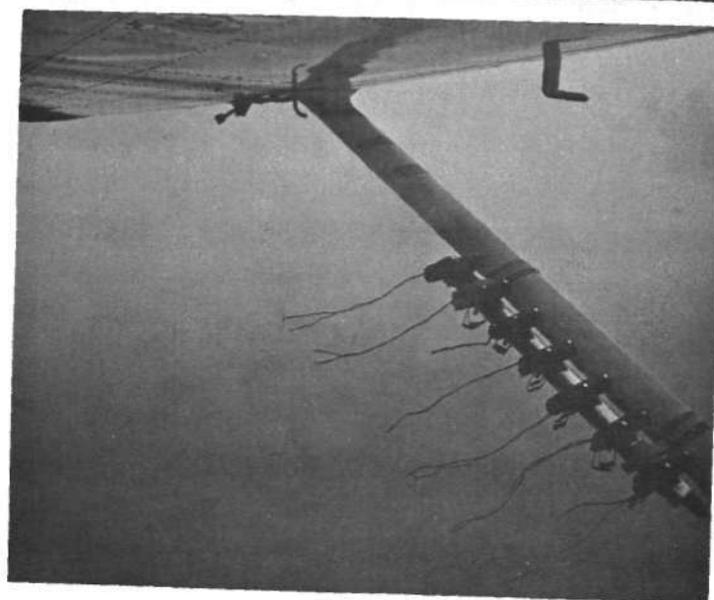
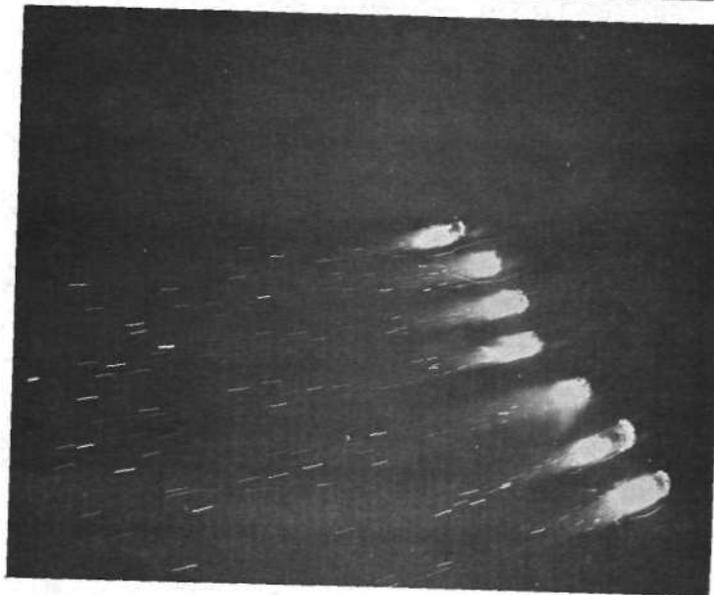
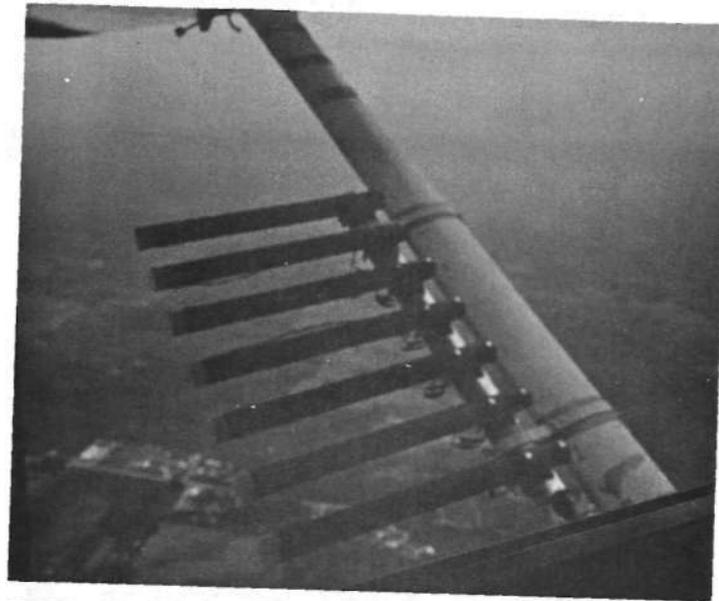


Figure 8. Left flare rack loaded with indium flares shown before, during, and after burning, 29 May 1969

It was at about this time that the main line of thunderstorms began to dissipate in Illinois, while the newer line of showers weakened and moved east (1920 CDT photograph of figure 7). The pilot then directed the Michigan mobile units to locations in the path of the inoculated shower and returned to Champaign at 1925 CDT.

The radar depiction facsimile map showed that the thunderstorms were not confined to Illinois, but existed as a scattered to broken squall line from northeast of the CIN southwestward along the front into western Texas. Figure 5c shows the 1900 CDT frontal position and the beginning of a wave in the Texas-Oklahoma region. The 500 mb short wave trough, still in relatively the same position as 12 hours previously, is evident in figure 6b.

By 0100 CDT on 30 May the front had moved to south-central Illinois, and the wave in the Texas panhandle had become more pronounced. Some thunderstorms still continued in Kansas and Oklahoma, but along the cold portion of the front all such activity had dissipated.

Showers from the secondary line of echoes were scattered and short-lived, and most passed over only two CIN raingages. Figures 9a-c show the rainfall from these cells, and figure 9 depicts the total rainfall. The cell southeast of Clinton in figure 9b was the one which received the tracer; less than 0.10 inch of rain fell at either gage from this shower. The rainwater sample bags at these two locations received rain from other cells, but the dilution effect of the additional water cannot be known until the indium analyses are completed by Michigan.

None of the sequential water samplers received rain from the secondary line of showers (which included the cell impregnated with indium). Consequently time-resolved data of the tracer-laden rainwater are not available.

The Weather Science, Inc., aircraft used in the first phase was also to provide meteorological data and accurate position records. It was learned after the 29 May mission that a faulty component in the data recording system had caused incorrect analog voltages to be recorded for every meteorological and aircraft position parameter. Thus the aircraft track, as well as the meteorological data, could not be used for this case. Since the radar operator and the pilot had been in constant communication regarding the plane's position, the location of the flare burn was known relatively well. The scattered nature of the showers allowed closer pinpointing of the desired cell, as some of the echoes failed to precipitate.

Rainout and Dry Fallout of Trace Metals

As mentioned previously, a study was initiated to investigate the use of natural trace metal aerosols as particulate tracers. This study involved the collection and analysis of a large number of samples to determine the areal and time variability of the rainout and dry fallout of certain trace metals.

Chemical analysis for trace metallic ions was performed by the Water Survey with an atomic absorption spectrophotometer. All of the precipitation

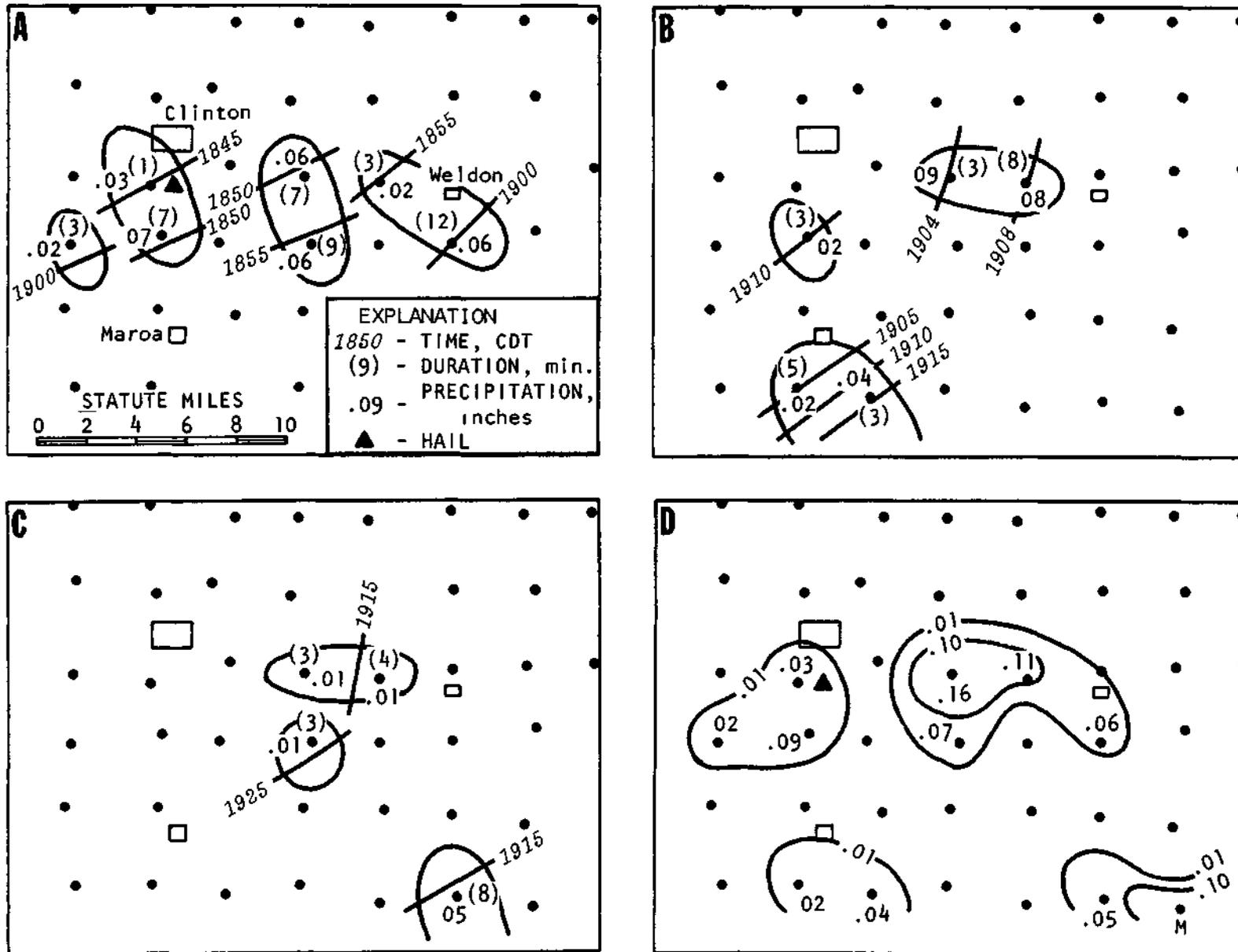


Figure 9. Rainfall associated with tracer release of 29 May 1969. Rain cells are shown in a-c, and total rainfall in d

samples were analyzed for sodium, potassium, calcium, magnesium, copper, and zinc, and most were analyzed for lithium. A 100 ml aliquot was extracted from the rainwater sample, but less than 50 ml was needed for the analyses, leaving 50 ml for re-analysis if necessary.

The accuracy of the spectrophotometry technique for each of the trace metals is expressed in terms of percentual concentration limit, or PCL. The PCL is defined as the concentration in ppm of analyte which gives a signal corresponding to 1% absorption under given operating conditions, and is equivalent to "sensitivity" expressed as ppm/1% absorption. The PCL for each metal analyzed is given in table 1 for the operating conditions used. All the metals except potassium, sodium, and lithium can be analyzed at PCL's two or three times lower than those given, if the situation warrants it. The actual limit of sensitivity is also several times lower than the PCL. The analysis of copper in any of the control sample analyses is limited to 0.5 ppm because of its presence in our deionized water in concentration up to that level.

TABLE 1

Percentual concentration limit in ppm for trace metals

Metal	Na	Ca	K	Mg	Cu	Zn	Li
PCL	.005	.260	.011	.028	.017	.008	.005

The handling of both the wet and dry fallout samples required several analytical steps, and it was necessary to determine to what degree contamination might occur during the sampling and handling processes. Potential sources of contamination included the polyethylene sampling bags, sample bottles, distilled water, hydrochloric acid, the spectrophotometer, dry fallout, and improper handling of the bags.

Contamination of the polyethylene sample bottles was checked in the following manner. One hundred milliliters of deionized water, acidified to 0.12 N HCl, was added to the 120-ml sample bottles. The water was analyzed after several hours by atomic absorption for the metals of interest. It was found that the sample bottles as they arrived from the manufacturer were contaminated, so it was necessary to wash them in HCl and to rinse them with deionized water before use.

The polyethylene liners were checked for contamination in the same manner as the sample bottles. The bags were flushed with acidified water and allowed to sit overnight. Fortunately, the liners were found to be clean as they arrived from the manufacturer.

Further tests showed that contamination was introduced if the inside of the liners or the sample bottles was touched in a place which would contact

sample water. In handling the bags, care was taken to touch only that portion of the liner which overlapped the outside of the collector.

The deionized water, HCl, and spectrophotometer were checked simply by measuring acidified samples of deionized water. These samples did not contain significant amounts of the trace metals.

As discussed earlier, the Water Survey installed four sequential samplers to measure the time variability of the rainout of indium and trace metals. Because of the complexity of cleaning the samplers, the standard procedure was to uncover these collectors immediately before a rain and re-cover them just after the rain. Although these samplers were serviced several times, no water was collected because of the sparsity of the rainfall. A series of sequential samples were collected, however, on the evening of 14 June at the Clinton laboratory by the Michigan personnel. The collection was not made during a tracer release, but the samples were suitable for chemical analysis.

The results of the analyses of trace metals collected 14 June are shown in figure 10. All five of the curves bear some resemblance to each other, each decreasing by a factor of approximately 2.5 during a 2.7 hour period. The concentrations of sodium, potassium, and zinc, however, increased on the sixth sample to values as high as at first, whereas calcium and magnesium did not. Sodium and zinc also reached a relative minimum at 2131 CDT while the other metals did not, but in general the curves are similar. The values of copper are not included in the figure because most of the concentration was below the instrument sensitivity. There are no radar data for this rain, and the rainfall rate has not yet been computed by the Michigan scientists. An examination of weather maps and the raingage charts from gages in the Clinton area indicated that the rain was relatively steady and associated with a low pressure wave moving along a stationary front.

The time variation of the rainout of trace metals as discussed above is not unlike the rainout of gross beta radioactivity described by Huff (1965). Huff reported that 35% of the convective storms investigated exhibited a beta activity curve which decreased in the first portion of the storm, later reached a minimum, and then increased again toward the end of the storm. The rainout patterns of sodium, potassium, and zinc are of this general shape, which Huff labeled type A. The calcium and magnesium curves resemble Huff's type C curves, which have relative maxima at the beginning and center of the storm followed by a considerable decrease toward the end. Only 9% of Huff's storms were of type C. Since the rainfall records are not yet available, it is not known whether the sampling encompassed the entire duration of the rainfall.

A study of the areal variability of the rainout of metals was conducted using the total precipitation samplers. As discussed previously, the rainfall on the network was very light, and the five rains sampled were either during non-operational periods or at night, making immediate collection of the rainwater impossible. The amount of dry fallout in the collectors and in the data sample is therefore probably appreciable.

The areal distributions of the rainout of six trace metals from rainfall on 30-31 May are presented in figure 11. These water samples were collected

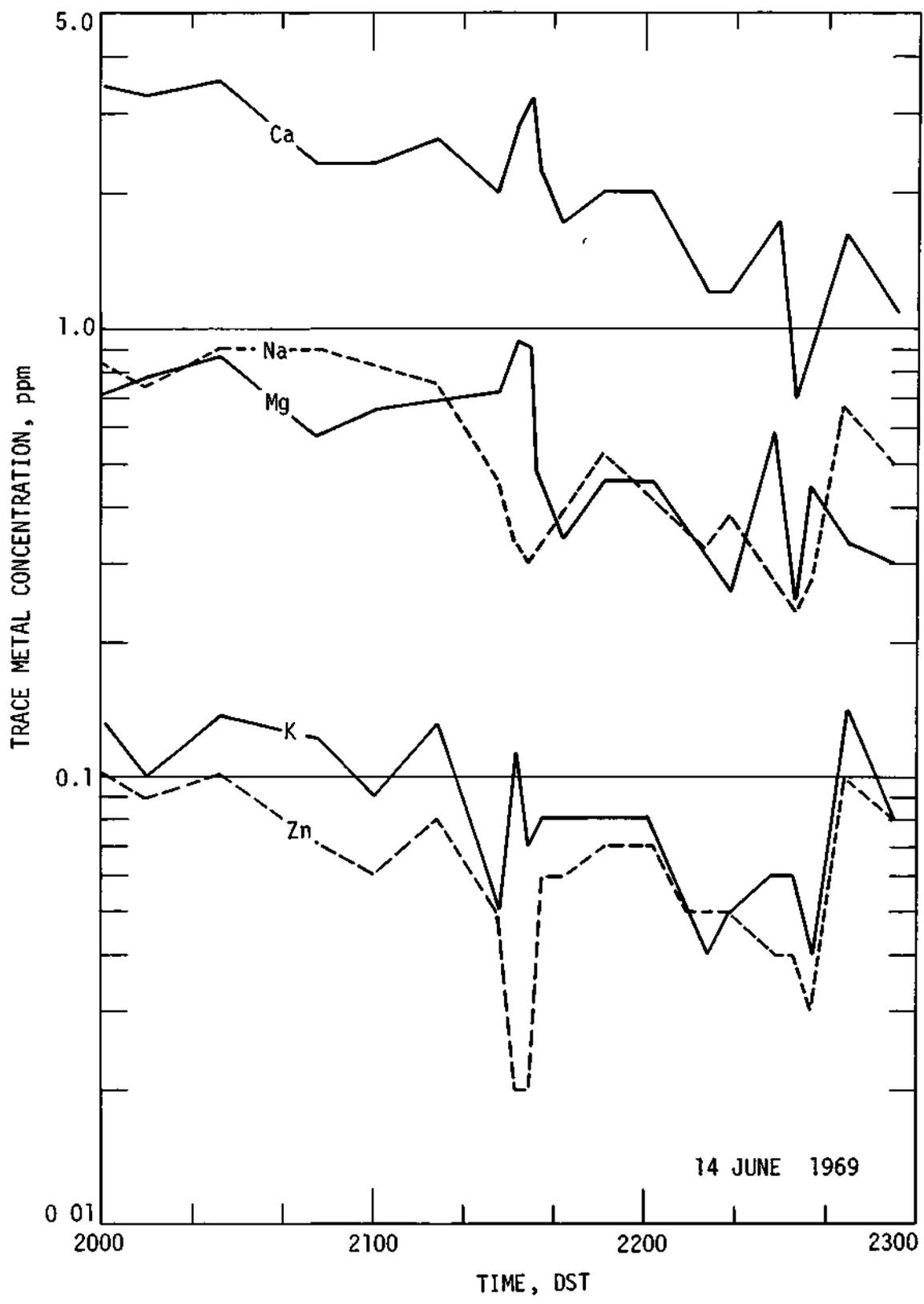


Figure 10 Time variability of trace metal concentrations in precipitation

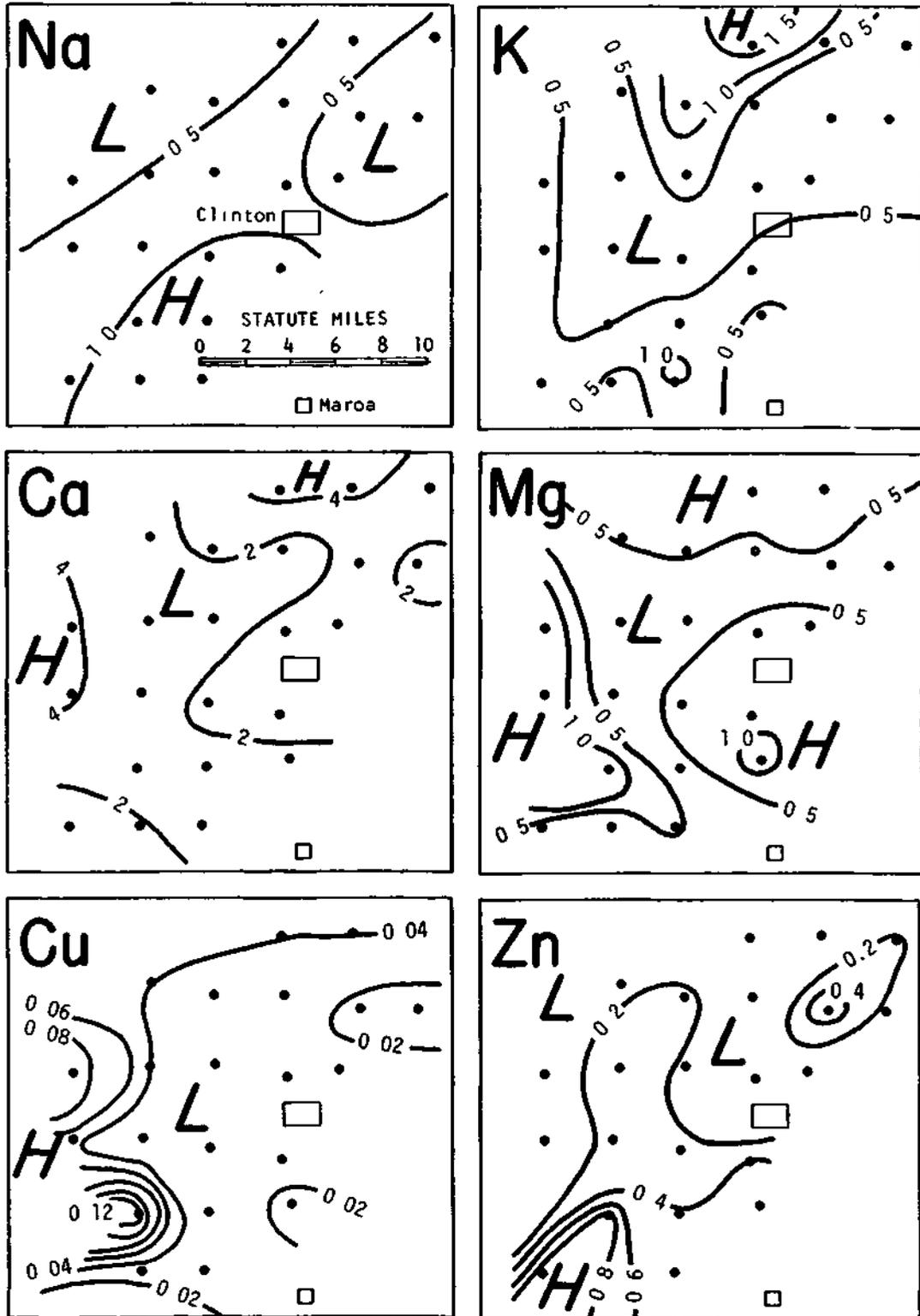


Figure 11. Areal variability of trace metal concentrations (in ppm) in precipitation on 30-31 May 1969

for background data by the Michigan personnel and represent only a portion of the network rainfall. The collector liners were installed on 30 May and picked up on 1 June.

Radar data are not available for the rain period, and the raingage data have not been processed yet. The synoptic analysis indicated that the rain resulted from warmer, moist air over-running a stationary front in southern Illinois. It is quite apparent from figure 11 that the distributions show only minor similarities. The calcium, copper, and magnesium exhibit highs of concentration on the western edge of the collecting area, and most of the metals had low concentrations in the west-central or northwest corner. The areal variability and lack of correlation between the concentration plots of the different metals may be real, or it may be somewhat distorted by possibly random distribution of dry fallout in the collectors.

Several preliminary tests have been conducted to evaluate the effect of dry fallout on the concentration of chemicals in rain samples. These tests consisted of leaving collectors exposed for various lengths of time at different locations, and then analyzing the contents of the polyethylene bags. In one test rain was allowed to fall into the liner, but in most cases dry bags were collected to measure the dry fallout. In order to determine the amount of dry fallout on the plastic liners, the bags were washed with acidified water and the contents of the solution analyzed by atomic absorption. In the study presented here, the samples were collected by the Michigan personnel, and the liners were washed with 1 liter of deionized water acidified with 50 ml of 6 N HCl.

During 9 to 14 June seven collectors with liners were placed successively in the Clinton test area. At the end of the period, the seven sample liners were collected, resulting in a series of bags exposed for increasingly longer times. The resulting concentrations of the trace metals with varying length of collector exposure are given in table 2.

TABLE 2

Trace metal concentrations versus sampler exposure time

<u>Time, hours</u>	<u>Metal concentrations</u>					
	Na	K	Ca	Mg	Cu	Zn
38.5	.41	.14	1.5	.33	.08	.05
48.7	.50	.33	2.1	.53	.08	.08
61.2	.40	.42	2.8	.76	.09	.07
67.5	.45	.27	2.6	.84	.08	.08
73.5	1.01	.33	3.8	.70	.08	.07
96.5	.40	.84	2.5	.73	.10	.07
120.5	.30	.56	3.2	.93	.11	.09

All the metals except sodium and copper show a general trend of increasing concentration with time, but there is considerable variability in the data. The sodium and copper concentrations even appear to be independent of the exposure time. The reason that the sodium behaved in this manner is not clear, as it was found to decrease with time in at least one other test. The lack of a relationship between concentration and exposure time for copper may be because of the sometimes high background of copper (about 0.05 ppm) in the deionized water used to wash the bags.

The dry deposition data were used to determine an approximate deposition rate for each of the metals. These depositions are expressed in terms of the increase in concentration per 24 hours for one liter of water in a collector, i.e., ppm/24 hr/liter. These unconventional units facilitated rapid comparison with collected water sample concentrations, even though they are not directly comparable unless the actual water samples collected were also one liter. However, since the volume of water collected for the field sample has not yet been calculated, the one-liter estimate, which is equivalent to 0.17 inch of rain, is a convenient approximation.

The dry fallout measurements were compared qualitatively with the lowest concentrations of trace metals found for each of the network rainfalls collected. It was determined that the 24-hour dry fallout in this particular test was greater, in general, than the lowest wet fallout measurements for sodium and copper; was approximately equal in magnitude for magnesium, potassium, and zinc; and was at least an order of magnitude less than calcium. This analysis indicates that, except in the case of calcium, the 24-hour dry fallout is sufficiently large to mask the lower concentration precipitation samples.

A more complete examination of all the dry and wet fallout data, with the actual rainfall amounts, will be necessary before the problem can be fully assessed, but it appears that the operational technique of sample collection must be considerably improved.

It may be possible, however, to minimize the dry fallout problem by limiting the exposure time of collectors to a few hours, as was attempted without success during the 1969 season, and by selecting specific metals with a low background such as calcium.

Raindrop Size Distributions

There were basically three reasons for collecting raindrop spectra data during the field project period. If information concerning circulation in a large convective cell (cumulonimbus) were to be gained from the presence of indium reaching the ground in the rainwater, then it would be necessary to know the scavenging efficiency of the indium particles by the precipitation particles. This required the raindrop size distributions as determined from the raindrop camera. Also, a comparison between the University of Michigan raindrop spectrometer and the State Water Survey raindrop camera was to be performed during the field project period.

Both instruments were operated simultaneously however, during only one storm (14 June 1969), the drop camera data for this storm have been reduced to drop size spectra, but the spectrometer data have not at the writing of this report. The intent was to obtain comparative data for several storms, but very few storms occurred. During the first phase, measurable rain was recorded at Clinton on 21 May, 25 May, 1 June, 4 June, and 14 June. The drop camera was operated on the last three dates, and the spectrometer on 1 June and 14 June. Unfortunately, on 1 June no measurable rain occurred during the period of drop camera operation. Rain was recorded, however, shortly after the camera ran out of film. The spectrometer was activated during this rain period. Since the spectrometer was operated during the first phase only, 14 June was the last day of data collection for that instrument. Finally, additional raindrop data for Illinois storms occurring in June were desired to increase the number of samples available for this period. Knowledge of drop distributions are important for determining rainfall rate (R) - radar reflectivity (Z) relationships for this region (Jones, 1956).

Briefly, the raindrop camera takes seven pictures, approximately 1 1/2 seconds apart, at the beginning of a 60-second period. The camera then becomes inactive for the remainder of the minute. Each frame represents a volume of about $1/7 \text{ m}^3$, so one minute of data represents a sampled volume of one cubic meter. The raindrops are then measured individually from a projected image of the film, and their number and size are punched onto data cards (Mueller and Sims, 1967).

Drop-size data have been analyzed for three storms, occurring on 4 June, 14 June, and 21 July 1969. The 4 June and 21 July storms were associated with thunderstorms, whereas the 14 June storm was a more uniform or steady rain. To indicate the relative sizes of the raindrops in these rains, the average size distributions as well as the Z - R relationships may be examined. Average drop distributions of similar rainfall rates for the three rain periods are shown in figure 12. (N_T is the average number of drops for the number of cubic meter samples, N_S , used in the analysis). The distributions are quite similar with the largest number of drops occurring at 1.5-1.6 mm. The Z - R equations for the storms are:

4 June.	Z =	$763R^{1.28}$	25	(Total number of cubic-meter samples taken during storm)
14 June:	Z =	$407R^{1.24}$	66	
21 July	Z =	$506R^{1.48}$	52	

The 14 June rain (steady rain) had the smallest Z value although it had the largest R. This indicates that there was a larger number of relatively small drops and fewer large drops than in the other two storms. This is expected since thunderstorm rains are generally composed of large drops for similar rainfall rates than those in non-thunderstorm rains (Stout and Mueller, 1968).

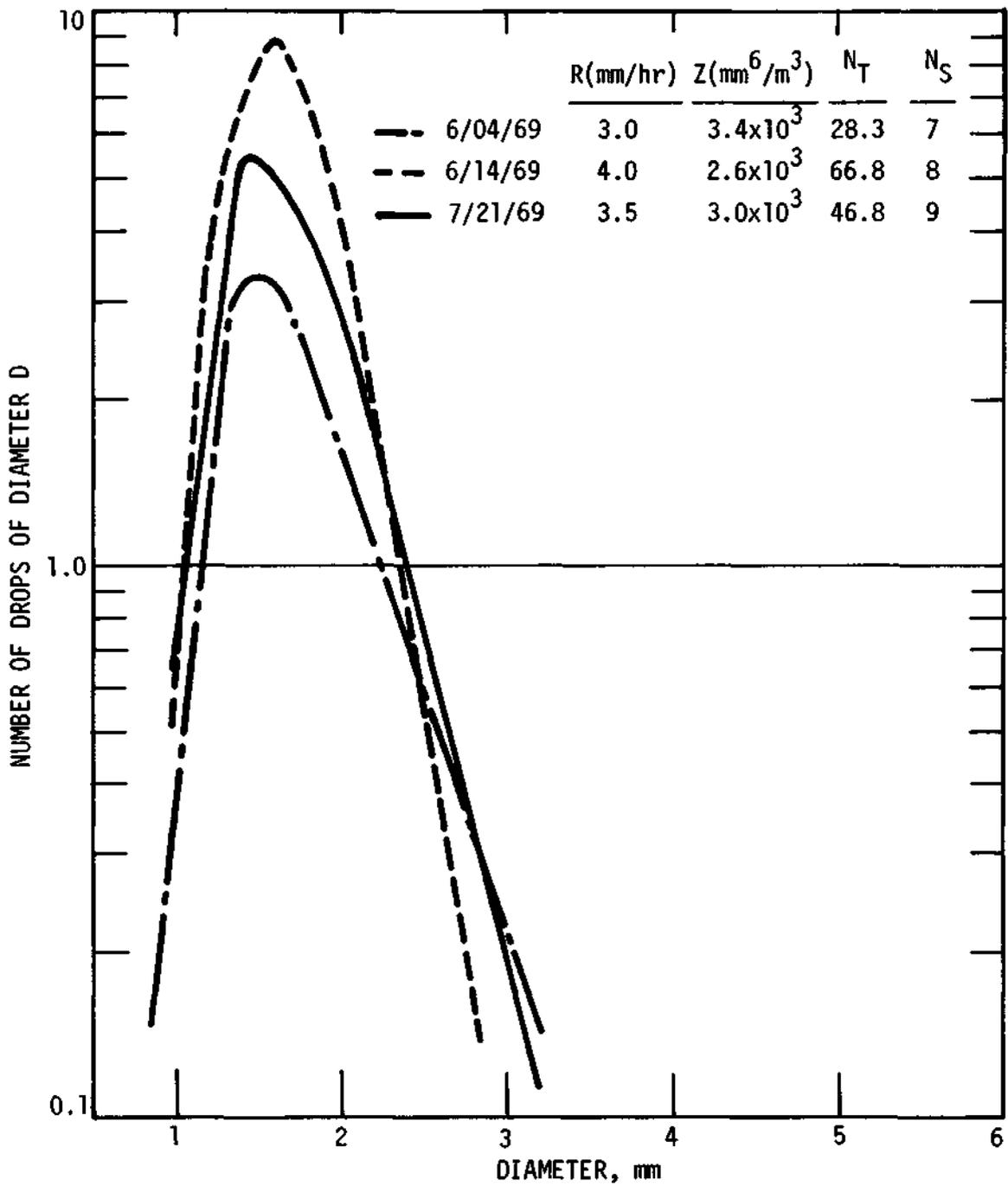


Figure 12. Average raindrop size distributions for similar rainfall rates from three separate rain periods occurring at Clinton, Illinois, in the summer of 1969

Summary

The goals of Project ITREX were not realized in 1969 because of abnormal weather conditions with much below normal numbers of daytime thunderstorms and precipitation amounts. Nevertheless, the desired staff was gathered, and essential equipment was installed and operated on several potential "go" days. These 1969 operations indicated that the project plans and data collection system were generally satisfactory to achieve the project goals if the weather had been near normal. One reasonably successful tracer experiment was accomplished on 29 May, but the rainwater analysis has not been completed at this time.

LABORATORY INVESTIGATION OF SUBMICRON
PARTICLE SCAVENGING BY RAIN

Introduction

A study of the removal of radioactive material from the atmosphere necessarily includes a determination of the physical processes responsible for scavenging the material both within and beneath the cloud. Experiments have been conducted in the laboratory and in the field, and theoretical investigations have been made in an effort to determine the processes dominating the scavenging mechanism. Only a limited amount of experimental research has been done concerning the scavenging of particulates under reasonably controlled conditions. However, this research has been productive and has demonstrated that particles greater than one micron (μm) in diameter are efficiently scavenged (Engelmann et al. , 1966).

The scavenging of submicron particles is, however, a relatively unexplored area. Approximately 80 to 90% of the particulate mass in the continental atmosphere is in the 0.1 to 20 μm diameter size range (Junge, 1963). About half of this mass and an even greater percentage of the radioactive debris is in the size range smaller than one micron diameter. Lockhart, Patterson, and Saunders (1965) made filter-pack measurements of the radioactive particulate concentration in the air in four size ranges down to 0.15 μm . They found that 60 to 70% of the activity was of a size $1.1 \mu\text{m} > D > 0.15 \mu\text{m}$, and most of this material in the $1.1 \mu\text{m} > D > 0.3 \mu\text{m}$ size range. Although the ratio of the activity of different sizes varied considerably, the data indicated that over 50% of the radioactive material is carried in the first decade of the submicron particle size range.

Aerosols are removed from the air in precipitation by three different mechanisms: removal of particles by the impaction of raindrops within and beneath the cloud, the consumption of particles serving as condensation nuclei, and the attachment of particles to cloud droplets and raindrops by Brownian motion. It has been calculated theoretically by Langmuir and Blodgett (1945) and shown experimentally (Engelmann et al. , 1966) that impaction is an efficient process for particles of a few microns or greater in size. However, Langmuir's work indicates that the washout coefficient for submicron particles is effectively zero. Engelmann's experimental data, if extrapolated to submicron particle sizes, would show essentially the same result. It has yet to be determined experimentally, however, what the collection efficiency is for submicron aerosols. The presence of charges on the drops and particles and the ambient electric field may aid the scavenging of submicron particles.

Because of the importance of delineating the mechanisms responsible for the removal of submicron particulates from the atmosphere, a laboratory experiment was begun at the Water Survey in 1968 to determine the collection efficiency of artificially produced raindrops for submicron particles in the size range from 1.0 to 0.1 μm diameter. Reported here are the various techniques that have been tried and the collection efficiencies of drops of 1.42 mm and 2.88 mm radius for spores of 1 μm diameter.

Analytical Technique

The technique used for determining the scavenging efficiency of raindrops is as follows. Artificial raindrops are generated in a drop acceleration tower. After reaching terminal velocity, the drops pass through an aerosol chamber, scavenging a portion of the particles therein, and are collected in a funnel at the chamber bottom.

Three different types of submicron particles were used in the scavenging studies. These were submicron polystyrene spheres, Escherichia coli, and Bacillus subtilis spores. The experimental procedure, and aerosol generation and sampling techniques of each are described below.

In generating the aerosol of polystyrene spheres (PS), the prime concern was the production of as high a number-concentration as possible because of the difficulty in detecting the spheres in the drop water. The apparatus developed for generating the PS aerosol is shown in figure 13. Compressed air enters the system from a pressure regulator and passes through a flow meter and into a bubbler, impactor, Millipore filter, and then into the nebulizer.

The PS suspension is nebulized and the resultant liquid aerosol passes through the impactor and settling chamber. The aerosol cloud then moves through the heated drying tube and over too 500 pc radioactive probes to neutralize electrical charge, and is then ducted into the scavenging chamber. When the generator is charged with a PS suspension of 0.033% PS of one micron diameter, the aerosol concentration in the experimental chamber will reach approximately 10^3 PS/cm³ with the number of doublets less than 2 or 3%. The concentration of PS in the chamber is determined by optically counting microscope slides of the aerosol collected with an electrostatic precipitator.

Once drops had fallen through the aerosol it was necessary to determine the PS concentration of the collected water. Two methods of particle counting were tried. The first involved applying one or more drops of the collected water on a microscope slide, allowing the drop to evaporate and then attempting to count the particles optically. As the drops dried, most of the material in the water was deposited in a ring at the edge of the drop. Unfortunately, there was always enough foreign matter in the water to encrust the PS sufficiently to make the particle counting uncertain, if not impossible. Despite attempts using doubly distilled, Millipore filtered water, the evaporating drop always left a "ring" partially encrusting the PS particles.

The second method tried in measuring the concentration of PS in the water was to filter the drop water and count the particulates on the filter. This technique is also extremely difficult because the concentration of PS in the drop water is so low that the small number of PS on the filter are difficult to count accurately. In the future the counting of the PS will be attempted with the use of a specially constructed filter holder which will pass the drop water through a very small portion of the filter to increase the area number-density of the PS.

The difficulty in detecting extremely small concentrations of particulates in suspensions led to the use of microbes as the scavenging aerosol. Microbes

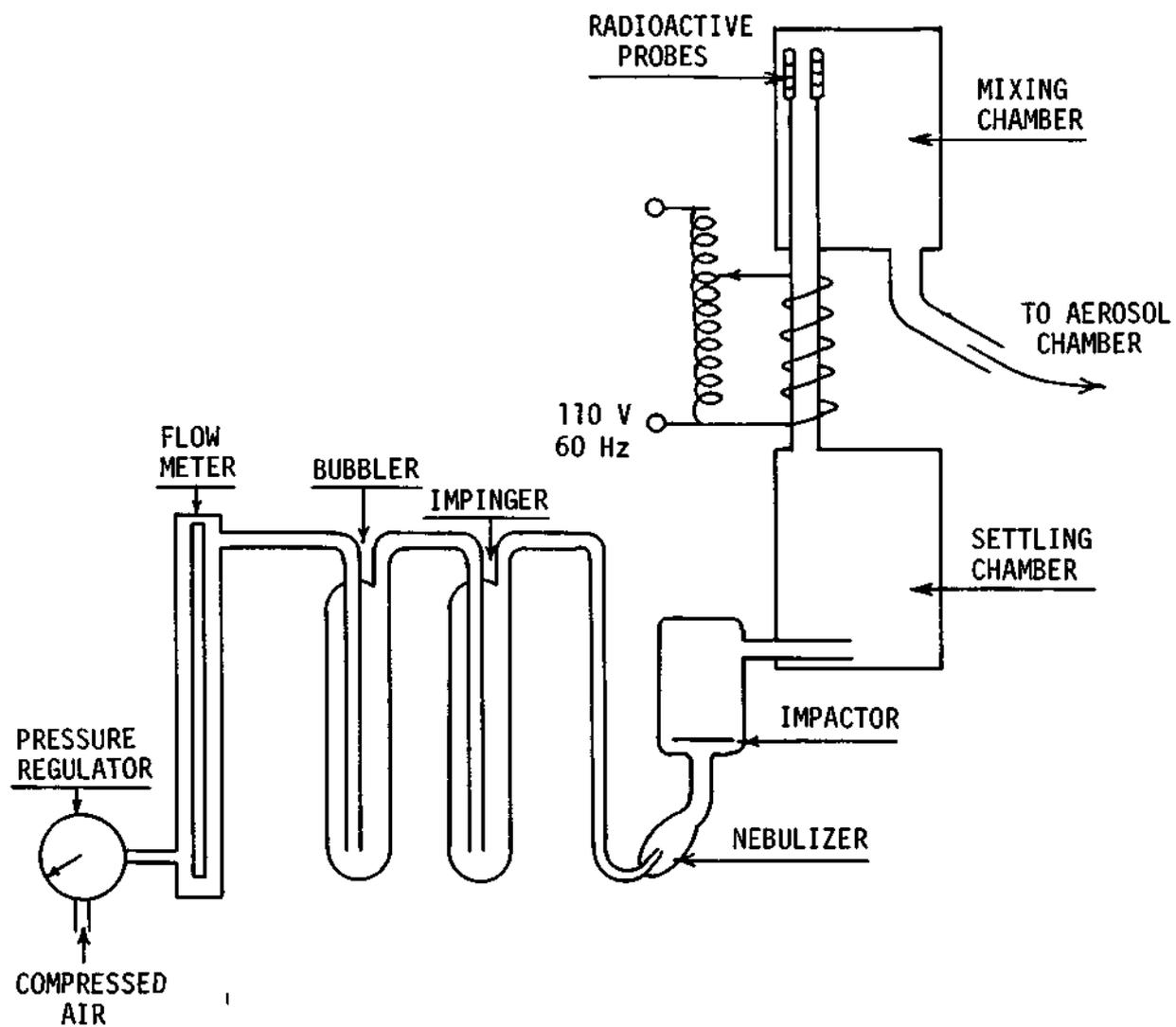


Figure 13 Aerosol generator for producing polystyrene sphere suspension

have the advantage of being detected individually. The microbes initially used in the scavenging studies were a combination of Aerobacter aerogenes and Escherichia coli. The aerogenes are rod shaped, being 0.5-0.8 μm in diameter and 1-2 μm in length.

The microbes were utilized as follows. A culture of them was grown, isolated, diluted, and sprayed into the aerosol scavenging chamber in a manner such as to impact on the wall of the chamber while the smaller drops evaporated leaving the spores suspended. A fan in the chamber was used to mix the aerosol thoroughly before passing the water through the chamber. A small narrow-beam spotlight made it possible to check for complete evaporation of the water from the aerosol and to inspect the falling drops.

The B. subtilis spore concentration was determined by passing a measured volume of air through three midget impingers. Three were used because the collection efficiency of each is approximately 95%. At least two impingers are thus needed to calculate the impinger collection efficiency, and to correct for it.

Two different types of droplet generators can be used depending on the size of drops desired. Between diameters of 1.5 and 6.0 mm the drops can be created by dripping water from the appropriate size tube or needle. Smaller sized drops, down to 0.5 mm, are produced by passing a stream of water through a small glass capillary which is vibrated laterally by a piezoelectric strip. With the proper adjustment of pressure, frequency, and capillary size, the desired diameter of drops' can be produced. The larger drops are produced at a rate that can be conveniently counted manually. The smaller drops from the vibration capillary are produced at the frequency of the vibrating strip and are spaced at about 2 diameters.

If the drops are allowed to fall down the drop shaft directly from the generator, many coalescences occur as revealed by a distinct sparkle from the resultant drop. The drops then fan out rapidly into a cone before they reach the bottom of the drop acceleration chamber. These problems were nearly eliminated by placing a small blower just below the drop needle. This introduces a random motion into the stream of drops, prohibiting them from all following the same path and thereby reducing coalescences to nearly zero. Most of the drops are then confined to a cylinder of, for instance, about 7 or 8 mm diameter for 0.5 mm diameter drops after falling 4.4 m.

One of the problems in the scavenging experiment is to measure the dry fallout of spores into the collector funnel. The technique used involved a control funnel to determine the amount of dry fallout in addition to a funnel to collect the water drops. The two funnels were exposed for the same length of time, after which the control funnel was washed to determine the amount of dry fallout. The dry fallout was then subtracted from the total washout in the drop funnel. Several techniques were tried for covering and uncovering the funnels. The workable technique was to enclose the funnels in metal cylinders with lids which were removed at the beginning of the experiment.

The sequence of events for an experiment was as follows. All of the equipment which comes into contact with the spores was autoclaved to remove contamination. This included the water, aerosol generator, collector funnels, and impingers. The aerosol was injected into the chamber and circulated with

a fan. Additional air was added after the aerosol droplets appeared to evaporate to insure that a saturated condition did not exist in the chamber. The fan was then operated another 30 seconds as the impinger sample of the aerosol was taken. The fan was stopped and the turbulence in the chamber was allowed to dissipate for approximately a minute. The cover was then removed from the aerosol chamber to allow the drops to enter, and the lids were removed simultaneously from the drop and control funnels. The drops passed into the chamber for a period of 30 to 60 seconds, after which the two funnels were quickly removed from the chamber, fanned to remove the airborne aerosol from them, and re-covered.

It should be emphasized that both the large and small drops spread out sufficiently to nearly fill the 10 cm diameter opening to the aerosol chamber. The drops were not directly following one another, and the aerodynamic flow around each drop should be independent of the others.

The water from the impingers, drop funnel, and control funnel was then analyzed. One ml of water was removed from each sample and divided evenly between 10 petri dishes, that is, 0.1 ml per dish. Dilutions were usually made of the drop water and first impinger because of their occasional high concentrations. The petri dishes were then incubated for 16 to 48 hours after which each plate was counted.

Results

The results of the *B. subtilis* spore scavenging experiments to date are presented in table 3, which gives the aerosol concentrations as well as the total wet and dry depositions. The collection efficiency (C.E.) of the raindrops for the spores is defined as the number of spores scavenged by the water drops divided by the number of spores in the volume of air swept out by the drops, times 100.

The average C.E. for both the 2.8 and 5.8 mm diameter drops is close to unity, but there is considerable variation about the averages. Only one experiment has been conducted to date with the 0.54 mm diameter drop, and the extremely low C.E. calculation should be considered questionable until verified by further experiments.

The relatively great variation in the C.E.'s is attributed to random errors in the procedure and is compounded considerably by the fact that the dry fallout is a significant portion of the total. This effect can be reduced by minimizing the exposure time of the funnels and maximizing the droplet flow rate. Changes in the experimental procedures have reduced the funnel exposure time and the amount of dry fallout in the most recent experiments using 0.54 mm drops. It is believed that variation in the experimental values of C.E. as determined for the 2.8 and 5.8 mm diameter drops can also be reduced by decreasing the exposure time and increasing the water flow rate.

TABLE 3

Results of B. subtilis spore scavenging experiments

<u>Drop diameter, mm</u>	<u>Aerosol concentration</u>	<u>Dry deposition, % of total</u>	<u>% Collection efficiency</u>
5.76	11.8	66	.63
5.76	53.9	31	.69
5.76	71.6	57	1.42
5.76	65.7	38	1.02
			<hr/>
		Average	.94
2.84	350.0	52	1.10
2.84	350.0	46	1.73
2.84	90.6	52	.91
2.84	90.6	75	.46
			<hr/>
		Average	1.05
0.54	274.0	17	.03*

Result questionable

REFERENCES

- Browning, K. A., and F. H. Ludlam, 1962. Airflow in convective storms. Quart. J. Roy. Meteorol. Soc., 88, pp. 117-135.
- Dingle, A. N., and H. F. Schulte, 1962. A research instrument for the study of raindrop-size spectra. J. Appl. Meteorol., 1, pp. 48-59.
- Engelmann, R. J., R. W. Perkins, D. I. Hagen, and W. A. Haller, 1966. Washout Coefficients for Selected Gases and Particulates. BNWL-SA-657, Battelle-Northwest, Richland, Washington, 22 pp.
- Gatz, D. F., A. N. Dingle, and J. W. Winchester, 1969. Detection of indium as an atmospheric tracer. J. Appl. Meteorol., 8, pp. 229-240.
- Huff, F. A., 1964. Study of Rainout of Radioactivity in Illinois. Second Progress Report, AEC Contract AT(11-1)-1199, Illinois State Water Survey, Urbana, 61 pp.
- Huff, F. A., 1965. Study of Rainout of Radioactivity in Illinois. Third Progress Report, AEC Contract AT(11-1)-1199, Illinois State Water Survey, Urbana, 66 pp.
- Huff, F. A., and W. E. Bradley, 1965. Study of Rainout of Radioactivity in Illinois. Fourth Progress Report, AEC Contract AT(11-1)-1199, Illinois State Water Survey, Urbana, 20 pp.
- Jones, D. M. A., 1956. Rainfall Drop Size-Distribution and Radar Reflectivity. Research Report 6, U. S. Army Contract DA-36-039 SC-64723, Illinois State Water Survey, Urbana, 20 pp.
- Jones, D. M. A., and L. A. Dean, 1953. A Raindrop Camera. Research Report 3, U. S. Army Contract DA-36-039 SC-42416, Illinois State Water Survey, Urbana, 19 pp.
- Junge, C. E., 1963. Air Chemistry and Radioactivity. International Geophysics Series, Academic Press, New York, 188 pp.
- Langmuir, I., and K. B. Blodgett, 1945. Mathematical Investigation of Water Droplet Trajectories. Report No. RL-225, G. E. Research Lab., 66 pp.
- Lockhart, L. B., Jr., R. L. Patterson, Jr., and A. W. Saunders, Jr., 1965. Filter Pack Techniques for Classifying Radioactive Aerosols by Particle Size, Part 3. NRL Report 6305, U. S. Naval Research Lab., Washington, D. C., 18 pp.
- Mueller, E. A., and A. L. Sims, 1967. Raindrop Distributions at Island Beach, New Jersey. Research Report 2, U. S. Army Contract DA-28-043 AMC-02071(E), Illinois State Water Survey, Urbana, 10 8 pp.

Stout, G. E., and E. A. Mueller, 1968. Survey of relationships between rainfall rate and radar reflectivity in the measurement of precipitation. J. Appl. Meteorol. , 7, pp. 465-474.

Sutherland, J. L., 1969. Project ITREX. Final Report, Sub-contract to Illinois State Water Survey, Weather Science, Inc., Norman, Oklahoma, 19 pp.

APPENDIX A

Summary of Daily Weather and Activities,
First Phase 15 May - 14 June 1969

15 May

This was the first day of operations for ITREX. The aircraft arrived the previous evening, and nearly all the network instrumentation was complete. Southwesterly flow prevailed over Illinois as a result of high pressure to the east, so no thunderstorm missions were planned. An orientation flight over the Central Illinois Network (CIN) was carried out to familiarize the aircraft crew with the area, test all communications, and make a final check of the automatic meteorological system (AMS) on the airplane. All systems functioned properly.

16 May

A cold front approached from the northwest, but was too distant to affect the Illinois weather. Seventeen polyethylene bags were placed in baskets on the CIN in anticipation of precipitation, and the raindrop camera was moved to Clinton and installation was begun at the laboratory.

17 May ("Go" day)

Eighteen additional bags were placed on the network prior to passage of the cold front in the afternoon. Showers and thunderstorms were expected to develop, and the aircraft was vectored to a line of weak echoes on the front. They weakened further however, and no tracer was released. The airplane flew for 90 minutes, and several hours of radar operation were logged.

18 May

Shallow cloudiness resulted from warm air over-running the cold front, which had moved south of Illinois. No significant weather was produced by the cloudiness however.

19 May

The bags deployed two days previously were collected from the network, and chemical analysis of the shower-produced rainwater was begun. A weak high pressure ridge moved through Illinois, and a cold front approached from the northwest. Installation of 11 microbarographs was begun on the CIN during this fair-weather day.

20 May

The cold front passed through the area during the afternoon, but with no accompanying precipitation. Installation and testing of the raindrop camera was continued at Clinton.

21 May

The WSI flight crew drove to Clinton to visit the Michigan laboratory and discuss the tracer experiments with the Michigan scientists. A large high pressure area moved across the Great Lakes producing fair skies. Installation of the microbarographs was completed, and an automatic weather station was installed on the CIN.

22 May

High pressure continued to dominate the northern portion of Illinois, while a small wave moved eastward on a stationary front in the southern part of the state. Cloudy and cool weather prevailed over central Illinois, with a few periods of light rain. Seventy-one bags were placed on the CIN to collect the rain, and 13 were recovered before nightfall. The plane made a 30-minute flight to check an AMS problem.

23 May

No precipitation was noted as high pressure continued over the area.

24 May

A cold front approached from the northwest, and the high pressure system continued to move east.

25 May

The cold front passed through the area during the afternoon, with the only precipitation being widely scattered showers. A one-hour flight was made to probe cumulus congestus clouds and to measure meteorological parameters inside and in the vicinity of the clouds.

26 May

The cold front which moved through on the 25th moved slowly northward but produced no significant weather. A computer program to compute air and water vapor fluxes from aircraft data was prepared.

27 May

High pressure over the Northeast caused southerly flow over Illinois. The air was too dry for any precipitation to form.

28 May

The high pressure center drifted into Virginia, and its effects were still felt in Illinois. A cold front extended from the Dakotas into Colorado and Utah, and showed promise of creating precipitation on the 29th or 30th. Thirty bags were brought in from the field, and were examined for dry fallout since no rain had fallen into them.

29 May ("Go" day)

In anticipation of afternoon rain, 87 polyethylene bags were placed on the network. A 90-minute mission was flown beginning at 1800 CDT, and 14 indium chloride flares were burned beneath a moderate rainshower. A cold front passed through the area during the early evening; showers remained until after midnight. A total of 175 minutes of CPS-9 film was exposed during the early evening. Forty-one of the bags deployed earlier were collected.

30 May

Fifty bags were installed on the CIN, and 68 were removed. The front became stationary in southern Illinois but caused no weather of significance in the central portion of the state.

31 May ("Go" day)

The stationary front became warm and moved north through the CIN in the afternoon. Thunderstorms formed over Champaign and moved eastward; however none formed farther west. No flight was made because it was evident that no activity would develop behind the squall line, although radar surveillance did continue for several hours. Nineteen sampling bags were collected, with rainwater from the thunderstorms.

1 June ("Go" day)

In advance of a cold front were showers during the morning. The aircraft investigated these but found no cloud bases below 10,000 ft and no rain heavier than light. Total flight time was 64 minutes, and almost 2 hours of radar film was collected.

2 June

Weak high pressure over the southern plains moved southeastward during the day, bringing partly cloudy skies to central Illinois. A total of 75 bags was brought in from the field, and 79 new ones were placed in collecting baskets. A problem in the aircraft's AMS was investigated also.

3 June

The AMS problem was found and a replacement part ordered. Skies remained partly cloudy, under the influence of high pressure centered over Arkansas. Twenty bags were exchanged for 20 new ones.

4 June

A cold front moved from Iowa into Illinois, but caused no precipitation over the CIN. Indications were that the front would become stationary in central Illinois and might produce showers or thunderstorms on the 5th. In view of this, 88 polyethylene bags were removed from the network and 92 were installed.

5 June ("Go" day)

The front did stall in central Illinois, and a wave began moving along it from Nebraska. Twenty-six additional bags were placed on the CIN. No flights were made since no storms developed, although 4 hours of CPS-9 radar film was collected.

6 June ("Go" day)

During the early afternoon radar echoes began to form northwest of the CIN, so 100 bags were exchanged for 100 clean ones on the network. The aircraft took off about 1415 CDT to investigate the showers, but found none of consequence. A squall line formed too far south of the CIN to use for tracer study, but the aircraft crew flew beneath it to investigate updrafts and fluxes. It dissipated shortly after their arrival in the vicinity. Approximately 3.4 hours of flight time was logged on this mission.

7 June ("Go" day)

The front remained quasi-stationary north of Champaign, with shower and thunderstorm activity expected south of the front. None developed, however, so no missions were flown.

8 June ("Go" day)

An early morning forecast indicated that the front would become cold and move through the area during the afternoon. Forty-six bags were changed on the network while a squall line approached from the northwest. The aircraft flew to the leading edge of the line, just north of Bloomington, and at that point the storms began to dissipate. No organized updrafts were found and after almost 6 hours of flying the aircraft returned to Champaign. Over 6 hours of radar film was taken.

9 June

A large high pressure area moved eastward across the Great Lakes. Because of the shower activity on the previous day, 44 bags were changed on the network. All systems remained ready for thunderstorms. An open house of the aircraft was conducted for interested persons from the University of Illinois Institute of Aviation.

10 June

High pressure continued to dominate the Illinois weather. Since no thunderstorms were forecast, the Michigan scientists traveled from Clinton to Champaign to inspect the airplane and meet with other project personnel.

11 June ("Go" day)

A quasi-stationary front from Wisconsin to Kansas drifted slowly southeastward, and a small low in northern Texas moved along the front. Severe weather was present along the northern portion of the front, and scattered thunderstorms were forecast for northern Illinois. None formed or moved into the central part of the state, so no flights were made. Ninety-two polyethylene bags were replaced over the network.

12 June ("Go" day)

A cold front extended from Wisconsin into Nebraska and Wyoming during the morning, and overtook the stationary front during the afternoon. Severe thunderstorms were forecast for central Illinois, but formed instead in western Indiana about noon. No activity occurred behind this squall line, but radar surveillance was continued until almost 1900 CDT. A 2-hour flight was made, beginning at about 1630 CDT. A total of 120 Bags were removed from the CIN and 114 were installed.

13 June

High pressure pushed the cold front south of Illinois. At the daily briefing it was decided to end this phase of ITREX on 14 June if no weather conducive to thunderstorms was forecast on the 14th.

14 June

Scattered showers, but no thunderstorms, were forecast for the evening. The longer-range outlook did not include any precipitation, so the aircraft was released from duty and the daily weather briefings were cancelled.

APPENDIX B

Summary of Weather and Daily Activities,
Second Phase 23 July - 8 August 1969

23 July

A cold front approached from the northwest, but produced no precipitation over central Illinois. The day was spent planning the tracer flights and working on the aircraft's meteorological instrumentation.

24 July

Several showers developed over the CIN during mid-morning, but dissipated with passage of a cold front about noon. A flight of approximately 90 minutes was made in the afternoon. The purpose was to check ground-air communications, deploy air samplers over the network, and release tracer gas as would be done on a day with thunderstorms. All systems functioned normally with the exception of the gas release mechanism. Ice formed on the nozzle, restricting gas flow from the tanks into the atmosphere.

25 July

High pressure centered over northern Missouri moved east during the day. The airplane was flown to St. Louis for repairs necessary on the distance measuring equipment.

26 July

A weak stationary front existed in southern Illinois; all of the state's precipitation was south of it. A cold front in the Dakotas moved southeast during the day giving an indication of rain on the 27th.

27 July

The cold front passed through central Illinois during the late morning, but precipitation failed to materialize. High pressure centered in Wyoming moved eastward.

28 July

High pressure encompassed most of the United States west of the Ohio Valley, with very little precipitation west of Indiana. The upper air ridge

showed no sign of weakening. Low ceilings were prevalent over Illinois as cold, unstable air flowed from the north behind the front.

29 July

The high pressure ridge extended from Minnesota into Oklahoma, and drifted only slowly eastward. Skies were partly cloudy, with little more than cumulus humilis being present.

30 July

A small trough aloft began to move through the northern part of the United States, coupled with a surface cold front through the Dakotas and Wyoming. High pressure centered over Illinois extended from the eastern seaboard to the cold front.

31 July ("Go" day)

The cold front had moved into central Wisconsin and central Iowa by morning; its forecast time of passage at Champaign was after midnight on the 1st. Thunderstorms were expected to develop in northern Illinois during the early evening in advance of the front. The air sampling network was readied but not used, as no storms developed. No flights were made, but 5 hours of CPS-9 film was collected.

1 August

The cold front passed through Champaign about sunrise, and continued southeast. High pressure began to fill behind the front, with a center in the Dakotas. A flight was made to northwestern Indiana to sample air in and around cumulus clouds downwind of the Chicago-Gary industrial complex.

2 August

The cold front moved over the Appalachians, while high pressure centered over Iowa remained stationary. Scattered cumulus clouds were present over most of Illinois.

3 August

The high remained stationary, but a new system began to develop in the upper Rocky Mountains.

4 August

General low pressure prevailed west of the Dakotas, but it remained fairly stationary as high pressure centered over Michigan did not move. Skies over Illinois remained fair.

5 August

A closed upper air low began moving across the United States - Canada border in Washington, and a surface cold front formed in Montana and Idaho. Indications were that the low aloft would move to the east and carry the front with it.

6 August

The cold front extended from the Dakotas into southern Nevada in the morning, and moved into Minnesota by evening. No precipitation was forecast for Illinois until the 7th, when the high pressure over the eastern third of the nation was expected to move off the coast. The aircraft crew made a flight to the St. Louis area to take air samples during the afternoon.

7 August ("Go" day)

A squall line was forecast to form from Michigan through central Illinois and to central Missouri during the afternoon, in advance of the cold front. Eleven air samplers were placed on the CIN, and the aircraft flew for several hours searching for promising storms. The radar vectored the plane to several echoes, but each dissipated soon after formation. No line ever formed, and the aircraft returned after several hours of flying.

8 August

The cold front passed through Champaign shortly before sunrise. Since the prospects of thunderstorms were poor for several days to come, the aircraft and the field crews were released.

APPENDIX C

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 and
W. E. Bradley
"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 Ce¹⁴⁴ and Sr⁹⁰ 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"
- COO-1199-15 -- Research Report No. 3 - July 1968 - John W. Wilson and
Parker T. Jones III
"Tracing Tropospheric Radioactive Debris by Isentropic
Trajectories"
- COO-1199-16 -- SMRP Research Paper No. 74 - June 1968 - Walter A. Lyons
and John W. Wilson
"The Control of Summertime Cumuli and Thunderstorms by Lake
Michigan During Non-Lake Breeze Conditions"
- COO-1199-17 -- Seventh Progress Report - November 1968
"Study of Rainout of Radioactivity in Illinois"
- COO-1199-18 -- Eight Progress Report - November 1969
"Study of Rainout of Radioactivity in Illinois"