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

STUDY OF RAINOUT OF RADIOACTIVITY IN ILLINOIS

Tenth Progress Report

Contract Number AT(11-1)-1199
November 1971

Sponsored by

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

Richard G. Semonin
Principal Investigator

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ABSTRACT

The field project phase (ITREX) of the contract research during 1971 was continued in the St. Louis area as an integral part of Project METROMEX, a field effort to determine the effects of an urban area on precipitation. Unique chemical tracers released into the updrafts of convective clouds were used to estimate the scavenging efficiency of the treated clouds.

This report describes the design of ITREX as it relates to METROMEX, and presents a few preliminary findings determined from the data collected during the field project period.

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PROJECT ITREX FIELD STUDY

Introduction

Contract research during 1971 has mainly involved extensive preparation and execution of Project METROMEX, a field project in the St. Louis area, designed to investigate precipitation scavenging of sub-micron particles as well as to determine effects of an urban area on precipitation processes and amounts. The current field work is a continuation of Project ITREX which was carried out during the previous two years in east central Illinois.

This report summarizes the project design of METROMEX and presents some preliminary findings based upon the large quantity of data collected during the field project period June-August 1971. Since the field work was only recently concluded, much of the data analysis is yet to be accomplished.

A total of four storms were treated with lithium injected into the rain systems by a Piper Navajo aircraft utilizing acetone generators as the tracer delivery mechanism. Three additional burns were made from the ground into a local industrial smoke stack also using an acetone generator. Considerable background data were collected to determine the occurrence of various elements, including lithium, in rainwater and in dry deposition. A summary of the data collected is presented in Table 1.

Project Description

Project METROMEX is a cooperative scientific effort designed to investigate the inadvertent effect on weather caused by an urban-industrial complex. The site chosen for this study was the St. Louis urban area, with the emphasis being on the scavenging of sub-micron particles by the precipitation process, the investigation of urban related changes in precipitation processes, and quantitative changes in surface precipitation. Project ITREX participated in this vital research through the use of tracer chemical releases into convective storms.

Scientists from seven groups worked together in the 1971 METROMEX field operation; these groups were: the Argonne National Laboratory; the University of Chicago; the University of Wyoming; the Battelle Northwest Laboratories; the Stanford Research Institute; the University of California at San Diego; and the Illinois State Water Survey.

Two funding organizations are sponsoring the Water Survey's portion of this research effort. The National Science Foundation is contributing funds toward the following goals: 1) to study the effects of urban environments upon the frequency, amount, intensity and duration of precipitation and related severe weather; 2) to identify the physical processes of the atmosphere, and the factors of the urban complex, which are the causative agents of the observed weather effects; and 3) to assess the impact of urban-induced inadvertent weather changes upon society.

The AEC contract is funding research to investigate the precipitation scavenging process through field measurement of tracer chemicals introduced by aircraft into storm systems, to evaluate their motion within the storms, and to carry out laboratory experiments on raindrop collection efficiences of sub-micron aerosols (Changnon, et al., 1971). Since all of the personnel involved in this present contract have devoted a considerable portion of their time during the past year in the preparation and execution of METROMEX, little progress has been made regarding further laboratory experimentation. However, three papers have appeared in various scientific journals during this period summarizing previous experimental results (see Appendices

A, B and C). The aircraft measurement of temperature distribution at low flight levels across the St. Louis urban area is also being accomplished to determine the influence of the city upon the thermodynamic structure of the atmosphere.

Weather During Field Operations

The first five days of June were dominated by a long wave trough along the Pacific coast with an associated cut-off low at 500 mb, and a weak ridge centered in the southern midwest. This resulted in westerly flow across the midwest. A short wave trough aloft along with a surface front passed through the midwest 01-02 June resulting in widespread precipitation in the area; approximately 0.3 inch of rain fell in the STL (St. Louis) region during this period.

This general pattern continued for the following nine days with short wave troughs and associated surface frontal systems passing through the midwest on 7, 9-10, and 13 June; precipitation occurred in the STL area on 7, 10, 11 and 13 June totaling approximately 1.8 inches. The cut-off low off the Pacific coast gradually migrated northeastward into north-central Canada during the ensuing six days, along with a gradual dissipation of the Pacific long wave trough. Indeterminate, weak flow existed over the midwest during this period; a short wave trough passed through the midwest on 15-16 June causing light precipitation in the midwest.

Another long wave trough with an associated cut-off low was established along the Pacific coast during the next six days; a weak ridge persisted in the southwest during this period. This resulted in northwesterly flow in the midwest with approximately 0.2 inch of precipitation occurring in the STL area during this 6 day sequence. The following four days saw a gradual migration eastward of the upper ridge in the southwest and a continuation of the trough off the Pacific coast.

A short wave trough passed through the midwest on 30 June - 01 July along with an associated surface front bringing moderate rainfall to the midwest.

Weak zonal flow existed over the area for the next 9 days as the Pacific trough weakened; however, a vigorous minor trough, coupled with a fast moving cold front brought moderate to heavy precipitation (1 inch in STL) to the midwest 04-05 July. Another minor trough with a slow moving cold front that became stationary through the midwest also brought heavy amounts of rain (1 inch in STL) during 10-11 July. Intensification of a ridge in the southwest coupled with the development of a long wave trough in the eastern 1/3 of the U.S. brought relatively strong NW flow aloft into the midwest for the following 10 days. Two major outbreaks of Canadian air during this period triggered moderate precipitation amounts on 13, 14, and 18 July, totaling approximately 1.5 inch in the STL area.

The above pattern persisted for the next 12 days into the first 3 days of August. Four major outbreaks of cold Canadian air during this period triggered frontal precipitation on 23, 28, 29 July and 03 August totaling approximately 1.25 inch. The long wave trough moved slowly eastward during the next 3 days and a strong, very slow moving surface ridge over the midwest with a supporting ridge aloft dominated the weather in the region. No significant amount of precipitation fell during this period in the area of interest. The ridge slowly broke down during the next 5 days as another long wave trough developed in the midwest resulting in a fresh outbreak of Canadian air into the midwest with light frontal precipitation of 0.03 inch occurring on 10 August.

The next 7 days saw a redevelopment of a weak ridge in the western portion of the country. A short wave trough associated with an east-west oriented stationary front through the middle of the country caused moderate precipitation in the STL area on 14 August. Zonal flow existed for the ensuing two days with light to moderate rainfall occurring in the STL region on 21 August as the axis of a long wave trough passed through the midwest. A moderately strong ridge in the west dominated the weather pattern for the remainder of the month with no significant amounts of precipitation falling in the midwest during this time.

Rainfall amounts ranged from 57% of normal in the northwest portion of the research circle to 23% of normal in the southeast during the month of June. Comparable figures for July ranged from 142% to 86%, and for August, 3% to 7%. The aircraft tracer operations were carried out during the latter part of July and August which unfortunately was an extremely dry period.

Radar Instrumentation

Two radar systems at the field operations headquarters at Pere Marquette (PMQ) in Grafton, Illinois, are being employed for METROMEX. An FPS-18 10-cm radar with PPI display is used to determine development regions of precipitation echoes as well as to deduce their movement with respect to the study area. A TPS-10 3-cm vertically scanning radar is also being utilized; this RHI display is used for the determination of the vertical development of individual storms within the research area. The time rate of change of storm tops is observed with this radar, providing the necessary information to decide whether or not to inject a particular rain cell with tracer material. Both radars are used to direct the aircraft toward cells of interest.

Surface Measurement Instrumentation

The research area in St. Louis (Fig. 1) is instrumented with 220 weighing-bucket recording raingages and a hailpad at each location, the spacing between gage locations being approximately 3 miles. The circular network design was selected after careful study and is considered optimal for evaluation of rain patterns.

Within this network is a smaller one consisting of 59 rainwater collection baskets (Fig. 1) with each one of these located at a raingage site. The samples collected from these sites will be used to determine background wet and dry deposition patterns in the area, as well as obtaining water samples on days when tracer material is burned into cells passing over the water collection network.

Included also in the research circle are two raindrop spectrometers, three thunder detectors, seven hygrothermographs, three time-lapse cloud cameras, six wind measuring sets and one electric field device. The drop spectrometers are the Joss variety (1967) and transform the mechanical momentum of a raindrop into an electrical impulse as the drop impinges on a circular area 10 cm in diameter. The impulses are recorded on magnetic tape for subsequent analyses as are the thunder detector data. The distribution of the various instrumentation is shown in Fig. 1.

A Piper Navajo aircraft was utilized during the final phase of the field work. The aircraft was used to inject tracer material into appropriate rain cells, and also to make measurements of temperature, condensation and ice nuclei, and moisture content as it made traverses across the St. Louis area.

Instrumentation Operation and Data Acquisition

The 220 recording raingages, hailpads, wind sets and hygrothermographs were maintained continuously by four field technicians from 10 June to 31 August 1971. Each gage was serviced weekly and the raingage charts as well as the hailpad foils were brought to PMQ for initial inspection before being sent to Champaign, Illinois for analysis. Half of the raingage network will continue to be operated after 31 August 1971; the full network will be reopened June 1972.

The FPS-18 radar was operated 24 hours per day, with a few short-duration exceptions, from 15 June to 29 August 1971. The PPI scope was photographed continuously in order to obtain a climatology of the echoes passing through the St. Louis urban area; this permits the determination of the urban influence upon rain cell morphology as echoes pass over the industrial complex. A signal integrator, which digitizes the return signal from the echoes, was operated when precipitation was occurring within, or approaching the research circle; the integrator data were recorded on magnetic tape.

The TPS-10 radar system was operated during the tracer portion of the field project when precipitation was within or approaching the research area. The TPS-10 scope was photographed when the above situation occurred. The remainder of the data collection is best described by going through the activity of a typical operational day.

Each morning between 0800 and 0900, the Radar-Operational Supervisor (ROS) analyzed the weather conditions. At 0930, a briefing was held and the ROS made a decision as to whether or not the day would be suitable for convective development and hence a possible aircraft tracer situation (GO day). Present at the briefing, in addition to Water Survey personnel, were members from the University of Wyoming and Battelle Northwest. The University of Chicago was notified by phone after the briefing as to the weather situation, and of activities planned for the day.

If a GO day was declared, the aircraft crew was alerted and told to prepare for a possible flight mission during the ensuing 10 hours. The four rainwater technicians were dispatched into the field and stood by until it was decided that rain was likely in the research area. When this occurred they were contacted by radio and told to put out their rainwater collector bags. After the bagging operation was completed, the baggers remained in the field awaiting further instructions from the ROS. The TPS-10 radar was turned on when cells came within range.

When precipitation over the water collection network was imminent, the aircraft was directed to the echo of interest and searched for an updraft in the cell; the acetone generators containing the tracer material were ignited when the cell was in an appropriate position relative to the network. When the precipitation from the treated cell left the network, the bags were picked up and returned to PMQ. The water samples were then prepared and sent to Champaign for chemical analysis.

On a tracer day then, with rain occurring in the water collection network, the following data were collected: 1) film data from the FPS-18 and TPS-10 radars; 2) integrator information from the FPS-18 radar; 3) raingage, wind and hygrothermograph data; 4) rainwater samples; 5) surface weather observations at PMQ; 6) raindrop spectra information; 7) thunder detector data; 8) meteorological measurements obtained by the aircraft and 9) cloud photographs from three sites. Teletype data, including radiosonde observations, as well as facsimile information, were also collected during the entire project.

During the first phase of the project, a number of background samples were obtained to determine the content of various elements in rainwater and in the air prior to the tracer phase (see Table 1). On several No GO days, the aircraft made traverses across the St. Louis area obtaining measurements of the particulate loading as well as measuring the standard meteorological parameters. A team from Stanford Research Institute (SRI) joined the METROMEX effort for the month of August. Their instrumentation consisted of a mobile lidar system, which was coordinated with the other phases of the project on a number of days to obtain vertical cross sections of the aerosol structure of the atmosphere in the research area.

Data Analysis

The emphasis of the AEC funded field effort is on the removal of aerosol by precipitation. In order to obtain a quantitative measure of scavenging, detailed analyses of individual rain cells are required.

The radar data provides a time and space description of the character of the precipitation. In particular, the stage of development of the storm under consideration may be deduced from the time-lapse photographs of the radar scopes as well as from the backscattered intensity signal recorded from the PPI radar. The RHI radar photographs are digitized using an analog-to-digital data reduction system and are then processed through a computer (Staggs and Lonnquist, 1970). The computer output is a plotted reconstruction of the three-dimensional echo as a function of intensity (Towery, et al. , 1970). The vertical growth rate of the storms under study is determined from the computer analyses and forms one of the more crucial measurements for the reconstruction of the storm. The flight track of the aircraft is then superimposed on the computer output to determine the exact relationship between the precipitation cell and the updraft as determined by the aircraft.

The PPI radar data is recorded on magnetic tape for post analysis by computer. The entire experimental research area is divided into small areas approximately one beam width wide and of adjustable length ranging from 0.3 miles to 2 miles. The intensity of the radar signal within each of the boxes, so defined, is integrated, digitized, and recorded. Computer software has been developed to yield total echo area as a function of intensity, echo movement, and deduced values of rainfall rate. Scope photography of the FPS-18 PPI facilitates the analysis of individual cases, and the photos, coupled with the RHI data, provide a complete time and space description of the cell morphology.

The aircraft data and surface meteorological network data are recorded on strip charts. All of these data are being reduced on the Water Survey's Autotrol chart reading device. Selected periods for intense study will be chosen to coincide with the release of tracer chemicals. The radiosonde data will be incorporated with the surface and aircraft measurements to provide a three-dimensional description of the atmosphere. These data will be used to: 1) depict the meteorological environment in which an individual experiment was undertaken; 2) provide the initial conditions for the numerical modeling of the case storms; and 3) provide the control data for model verification.

The drop-size data are recorded on magnetic tape in digitized form and are being transformed to a computer compatible tape for analysis. The output will produce size distributions and with further processing will yield estimated scavenging efficiencies. The drop-size spectra data will be analyzed in two ways. The total drop-size distributions obtained for the 5-year period upwind of the urban input area will be compared with those from the downwind regions to determine the presence of a climatological difference in the distributions. Secondly, individual storms will be analyzed for differences in spectra as they pass over the urban complex.

The results from Project ITREX 1970 have shown the efficacy of the atomic absorption analysis of the rainwater for the tracer chemicals. The method has the distinct advantage of producing results in a short period of time. An AAS system purchased under the current AEC grant is being utilized for the chemical analyses.

Preliminary Case Studies

A small portion of the data collected during METROMEX '71 has been analyzed; the following are some preliminary results for three operational days.

Wet no-burn case - 18 June. The synoptic situation on 18 June resulted in southerly flow into the midwest around a high pressure area in the southeast U.S. Dewpoints in the region were in the high 60's with surface temperatures reaching into the low 90's. Isolated to scattered airmass showers occurred in the area as a result. Figs. 1-7 show the isohyetal patterns of rainfall rates in the St. Louis urban area during a 96 minute period. The rainfall rates are four minute averages ending with the time indicated in the figure.

There are two main points of interest regarding these data. The initial rain cell (Fig. 1) centered around raingage #18 is located in the immediate vicinity of the Portage des Sioux power plant, one of the largest in the St. Louis area, located on the Mississippi River between gages #17 and 18. This cell became one of the major cells of the day. The second point is that the other two major precipitation areas developed and grew in and downwind of the urban area (Figs. 3-6). One originated in the vicinity of the Wood River Power Plant, and the other in a heavily industrialized area of East St. Louis. Since there were no other major developments elsewhere in the network, the above strongly suggests that the urban effect on precipitation was operating on this day.

Urban temperature profile - 05 August. 05 August was characterized by a large stationary Canadian high pressure region on the surface centered over northern Wisconsin, bringing northeasterly flow into the midwest. The atmosphere was sufficiently stable to eliminate the possibility of any convective development in the area of interest. The Piper Navajo aircraft flew an "X" pattern over the St. Louis

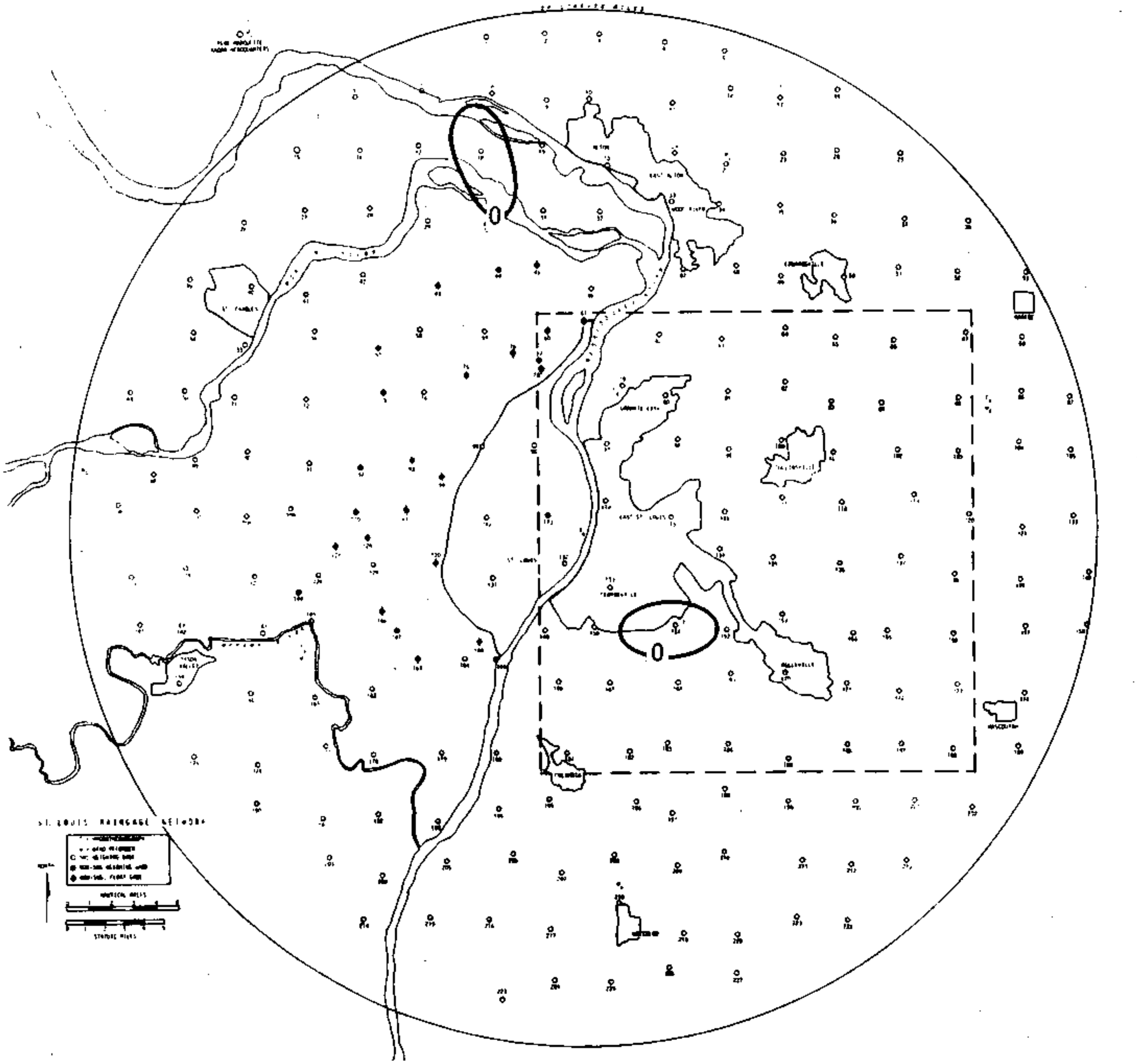

Figure 1. Four minute average rainfall rates in inches/hr for 18 June 1971 for the time ending 1220 CDT

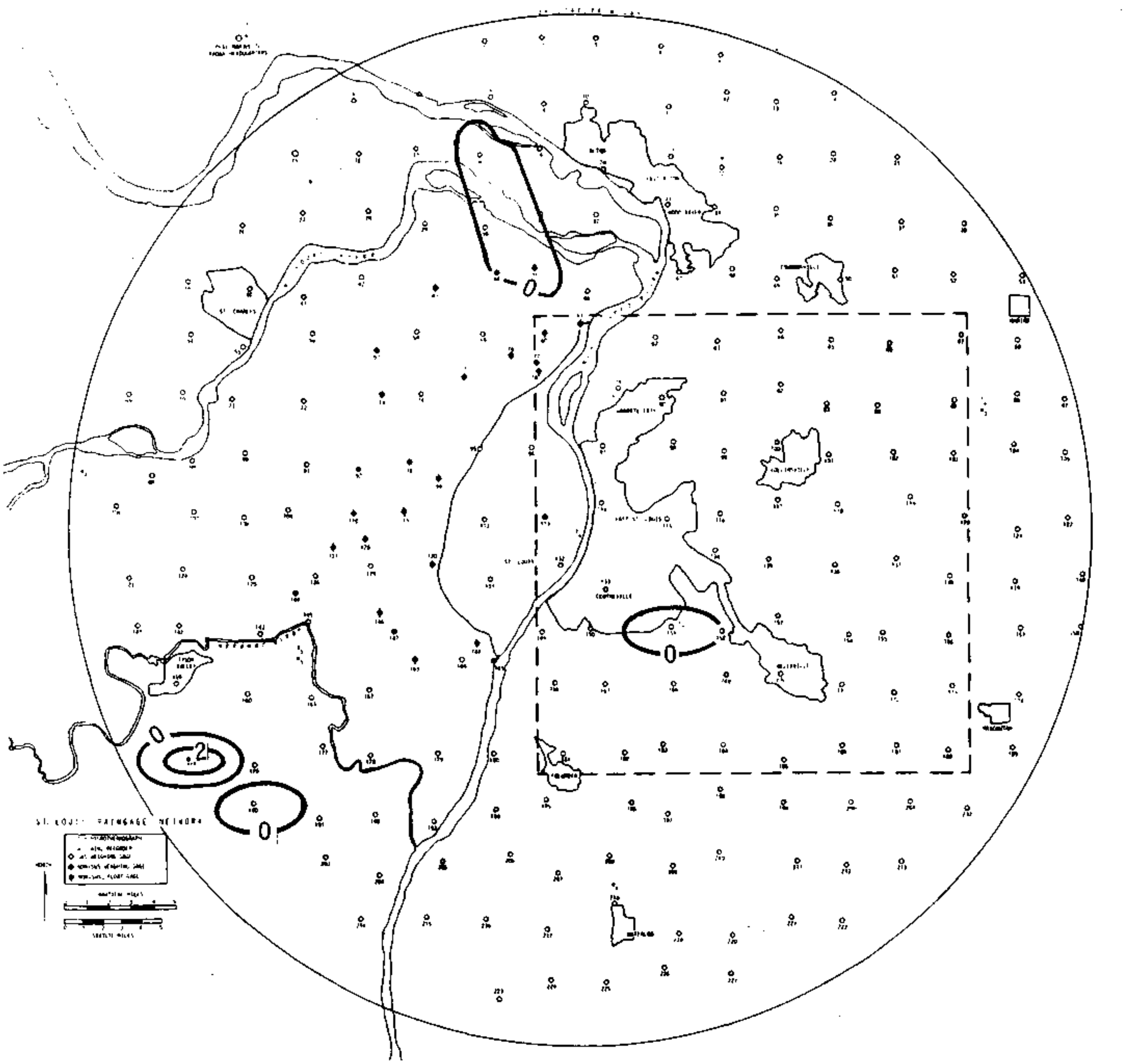

Figure 2. Four minute average rainfall rates in inches/hr for 18 June 1971 for the time ending 1236 CDT

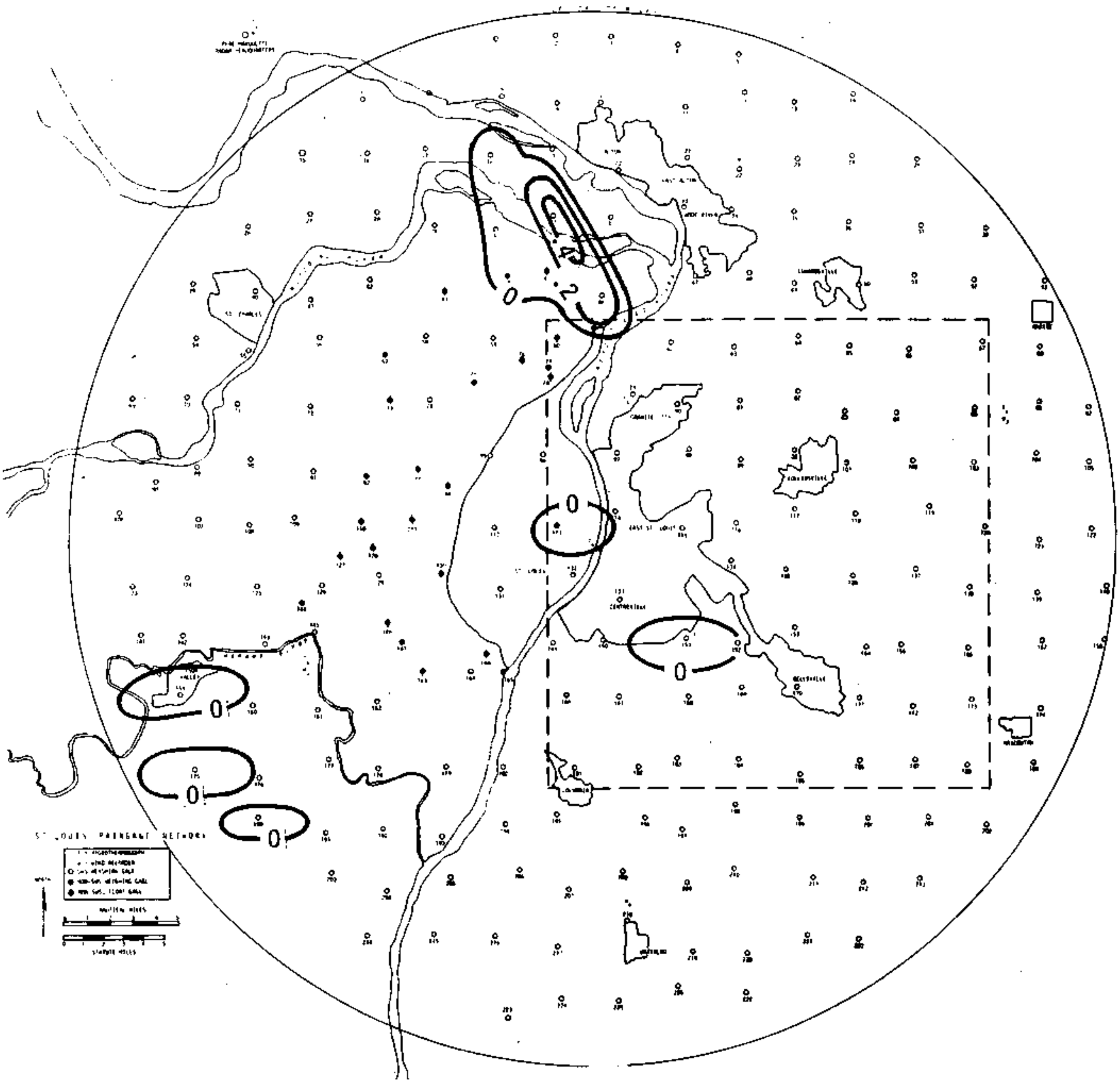

Figure 3. Four minute average rainfall rates in inches/hr for 18 June 1971 for the time ending 1252 CDT

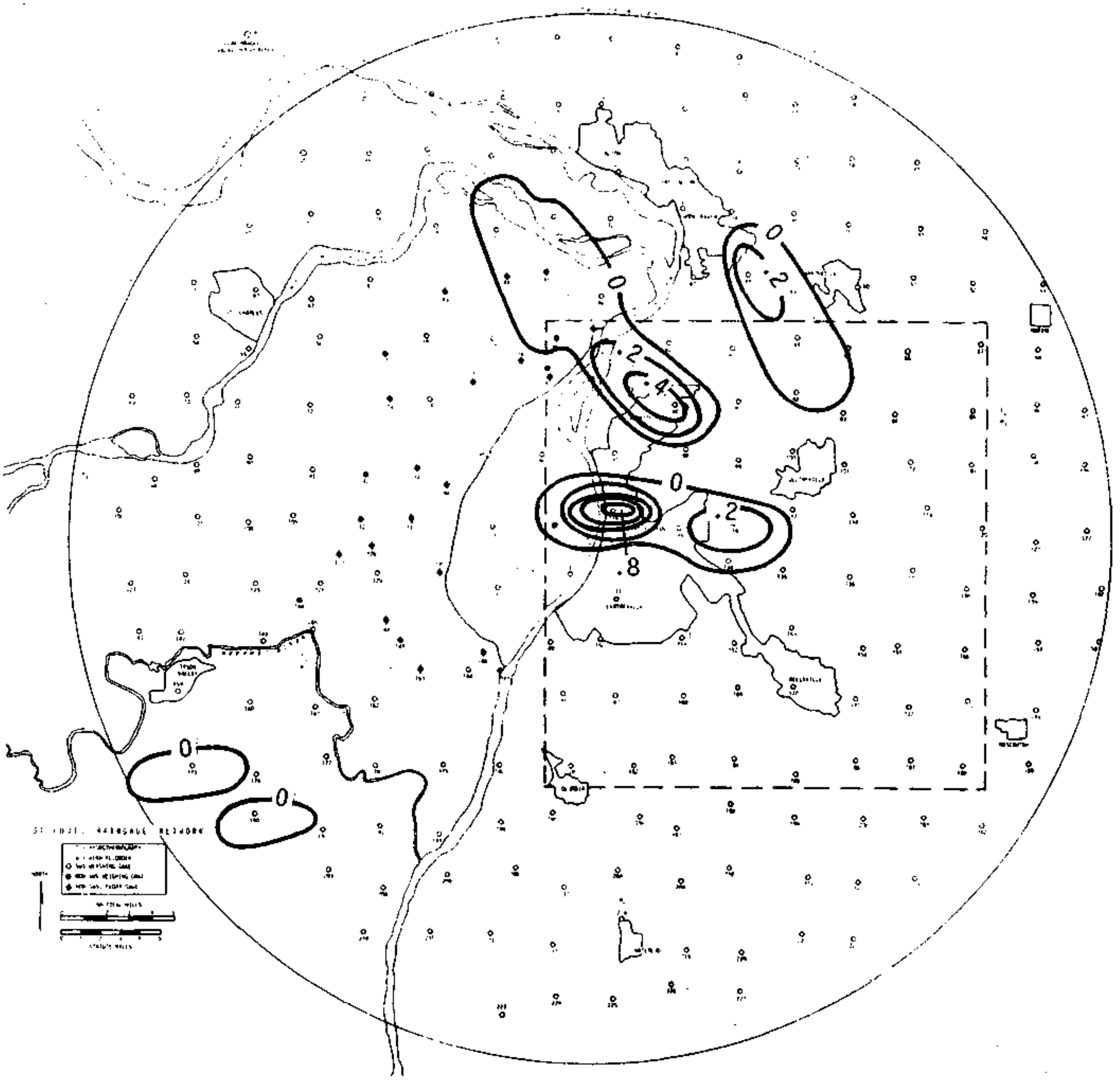

Figure 4. Four minute average rainfall rates in inches/hr for 18 June 1971 for the time ending 1308 CDT

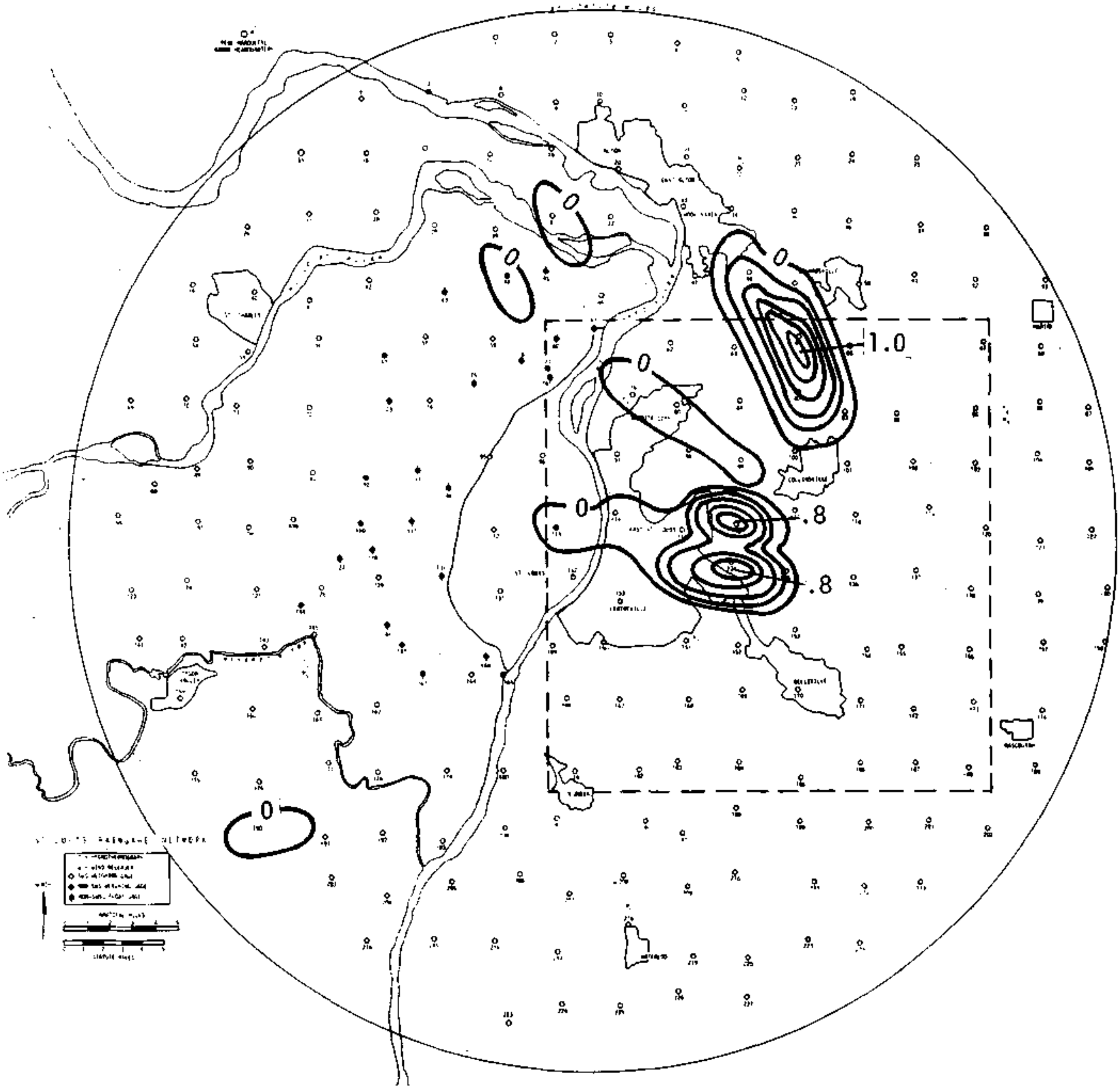

Figure 5. Four minute average rainfall rates in inches/hr for 18 June 1971 for the time ending 1324 CDT

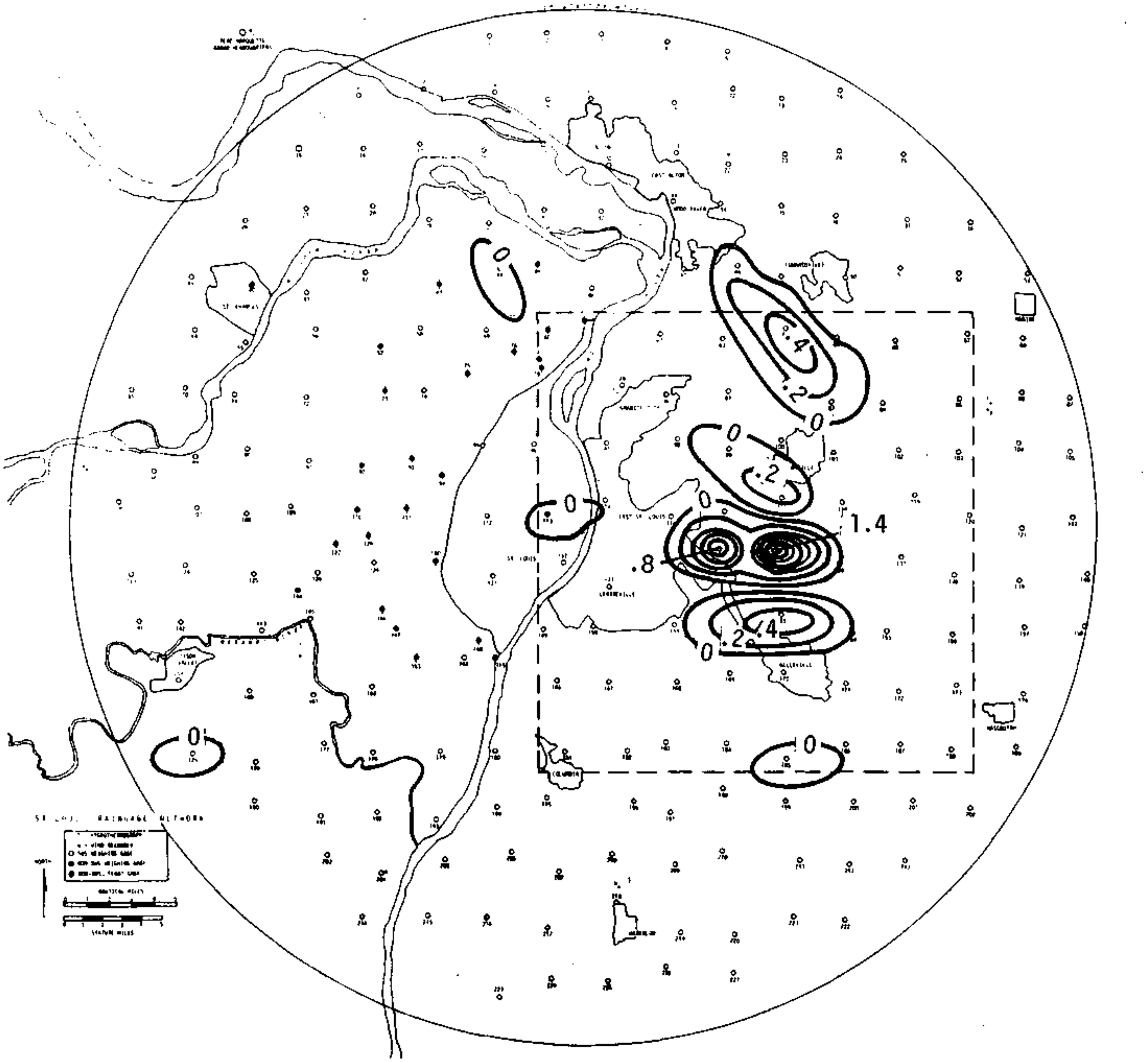

Figure 6. Four minute average rainfall rates in inches/hr for 18 June 1971 for the time ending 1340 CDT

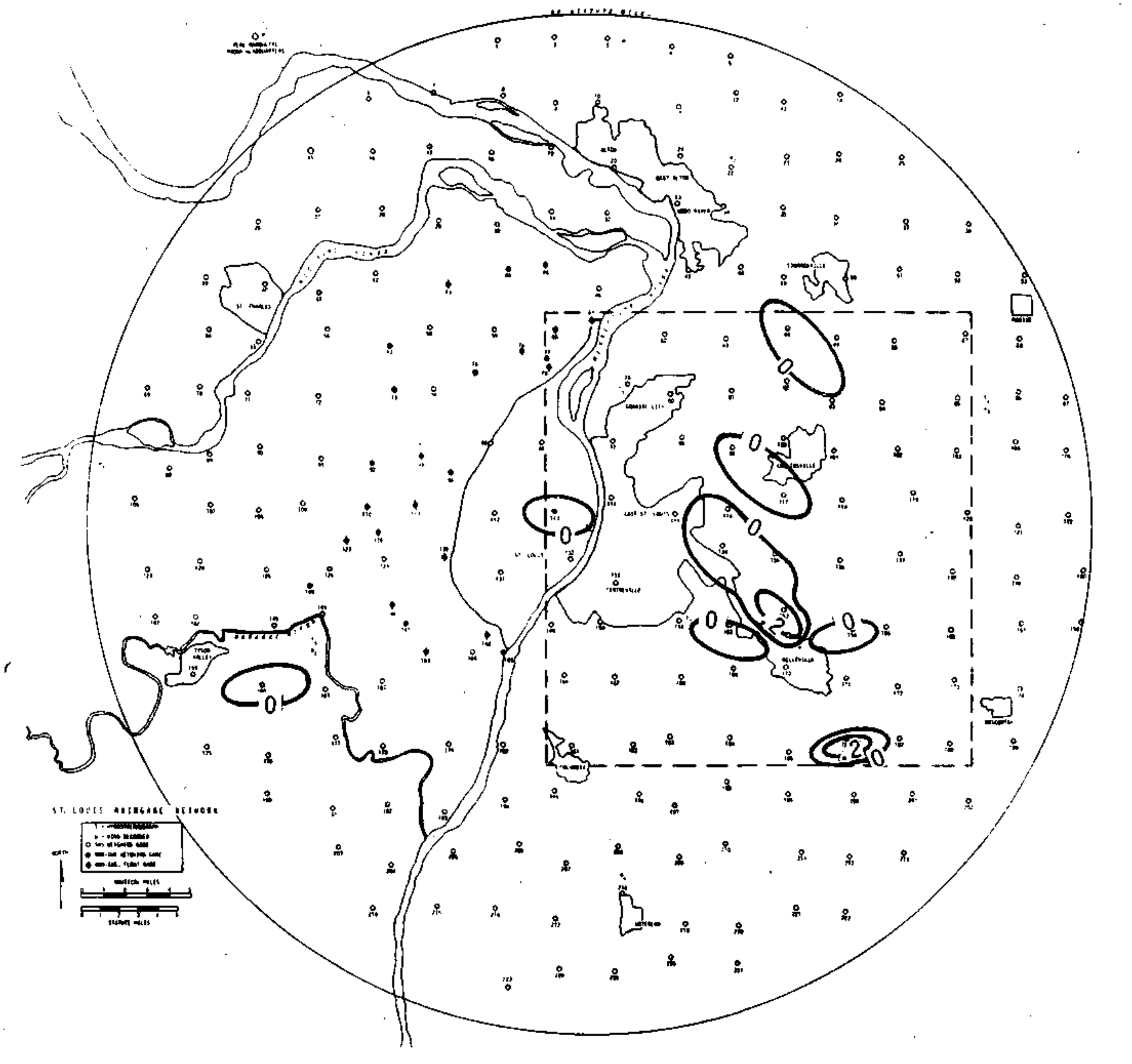

Figure 7. Four minute average rainfall rates in inches/hr for 18 June 1971 for the time ending 1356 CDT

area at an elevation of 1800' MSL. Fig. 8 graphically depicts the temperature profile at flight elevation as the aircraft traversed the city, first in a northeast-southwest direction, then in a south-north path. The dashed lines are isotherms at the surface based upon nine reporting stations.

The northeast-southwest temperature trace aloft, the surface isotherms, and the low level wind flow strongly infer the presence of a heat island effect. The temperature aloft slowly but steadily increases towards the west-southwest, while the surface isotherms follow the same pattern. Since the low-level wind flow was from the northeast at 10-15 knots , it is reasonable to expect the temperature patterns that resulted; the "heat bubble" from the city was' advected downwind (southwest) of the urban region by the low-level wind flow.

Lithium burn - 14 August. A lithium burn was made by the aircraft on 14 August into a moderately strong convective cell with maximum tops to 36K during its north-south passage across the eastern third of the water collection network. The cell was part of an east-west oriented, pre-frontal squall line which moved in a southerly direction in advance of a slow moving cold front.

The aircraft path during the lithium burn is shown in Fig.9. The patterns of lithium, sodium, magnesium, and calcium concentrations, and total rainfall are depicted in Figs. 10-14. The low level wind flow was from the north-northeast during the time of burn (124-1-1341 CDT). Rainfall rate patterns are shown in Figs. 15-18; the calculated rates are five minute averages ending with the time indicated in the figure. A detailed analysis of this case and the other burn days will be forthcoming. A summary of samples collected for chemical analysis during the period June-August is given in Table 1.

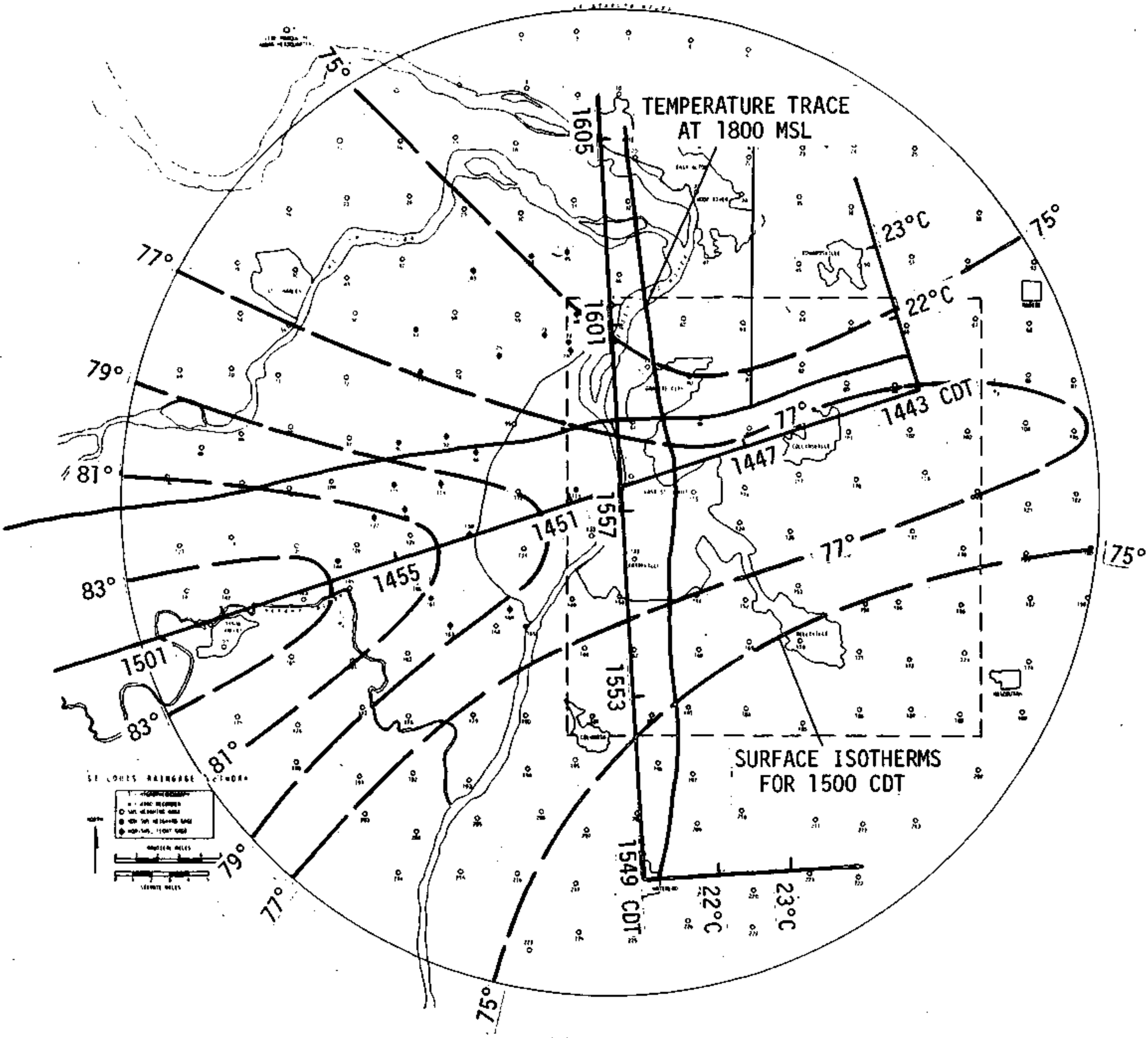

Figure 8. Temperature profiles at the surface and at 1800 ft MSL on 05 August 1971

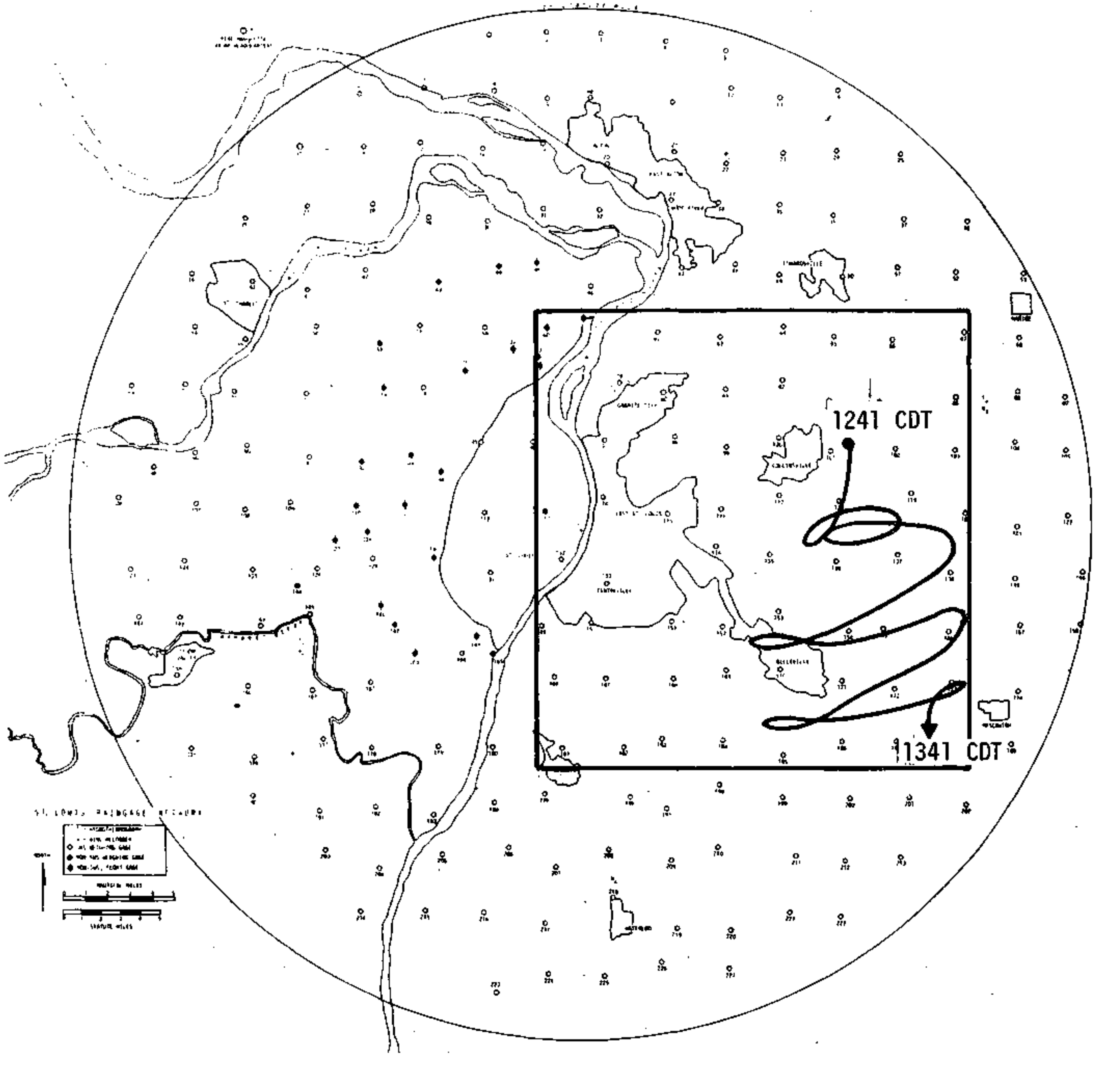

Figure 9. Flight track on 14 August 1971 during lithium burn into a convective cell

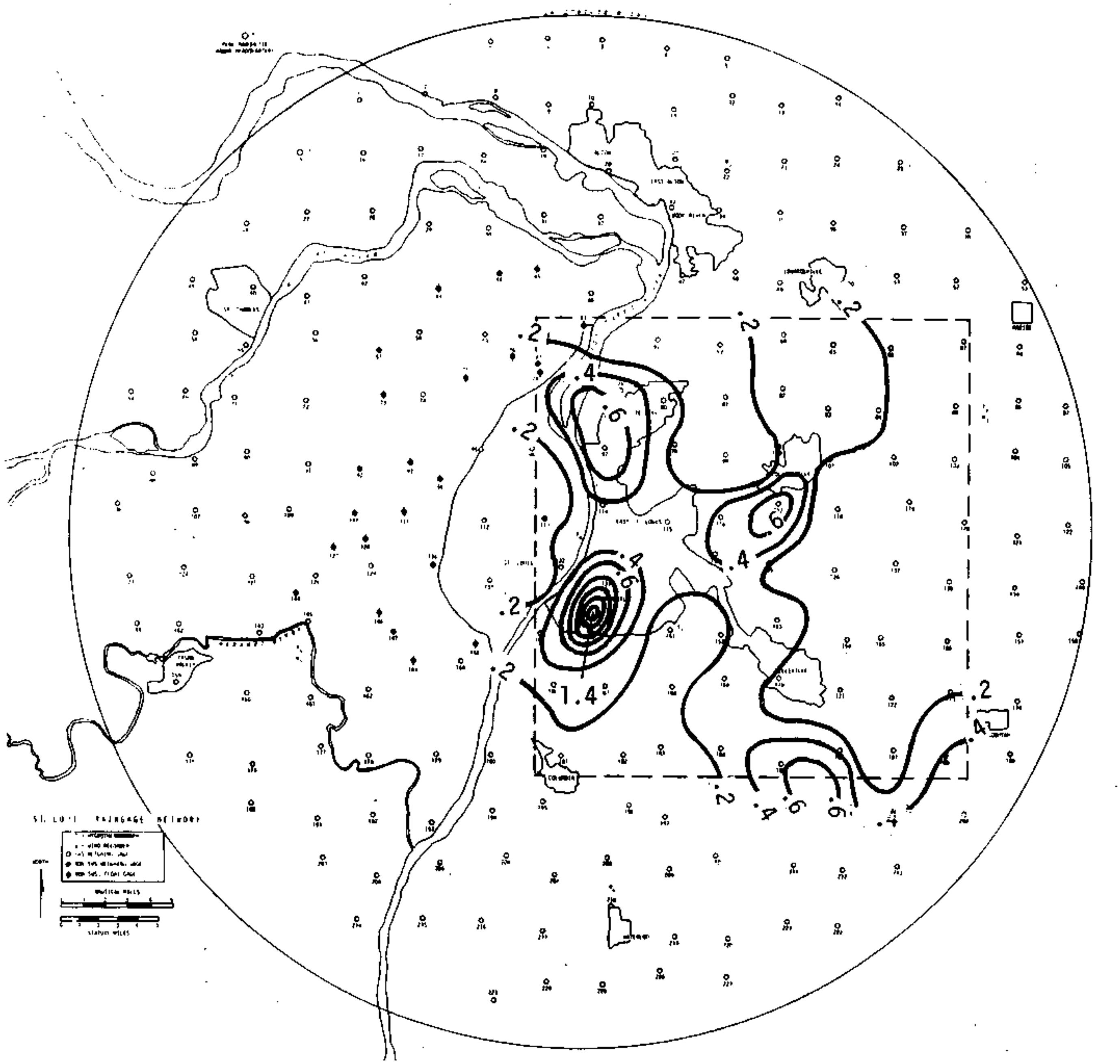

Figure 10. Lithium concentration pattern in parts/billion for 14 August 1971.

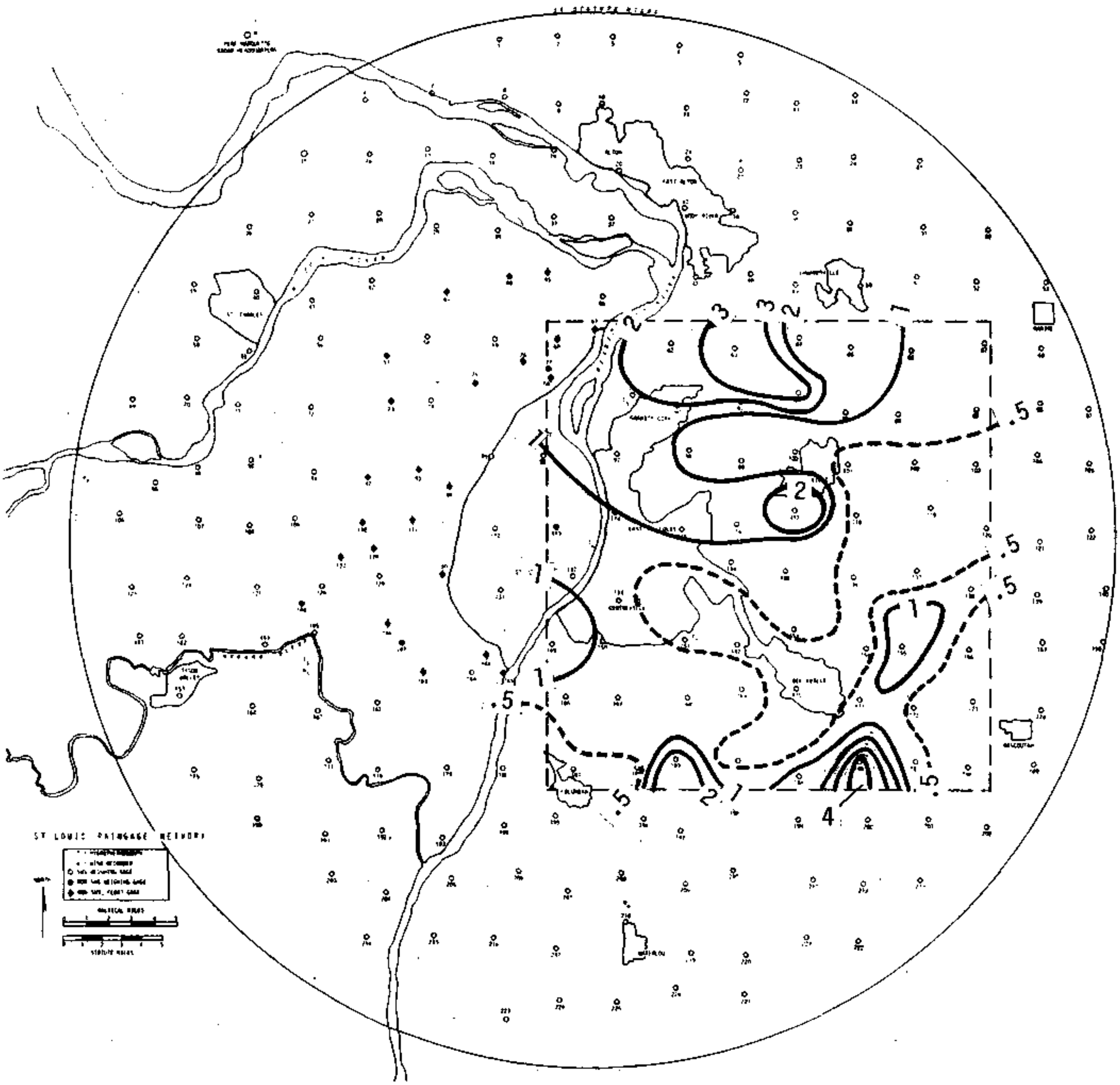

Figure 11. Sodium concentration pattern in ppm x 10^1 for 14 August 1971

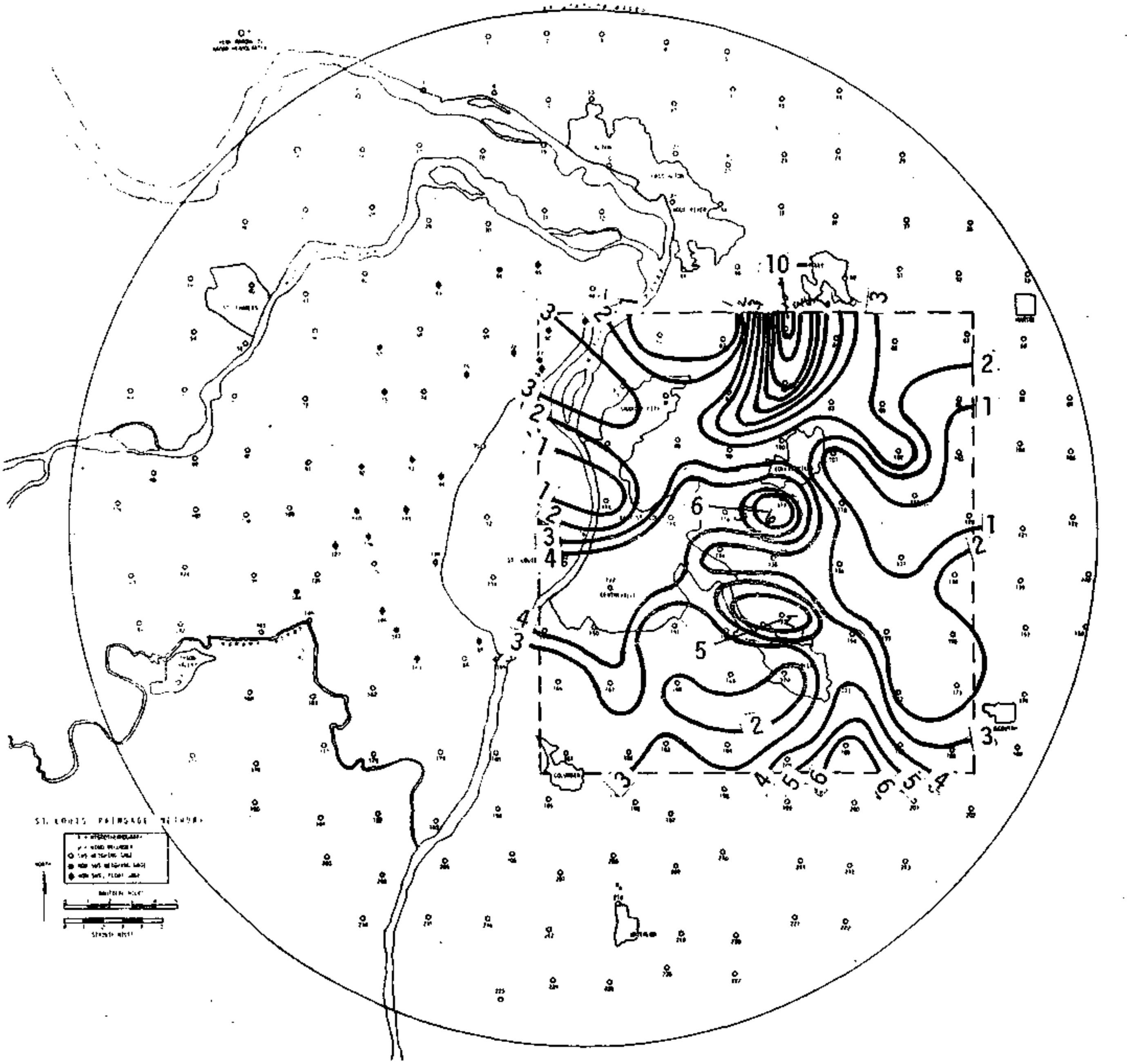

Figure 12. Magnesium concentration pattern in ppm x 10^1 for 14 August 1971

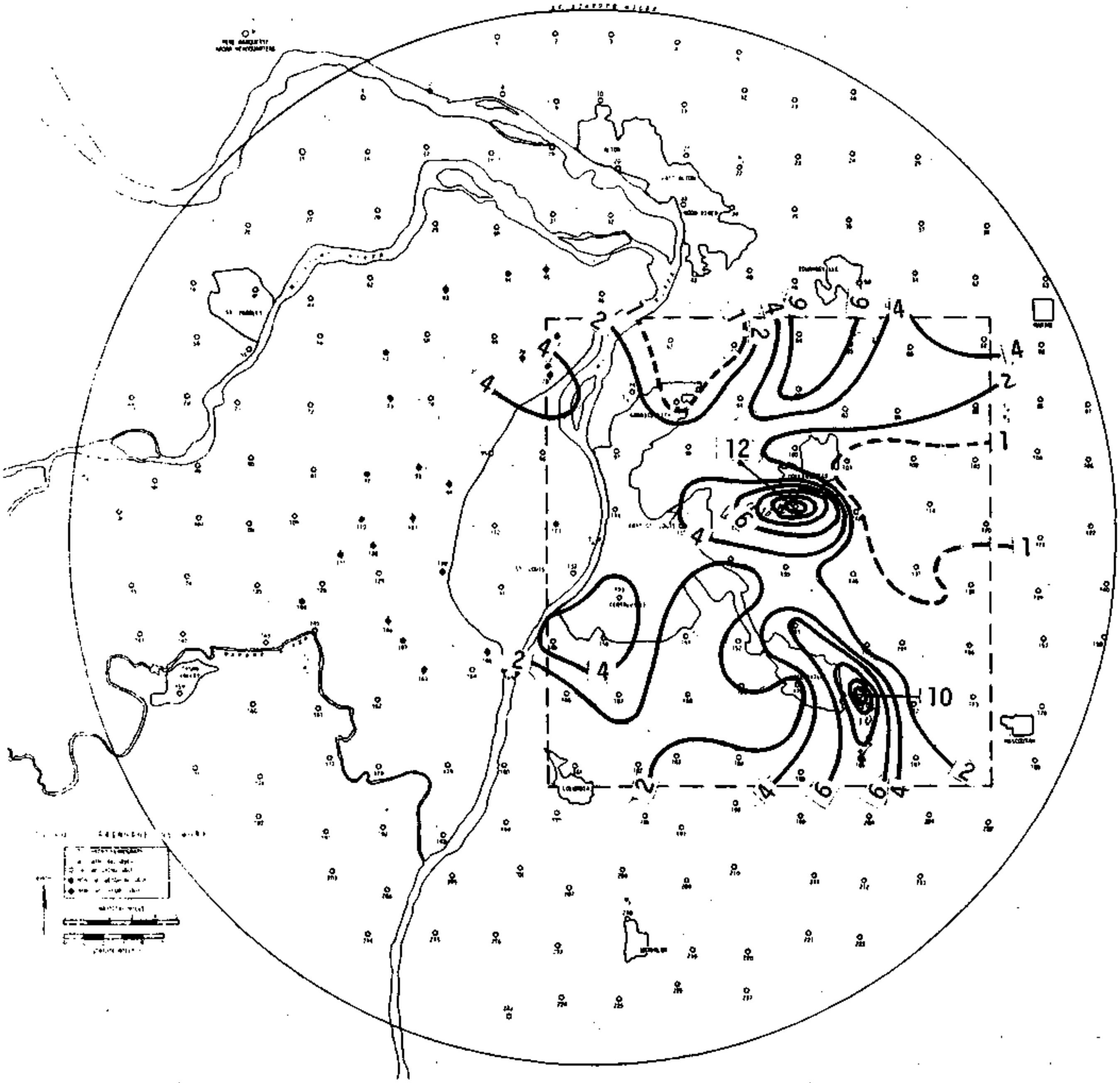

Figure 13. Calcium concentration pattern in ppm for 14 August 1971

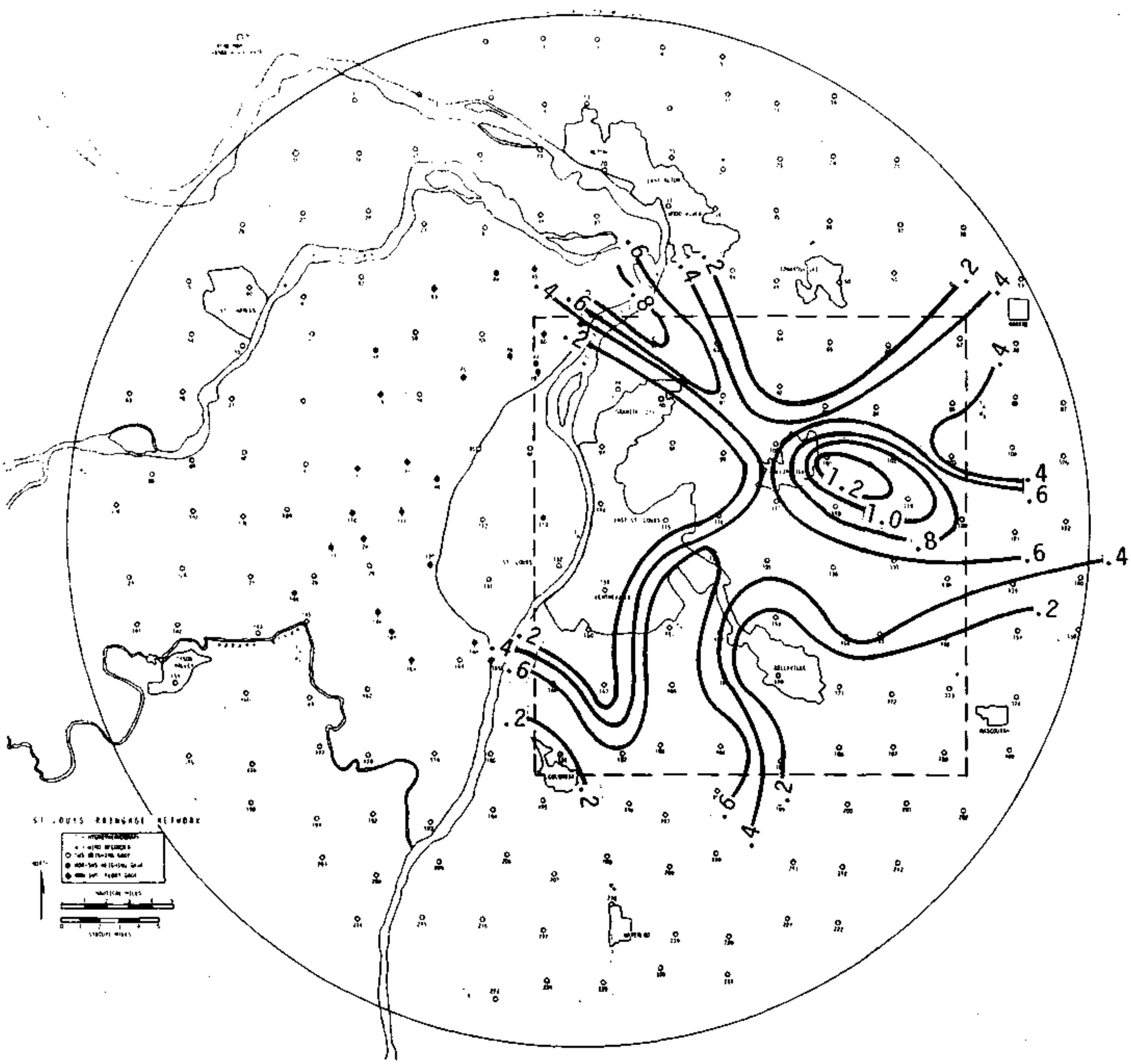

Figure 14. Total rainfall amounts in inches for lithium treated storm 14 August 1971

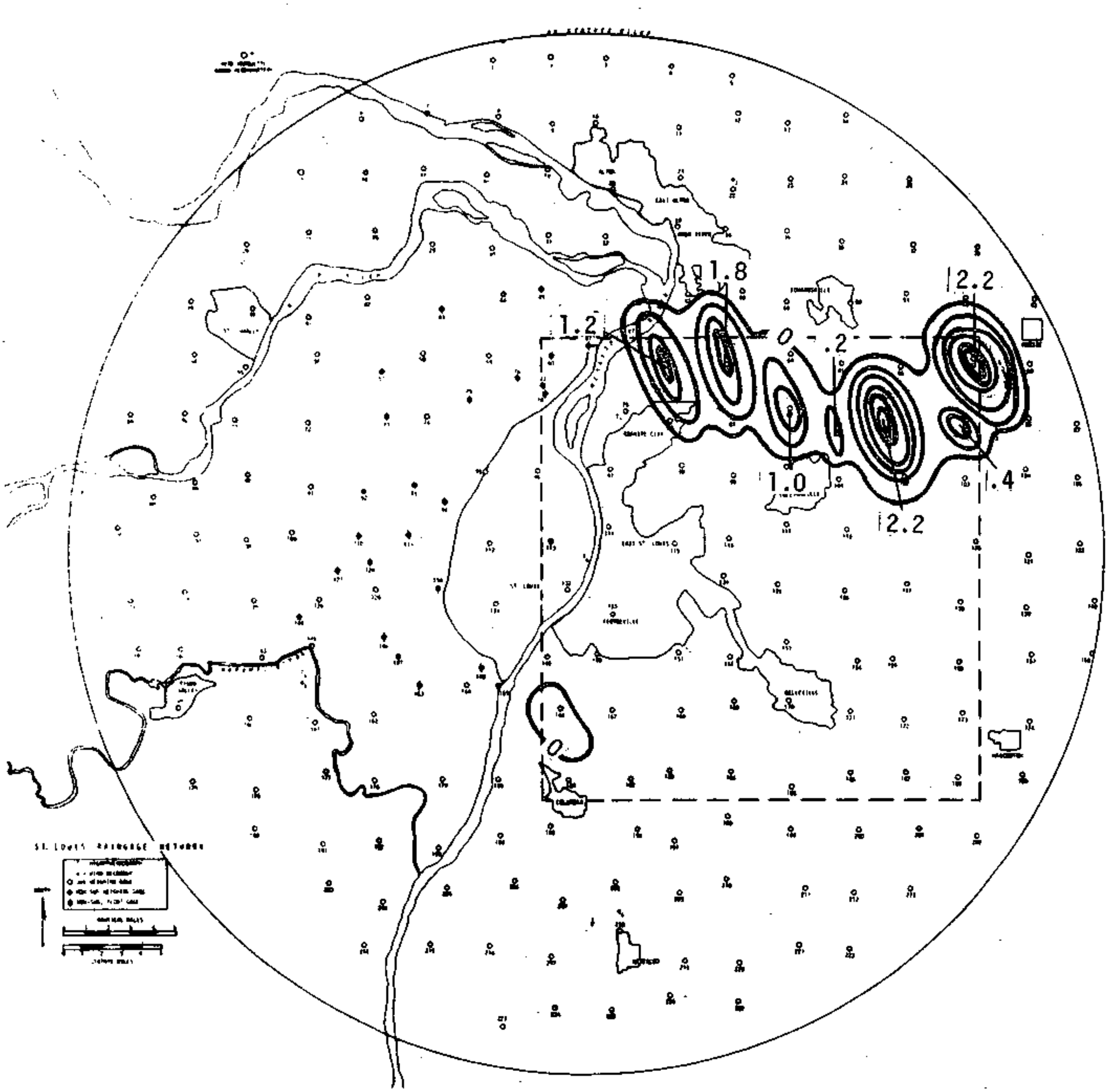

Figure 15. Five minute rainfall rates in inches/hr for 14 August 1971 for the time ending 1240 CDT

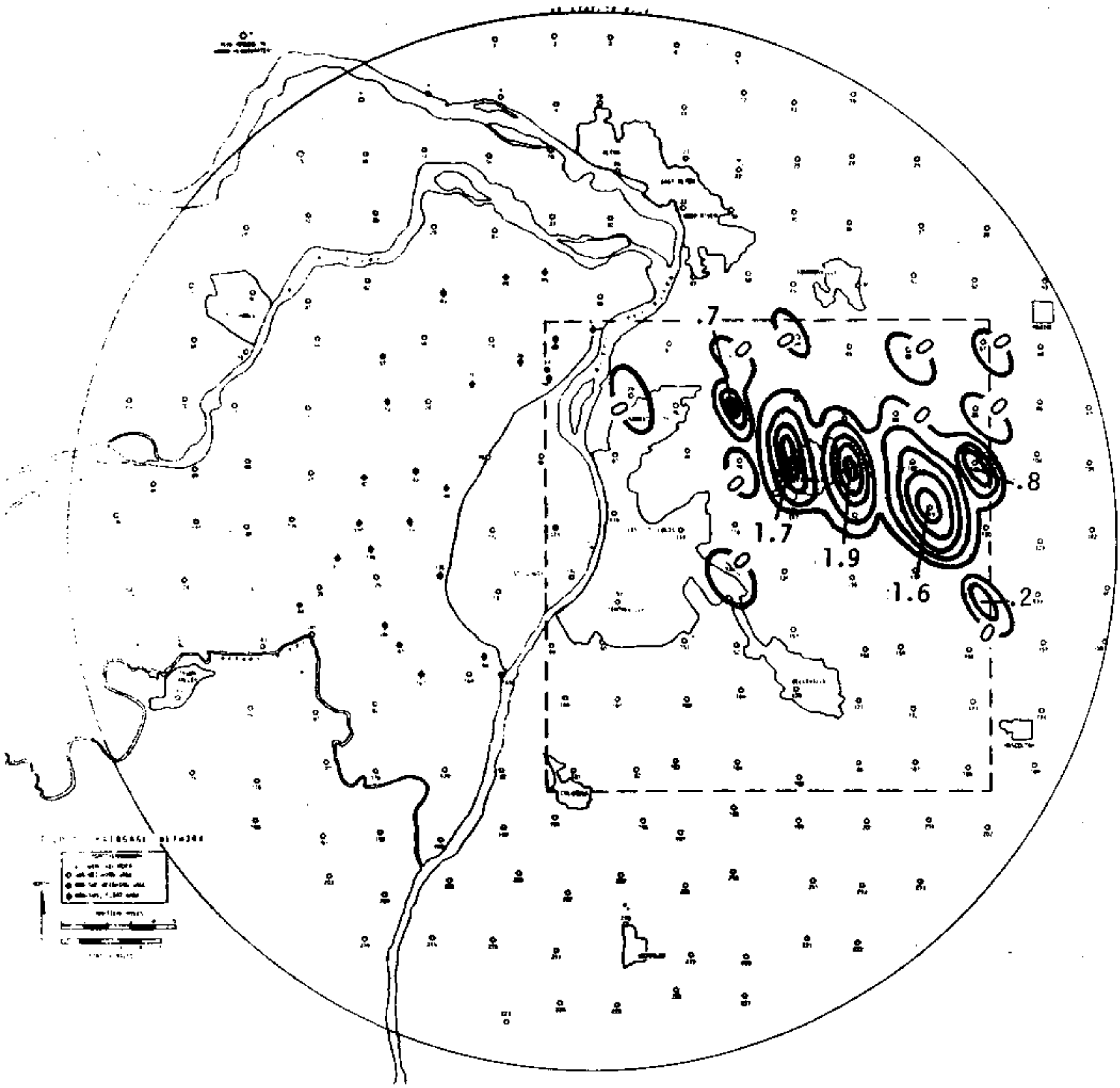

Figure 16. Five minute rainfall rates in inches/hr for 14 August 1971 for the time ending 1300 CDT

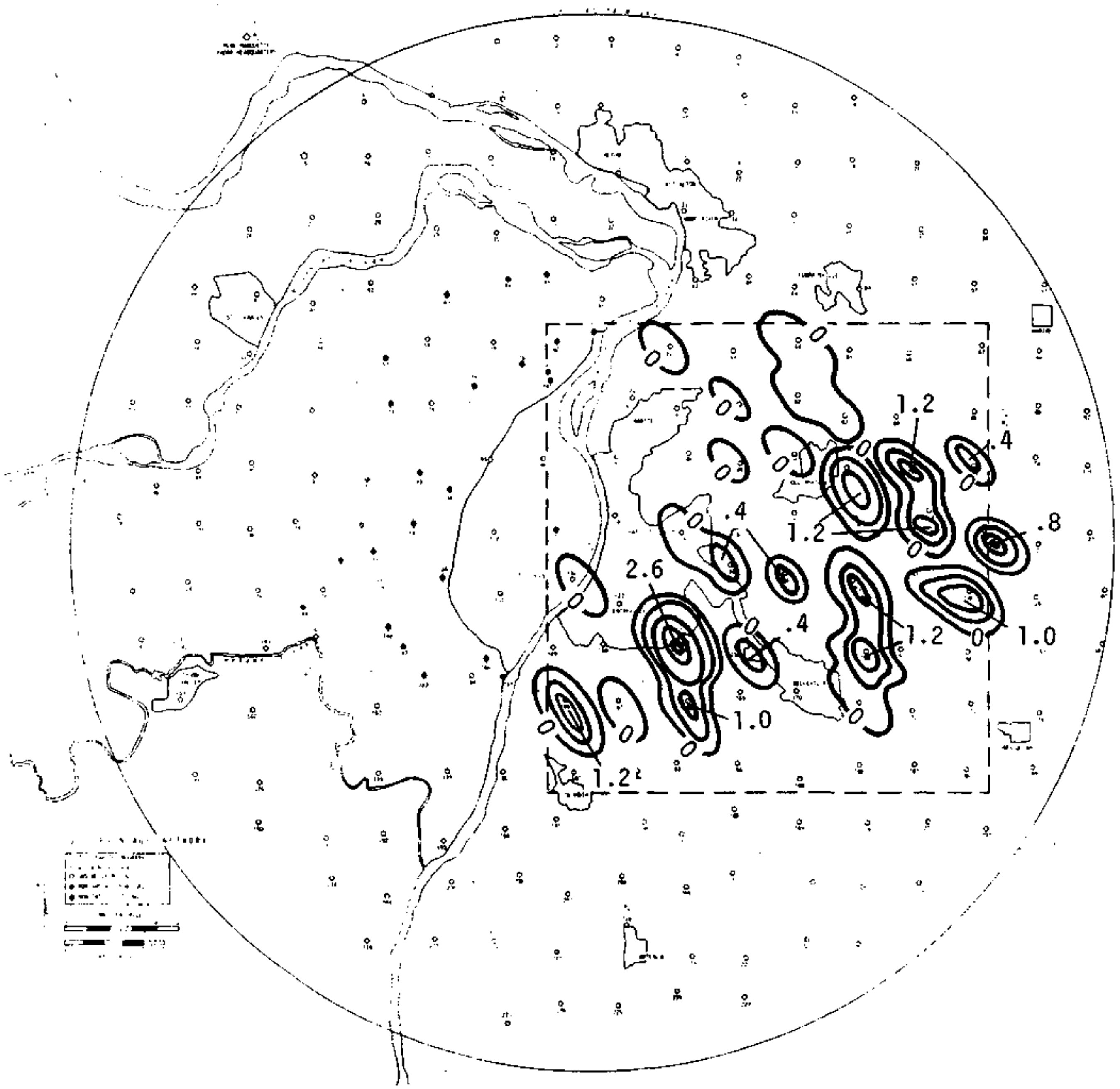

Figure 17. Five minute rainfall rates in inches/hr for 14 August 1971 for the time ending 1320 CDT

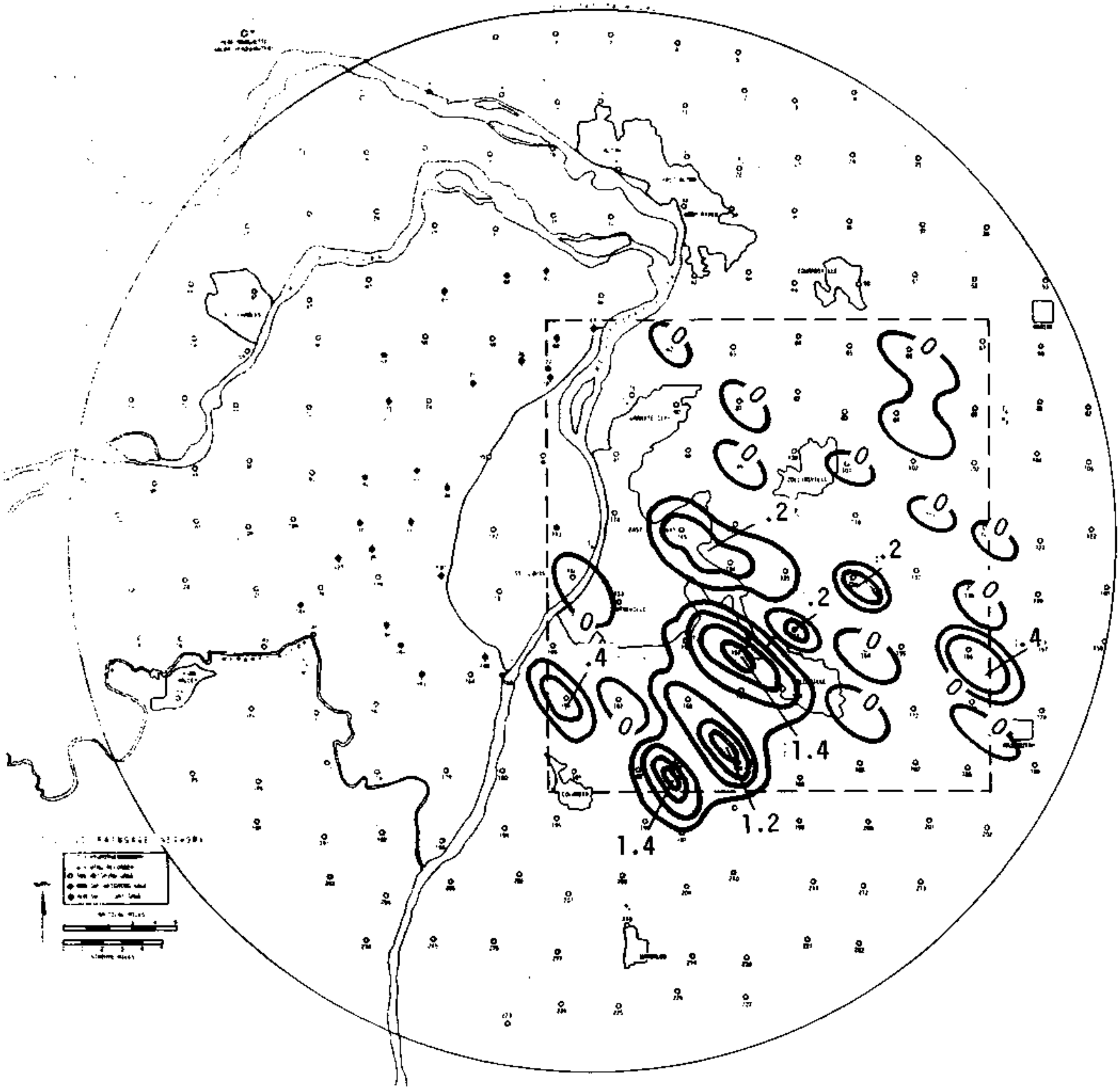

Figure 18. Five minute rainfall rates in inches/hr for
14 August 1971 for the time ending 1340 CDT

Table 1. Summary of Samples Collected for Chemical Analysis

METROMEX-1971

Date	Burn (stack, aircraft)	# Samples Dry	Wet
6/19/1971		56	0
6/21	Stack	56	0
6/25		58	0
6/29		52	0
6/30		34	25
7/03		56	
7/04		0	58
7/08		39	3
7/09	Stack	58	0
7/14	Stack	0	59
7/17		0	57
7/19		57	0
7/25		56	0
7/28		14	2
7/29	Aircraft	0	44
8/03	Aircraft	15	43
8/08		45	10
8/09		56	1
8/10		13	46
8/13		14	2
8/14	Aircraft	5	53
8/21	Aircraft	2	55
8/22-24		57	1
8/24-25		58	0
8/25-28		58	0
TOTALS	Stack - 3 Aircraft - 4	795	523 (including 64 traces)

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APPENDIX A

PUBLISHED PAPER

J. Atmos. Sci., 28, 416-418

INTERACTIONS BETWEEN EQUAL-SIZED DROPLETS DUE TO THE WAKE EFFECT

Reprinted from JOURNAL OF ATMOSPHERIC SCIKNCES, Vol. 28, No. 3, April, 1971, pp. 416-418
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Interactions between Equal-Sized Droplets Due to the Wake Effect

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(Manuscript received 8 September 1970, in revised form 4 December 1970)

ABSTRACT

In the development of raindrops from cloud droplets in warm rain, the collision-coalescence process is considered to be the main growth mechanism for droplets of unequal size greater than 20 µm in diameter. However, due to the wake effect, the possibility of equal-sized droplets colliding does exist for some maximum vertical separation of the droplets. An empirical study has been performed which led to the determination of the maximum vertical separation required, as a function of droplet size, for equal-sized droplets to be influenced by the wake effect.

1. Introduction

In early studies of the collision-coalescence process (Langmuir, 1948; Houghton, 1950), it was assumed that water droplets of nearly equal size in a cloud are in equilibrium with each other, and therefore, coalescence would not take place unless larger droplets were present. However, later experiments (Telford *et al.*, 1955; Woods and Mason, 1965) indicated that collision efficiencies for equal-sized droplets were non-zero and, in fact, were unexpectedly quite large. These results emphasized the importance of the asymmetric flow around a droplet in the collision-coalescence process for droplets of equal size. A recent empirical study (Steinberger *et al.*, 1968) noted, for Reynolds numbers as low as Re =0.06, which is the approximate Reynolds number of a 30-µm diameter droplet falling at terminal velocity, that asymmetric flow exists around the droplet. Conceivably then, two 30-µm droplets falling one above the other could approach each other if their vertical separation were sufficiently small, allowing the decreased drag force in the wake of the leading droplet to act upon the upper droplet. It then becomes important to determine the maximum vertical separation possible, as a function of droplet size, for equal-sized droplets to approach one another. In this paper the experimental procedure used to determine this distance is described along with the results.

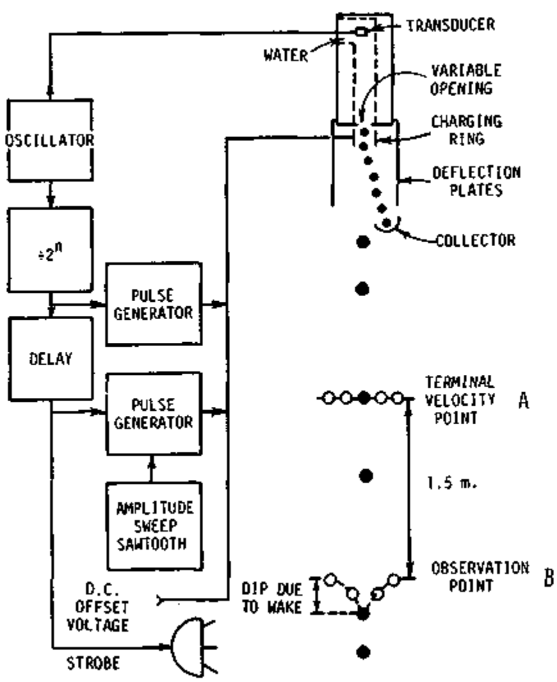

FIG. 1. Block diagram of apparatus to study wake effect.

2. Experimental apparatus and procedure

The experimental apparatus is shown in Fig. 1. A stream of water droplets is produced by forcing water through a small opening of the desired size. A vibrating piezo-electric strip (transducer) in the water supply upstream of the water exit point forces the exiting water jet to break up into uniform-sized, equally spaced droplets. Single, charged or uncharged, droplets can be removed from the stream by applying a voltage pulse at controlled intervals to a charging ring placed around the jet break-up point, and then passing the stream through an electric field (Lindblad and Schneider, 1965). A pair of droplets may be produced in a similar way by applying a second, independent pulse to the ring; this was the technique used. The vertical separation between droplets in the pair can be controlled by varying the time between the first and second pulses. The horizontal position of the two droplets is determined by the pulse amplitude which controls the difference between droplet and stream charge. An automatic sawtooth-amplitude sweep was incorporated in the circuitry to allow the upper droplet in the pair to move from one side of the lower droplet, over it, then to the other side in a sweeping fashion at a rate of 5 cycles per minute. The spacing between pairs was always set much greater than the distance between two droplets in a pair.

When viewed under stroboscopic light, the droplets appear to stand in space with the upper one sweeping back and forth across the lower one as is shown at point A in Fig. 1. If a wake effect is present, it is observed as a dip in the sweeping trajectory of the upper drop as it passes over and accelerates toward the lower drop (point B in Fig. 1). The droplets investigated were allowed to reach terminal velocity before the presence of a wake effect was determined. The distance at which terminal velocity is reached was calculated and was also measured for each pair (Cataneo and Semonin, 1969). The experimental apparatus permitted the droplets to be observed for wake effect at a maximum distance of 1.5 m below the point at which they reached terminal velocity. If a wake effect was observed, the vertical separation was increased until the effect was no longer visible. The vertical separation at which the phenomenon visually disappeared was then measured at the terminal velocity point. The minimum detectable dip induced by the wake was approximately one droplet diameter. Charge on the droplets was varied from 10^{-12} to 10^{-16} coulombs per drop and had no effect on the maximum separation measured. The experiment was carried out for four droplet sizes, 115, 195, 325 and 700 µm in diameter. The maximum vertical separation as a function of droplet size is plotted in Fig. 2.

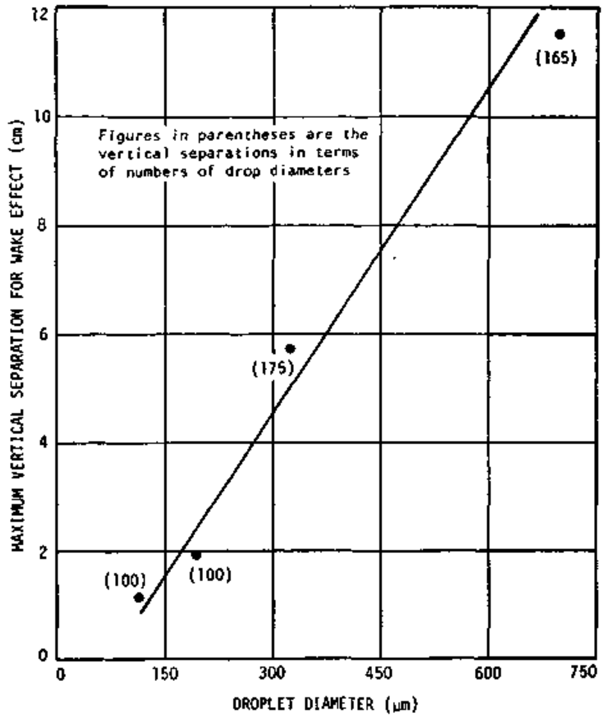

FIG. 2. The maximum vertical separation for presence of a wake effect as a function of droplet size.

3. Discussion

The results were indeed surprising. The wake effect was noted as far away as 11.5 cm for the 700-µm diameter droplets, the distances decreasing directly with droplet size. However, even for the 115-µm droplet, the distance was still quite sizable (1.15 cm, or 100 diameters). The implications of these results with respect to precipitation physics are far reaching in regard to the mechanism for warm rain production.

The wake effect at the maximum vertical distance was observed to be present at radial distances of 2-3 droplet diameters from the bottom droplet. There appears then to be a right circular cone of influence downstream of the leading droplet, which is the cone apex, whose radius at the maximum vertical distance is 2-3 droplet diameters. If we consider that the number density of 100-µm diameter drops may be 3-4 cm^{-3} in a cumulus congestus cloud (Mason, 1957), and if we consider the "wake cone" to be as described above, then the probability of any one of four 100-µm droplets in a cubic centimeter volume being influenced by the wake of one of the other three is ~ 1%. If we extrapolate this probability to a cubic meter, the number of drops being influenced by the wake effect becomes 10^4. Of course, the number density of larger size droplets in a cloud decreases sharply so that the influence of the wake effect becomes increasingly small. However, the wake effect for smaller sizes, where Re < 1, becomes important since their number density in clouds is greater. We are presently investigating these droplet sizes.

4. Conclusions

An experimental procedure has been devised to investigate the magnitude of the wake effect for equal-sized water droplets. From the data obtained to date, we conclude that this phenomenon has a large influence on the collision-coalescence process, and may be a major factor in the rapid growth of raindrops from cloud droplets.

Acknowledgments. This research was sponsored by the Atomic Energy Commission under Contract AT(11-1)-1199 and by the National Science Foundation under Grant GA-4576. We would also like to express our appreciation to Mr. Terry Flach, Mr. Wayne Smith, Mr. David Vercellino and Mr. Timothy Thornburn for their assistance in this project.

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APPENDIX B

PUBLISHED PAPER

Proc. Conf. Air Pol. Meteor., 1971, 65-68

THE WASHOUT OF ATMOSPHERIC PARTICULATES BY RAIN

THE WASHOUT OF ATMOSPHERIC PARTICULATES BY RAIN

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INTRODUCTION

An aerosol can be scavenged from the atmosphere by rain through two basic processes. The first is termed "rainout" and is the result of the aerosol serving as a cloud nucleus or undergoing capture by cloud water or ice particles. The second process is termed "washout" which is due to the removal of the subcloud aerosol by raindrops as they fall. Ultimately, the aerosol returns either to the ground in the raindrops produced by the cloud or to the atmosphere as the cloud dissipates.

The relative importance of these processes in atmospheric cleansing is difficult to assess. Each depends on the nature of the aerosol, its size distribution, concentration, wettability, and activity as nuclei. The rainout process also depends on the nature of the cloud itself; its depth, temperature and water distributions, electrical activity, and other parameters. Because of the complexity of these in-cloud variables, only estimates of the significance of this mechanism can be made (1).

The washout process depends on the characteristics of the rain, including the raindrop size distribution, and the amount of aerosol collected by the individual raindrops. Theoretical and experimental investigations of the important parameters have been performed and a realistic measure of the magnitude of washout can be made. In this paper, the annual amount of aerosol removed by washout is calculated for two locations. The effects of collection efficiencies, as reported by various authors, and the shape of the raindrop size distributions are examined.

THE COLLECTION AND WASHOUT OF AEROSOLS BY RAINDROPS

The below-cloud removal of an amount of aerosol by a raindrop depends on the collision with and capture of the aerosol particles. The collision efficiency, E_1, is defined as the number of aerosol particles which impact the raindrop divided by the number of particles in the volume swept out by the drop. All the particles which strike the drop need not be captured by it. The capture or coalescence efficiency, E_2, is the number of particles which remain with the drop after impact divided by the total number which impact it. The collection efficiency, E, is then the product of the collision and capture efficiencies.

The collision efficiency of raindrops for micron-size particulates has been examined by various researchers. Typical of the theoretical results are the original calculations performed by Langmuir (2) and those later reported by Mason (3). Collection efficiences have also been measured experimentally by Englemann (4) using Zinc Sulfide particles and by Adam and Semonin (5) using B. subtilus spores. These results are summarized in Fig. 1. The experimental results are always well below the theoretical results for the same size particle. This disparity between theory and experiment may partially be explained by the fact that the theoretical values are collision efficiences and the capture efficiency is assumed equal to one. The experimental results are total collection efficiences.

The concentration of aerosol in a volume of air through which raindrops are falling at any time is given by

$$C(t) = C_0 e^{-\lambda t}$$

where Co is the initial aerosol concentration and X is the washout coefficient. The washout coefficient is

$$\lambda = \int_0^\infty \frac{\pi}{6} N_D D^2 V_D E \, dD \qquad (1)$$

where N_D and V_D are the number and terminal velocity of raindrops with diameter D, and E is the particulate collection efficiency. If a source of aerosol exactly replaces that which is removed by the rain, the amount removed or supplied over a period of time, T, can be expressed by

$$\Lambda = \int_0^T \int_0^\infty \frac{\pi}{6} N_D(t) D^2 V_D E \, dD \, dt \qquad (2)$$

where A, the total washout is expressed as a multiplier of the inital concentration. If the annual variation in raindrop size distributions along with the concentration of a component of the atmospheric aerosol is known, then the amount of material deposited on the ground can readily be determined.

RAINDROP SIZE DISTRIBUTIONS AND RAINFALL RATES

Raindrop size distributions have been measured over one-year sample periods at various locations by

Mueller (6) and Mueller and Sims (7). A raindrop camera, as described by Jones and Dean (8) was used to make these measurements. It consists of a 70-mm camera and an optical system to photograph the raindrops in a sample, volume. A series of seven photographs, corresponding to a one cubic meter sample, were taken during each minute of rain. Drops as small as 0.1 mm can be measured and the overall accuracy is +0.2 mm.

Drop distributions from Island Beach State Park, New Jersey and Miami, Florida, were used in the following calculations. A total of 3135 cubic meters of rain were sampled at Island Beach from October 30, 1960 to May 24, 1962. The Miami raindrop distributions were measured from August 20, 1957 to August 14, 1958 and contained 2506 cubic meters. The camera sampled approximately 15% of the total rain during the sample periods at both locations. From the measured results, 30 average distributions were calculated as a function of rainfall rate as shown in Figs. 2 and 3. The distributions at the two locations are quite different. Miami has a, larger mean droplet diameter and strikingly fewer small drops when compared with Island Beach.

A recording raingage was operated simultaneously with the raindrop camera at both locations. Jones (9) has used the raingage records to determine the frequency of occurrance of rainfall rates over the period sampled by the camera. These frequencies, normalized to one year are shown in Fig. 4. The total rainfall at Miami was 119 cm while at Island Beach it totaled 106 cm. Rain fell at Island Beach 4.2% of the year and at Miami only 2.3% of the year, indicating that moderate and heavy rains were more appreciable at Miami.

RESULTS

The collection efficiencies and raindrop size distributions have been used in the integration of equations (1) and (2) to yield the washout coefficient for various rainfall rates and the total annual washout. The washout coefficients for typical particle sizes at Miami and Island Beach are presented in Fig. 5. Washout coefficients for semiarid Sunnyside, Washington, using Mason's collection efficiencies (1) are also shown. The washout coefficients are larger at Island Beach than at Miami, though wider variations are introduced by the various sets of collection efficiency data used. Washout coefficients calculated from experimentally determined collection efficiencies are well below those calculated from theoretical efficiencies.

The annual washout was determined by integrating equation (2) in such a way that the rainfall rate followed the frequency of occurrance in Fig. 4. This parameter can be interpreted in the following manner. Assume aerosol laden air passes through a volume with an average velocity, v, and average concentration, C. The amount of aerosol which enters the volume over a period of time, T, is vCT. If a time period of one year is assumed, the amount of one size of aerosol particle washed from the volume is AC, assuming AC<<vCT. If, for instance, a flow velocity of 1 m/sec is assumed, the portion of aerosol annually removed by rain is 3.17×10^{-6} A percent. This percent removal is also tabulated in Table 1. The amount of material removed is quite small, ranging from 10^{-3} to 10^{-6} percent.

The calculation of the total amount of material removed from the atmosphere and the concentration of aerosol constituents in the rainwater is an obvious extension of this work. Unfortunately, several parameters are missing. The most important of these is the vertical distributions of aerosol concentration and aerosol size. Another missing parameter is a more comprehensive tabulation of collection efficiency for various aerosols. The effects of electric charge on the aerosol and the raindrops must also be considered (5), (10).

A total atmospheric aerosol budget could be determined if the required measurements were made simultaneously. These include the vertical aerosol distribution, the raindrop size and charge distribution, and the concentration of the aerosol in the rainwater. Calculations of the type considered in this paper would then yield the fraction of the aerosol in the rainwater which can be attributed to washout below cloud base. The remainder would then be related to the in-cloud rainout mechanism and its importance in atmospheric cleansing could be assessed.

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Table 1. The Annual Washout and Percent Removal at Atmospheric Aerosols.

Particle Dia. (microns)	Collision Efficiency Reference	Miami Washout	Miami % Removal ($x10^3$)	Island Beach Washout	Island Beach % Removal ($x10^3$)
4.0	Mason	119.0	0.377	174.0	0.553
6.0	Mason	307.0	0.974	417.0	1.32
8.0	Mason	433.0	1.37	574.0	1.82
12.0	Mason	619.0	1.95	801.0	2.54
20.0	Mason	760.0	2.41	988.0	3.13
4.0	Langmuir	117.0	0.370	127.0	0.402
6.0	Langmuir	296.0	0.940	378.0	1.19
8.0	Langmuir	445.0	1.44	572.0	1.81
12.0	Langmuir	618.0	1.95	793.0	2.51
20.0	Langmuir	747.0	2.37	955.0	3.03
6.9	Englemann	37.6	0.120	76.5	0.240
9.76	Englemann	153.0	0.487	236.0	0.749
13.8	Englemann	510.0	1.61	474.0	1.50
1.0	Adam ε Semonin	0.438	0.0014	1.06	0.0034

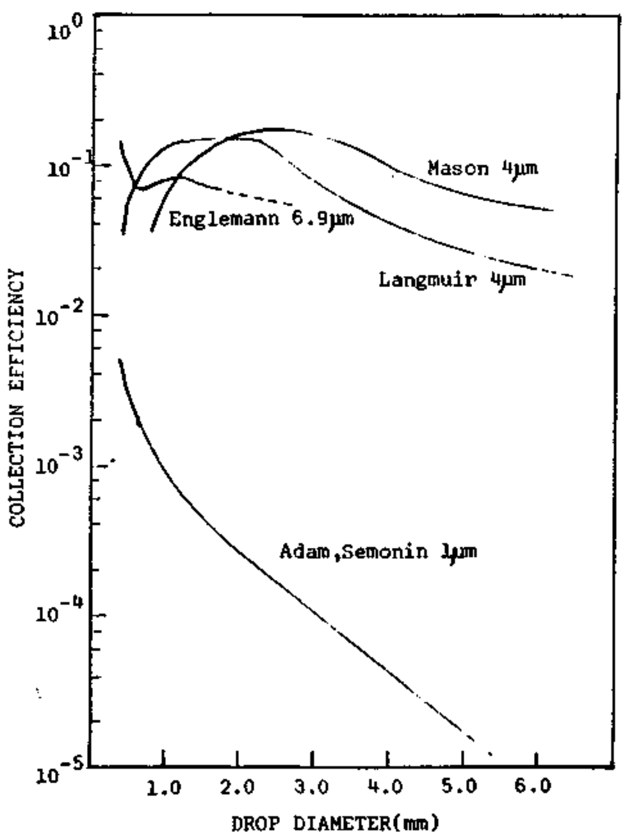

Fig. 1 Collection efficiencies of raindrops for particles.

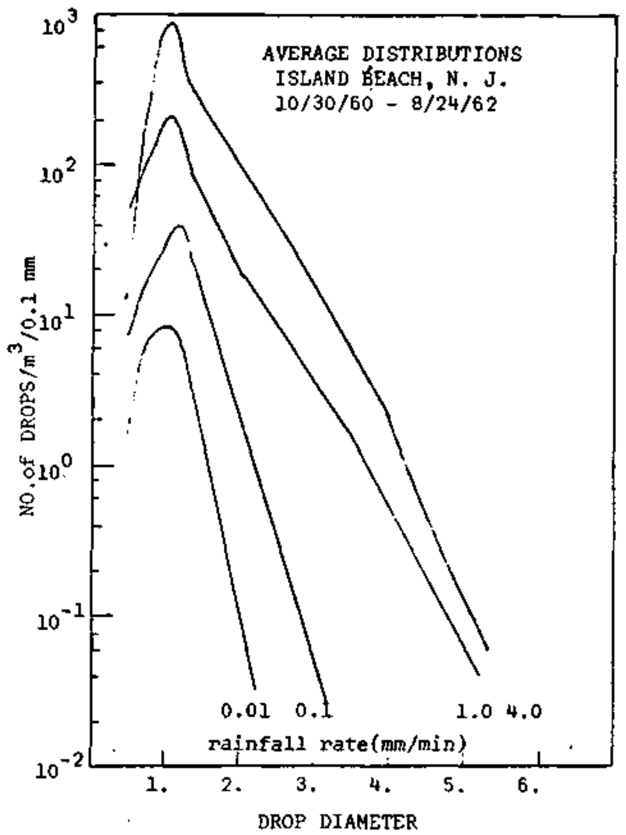

Fig. 2 New Jersey raindrop distributions.

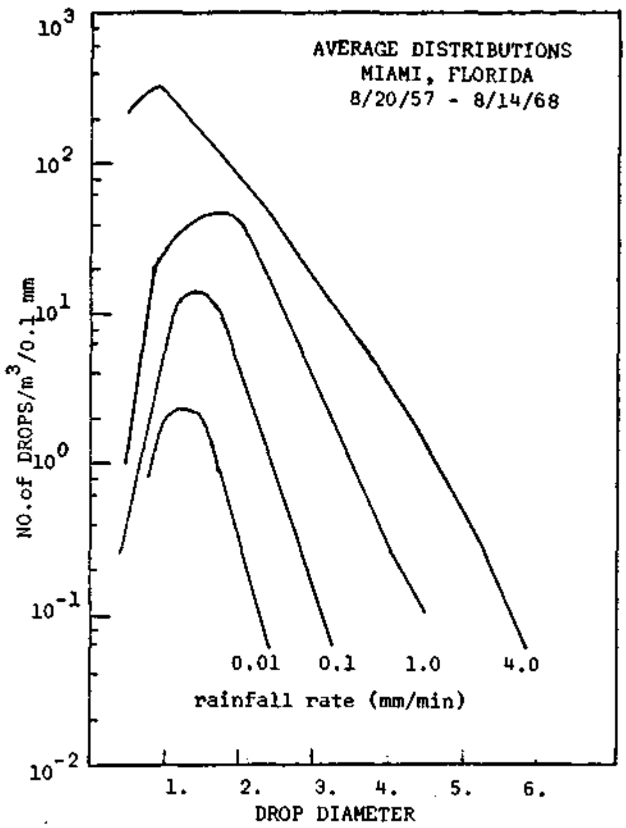

Fig. 3 Florida raindrop distrubutions.

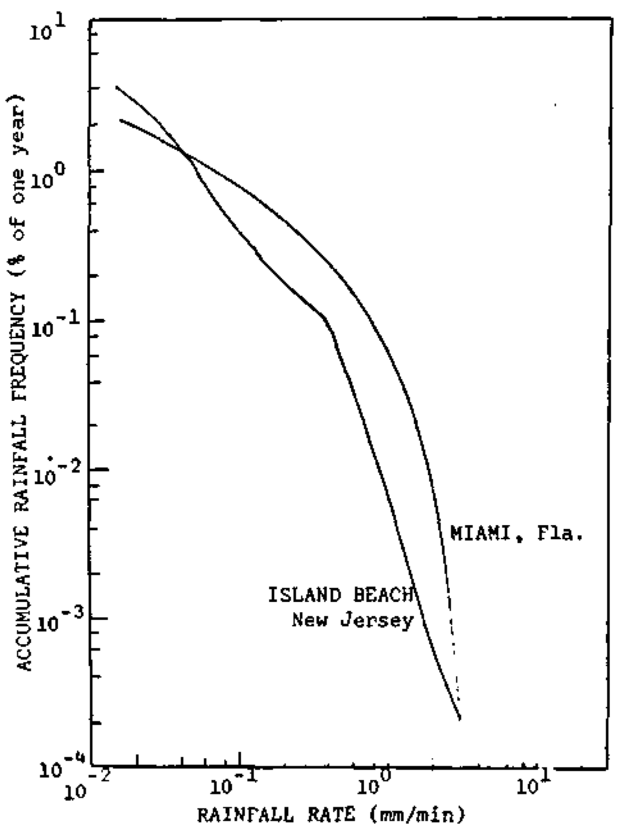

Fig. 4 Frequency of occurrence of rainfall.

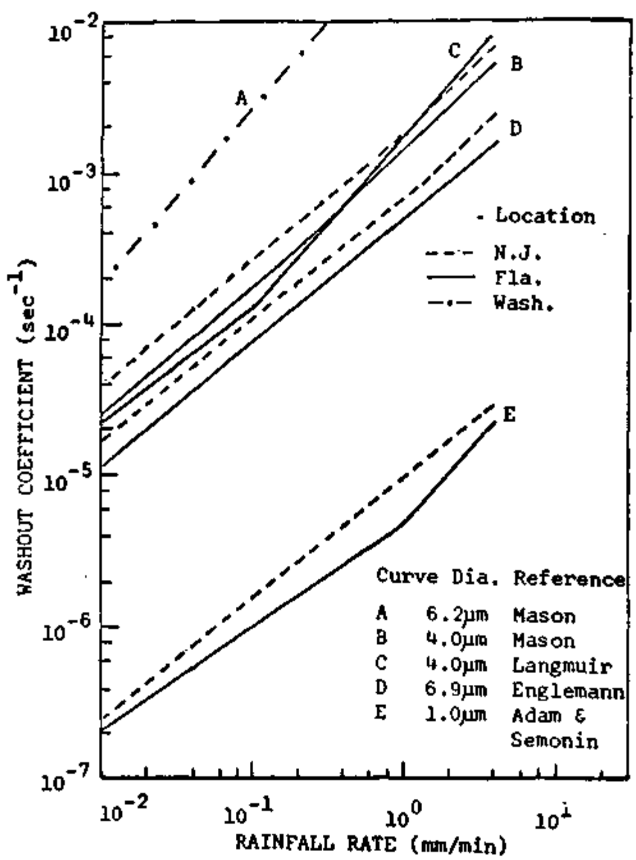

Fig. 5 Washout coefficients at various locations.

APPENDIX C

PUBLISHED PAPER

Proc. Precip. Scav. Conf. Richland, Wash., 1970, 151-159

COLLECTION EFFICIENCIES OF RAINDROPS FOR SUBMICRON PARTICULATES

COLLECTION EFFICIENCIES OF RAINDROPS FOR SUBMICRON PARTICULATES

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ABSTRACT

An experimental technique was developed to determine the scavenging efficiency of raindrops falling at terminal velocity for submicron biological aerosols. The experimental apparatus and methods used to produce the simulated raindrops and the aerosol are discussed. Experimentally determined scavenging efficiencies of raindrops for 1-µ *Bacillus subtilis* spores are presented. The amount of material scavenged is very small, approximately 0.004% for a 2-mm-diameter drop. The scavenging efficiency increases exponentially with decreasing drop sire to a value of 0.4% for a 0.4-mm drop. Also, electric charge on the drop dramatically increases the collection of the submicron particulates.

Aerosols are removed from the air by precipitation in three ways: impaction and capture, or scavenging, of particulates by raindrops; consumption of particulates as condensation nuclei; and attachment of particulates to cloud droplets and raindrops by Brownian motion. Both theoretical calculations[1] and experimental data indicate that impaction and capture is an efficient process for removing particles larger than a few microns in diameter. Extrapolation of these results to submicron particles, however, shows virtually no removal by this method. A new technique has been devised to extend the range of experimentally determined scavenging efficiencies of a raindrop for particles below 1 µ. This technique uses a biological aerosol of the type used by Sood and Jackson[3] to measure the collection efficiency of natural snow.

EXPERIMENTAL PROCEDURE

Apparatus

Figure 1 is a diagram of the experimental apparatus, which consists of an aluminum raindrop-acceleration tower 12.2 m tall and 1 m in diameter. At the

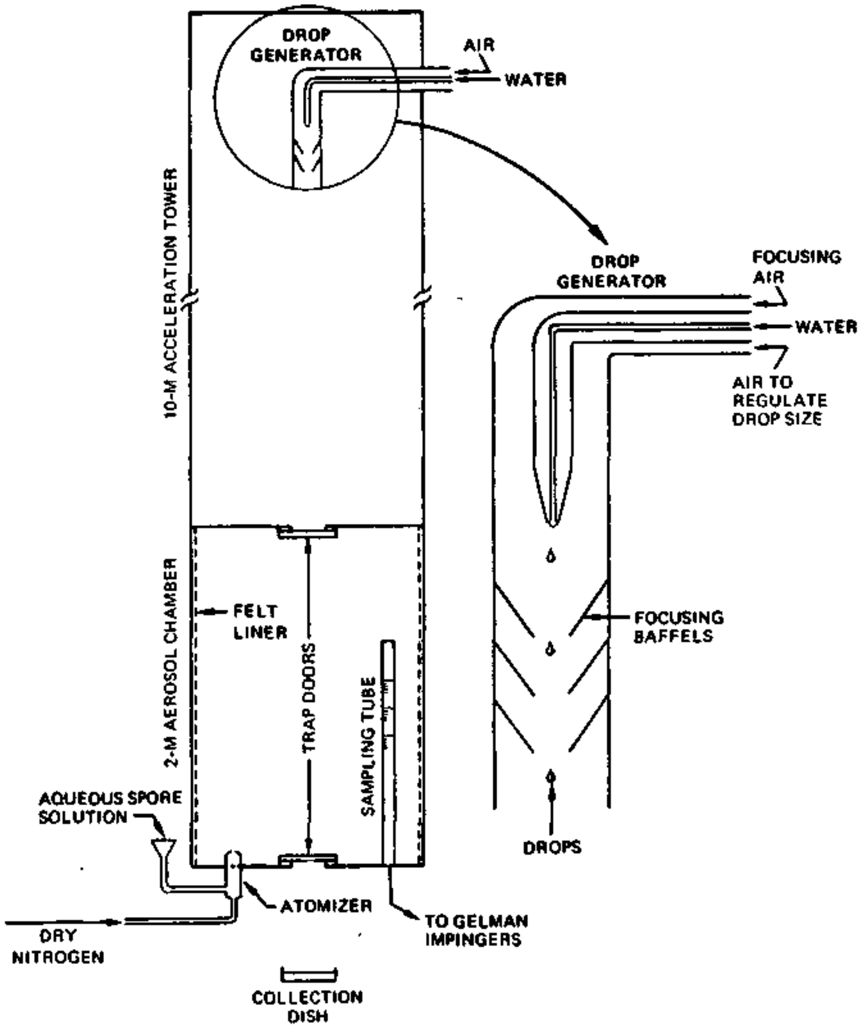

Fig. 1 The experimental apparatus used to measure raindrop scavenging efficiencies.

bottom, a 2-m section of the tower is sealed to form an aerosol chamber. At the center of the top and the bottom of the chamber are 12-cm-diameter sliding doors, which can be opened manually to allow the simulated raindrops to pass through them. The walls of the chamber are lined with felt to absorb moisture from the atomized-particlc suspension. A sampling tube extends vertically through the chamber and is attached to two midget impingers in series, a flow meter, and a vacuum pump for determining the aerosol concentration. An electronic probe to monitor aerosol charge, with a resolution of 10^{-16} coulombs per particle, is mounted at the bottom of the chamber.

A drop generator is mounted within the tower at a height chosen to ensure that the drops reach terminal velocity before entering the aerosol chamber. Simulated raindrops are generated in two ways. Large drops, with diameters greater than 2 mm, are produced at a slow rate by dripping water from a hypodermic needle. An electrically operated shutter opens in sequence with the aerosol chamber doors to allow one drop at a time to fall from the generator, through the tower and aerosol chamber, and into a collector.

Smaller drops are produced by the drop generator shown in Fig. 1. Water flows through the capillary tube to form a drop at its tip. A controlled airstream passes over the capillary, and the aerodynamic drag, along with gravity, pulls the drop from the tip. The diameter of the drops varies from 0.1 to 2 mm depending on the size of the capillary tube and the air flow rate. The drops fall from the drop generator into another airstream and through a series of baffles that accelerate the drops and focus them toward the center of the tower.

Two independent drop-size measurements are made. Volume collections of drops are used to determine the mass of a known number of drops. Drops are also collected in immersion oil and measured optically. Both drop generators produce drops with diameters that vary less than 5%.

The drops from either type of generator can be electrically charged. The drop at the end of the capillary tube is inductively charged by putting an electrical potential between a metallic ring surrounding the drop and the water reservoir. The amount of charge on the drop is determined by the electrical potential and the capacitance between the drop and the ring at the time of breakoff. The charge on one drop is measured by collecting a drop in a shielded metal cup connected to a coulombmeter.

The amount of submicron particulate matter scavenged by a single simulated raindrop passing through the aerosol chamber is very small. Detection difficulties have been overcome by the use of bacteria spores as the aerosol constituent. After passing through the aerosol, the scavenging drop is collected on a petri dish and incubated, and the colonies originating from the individual spores are counted. *Bacillus subtilis* spores are being used currently. These spores are rod shaped, with a diameter of 0.7 μ and an length of 1.2 μ. An aerosol of the spores is formed by atomizing an aqueous solution of the spores with pressurized air. The solution is diluted as much as is necessary to ensure one spore at most in each atomized drop. After it is generated, the aerosol is allowed to stand for several minutes. This dries the spores and allows any large droplets to settle out. Microscope slides are then placed on the bottom of the chamber to collect spores for a short time. The slides can be examined to make certain that the spores are monodisperse.

The aerosol concentration is determined at various times during an experimental run by passing a known volume of air through a midget impinger for which the collection efficiency for the spores has been determined. After a sample is taken, the water in the impingers is diluted, plated on petri dishes, and incubated. After several hours the number of colonies on the dishes is counted, and the original aerosol concentration is calculated. Unlike other techniques this method determines only the concentration of viable spores. Fortunately these are also the only spores that are detected, by a similar culturing technique, in a scavenging raindrop. Typically, the spore concentration decays with time as is shown in Fig. 2.

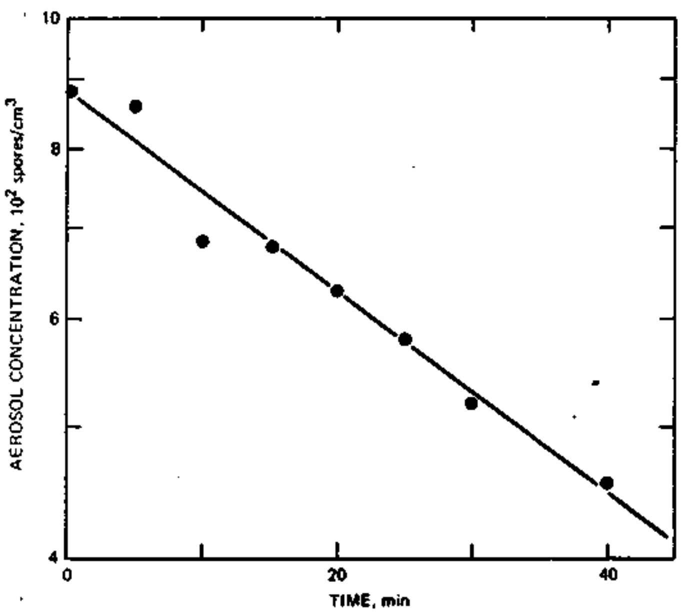

Fig. 2 The decay of *a B.* subtilis spore aerosol with time.

Procedure

A typical experimental run proceeds as follows. The drop generator, adjusted to produce a drop of a specific size, is positioned so that the drop falls at terminal velocity through the aerosol chamber. The aerosol is generated, and an initial concentration sample is taken. The next approximately 15 to 30 min of high aerosol concentration is divided into various series of drop collections. A measurement of the aerosol concentration is made at the beginning and end of each series. Within each series a large number of drops, with or without electrical charge, pass through the aerosol, and each drop is collected on a petri dish placed 1.2-m below the aerosol chamber. Drops larger than 1 mm are collected one per petri dish, but several smaller drops that do not overlap are collected per dish. The time interval between the last concentration measurement and the catching of a drop is recorded so that an accurate concentration of the exponentially decaying aerosol can be calculated.

The plates arc then incubated long enough for each spore to develop into a colony of cells approximately 30 μ in diameter. At this size the cells are large enough to be counted easily with a microscope but do not overlap and lose their individual identity. Figure 3 shows a typical colony that developed in collected drop water. The area of the agar surface struck by the drop is marked as the water is absorbed, and the colonics within the marked area arc counted. The number of background colonies outside the drop areas, usually averaging 1 or 2 per plate, is also counted and subtracted from the drop-deposited spores in proportion to the drop vs. nondrop area ratios. The collection efficiency for each drop is then calculated.

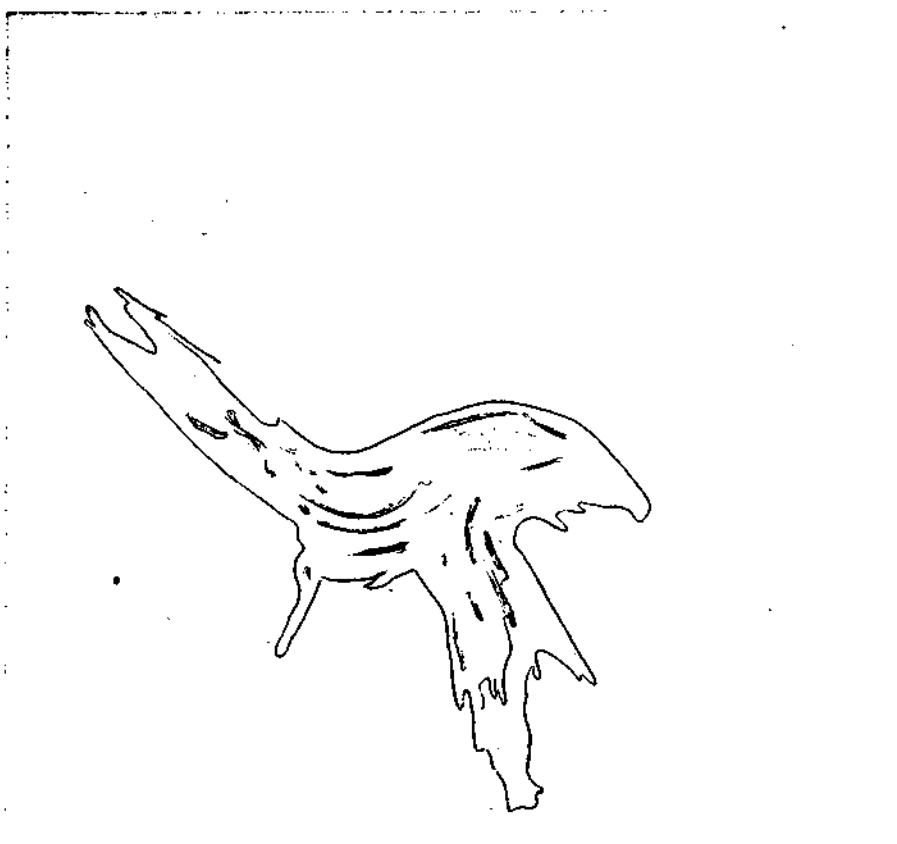

Fig. 3 A colony of cells grown from a single *B, subtilis* spore scavenged by a simulated raindrop.

For accurate experimental results, several precautions should be taken. The area around the tower is cleaned and disinfected to ensure a clean environment before the experimental run begins. The tower and aerosol chamber are ventilated with clean air to remove residual spores and dust. All glassware, the agar, and the drop generator and its water are autoclaved. Control air samples are taken around the tower and chamber to obtain a background spore concentration. The concentrated spore solution is prepared and atomized by someone other than the experimenter to further reduce the possibility of contamination.

EXPERIMENTAL RESULTS

The ratio of the number of particulates scavenged from the aerosol to the total number in the volume through which the drop falls is defined as the scavenging efficiency. The scavenging efficiency (%) is calculated by

$$E = \frac{n_d \times 100}{\pi r^2 L\, c(t)} \tag{1}$$

where n_d is the number of spores collected by a drop, r is the drop radius, L is the length of the aerosol chamber, and c(t) is the aerosol concentration at the time the drop passes through it. In the analysis of the experimental results, we determine the aerosol concentration for each drop by assuming an exponential decay between impinger samplings.

The scavenging efficiency for uncharged drops is shown in Fig. 4. Single drops of various sizes were produced, collected, and examined individually. We determined the results reported here by averaging the collection efficiencies typically from over 50 drops with greater than 2-mm diameters to several hundred smaller ones. The scavenging decreases exponentially with particle size and approaches the experimental resolution near 2.5 mm. This resolution is set by the background contamination of spores and corresponds to approximately 1 spore in 10 drops examined.

Only the increase in collection due to large charges on the raindrops has been examined so far. Although the charges are large, they are still well below the Rayleigh limit, and natural raindrops with charges of this magnitude have also been observed. Figure 5 shows the increased scavenging of the uncharged aerosol by drops with various diameters and charges. The scavenging efficiency increases linearly with increasing charge from its value in the uncharged case (correlation coefficient greater than 0.95).

In all cases the aerosol charge was monitored, and no charge greater than 10^{-16} coulomb/spore was observed. Only for the 3.7-mm drops was there any tendency for increased collection with one sign of charge. This might possibly indicate that the aerosol had a small net charge for this run.

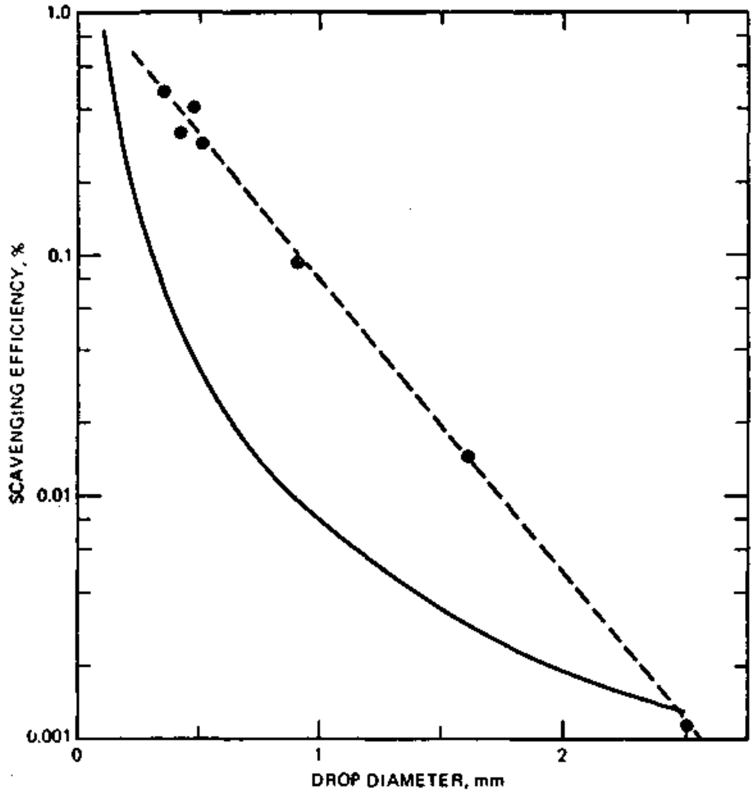

Fig. 4 The scavenging efficiency of uncharged drops for a *B. subtilis* aerosol. •, experimental results. —, experimental resolution.

CONCLUSIONS

The experimental technique described is both accurate and sensitive for measuring scavenging efficiencies. The ease of generation, detection, and decontamination, the uniformity of size, and the ease of concentration determination make bacteria spores well suited for use as the aerosol particulate. The only difficulty is finding other suitable spores, smaller than 1 μ, which are viable after atomization and which have a relatively low natural occurrence.

The measured scavenging efficiencies make it possible to estimate the amount of material in the 1-μ size range that will be removed by natural rain. It is assumed that rainfall has a Marshall Palmer drop distribution for drops greater than 0.5 mm, that the drops are uncharged, and that they fall at terminal velocity. A single calculation then shows that less than 1% of the particles are

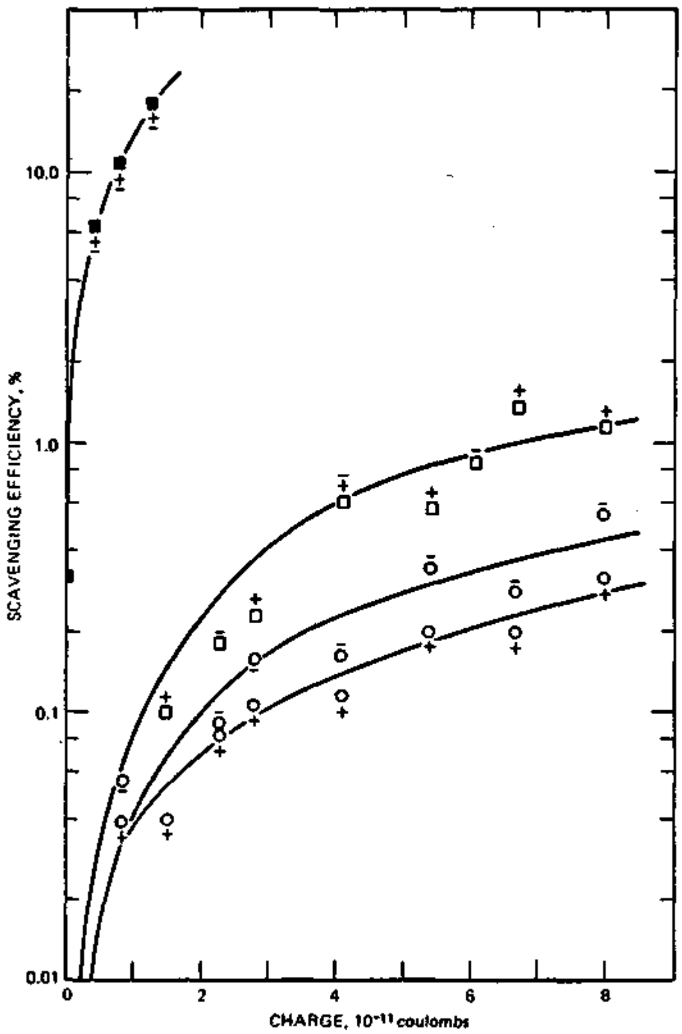

Fig. 5 The effect of raindrop charge on the scavenging of an uncharged aerosol. ■, 0.4-mm diameter, 2×10^{-10} coulombs. □, 2.8-mm diameter, 9.7 x 10 coulombs. O, 3.7-mm diameter, 1.5 x 10^{-9} coulombs.

removed per hour in a 5 mm/hr rain and that only 4.5% are removed in a 100 mm/hr rain.

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APPENDIX D

REPORTS PREPARED UNDER THE CONTRACT NUMBER AT(11-1)-1199 U.S. ATOMIC ENERGY COMMISSION

APPENDIX D

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

C00-1199-2 -- Second Progress Report - January 31, 1964 - F.A. Huff "Study of Rainout of Radioactivity in Illinois"

C00-1199-3 -- SWS Reprint Series No. 46 - F.A. Huff "Radioactive Rainout Relations on Densely Gaged Sampling Networks"

C00-1199-4 -- SWS Reprint Series No. 45 - F.A. Huff and G.E. Stout "Distribution of Radioactive Rainout in Convective Rainfall"

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

C00-1199-6 -- Research Report No. 1 - March 1965 - F.A. Huff "Radioactive Rainout Relations in Convective Rainstorms"

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

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

C00-1199-9 — Reprint - Vienna Paper - Symposium on the Use of Isotopes in Hydrology - G.E. Stout and F.A. Huff - November 14-18, 1966 "Rainout Characteristics for Hydrologic Studies"

C00-1199-10 -- Fifth Progress Report - December 1966 - W.E. Bradley and P.J. Feteris "Study of Rainout of Radioactivity in Illinois"

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

C00-1199-12 — Reprint - TELLUS - October 1967 - F.A. Huff and G.E. Stout "Relation Between Ce^{144} and Sr^{90} Rainout in Convective Rainstorms"

C00-1199-13 — Conference at Chalk River Laboratories, Canada - September 11-14, 1967 - F.A. Huff and G.E. Stout "Time Distributions of Radioactivity and Chemical Constituents in Rainfall"

C00-1199-14 — Sixth Progress Report - November 1967 - F.A. Huff, W.E. Bradley, and P.J. Feteris "Study of Rainout of Radioactivity in Illinois"

C00-1199-15 -- Research Report No. 3 - July 1968 - John W. Wilson and Parker T. Jones, III "Tracing Tropospheric Radioactive Debris by Isentropic Trajectories"

C00-1199-16 -- SMRP Research Paper No. 74 - June 1968 - Walter A. Lyons and John W. Wilson
"The Control of Summertime Cumuli and Thunderstorms by Lake Michigan During Non-Lake Breeze Conditions"

C00-1199-17 — Seventh Progress Report - November 1968
"Study of Rainout of Radioactivity in Illinois"

C00-1199-18 — Eighth Progress Report - November 1969
"Study of Rainout of Radioactivity in Illinois"

C00-1199-19 -- Ninth Progress Report - November 1970
"Study of Rainout of Radioactivity in Illinois"

C00-1199-20 — Tenth Progress Report - November 1971
"Study of Rainout of Radioactivity in Illinois"