C00-1199-30

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

STUDY OF RAINOUT OF RADIOACTIVITY IN ILLINOIS

Eleventh Progress Report
Contract Number AT(ll-l)-1199
May 1973

by

J. R. Adam, R. Cataneo, D. F. Gatz, and R. G. Semonin

Sponsored by

United States Atomic Energy Commission
Earth Sciences Branch
Division of Biomedical and Environmental Research
Washington, D.C.

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Principal Investigator

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ABSTRACT

The field project phase of the contract research during 1972 was continued in the St. Louis area as an integral part of Project METROMEX, an effort designed to determine the effects of an urban area on the precipitation process. Unique chemical tracers released into the updrafts of convective clouds were used to obtain insight into the scavenging mechanism of the treated clouds.

This report describes the field project design as it relates to METROMEX, and presents some analyses and conclusions related to the 1971-72 data collection periods.

ACKNOWLEDGMENTS

In an effort the magnitude of METROMEX, there are, of course, several individuals whose contributions make the project "go".

We would like to specifically acknowledge the following State Water Survey members: Stanley A. Changnon for his general guidance and direct support through his National Science Foundation grant of the surface instrumentation network which was invaluable in the preparation of this report; Bernice Ackerman for the use of her pibal-radiosonde network data which was essential to the case studies analysis; Ronald Grosh for his significant contribution towards the case studies section of this report; Anthony Rattonetti and staff for their diligence and perseverance in the chemical analyses; and Terry Flach for the preparation of the raindrop spectrometers used in the field.

We would also like to thank Harry Moses and Paul Franzen of Argonne National Laboratory for the loan of their instrumentation; Roscoe Braham of the University of Chicago for the TPS-10 radar data used in the case studies analysis; Augie Auer and Dick Dirks of the University of Wyoming for collecting airborne water samples; Herb Volchok of the U. S. Atomic Energy Commission Health and Safety Laboratory for the use of his wet-dry samplers; Bill Finnegan of Naval Weapons Center for the use of his ground tracer generator; and St. Louis University for allowing us to utilize their grounds. '

During METROMEX 1971 and 1972, several student employees toiled hard and long hours in various phases of the field effort. We would like to express to the following our appreciation for jobs well done:

1971 Radar Operators--Gary Alessi, Gary Ernst, Steven Hilberg, Kenneth LaPenta

Rainwater Collectors–Thomas Cislo, Walter Drag, Elliot Mulberg, Patrick Reddy

Special Instruments Technician–Charles Knight

1972 *Radar Operators–Alan D'Hautecourt, Gary Ernst, Ronald Grosh, Steven Hilberg, Kenneth LaPenta, Elliot Mulberg*

Rainwater Collectors–Nolan Doeskin, Curt Holderbach, David Manton, James Perkovic, Patrick Reddy

Special Instrument Technicians–John Appleman, Walter Drag, William Grobl, Lawrence Heitkemper, Charles Knight, Michael Soper, John Werner

CONTENTS

Page

Page

LIST OF FIGURES

Page

Page

Page

LIST OF TABLES

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1.0 INTRODUCTION

The removal of gaseous and particulate matter from the atmosphere by precipitation is a vital study area with direct application to the determination of the deposition of airborne radioactive materials, air pollution meteorology, and weather modification. A brief summary of the state of knowledge of precipitation scavenging has been given by Slade (1968), and recently updated in a symposium entitled, "Precipitation Scavenging (1970)" sponsored by the Pacific Northwest Laboratories, Battelle Memorial Institute, and the Fallout Studies Branch, Division of Biology and Medicine, U. S. Atomic Energy Commission". The meteorological system associated with precipitation, including its widespread cloud cover, also plays an important role in the transport and diffusion of atmospheric contaminants. The scavenging of materials by precipitation, however, is of particular interest because it represents a mechanism for concentrating gases and aerosols. The very existence of a cloud (due to the convergence and phase change of water vapor) is a manifestation of its importance to the transport and diffusion of atmospheric constituents.

The problem of precipitation scavenging is compounded by the complexities associated with two areas of geophysical research. The first of these concerns the problems related to the dynamics of the atmosphere on scales varying from that of cloud dimensions to the global circulation. The second and equally complex problem area is related to the chemistry of the aerosol and gaseous concentrations in the atmosphere. The combination of these two study areas, with water introduced as a catalyst, is a research topic worthy of sophisticated equipment and considerable effort in the carrying out of field experiments.

Copies of the proceedings are available as CONF - 700601 for $6.00 from the National Technical Information Service, U. S. Department of Commerce, Springfield, Virginia 22151.

Very commonly the precipitation scavenging is divided into at least two separate mechanisms: 1) that which removes aerosols in the region between cloud base and the surface; and 2) that which removes aerosols by in-cloud processes. The former is referred to as washout whereas the latter is termed rainout.

Within the total scavenging problem, the atmospheric aerosol and gaseous material can be removed by two generalized processes. The removal of either particles or gases can be achieved by dry processes such as gravitational settling enhanced by the atmospheric turbulence near the ground surface. In addition an important sink for the gaseous and particulate constituents in the atmosphere is the collection by various obstacles to the atmospheric flow such as trees, and vegetation in the lower atmosphere. The removal mechanism has been theoretically described by Toba and Tanaka (1963) and more recently by Semonin (1972).

A more subtle sink that must be considered as a dry process is the conversion of an aerosol or gaseous constituent from one chemical species to another by chemical reactions. This does not preclude the removal of the gas phase material by attachment to particulates within the effluent of an urban-industrial region.

All of these removal processes are at work under non-precipitating conditions and are responsible for the quasi-stable background concentration of gaseous and particulate matter in the atmosphere.

The introduction of trace materials into rainwater is the result of one or more possible mechanisms which must be separated to fully understand the scavenging problem. There are three primary processes involved in precipitation scavenging: 1) the phase change of water vapor, that is, condensation and/or deposition on particles; 2) particle-to-particle or particle-to-drop diffusion; and 3) the aerodynamic collection by the liquid and frozen particles within a cloud.

The research undertaken under this contract as a part of the METROMEX field project involves measurements which will lead to a better understanding of the removal of particulate matter from the atmosphere by both dry and wet mechanisms. The sophistication of the techniques for data collection are constantly undergoing improvement along with the analytical methods for the

detection of various chemical species. A more detailed and comprehensive report of the work accomplished during the last three contract periods will appear in a forthcoming research report under this contract.

2.0 PROJECT DESCRIPTION

The METROMEX research program is a cooperative scientific effort designed to investigate the inadvertent effect on weather caused by an urban-industrial complex. The program is divided into various areas of research as described by Changnon et al., (1971), but the largest cooperative effort is the METROMEX Field Project located in and around the St. Louis, Missouri urban area. The specific goals of the Water Survey's participation in the field project include 1) the release of unique chemical tracers into convective storms to determine their scavenging efficiency; 2) to assess the washout ratios of various atmospheric constituents as a function of meteorological conditions; 3) to determine the effects of urban structures on the low level wind; 4) to study severe local weather phenomena so as to describe the temporal-spatial variations of these events in the St. Louis urban area with special reference to their relationships under varying synoptic weather conditions; and 5) to study rainfall and radar data to assess the magnitude and location of the urban-related precipitation changes with specific reference to time-space analyses of rainfall and synoptic weather analyses.

Scientists from six groups participated in the 1972 METROMEX field operation. It was not possible for all of these groups to be represented in the St. Louis area during the entire 3-month operational period, and therefore, the month of August became the primary data collection period for many of the groups involved. The participating groups were: the Argonne National Laboratory, the University of Chicago, the Battelle Northwest Laboratory, the Stanford Research Institute, the Water Survey, and the University of Wyoming. The AEC Health and Safety Laboratories in New York and the Naval Weapons Center in California provided equipment for the project and personnel to instruct Survey employees on its operation in the field. This type of participation is most welcome and demonstrates that the program in St. Louis appears attractive to many scientists although they are not able to participate themselves.

The Water Survey's portion of the research effort is funded from three sources. The State of Illinois contributes funds and equipment for the

operation and maintenance of the extensive surface network as well as the field headquarters for the project. The National Science Foundation contributes support towards the operation of additional surface instrumentation for the detection of severe storms, as well as the operation of radars for the assessment of their utility in delineating the urban effects on precipitation. The AEC contract is funding the research to investigate precipitation scavenging through field measurements of tracer chemicals introduced by aircraft and surface locations into storm systems that traverse the rainwater sampling network, and measurements to assess the washout ratio variations as related to chemical species, and particle size.

3.0 . FIELD INSTRUMENTATION AND OPERATIONS

A very extensive network of various instruments has been deployed within the 3900 mi^2 research area surrounding St. Louis. The care and operation of the diverse equipment is directed from the project field headquarters located north of the research area. Since many of the instruments and their applications are unique, each of them is described in the following sections as well as their placement and utility on the project.

3.1 Raingage Network

The network of recording raingages was installed at a density of one per 9 mi^2 within a circle of 26 miles radius centered on the Arch in downtown St. Louis. The data from the 220 gages employed in 1971 indicated that additional observations were necessary to fully describe the morphology of precipitation entities as they move to the east and northeast. Consequently, the network was expanded to include an additional 1700 mi^2 downwind from St. Louis with a gage density of 1 per 81 mi^2. In 1972, therefore, the research area was elongated to the east and northeast encompassing a total of 3900 mi^2 as shown in Fig. 1.

A standard weighing bucket raingage is used on the entire network. A typical field installation is shown in Fig. 2. As the precipitation is captured by the 8 inch diameter opening it falls into an 8 quart metal pail which moves a pen, through a system of levers and linkages, across a clock-driven cylindrical drum. A typical gage will record 6 inches of precipitation which raises the recording pen to the top edge of the chart

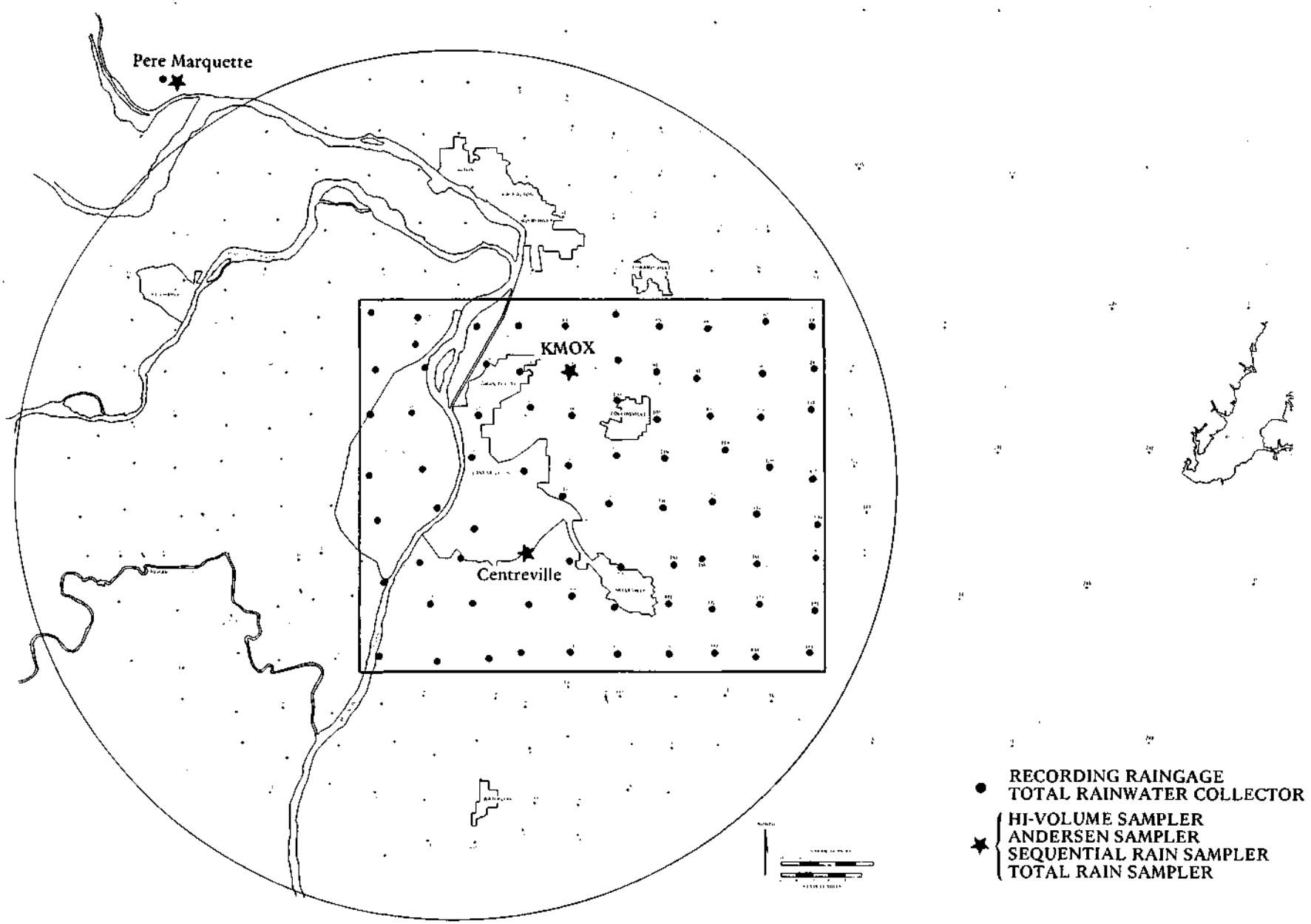

Figure 1. Raingage, rainwater, and air chemistry network for the 1972 METROMEX field project

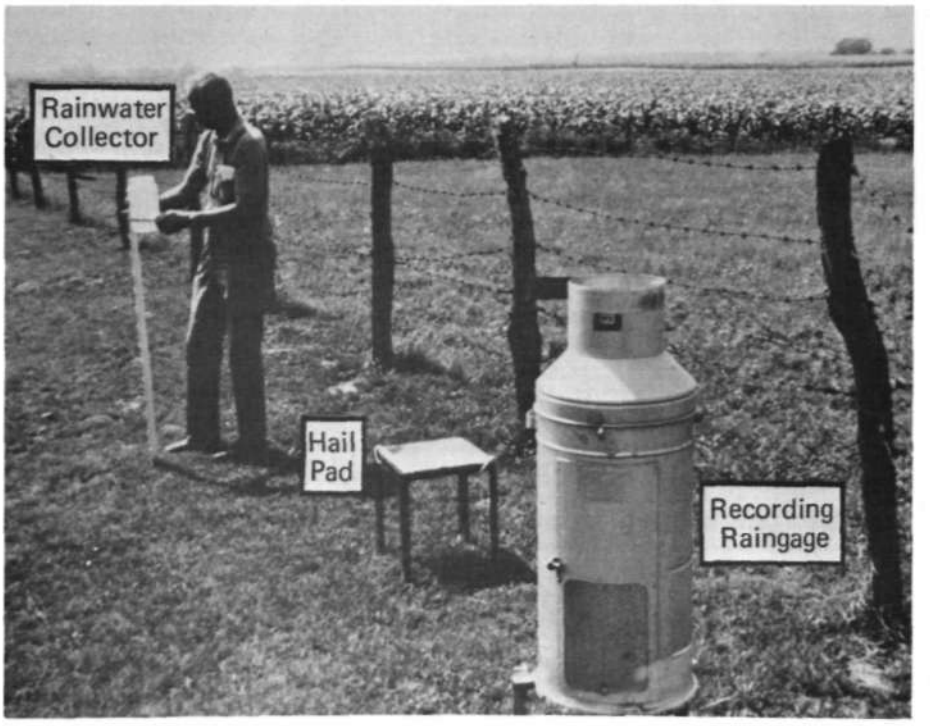

Figure 2. A typical network instrument site with a rainwater collector, hailpad, and recording raingage

paper and subsequent precipitation will cause the pen-arm to reverse, thus permitting the recording of a maximum of 12 inches of precipitation.

The recorder drum is rotated by a standard hand wound clock mechanism which requires maintenance and care of such a device throughout the field operation. Various gears are available which permit 6 hour to 1 week records per chart. As a compromise to the research involved in St. Louis, all of the gages with a few exceptions, operate with 24 hour gears and the charts are changed weekly. A few gages are placed judiciously throughout the network with weekly gears which allow the determination of particular storms to be distinguished with relative ease since the 24 hour charts may revolve many times prior to the onset of precipitation; such a situation may cause confusion with regard to the actual day or period of the storm occurrence.

The 240 charts which are generated weekly during the summer months are transported to the Survey facilities in Champaign for further checking and processing within a week from the time they are changed in the field. The charts are edited and storm periods are indicated on the chart. All of the charts are subsequently digitized onto magnetic tape by means of a hand operated curve follower, and transmitted to the computer system of the University of Illinois for final calculation of storm rainfall rates and amounts.

These data are not only invaluable in detecting the urban effects upon precipitation, but are also essential for the final analysis of the tracer experiments conducted over the network. The usefulness of this network facility with regard to the tracer experiments will be amplified in a later section of this report.

3.2 Field Headquarters

In 1970 the State of Illinois acquired title to the property formerly occupied by the U. S. Army as a Nike system missile complex. In turn the Water Survey was permitted to occupy the headquarters portion of the property as the field headquarters for the METROMEX project.

The property consists of a group of buildings which provide adequate space for the radar operations, shop and storage facilities, radar equipment, office and housing space, and living quarters for a fulltime caretaker and his family. A plan map of the area is shown in Fig. 3. An important part

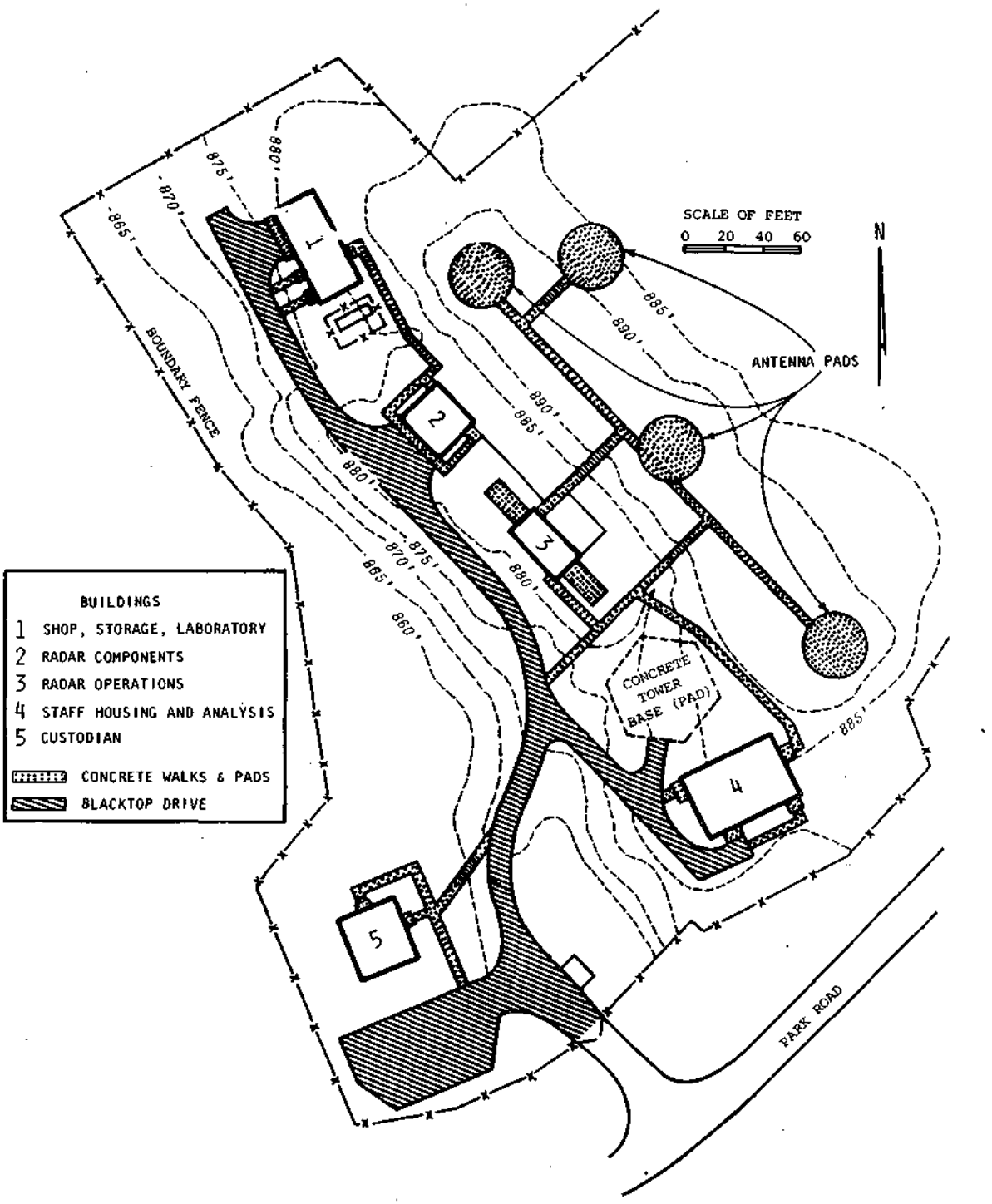

Figure 3. Plan of the METROMEX field project headquarters and radar site at Pere Marquette State Park near Grafton, Illinois

of this facility is the weather central for the entire METROMEX project where teletype and facsimile weather data are displayed and interpreted for all groups. Work areas were established for special operations by other researchers, for example, a radio was installed near a special repeater scope for the radars to aid the University of Wyoming with their aircraft flight program.

Standard weather observations are obtained on a 24-hour schedule at this headquarters during the summer months. Special pibal observations are also taken once daily to assist in the overall planning for the daily operations. The strategy for the daily work involves the interpretation of the weather maps to provide a forecast along with a determination of the low-level winds so that decisions concerning the type of mission to be attempted can be optimized.

Radio facilities are available at the headquarters for communications with the Survey aircraft and ground crews concerning the development and progress of storms under study.

The field crews used for the rainwater sampling network, and technicians required for the maintenance of much of the field and radar equipment are housed at the headquarters. The living quarters consist of a bunk area suitable for 14 personnel, a kitchen area with all the necessities for preparing and eating meals, and a general area which serves as the office space for senior scientists.

The floor space useable for the conduct of the weather and radar operations was expanded by the addition of a mobile van on the southeast side of the radar operations building. This addition has made additional room available, in proximity to the radar, for the real time analysis of the current weather and other routine work essential to the conduct of the everyday operations. It is anticipated that another trailer will be added to the northwest side of the radar operations building for the 1973 operations to further free the radar room from the congestion arising from the various radar data recording systems.

The site, in addition to its prime function of a field headquarters, represents a useful rural site for background measurements from specialized instrumentation. This use of the site requires knowledge of the prevailing

flow patterns since there is evidence to show that the urban influenced atmosphere is transported to Pere Marquette under southeasterly low-level flow.

3.3 Rainwater Collection Network

The 1972 rainwater sampling network (Fig. 1) consisted of 81 single-sample collectors in a 700 mi^2 area encompassing and extending eastward from the cities of St. Louis and East St. Louis.

Samples were collected in pre-washed linear polyethylene bottles mounted approximately 1.5 m above the ground on metal fence posts. The bottles were attached to the post in such a way as to avoid any possibility that raindrops splashing off the metal support would enter the sample bottle.

On days designated for collection (either "wet" or "dry") sample bottles were distributed to the network of sites and left exposed, with caps off, by a team of field technicians in automobiles. In the case of a "dry" experiment, where the purpose was to measure the dry component of the deposition, bottles were distributed one day and picked up the following day. When the purpose of the experiment was to measure wet deposition, samples were often picked up immediately following the rain. Radio communications between the Pere Marquette field headquarters and the field crew were used to give, or change, sample collection instructions based on current and expected weather.

Sample bottles were used once only; after collection in the field they were returned to the Pere Marquette headquarters, and then transferred to the Survey chemistry laboratory in Champaign for processing and analysis.

The results of the 1972 rain sampling operations are summarized in Table 1. Between 12 June and 27 August 1972, 3637 samples were collected. Of these, 1745 (48%) contained rain samples, and 1892 were dry.

About 25% of the samples were collected on days when tracers were released either from the ground or aloft. The tracer experiments produced 585 wet and 307 dry samples that may contain tracer materials. Results of the tracer experiments are presented later in this report.

TABLE 1

Summary of wet and dry samples from 1972 METROMEX network operations.

Experiment Number	Date	Samples Wet	Samples Dry	Tracer Operation
100	6/12-13	1	79	
101	6/15-16	0	81	
102	6/19-20	81	0	
103	6/20-21	1	80	
104	6/21-22	1	79	
105	6/22-25	5	75	
106	6/25-26	0	80	
107	6/26-27	0	81	
108	6/27-28	20	61	
109	6/29-29	2	79	Ground-lithium
110	6/29-30	1	80	
111	6/30-7/2	81	0	
112	7/ 2- 3	72	9	
113	7/ 3- 5	79	2	Ground-lithium
114	7/ 7- 9	15	66	Air-lithium
115	7/ 9-10	38	43	
116	7/10-12	0	81	
117	7/12-15	65	16	
118	7/15-16	64	16	Ground-lithium
119	7/16-17	67	14	
120	7/17-18	81	0	Air-lithium indium
121	7/18-19	45	36	
122	7/19-21	20	61	
123	7/21-24	8	73	
124	7/24-24	60	21	Air-lithium
125	7/24-26	15	66	
126	7/26-27	81	0	
127	7/27-29	81	0	
128	7/29-8/1	0	81	
129	8/ 1- 3	81	0	
130	8/ 3- 3	81	0	Air-lithium indium
131	8/ 3- 4	45	36	
132	8/ 4- 6	81	0	Ground-lithium
133	8/ 6-10	2	78	
134	8/10-11	0	81	
135	8/11-11	54	27	Air-lithium indium
136	8/11-13	50	30	
137	8/13-19	28	52	
138	8/19-20	21	60	Ground-lithium
139	8/20-20	45	36	Ground-lithium
140	8/20-22	81	0	
141	8/22-23	81	0	
142	8/23-24	72	9	
143	8/24-26	11	70	
144	8/26-27	28	53	
Totals		1745	1892	

3.4 Sequential Rain Samplers

The network of single-sample collectors provides detailed information about the spatial distribution of the rain-deposited tracer, but no information about the time history of the deposition. Data on tracer deposition with time during the rain is needed to infer in-cloud residence times'. Such residence times, together with observed relationships between rainfall rate and tracer concentration or deposition, are important clues to the dominant physical mechanism responsible for the incorporation of the airborne tracer material into the rain. To provide data on the time history of deposition, 3 sequential rain samplers (Gatz, 1971), each capable of collecting 70 samples from 5 cm of rain, were operated in the research area, as shown on the map in Fig. 1.

The sequential samplers were visited daily and checked for adequate ink and paper supply in the strip chart recorder, proper operation of rain-sensing and sample-sequencing mechanisms, and correct clock time. When conditions indicated the possibility of rain within 2-3 days, the technician thoroughly cleaned the collecting funnel—and all parts that come in contact with the collected water—with distilled water, and obtained 2 blanks consisting of distilled deionized water sprayed over the collecting funnel and collected at its drain. On days when rain was expected, the technician removed the funnel cover, collected blanks, checked the samplers for proper operation, and removed the caps from the sample bottles. After a rain event, caps were replaced, the bottles carefully labeled in order of collection and removed, blanks taken, data charts and tapes removed, and the funnel cover replaced.

The 1972 sampling season produced 238 sequential rain samples from 10 rains. Results of chemical analyses of a portion of these samples are presented and discussed later in this report.

3.5 Other Rain Samplers

Small numbers of two other types of rain samplers were operated for special purposes in 1972. Three ten-inch diameter polyethylene funnel collectors which drained into polyethylene bottles were used to obtain samples of adequate volume for the determination of about 10 trace metals for calculation of washout ratios. These samplers were located at Pere Marquette,

the KMOX radio transmitter, and Centreville (the starred locations in Fig. 1). They were visited daily to pick up samples and clean the funnels. Dry deposition samples were collected twice weekly, on Tuesdays and Thursdays, provided that rain did not occur.

Another type of rain sampler operated, shown in Fig. 4, was the wet/dry sampler built and provided by the AEC's Health and Safety Laboratory (HASL). These devices are equipped with a motor-driven cover that exposes one collector only during dry weather and another only during rain. The resulting samples provide a means of determining what fraction of the total deposit of any element of interest came down in dry form vs. wet (i.e., in precipitation). There were two of these samplers — one located at Pere Marquette and the other at Centreville. These samplers were checked daily for proper operation. After each rain, both the wet and the dry samples were removed and replaced with clean sampling bottles (the same as used in the rainwater collection network).

3.6 Aerosol Sampling

Samples of atmospheric aerosols were collected for chemical analysis by two different methods, that is, on filters to obtain 24-hour average concentrations, and in multi-stage Andersen impactors, to obtain information on size distributions, for about 10 trace metals.

3.6.1 Filter Samples

Samples were collected on 15 cm diameter Whatman-Ul filter paper, using high volume (about 40 m^3/hr) positive displacement pumps. Filters were changed daily. Sample volumes were estimated from pressure drop measurements, calibrated against a precision rotameter-type flow meter, made at the start and end of each filter exposure. Sample volumes obtained in this way are considered accurate to ±5%. Filters were handled only with clean teflon-covered forceps and stored with sample-side folded together in polyethylene bags until analysis.

3.6.2 Andersen Impactor Samples

Two different types of Andersen impactors were used in 1972. As in 1971, 3 of the 9 stages (with backup filter), 1-cfm samplers, were used. These were

Figure 4. A wet/dry sampler for precipitation and dry fallout chemistry studies

located at the Pere Marquette, KMOX, and Centreville sites. Sample durations were usually about 7 days. To provide a few samples from air having a constant wind direction (±45°), sampling was occasionally interrupted for periods of 1 or more days when the wind direction changed.

Since experimentation demonstrated (Fig. 5) that Whatman-41 filter paper was essentially equivalent to the thin polyethylene discs previously used by us and others (Nifong, 1970) as collection substrates, the 1972 9-stage impactor samples were collected on Whatman-41 filter paper.

The second type of Andersen impactor used in 1972 was a 5-stage (with backup filter) "high-volume model", operating at 20 cfm. This sampler also used Whatman-41 collection substrates. Because of the higher flow rates, shorter sampling periods were possible. Thus, this sampler was operated for 24-hr periods. It collected a series of seven consecutive 24-hr samples from each of the three primary instrument sites, namely Pere Marquette, KMOX, and Centreville.

Altogether, 36 separate size distribution samples were collected. Their analysis has been delayed by the need to process rain samples first, and the results are not yet available.

3.7 . Aircraft

The aircraft utilized during METROMEX 1972 was a Piper Navajo operated by Atmospherics Incorporated under a subcontract. The main mission of the aircraft during the six week period of 6 July - 15 August, as it was in 1971, was to inject tracer material into convective cells that passed over the rainwater collection network. Four additional priorities were also included. These were: 1) measure cloud bases over the urban area and the surrounding rural area to determine possible differences in the two; 2) map convective cells as to updraft speed, size, location and duration; 3) identify and locate major particulate sources in the St. Louis urban area; and 4) determine general cloud structure characteristics in the region. The aircraft (Fig. 6) logged 44- flights during METROMEX 1972 including five tracer missions.

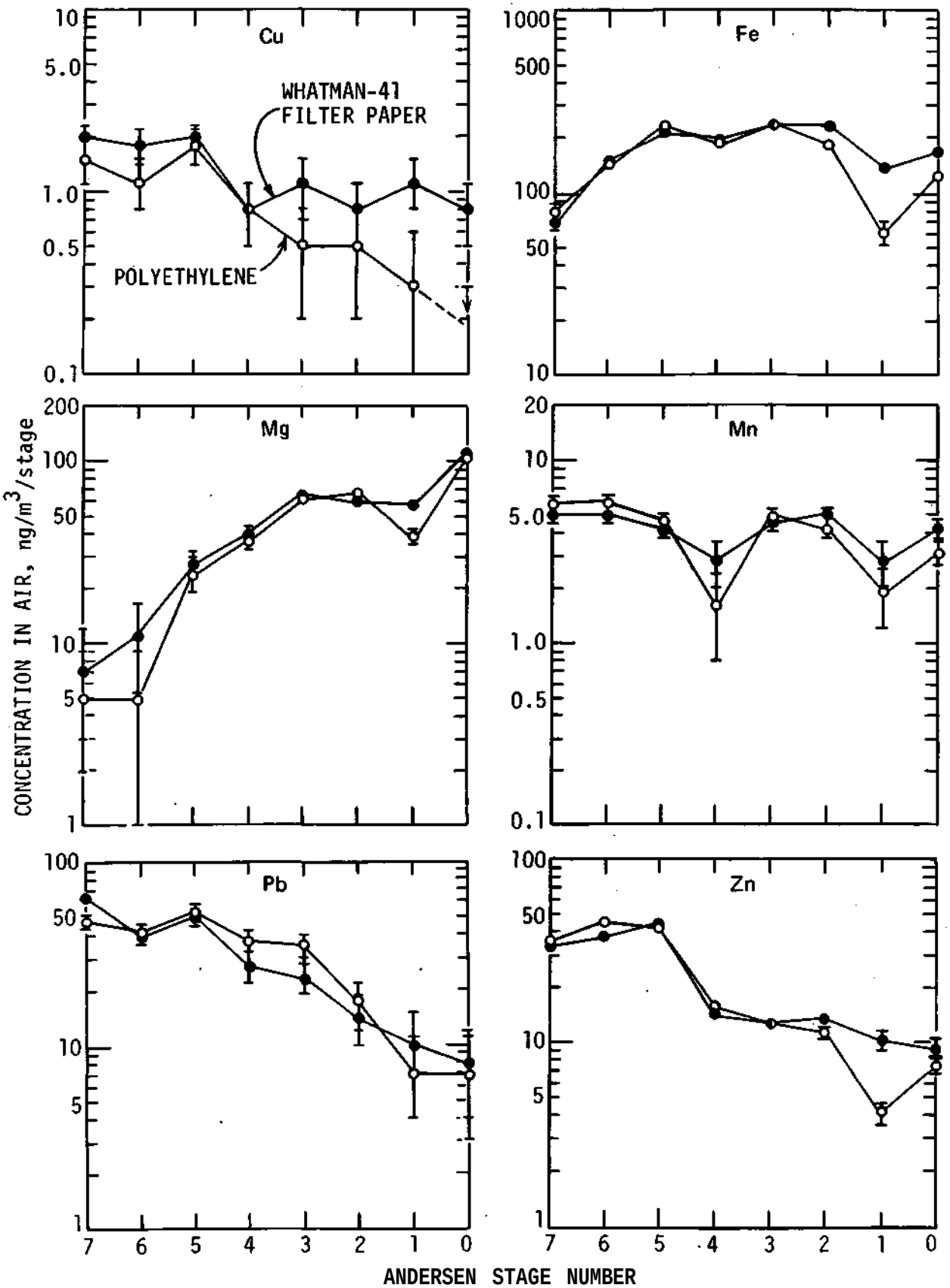

Figure 5. Comparison between Whatman-41 and polyethylene filters for various elements as a function of Andersen impactor stage number

Figure 6. The Atmospherics Inc. Piper Navajo used for the METROMEX project. The tracer release generators can be seen on the wing tips

3.8 Tracer Generator

A surface tracer generator (Fig. 7) was used which simulated a near surface source of particulates. The device was on loan from Naval Weapons Center, China Lake, California for the June-August 1972 period and was utilized for six ground tracer releases into convective. cells during this time.

3.9 Dropsize Network

Raindrop size distributions were measured at several locations in the St. Louis region to determine the possible effects of the urban area on raindrop spectra. The instrument used for the measurements is a raindrop spectrometer which converts the momentum of a drop impacting on a sensor, to a voltage; the voltage is then converted to dropsize.

Two commercial devices were located at PMQ and Centreville for most of the 1972 period; several similar in-house-manufactured spectrometers were located at other sites in the research circle (Fig. 8).

3.10 Upper-Air Network

This operation was designed to provide low-level wind data for trajectory analysis and temperature-moisture profiles to determine boundary layer stability. Temporal changes in the thermodynamic structure as cloud systems develop and move through the urban area are also observed by serial radiosonde ascents. This effort is in direct support of the tracer experiments (2.7 and 2.8).

To accomplish the above, ten pibal-radiosonde teams from the Air Weather Service were utilized. Three types of operation were implemented during the 12 July - 11 August period; the first involved convective shower and tracer experiments, the second was related to nighttime urban circulation., and the third was concerned with non-rain daytime situations. A total of 23 days were worked in 1972 and distributed among the three mission objectives above.

3.11 Radar

3.11.1 FPS-18

The purpose of this 10-cm radar was two-fold; to examine storm morphology as it is influenced by the urban area, and to vector the tracer aircraft to

Figure 7. The surface tracer generator provided by the U. S. Navy, Naval Weapons Center for the release of tracer lithium

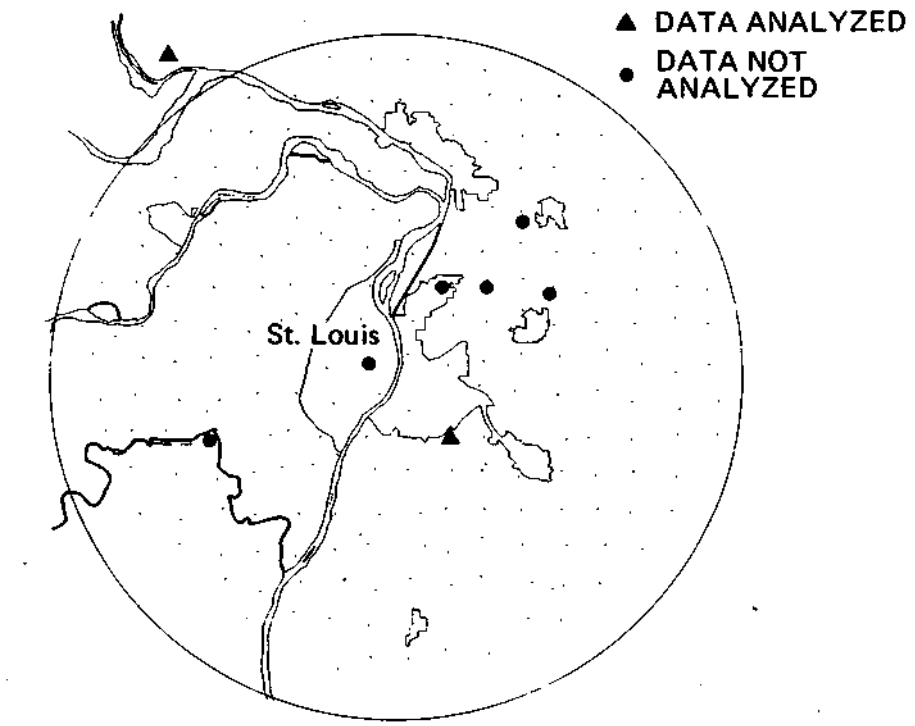

Figure 8. The location of spectrometers used to determine the number and size of the drops during precipitation

appropriate convective cells during the aircraft portion of METROMEX. The precipitation echo data obtained with this radar was recorded on film as well as on magnetic tape for later analyses.

The FPS-18 radar was operated 24 hrs/day during the June-August 1972 period with few exceptions.

3.11.2 TPS-10

This 3-cm vertical scan radar was used in conjunction with the FPS-18 to examine the morphology of echoes as they passed over the urban area, and it also served to inform the aircraft of the location of convective cells which were in the growing stage. For the tracer portion of METROMEX, it was important that the tracer material be placed in a cell that had a projected life-time sufficiently long to carry it over the rainwater collection network. This radar was an indispensable tool for the real-time evaluation of storm development pertinent to the tracer operations.

3.12 Supplementary Weather Instrumentation

To assist in describing the small-scale meteorological picture associated with scavenging studies and the occurrence of urban-related weather phenomena, a network of various surface instruments was established which is briefly described below.

3.12.1 Wind Network

The surface network of wind-measuring equipment especially installed for Project. METROMEX consisted of six instruments. The six instruments were sited as shown on Fig. 9 and were generally on the perimeter of the METROMEX research circle. No location changes were made between 1971 and 1972.

The wind recorder at radar headquarters (PMQ) was a Bendix-Friez Aerovane system with electric synchronous strip chart drive. Occasional missing records were due to failure to change the chart and to malfunction of the inking system of the recording pens. The speed and direction sensor was located approximately 15 ft above the crest of the ridge upon which the radar antennae were located. The northwest-southeast orientation of the ridge would tend to bias the recorded wind directions toward the axis of the ridge to an unknown extent, while the crowding of streamlines rising over

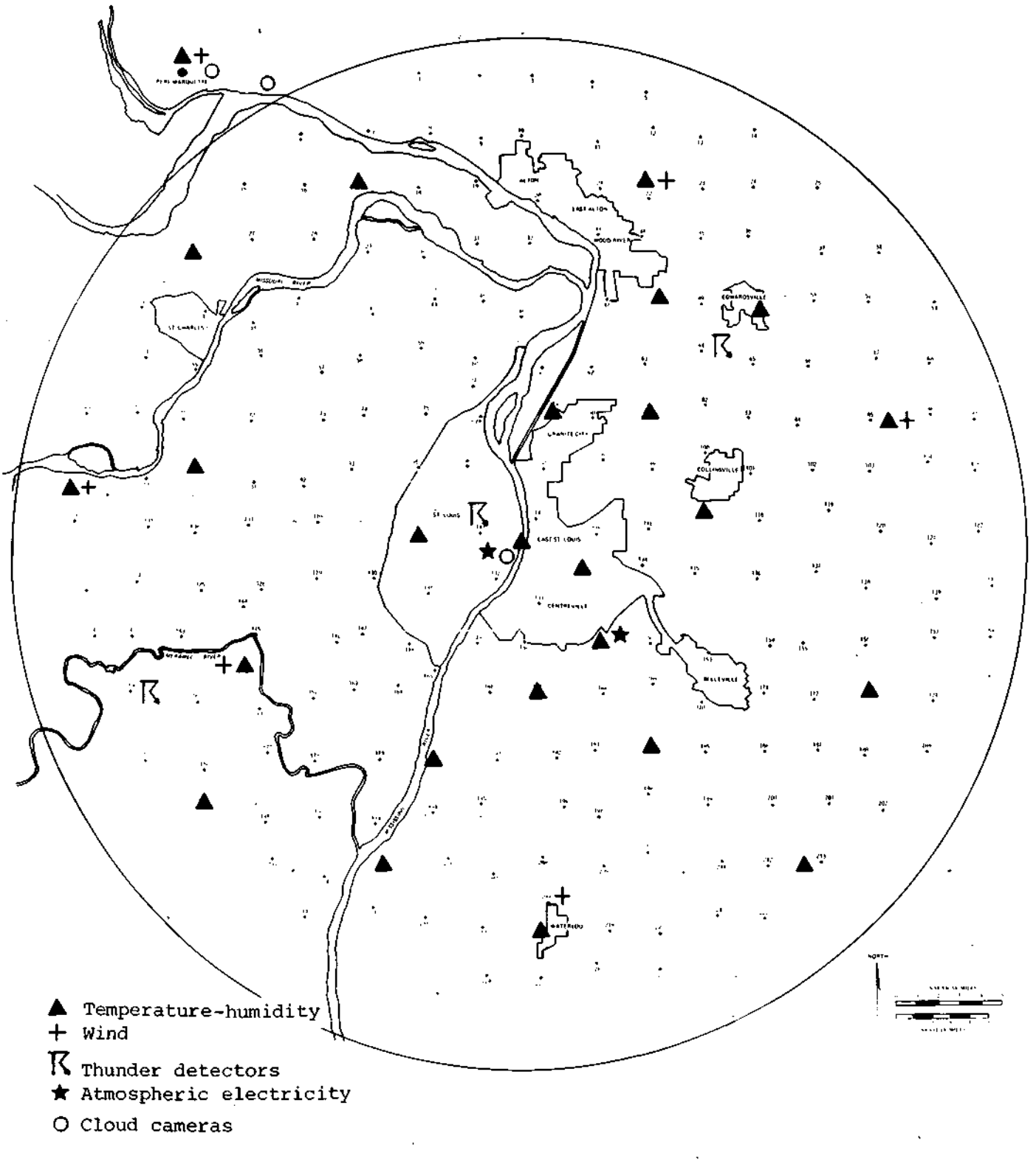

Figure 9. The site locations for temperature and humidity measurements, wind observations, and thunder detection

the ridge would tend to increase the recorded wind speed above ambient by approximately 5%. Although there are no obstructions to the wind field about the sensor, the radar headquarters site was surrounded by heavily wooded lower-lying hills. The sensor was sited on the highest hill within Pere Marquette State Park.

The wind recorder at Alton Civic Memorial Airport used a speed sensor consisting of a beaded-rim, truncated-cone, three-cup design whose rotation was translated into a proportional dc voltage through an attached generator. The voltage was recorded as speed-equivalent in analog form in mph on a mechanical clock-driven strip chart moving at three inches per hour. Starting speed of the cups was about 1 to 2 mph and recording accuracy was ±1 mph. The wind direction was sensed by a wind vane approximately 3 ft in total length and its position was detected by 8 electrical contacts fastened to the vane shaft. The brush and contact arrangement was such that shorting contact was made between two adjacent direction contacts when the wind direction was between them. Thus, the intermediate directions as well as 8 points of the compass were indicated on the 8 event-recording pens that write upon the strip chart recorder used for both speed and direction. Chart speed was 3 inches per hour. The two sensors were mounted on a mast approximately 12 ft above grade. No obstacles taller than the recorder housing at 4 ft height were closer than 300 ft to the wind instrument. Data were lost occasionally because of clock failure, low voltage (12vdc) to the direction recorder, or failure of the ink supply to the 9 recording pens.

The wind instrument at the Nagel Farm was sited in a clear, flat area with the closest obstacle an east-west railroad embankment approximately 100 ft to the south. The area was open in all other directions for at least 300 ft with a grove of trees to the east at that distance. The instrument is a duplicate of the Alton system and subject to the same data losses.

The Waterloo wind set was sited on a very slight prominence surrounded only by lawn to the north and corn or soybean fields to the south. A small fruit tree was growing approximately 30 ft southeast of the wind instrument. All other obstacles were less than half the height of the sensors at approximately 15 ft. This installation was a duplicate of the Alton installation.

A wind set was sited at the Weiss Airport in the Meramec River Valley southeast of downtown St. Louis. The instrument was located about 30 ft

west of the north-south runway and had no obstructions within 300 ft of the site. The area is relatively protected since the airport is located in a wooded valley. No particular wind direction would appear to be favored, however. The instrumentation was a duplicate of the Alton equipment with the sensors about 12 ft above grade.

The sixth wind set was installed at the Spirit of St. Louis Airport in the flat floodplain of the Missouri River west of downtown St. Louis. The edge of the river valley was in the distant south with no obstructions within 500 ft of the instrument. The sensors were about 12 ft above grade. The equipment was a duplicate of the Alton wind set.

3.12.2 Hygrothermograph Network

Surface temperature and humidity were recorded by hygrothermographs in standard cotton region shelters in both 1971 and 1972. Eight sites were in operation during the summer months of 1971. There were six hygrothermographs manufactured by Weather Measure Inc. These instruments had a bi-metal temperature sensor and a hair bundle humidity sensor. Recording was performed onto a combined chart with 168-hr duration. The nearly-logarithmic indication of humidity obtained from the hair-length changes was converted to nearly linear recording by a set of cams in the humidity linkage. The remaining two instruments were manufactured by the Bendix Corporation. These instruments used a bourdon type for a temperature sensor and a harp of hairs as the humidity sensor. But for these two differences the two types of instruments are similar.

Twenty-five hygrothermograph instruments in Cotton Region shelters were in operation during part or all of the summer months of 1972. These sites are also shown in Fig. 9. Seventeen of these instruments were of Weather Measure manufacture and 8 were the design of the Bendix instrument. Both types of instruments are capable of temperature recording accuracy of ±1°F.

The checking of the accuracy of the instruments was insufficient to insure that the recordings were comparable during 1971. Hence, the data have been used only for special studies wherein limited inter-instrument comparisons were made.

Comparison of a sling psychrometer reading was made each time of chart change during the summer of 1972. These comparisons have been used to prepare

corrections in temperature and humidity for the data reduced from the original recordings. The data have been used in the preparation of special studies of particularly interesting cases recorded during the summer months.

3.12.3 Thunder Detectors

Thunder detection instruments were designed, constructed and installed at three locations in the research area as shown in Fig. 9. Each of the sites included four microphones installed in a square array. The microphones were coplaner with a spacing of 125 ft. The filtered audio signal from thunder is recorded on a four channel recorder for subsequent analysis.

The analysis is carried out by comparing the arrival time for the thunder at each of the four sensors and it is therefore possible to determine the direction from which the thunder traveled. Furthermore, the entire instrument is triggered by the detection" of lightning within a limited area surrounding a station. Each of the lightning strokes is recorded and the frequency of lightning can be determined.

These data are useful to the scavenging work for the determination of the electrical activity associated with storms which are under investigation. The entire effort was funded by the National Science Foundation through the Survey.

3.12.4 Hail Sensor Network

In addition to the siting of 250 raingages throughout the research area, an equal number of hail detectors were placed at each location. The hail sensors are simple 1 ft^2 polystyrene pads covered with aluminum foil (see Fig. 2). When hail impacts on the foil, an imprint is made in the surface which can be measured. The size of the hailstones is determined by measurement of the impact diameter and the use of a calibration curve. The number density of the stones per unit area is obtained as well as the size distribution. The time of occurrence is ascertained from the nearby raingage by noting the spikes on the raingage chart. For this purpose, the evaporation funnel has been removed from all of the recording gages to allow the hail to fall unimpeded into the weighing bucket.

The data generated from this network are used to depict the areal extent of hailfalls and their association with other storm characteristics

such as rainfall. Careful analysis of the hail information in association with the time data from the recording raingage charts permits the depiction of hailstreaks, that is, hail entities analogous to the raincells used for other tests of the validity of the urban effects on precipitation.

This phase of the Survey research is sponsored by the National Science Foundation. The results from the studies are useful to the AEC work since another measure of the relative severity of storms treated with tracer material is determined.

4.0 LABORATORY PROCEDURES

This section outlines procedures used in handling and analyzing precipitation and aerosol (filter) samples collected in 1971 and 1972. Analysis of 1971 samples was divided among two laboratories. Air filter and precipitation analyses for computation of scavenging ratios and sequential rain sample analyses were carried out at Argonne National Laboratory. Precipitation samples collected in the large network (Fig. 1) of single-sample collectors were analyzed for tracer elements and a few other elements at the Survey. Analyses of all 1972 samples are being done at the Survey.

4.1 1971 Samples

4.1.1 Rain Samples

At Argonne National Laboratory, rain samples were filtered through 0.5 μm pore diameter low-blank membrane filters to separate the soluble and insoluble components. After all of the liquid portion of each sample had passed through the filter and into a clean polyethylene bottle, the filtrate (soluble fraction) was removed from the filtration chamber and stored frozen until analysis. Care was taken to remove as much of the insoluble material as possible from the walls of the polyethylene sample bottle, using a stream of distilled deionized water -from a squeeze bottle, and when necessary, a clean Teflon* scraper. Less than 5% of the insoluble material

* DuPont trade mark.

was left in the bottles, by visual estimate. All water rinses and a final isopropyl alcohol rinse of the filter funnel were collected in a separate waste bottle and discarded.

Dissolution of the filter samples was carried out in a clean 100-ml Teflon beaker. A few drops of acetone were added to dissolve the membrane filter, and the sample was again dried, using low heat on a hot plate. Then 10 ml of concentrated HNO_3 were added and the sample heated on the hot plate until the reddish-brown NO_2 fumes disappeared. Ten ml of concentrated $HClO_4$ and 5 ml of concentrated HF were added carefully and heated to strong fumes of $HClO_4$. Another 5 ml of HF were added and the sample was again heated to strong fumes of $HClO_4$, followed by evaporation to reduce the sample volume to about 5 to 10 ml. This was transferred, with beaker washings, to a 50-ml volumetric flask, diluted to volume with distilled water, and transferred to a polyethylene bottle for storage until analysis.

The insoluble portions of the precipitation samples were analyzed for Cu, Fe, Pb, Mg, Mn, and Zn directly by aspiration of the sample into the air-acetylene flame of an Instrumentation Laboratory Model 353 atomic absorption spectrophotometer. Approximately 2 ml were needed for each element. A standard La addition was used to suppress interference in the Mg determination.

The same procedure was followed in the case of the soluble portion except that in these samples Cu and Pb were first concentrated by a solvent extraction with dithizone into ethyl propionate (Sachdev and West, 1969). After confirming that rain contains virtually no soluble Fe, no Fe analyses were performed on the soluble rain fractions.

The instrument response was read in absorbance units and converted manually to concentration with calibration curves constructed from analyses of standard solutions. The standards used with acidified samples were made approximately 10% acid with equal parts of concentrated HNO_3 and $HClO_4$ to match the sample matrix. Standards for the extracted elements were prepared in water and underwent extraction in the same way as the samples. Single-element hollow cathode lamps were used as light sources for the analyses.

At the Survey, rain samples were analyzed directly, without filtration, for Li, Na, K, Ca, and Mg. In general, the analysis procedures followed

were similar to those used on the soluble portion of rain samples at Argonne. However, Li, Na, and K were determined by emission instead of absorption.

4.1.2 Air Filter Samples

The air filter analyses began by removal of about 1/2 of each sample to be analyzed. After removal of the outer ring of the filter (through which no air had passed), the filter was cut using a clean surgical knife. The fraction of the whole filter that was analyzed was determined from the relative masses of the analyzed fraction and the entire filter. Before weighing, care was taken to insure that the filter weight had stabilized for the ambient temperature and humidity conditions.

Except for the elimination of the addition of acetone (used to dissolve the membrane filter) the dissolution and analysis of the air filters followed the same procedure as given earlier for the rain insoluble fraction.

4.2 1972 Samples

All rain samples collected in 1972 have been filtered to separate soluble and insoluble fractions. The procedure was the same as described above, except that the final rinse with isopropyl alcohol was eliminated, and the filtrates were stored in liquid form until analysis.

Analyses of the soluble fraction of all rain samples for Li are completed and analyses for additional soluble elements are now underway.

Analysis of both the rain-insoluble fractions and air filters have been delayed pending development of a filter-dissolution procedure that does not include $HClO_4$, which our laboratory facilities are unable to handle safely. The new procedure is nearly operational, however, and routine analyses will begin shortly.

5.0 1971 AND 1972 OPERATIONS AND ANALYSIS

5.1 Aircraft Program-1971

In addition to the main priority of injecting tracer material into convective cells, the AI aircraft performed flights on fair weather days to measure condensation nuclei, ambient temperature and wet bulb depression

at approximately 1300 ft AGL in an X-pattern flight path over the St. Louis area (Fig. 10). These missions were carried out on 19 days during the six week aircraft period. Standard aircraft parameters were also measured along with the above variables. The data were recorded continuously on a strip chart recorder.

A summary of flight results is presented in Figs. 11-20. The abscissa in each of these figures represents points along the flight path shown in Fig. 10. Some observations and tentative conclusions relating to these figures follow.

5.1.1 Temperature Distribution

Ambient temperature at flight level was measured with a Rosemount probe; flight level temperatures averaged for five flights are plotted in Fig. 11; missing data on portions of cross-section flights on other days precluded their use. The data in the figure have been corrected for differences in topography using a dry adiabatic lapse rate (1°C/100 m). No attempt was made to correct for deviations in aircraft altitude from 1800 ft MSL, it is assumed that these deviations are random and are "averaged out" over a sufficient number of samples. Plotted also in the figure are the temperature deviations along the flight path from an average rural value of 22.7°C.

The data indicate an average positive temperature anomaly of 0.7°C in the urban area when compared to rural values; this is present in both the WSW and N legs of the flight track. It is very apparent that, for the data presented, the temperature at 1800 ft MSL increases as the urban area is approached, maximizes over the city, and then decreases to rural values.

5.1.2 Wet Bulb Depression

Wet bulb depression was measured with an electronic psychrometer manufactured by Mee Industries. Wet bulb depression corrected for topography and averaged over five flights is shown in Fig. 12. Topography corrections used were 1°C/100 m for temperature, and 1°C/235 m (moist adiabatic lapse rate) for wet bulb temperature. To properly assess possible moisture changes as the aircraft traversed the urban area, values for mixing ratio were calculated from ambient temperature and the corresponding wet bulb temperatures;

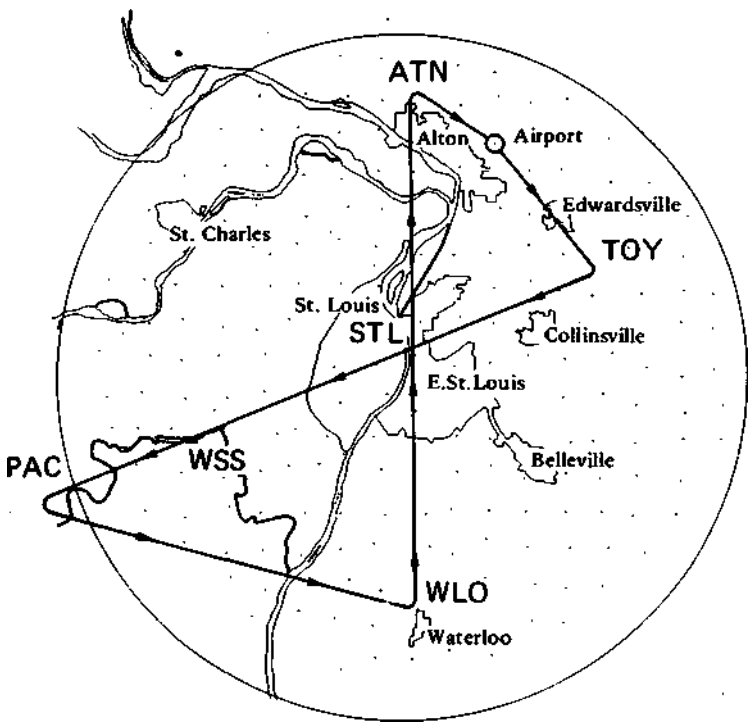

Figure 10. Flight path for routine data acquisition over St. Louis

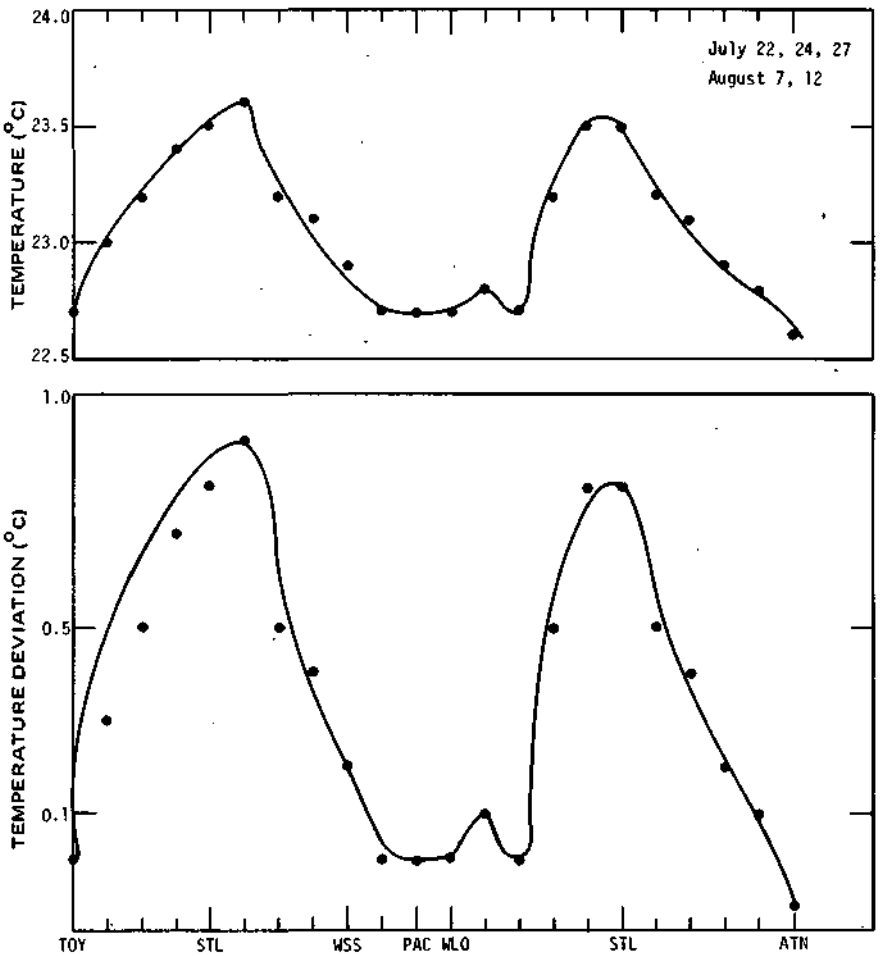

Figure 11. Temperature and temperature deviations from the rural values along the flight path shown in Fig. 10

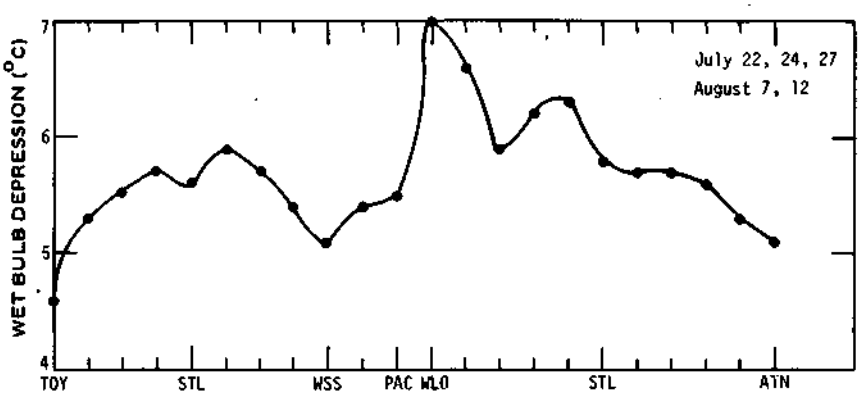

Figure 12. The wet bulb temperature depression along the X-pattern flights over St. Louis

Fig. 13 depicts these two variables with respect to location in the X-pattern configuration shown in Fig. 10.

The WSW-ENE leg from Troy to Pacific does not indicate a marked difference in mixing ratio across the city as was obvious in Fig. 11 with temperature; the minor perturbations are too small to be attributable to any particular source. The steady increase in mixing ratio going from south to north shows little resemblance to the corresponding temperature changes for the same periods although there is an obvious decrease in moisture south and west of the urban region. If one accepts the temperature changes as being indicative of an urban influence for these data, and there appears to be substantiation for this, then the dissimilarity of the temperature and mixing ratio traces precludes any obvious urban influence on the moisture field.

5.1.3 Condensation Nuclei Data

The condensation nuclei (CN) data reported on here were taken with an Environment I continuous condensation nuclei counter; the relative humidity attained during measurements was approximately 350%, so, it may be assumed that essentially all sub-micron particles are activated and counted with this instrument. One obvious purpose for the analysis of the CN data obtained with the aircraft is to attempt a delineation of the urban effluent under varying meteorological conditions. Figure 14 shows the average CN values for dates indicated; there are two regions of maximum CN values, one located immediately across the Mississippi River from downtown St. Louis obtained as the aircraft was heading WSW from Troy (Fig. 10), and the other a general increase from Waterloo to approximately 8 miles south of the downtown St. Louis area. Two distinct minima exist also; one is at Pacific which is approximately 30 miles WSW of the Arch and the other is located immediately north of Granite City. The Pacific minima is not unexpected, nor is the general decrease from the Arch to that point; but, the marked decrease from approximately 3 miles south of the Arch to just north of Granite City is rather surprising; however, examination of heavy industrial area locations indicates a distinct lack of activity in this region (Fig. 15).

The following four figures depict the CN profile under varying low level wind conditions. The value at Pacific in Fig. 16 appears to be

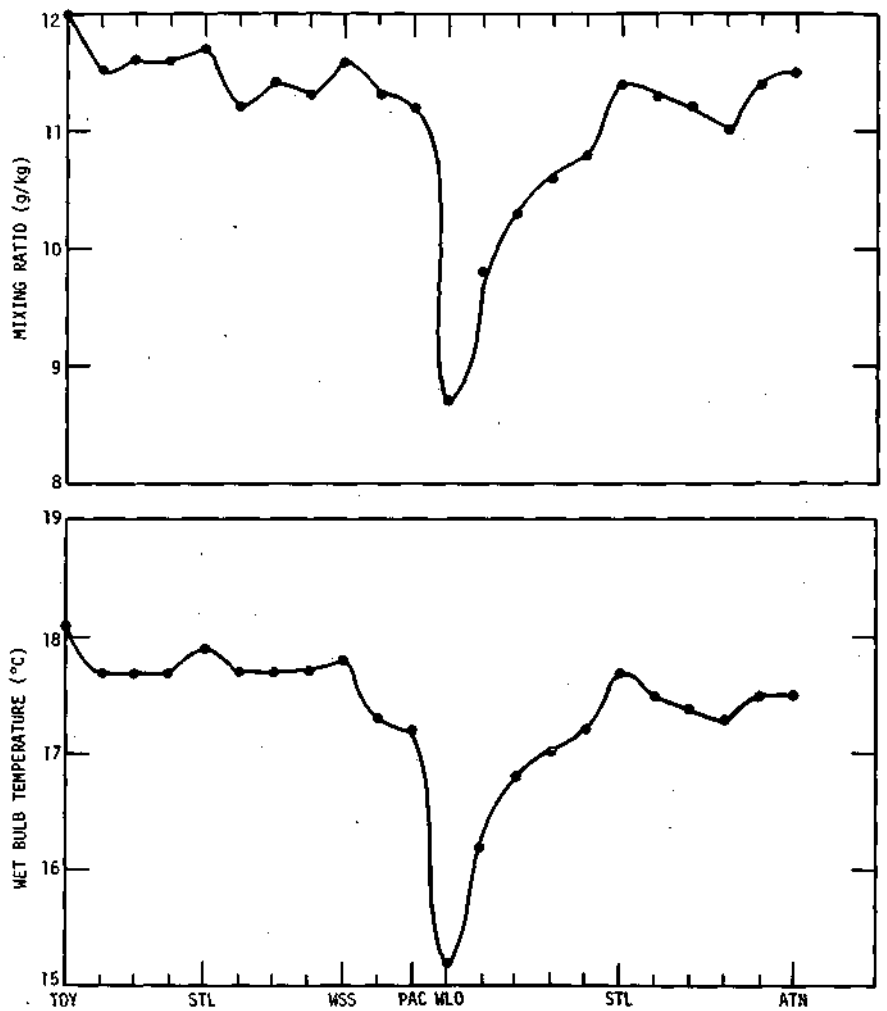

Figure 13. The mixing ratio and wet bulb temperature derived from the data in Fig. 12

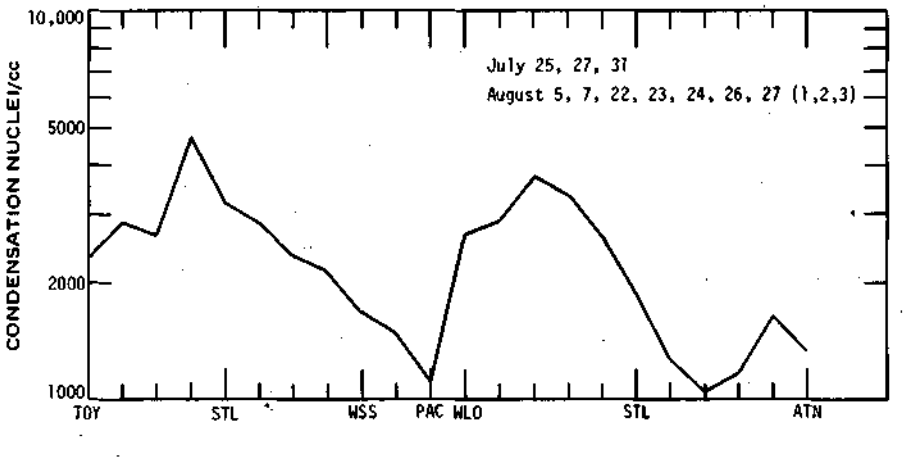

Figure 14. The average condensation nuclei concentration along the flight path shown in Fig. 10

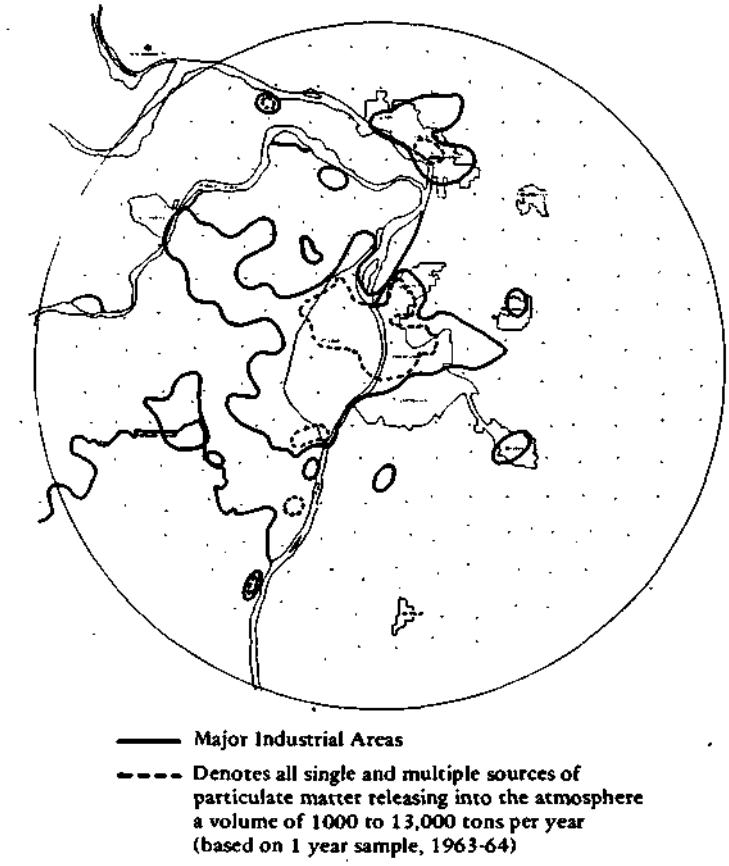

Figure 15. Major industrial areas of the St. Louis area and area sources of particulate matter greater than 1000 tons emitted per year

consistent with the average wind flow on the days examined; with northeasterly flow at low levels, the Pacific CN value is relatively higher when compared to surrounding readings, than on days with a different flow pattern (Figs. 17-19). This is reasonable in light of the position of this region with respect to the downtown St. Louis area. With NNW flow (Fig. 17) the aircraft appears to be crossing the urban plume as it tracked WSW reaching a minimum at Pacific; it is difficult to determine whether Pacific is or is not in the urban plume; if so, then it is likely on the extremity of the plume in light of the low reading. However, there is little doubt that Pacific is well within the plume in Fig. 16 with ENE flow and CN values very slowly decreasing from the Arch (St. Louis) to Pacific; the Pacific reading is an order of magnitude higher than the lowest reading of the day. It should be noted that the values drop off rather sharply as the aircraft left Pacific heading ESE toward Waterloo, apparently indicating an extremity of the urban plume. The much lower value between Waterloo and St. Louis is strong evidence that this location is out of the plume.

5.2 Weather Summary-1972

The first eight days of June were dominated by an upper ridge to the west of St. Louis (STL) keeping the Midwest in dry northwest flow aloft. This dry pattern resulted in only a trace of precipitation in the STL area during this period as frontal passages produced little or no rain in the Midwest. This general 500-mb pattern of a weak to moderate ridge to our west and a long wave trough along the east coast continued for the next four days with 0.2-inch of precipitation occurring during this time.

The upper air pattern began to change on 13 June as a long wave trough developed to the lee of the Rockies and passed through the Midwest during the ensuing four days. A slow moving Pacific cold front associated with the long wave trough passed through the STL area late on 14 June causing widespread, showery precipitation with approximately 0.3-inch falling at STL. Another long wave trough appeared off the Pacific coast and progressed across the country from 17-21 June in a manner similar to the last system with an associated cold front passing through the Midwest 19-20 June; approximately 0.5-inch of rain fell in the STL region as a result of this system.

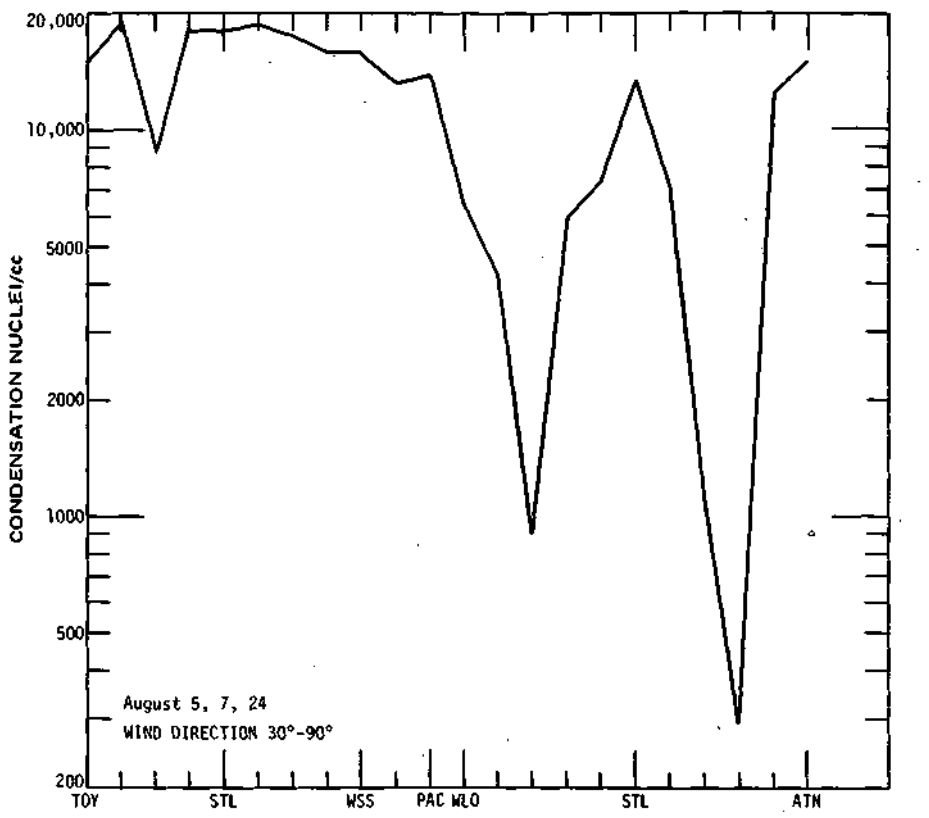

Figure 16. The average condensation nuclei concentration at 1800 ft. MSL with low level winds from 030° to 090°

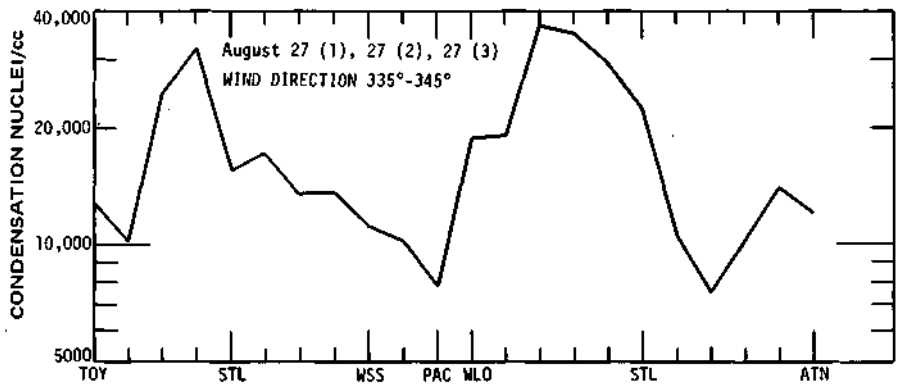

Figure 17. Same as Fig. 16 with low level wind from 335° to 345°

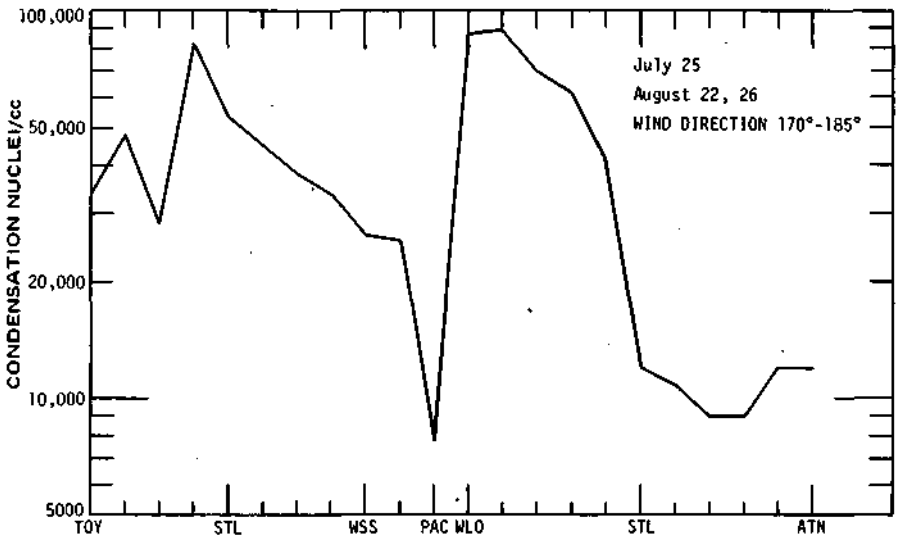

Figure 18. Same as Fig. 16 with low level wind from 170° to 185°

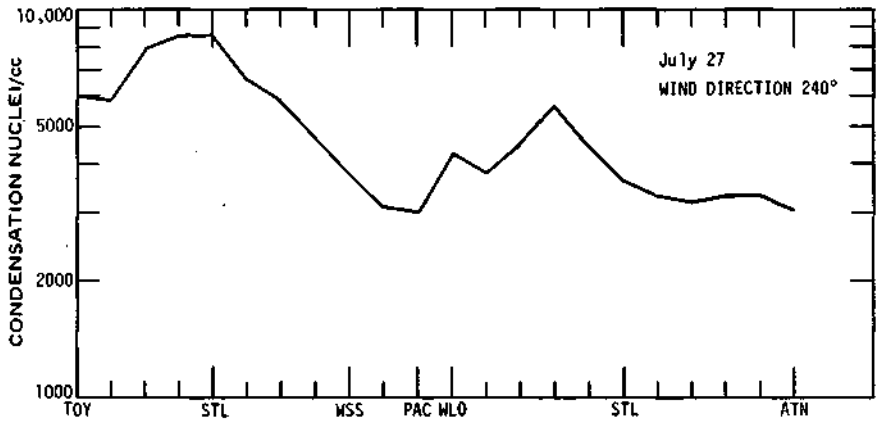

Figure 19. Same as Fig. 16 with low level wind from 240°

The next six days were characterized by two long-wave troughs along both coasts with a ridge of high pressure in between. This resulted in dry, stable conditions in the Midwest with northerly flow; during this period there was approximately 0.02-inch of precipitation in the STL region. Both troughs migrated slowly eastward during the next several days; however, the general pattern of a lack of moist flow from the south-southwest continued with only an additional 0.07-inch of rain occurring for the month in the STL area on 28 June as a weak Pacific cold front passed through the Midwest.

Precipitation for the month at STL (Lambert Field) totaled 1.19 inches which was 28% of normal for June. The upper air flow pattern resulting in generally northwesterly flow over the Midwest brought about the sparse rainfall in the region.

The passage of a short-wave trough with an associated cold frontal passage on 2 July brought over an inch of precipitation to the STL area. Overrunning from that same front, as a minor disturbance along the front in Oklahoma moved east-northeast, caused additional rainfall on 4 July in the form of light rainshowers in the STL area amounting to approximately 0.2-inch.

A weak but persistent upper ridge in the southwest coupled with a slow moving surface high pressure system, brought generally dry conditions with northwesterly flow to the Midwest. A minor short-wave trough interrupted the dry flow and caused light precipitation on 7-8 July, however, this was of little consequence. A rather flat, dry upper air pattern developed and persisted for the ensuing six days with only a trace of precipitation from 9-14 July in STL. However, a developing long-wave trough in the mid-section of the country associated with a slow moving pacific cold front produced moderate precipitation amounts in the STL area on 15 July; this pattern continued for the next four days as the Midwest remained in southerly flow aloft bringing moist, Gulf air into the region. During this period, approximately 1 inch of rain fell in STL.

The ensuing six days were marked by a retrogression westward of the long-wave trough and a building of a ridge in the southeast United States; this resulted in subsiding air and a cut-off of Gulf moisture to the Midwest. As a result, less than 0.1-inch of precipitation fell in the STL region during this time. As the upper ridge to the southeast began to flatten

bringing about zonal flow across much of the country, a vigorous short-wave trough coupled with a stationary front in the Midwest brought moderate-heavy precipitation to the STL vicinity on 26 July. Lighter amounts were added during the next two days associated with a stationary front just south of STL; nearly 0.75-inch fell in STL during this period.

The next five days were characterized by an upper ridge to the southwest and a strong surface high over the Midwest resulting in dry, subsiding airflow and no precipitation in the STL area. The weather during the next two days was influenced by another active short-wave trough passing through the Midwest coupled with a slow moving cold front; the result was approximately 1.5-inches of rain in STL from 2-3 August.

The following six days were marked by the development of a long-wave trough in the eastern third of the United States which moved very slowly eastward during this period. A complex surface frontal system produced moderate-heavy rainfall amounts in the Midwest on 6 August; a cold frontal passage on 8 August added approximately 0.04-inch. The total rainfall during this period was approximately 0.85-inch. As the trough axis passed through the Midwest, northwesterly flow prevailed for the next two days with no precipitation; a short-wave trough superimposed upon the northwesterly flow aloft triggered thunder showers on 11 August.

The weather pattern returned to a prolonged, dry situation as a ridge in the middle of the country was maintained for the next nine days; the main frontal activity stayed well to the north of STL resulting in <0.02-inch of rain during this time. On 21 August the pattern began to shift as a long-wave trough developed in the middle of the country; this continued until 25 August bringing moist southwesterly flow into the Midwest resulting in approximately 0.4-inch of rain during this period. The remainder of the month was characterized by a strong ridge aloft in the south-central portion of the country resulting in no precipitation in the STL area.

The July precipitation at Lambert Field totaled 3.10-inches which was 94% of normal, while August rainfall amounted to 2.69-inches which was 89% of normal.

5.3 Aircraft Program-1972

The aircraft utilized during METROMEX '72 was a Piper Navajo operated by Atmospherics Incorporated under a subcontract. The main mission of the

aircraft during the six week period of 6 July - 15 August, as it was in 1971, was to inject tracer material into convective cells that passed over the rainwater collection network. Four additional priorities were also included. These were: 1) measure cloud bases over the urban area and the surrounding rural area to determine possible differences in the two; 2) map convective cells as to updraft speed, size, location, and duration; 3) identify and locate major particulate sources in the St. Louis urban area; and 4) determine general cloud structure characteristics in the region. The aircraft logged 44 flights during METROMEX '72 including 5 tracer missions. Two tracer materials were used during those missions, a solution of lithium chloride which was burned as an aerosol, and solid indium oxide flares.

5.3.1 Cloud Base Study

A total of 82 cloud base measurements observed over 19 days, were made by the AI aircraft to fulfill priority (1) above; these are plotted in Fig. 20 and isopleths of equal cloud base height are drawn. The data strongly suggest the presence of higher cloud bases in the urban area when compared to rural values, the maximum difference being approximately 2000 ft. The times of observation ranged between 1300-1600 CDT; the measurements were random with respect to time and place.

5.3.2 Updraft Measurements

Updraft measurements were made on 12 days and are plotted in Fig. 21; there is no apparent pattern to the distribution; as a result, no isopleths are drawn. However, the following observations were made by the AI crew.

1) Most inflow locations, even in vigorous thunderstorm activity, gave the impression of small diameter areas with short duration inflow pulses.
2) Most significant inflow areas, observed and measured, were on the leading edge of squall lines with little activity on the trailing edge; inflow on the lack of cumulus cells was found on systems that contained vigorous feeder clouds.
3) No significant inflow areas were found on weak or dissipating mature thunderstorms.

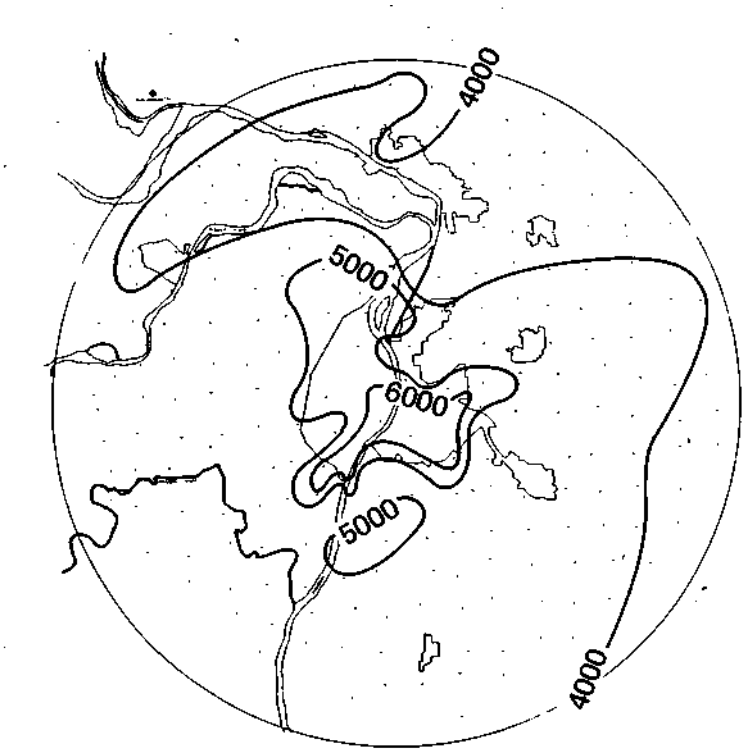

Figure 20. Isopleths of cloud base height (feet) as determined from 82 clouds observed on 19 days

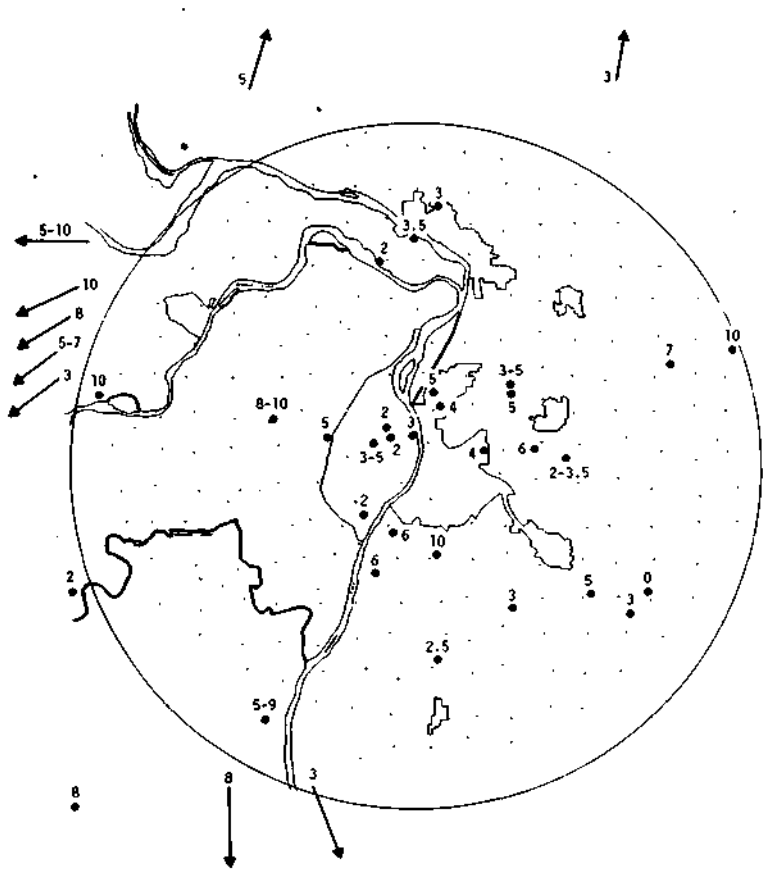

Figure 21. The updraft speeds (100 's fpm) observed on 12 days. The arrows indicate observations off the scale of the map.

4) With a few exceptions, there were no significant inflow areas at mid-levels of moderate to large cumulus cells. In those cases, inflow was evident very near the rising turrets and approximately 2000-3000 ft below cloud top.

5.3.3 Particulate Source Mapping

The locations of 25 major particulate sources in the Alton-St. Louis area are shown in Fig. 22. With a few exceptions, all of the major sources are located along the Mississippi River. There is an obvious lack of major industry in an area between Granite City and Wood River.

5.3.4 Cloud Structure

It was observed by the AI crew that major differences exist in the urban affected clouds when compared to those which are not affected. Further, these differences appear to be evident in the lower 2000 ft of cloud. At these levels, the urban clouds appear to be diffused, fractured and without well defined inflow areas. This appearance seems to be related to the type and concentration of particulates released by the industries in the area. The diffused cloud base may be the result of differing activation supersaturations required by the variety of particulate emissions.

It was noted that well defined solid bases existed on days when cumulus clouds developed totally above the top of the haze layer where particulates were at background levels found in rural regions.

5.4 Surface Data

5.4.1 Wind - 1971 and 1972

5.4.1.1 Data Collection and Analysis

The operational period for the wind sets was from mid-June until 1 September 1971 and from 1 June until 1 September 1972 with the radar headquarters equipment in continuous operation since mid-June 1971. Abstraction of the data on a complete basis was limited to the summer months of June through August of both years.

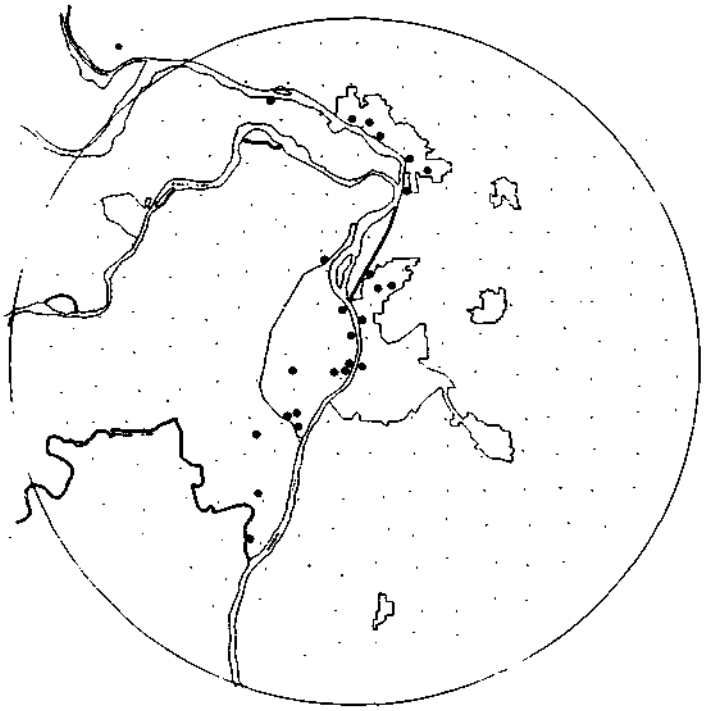

Figure 22. The locations of 25 major particulate point sources in the St. Louis industrial region

The abstraction of the data was patterned after the procedure in use at the St. Louis station of the National Weather Service, i.e., hourly values of speed and direction averaged over the period from 10 minutes before the hour until the hour.

5.4.1.2 Results

June 1971. All stations indicated a predominantly southerly component to the winds recorded during the month, although there was considerable dispersion toward easterly and westerly components in this southerly flow. Pere Marquette was the only station indicating some northerly winds. Wind roses have been plotted in Fig. 23 with the roses centered upon the station locations. The recorder at Waterloo, Illinois was installed too late in the month to establish average wind directions.

Wind speeds averaged between 4 and 9 mph with the highest wind speeds of 16-18 mph measured at PMQ and Alton from northerly directions.

July 1971. The recorder at Weiss Airport was inoperative during most of this month and its record is not shown.

The wind directions shown in Fig. 24 for July 1971 were confused, with strongly northerly and southerly winds at PMQ, northerly and southeasterly at Alton and Nagel. Waterloo and Spirit of St. Louis indicated somewhat of a predominance of southerly flow. The highest wind speeds were measured at Alton and PMQ at 22-24 mph.

August 1971. All of the stations were in operation during August 1971. The patterns of wind direction from these stations are interesting with both north and south directions predominating at PMQ, NNE and south at Alton, NNE at Nagel, and Waterloo, SSE at Weiss, and SW at Spirit of St. Louis. However, at all stations the northerly winds were the strongest measured. The wind roses for August 1971 are shown in Fig. 25.

June 1972. The wind roses for June 1972 are shown in Fig. 26. Alton and Nagel had NNW winds and PMQ had NNE winds of some noteworthiness. The winds at Spirit of St. Louis had no predominate direction. The predominate wind at Weiss was from the WNW and the SW at Waterloo.

July 1972. The predominate wind direction at all stations in July was from a southerly quadrant as shown in Fig. 27. All stations except Weiss had a westerly component. Weiss had a southeasterly component.

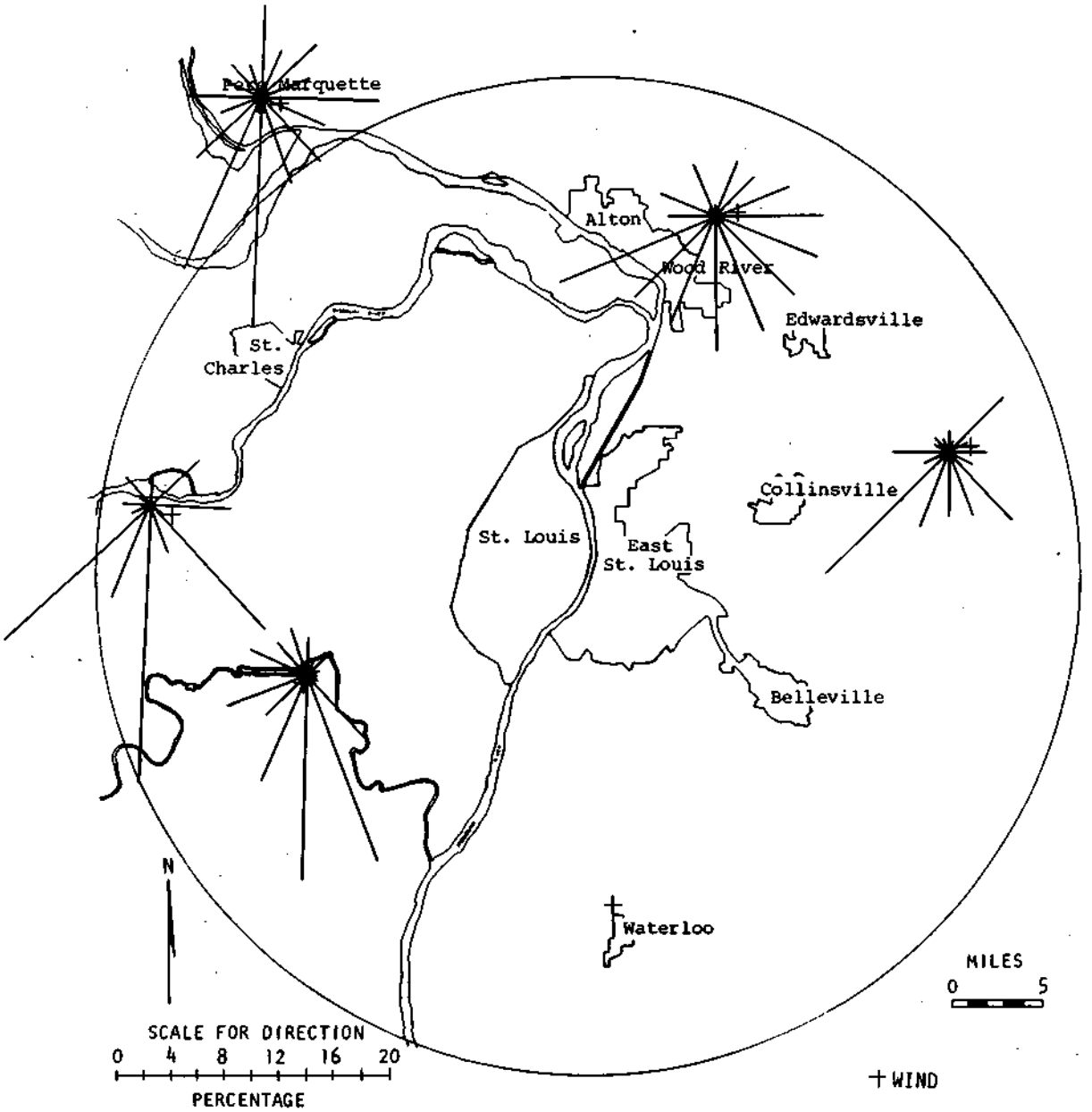

Figure 23. The June 1971 wind roses for METROMEX wind recording sites

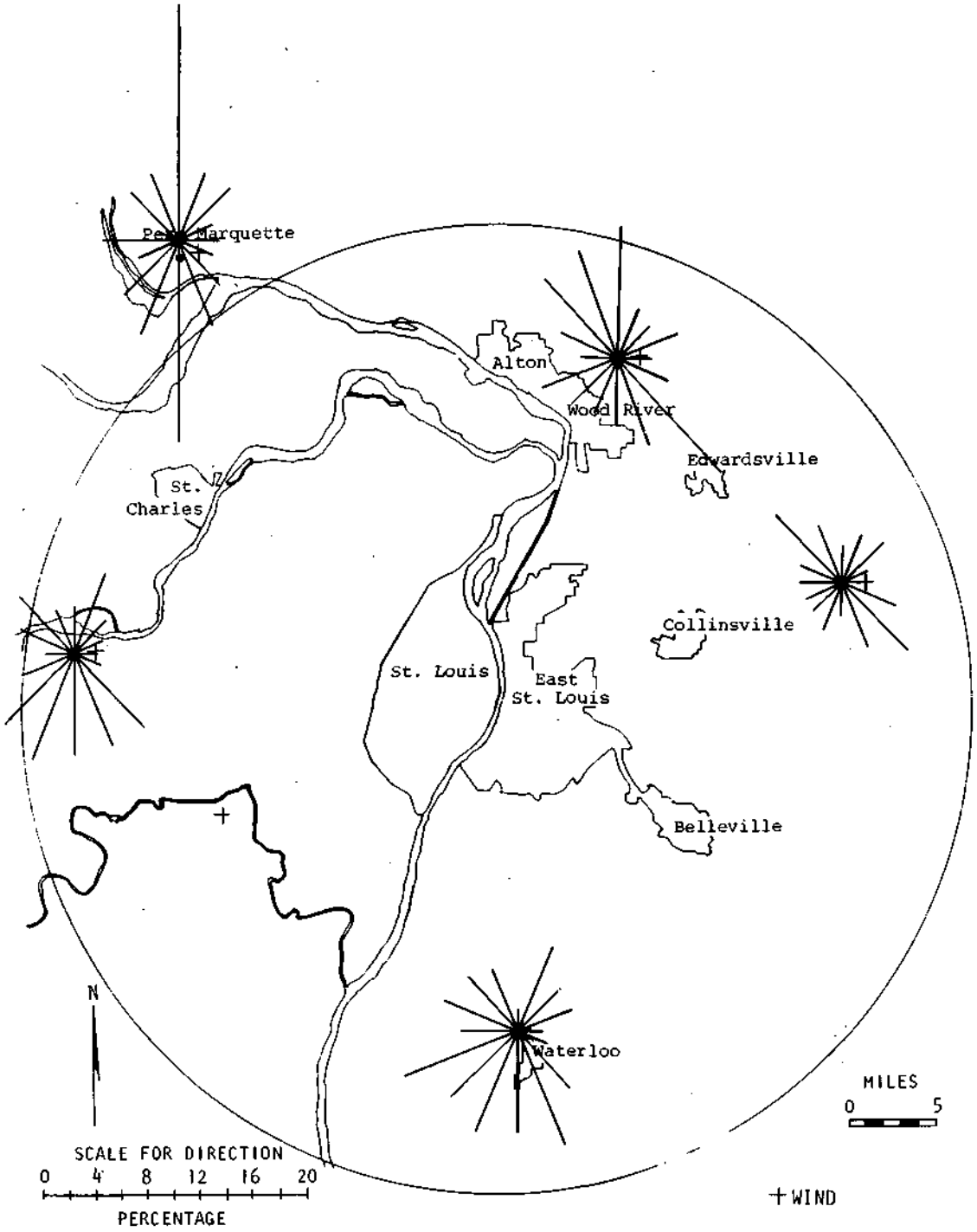

Figure 24. Same as Fig. 23 for July 1971

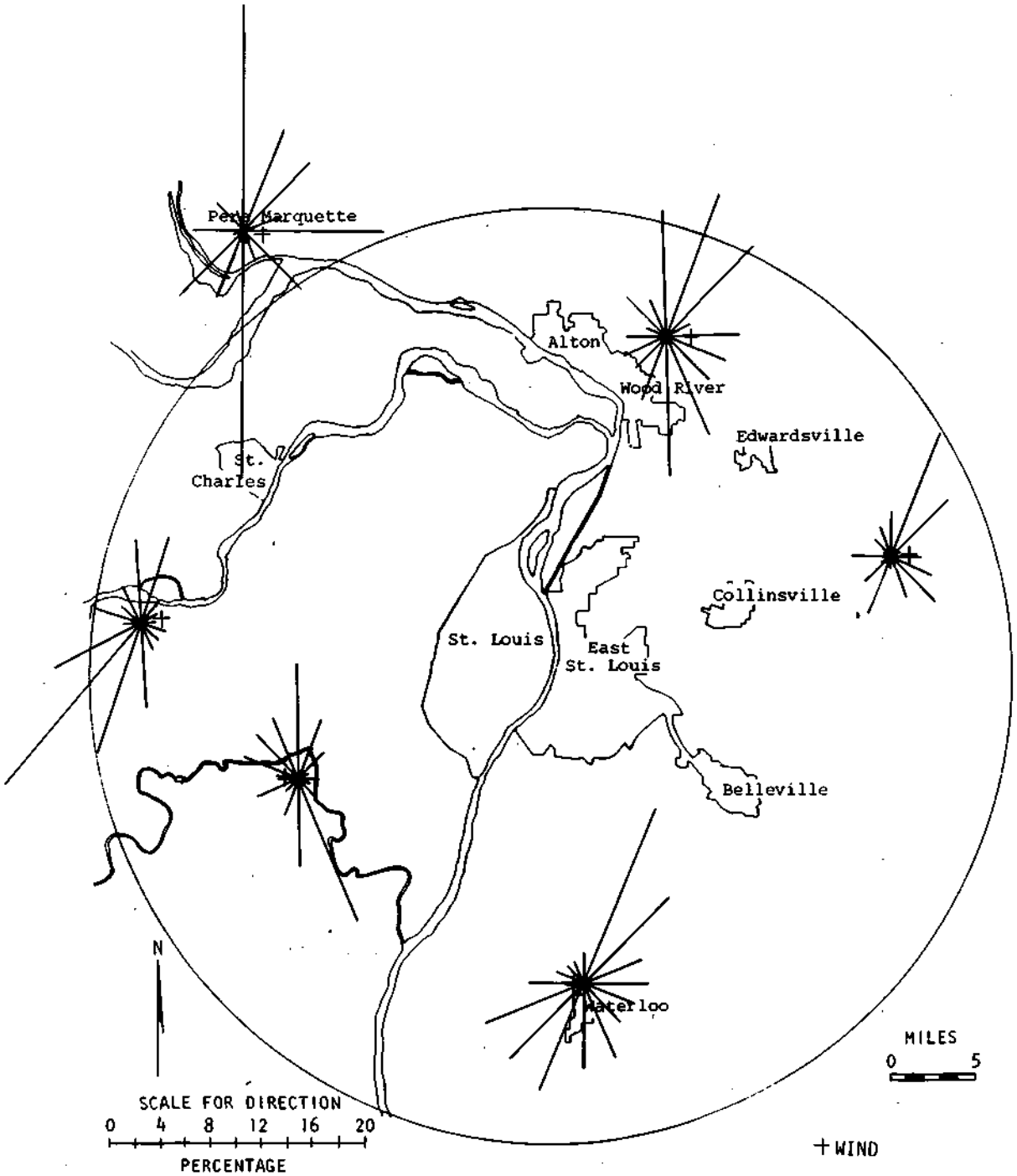

Figure 25. Same as Fig. 23 for August 1971

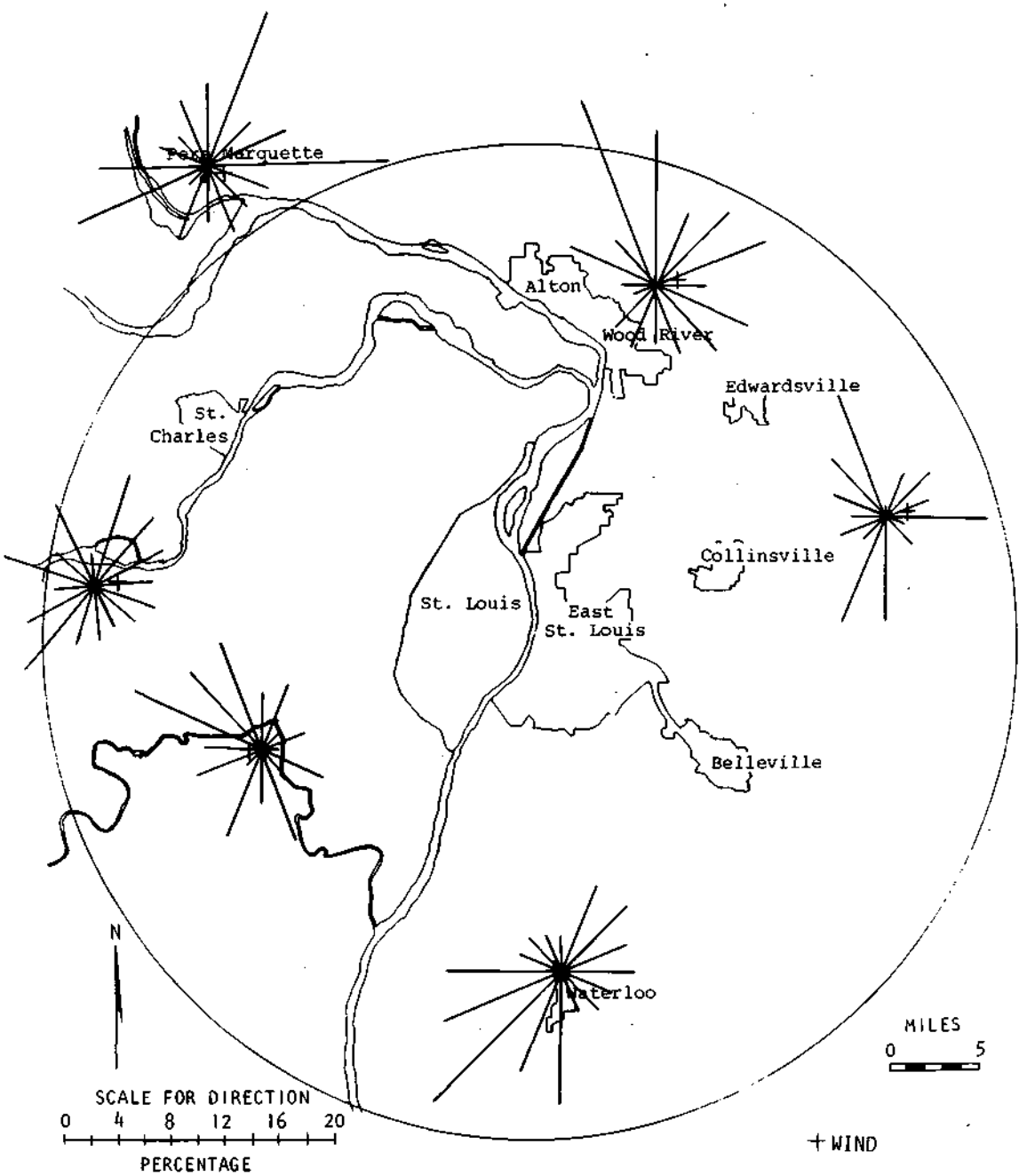

Figure 26. Same as Fig. 23 for June 1972

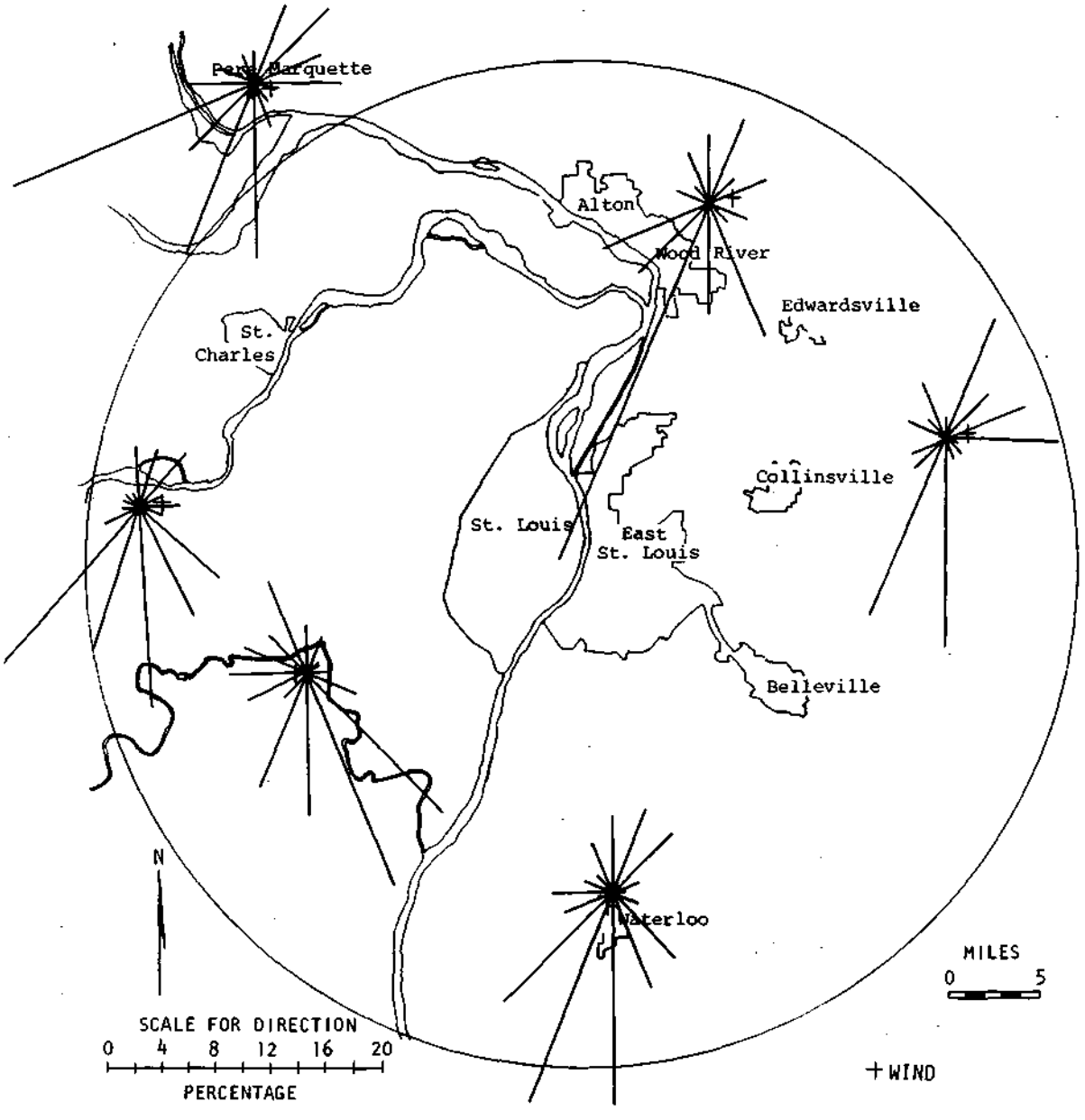

Figure 27. Same as Fig. 23 for July 1972

August 1972. The predominate wind direction at all stations in August as in July was from a southerly direction as shown in Fig. 28. The peculiar omission of the south wind at PMQ, Spirit of St. Louis, and Alton raises the question of bias on the part of the person extracting the data from the charts. The bias seems to obtain at the primary points of an 8-point compass, i.e., 0°, 45°, 90°, 135°, 180°, 225°, 270°, and 315°.

5.4.1.3 Discussion

The wind set at Nagel averaged a state of calm winds approximately 40% of the time during the summer of 1971. During 1972, the June average was 30%, July 18%, and August 4%. Such protracted periods of little wind movement seems unreasonable even though the Nagel site is somewhat sheltered. The 4% calm average in August 1972 seems to be more normal. Unless the lower speed winds came from preferred directions, there should be no bias in the direction data from Nagel. In contrast, the recorders at PMQ, Waterloo, and Alton in much more exposed sites rarely experienced calm conditions.

In general, the two years of wind direction data do not show a trend which would indicate that a particular site was biasing winds blowing over it, except in the case of Weiss in August of both 1971 and 1972. The Weiss data do not agree with the surrounding stations in indicating a southwesterly wind, but showed a southeasterly wind instead. The expected biasing at PMQ and Nagel did not appear in direction. The Nagel instrument may have been insensitive, but this seems to have been corrected during summer 1972.

5.4.2 Temperature and Humidity - 1971 and 1972

5.4.2.1 Status of Analysis

All summer data from the 25 sites have been extracted from the charts and the corrections made. These corrected data will be punched into cards and machine calculation of the dew point temperature for each hour at each station will be performed and punched into the cards.

The temperature data at three-hourly intervals from the hygrothermograph network of summer 1972 have been averaged for each station by months. In general, the three-hourly temperature fields in the three months differ only in the temperature values; the patterns are the same. Only 17 sites were in

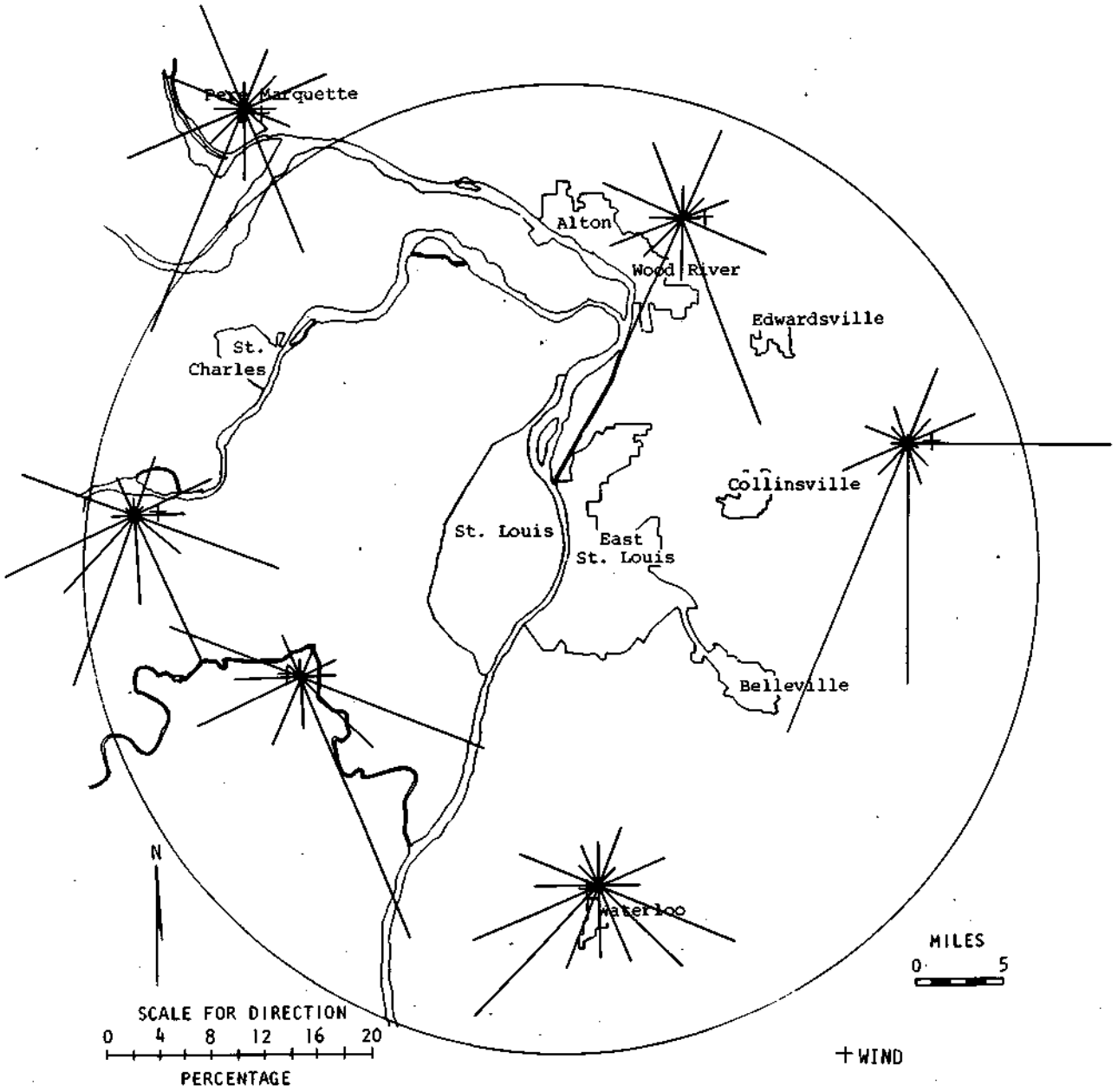

Figure 28. Same as Fig. 23 for August 1972

operation with sufficiently complete records during June and July. The additional 8 sites were instrumented in. late July. Hence, the greatest detail of pattern is to be found in the August data, and two hours from that month have been chosen as illustrations of the patterns found in the research circle. This is the mean pattern for 0600 CDT, representing the hour of transition between nocturnal radiational cooling and solar isolation, and the mean distribution for 1500 CDT, representing the hour of maximum solar isolation (Figs. 29 and 30, respectively).

5.4.2.2 Some Results

August Mean Temperature Field at 0600 CDT. The greatest contrasts in temperatures between the urban complex and the surrounding rural areas occurs at dawn (10.6°F) as has been noted by other investigators. The measuring site at the Gateway Arch had the highest minimum temperature of 72.2°F. The coldest temperature (61.6°F) was measured at two rural sites, stations 17 and 117. The first station is at the Machens siding in the Missouri-Mississippi floodplain southeast of Pere Marquette and the second south of the city of Collinsville. A third cold spot was found south of St. Louis along the Mississippi River, The radar headquarters site in Pere Marquette State Park (PMQ) exhibits the expected reversal of temperature between day and night in comparison with plain or valley locations; the hill temperature remains relatively warm at night and relatively cool during the day. It will be noted that the river valley stations at Weiss Airport, and the Spirit of St. Louis Airport were relatively warm at 0600 CDT.

August Mean Temperature Field at 1500 CDT. The difference between the warmest and coldest temperatures measured at 1500 CDT was 6.2°F. The urban heat island is still very much in evidence, but the warmest temperature measured was at Spirit of St. Louis Airport in the Missouri River Valley. The Machens elevator had a maximum temperature nearly as warm. This station had the greatest range in temperature of all measuring sites, 25.8°F.

5.4.2.3 Discussion

There were height differences between the several measuring sites, particularly between PMQ and the valley stations south of it. However, a thermal wind should be expected to be measurable in the areas of rapid areal

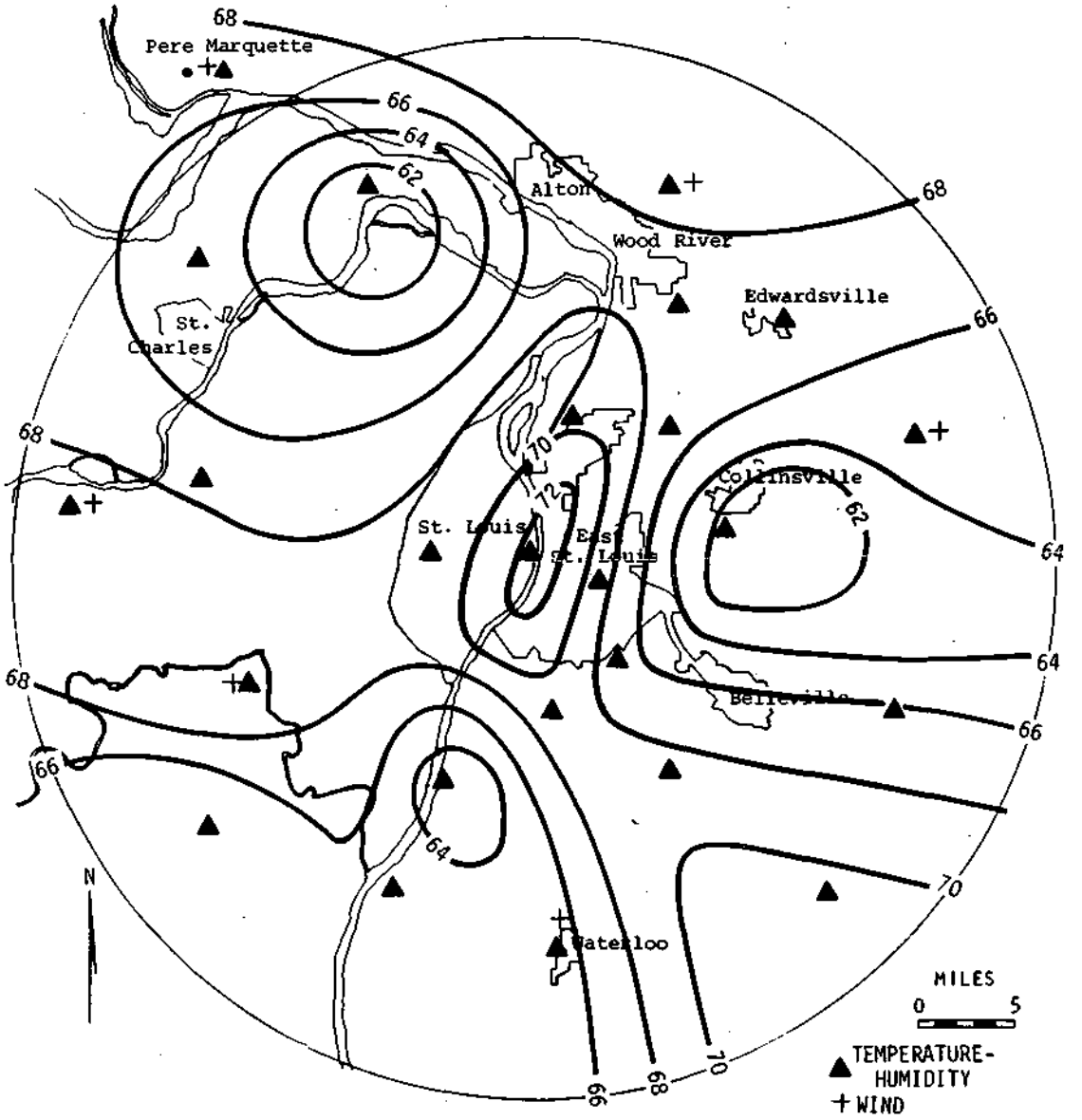

Figure 29. Mean surface temperature distribution (°F) at 0600 CDT for August 1972

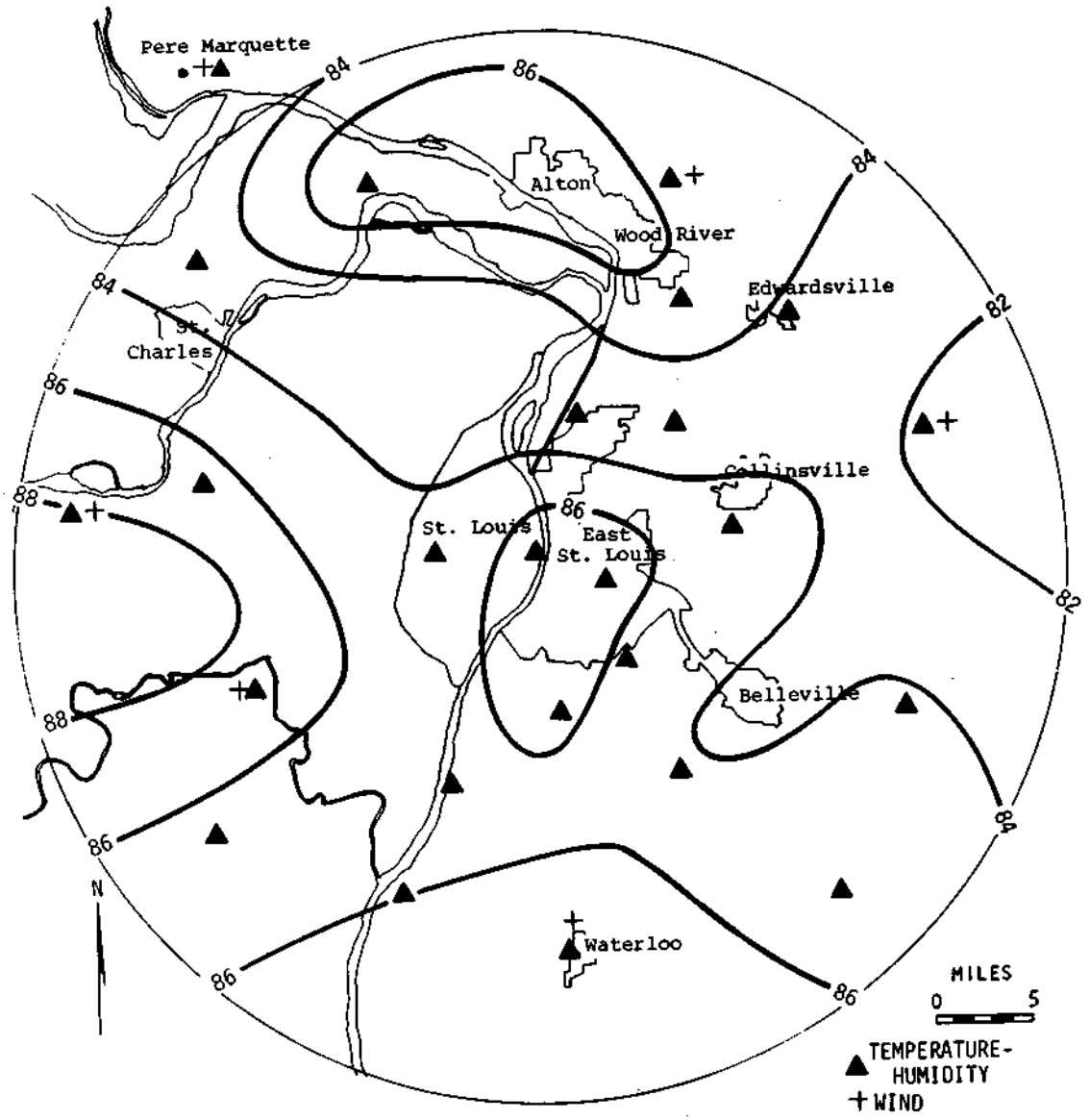

Figure 30. Same as Fig. 29 for 1500 CDT

temperature changes. Unfortunately, all of the wind-measuring sites were in areas of weak thermal gradient during 1972. It would be most desirable to place an anemograph between the Collinsville site and the Arch to take advantage of the strong gradient which should produce a wind vector counter to the prevailing southerly or southwesterly flow. The expected vector should be from the east, blowing from the colder, more dense air toward the relatively warm city.

5.4.3 Pere Marquette Nuclei Data - 1971

During METROMEX 1971 hourly, ground level, condensation nuclei measurements were made at PMQ for the period June-August; the data were taken with a Gardner Associates, small particle detector, type CN. The measurements were made hourly at one level of supersaturation (approximately 250%).

Average nuclei values per tens of degrees wind direction at PMQ are shown in Fig. 31 for the period June-August 1971; there is an obvious increase in nuclei counts in the southeast quadrant which is in the direction of the STL industrial complex. Major industrial regions in this area are shown in Fig. 15. There is little doubt that the concentration of particulate matter produced by the STL urban industrial complex is significantly above rural levels, and is detectable at a distance of at least 25 miles.

5.4.4 Raindrop Distributions - 1972

Since climatological data in the St. Louis area strongly indicate downwind increases in rainfall amounts, the microphysics of the precipitation process are likely altered. To investigate this possibility, raindrop distributions were measured in the St. Louis region; the distribution of raindrop spectrometers is shown in Fig. 8.

As of this report, data from two of the eight sites have been analyzed; these are at the Centreville (CEN) and Pere Marquette (PMQ) sites. The CEN location is within the climatological downwind precipitation maximum area while PMQ is sufficiently far from industrial areas to be considered rural. At these two locations, Joss spectrometers were utilized to determine the raindrop distributions; these devices were purchased commercially and function by converting the momentum of a raindrop impinging upon a sensor surface,

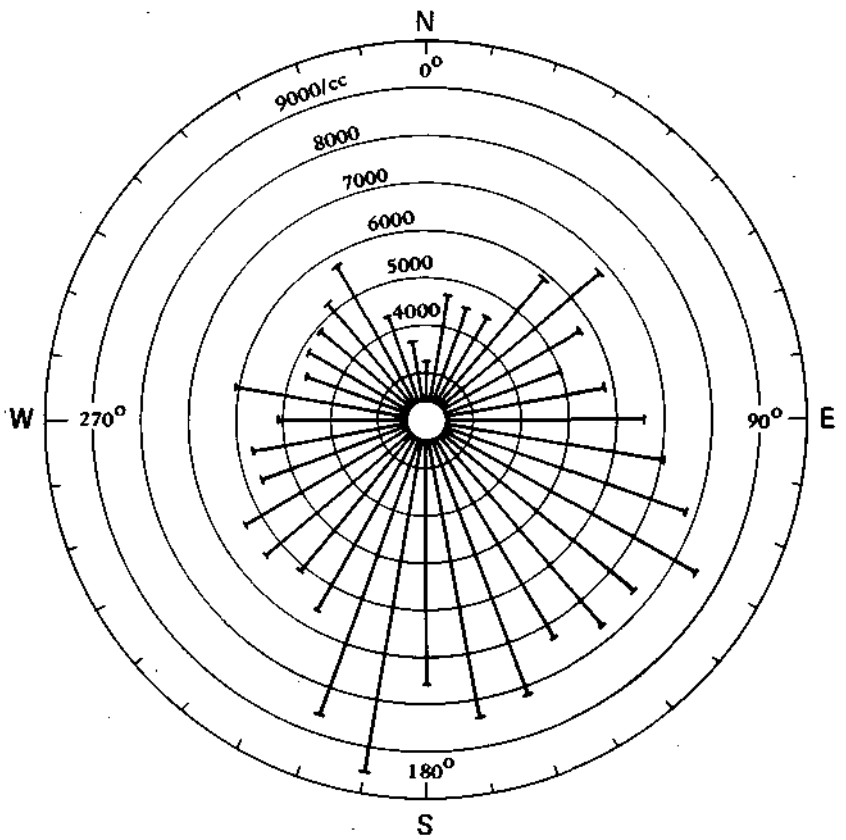

Figure 31. June, July, August 1971 average condensation nuclei concentration at Pere Marquette State Park field headquarters as a function of wind direction

to drop size. At the remaining sites, in-house manufactured devices operating on a similar principle were used.

During the METROMEX '72 field project period, raindrop data from 10 days at PMQ and 14 days at CEN were analyzed and compared; this represented a total of 275 five minute rain samples at PMQ and 422 at CEN. The total number of drops per 0.1 mm diameter intervals for all drops combined is shown in Fig. 32; if these totals are divided by the number of 5 minute samples, the average number of drops per 0.1 mm interval results, and is demonstrated in Fig. 33. The data clearly indicate an obvious difference in the distributions; the PMQ data show a much larger drop concentration although the average rainfall rate for that site is approximately 1/2 that of CEN. The radar reflectivity (Z) values also reflect the distribution differences; this parameter is approximately 5 fold greater for CEN. This is expected since Z is dependent on drop diameter to the sixth power and Figs. 32 and 33 reveal consistently greater numbers of the larger drop sizes.

During 8 rain events on 4 rain days, data were collected at both PMQ and CEN; this allowed a comparison of drop spectra for similar rain periods. Figures 34 to 41 compare average raindrop distribution for both locations for certain average rainfall rates. With the exception of Fig. 37, these individual comparisons agree with the general results, shown in Figs. 32 and 33, of greater drop concentrations at PMQ for similar rainfall rates.

Based on the data analyzed to date at these two locations, it is concluded that the downwind, urban affected location (CEN) has fewer, larger raindrops for similar rainfall rates as well as generally higher rainfall rates than the site at PMQ.

5.4.5 pH Measurements - 1972

5.4.5.1 Introduction

The rainwater chemistry project within METROMEX requires the collection of numerous precipitation samples over an area of 750 mi^2 to determine the spatial variability of several elements. Among these are the elements which are released into convective storms traversing the sampling network located immediately downwind of the St. Louis-East St. Louis metropolitan area.

During the summer of 1972, rainwater samples suitable for pH determination were obtained on 14 days. The maximum number of samples obtained from a given

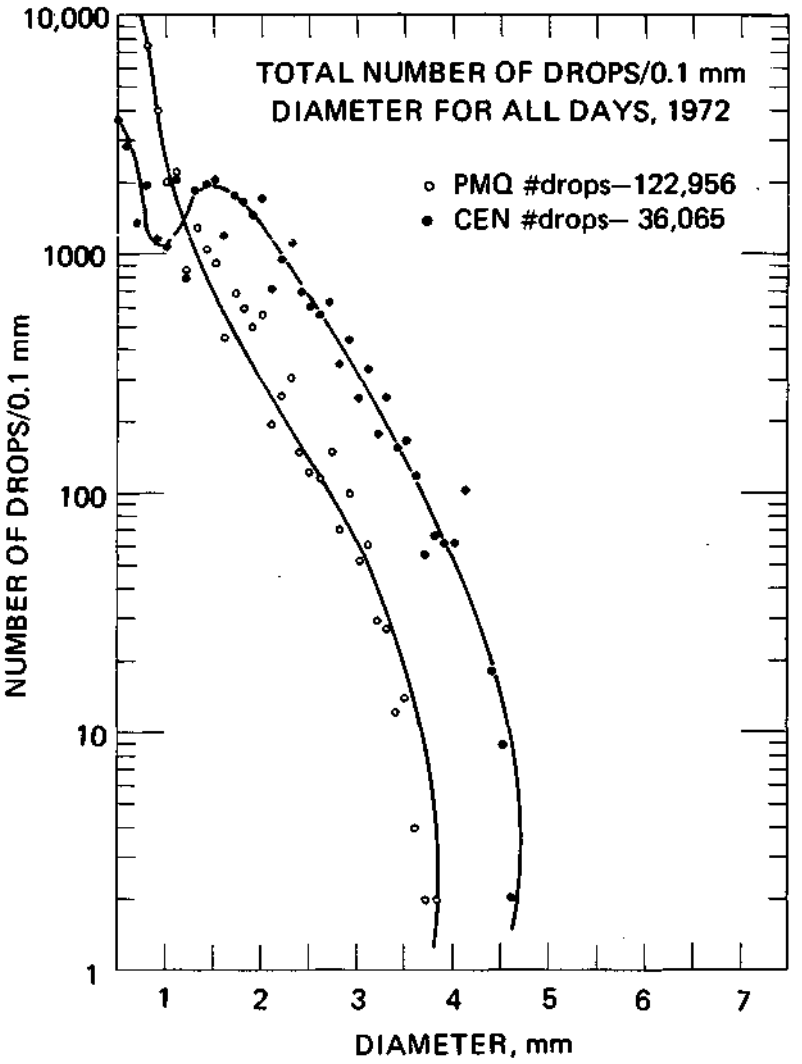

Figure 32. Accumulated raindrop size distributions obtained at Centreville and Pere Marquette during 1972

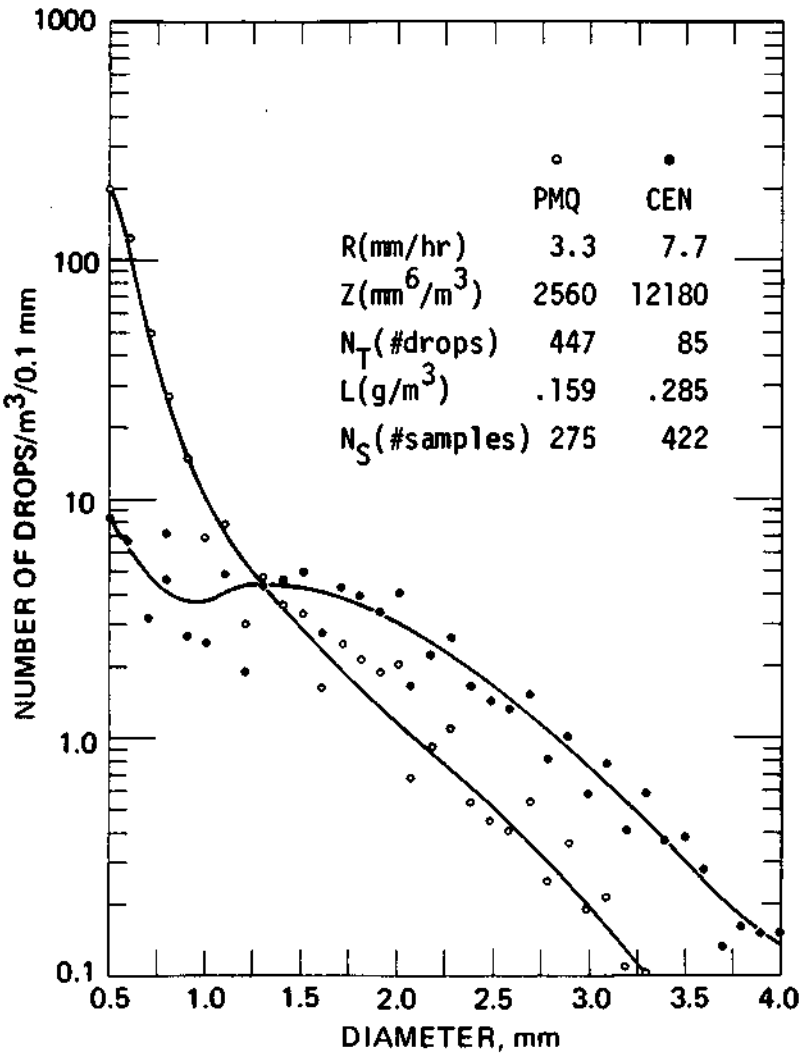

Figure 33. Average raindrop size distribution and derived variables obtained at Centreville and Pere Marquette during 1972

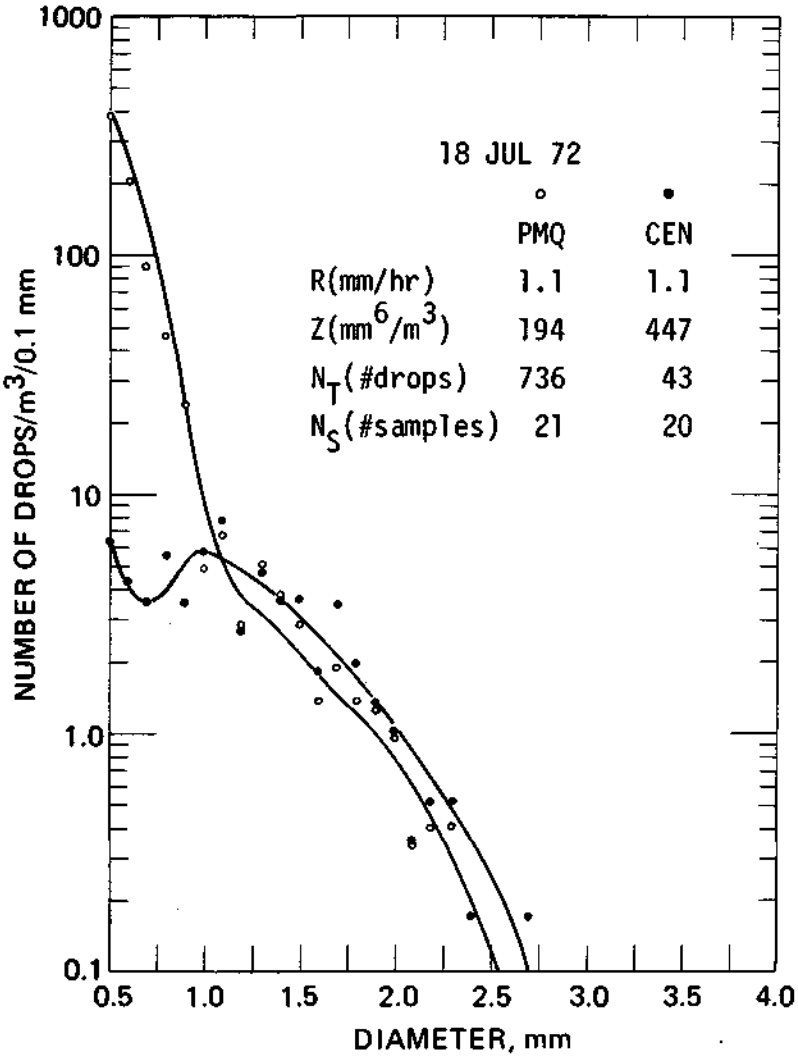

Figure 34. Raindrop size distribution observed in a storm of 18 July 1972 at both Centreville and Pere Marquette

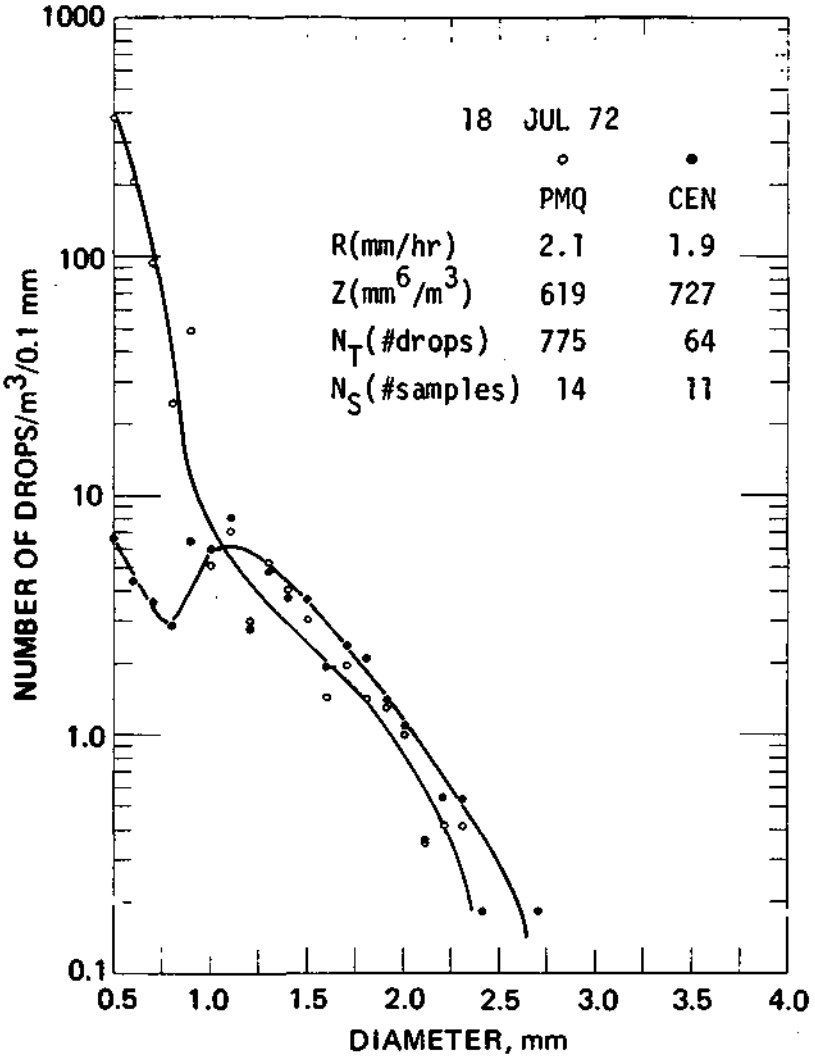

Figure 35. Same as Fig. 34, but a different rainfall rate

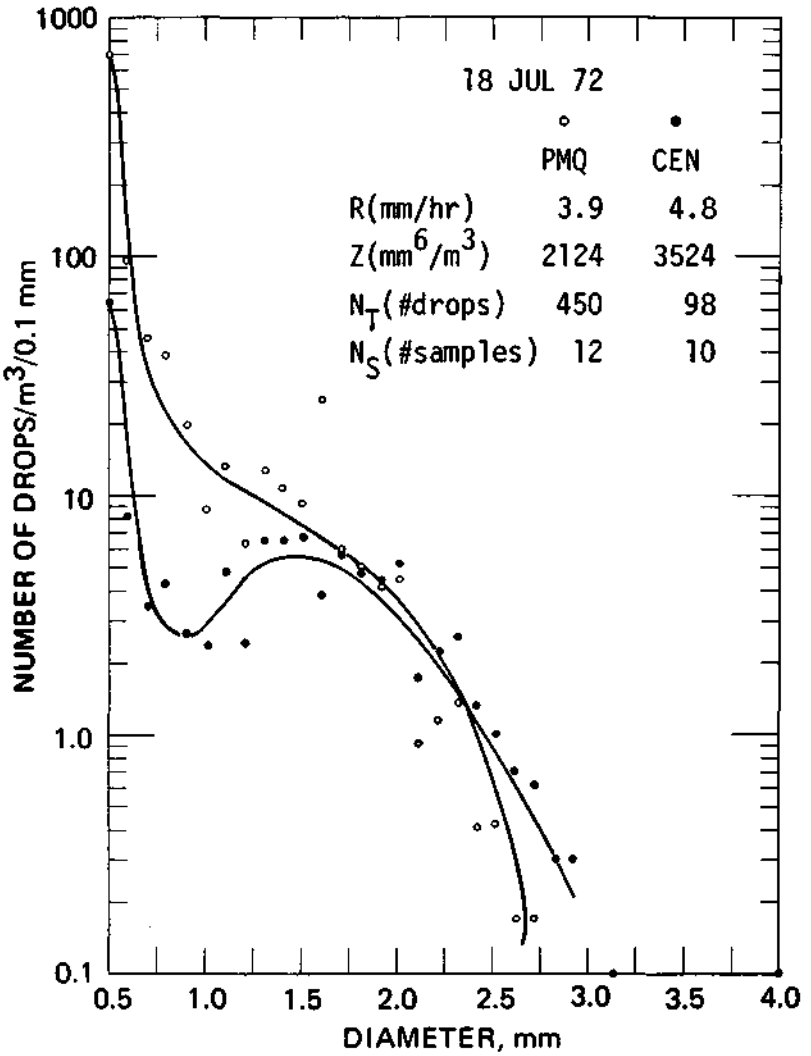

Figure 36. Same as Fig. 34, but a different rainfall rate

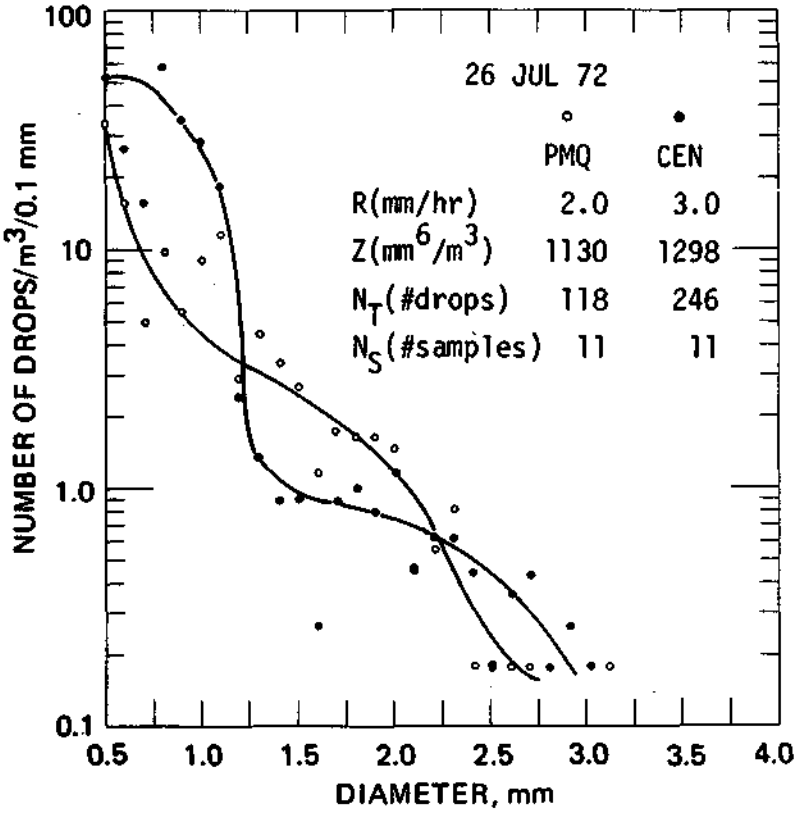

Figure 37. Same as Fig. 34 for 26 July 1972

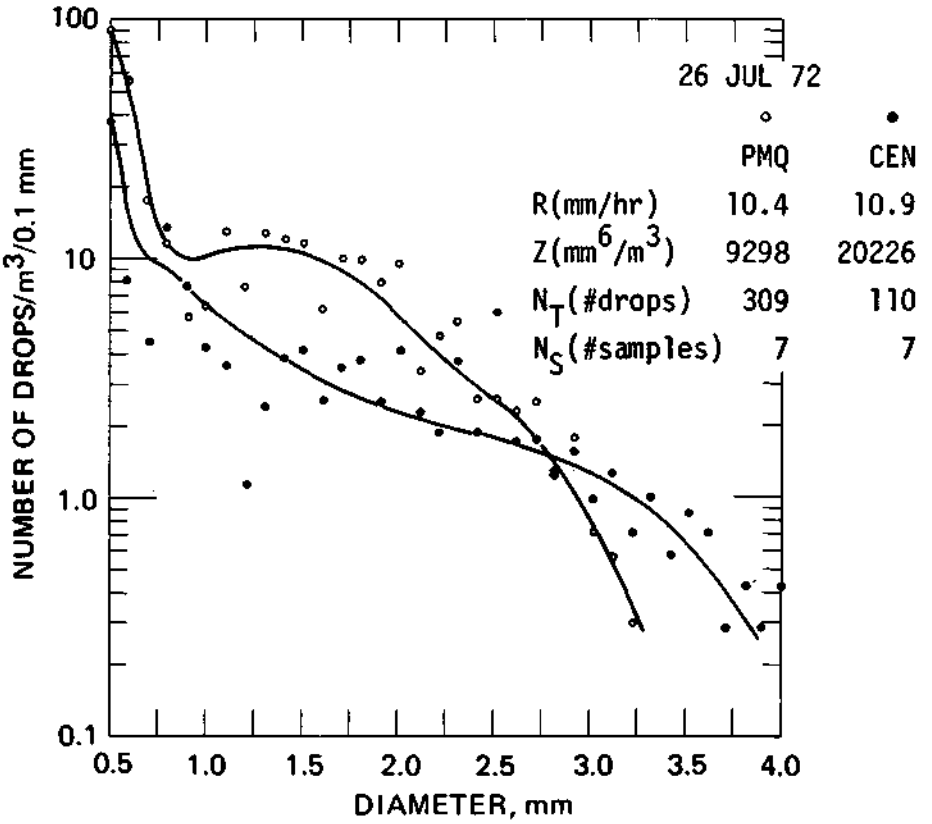

Figure 38. Same as Fig. 37 , but a different rainfall rate

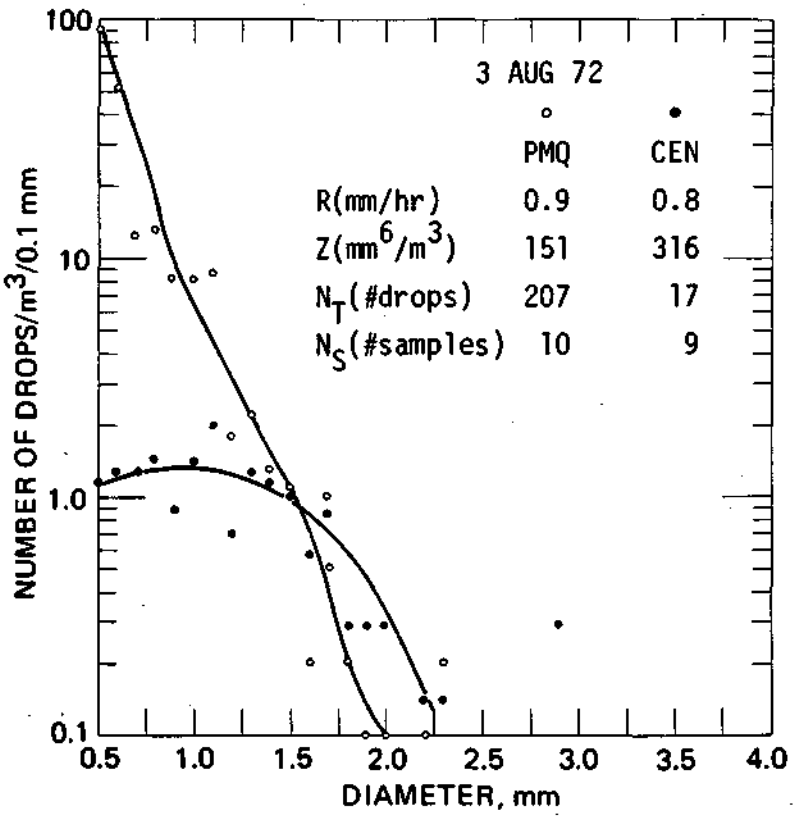

Figure 39. Same as Fig. 34 for 3 August 1972

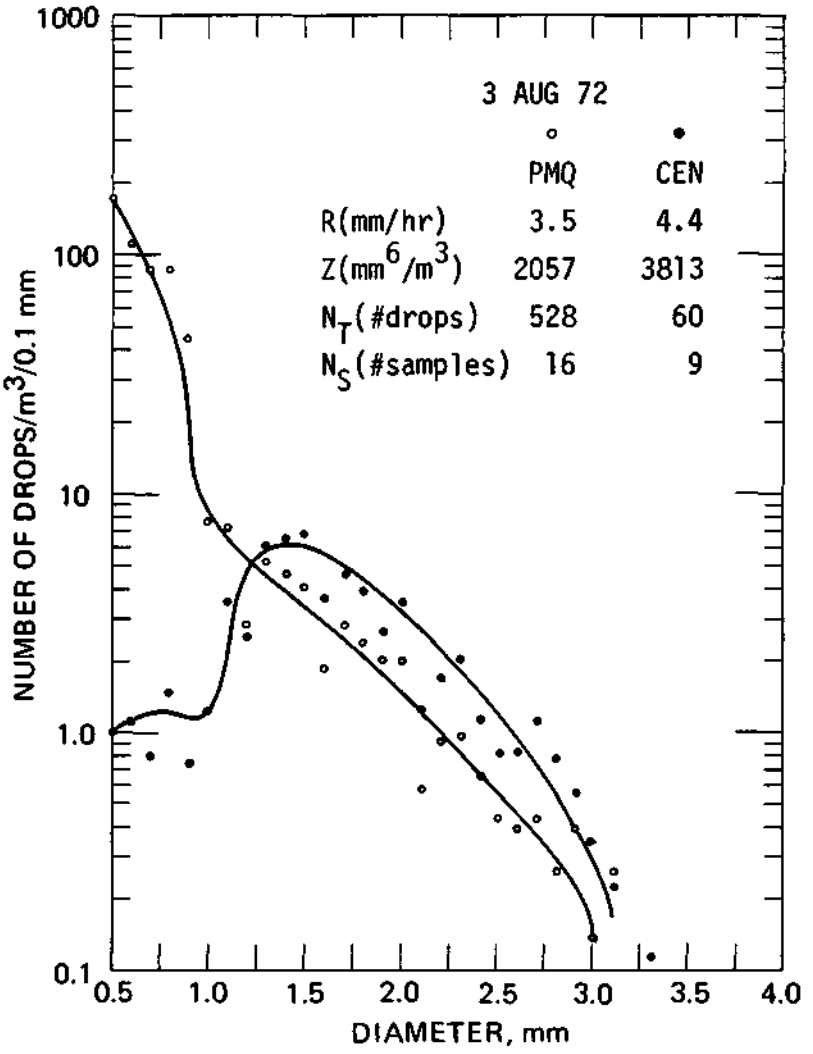

Figure 40. Same as Fig. 39, but a different rain fall rate

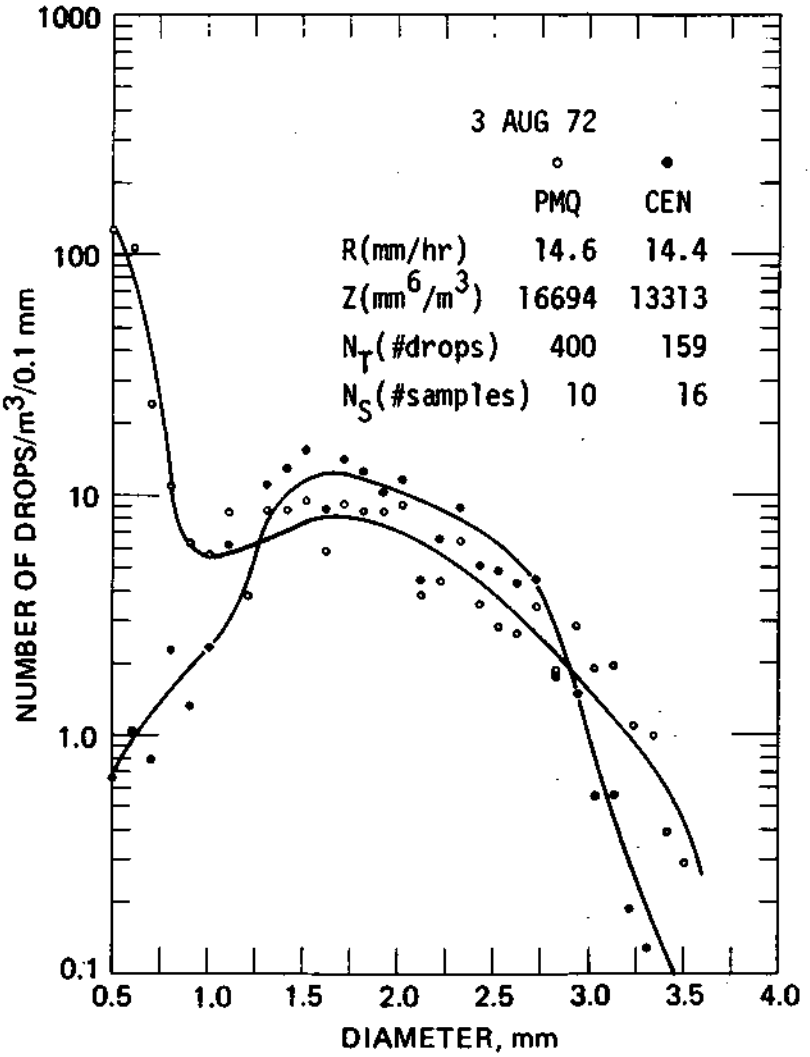

Figure 41. Same as Fig. 39, but a different rainfall rate

storm was 81 with lesser numbers available on the other days depending upon the degree of organic contamination in the sample. Subsequent to the pH measurement the water sample was prepared for the chemical determination of many elements removed by the precipitation process.

5.4.5.2 Sample Acquisition and Handling

The samples were collected in plastic bottles within a network downwind of the city of St. Louis covering an area of 750 mi^2. Each of the sampling locations represented an area of about 9 mi^2 . Within a short period of time after the cessation of rain (a few hours), the samples were removed from the field, immediately capped, and transported within 24 hours to Champaign, Illinois for analysis.

Upon receipt of the samples in the laboratory, the container was weighed with its precipitation sample and a pH measurement was obtained. If the individual sample appeared to be contaminated with various organic materials, as indicated by a visual inspection, the determination of pH was not made. The pH of contaminated samples was uniformly greater than 7. While the samples deemed unfit for this analysis, due primarily to bird droppings were discarded, the rain falling through trees and on man-made edifices would be so contaminated. Consequently, such natural phenomena may counteract the acid content of the precipitation and tend to neutralize it towards more alkaline values.

5.4.5.3 Results

The average pH at each of the sampling sites was calculated and is shown in Fig. 42. The values ranged from 4.26 to 6.82 resulting in a network average for all data points of 4.94. The average pH values shown, in Fig. 42 are weighted by the total water in each of the individual samples.

It is difficult to identify any particular pattern associated with the known industrial areas within the network. There is a relative low pH value near Granite City and in the southern East St. Louis area but comparable values of pH are observed in the extreme southeast portion of the network in a rural environment.

Due to the natural abundance of CO_2 in the atmosphere, precipitation will become more acid due to the solubility of this gas. As an approximate

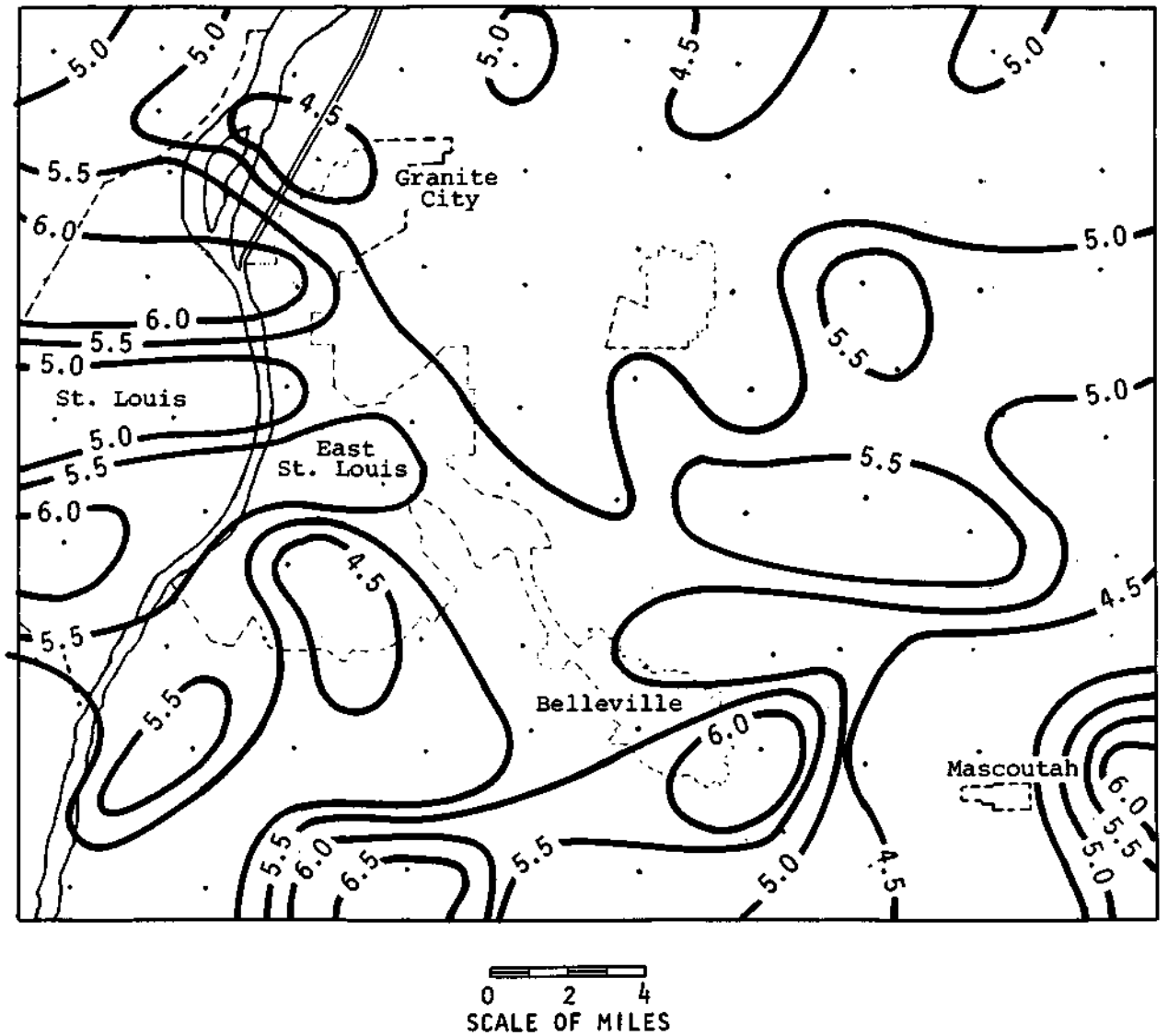

Figure 42. Mean pH from 14 rain events during 1972. Dots are sampling locations for chemistry sub-network of the METROMEX research area.

measure, the pH of water falling through the atmosphere would be about 5.70 due to the formation of natural carbonic acid. Therefore, many of the values shown in Fig. 42 are more alkaline than the natural rainwater without the addition of man-made effluents. In general, the remaining pH values after adjustment for CO_2 solubility would increase, but there are few areas in the network where the pH would be entirely accounted for by the natural acid content of the rainfall.

The average precipitation for the samples included in this analysis is shown in Fig. 43. A comparison between the precipitation pattern (measured in milliliters) and the pH values shown in Fig. 42 show little resemblance and appear to be totally independent.

The deposition of the hydrogen ion can be deduced from the observations of the pH and the precipitation and the mean values for the summer of 1972 are shown in Fig. 44. The resulting pattern appears to be dominated by the distribution of the pH values rather than by the rainfall. This, of course, may be expected since the pH, by definition, is proportional to the logarithm of the hydrogen ion concentration, whereas the precipitation enters the calculation in a linear manner.

5.4.5.4 Summary

The pH was measured in several storms during 1972 and average values of pH, precipitation, and hydrogen ion deposition were determined. These initial data indicate the mean pH over an area of 750 mi^2 is 4.94 and varied from 4.26 to 6.82. These values are not adjusted for the CO^2 saturation which would naturally acidify rainwater to a pH of 5.70, There does not appear to be a distinct correlation between the precipitation and the pH. Likewise, there appears to be no relationship between the areal pH pattern and the industrial areas of the St. Louis metropolitan region.

These observations will be continued during the remaining years of the METROMEX program to ascertain the affects of rainfall acidification on the quality of the soils in the rich agricultural areas in Illinois, and its possible destructive action on buildings and other man-made edifices within the region.

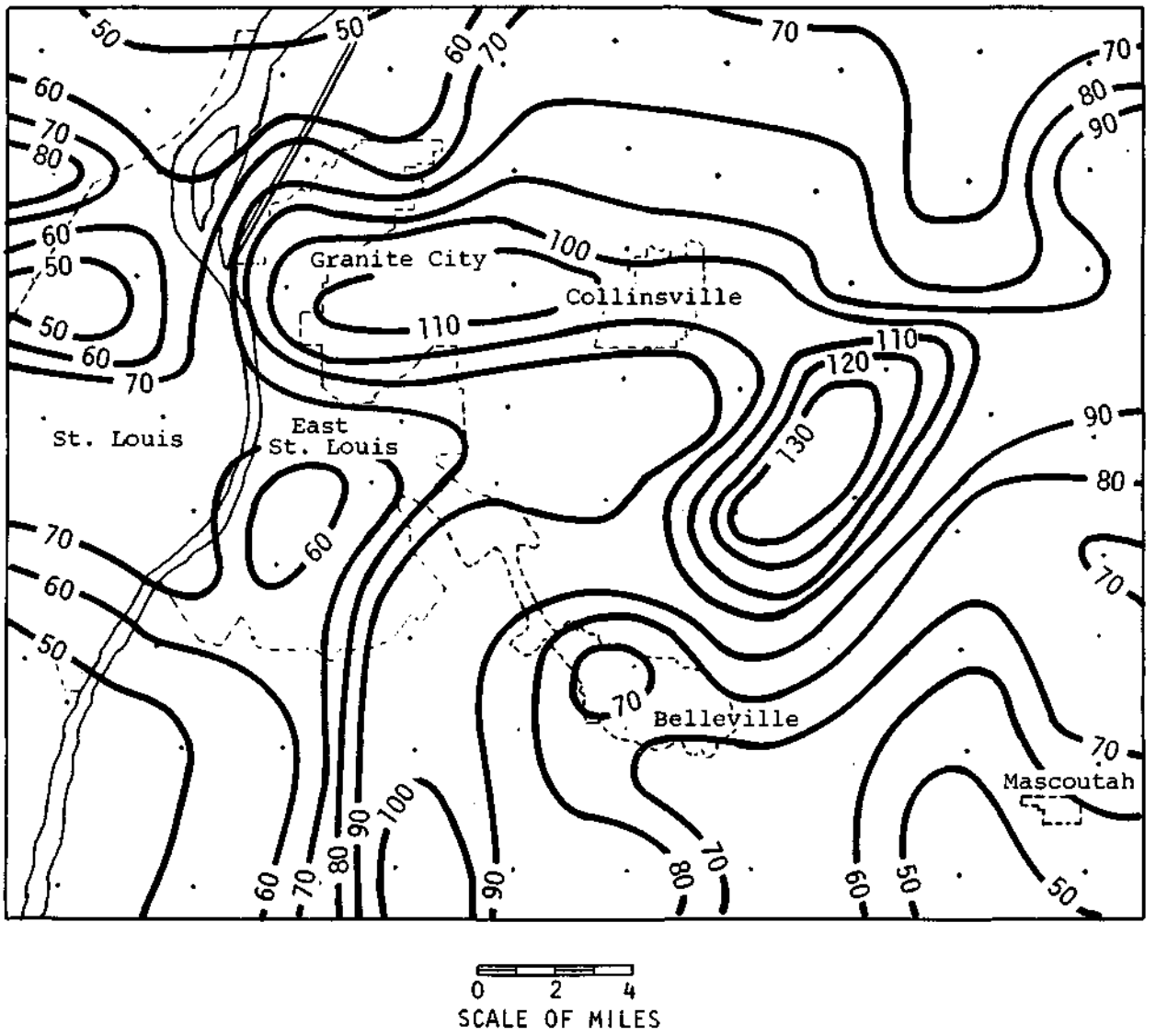

Figure 43. The average precipitation (milliliters) of the 14 rain events of Fig. 42

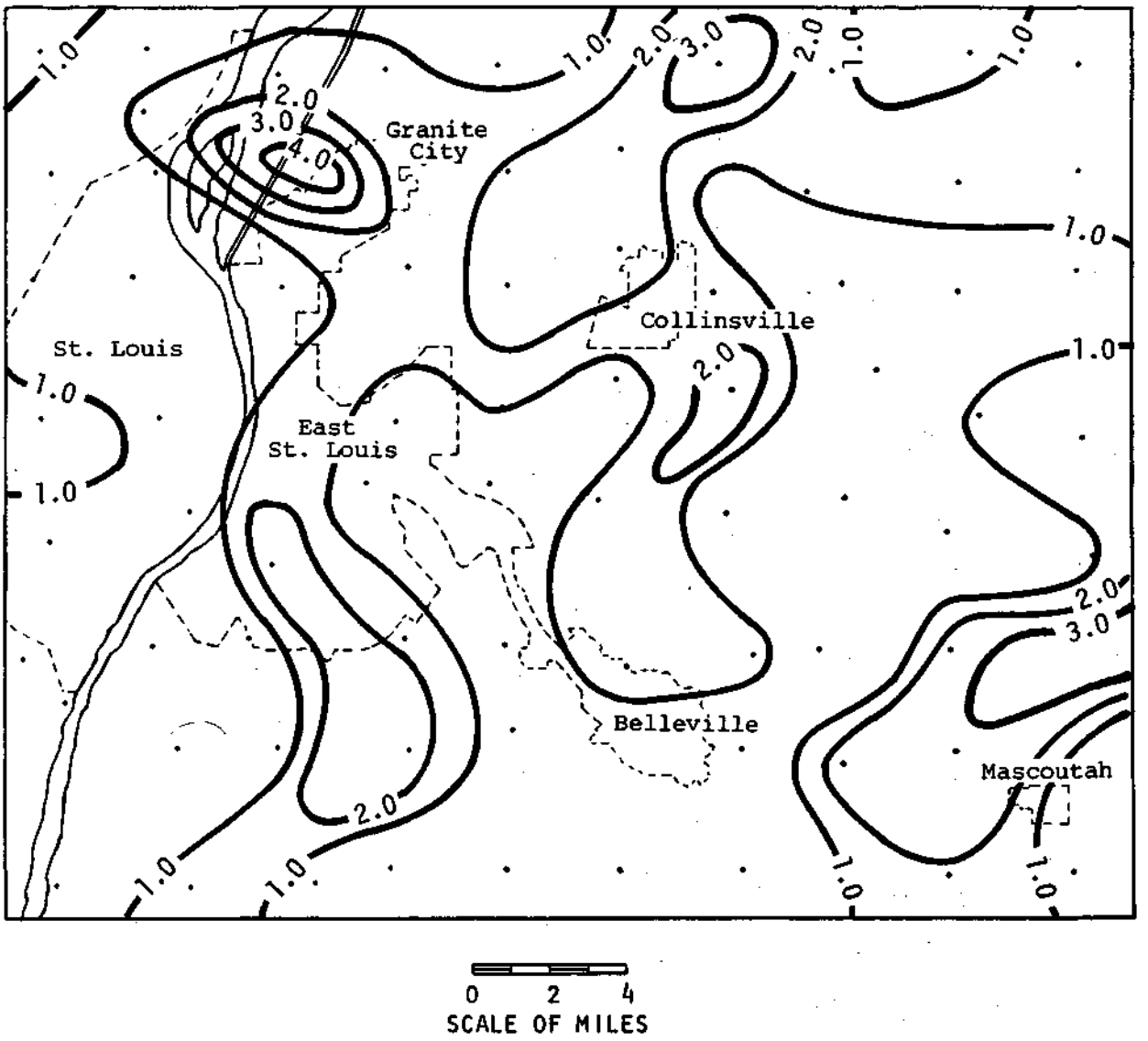

Figure 44. Average hydrogen ion deposition (milligrams) calculated from the observed pH and rainfall for 14 rain events in 1972

5.5 Scavenging Studies - 1971

5.5.1 Scavenging Ratio Measurements

The scavenging ratio*, W, is defined by

$$W = \frac{k\rho}{\chi} \qquad (1)$$

where k is the concentration of a material in precipitation (μg/g), x is its concentration in air (μg/m^3), and ρ is the density of air, taken as a constant, 1200 g/m^3 (20C, 760 mm Hg).

Deposition in precipitation is one segment in the system of pathways by which airborne materials travel through the environment. Accurate assessments of how much material travels this way, and how fast it travels, are desirable, if not essential, for the accurate evaluation of pollution hazards or control strategies.

Scavenging ratios have been found to be relatively constant (varying by a factor of the order 10) for a wide variety of airborne materials (Engelmann, 1971). This makes them useful for prediction of deposition in precipitation when the scavenging characteristics of the material of interest have not been studied in detail. Unfortunately, this is all too often the case..

Even when the physical and chemical properties (e.g., particle size distribution and solubility) of an aerosol are known, there is no general theory of scavenging yet available from which deposition at a point may be predicted.

The scavenging ratio method for predicting precipitation deposition is based on measuring or assuming values for W and χ, from which k may be calculated (Eq. 1).

The objective of this work is to improve the scavenging ratio method for predicting deposition in precipitation. The first part of the work consists of the measurement of washout ratios for six elements, based on daily precipitation and air filter samples collected as part of a comprehensive meteorological field observation program (Project METROMEX). The detailed

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* This ratio has also been called the "washout ratio", but that term is a misnomer, because the ratio is a relative measure of the efficiency of all scavenging processes — not only washout, which is usually defined to include one, below-cloud scavenging mechanism.

data on precipitation, cloud micro-physics, and winds available from this program permit us to look for relationships between daily scavenging ratios and meteorological parameters. The second part of the work consists in attempting to generalize the results to other materials, through measurements of the rain-solubility and particle size distribution of the elements for which scavenging ratios are measured.

Sample collection and analysis procedures for precipitation and air filter samples were discussed earlier in this report. We go on now to the results obtained from the samples collected in METROMEX 1971, as part of the Argonne National Laboratory field effort.

5.5.1.1 Concentrations in Rain

A map showing the locations of the sites is given in Fig. 45. Results are summarized in Table 2 for the concentrations of Cu, Fe, Pb, Mg, Mn, and Zn in rain (corrected for dry deposition). Both the rainfall-weighted mean, k , and the arithmetic mean, $\bar{k}_a$, as well as the standard deviation of the arithmetic mean $\sigma_{\bar{a}}$, are presented for each sampling site, along with the number of samples, n, at each site. The rather small numbers of samples at most stations reflects the fact that St. Louis had an abnormally dry summer in 1971. The station at Pere Marquette collected the highest number of samples, 13. This resulted both from a longer period of operation and from being the only station in operation during July, when rainfall was more nearly normal.

Table 2 shows that mean concentrations of each of the elements are consistently higher (by up to about 10 times) at the close-in downwind stations (Centreville, KMOX, and Coldwater Creek) than at the more remote upwind stations (Tyson and Pere Marquette). This is true for both the rainfall-weighted and the arithmetic means. Concentrations of rainfall constituents usually decrease with increasing rainfall amounts, so the rainfall-weighted concentration would be expected to be smaller than the arithmetic mean. The results in Table 2 are inconsistent on this point, although Pb and Mg are consistently just the opposite of the normally expected pattern. However, these results may be due to the small number of samples at most stations.

TABLE 2

Rainfall-Weighted Mean and Arithmetic Mean Concentrations in Rain METROMEX 1971

Station		Concentration, ppb Cu	Fe	Pb	Mg	Mn	Zn
Centreville	$\bar{k}_r$ [a]	7.2	640	55	303	10	73
n = 5 [a]	$\bar{k}_a$	14.0	760	23	204	15	184
	$\sigma_{\bar{a}}$	7.7	360	14	142	9	81
KMOX	$\bar{k}_r$	2.1	350	38	260	8	21
n = 4	$\bar{k}_a$	0.6	610	12	212	4	47
	$\sigma_{\bar{a}}$	0.6	310	12	84	2	28
Coldwater Creek	$\bar{k}_r$	10.0	540	108	330	17	126
n = 4	$\bar{k}_a$	6.1	330	58	256	10	67
	$\sigma_{\bar{a}}$	5.0	328	51	120	8	58
Tyson	$\bar{k}_r$	0.9	(b)	23	46	6	(b)
n = 3	$\bar{k}_a$	0.3		5	23	5	
	$\sigma_{\bar{a}}$	0.3		5	13	3	
Pere Marquette	$\bar{k}_r$	1.5	210	10	103	7	9
n = 13	$\bar{k}_a$	1.7	180	9	125	7	24
	$\sigma_{\bar{a}}$	0.2	87	2	37	3	13

$\bar{k}_r$ is the rainfall-weighted mean, $\bar{k}_a$ is the arithmetic mean, $\sigma_{\bar{a}}$ is the standard deviation of the arithmetic mean, and n is the number of samples at each station.

Contamination or error suspected.

In assessing the influence of the urban area on the concentrations in rain, one should ideally divide his samples at each station into (1) those collected from rain systems that ingested air that had recently passed over the urban area and (2) those collected from rain systems that had not ingested such air. Because of the small numbers of samples at each station, however, it is not practical to do this with the 1971 data. A similar, but somewhat less satisfactory procedure is to take the data from all stations and divide them in the same way. The results of such a data stratification, and a test of the significance of the observed differences, using Student's t-test, is given in Table 3. The two areas assumed to be the source(s) of the "urban plume(s)" are outlined in Fig. 45. The direction of movement and the horizontal spreading of the plumes from these sources was estimated subjectively from all available local wind observations in the mixing layer. These consist of about 10 surface stations and the NOAA Environmental Meteorological Support Unit (EMSU) low-level sounding at the Arch.

TABLE 3

Concentrations in Rain-Upwind/Downwind* Comparison, All Stations

	$\bar{k} \pm \sigma_{\bar{k}}$, ppm		
	Upwind n = 18	Downwind n = 10	t-value**
Cu	0.0015 ± 0.0007	0.0088 ± 0.0042	2.13
Fe	0.15 ± 0.06	0.67 ± 0.22	2.70
Pb	0.0070 ± 0.0021	0.034 ± 0.021	1.66
Mg	0.11 ± 0.03	0.24 ± 0.08	1.72
Mn	0.0061 ± 0.0021	0.011 ± 0.005	1.02
Zn	0.026 ± 0.011	0.13 ± 0.05	2.47

* From measured winds. See text for detailed definition of terms.

** t-value for significance at 10% level = 1.71.

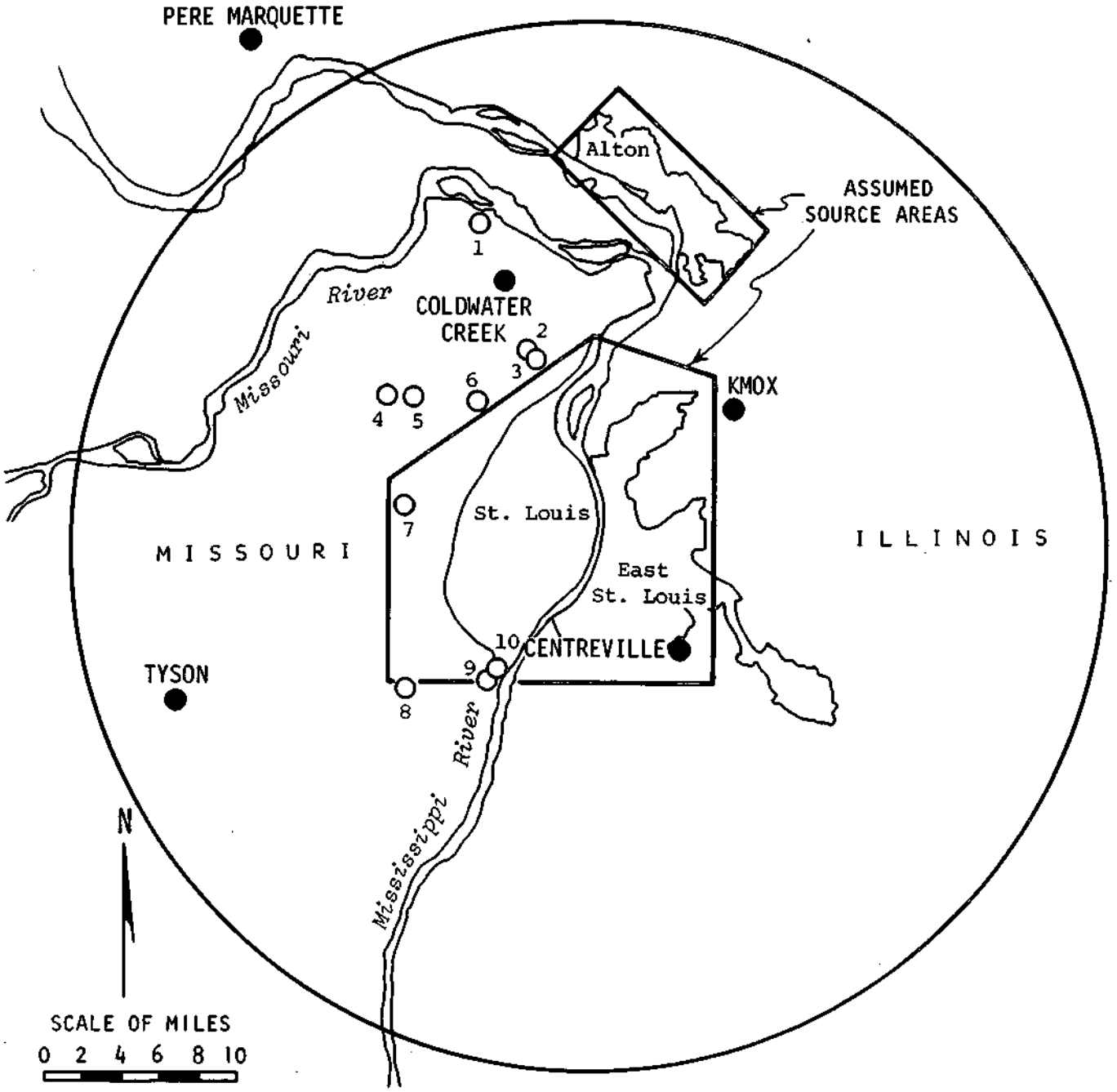


Figure 45. Map of research area showing assumed emission source areas, air and rain sampling stations (solid circles), and St. Louis County Health Department high volume air sampler sites (open circles)

The rain sample was designated to be "downwind" if it was located in the urban plume, just defined, at the time of, or up to one hour prior to, onset of rain at the collecting stations. Otherwise, the sample was designated "upwind".

The concentrations in upwind rain are consistently less than in downwind rain, and the differences are significant at the 10% level for all the elements measured except Pb and Mn. However, the upwind data are biased toward the more remote locations (13 of 18 observations were from Pere Marquette and Tyson), so the differences may reflect the effects of distance from the city as well as urban or non-urban trajectory of the ingested air.

5.5.1.2 Concentrations in Air

The mean concentrations $\overline{x}$, of the same six elements in air sampled on rain days* at the various filter sampling sites are presented in Table 4. All filters collected on rain days were analyzed, even if rain was collected at only one station. The table shows that concentrations in air, like those in rain, increase from the more distant, upwind locations to the closer-in, downwind locations. Again, the downwind concentrations are higher by factors of up to about 10. For each element except Pb, KMOX had the highest concentration, followed by Coldwater Creek, Tyson, and Pere Marquette. For Pb, KMOX and Coldwater Creek were reversed.

For comparison, monthly and mean trace metal concentrations measured in the 10-station St. Louis County Health Department network for summer, 1971, are given in Table 5. Station locations are shown in Fig. 45. Agreement between the two networks is generally excellent. A possible discrepancy occurs in the case of Cu, where the St. Louis Co. concentrations are generally higher. As Hoffman and Duce (1971) have pointed out, however, brush wear on copper armatures can be a significant source of Cu contamination on filter samples collected on the type of high-volume sampler used in the St. Louis Co. network. This problem should be less severe in the samplers used. The

* Exception: Four of the 12 KMOX filters were collected on days when no rain samples were collected at any of the 5 stations.

TABLE 4

Mean Concentrations in Air on Sampled Rain Days, METROMEX 1971

Station		Concentration, $\mu g/m^3$					
		Cu	Fe	Pb	Mg	Mn	Zn
KMOX[a]	$\bar{\chi}$	0.0297	1.13	0.476	0.463	0.043	0.294
	$\sigma_{\bar{\chi}}$	0.0052	0.12	0.039	0.093	0.004	0.063
	n	8	8	8	8	8	8
Coldwater Creek	$\bar{\chi}$	0.0181	1.01	0.574	0.325	0.031	0.162
	$\sigma_{\bar{\chi}}$	0.0059	0.11	0.157	0.036	0.005	0.071
	n	8	8	8	8	8	8
Tyson	$\bar{\chi}$	0.0020	0.52	0.314	0.157	0.017	0.064
	$\sigma_{\bar{\chi}}$	0.0003	0.08	0.058	0.034	0.003	0.027
	n	8	8	8	8	8	8
Pere Marquette	$\bar{\chi}$	(b)	0.28	0.15	0.125	0.013	0.047
	$\sigma_{\bar{\chi}}$		0.06	0.03	0.020	0.001	0.014
	n		14	14	14	14	14

If 4 samples collected on dry days are included, the means at KMOX are: Cu, 0.0246; Fe, 1.13; Pb, 0.489; Mg, 0.397; Mn, 0.044; and Zn, 0.277 $\mu g/m^3$.

Cu undetectable on all filters.

TABLE 5

St. Louis Co. Trace Metal Concentrations in Air, 1971

Concentration in Air, $\mu g/m^3$

Station No.	Month	Cu	Fe	Pb	Mn	Zn
1	June	0.357	1.13	0.40	0.054	0.04
	July	0.235	1.58	0.80	0.031	0.00
	August	0.375	1.16	0.54	0.029	0.47
	Mean	0.322	1.29	0.58	0.038	0.17
2	June					
	July					
	August	0.130	1.87	1.76	0.052	0.67
	Mean					
3	June	0.022	1.11	0.85	0.072	0.09
	July	0.015	1.11	1.63	0.043	0.00
	August	0.070	1.78	0.79	0.042	0.56
	Mean	0.036	1.33	1.09	0.052	0.22
4	June					
	July	0.008	0.95	1.08	0.029	0.19
	August	0.041	1.70	1.48	0.045	0.03
	Mean	0.024	1.32	1.28	0.037	0.11
5	June	0.125	1.42	1.02	0.079	0.12
	July	0.079	0.99	1.11	0.015	0.00
	August	0.148	3.04	1.38	0.117	0.00
	Mean	0.117	1.82	1.17	0.070	0.04
6	June	0.025	1.26	1.40	0.067	0.12
	July	0.017	1.14	2.20	0.047	0.28
	August	0.029	1.96	1.38	0.070	0.41
	Mean	0.024	1.45	1.66	0.061	0.27
7	June	0.084	1.48	1.93	0.082	0.20
	July	0.034	1.25	3.14	0.013	0.00
	August	0.032	1.44	1.68	0.048	0.00
	Mean	0.050	1.39	2.25	0.048	0.07
8	June					
	July	0.033	1.07	2.88	0.029	0.20
	August	0.075	0.96	0.84	0.043	0.17
	Mean	0.054	1.02	1.86	0.036	0.18
9	June	0.101	1.33	0.64	0.056	0.11
	July	0.036	1.94	1.36	0.045	0.16
	August	0.062	2.47	0.66	0.039	0.00
	Mean	0.066	1.91	0.89	0.047	0.09
10	June	0.002	4.00	1.06	0.096	0.24
	July	0.034	2.38	1.28	0.042	0.09
	August	0.059	4.23	0.60	0.068	0.07
	Mean	0.032	3.54	0.98	0.069	0.13

motor that drives the vacuum pump was enclosed in a housing open only near the ground, while the filter paper was exposed at height of about 2 m.

Again, it is of interest to examine upwind-downwind differences in concentration, as an indication of the effect of the city on air quality. Table 6 shows mean concentrations of the various elements (and standard deviations of the means) for all stations, divided into two categories that indicate whether the sampling site was upwind or downwind* of pollution source regions (see Fig. 45) during the sampling period. Downwind concentrations are greater for each element, with significance at the 10% level or better for all elements except Pb.

TABLE 6

Concentrations in Air-Upwind/Downwind* Comparison for All Stations

	$\bar{\chi} \pm \sigma_{\bar{\chi}}$, $\mu g/m^3$		
	Upwind n = 24	Downwind n = 17	t-value**
Cu	0.0045 ± 0.0013	0.021 ± 0.004	3.99
Fe	0.60 ± 0.07	0.89 ± 0.12	2.85
Pb	0.30 ± 0.06	0.44 ± 0.06	1.63
Mg	0.20 ± 0.02	0.34 ± 0.06	2.51
Mn	0.021 ± 0.003	0.034 ± 0.004	2.67
Zn	0.091 ± 0.022	0.21 ± 0.05	2.51

* From measured winds. See text for detailed definition of terms.

*** t-value for significance at 10% level =1.68.

Here again, the upwind observations are somewhat biased toward the remote stations—16 of 24 samples are from Tyson and Pere Marquette. In

* A sample was designated "upwind" if its collection station was in the urban plume (defined earlier) for 2-hr or less during its 24-hr collection period. All other samples were designated "downwind".

this case it is possible to compare concentrations at a single station when it is upwind versus when it is downwind of the source regions. This is done in Table 7 for the KMOX station. The total number of samples—12—is still not as large as would be desirable, but the results, while still preliminary, reveal some interesting variations from the pooled samples.

At KMOX, only Cu and Mg have significantly (10% level) higher concentrations in air from source regions. Many speculations about the cause of this occurrence are possible, but for now it is probably sufficient to say that the answer probably lies in the rather poorly known distribution of sources of the various elements in the St. Louis area.

TABLE 7

Concentrations in Air-Upwind/Downwind* Comparison for KMOX Only

	$\bar{\chi} \pm \sigma_{\bar{\chi}}$, $\mu g/m^3$		
	Upwind n = 5	Downwind n = 7	t-value**
Cu	0.015 ± 0.003	0.032 ± 0.006	2.29
Fe	1.1 ± 0.1	1.2 ± 0.1	0.62
Pb	0.52 ± 0.06	0.47 ± 0.04	0.61
Mg	0.26 ± 0.02	0.50 ± 0.10	1.84
Mn	0.042 ± 0.004	0.045 ± 0.005	0.42
Zn	0.27 ± 0.03	0.28 ± 0.07	0.18

* From measured winds. See text for detailed definition of terms.

** t-value for significance at 10% level = 1.81.

5.5.1.3 Scavenging Ratios

It is worthwhile to repeat that the emphasis is on measuring daily scavenging ratios, based on daily measurements of concentrations in rain and air—not on the mean concentrations just reported (Tables 2 and 5). Furthermore, it should be stressed that the arithmetic mean of daily scavenging ratios over the period of a month, say, is not equivalent to the

ratio that would be computed from analyses of monthly air filter and precipitation samples collected at the same place. The mean of daily ratios gives equal weight to each rain regardless of amount, whereas a ratio computed from monthly samples weights the rain samples according to amount.

The scavenging ratio data, or the concentrations in rain and air found in the literature are frequently this type of "rainfall-weighted" composite. It is of interest to compare scavenging ratios computed both ways from the same data.

Both types of scavenging ratios for Cu, Fe, Pb, Mg, Mn, and Zn from METROMEX 1971 are presented in Table 8. The composite means range from about 1/4 to 3/4 of the respective arithmetic mean values for the various elements. This shows how the composite means are influenced by the heavier rains, which frequently have low k-values and hence, low W's.

TABLE 8

Arithmetic Mean and Rainfall-Weighted Mean Scavenging Ratios, METROMEX 1971

		Cu	Fe	Pb	Mg	Mn	Zn
Arithmetic Mean	$\overline{W}_a$	724	979	147	1282	721	696
	$\sigma_{\overline{W}}$	251	233	32	267	158	168
Rainfall-Weighted Mean	$\overline{W}_r$	202	520	116	660	405	294
Number of Samples		8	19	17	21	18	20

A comparison of scavenging ratios at a given station in the two cases when the station is (1) upwind and (2) downwind of pollution sources is again the most desirable. Again, however, the data are presently too few to yield anything meaningful at any one station, and the data are grouped from all 5 stations into upwind and downwind* cases. The comparison is

* Daily scavenging ratios were included only if both component measurements fell into the same class: i.e., upwind or downwind.

shown in Table 9. It shows very small differences for all the elements, with no differences significant at the 10% level.

TABLE 9

Scavenging Ratios-Upwind/Downwind* Comparison for All Stations

	$\overline{W} \pm \sigma_{\overline{W}}$ (n)		
	Upwind	Downwind	t-value**
Cu	540 (1)	790 ± 410 (5)	
Fe	870 ± 450 (8)	930 ± 150 (8)	0.13
Pb	190 ± 70 (7)	140 ± 70 (7)	0.59
Mg	1200 ± 380 (10)	1400 ± 490 (8)	0.26
Mn	840 ± 280 (9)	520 ± 210 (6)	0.78
Zn	970 ± 330 (7)	710 ± 250 (9)	0.61

* From measured winds. See text for detailed definition of terms.

** t-value for significance at 10% level $\simeq$ 1.76

The two previous tables both show some rather marked differences between scavenging ratios for the various elements. To examine these differences more carefully, tests for significance of the differences between means of each element pair were performed using Student's t. Because the test for upwind-downwind differences showed no significance, all samples were grouped together for the present test. The computed t-values are shown in Table 10. The results show that the scavenging ratio for Pb is significantly different from those of all the other elements determined, at the 1% level or better. In addition, the scavenging ratios for Mg and Mn were found significantly different at the 10% level.

Inspection of the scavenging ratio data gave evidence that some pairs of elements were positively correlated and others perhaps negatively correlated. To examine this possibility in a quantitative way, correlation coefficients were computed for all pairs of elements. The results appear in Table 11. The pairs Cu-Mg, Cu-Mn, Fe-Mg, and Fe-Mn, had correlation coefficients greater than 0.80, and all were significant at the 5% level or better.

TABLE 10

Results of t-Tests for Significance of Differences Between Mean Scavenging Ratios

t-values

Cu W = 724	0.62	3.10	1.23	0.01	0.09
Fe 979		3.25	0.82	0.88	0.96
	Pb 147		3.71	3.36	2.89
		Mg 1282		1.69	1.62
			Mn 721		0.10
				Zn 696	

t-value for significance at 10% level ≅ 1.7, at 1% level ≅ 2.8

TABLE 11

Correlation Coefficient Matrix for Scavenging Ratios from All Stations, METROMEX 1971

Cu	0.45	-0.30	0.83	0.94	0.23
	Fe	-0.31	0.82	0.84	-0.20
		Pb	-0.13	-0.24	0.34
			Mg	0.50	-0.04
				Mn	-0.01
					Zn

In examining the possible causes of these high correlation coefficients, it is useful to review what scavenging ratios and their correlations mean in physical terms. The ratios are computed from concentrations in air and precipitation measured near the ground. Actually, of course, the cloud "sees" a different concentration entering at its base, and the physical processes that remove particles from the air in the cloud act on that concentration of particles. The physical processes that remove particles from the air below the cloud act on the entire vertical concentration profile up to cloud base.

To reflect the actual physical processes taking place, scavenging ratios should take into account the vertical concentration profile from ground level to cloud base. However, obtaining vertical profiles is a difficult sampling problem, even in a research situation, and completely impractical for operational use. Thus, if scavenging ratios are to be useful for prediction purposes, the necessary measurements must be conveniently made, i.e., at or near ground level. Ground level concentrations will be useful in predicting wet deposition if they are a reliable index of concentrations in the layer below cloud base.

However, we must look more closely at the situation in examining relations between elements. If the scavenging ratio of element A is larger than that of element B, it can mean (1) that the scavenging efficiency for A is greater than that for B, or (2) that the estimate of X_A at cloud base, using ground level measurements, was disproportionately low, relative to the estimate of X_B. As an example of the latter situation, consider two elements whose washout ratios are equal, based on measurements at cloud base. Now, however, suppose that the concentration of element C is constant with height from the ground to cloud base, while the concentration of element D decreases by a factor of 10 between the ground and cloud base. Then the estimate of the mean X_D below cloud base, from a ground-level measurement, will be disproportionately high, relative to X_c . Because x is in the denominator of the washout ratio, the resulting W_D will be larger than W_C. Thus, if a given element's washout ratio is high relative to some other element, it can mean either that the first element is more efficiently removed from the air, either in cloud or below cloud, or that its estimated x is disproportionately

low, relative to the second element. Conversely, a (relatively) low washout ratio means less efficient removal, or a disproportionately high estimate of x.

In general, a positive correlation between two variables indicates that large values of the two parameters tend to occur together and small values tend to occur together. In terms of the possible interpretations of correlations between scavenging ratios, positive correlations can indicate either (1) parallel variations of scavenging efficiency from rain to rain, or (2) parallel deviations between actual and estimated concentrations in air.

One possible explanation for a high correlation between scavenging ratios is that the two elements have similar particle size distributions. The efficiencies of both nucleation collection and impaction are very dependent on the size of the collected particles, so that elements with very similar size distributions could have removal efficiencies that would vary in a parallel manner from rain to rain in response to varying updraft speeds, moisture, nuclei concentrations, and raindrop size distributions of the precipitation systems.

Another possible explanation of highly correlated scavenging ratios is a common source. For example, industrial plants or power plants with high stacks could cause parallel deviations between ground-level concentrations and those for the sub-cloud layer for certain elements.

Both of these possible explanations find support in the literature. In heavily industrialized northwest Indiana, Nifong (1970) found Fe, Mg, and Mn to occur predominantly on large (5-10 μm diameter) particles, and had common sources in the steel industry. Common sources were also found by Dams et al., (1971) for Fe-Mg-Mn, in northwest Indiana but not for Cu-Mg-Mn. An Fe-Mg-Mn association may also be expected in the St. Louis area from the fact that there are steel making operations in the area. However, the measurements to confirm this expected association, and to determine whether one also exists for Cu-Mg-Mn are tasks for the future.

For the 1971 data, a number of possible relationships between scavenging ratios and weather parameters were investigated. The parameters included in this investigation were (1) rainfall, (2) atmospheric stability., and (3) synoptic rainfall type.

There is a well known inverse relationship between contaminant concentrations and rainfall amount. Thus, it might reasonably be expected that scavenging ratios would also vary inversely with rainfall amount. Correlation coefficients for scavenging ratios and corresponding rainfall amounts were computed and are given in Table 12. The correlation coefficients vary from -0.69 to 0.65, but because of the small number of data points and large variability, none is significant at the 10% level.

TABLE 12

Correlation Coefficients Between Scavenging Ratios and Rainfall

	r (n)		
	Upwind*	Downwind*	Total
Cu	--	-0.69 (5)	-0.61 (6)
Fe	-0.44 (8)	-0.16 (8)	-0.34 (16)
Pb	0.27 (7)	0.52 (7)	0.37 (14)
Mg	-0.13 (10)	-0.38 (8)	-0.24 (18)
Mn	-0.41 (9)	-0.46 (6)	-0.38 (15)
Zn	0.65 (7)	-0.03 (9)	0.35 (16)

* From measured winds. See text for detailed definition of terms.

Relationship to stability was investigated by computing correlation coefficients between scavenging ratios and mixing depth as determined from the NOAA low level sounding at the Arch, and by grouping the ratios according to whether the rain fell during daylight (unstable) or darkness (stable). The results (not shown) indicated no relationships between scavenging ratios and either stability parameter, but the data set was very limited. This relationship should be investigated further when more data are available.

Relationships between scavenging ratios and particle parameters offer a possible method of extending the present results to other aerosol materials. Thus, relationships between (1) scavenging ratios and

solubility, and (2) scavenging ratios and particle size distribution were examined.

Solubility was suspected as a possible indicator of susceptibility to scavenging by nucleation collection. Thus, correlation coefficients were calculated for scavenging ratios and the soluble fraction of each element in individual rains. The results are given in Table 13. The computed coefficients range from -0.58 to +0.23, but only Mn (r = 0.42) was found to be significant at the 10% level.

TABLE 13

Correlation Coefficients Between Scavenging Ratios and Percent Soluble Fraction for All Stations

ELEMENT[(a)]	n	CORRELATION COEFFICIENT, r
Cu	8	-0.58
Pb	17	+0.23
Mg	21	-0.13
Mn	18	-0.42
Zn	20	+0.22

Fe is not included because it is virtually 100% insoluble in rain.

Analysis of Andersen impactor samples collected in 1971 are not yet completed, so primary information is unavailable on particle size distributions for the various elements in the St. Louis area. However, the literature contains information on particle size distributions measured in other areas from which some tentative conclusions may be drawn about the relationship between washout ratio and particle size distributions. The large-particle preference of Fe, Mg and Mn in northwest Indiana found by Nifong (1970) has already been mentioned. The same work showed Zn to occur primarily on the small-particle stages (~1 um diameter) of the Andersen impactor, while Cu showed both large-particle and small-particle components, with emphasis

usually on the latter. Others have published data on mass median diameter" (MMD) for other locations. A partial summary of these data is given in Table 14. The data were also used to construct the graph shown in Fig. 46 relating the rainfall-weighted mean scavenging ratios to particle size ranges reported in the literature. (Literature values for St. Louis are shown as solid triangles on the range bars for each element.) Although Table 10 shows only Pb to have a scavenging ratio significantly different from all the other elements, the graph shows steadily increasing ratios with increasing MMD for the various elements measured.

TABLE 14

Literature Values of Mass Median Diameters for Selected Elements and Locations

	MMD (μm)						
Location	Cu	Fe	Pb	Mg	Mn	Zn	Reference
Cincinnati	1.2	3.7	0.2[(a)]	4.5			Lee et al., (1968)
Cincinnati	1.3	2.5	0.5		2.1	1.1	Thompson (1972)
Fairfax, Ohio		1.4	0.4[(a)]	7.2			Lee et al., (1968)
Philadelphia	1.2	2.4	0.6		2.3	1.3	Thompson (1972)
Washington, D.C.		2.3	0.4			1.2	Thompson (1972)
Chicago	1.5	3.6	0.7		1.9	1.0	Thompson (1972)
Denver	1.6	2.5	0.5		1.8	1.7	Thompson (1972)
St. Louis	1.1	3.2	0.7		2.2	1.2	Thompson (1972)

Estimated

* That size for which half the total mass of a given element occurs on larger particles and half on smaller particles.

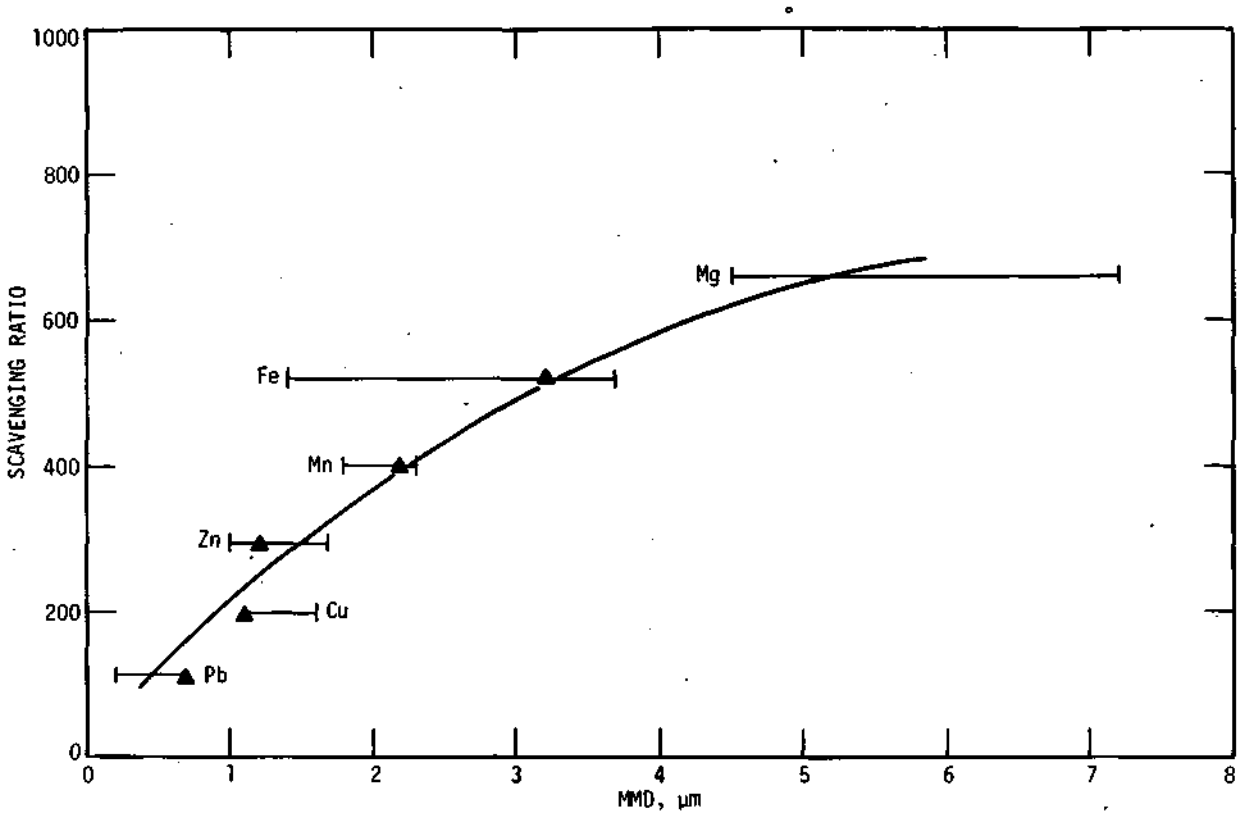


Figure 46. The precipitation scavenging ratio as a function of the mass median diameter (MMD). The triangles represent values for St. Louis from the literature and the range for other urban locations are indicated by the horizontal bars

It is reasonable that scavenging ratios should increase with particle size over the approximate diameter range from 0.5 to 5 μm. Both nucleation and impaction are effective collection mechanisms in this size range, and the efficiencies of both increase with particle size in this range (Fletcher, 1962).

5.5.1.4 Summary

In summary, scavenging ratios for Cu, Pb, Fe, Mg, Mn, and Zn were measured during METROMEX 1971. The observed values ranged from about 150 for Pb to about 1280 for Mg. No significant upwind/downwind differences were found for scavenging ratios, although concentrations in both precipitation and air showed variations ranging up to about 10-fold. The ratio for Pb was shown significantly smaller than each of the other elements. In addition, plots of scavenging ratio versus literature values of the MMD for the various elements show a continuous increase in scavenging ratio with particle size. Little or no relationship was found between scavenging ratios and any of the several meteorological or particle parameters examined, including atmospheric stability, rainfall amount, synoptic rain type, and solubility of aerosol materials in rain.

5.5.2 Aircraft Filter Collections

5.5.2.1 Introduction

Earlier in this report it was pointed out that the vertical distribution of a given material in the atmosphere can affect its scavenging ratio. The prediction of concentrations in precipitation based on measured scavenging ratios and ground-level concentrations in air may be perfectly valid, but a knowledge of the vertical distribution is needed to provide clues to the dominant physical mechanisms of particle collection and their relative and absolute efficiencies.

To provide a few samples collected aloft for comparison with surface concentrations, a program of aircraft filter collections over St. Louis was carried out during METROMEX 1971. This was accomplished through the cooperation of scientists at Los Alamos Scientific Laboratory (LASL) using RB-57C sampling aircraft and flight crews of the 58th Weather Reconaissance

Squadron, U. S. Air Force. This team had extensive experience in the collection of filter samples by aircraft, including recent low-altitude flights in urban areas and in the vicinity of other pollution sources.

While the primary purpose of the aircraft sampling program was collection of filters for chemical analysis, a second analysis priority was particle-size determination using the LASL scanning electron microscope (SEM). These determinations were planned for the filters collected aloft, and in addition, filters of the same type exposed at ground level during the approximate period of sample collection aloft. With such a set of samples, it was possible to examine upwind/downwind as well as vertical differences in the particle size distribution and vertical differences in trace element concentrations.

This is a summary of the samples collected during July and August, 1971 and a report of results obtained from the collected samples up to 1 February 1973.

5.5.2.2 Sampling Procedures

The sampling aircraft operated from its home base at Kirtland AFB, Albuquerque", New Mexico. On sampling days the aircraft flew to St. Louis , collected a single sample, landed at Scott AFB, Belleville, Illinois for refueling, and returned home.

Eight sampling flights were scheduled, one each week between 8 July and 26 August 1971. Various operational difficulties altered the original schedule somewhat, but a total of eight sampling flights were accomplished, including one thunderstorm-anvil sampling mission carried out in the Albuquerque, New Mexico, area for the purpose of determining the particle content of air that had been circulated through a thunderstorm.

While sampling in the St. Louis area, the aircraft flew along the flight path shown in Fig. 47, at an altitude of about 610 m (2000 ft.) above ground level, at speeds of about 103 m/sec (200 kt.), for periods of 1/2 hour. Sampling was carried out using the LASL B-57 Particulate Debris Sampler (for further description see Kelsey, 1971), carried by RB-57C aircraft. Figure 48 shows a sampling aircraft in flight. The Kronisol-impregnated IPC-1478 filters used to collect the samples were

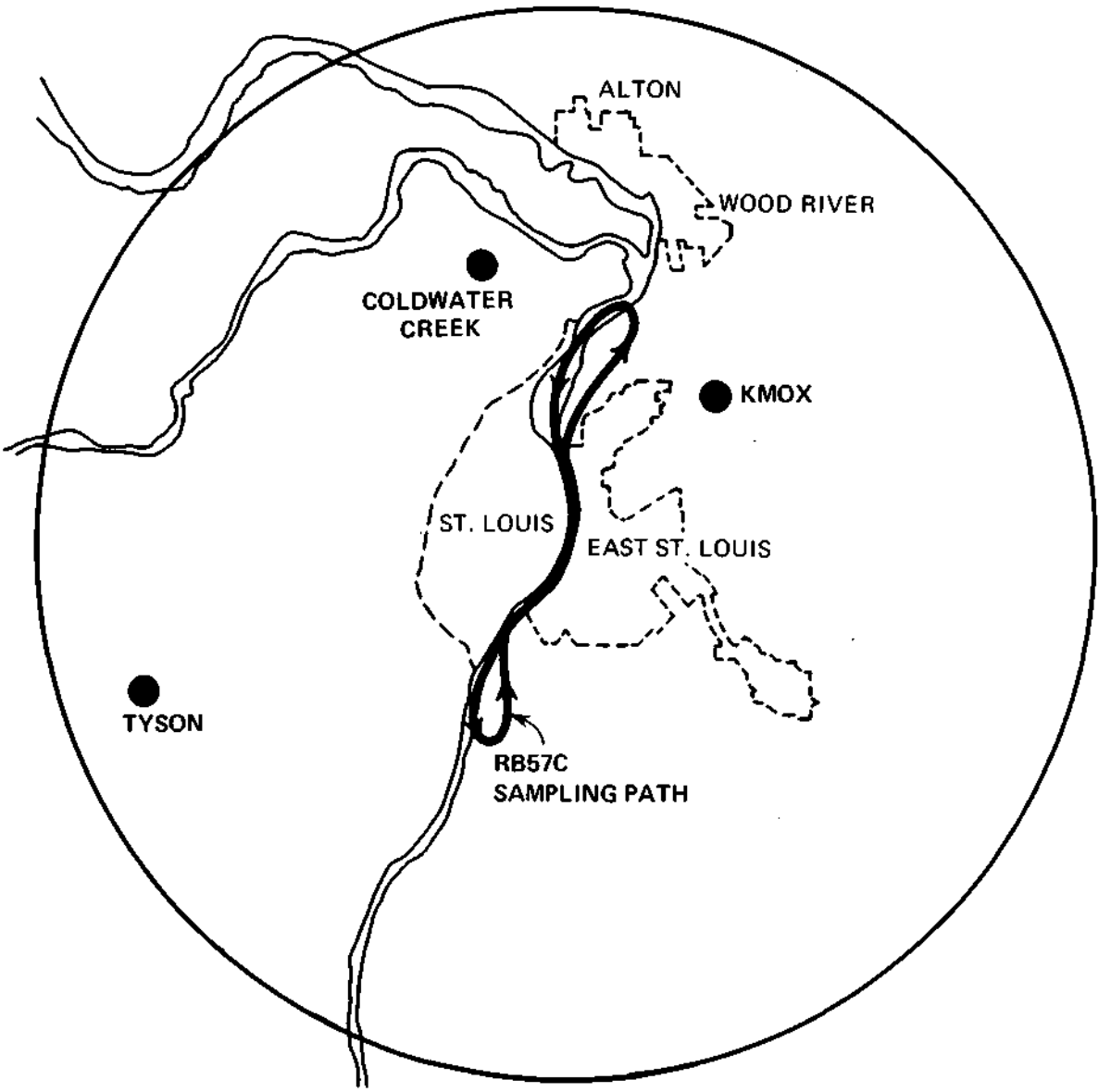


Figure 47. Map of research area, showing aircraft sampling track, surface stations, and primary urban-industrialized regions

Figure 48. A U. S. Air Force RB-57C aircraft in flight, showing wingtip filter samplers (U. S. Air Force photo)

loaded, unloaded, and handled by LASL or USAF personnel following their usual precautions to avoid contamination. Portions of each filter were removed for chemical and particle size analysis using a standard procedure designed by LASL to insure that the portions removed are as representative as possible of that collected by the aircraft (Guthals and Smith, 1972). Sample volumes for the aircraft samples were estimated from the area of the sampler inlet, the aircraft speed, and the duration of the sample, corrected for a sampling efficiency of 0.554 (Kelsey, 1971).

On selected aircraft sampling days, ground level samples were collected on IPC-1478 filters at three sites (Tyson, KMOX, and Coldwater Creek) shown in Fig. 45, using high-volume (approximately 50 m^3/hr) positive-displacement pumps. Volumes for these samples were estimated from the pressure drop, read at the beginning and end of each sample and calibrated against a precision rotameter-type flow meter. Additional samples were collected for chemical analysis on 25 mm diameter Whatman-41 filter paper. Collection times varied from 1.5 to 10 hr, bracketing the period of aircraft sampling.

5.5.2.3 Analysis Procedures

For particle size analysis, a small portion of each filter sample was coated with either pure gold or a gold-palladium alloy. After coating, the filters were affixed to the SEM mounts with silver glue. A series of photographs was made, at magnifications of 1000X, of various portions of each filter. An example is shown in Fig. 49. Particle size determinations were made from the photographs, classifying particles in radius ranges of 0.1-0.5 μm, 0.5-1.0 μm, and so on up to 5.0 μm, and then in 2.5 μm classes up to the largest particle observed.

The procedure for chemical analysis of the filters consisted of wet-ashing the filter in a mixture of nitric, perchloric, and hydrofluoric acids, followed by trace metal analysis using atomic absorption spectrophotometry. Details of the procedure were given by Gatz et al., (1972). The elements Ca, Cu, Fe, Pb, Mg, Mn, K, Na, Zn, and others are detectable on the filters using this procedure.

Defined as the actual mass-flow rate divided by the product of the free-stream air density, the aircraft velocity, and the area of the sampler inlet.

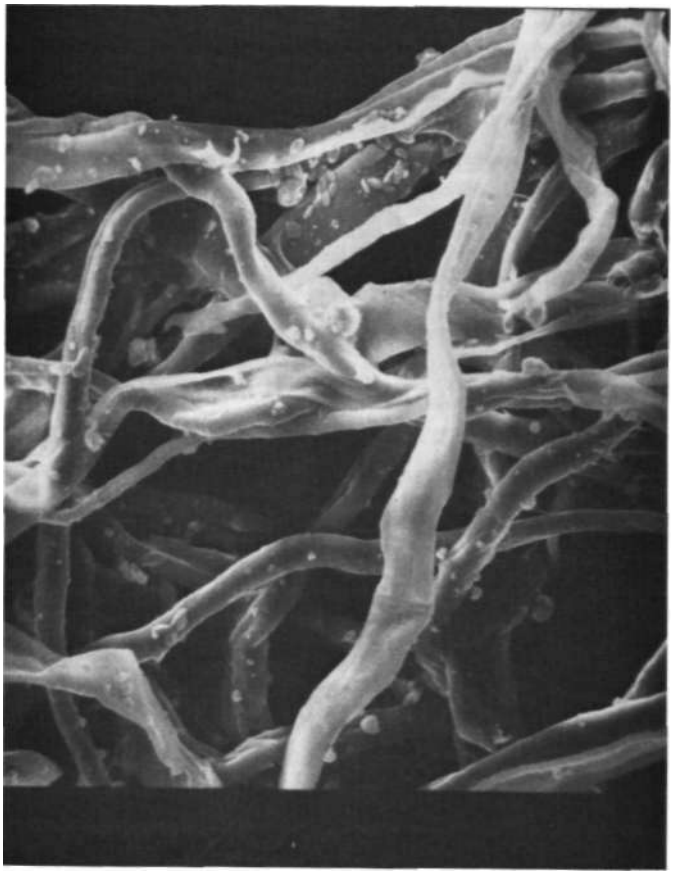


Figure 49. A scanning electron photomicrograph of IPC-1478 filter fibers and collected particles. Magnification 1000X (Los Alamos Scientific Laboratory photo)

5.5.2.4 Error Estimates

This section is included in an attempt to provide a perspective from which to judge whether the data justify the conclusions drawn. Unfortunately, the various uncertainties are difficult to quantify. Thus, the errors given are subjective estimates of the maximum error that may reasonably be expected. In slightly more precise terms, they may be viewed as approximately 2 to 3 times the expected standard deviation.

Particle Size Distributions. Errors in particle size distributions may arise from three main sources: sample volume estimates, sampling procedures, and counting procedures. Incorrect sample volumes would affect the position (absolute error) of the distribution curve on the concentration scale, but not the shape (relative error) of the distribution. The various errors that are possible in the sampling and counting procedures could affect both the position and the shape (i.e., cause both absolute and relative errors).

The error in the aircraft sample volumes due to errors in measurement of inlet area and aircraft speed are expected to be well within 5%. Kelsey (1971) did not discuss the error in the determination of sampling efficiency, but examination of his data indicates that the error is perhaps 5 to 10%. Errors associated with sampling and counting are likely to be much larger.

Kelsey (1971), for example, found that the velocity profile of air passing through the filter is "extremely non-uniform". Indeed, he shows that the. velocity varies by a factor of 4 across the face of the filter. Thus, error may be introduced if the small section of the filter selected for analysis is not representative of the entire volume of air sampled. According to Guthals (1972), this error has been minimized through long experience with the sampling system. Still, an error of 10 to 20% from this source may reasonably be expected.

Filter collection efficiency and its variation with particle size are additional sources of possible errors. Figure 50 shows collection efficiencies given in the literature as a function of particle size and face velocity for IPC-1478 filters impregnated with the organic adhesive Kronisol (dibutoryethyl phtalate). For the St. Louis aircraft sampling conditions—103 m/sec (200 kt.) at an altitude of 610 m (2000 ft) above ground level—the face velocity was about 7.12 m/sec (1400 ft/min). Interpolating between the 5.08 m/sec (1000 ft/min)

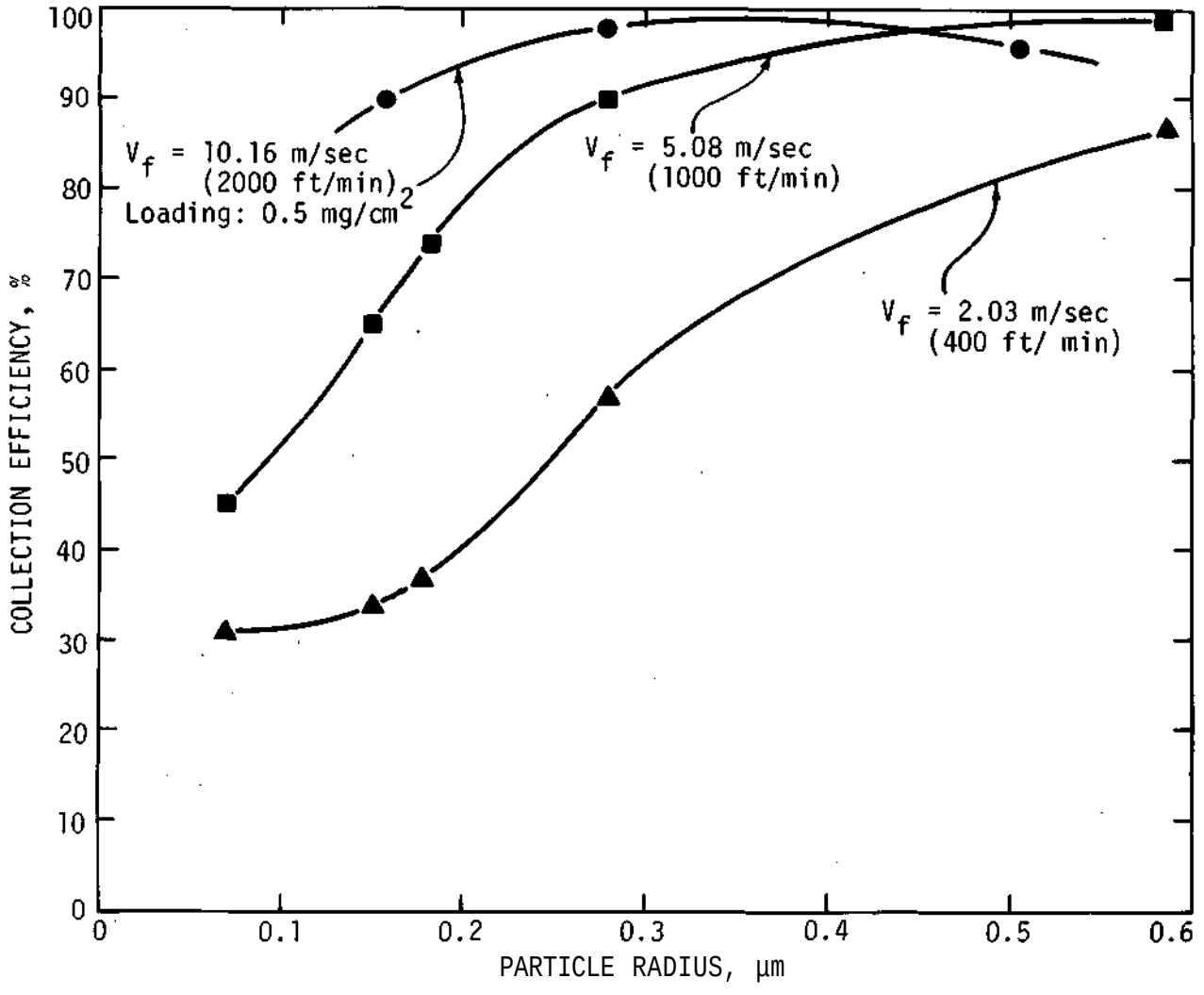


Figure 50. Variation of collection efficiency of Kronisol-impregnated IPC-1478 filters with particle size and face velocity, V_f, for polystyrene latex spheres. Ambient laboratory pressure. Filter loading unspecified unless shown. V_f=10.16 data from Stafford and Ettinger (1971); other data from Stern et al. (1960).

and 10.16 m/sec (2000 ft/min) curves in Fig. 50 gives collection efficiencies for particles larger than 1 μm are unknown. In Fig. 50 the data of Stern et al., (1960) at 5.08 m/sec suggest that collection efficiency increases continuously with particle size. However, the data of Stafford and Ettinger (1971) for 10.16 m/sec suggest that efficiency may decrease somewhat with increasing particle diameter above 1 μm. Overall, on the aircraft filters, the variation of collection efficiency with particle size is not likely to cause errors of greater than 25% in the size range.

The ground-level samples, collected with face velocities of about 2.03 m/sec (400 ft/min) have collection efficiencies ranging from about 30 to 90% over the radius range 0.1 to 0.5 μm. Thus, the particle members in the smallest size category must be systematically increased by a factor of about 2 to be comparable to the aircraft filter results. For particles larger than 0.5 μm radius, the two types of samples should have approximately the same collection efficiencies.

Corrections" for collection efficiency have not been applied to the size distributions presented in the section on results.

Counting procedures are the probable source of greatest error. The IPC-1478 filter mat is made of viscose, having a thickness of 0.084 in and a fiber volume comprising 10% of the mat (Stern et al., 1960). Thus, the filter is a loose, and relatively thick, mat of fibers. A photomicrograph of a filter is shown in Fig. 49. The nature of the filter causes the collected particles to be distributed non-uniformly across the face of the filter; that is, only where there were fibers to intercept them. This increases the danger that the SEM fields selected for photography may be biased toward those containing sharply-focused, visible fibers and particles, and thus be unrepresentative of the filter as a whole.

By far the greatest potential source of error, however, arises from the inability of the SEM photographs to show the entire depth of the filter. Only the fibers and their collected particles near the surface are visible; particles collected deep within the filter mat go uncounted. The systematic errors introduced in this way undoubtedly range from 500 to 1000% and upwards, and perhaps vary slightly with particle size.

With errors of this magnitude, one well might question the usefulness of the particle size data. Certainly the absolute concentrations determined

have little relation to reality. This will be demonstrated later by comparison of these results to Junge's (1963) model for the size distribution of continental aerosols.

On the other hand, certain relatiye comparisons are valid. First of all, comparison of the number concentration and distribution shape among samples collected at the same face velocity should be valid assuming no gross errors due to selection of fields for photography. Thus, ground-level samples may be compared among themselves, and the same is true for aircraft samples. Further, comparison of ground samples and aircraft samples is possible if differences in collection efficiency at each particle size are taken into account.

Chemical Analyses. Errors in reported concentrations may arise from errors in sample volume estimates or from analytical errors. Sample volume errors may be 20%, as previously discussed. With analytical errors included, the overall error is estimated as ±25%.

5.5.2.5 Particle Size Distributions

Ground-level and aircraft filters for 17 and 19 August 1971 have been analyzed for particle size distribution, and a single aircraft filter, collected on 13 August, has been analyzed for trace metals in particulate matter.

Figure 51 shows particle size distributions near ground level and aloft on 17 August. All distributions show a severe departure from Junge's (1963) model for continental aerosols. This is undoubtedly due to the fact that the filter medium collects most particles too deep within the mat to be seen by the SEM. The deviation from the model appears to be least in the largest particles (about 5 to 10 μm radius).

Winds during the collection period were mostly NNE to E at about 2 m/sec, but occasionally lighter and more variable in direction. With such winds, the station at the KMOX transmitter (see Fig. 45) would have been upwind of all major sources. The Coldwater Creek station would have been upwind of the St. Louis/E. St. Louis source area, but downwind of the Alton/Wood River source area. Tyson would have been distantly downwind of all sources. The aircraft sampling track would have been over or downwind of sources at all times, with the possible exception of its southernmost extremity.

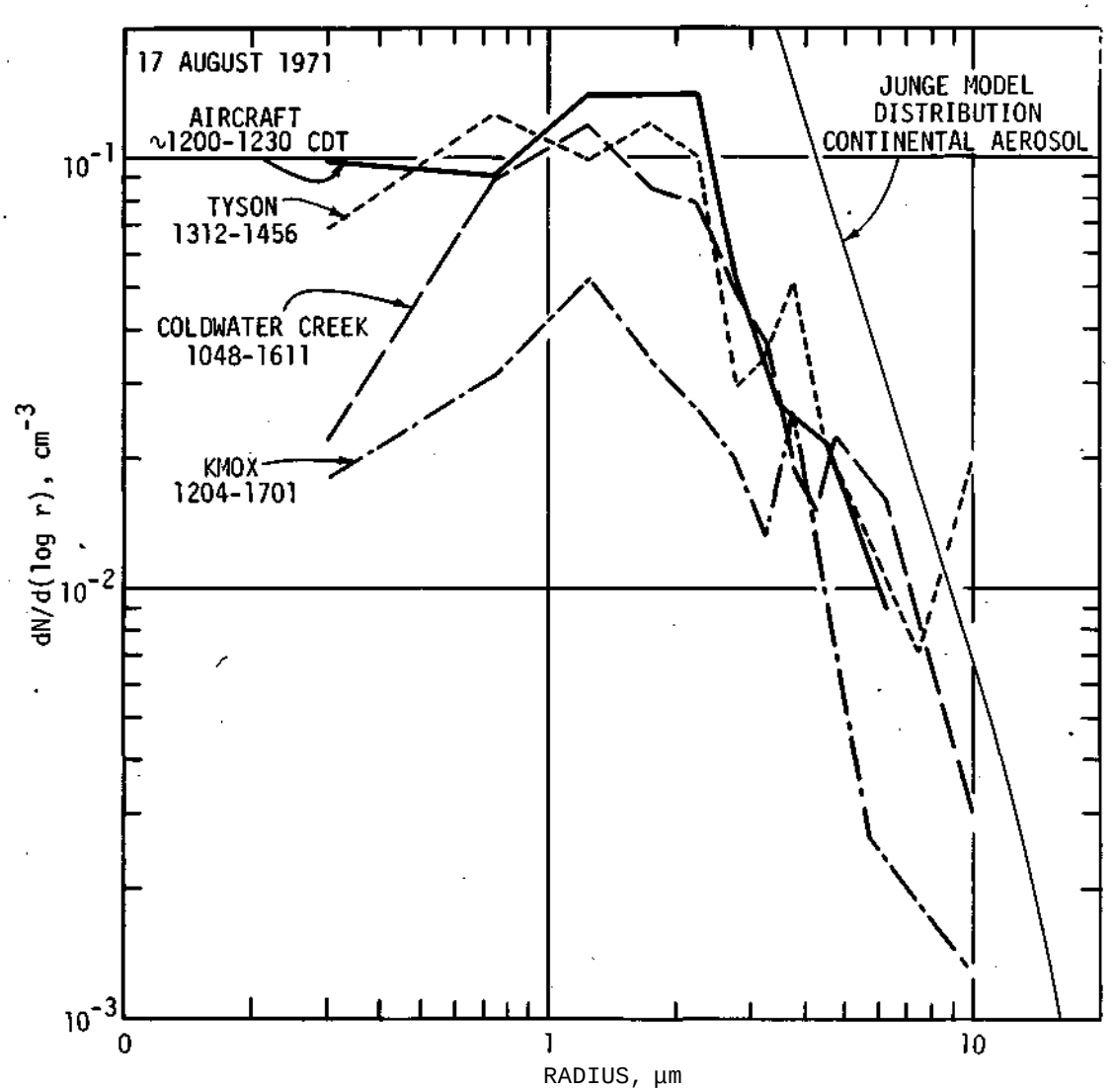


Figure 51. Particle size distributions for 17 August 1971'

The observed distributions in Fig. 51 all have approximately the same shape. The observed drop-off in concentration in the smallest size category on the ground-level samples is not seen in the aircraft sample, but this may be attributed to differences in collection efficiency due to differences in face velocity of the two sampling systems, at least for the KMOX and Tyson samples.

The number concentrations appear to increase from upwind (KMOX) to intermediate (Coldwater Creek) to downwind (aircraft and Tyson). It is somewhat surprising, however, that the aircraft sample and the one collected at Tyson are so similar. After all, the aircraft sample was collected directly over, and immediately downwind, of major sources, while Tyson is at least 24 km (15 mi) downwind of any major sources. In view of the possible errors, however, one should probably not attach great significance to this result.

On 19 August winds were from the S to SW at about 2 m/sec. All distributions (shown in Fig. 52) again show major departures from Junge's model, but the distribution shapes are again similar to each other, and they are similar to those found on 17 August.

Sampling site orientations with respect to pollution sources are now reversed, to a large extent. Tyson is upwind, Coldwater Creek upwind to intermediate, and KMOX downwind. The aircraft track would have been mixed—perhaps 1/3 upwind and 2/3 downwind.

Among the ground-level stations, number concentrations at KMOX are greater than at Coldwater Creek among particles smaller than 4 μm radius, as expected, but Tyson shows unexpectedly high concentrations below 2 μm.

5.5.2.6 Chemical Analyses

One aircraft filter has been analyzed for trace metals. The results appear in Table 15, along with comparative data from the St. Louis County Health Department. The St. Louis County samples were collected near ground level or rooftop level, and are mean concentrations for August 1971. Thus, they are not strictly comparable to the 1/2-hour aircraft sample collected at 610 m above ground level. Nevertheless, the comparison serves as a rough indication of aircraft data quality. Table 15 shows that concentrations measured from the aircraft filter correspond roughly to the highest monthly means at the St. Louis County stations.

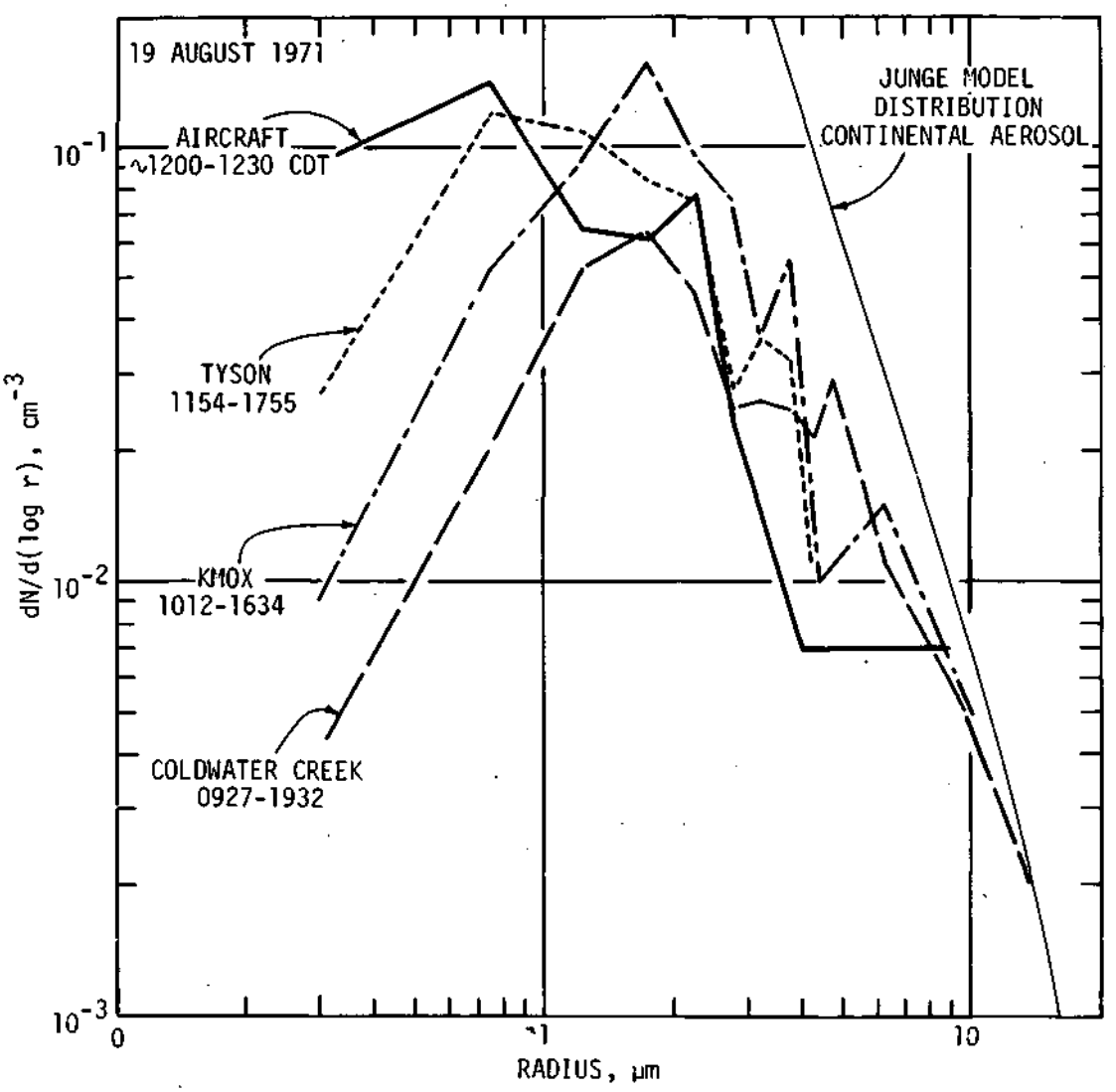


Figure 52. Particle size distribution for 19 August 1971

TABLE 15

Trace Metal Concentrations from Aircraft Sample
with Comparative Data

	Cu	Fe	Mg	Mn	Pb	Zn
Concentration aloft, 13 Aug. 1971, 2000 ft AGL, $\mu g/m^3$	0.139	5.53	4.54	0.198	1.87	0.583
Detection limit, $\mu g/m^3$	0.0008	0.03	0.004	0.003	0.004	0.007
St. Louis Co. Health Dept. Monthly Mean, Aug. 1971, $\mu g/m^3$						
Northland	0.029	1.96		0.070	1.38	0.41
Sinks	0.375	1.16		0.029	0.54	0.47
Chambers	0.070	1.78		0.042	0.79	0.56
SLCHD	0.032	1.44		0.048	1.68	0.00
Mt. St. Rose	0.062	2.47		0.039	0.66	0.00
Harold	0.148	3.04		0.117	1.38	0.00
ACIC	0.059	4.23		0.068	0.60	0.07
T-4	0.075	0.96		0.043	0.84	0.17
St. Ann	0.041	1.70		0.045	1.48	0.03
State Farm	0.130	1.87		0.052	1.76	0.67

The surface winds on 13 August were from the SE to SSE, with speeds between 0.9 and 2 m/sec at sampling time. The low-level sounding taken from the Arch at 12 30 CDT indicated a mixing depth of 2070 m with the average winds to that height from 170° at 1.4 m/sec. These data indicate a very limited horizontal air motion, which would allow greater than normal pollutant concentrations to build up over the St. Louis area. Thus, the finding that concentrations at 610 m correspond approximately to the highest monthly means among the St. Louis County surface stations is not at all surprising.

5.5.2.7 Conclusions

The usefulness of particle size distributions determined from SEM counts of IPC-1478 filter collections are limited to relative comparisons

between sampling locations. Such comparisons show that particle concentrations increase from upwind to downwind locations. No differences were apparent between particle concentrations at 610 m above the surface and surface stations having the same orientation with respect to pollution sources.

Concentrations of trace metals at 610 m on 13 August 1971 appear to be reasonable in comparison with monthly means from St. Louis County surface stations, in view of the weather conditions at sampling time.

6.0 CASE STUDIES

6.1 Introduction

Thunderstorms which occurred on two days (August 3 and July 18) in the summer of 1972 and passed through the METROMEX data collection network were selected for detailed analysis. On both days heavy rainfall totals were recorded in and downwind from the St. Louis urban area. These storms have been studied using a variety of meteorological instrumentation including surface networks, radars, upper air observations, and aircraft. The objective of the analysis was to determine the components in the moisture budget of the storms, the airflow structure of the storms and the efficiency of the storms in scavenging tracer materials released into the storms by aircraft. It is hoped that the results of these case studies will ultimately help to reveal the causes of the climatological precipitation maximum noted downwind from St. Louis.

The following gives a preliminary description of the air flow structure of the tracer treated thunderstorms at selected observation times. The general meteorological conditions which prevailed on the two days will also be described.

6.2 3 August 1972 Case

6.2.1 Storm Rainfall

On August 3 between 1119 and 2203 CDT a number of rain storms passed over the St. Louis area. The resulting rainfall pattern, Fig. 53, shows a very striking maximum downwind from the St. Louis urban area where totals

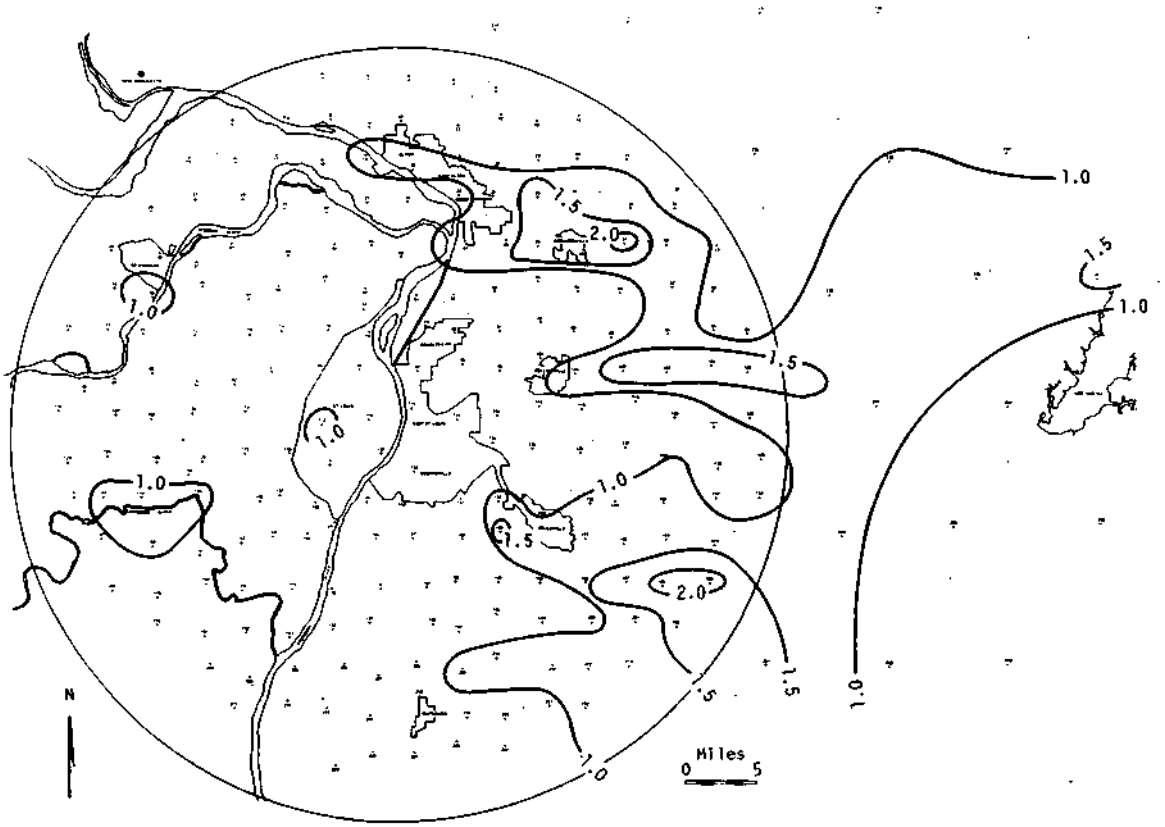


Figure 53. The total storm rainfall (inches) on Z August 1972 (1124-2055 CDT).

exceeding 1 in. were received over a large area. Totals of over 2 in. were recorded at two locations, one southeast and one northeast of St. Louis. Both of these rainfall maxima are within the Survey research circle where urban influences can be expected to affect rain systems which have passed over the urban area. In short, August 3 appears to be an ideal day for the study of urban effects on precipitation.

6.2.2 Synoptic Conditions

On August 3 the 500 mb (0700 CDT) flow over the METROMEX area was essentially zonal, but rather high wind speeds were observed over Topeka, Kansas, and a small amplitude short wave was approaching the METROMEX area from that state. Precipitable water in most of Missouri and Illinois was greater than 1.50 in. and moisture gradients were fairly strong. An interesting series of small pockets of very dry air was observed at the 500 mb level during a period of a few days before and after August 3. Charts from 1900 CDT show that the short wave and high velocity winds moved through the METROMEX area during the course of the day releasing the convective instability in conjunction with events noted at the surface.

Surface charts indicate that two rather closely spaced cold fronts had approached the area from the northwest on the previous day as a cold area of high pressure pushed into North Dakota. By 0700 CDT on August 3 it was hard to distinguish two fronts, but there was a trough from central Missouri through central Indiana and a cold front close behind it reaching from lower Michigan through northern Missouri to the panhandle region. The NWS radar chart showed a broad band of precipitation associated with these features.

By 1600 CDT the front had moved into central Illinois and central Missouri with the trough still ahead of it in southern Illinois and southern Missouri, (see Fig. 54). A line of thunderstorms developed along the front, from Terre Haute, Indiana, to Columbia, Missouri, and began moving to the south. One of a series of small lows that moved along the trough line from southwest to northeast was over the METROMEX area during the period of thunderstorm activity.

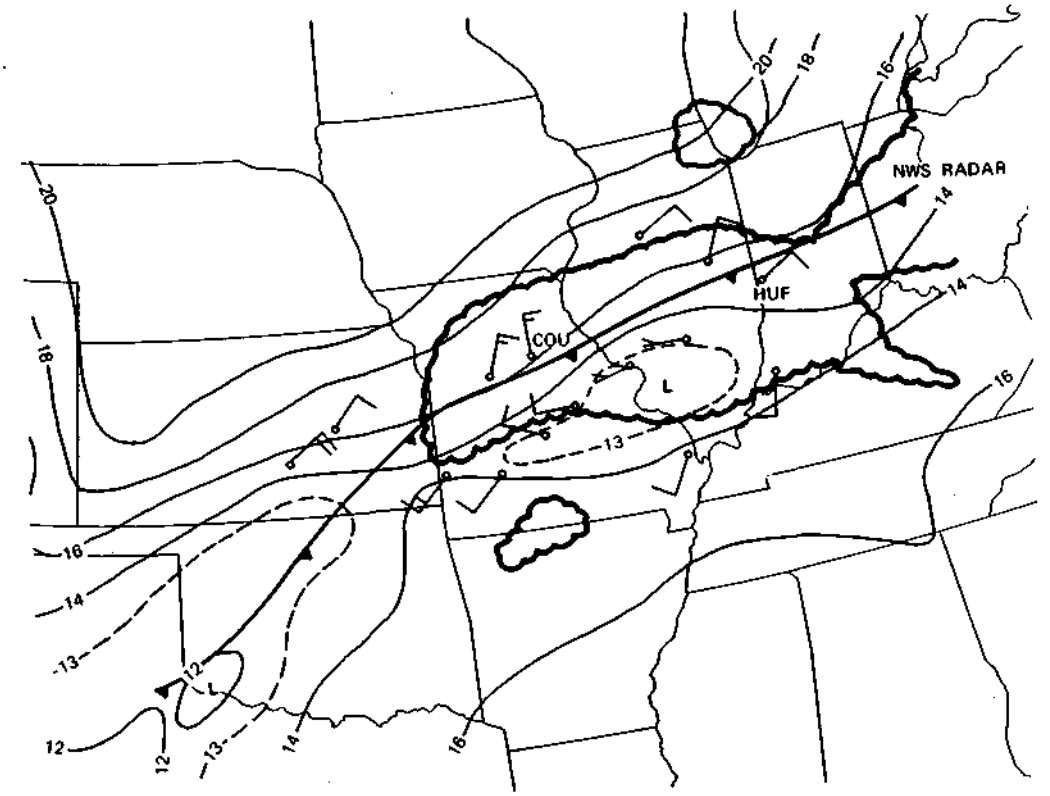


Figure 54. The surface analysis at 1600 CDT, 3 August 1972. The NWS radar echo area is indicated through Missouri, Illinois, and Indiana.

6.2.3 PPI and Raingage Network Data

Photographs of the PPI scope of the FPS-18 radar indicate that an echo which passed over southern St. Louis merged with other echoes southeast of the city at about 1545 CDT. The merging of echoes continued until about 1600 CDT.

The charts from 227 recording raingages in the METROMEX primary research circle were analyzed for 5 minute rainfall rates. Maps of 5 minute rainfall rate were plotted and analyzed to study rain cell movement from one 5 minute period to another. Figure 55 shows contours of rainfall rate for the period 1600-1605 CDT. Rainfall rates for the rain cells labeled 13 and 14 exceed 1.0 in/hr and 1.5 in/hr, respectively, during this 5 minute period. These two rain cells are part of the merged system and represent peak rainfall conditions observed in the raingage network. Unfortunately, part of the merged system was just beyond the network boundary.

During the echo merging period three interesting developments took place: 1) the total rain flux in the network doubled from the rain cells that comprised the merged system; 2) hailstreaks were observed at four nearby locations; and 3) between 1555 and 1605 CDT, when rain cell 13 was at an intense state, it made a dramatic change in its direction of propagation. Rain cell 13 had been moving from the west toward the east at a rate of about 12 mph. It moved sharply to the left of its original path during a 10 minute interval and in an apparent cyclonic manner. The centroids of the areas enclosed by the most intense rainfall rate contours were plotted for the various map time intervals (Fig. 56), the rotation is readily noted. The cell duration was forty minutes. The motion of a nearby slow moving (7 mph) but intense rain cell 14 was not similarly affected.

6.2.4 RHI Observations

The vertical structure of the storm was continuously observed and photographed on the University of Chicago's 3 cm TPS-10 radar located in Greenville, Illinois. Figure 57 is a tracing made from a RHI scope photograph taken at 1600 CDT. The radar was scanning along azimuth 232° and the large echo with a top near 40,000 ft was directly associated with rain cell 13. The large overhanging structure observed beyond a range of

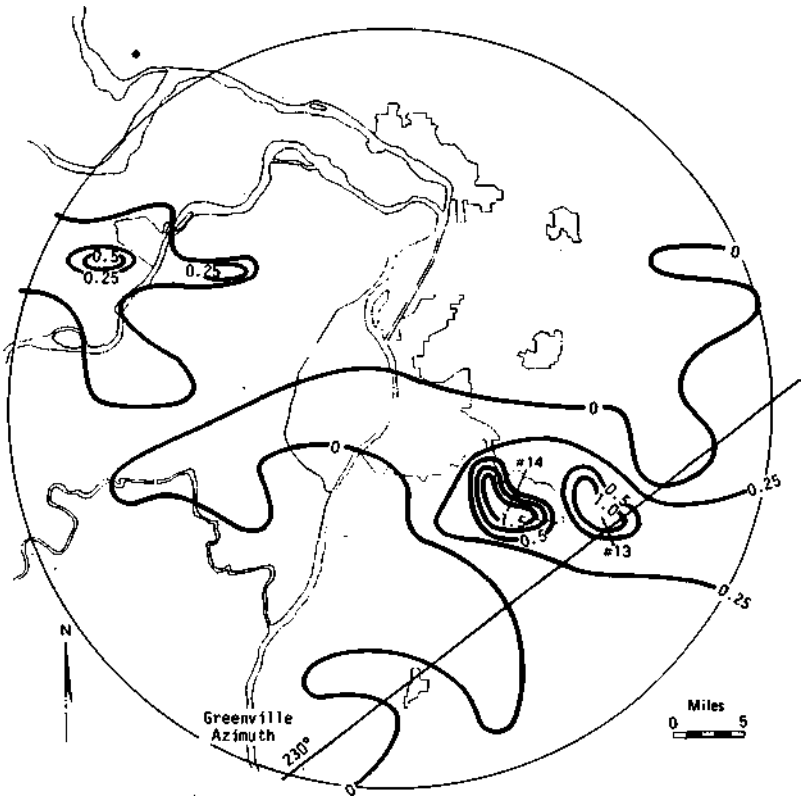


Figure 55. The analysis of 5 minute rainfall rate (in./hr.) for the period 1600-1605 CDT, 3 August 1972.

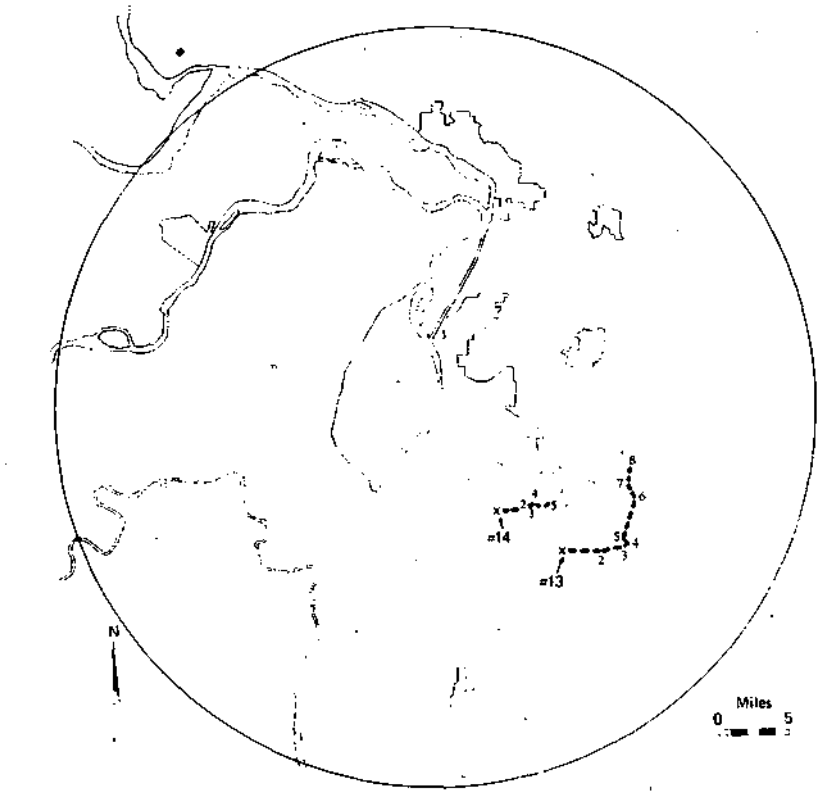


Figure 56. The paths of rain cells 13 and 14 between 1535 and 1615 CDT, 3 August 1972.

38 miles and at an elevation of 15,000 ft is likely associated with the storm's updraft (Browning and Ludlam, 1962).

6.2.5 Aircraft Flight Path and Observations

Observers in the aircraft releasing the tracer materials noted that most of the inflow was taking place along the trailing W and SW sides of the storm, where the overhang was located. The flight path of the airplane is shown in Fig. 58. The plane flew at an elevation of approximately 5,000 ft (msl) and attempted to stay in updrafts as much as possible while dispersing the two tracers. Figure 58 indicates that the plane was in excellent position for viewing the updraft area of the storm at 1602 CDT. Small updraft areas about 0.5 mile in length with vertical velocities of about 500 fpm were recorded by the aircraft's instrumentation. The tracer materials, lithium and indium, were released from the aircraft into the storm between 1526 and 1623 CDT. The analysis of the surface deposition as measured by 81 special bottles placed in the rectangular rain water collection network has not yet been completed. When the tracer analysis is completed it is expected to aid in the depiction of the outflow structure of the storm.

6.2.6 Upper Air Observations

The pibal and radiosonde release points being used on August 3 are shown in Fig. 59. A total of 31 double theodolite pibals and 14 radiosondes were released between 1100 CDT and 1900 CDT. The pibals were released from 7 sites every half-hour if it was not raining, and the radiosondes were released from 3 sites (site 31, 32, 33 in Fig. 59) every hour. The radiosondes were tracked with single theodolites.

The low level wind field in the vicinity of the storms was obtained from the theodolite observations. Figure 60 shows the streamline analysis at 500 m (msl) for 1600 CDT. Unfortunately there were no observations to the south of the storm. The storm was fed from behind by the basic westerly to west-southwesterly flow, but there was also a broad feeder current that approached the storm from the north and northwest. The feeder current from the W or SW would have to rotate in a cyclonic sense in the immediate vicinity of the storm to enter the storm.

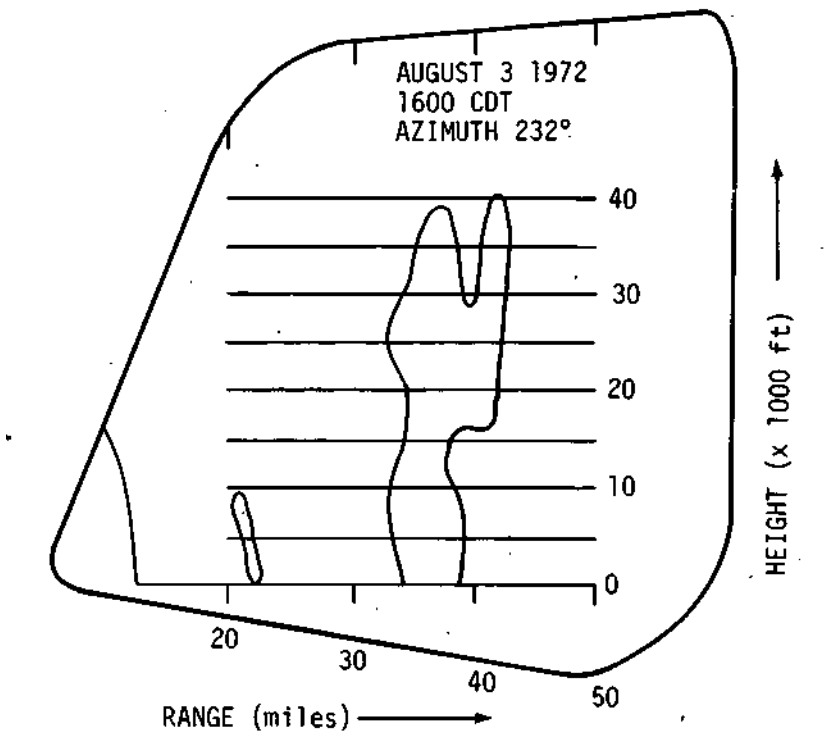


Figure 57. An RHI radar photograph of rain cell 13. Note the overhang at 16,000 ft. visible at 41 miles.

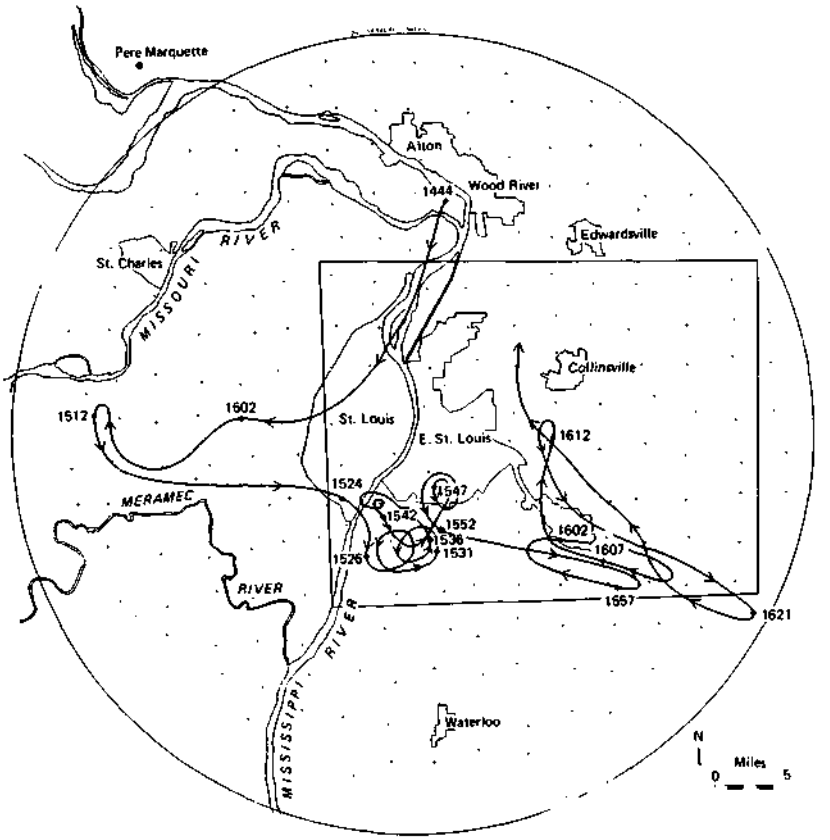


Figure 58. The flight path of the aircraft following the updraft areas associated with one of the storms of 3 August 1972

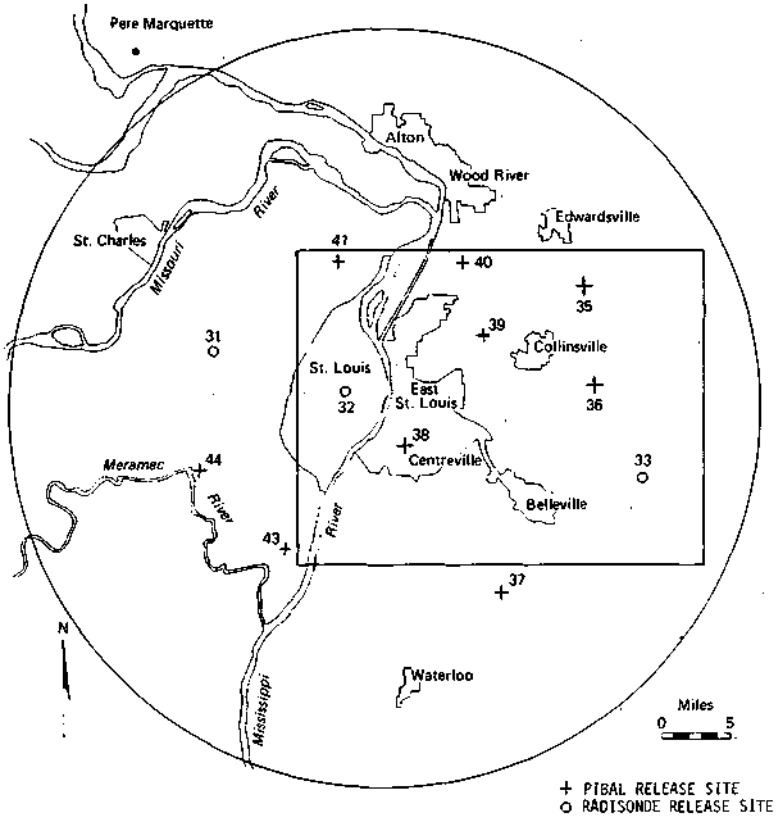


Figure 59. The sites utilized for upper air observations for the METROMEX research project in 1972

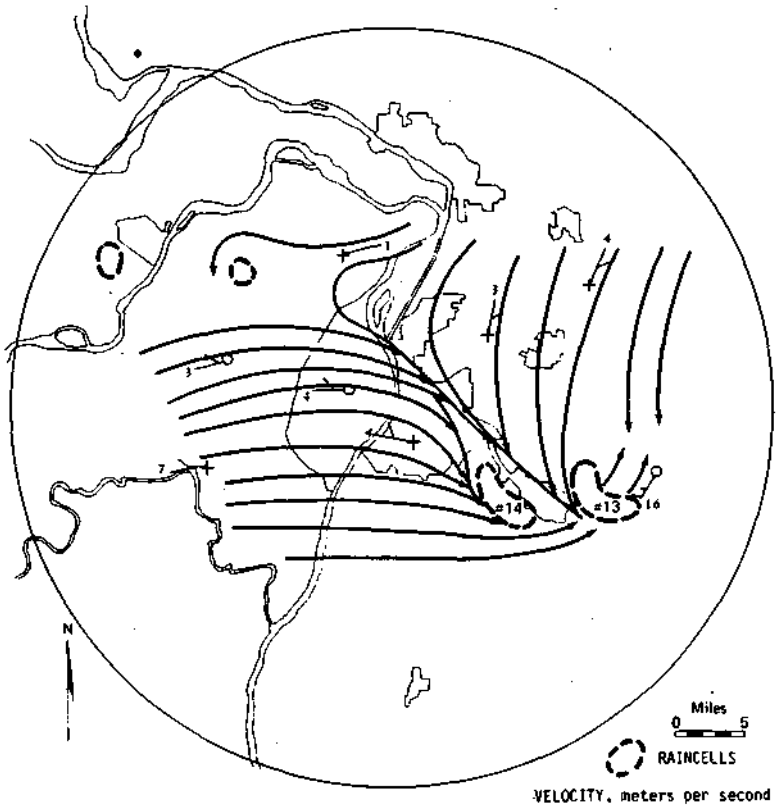


Figure 60. The streamlines at 500 m MSL at 1600 CDT, 3 August 1972

The strong southerly wind observed at site 33 in front of the storm was the result of the radiosonde being caught in the. cool outflow from rain cell 13. The radiosonde was only 300 m above ground and reported temperatures in the low 70's and relative humidities of about 80%.

The northerly current observed at 500 m was also indicated at the 1000 m level and in the confused ground level wind field depicted by a network of 17 surface wind stations.

At 1500 m the flow was from the WSW with only a slight deflection toward the convective area (see Fig. 61). Between 2000 m and 3000 m, the flow was west-south-westerly and there was no indication of any flow deflected toward the storm region.

At all map levels (0-3000 m) there was a tendency for winds to be stronger in the southwestern part of the research area than in the northeastern part.

The radiosonde observations from sites 31 and 32 indicate warm (temperature greater than 80°F), dry air (RH about 50%) at 500 m over the city. Figure 60 indicates that this same dry city air was transported into the updraft region of the storm.

6.2.7 Air Flow Structure

The inflow structure of the August 3 storm as viewed at 1600 CDT in this preliminary analysis consisted of two main parts in the lower atmosphere. There was a large inflow area below approximately 1000 m and apparently to the north and west of the storm. Above the 1000 m level less air was deflected into the storm from the surrounding environment. However, the general convergence observed in the storm's environment up to at least 3000 m indicates that significant amounts of air were still entering the rear of the storm at these levels which, it should be noted, are well beneath the overhang. This conclusion is supported by the fact that there was no distinct evidence of potential type flow developing around the edge of the storm-, that is , there is no observed increase in wind speed along the lateral edges of the storm perpendicular to the direction of the ambient air flow (Fankhauser, 1971).

The low level flow that approached from the north acquired positive relative vorticity upon entering the storm. The updrafts resulting from this

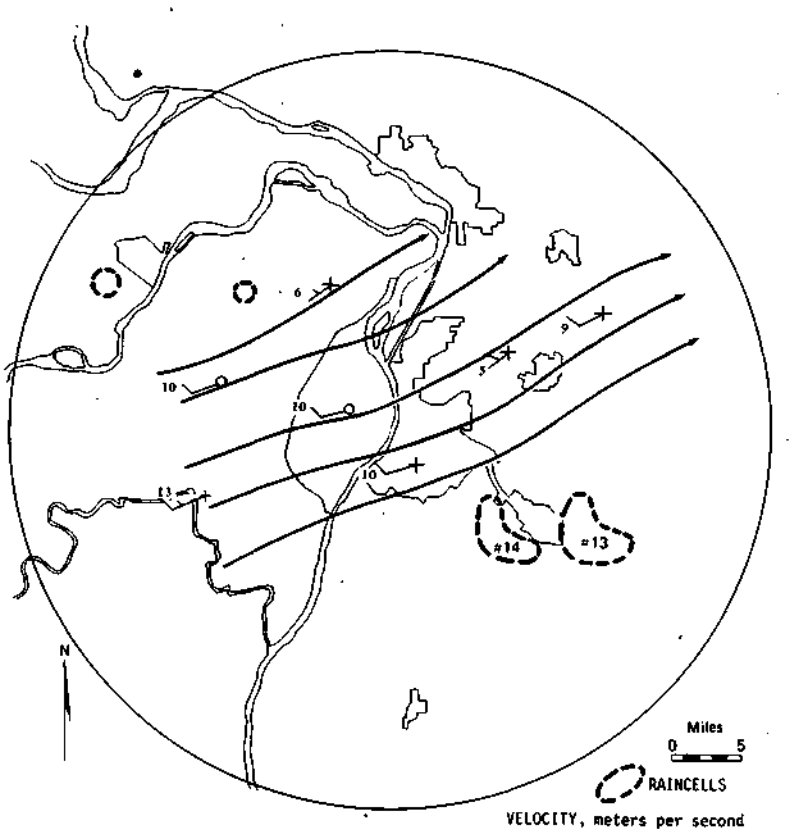


Figure 61. The streamlines at 1500 m MSL at 1600 CDT, 3 August 1972

inflow were mostly small and not particularly strong when encountered by the aircraft at 1500 meters. This may imply that the updrafts combined into a more organized, possibly rotating, structure at some higher level.

The outflow from the storm system was found in front of the storm and exhibited a rain shaft that seemed to rotate in a cyclonic manner. This occurred during a period when PPI echoes were merging, rain cells were intensifying, and downdrafts were doubtlessly consolidating.

What the effect of the urban environment was on this storm is not yet clear, of course. However, it is clear that rather dry air was ingested by the storm from over the city. It is felt that the energy available to the storm's updraft may have been lessened by the inflow of dry air. The ability of the updraft to support liquid water may have been lessened, and precipitation intensified by this.

6.3 18 July 1972 Case

6.3.1 Storm Rainfall

On July 18 a series of rain storms moved through the St. Louis area between 1124 and 2055 CDT. Rainfall totals exceeding 1 in. were recorded in 5 different areas in the METROMEX circle (see Fig. 62). Two of these areas were downwind from the city and were therefore prime subjects for the study of urban effects on rainfall.

6.3.2 Synoptic Conditions

The St. Louis area was under the leading edge of a 500 mb trough which covered much of the central and western United States and ridges were observed over both the Pacific and Atlantic coasts. Flow over the METROMEX area was from the south-southwest at all levels at 0700 CDT. A strong moisture gradient existed to the northwest of St. Louis. The precipitable water was 1.62 in. and 0.51 in. at Salem, Illinois, and Omaha, Nebraska indicating a strong moisture gradient northwest of St. Louis. Between 0700 CDT and 1900 CDT a pocket of cold air moved over the METROMEX area.

A careful analysis of consecutive surface synoptic charts revealed that at 1300 CDT (Fig. 63), there was a cold front which had become stationary along the Iowa-Missouri border. This front was identified by the 65°F

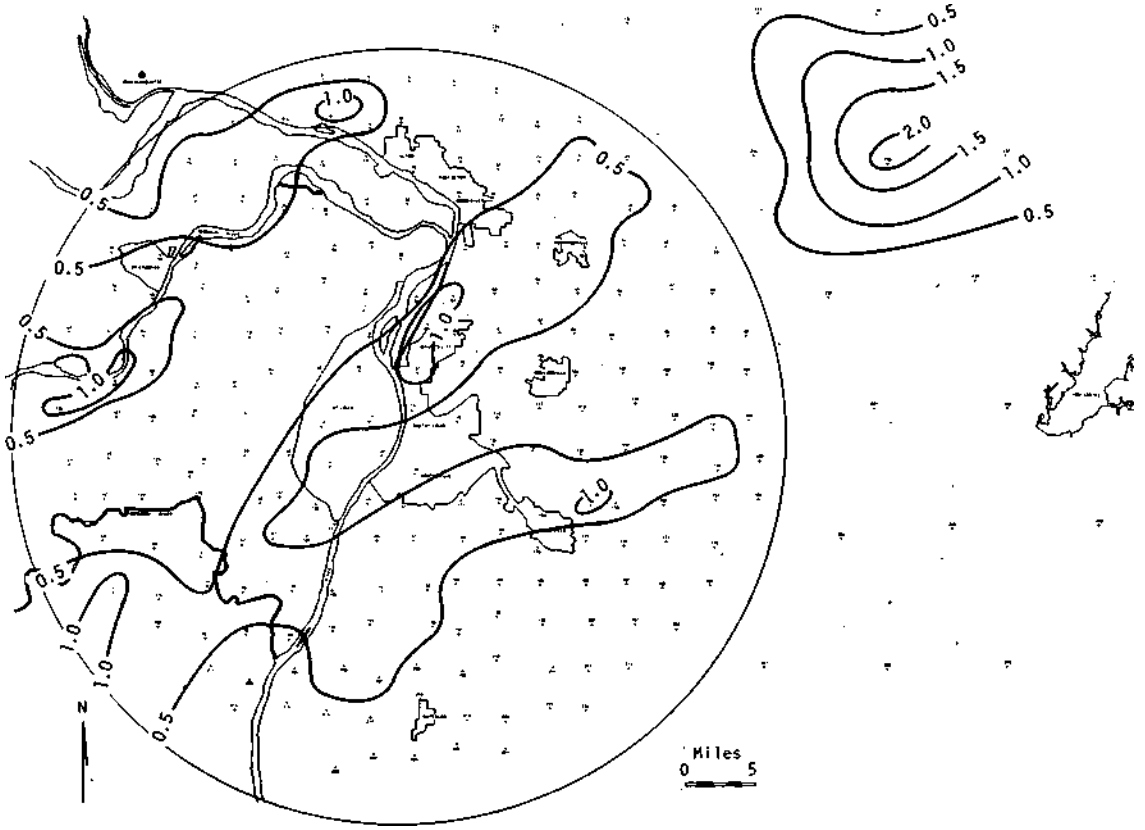


Figure 62. The total storm rainfall (inches) on 18 July 1972

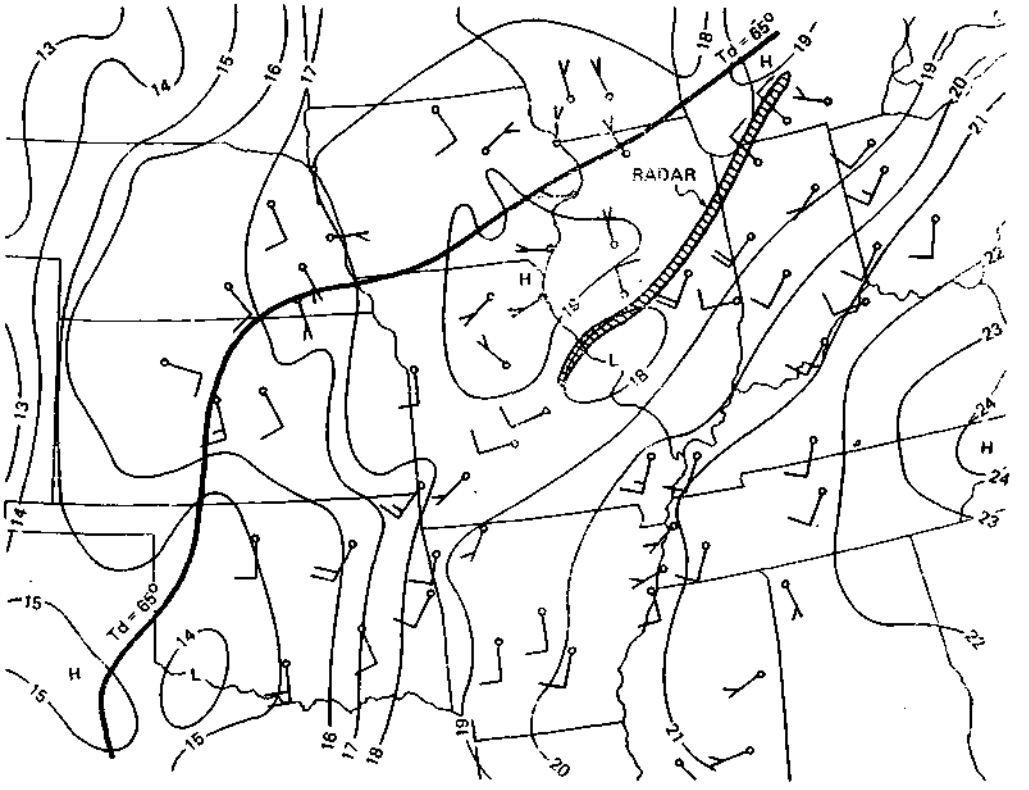


Figure 63. The surface analysis at 1300 CDT, 18 July 1972, with the radar indicated squall line

isodrosotherm which marks the northern boundary of the warm tropical air covering Missouri. A trough of low pressure was found south of the front passing through central Missouri and central Illinois. A squall line which had formed along the front on the previous evening approached St. Louis on 18 July, in association with the trough. The squall line is clearly distinguished from the frontal clouds on satellite photographs.

The southward movement of the trough was halted over the St. Louis area by the westward movement of the Bermuda high. As a result showers moving along the trough continued to pass through the St. Louis area until the trough dissipated around 2100 CDT.

6.3.3 Raingage Network Data

The observation time selected on July 18 for inclusion in this report was 1400 CDT. Figure 64 shows the 5 minute rainfall rates observed in the raingage network between 1400-1405 CDT on July 18. The squall line entered the city at this time. Rain cell 8 was precipitating at a rate of more than 0.25 in/hr. The path of the centroid of the area of greatest rainfall rate is seen in Fig. 65. Two small deflections to the right of the mean direction of motion can be observed. The cell lasted about 55 minutes and was responsible for a large part of the rainfall associated with the maximum at Granite City in Fig. 62. The cell moved toward the east-northeast at a mean velocity of 24 mph.

6.3.4 RHI Observations

A tracing from a photograph taken of the RHI scope of the University of Chicago's TPS-10 radar is seen in Fig. 66. The observed echo between 45 to 50 miles was directly over rain cell 8. The storm top was less than 20,000 ft at this time with a small forward overhang at about 6000 ft.

6.3.5 Aircraft Flight Path and Observations

Reports from the aircraft on July 18 indicated that rainstorms in the squall line were received inflow along the southeast side of their leading edge. The updraft areas and velocities were not large (0.5 mile length and 600 fpm).

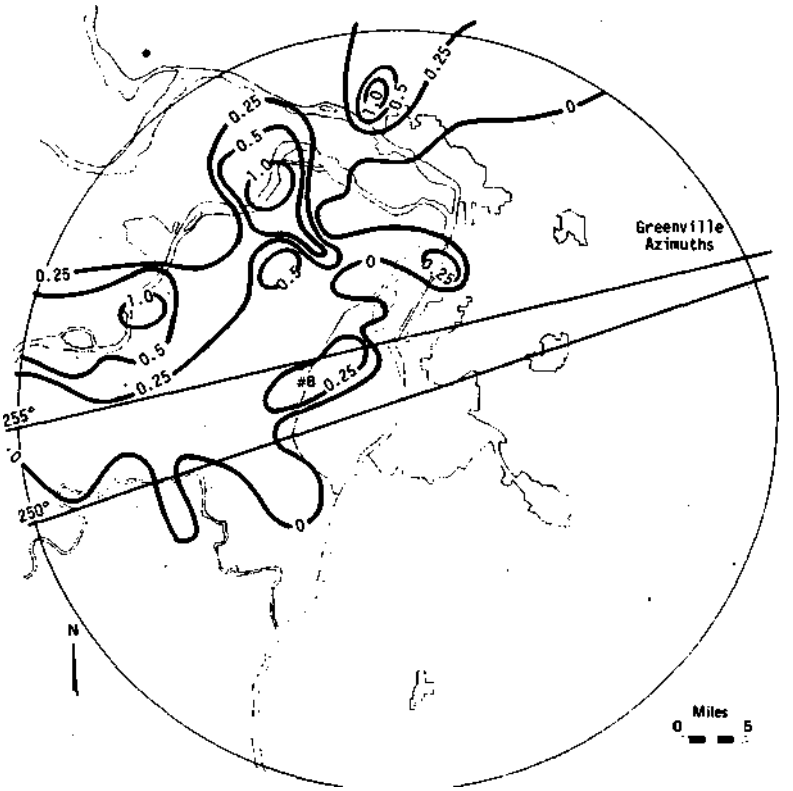


Figure 64. The analysis of 5 minute rainfall rate (in./hr.) for the period 1400-1405 CUT on 18 July 1972. Two azimuths are shown which indicate the region studied with RHI radar.

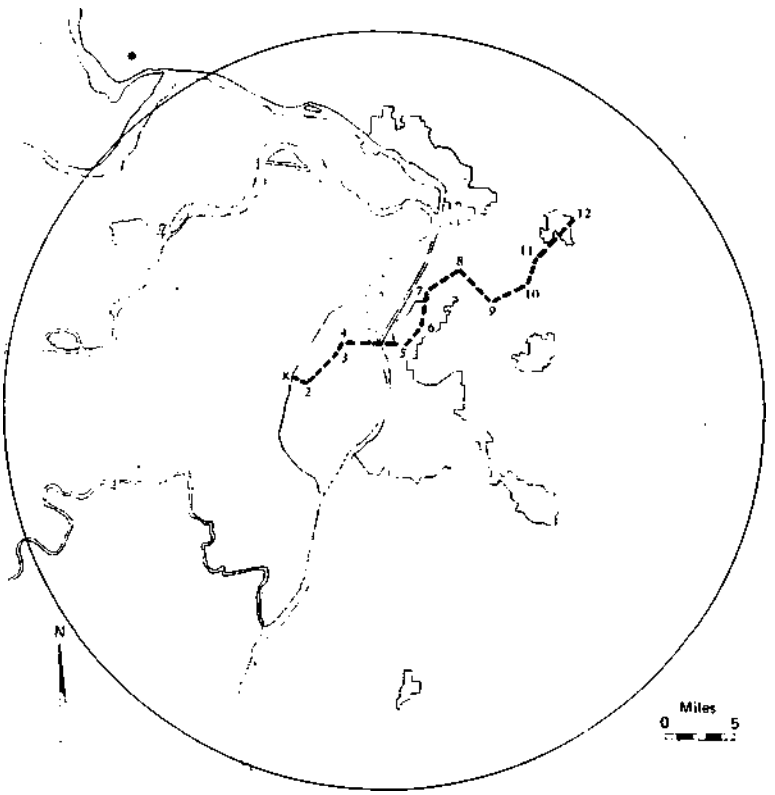


Figure 65. The path of rain cell 8 derived from 5 minute rainfall rates for the period from 1400 to 1500 CDT on 18 July 1972

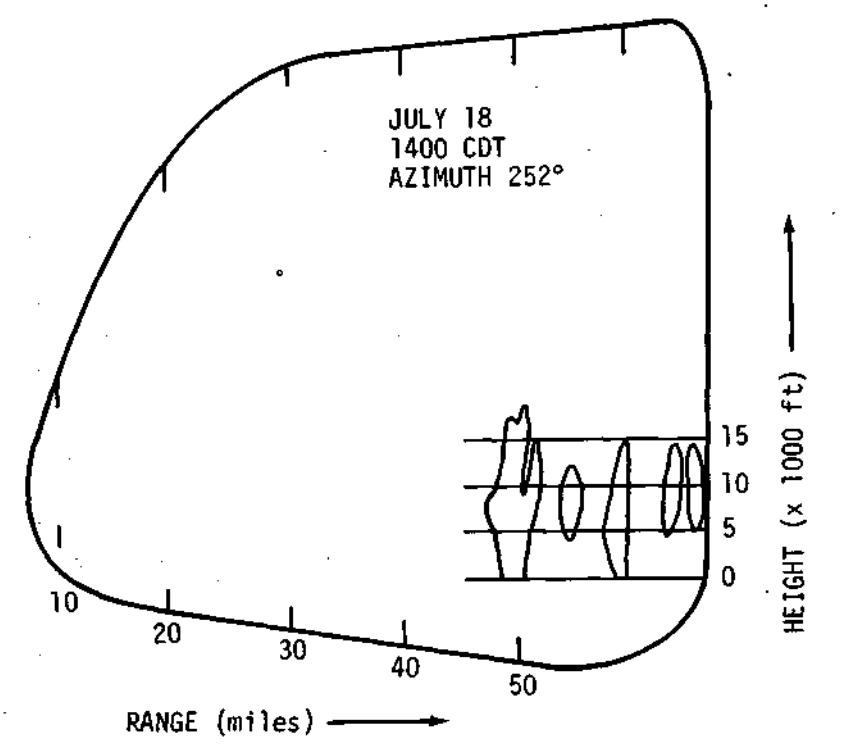


Figure 66. A 3-cm RHI radar photograph of rain cell 8 located at approximately 50 miles and extending to 20,000 ft. A slight overhanging echo at 6,000 ft. was observed at 47 miles.

The flight path is seen in Fig. 67. The tracer materials were dispersed between 1415 and 1510 CDT while the plane flew in updrafts at an elevation of approximately 5000 ft.

6.3.6 Upper Air Soundings

On July 18 a total of 23 pibals and 14 radiosondes were released in accordance with a time schedule similar to that of August 3.

The 500 m streamline chart, Fig. 68, obtained from the 1400 CDT soundings, shows a strong line of convergence and shear in front of the squall line rain cells. The convergence resulted from the cold outflow of the rain cells embedded in the south-southwesterly flow of the surrounding environment.

At 1000 m (Fig. 69), the outflow structure was not observed, and the inflow structure was along the southwest side of the storm. The pibal released from site 44 was affected by an echo that had not reached the ground. The radiosonde from site 31 was not affected by rain cell 8, and there was no evidence of any organized inflow on the trailing side of the storm.

The streamline charts between 1500 m and 2500 m do not indicate flow moving toward the storm.

The radiosonde released from site 32 was in the inflow current at the 1000 m level. The inflowing air had a relative humidity of about 50% and a trajectory from over the southern part of St. Louis.

6.3.7 Air Flow Structure

The storm observed in the squall line of July 18 was characterized by an inflow current that approached the storm from the front on the right hand side at an elevation of about 1000 m. This inflow current passed over a lower outflow current that was also to the front of the storm. The overhang observed by the radar at 6000 ft implies that air entered the storm at this level. There was no indication that the inflow was deflected into the storm from the nearby environment at levels above 1500 m. Thus, the main source region for updraft air was below 1500 m and to the southeast of the storm. As in the August 3 case, the rather dry air entered the storm in the main inflow current. This lessened the ability of the storm to support precipitation sized particles and led to the initiation of the rainout stage of cell 8.

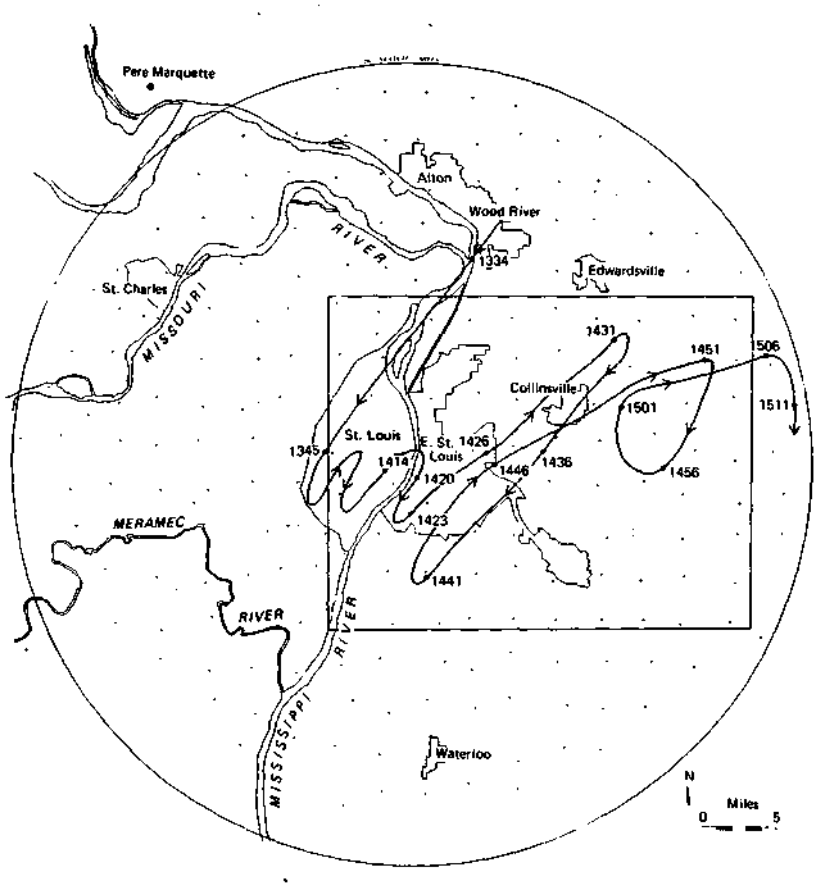


Figure 67. The flight path of the aircraft in front of the eastward moving storm system on 18 July 1972 (all times are CDT).

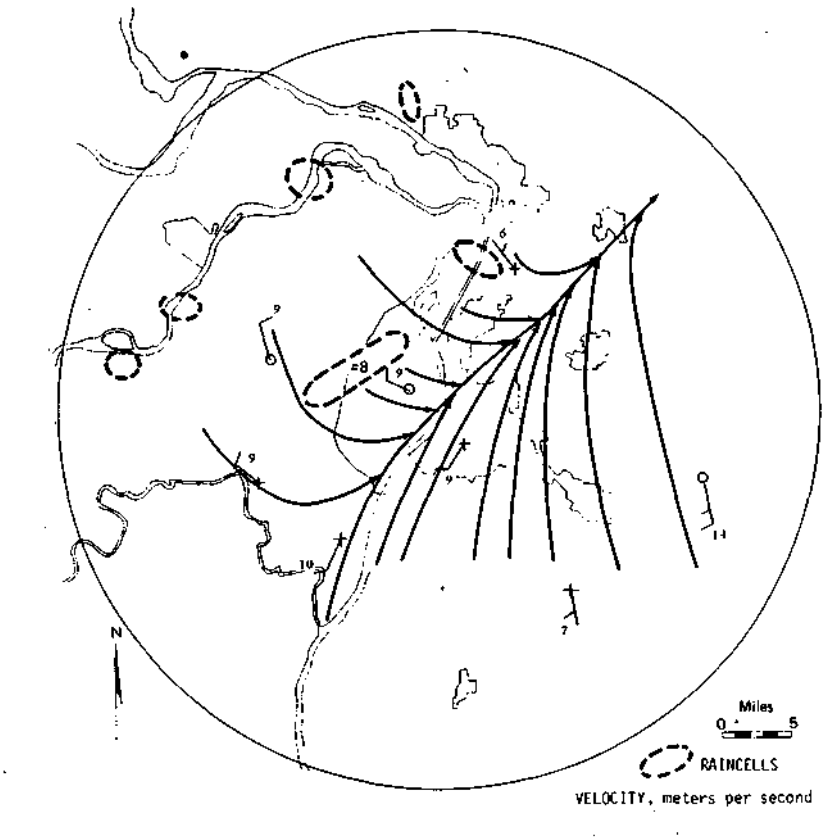


Figure 68. The streamlines at 500 m MSL at 1400 CUT, 18 July 1972

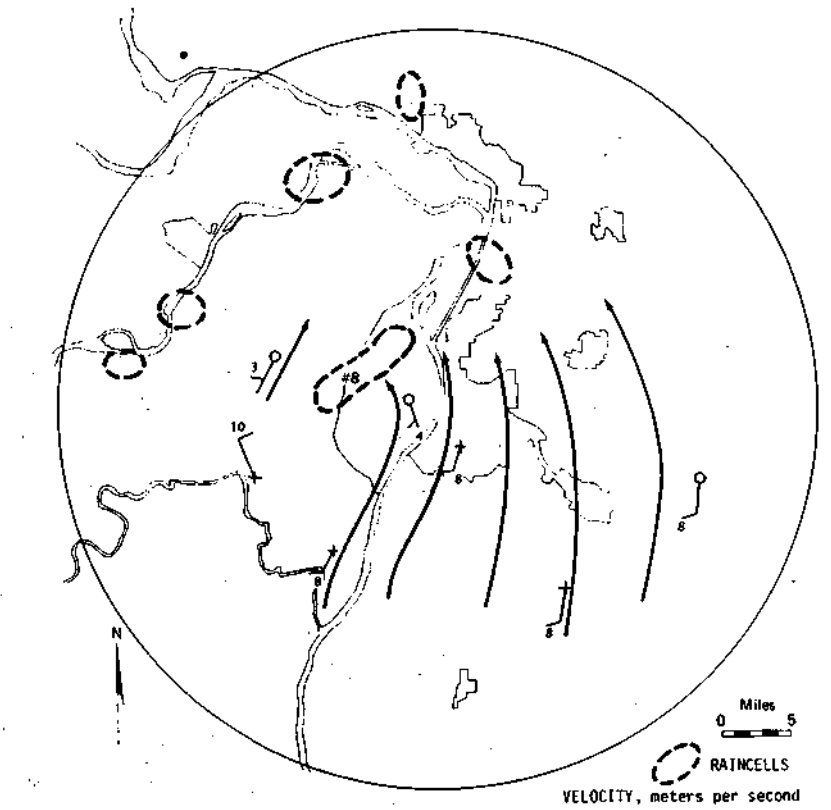


Figure 69. The streamlines at 1000 m MSL at 1400 CDT, 18 July 1972

6.4 Summary of Meteorological Analysis

Two storms which passed over the St. Louis urban area producing heavy rainfall, were studied at selected time intervals. The storms occurred during different synoptic conditions and had markedly different air flow structures. However, both storms seemed to derive most of their inflow from low levels. In both cases, the air which it passed over the city was rather dry as it entered the storm. This may have weakened the updrafts of the two storms and led to increased precipitation activity.

6.5 Some Li Tracer Chemistry Results

Because natural background Li is detectable by the analytical techniques, the first requirement for assessing time of tracer deposition is to identify the tracer Li in the presence of the natural background. Along with such physical evidence as the time and place of the tracer release in relation to the position of the sampling station and the winds in the air layer below the release level, there are certain features of the chemical analyses of sequential rain samples that provide independent evidence of the presence of tracer materials.

High concentrations are tracer material, although not as definitive as desirable because of the large natural variability of elemental concentrations in precipitation.

Deviations from normal rainfall rate-concentration relationships with time are somewhat stronger evidence, although again there is the uncertainty of natural variability to contend with.

Concentration ratios, involving Li and another element, especially a soil-derived element, may provide the best clue to the presence of tracer Li in rain. Deviations of measured ratios from such norms as ratios observed in non-tracer rains, in local soils, or earth's-crust values, should provide strong evidence for the presence of tracer Li.

Sequential rain samples were collected on 18 July and 3 August 1972—days when tracer Li was released into thunderstorms. Note that the elemental concentrations presented are for the soluble portion of the samples only. This portion is expected to include all tracer Li because LiCl, the specific chemical form of the tracer released, is very soluble in water. Soil-derived

Li and the other elements determined are expected to be at least partially insoluble in rain.

On 18 July, the sequential sampler at the KMOX radio transmitter (Fig. 45) collected 33 samples. The rain began at 1434 CDT, ended sometime after 1734 CDT, and produced 18.9 mm of rain, as measured by the sampler. The concentration, rainfall rate, and element ratios for the main portion of this rain are shown plotted against time in Fig. 70. Lithium tracer was released from 1414-1510 CDT.

The upper portion of Fig. 70 shows the time history of soluble Li concentration during the rain. The first sample had the highest sample found in any sequential sample during the entire 1972 season—2.24 part per billion (ppb). During the next 16 min of rain, the concentration dropped by a' factor of about 10, with a number of short-period fluctuations that appear to be related to the 2 or 3 overlapping showers that occurred during that period. Additional fluctuations occurred later in association with small showers at about 1500 CDT and 1525 CDT.

Do the concentration data suggest that tracer Li was deposited in this series of samples? The magnitude of the concentration in the first sample suggests that it may possibly include some tracer, and indeed the tracer release was going on during the same time. The time-pattern of concentration and rainfall rate variations is very similar to that seen in non-tracer rains and for other materials, so this is not evidence for the presence of tracer Li.

The bottom section of Fig. 70 shows time variations of the K/Li ratio in the soluble portion of the rain. The smallest ratio (20) occurred in the first sample. By comparison with the ratio of K/Li in the earth's crust (400), this indicates an excess of Li over that expected from soil if K is an index of the soil contribution. Figure 70 further shows that the K/Li ratio was less than the earth's crust value during most of the rain. Examination of the ratios (not shown) of Li and a number of other at least partially soil-derived elements (Ca, Na, Mg) gave the same result. Thus, of the three indicators, both the magnitude of Li concentration and the magnitude of the K/Li ratio suggest the presence of tracer, while the concentration pattern appears normal. This evidence, together with the timing of the

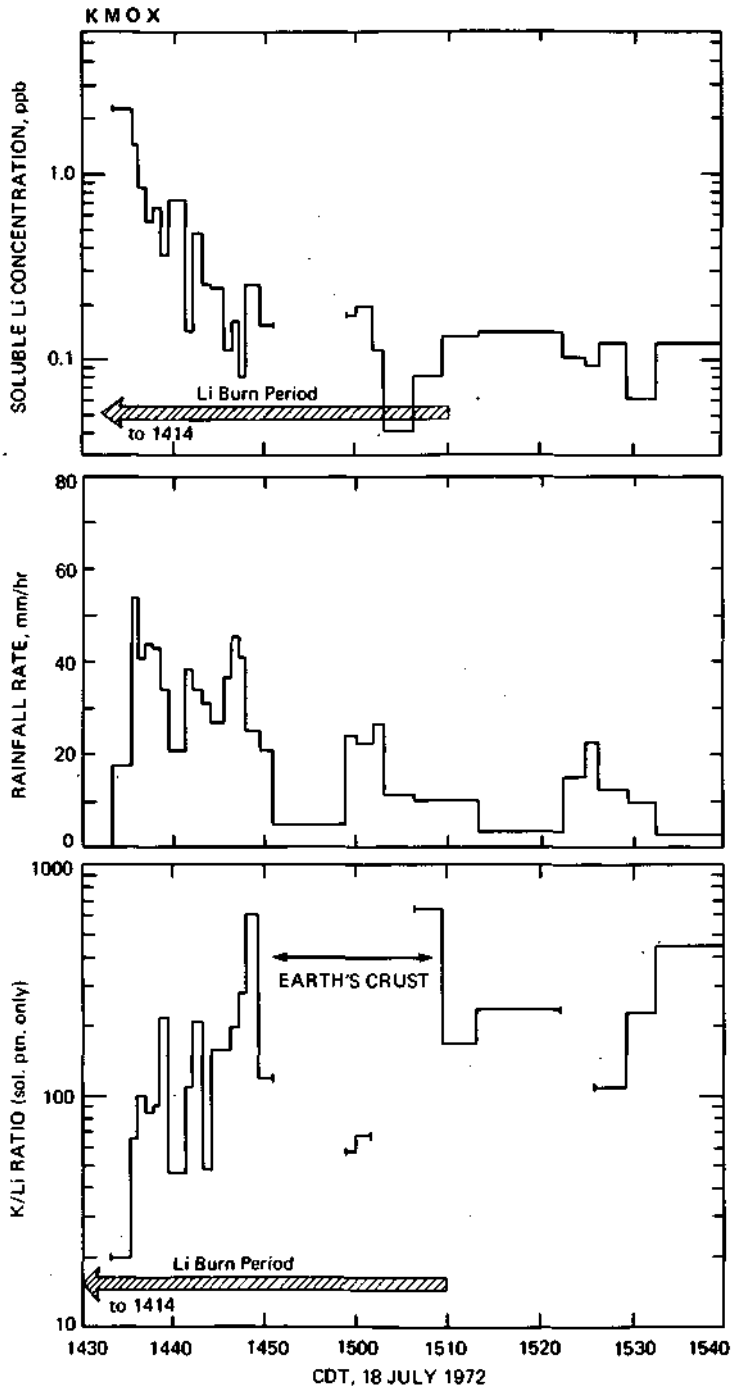


Figure 70. Variation of soluble Li concentration, rainfall rate, and K/Li ratio at KMOX during rain of 18 July 1972.

release, strongly suggests that tracer Li fell at this station. The K/Li ratio pattern suggests that the deposition occurred largely as a series of bursts at the onset of each of several showers.

Another interesting question, although not related to the identification of tracer Li, concerns the cause of the observed variations in the K/Li ratio and their relationship to the rainfall rate, Fig. 70 shows a pattern of low ratios early in a shower and high ratios during or following the heaviest rain, in each of the 5 showers. This suggests that soluble Li is removed from air preferentially over soluble K at the leading edge, or during the early portion of the rain showers. Although no particular explanation for such occurrences is offered now, the systematic variation in chemical parameters with rainfall rate offers the hope of elucidating the complex interactions between scavenging and precipitation-formation processes in convective showers. Both Engelmann (1971) and Gatz and Dingle (1971) have previously suggested mechanisms to explain concentration variations during convective rains.

The two remaining sequential series were collected at KMOX and Centreville on 3 August 1972 (see Fig. 45). Tracer Li was released on this day between 1526 CDT and 1536 CDT.

At KMOX, rain began at 165 3 CDT, more than 1 hr after the Li release, and ended sometime after 1715 CDT. The total rainfall in the sampler was 15.0 mm, and produced 24 samples. Concentration, rainfall rate, and ratio data are shown in Fig. 71.

The soluble Li concentration followed the familiar pattern in its time variations with rainfall rate, with one exception. The concentration dropped sharply while rainfall rate increased during the first shower (about 1705 CDT). It recovered somewhat at the end of the first shower, only to drop off again during the second, multi-peaked shower (1710-1715 CDT). The exception to the usual pattern was the sudden and brief (1-sample) increase in Li concentration that occurred at the end of the second shower.

The magnitude of the observed concentrations, even in the anomalous sample, is that of natural background Li. The concentration pattern is normal, except for the single anomalous sample. If real, this anomaly in the concentration-time pattern definitely suggests the presence of tracer Li. The fact that the high concentration was seen in only one sample,

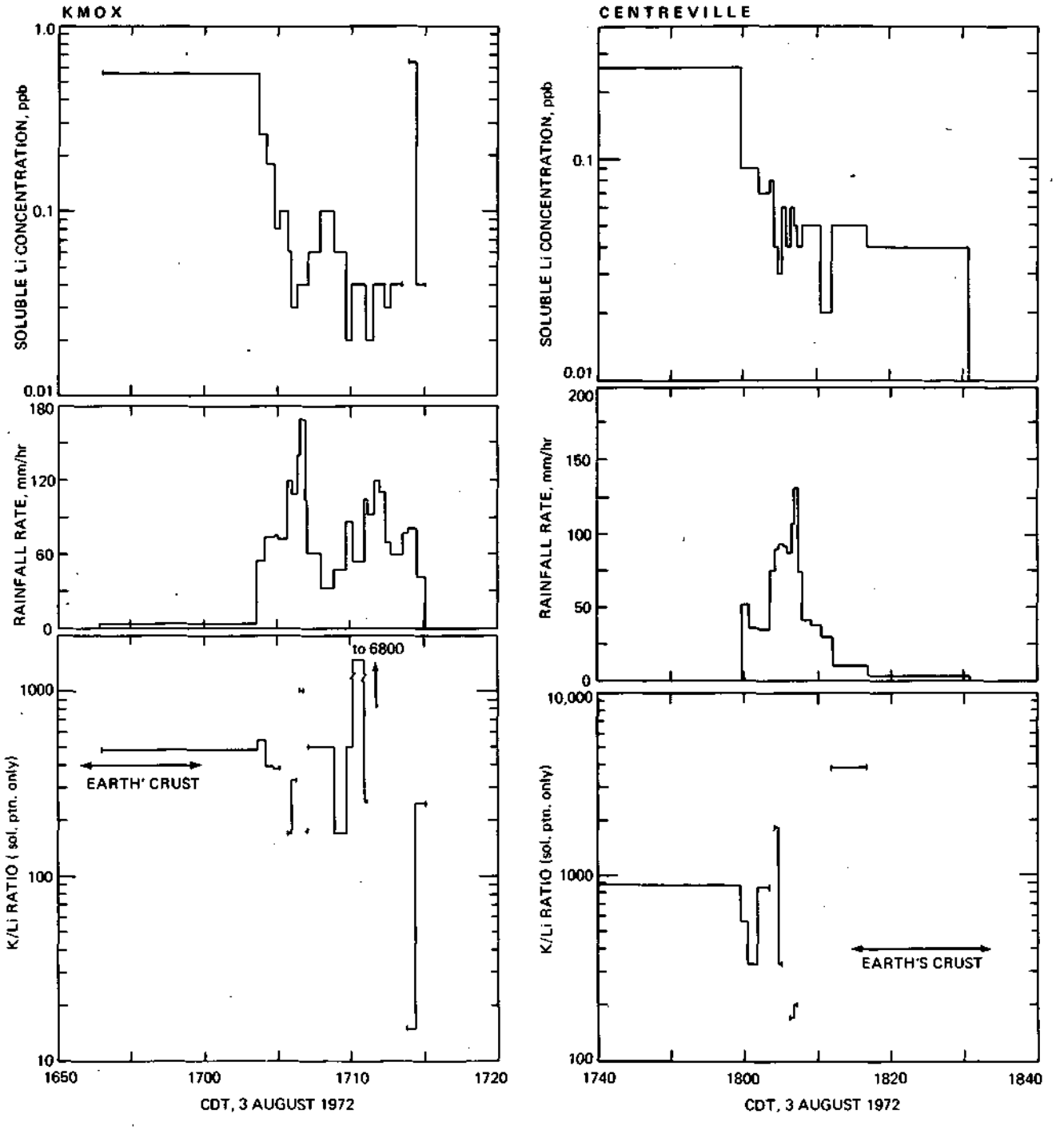


Figure 71. Variation of soluble Li concentration, rainfall rate, and K/Li ratio at KMOX and Centreville during rain of 3 August 1972.

however, leads to the suspicion of error or contamination during sample-handling or sample analysis.

Information provided by K/Li ratios also leads, with the exception of the same anomalous sample, to the conclusion that tracer Li was not deposited at KMOX in this rain. Again, if the Li concentration of the anomalous sample is correct, then the low K/Li ratio is also correct, and suggests tracer Li in that sample.

It is worth noting that the pattern of low K/Li ratios early in an individual shower, followed by higher ratios during or following the heavy rain, was repeated here with natural Li during both main showers.

The 3 August rainfall at the Centreville station began at 15 36 CDT ■ as the Li release ended, and ended at 1839 CDT. The total rainfall in the sampler was 15.4 mm and produced 19 samples. Results from one hr of this period containing the only heavy shower, are shown in Fig. 71, using the same format as before. Both the magnitude and the time pattern of the soluble Li concentration are normal. Furthermore, the magnitude of the K/Li ratios suggest that only natural Li was present. Again, some interesting variations in the K/Li ratio occurred in relation to the rainfall rate, but because of missing ratio data (attributable to some less-than-detectable K concentrations) the exact pattern is not clear.

The first two samples in this series (not shown in Fig. 71) were collected soon after tracer release. No evidence for the presence of Li tracer in this sample was found, either in their Li concentrations, or in their K/Li ratios.

7.0 SUMMARY OF PROGRESS

The identification of the sources and sinks of pollutants in an urban-industrial region is essential for the complete understanding of the impact of man's activities on his environment. The industrial and urban wastes emitted to the atmosphere are of particular importance in the study of the effects of the urban-induced alterations of the atmospheric properties conducive to the formation and continued production of precipitation. The tracer chemical studies are directed toward learning about the interaction of simulated urban effluents and cloud and precipitation processes, as well as providing an insight into the internal motions of convective storms.

The atmospheric particulates enter the cloud and precipitation processes through two means: 1) directly, by condensing water vapor and forming cloud droplets which become involved in the precipitation, and 2) indirectly, through in-cloud and sub-cloud scavenging (or removal) processes. Tracers are released which are of both forms, that is, a substance which will form cloud droplets, and a substance which is dry and will be removed by capture mechanisms only. The former is a lithium solution emitted to the atmosphere by a generator similar to those used for cloud seeding, and the latter is indium which is released from flares.

The tracer chemicals were released in 1972 from ground level and from aircraft. The ground level releases were used to simulate the emissions of materials from sources such as automobiles and small industries, whereas the aircraft releases were conducted in the area of the updraft of convective storms to examine the removal process of precipitating clouds.

During 1972, 45 experiments were conducted in the St. Louis area. Of these, 6 were ground releases of lithium and 5 were airborne releases of both lithium and indium. The remaining 34 experiments were designed to obtain background data on the pre-existence of the tracers in natural rainfalls and in the dry fallout which normally occurs in the area.

The 45 experiments, which consisted of time periods varying from a few hours to a few days, yielded a total of 3637 samples collected in the network of 700 square miles downwind of the city of St. Louis. The network contains 80 samplers which were serviced by 5 trained personnel. Of the total samples 1745 were rainfall and the remaining 1892 were dry fallout.

The addition of the pibal and radiosonde operations in support of the tracer experiments permit detailed study of the treated storms, such as the interaction between the mesoscale storm system and the synoptic scale flow. The low-level flux of vapor into the storm is amenable to calculation using the upper-air and aircraft observations. Such estimates of the precipitation efficiency will assist in the determination of the scavenging efficiency as a function of the maturity of the storm.

Preliminary results from the analysis of these data continue to show interesting and unexpected results. Tracer material injected into a cloud system will be found removed by neighboring clouds and in short periods of time, indicating very efficient removal mechanisms. The ground-released

material is found to be incorporated into cloud systems with relative ease and removed by rain processes in an efficient manner.

The final results from this aspect of the research will not be known until the many years of field effort are completed because of the complexities of the complete chemical analysis of the samples and the detailed analyses of the surrounding meteorological conditions. These results will be of great value in determining the ability of the atmosphere to remove man-made materials and maintain an equilibrium value of pollutant particulates.

8.0 LABORATORY STUDIES

8.1 Introduction

The scavenging efficiency of raindrops for micron size particles has been measured in the laboratory as well as in the field. The laboratory measurements were conducted in a 10 m tall drop acceleration tower attached to a 2 m tall aerosol chamber. The chamber was filled with a monodisperse aerosol and the size and concentration of the particles were determined. Raindrop size water drops were produced at the top of the tower, and reached terminal velocity prior to passage through the aerosol chamber. The drops were analyzed and the amount of aerosol material in each was determined. The scavenging efficiency was calculated as the percentage of the aerosol removed from the volume through which the drop passed. A comprehensive discussion of this experiment can be found in the symposium proceedings entitled "Precipitation Scavenging (1970)" (see footnote on page 1).

8.2 Aerosol Generation Studies

8.2.1 Biological Aerosol

Generation of a known size of monodisperse aerosol in the 0.1 to 1 μm (all measurements are equivalent diameter) size range is difficult and requires continuous monitoring of the aerosol to determine the size and distribution, concentration, and coagulation rate of the particles. The previously reported scavenging efficiency measurements used a dry Bacillus subtilis spore aerosol.

An aqueous suspension of the spores was atomized into droplets which were subsequently evaporated to leave a dry spore aerosol. Monodispersity was insured if the spores were all of equal size and the concentration of spores in the suspension allowed no more than one spore in each atomized droplet. Detection of the spores in the scavenger drop was relatively simple even though their concentration was small. The drops were captured on agar coated petri dishes after passage through the aerosol chamber. The dishes were incubated and the individual spores in the drops developed into colonies of cells which could be counted optically after 24 hours.

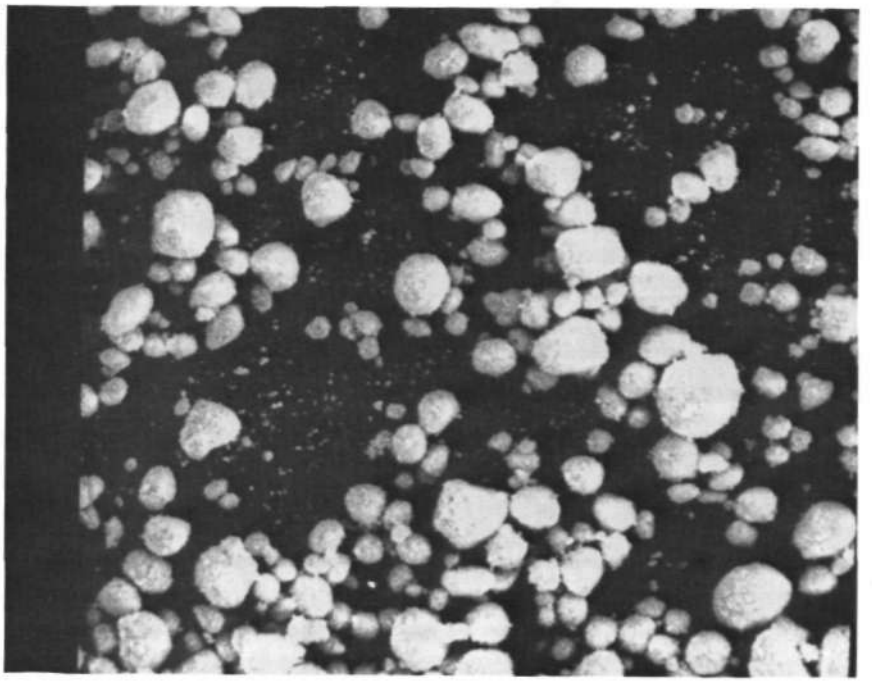
The ease of production of a monodisperse aerosol and the simple detection of minute concentrations of the aerosol in the drops makes this method very attractive for the current activity. However, after an examination of the biological spores and viruses available commercially, none were found which were smaller than 1.0 urn, viable as an aerosol, and nonpathogenic. Consequently, the search for other materials suitable for aerosol production led to the study of various chemical compounds.

8.2.2 Chemical Aerosol Production

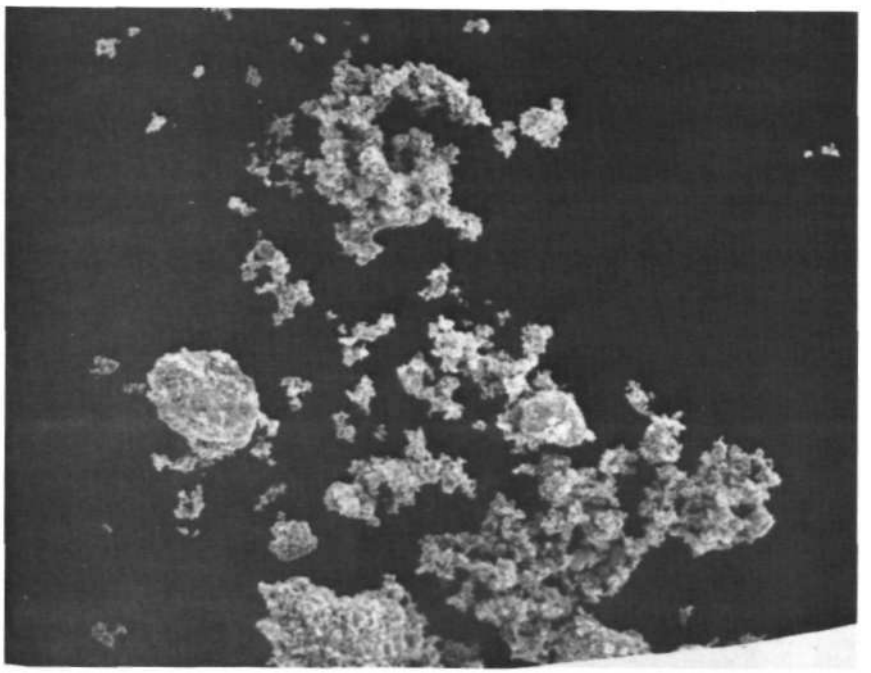
Chemical powders are available in the size range of interest*. An iron oxide powder with a mean diameter of 0.5 μm and a zinc oxide with a mean diameter of 0.66 μm were obtained for testing. The iron is used commercially for magnetic tape coatings and the zinc is used as an electro-luminescent surface for cathode ray tubes. These chemicals were chosen because they could be detected in water at low concentrations using flameless atomic absorption spectroscopy (FAA).

Electron microscope photographs of the powders are shown in Fig. 72. Both the iron and zinc powders were clumped into clusters of approximately 50 μm. Several attempts were made to produce submicron particle dispersions in water, and the most successful of these was to place a sample into an ultrasonic bath. Excitation of the sample dispersed the particles, but

* Duke Standards Co., Palo Alto, California.



IRON OXIDE (x200)



ZINC OXIDE (x200)

Figure 72. Commercially available iron oxide (top) and zinc oxide (bottom) tested as an aerosol for laboratory experiments. Magnification was 200X.

clumping occurred extremely rapidly when the excitation was stopped. The large spread in particle sizes (the standard deviation was 0.25 μm) and their irregular shape led to the rapid agglomeration which prohibited further use of these particles.

8.2.2.1 Ultrasonic Nebulization

An ultrasonic nebulizer has been loaned to the Cloud Physics Laboratory for evaluation*. This device is shown schematically in Fig. 73. It consists of a radio frequency power source and an atomization cell containing a piezoelectric element. A thin sheet of a chemical solution or suspension to be atomized is passed over the piezoelectric transducer. When the transducer is activated by the power source, the surface oscillations disrupt the fluid film creating an aerosol of small droplets. The droplets are removed by a carrier gas flowing through the cell. The output section of the cell contains a five stage impactor which removes droplets larger than 30 μm which are produced randomly by the violent agitation of the fluid. Aerosol particles are produced at a rate of approximately 10^7 per second.

For a monodisperse droplet generator, the dry aerosol size distribution is proportional to the concentration of salt in the solution. Likewise, for a given concentration, the aerosol size distribution is proportional to the droplet size distribution produced by the nebulization.

Aqueous solutions of various salts have been atomized by the nebulizer. If the concentration of the salt is kept below 1.0% by weight, the droplets produced have a size distribution similar to that of Fig. 74 for $CuSO_4$. This distribution is more broad than would be expected from the promotional material describing the device. Higher concentrations quench the action of the transducer and no aerosol is generated. The aerosol was passed into a chamber and dried by dilution with dry air. The resulting particle size distribution is shown in Fig. 75 which are similar to those obtained by Matteson and Stober (1967) using electrolyte solutions in an air blast nebulizer.

* A Tomorrow Enterprises, Portsmouth, Ohio ultrasonic nebulizer on loan from the Materials Research Spectroscopy Laboratory, University of Illinois.

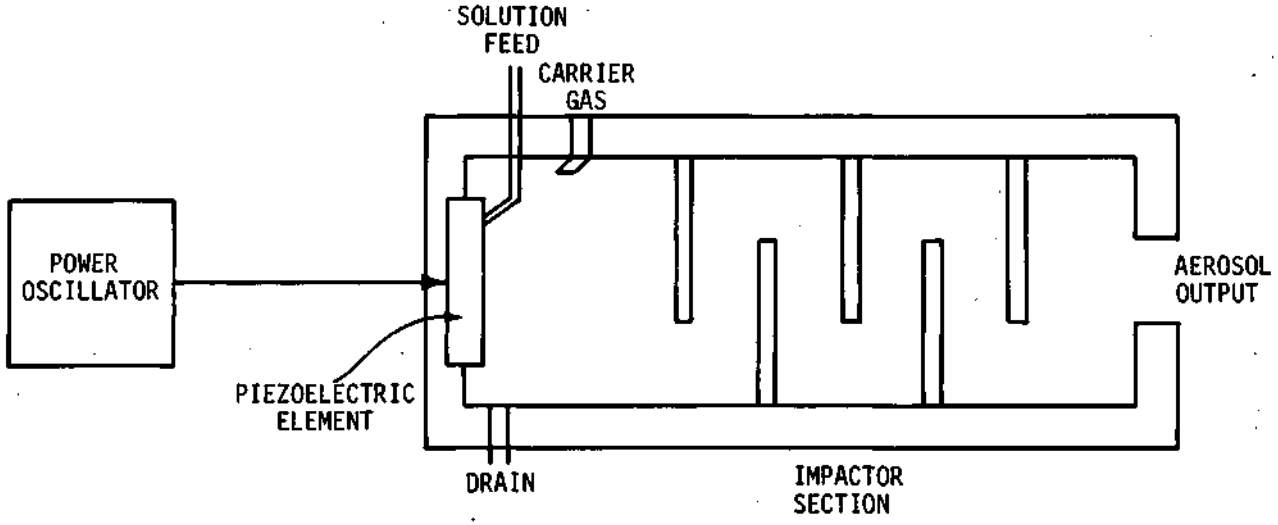


Figure 73. Schematic diagram of the ultrasonic nebulizer aerosol generator.

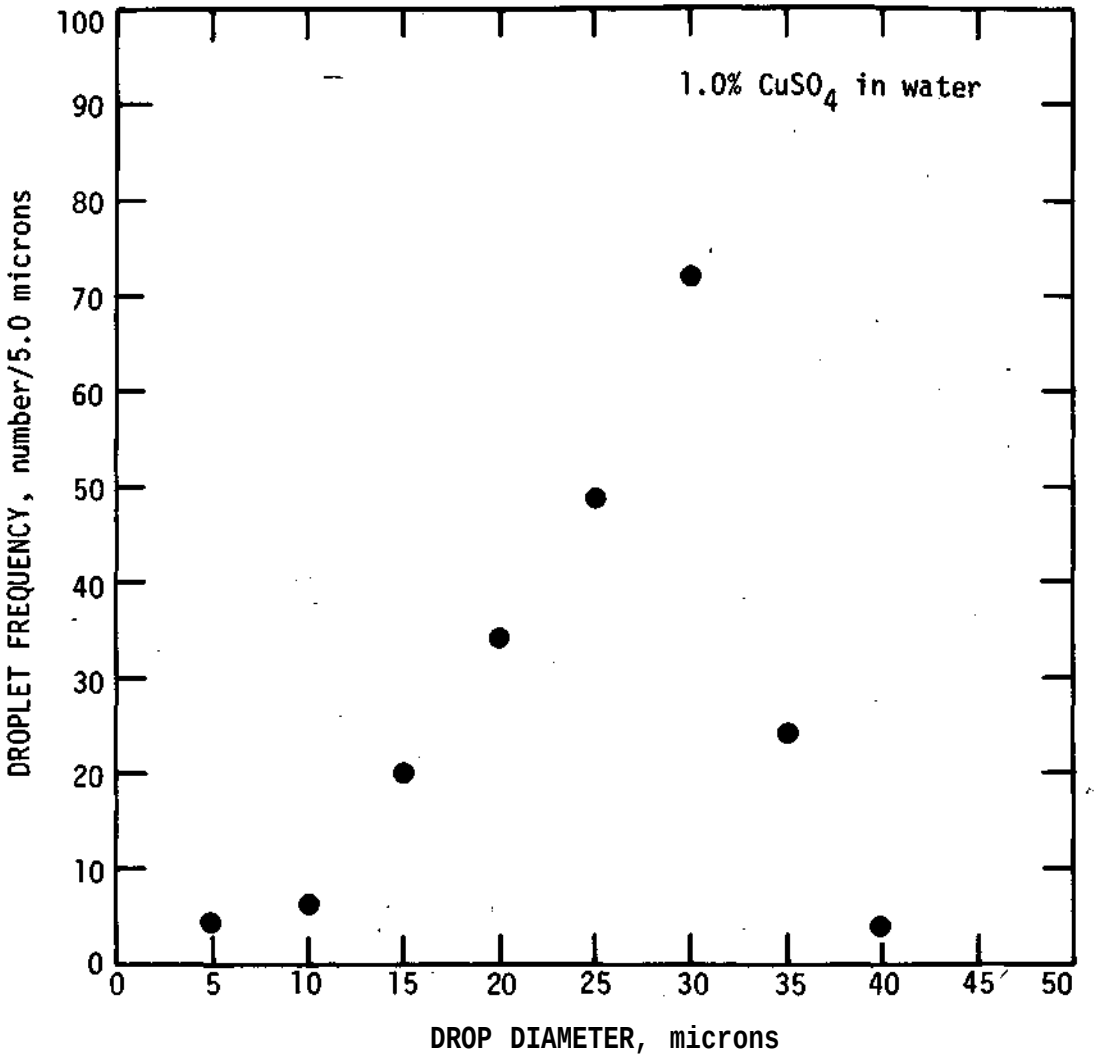


Figure 74. Frequency diagram of droplet distribution obtained from the ultrasonic nebulizer.

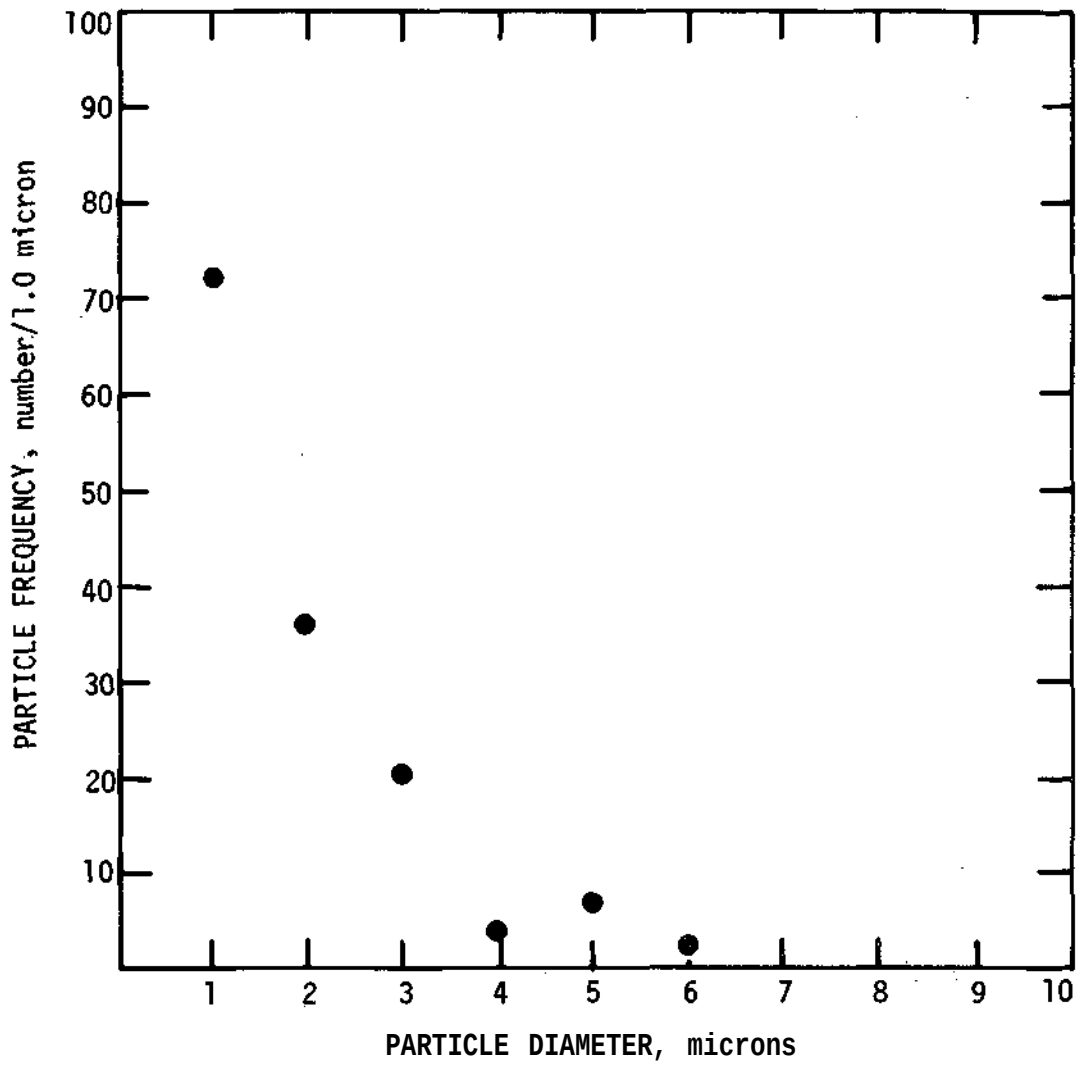


Figure 75. Aerosol particle size distribution resulting from the evaporation of the droplets shown in Fig. 74.

The size of the droplets and particles were measured using an optical microscope. An oil-coated slide (to inhibit evaporation) was inserted into the aerosol chamber and allowed to collect the droplets or particles. A representative area near the center of the slide was examined under the microscope and approximately 200 particles were counted. The water droplets were relatively large and easily measured on the oil film. The dry aerosol particles were easily recognized because of their distinctive crystaline shape, but measurement of their exact size was difficult.

8.3 Near-Forward Laser Scattering Particle Detection

Dow latex spheres were used as an aerosol in the initial stages of this scavenging research. They are available in almost any size in the 0.1 to 1.0 ym range. An aerosol of the spheres can easily be generated using an air blast atomizer or the ultrasonic nebulizer described earlier. The detection of minute quantities of these spheres in water is difficult and require electron microscopy or Coulter Counter techniques. Though these are available they are complicated and time consuming for the determination of small concentrations of particles.

Optical detection of the particles is possible only in the cleanest conditions and then only for the 1.0 µm or larger spheres. Another approach, however, is to use a standard microscope and illuminate the sample with coherent laser light. The intensity of the light scattered by the particles is great at small angles with respect to the beam as was measured by Phillips et al. , (1970). The apparatus shown in Fig. 76 was tested as a method of detecting individual spheres. When light was scattered forward at less than 20 degrees, the intensity of the light tends to increase with increasing particle size. While this method is not used to size individual spheres, multiple clumps of spheres can be distinguished from individuals. Using this technique, the number of 0.22 µm spheres have been counted readily.

8.4 Summary of Progress

The search for suitable aerosols is continuing. Chemical salt aerosols are the most attractive because such properties as wettability,

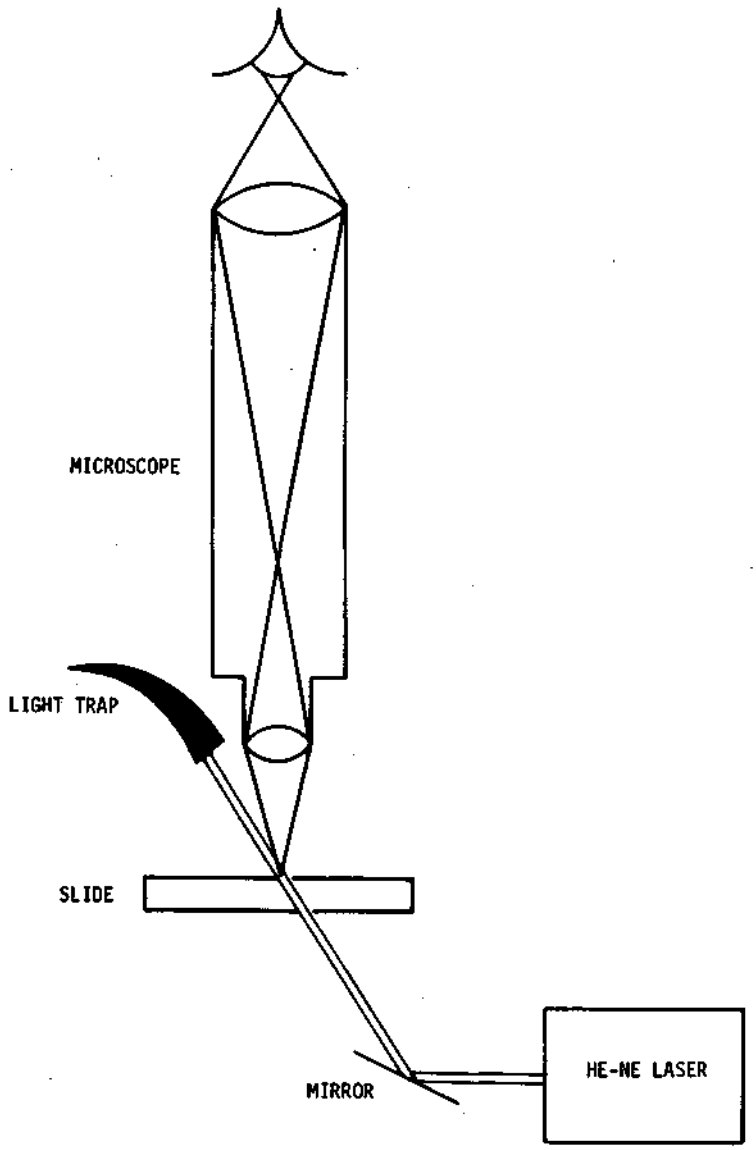


Figure 76. Laboratory setup for particle counting by laser beam scattering by aerosols.

effect on surface tension of the drop, adhesion to the drop surface, and the shape of the crystals in the aerosol can be controlled and varied. The objections to the use of these aerosols are: 1) the analytical detection limits of the chemical species in individual scavenger drops may be too high for sensitive measurements; and 2) the generation of dense, monodisperse carrier aerosols is difficult to achieve.

The atomization of insoluble particles, including latex spheres, has been reexamined in light of the forward scattering counting technique. These results seem worth pursuing and scavenging efficiency measurements are now under way to verify its applicability.

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APPENDIX A
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