

ILLINOIS STATE WATER SURVEY

METEOROLOGY LABORATORY

at the

University of Illinois
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ATMOSPHERIC PARTICULATES IN
PRECIPITATION PHYSICS

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ABSTRACT

The temporal and spatial distribution of giant nuclei in the atmosphere over Illinois was investigated to determine whether their presence was directly related to precipitation processes occurring in that area. A semiautomatic aerosol sampling device was designed and constructed to obtain up to 20 individual samples. A spot test technique was used to determine the chemical types of the particulates captured on a membrane filter.

The results of the analyses indicated that the size and number distribution of particulates does not correlate with precipitation occurrences. There is no obvious relationship between the chloride particle concentration and the prevailing air mass type. Certain industrial areas were examined and found to be reasonably good sources of giant hygroscopic nuclei. However, a cursory examination of rainfall records does not indicate that they are important in modifying the precipitation patterns in the vicinity of the sources.

1.0 INTRODUCTION

1.1 Hygroscopic Nuclei and Precipitation

The temporal and spatial distribution of giant nuclei in the atmosphere over Illinois was investigated to determine whether their presence was related to precipitation processes occurring in that area. Certainly if giant nuclei exist during periods of precipitation, then the coalescence process must be considered as a possible mechanism in the development of precipitating clouds in the Midwest. The complete lack of nuclei of oceanic origin implies that the coalescence process, if operable in the Midwest, is a result of the presence of giant nuclei of continental origin. Therefore, if the sources of the nuclei can be determined, then anomalies in the rainfall surrounding the sources may be expected.

Many methods, only a few of which will be discussed here, have been developed for investigating atmospheric particulates. Size determinations have been made indirectly by analyzing bulk samples of cloud or fog water for certain constituents and dividing by the number of drops represented in the sample. Work of this nature was first performed by Kohler (1936) who, through titration, determined the amount of chloride present in rime samples collected at a mountain observatory. The corona method was used to resolve the mean drop size.

Several direct measurement methods also have been devised. Woodcock (1952) caught nuclei on glass slides which had been exposed

from an aircraft flown over the ocean. Microscopic measurements and counts were made of the individual particles through the use of an isopiestic method. Twomey (1959) collected particles on spider webs and observed with a microscope the relative humidity at which the solid-liquid phase transition occurred. From this, the chemical composition of the particles was established.

Variations of the chemical spot test analyses described by Feigl (1958) have resulted in other direct measurement techniques. Cadl (1959) made use of "chemical micrurgic" methods in which tedious micromanipulative procedures were employed to add droplets of reagent to particles which had been collected on glass microscope slides or cover glasses by settling, impaction, or thermal precipitation.

Seeley (1952) developed a highly simplified method of spot testing which involved the impaction of particles on a gelatin impregnated with a reagent known to be specific for a certain ion. Lodge (1954) modified Seeley's method by replacing the gelatin with a thin, cellulosic, porous membrane filter. Particulate matter collected on this membrane was assayed by floating the filter on a specific reagent and viewing the reactions under a microscope in order to make size and number determinations of the ions present.

Of the analytical methods surveyed, the one adapted by Lodge seemed to be the most applicable to an air-borne sampling program by which the time and space variations of specific particulates was to be determined. Such applicability was demonstrated by Lodge (1955), when B-17 aircraft flights were conducted by the University of Chicago near Puerto Rico for the purpose of measuring the concentration of chloride particles at various altitudes. Therefore, the membrane filter technique was used throughout this project.

From the results of previous work (Lodge, 1955) a volume of one cubic meter appeared adequate to provide numerous particles for the determination of the number and size distribution. At the same time, a minimum sampling resolution of one sample per five to ten miles was considered desirable. In order to examine more than one constituent simultaneously, it was required that the exposed filter paper surface be large enough to be subdivided into two or more smaller samples with enough particles on each for the determination of the number and size distribution per unit volume.

An examination of the mineral content in rainwater collected simultaneously with particulate samples from the atmosphere would provide information concerning the extent to which the nuclei were used in precipitation production. The relationship between these two parameters is governed by so many variables that nothing more than a very qualitative connection was expected.

2.0 AEROSOL AND RAINWATER SAMPLING EQUIPMENT

2.1 Millipore Filter Tape Sampler

A laboratory experiment was conducted at the University of Illinois Aeronautical Engineering Shock Tube Laboratory to determine the flow characteristics of the membrane filter material to be used in the sampling program. Plastic tubes with two-inch inside diameters were assembled with break-away sections so that discs of filter material could be placed in position to facilitate the measurement of the air resistance at different pressures. Through these tests, it was ascertained that a vacuum of ten inches of mercury with a volume flow of approximately ten cubic feet per minute through a circular filter two-inches in diameter, would satisfy the desired sampling resolution.

Since the data were to be collected under a variety of synoptic conditions using an aircraft to support the sampling device, consideration was given in the development of the instrument to the mounting on the aircraft, power requirements, drag forces on the aircraft, and weight. In addition, it was necessary to provide a means by which the filter material could be automatically supplied while in flight from an unexposed storage area to the exposure area, and subsequently, a means of storing the exposed samples.

A suitable filter medium was investigated first. A membrane filter with 0.45 micron pore size, which was given added strength by the addition of a web of monofilament nylon on one side, was obtained from the Millipore Filter Corporation, Bedford, Massachusetts. While the nylon gave added tensile support to the material it also affected the flow rate; however, the change of flow rate was not a major problem. The optical properties of the nylon made the use of this type of filter paper impossible, since the reaction sites were observed with crossed-polarized light or in a dark-field illumination. It was found that, when viewed through the microscope, many of the reaction spots were indistinguishable in the vicinity of the nylon webbing.

With the cooperation of the Millipore Filter Corporation, standard filter material with 0.45 micron pore size without the nylon backing was supplied on reels which, when fully loaded, contained a usable filter area of 68 linear inches, three inches wide. The edges were taped to provide tensile strength for the automatic movement of the paper during flight. Because the membrane filter is fragile, it was necessary to provide a substantial backing while the ten inches of vacuum was supplied to it, and a stainless steel screen was provided for this purpose. The screen also outlined the exposed area for subsequent laboratory analysis.

With the completion of the preliminary investigations of a suitable filter medium, considerations were given to the physical

design of the device to perform the required functions of advancing the material and storing it while in flight. The sampling equipment consisted of a supply reel for the filter paper, the filter head and vacuum lines, a metering system for the paper advance, take-up reel for the storage of exposed samples, and a paper drive mechanism.

The use of a simple spiral take-up spool permitted the collection of 70 inches of exposed filter paper without contamination of succeeding samples. The paper was pulled from the center by a teflon coated leader paper over a set of rollers arranged in a spiral shape.

The tape sampling system described above is capable of exposing and storing 20 to 30 air-borne samples. A one-cubic-meter sample of the atmosphere can be filtered in approximately two to three minutes, and the transfer from an exposed sample to a fresh area of the filter paper can be made in about 30 seconds.

The tape sampling system was discarded after several flights since it was discovered that the filter paper was contaminated in the process of taping the edges with the vinyl tape. The paper was checked and found to contain chloride particles in large numbers, but none of the other ions that were being examined were found. Therefore, although the system was not satisfactory for the chloride test, it was adequate for testing other particulates. The system would have been adequate if the application of the vinyl tape could have been automated to avoid handling of the filter paper.

The physical dimensions of the equipment in the sampling system dictated a housing of suitable aerodynamic shape approximately one foot in diameter and three feet long. A fiber glass housing was molded in the shape of a cylinder one foot in diameter and one foot long with a cone on either end tapering to a point through a distance of one foot. The vacuum line from the aircraft engine and the electrical wiring to the sampling mechanism were installed in the wing of the aircraft.

The front end of the sampler tube was exposed to the airstream at a point approximately 18 inches below the wing surface and four inches in front of the leading edge of the wing of a Piper Tri-Pacer aircraft. This location eliminated, for all practical purposes, the aerodynamic effects of the wing, contamination effects from the aircraft exhaust, and problems of particle size fractionation due to the aircraft propeller.

An aerometeorograph, for the measurement of temperature, humidity, and pressure, was mounted under the other wing in approximately the same relative position as the aerosol sampler. The data collected with this instrument provided information on the thermodynamic structure of the atmosphere during particulate sampling missions,,

A standard displacement type flow measuring device calibrated in cubic meters was used to measure the volume of atmosphere sampled. Since atmospheric air which was filtered underwent a rapid expansion upon passing the filter, the direct measurement of filtered air was not accurate. The adiabatic assumption was made to determine a correction factor which was applied to the measured volume in order to obtain the true atmospheric volume. Under average conditions of a ten-inch pressure differential across the filter and flying at an altitude of 5,000 feet the correction was approximately 0.75. In other words, of one cubic meter was measured, the true atmospheric volume sampled was approximately three-quarters of a cubic meter.

2.2 Disc Sampler

After the discovery of the contamination problem with the tape sampler, a major modification of it, as described by Semonin and McCrady (1961), was undertaken. A Bausch and Lomb "300" slide projector was adapted to serve as the basic assembly of the aerosol sampler. The filter material used in the sampler was, again, the Millipore filter, but in disc form. This particular filter medium can be obtained in several pore sizes indicating the minimum particle size which will be retained by the membrane.

The filter holders, designed to replace the 35-mm slides, were loaded in the magazine in the laboratory to prevent contamination of the samples, and the magazine was inserted in the aerosol sampler immediately prior to take-off. On all flights one sample was loaded in the magazine and was not exposed to the airstream. The analysis of this control sample was compared to the analysis of the exposed samples, and the results indicated that the unexposed samples were not contaminated during sampling intervals.

The entire modified sampler system was mounted in the same fiber glass structure that housed the previously described tape sampler. The disc sampler was capable of obtaining 20 discrete samples, and with minor modifications, it could obtain a maximum of 40 samples.

2.3 Rainwater Collector

An iron frame was fabricated to support 0.005-inch polyethylene sheets, used as the rainwater collecting surface, four feet above the ground. The surface was rinsed with distilled water just prior to the collection of a sample. The chemical analysis of the rinse water indicated that three washings were required to cleanse the collector of impurities. The rainwater was collected in glass bottles and immediately sealed; chemical analyses were accomplished as quickly as possible after collection.

3.0 CHEMICAL ANALYSIS

3.1 Laboratory Procedures

The spot test techniques used for the determination of the particulates collected were a specific application of those developed and used extensively by Peigl (1958). Seeley (1952) modified certain of the tests using gelatin-covered glass slides that were exposed to an airstream. The particles reacted with the reagent which was impregnated in the gelatin. The reactions were then examined under a microscope for size and number determinations. Lodge (1954) modified this technique and replaced the gelatin with a membrane filter. The filter retained all particles above the pore size of the filter paper. It was subsequently assayed in a manner similar to that described by Peigl.

The tests that were used throughout this research are extremely sensitive; and therefore, only a minimum handling of the filter materials could be tolerated. The loading of a magazine prior to a flight was accomplished in a dust free area of the laboratory using rubber gloves, forceps, and a dental pick. After the filter magazine was loaded, it was placed in a sealed container and taken to the aircraft where it was immediately installed in the sampling mechanism which was sealed from the environmental air. After a flight, the same procedures were invoked in transporting the magazine to the laboratory. The individual filters were removed from the magazine and stored in plastic petri dishes until the tests were performed.

All of the tools and equipment that were used in contact with the filter material either prior to or after exposure were thoroughly scrubbed to prevent contamination of the sample. Filter samples were periodically checked in the laboratory for possible contamination.

3.2 Specific Tests Used

In general the spot tests that were performed were all similar to those described by Lodge (1954), Lodge and Tufts (1956), and Tufts (1959). The filter to be examined was floated on a reagent specific for the ion being sought. After a certain development time, which varied for different constituents, the filter was usually washed of excessive reagent, in most cases dried in a desiccator, and examined under a microscope. The filter paper, once it had been examined for a particular ion, was useless for further tests.

Several conferences were held with Dr. James P. Lodge and Miss Barbara J. Tufts during the development of laboratory techniques.

Many of the difficulties encountered in our laboratory were likewise experienced by Lodge and Tufts.

Three of the seven tests that were investigated proved to be unreliable as indicated in the following paragraphs. Of the remaining four, only two were used extensively since the results of many flight samples indicated that nitrate and calcium compounds were practically nonexistent. The sulfate and chloride tests were used exclusively for the remainder of the program.

Ammonium - The Millipore filter was placed on a blotter saturated with a standard solution of Nessler's reagent. The filter absorbed the reagent by capillary action in a very short time; and the particles reacted in approximately one minute. The filter was then washed three times by floating on distilled water for five minutes per wash. The sample was then placed on a clean blotter and introduced into a desiccator for drying.

The reaction sites appeared as circular groups of orange to pale yellow crystals. The filter was handled rapidly while the reagent was on it, because it picked up ammonia from the air very rapidly. The test faded fairly quickly, and had to be examined within a few hours.

This test was one of the most difficult for obtaining reproducible results over a period of time. The reaction sites were so widely dispersed over the filter that it became impossible to make quantitative measurements of the diameter of the spot. After some effort was made to make the test objective and more definitive, it was noted that reactions were observed on filters that had never been exposed to the atmosphere except during handling in performing the analysis. Therefore, this test was abandoned since it became impossible to make any quantitative determinations of the presence of ammonium in the collected samples as distinguished from reactions that might have been from contamination.

Potassium - The exposed sample was floated on a one percent aqueous solution of sodium tetraphenyl boron for five minutes. The reacted sample was then washed twice by floating on distilled water for five minutes per wash. After the excess reagent was removed by washing, the filter was placed on a clean blotter and placed in a desiccator for drying.

The reaction sites, when viewed under dark-field illumination, appeared as large white granular spots. The growth factor for the potassium test as determined by Tufts (1959) was 10.3.

Further testing of this particular reagent showed that it reacted with certain ammonium compounds; since it was not specific, the test was discarded.

Calcium - The filter was floated on a 30 percent aqueous solution of ammonium ferrocyanide for 15 minutes. The reacted filter was washed by placing it on a thoroughly wetted blotting paper. The filter was moved to fresh areas of the blotting paper until all of the yellow color was removed from it. The sample was then dried in a desiccator.

Calcium salts gave a rather diffuse circular area of fine white crystals which were more sharply visible under polarized light. Extreme care was exercised in handling the filter paper when it was wet with the reagent since the reaction sites had a tendency to run on the filter paper making the counting, sizing, and determination of the growth factor nearly impossible.

Magnesium - The filter was floated for 15 minutes on a 0.3 percent solution of p-nitro-benzeneazo-1-naphthol in 7.5 percent aqueous solution of sodium hydroxide contained in a petri dish. The excess reagent was washed from the filter by floating on distilled water three times at five minutes per wash. The filter was then immediately placed on a microscope slide and viewed. This test did not require drying.

Blue circles were formed and were best observed on the undried filter by reflected light. Since the sites tend to fade on standing they were examined immediately. This test proved to be satisfactory, and the reaction spots had a growth factor of 1.88.

Nitrate - The Millipore filter was floated on a reagent of four percent nitron solution in 10 percent acetic acid for 20 minutes. This test did not require washing, but was dried immediately.

The reaction sites were in the form of stellar radiates. These were best seen under polarized light. The sizing of the reaction sites was somewhat subjective since there was no hard boundary to the stellar radiates. This test worked quite well, and the reactions had a growth factor of $D = 3.73d + 6.29$, where D is the halo diameter and d is the particle diameter.

Sulfate - The sample was floated on a warm, saturated, lead nitrate solution for 20 minutes. The excess lead nitrate was removed by washing the filter twice by floating on distilled water. The reacted sample was then dried by placing on a blotter in a desiccator.

The reaction gave white, circular halos with a coarse center and a diffuse perimeter. These reactions were best seen under dark-field illumination. The growth factor for the sulfate test was 1.68.

Chloride - The sample was assayed by floating the filter on a five percent aqueous solution of mercurous fluosilicate containing 3 drops of 30 percent fluosilicic acid per 5 ml for 5 minutes. The sample was then washed twice by floating on distilled water and dried in a desiccator.

The halos formed were mostly circular. Halos larger than 10 to 15 microns were white, comparatively coarse, highly refractive crystals; those smaller than 10 to 15 microns in diameter usually exhibited a sky-blue color. Since the blue color fades on standing, the halos were viewed within a few hours from reaction time.

The chloride test was by far the most satisfactory that was used, and the results were consistently reproduceable. These halos were best viewed with crossed-polarized light and had a growth factor of 4.73.

3.3 Microscopy Techniques

The reaction spots resulting from the chemical techniques outlined above were, for the most part, easily detected under crossed-polarized light with an optical microscope. In some instances, dark-field illumination was used to provide better contrast for some of the weaker reactions. After considerable time was spent in searching for a single illumination system that would serve to distinguish all of the reactions, it was decided that a phase-contrast microscope was best suited for the purpose.

The phase-contrast illumination was used in a way that would yield a semidark field which provided great contrast for most of the spot test reactions. A Whipple disc inserted in the eyepiece was used to measure the size of the reaction sites. A calibrated stage permitted the control of the area being counted so that there was no problem in counting the same area more than once. A magnification of 300 times was found most convenient for resolving particles in the size range collected. This magnification was achieved through the use of 15-power, wide-field eyepieces and a 20-power objective lens.

Some difficulty was encountered in distinguishing the smaller particle reaction sites regardless of the illumination system used. Certain particles collected from the atmosphere which were not soluble in the reagent remained on the filter paper and were extremely difficult to distinguish from reacted sites when their diameters were less than two microns. Therefore, the particle sizes investigated were restricted to those greater than 1.5 microns in diameter.

After being dried, all the filter papers were treated, except magnesium, with immersion oil which made them transparent, since the

index of refraction for the filter material and the oil is 1.515. It was found that, with care, the slides could be retained for quite a long period of time without impairing their quality. The magnesium test had to be viewed while wet with reagent. Since the paper was opaque when wetted with water, it had to be viewed with surface lighting.

3.4 Rainwater Chemical Analysis

The rainwater samples collected were analyzed for the following mineral constituents:

<u>Cations</u>	<u>Anions</u>
Ammonium	Alkalinity
Hardness	Chloride
Sodium	Nitrate
Potassium	Sulfate

In addition, the total dissolved mineral content of each sample was estimated by converting the 25°C resistivity to parts per million using an empirical factor.

With the following exceptions, the analytical methods used in this study were similar to the methods used by Larson and Hettick (1956) in a previous study made by this laboratory of the mineral composition of rainwater. Chlorides, previously determined by the mercuric nitrate procedure (Clark, 1950), and sulfates, previously determined gravimetrically, were determined by turbidimetric methods. Nitrates, previously determined by the reduction method described in Standard Methods of Water Analysis (191|-6), were determined by the phenoldisulfonic acid method (Taras, 1950).

4.0 RESULTS AND DISCUSSION

4.1 Local Sampling Flights

Seventy flights were conducted in the local area surrounding Champaign, Illinois. Chloride determinations were made from the nuclei collected on 32 flights. Sulfate tests were performed on filters obtained during 37 flights. The results of the chloride analyses for samples collected between 1000 and 3000 feet above the local terrain are given in Table 1. The tabulation of the air mass type was made using the U. S. Weather Bureau daily weather maps. The number of days since the last air mass change presents a qualitative measure of the modification which the existing air mass has undergone prior to sampling.

The last column of Table 1 illustrates the occurrence of precipitation within one day's trajectory of the sampling point. There appears to be little correlation between the air mass type, precipitation upwind, and the chloride particle concentration. Detailed analyses of a limited number of cases are in preparation for submission to a professional journal for publication.

A portion of the filter paper from 37 local flights was tested for sulfate ions. The total number per cubic meter varied from zero to 250. Since the mean was approximately 27, it was apparent that sulfate ions could not be a large contributor to the hygroscopic nuclei population; therefore, the remainder of the samples was not tested.

4.2 Pollution Flights

Local sources of giant, hygroscopic nuclei may have an effect on the rainfall distribution in the area surrounding the source. Peoria, a city in north central Illinois with a population of approximately 113,000, was chosen for study in an effort to determine whether these sources are present. The industrial complex for the Peoria metropolitan area extends from the northern extreme of the city to Pekin approximately 15 miles south. The wind was determined from the 0600 CST radiosonde observation at Peoria, and the flight plan was established from these data. It was found that the only constituent present in the size range which was under investigation was chloride, and this was not in great abundance. Tests for nitrate and calcium compounds were made, but the results indicated concentrations too small to be considered significant. A cursory examination of the long term rainfall patterns in the Peoria area do not reveal any anomalies which can be attributed to local nuclei influence.

Further data of this type were collected near Chicago and St. Louis. An example of a flight made on May 9, 1960, in the Chicago area, is shown in Figure 1. The winds in the sampling area were dominated by a low pressure system on the eastern seaboard. The winds at the 500-foot level in the Chicago area were northerly, from five to ten miles per hour. In general, the sampled air had a trajectory from the New England coast to upper Lake Huron, curving southwestward and then south to the southern Lake Michigan region.

It is noticed, with reference to Figure 1, that the greatest concentration of chloride particles, as shown by sample number one, was observed over the south Chicago area, decreasing northeastward near the lakeshore of midtown Chicago, where sample number two was obtained. Sample number three, taken over Lake Michigan on a southerly flight path, showed a decrease in chloride particle concentration compared with those taken over the city. Sample

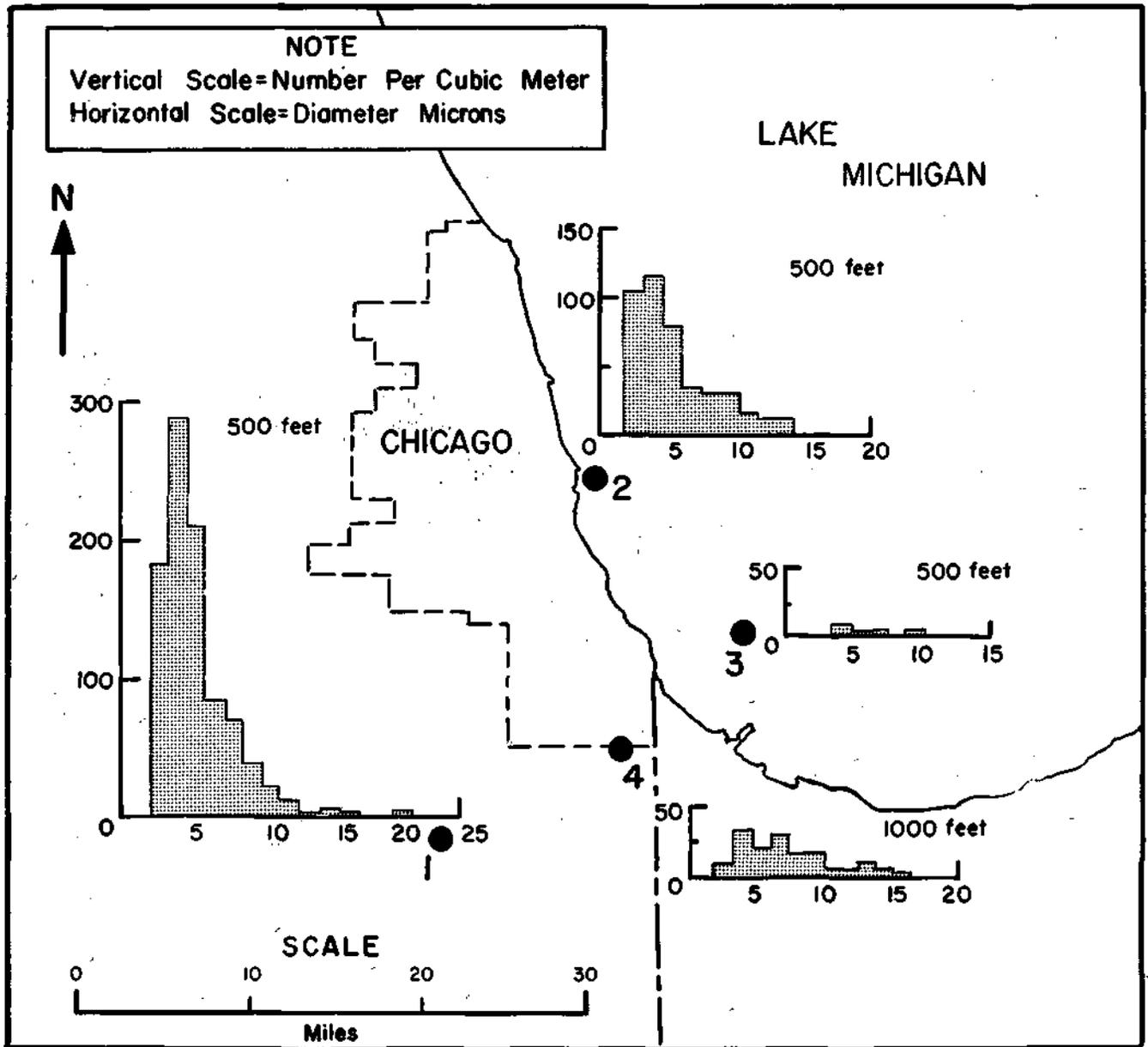


FIG. 1 AN EXAMPLE OF CHLORIDE DATA COLLECTED ON MAY 9, 1960.

number three was comparable to those obtained on the return flight to Champaign, 140 miles south of Chicago. The fourth sample, taken on a southwesterly heading at 1000 feet above terrain, illustrates a decrease with height of the particle number compared with sample number one. Forty miles south of Chicago, the chloride content dropped to nearly zero, illustrating the lack of chloride particles in the air mass.

TABLE 1

Local Chloride Sampling Flights

<u>Date</u>	<u>No. of Cl/m³</u>	<u>Air Mass</u>	<u>Days to last Air Mass Change</u>	<u>Precip. Upwind</u>
7/8/59	0	mT	1	No
7/28/59	758	mT	3	Yes
7/29/59	101	mT	4	Yes
9/25/59	4,139	cP-mT	1	Yes
10/12/59	6	cP	4	No
10/13/59	1,958	cP	5	Yes
10/29/59	6	<u>mT</u> cP	1	Yes
11/2/59	179	cP-mT	1	No
11/3/59	4,111	cP	3	No
1/5/60	351	cP	10	Yes
2/2/60	67	cA	1	Yes
2/16/60	260	cP	3	Yes
2/16/60	63	cP	3	Yes
2/17/60	860	cP	4	Yes
2/17/60	31	cP	4	Yes
2/23/60	399	cA	7	Yes
2/29/60	340	cA	5	Yes
3/1/60	214	cA	6	Yes
3/7/60	200	cA	12	Yes
3/22/60	100	cA-cP	1	Yes
4/13/60	34,426	cP-mT	1	No
4/14/60	86,964	mT	1	Yes
4/15/60	6,110	mT	2	Yes
4/18/60	1,472	cP	1	Yes
4/19/60	357	cP	2	Yes
5/16/60	0	cP-mT	1	Yes
5/27/60	0	mP	1	Yes
6/8/60	0	cP	4	No
6/27/60	6	mT	1	Yes
6/29/60	7	mT	3	Yes
7/11/60	15	mT	3	Yes
7/12/60	0	mT	4	Yes

Changnon (1961) has shown that a rainfall anomaly exists in the Chicago area, However, there are so many possible parameters to influence the precipitation over Chicago that it would be an insurmountable task to separate the effect due to nuclei. However, these flights do show the usefulness and resolution achieved with the sampling system. Using the sampler described, it is possible to discern pollution patterns from industrial sources as well as purposely introduced material that can be used as tracers. With this technique, and employing the appropriate tracing materials, air trajectories under steady conditions can be determined.

4.3 Cross-Country Flights to New Orleans

In an effort to determine the gradient of chloride particle concentration between Illinois and the Gulf of Mexico, two flights were made to New Orleans, Louisiana. The flights were made at an altitude of 5000 feet above terrain, and samples were collected every 70 miles enroute.

The results of one of these flights are shown in Figure 2. The cold front was relatively inactive, with only scattered cumulus congestus, when penetrated enroute to New Orleans. The chloride particle content, however, showed a distinct increase in the smaller size ranges south of the front. The eighth sample was obtained within 20 miles of the New Orleans airport, and a broadening of the size distribution was noted,,

On the following day a flight was made to determine the vertical profile of the particle concentration near the Gulf of Mexico, The size' distribution in the lower 2000 feet resembled sample number eight in Figure 2| but, with an increase in altitude, the larger sizes were not evidenced. The decrease with height of the chloride particle concentration was similar to that noted by Lodge (1955) near Puerto Rico,

The data obtained during these flights are being prepared for publication,

4.4 Rainwater Analyses

The analyses of the rainwater samples collected showed a relationship to the chloride particle content. When the particulate concentration was low or nearly zero, the rainwater chloride content was low. However, the problem of determining the importance of particulates in precipitation from rainwater analyses is compounded by the complexity of individual storm systems. The origin of the moist air entrained by the storm in its initial stages, while of some importance for subsequent chemical analyses, is difficult to determine. It is surprising, in view of the problems

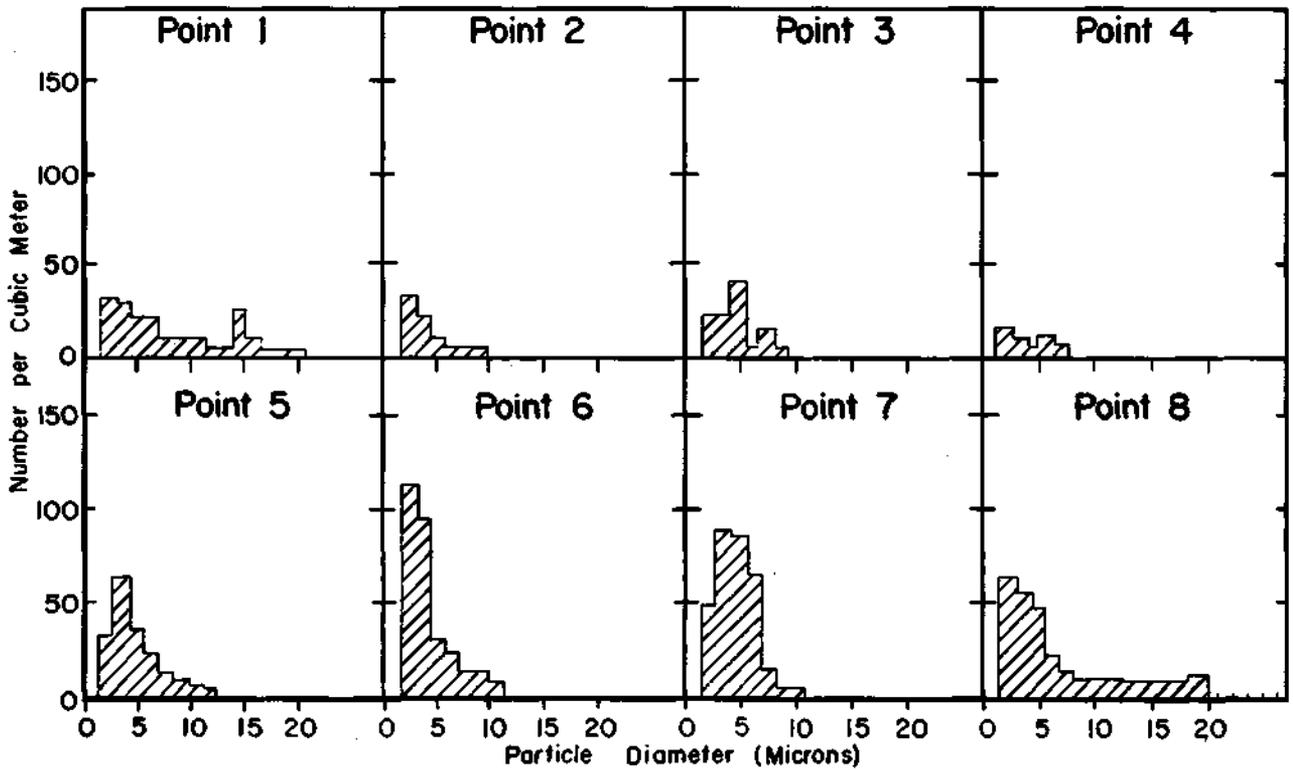
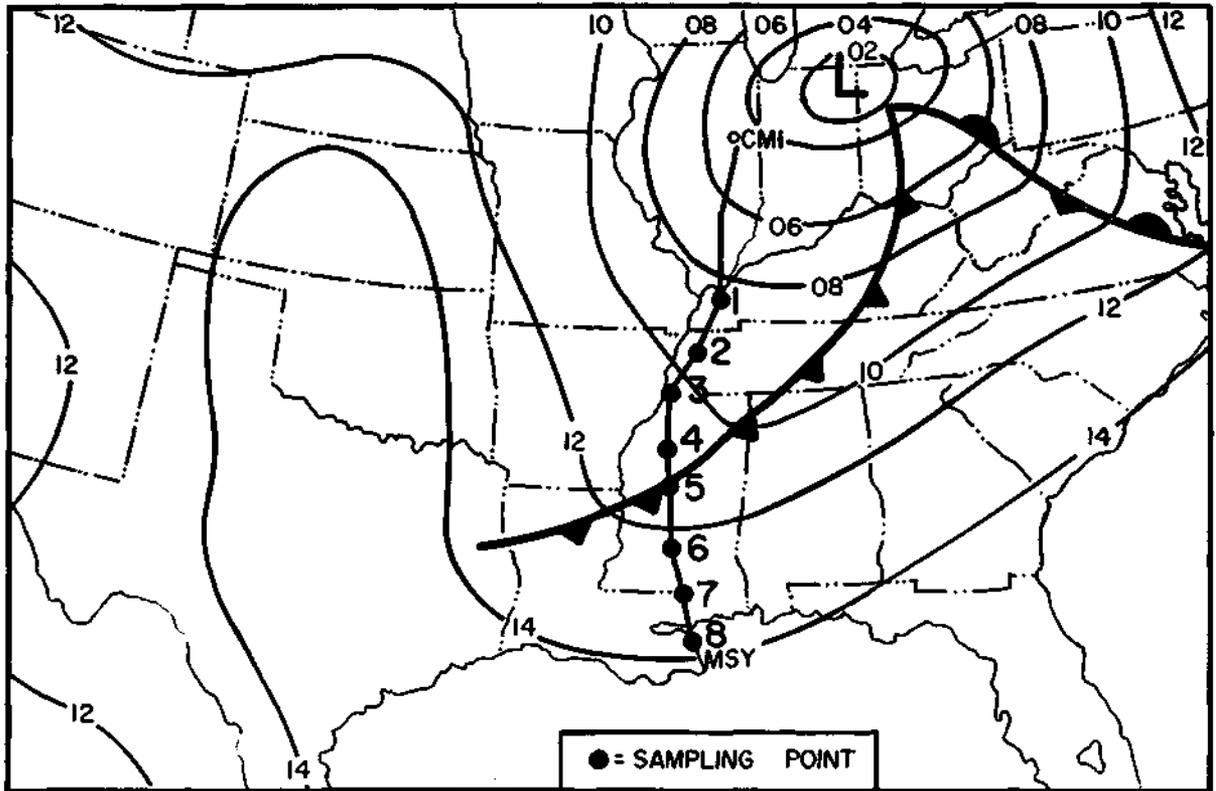


FIG. 2 SURFACE WEATHER CONDITIONS (0600 CST), FLIGHT PATH, SAMPLING POINTS, AND CHLORIDE CONCENTRATION ON JUNE 14, 1960.

TABLE 2

Rainwater Mineral Content Analyses

<u>Date</u>	<u>No.</u>	<u>Cl⁻</u> <u>epb</u>	<u>SO₄⁼</u> <u>epb</u>	<u>NO₃⁻</u> <u>epb</u>	<u>Alk</u> <u>epb</u>	<u>Na⁺</u> <u>epb</u>	<u>K⁺</u> <u>epb</u>	<u>NH₄⁺</u> <u>epb</u>	<u>Hardness</u> <u>epb</u>	<u>Total</u> <u>Mineral</u> <u>Content</u> <u>ppm</u>	<u>Total</u> <u>Anions</u> <u>(-) epb</u>	<u>Total</u> <u>Cations</u> <u>(+) epb</u>
5/16/60	1	4.2	36.0	6.1	5.8	6.1	1.0	5.6	38.2	4.62	52.1	50.9
5/16/60	2	1.1	20.6	5.0	-6.4	1.3	0.3	0.6	11.6	4.98	26.7	20.2
5/19/60	3	2.5	23.1	5.6	-5.0	1.7	0.5	5.6	13.8	5.29	31.2	26.6
5/19/60	4	0.6	21.4	2.9	-9.0	2.6	0.5	0.6	6.6	4.44	24.9	19.3
5/19/60	5	4.2	--	30.9	-47.0	6.1	1.3	12.2	28.6	19.4	Analysis Incomplete	
6/13/60	6	3.7	70.1	18.4	-14.0	1.3	0.8	3.9	56.8	11.1	92.2	78.8
6/13/60	7	3.7	86.5	35.7	-68.6	0.9	2.0	15.0	36.8	19.6	125.9	123.3
6/16/60	8	5.6	51.4	13.2	28.8	5.2	1.5	22.2	64.0	7.61	99.0	92.9
6/16/60	9	3.4	37.6	8.4	5.0	3.5	0.5	9.4	35.0	4.29	49.9	48.4
6/20/60	10	6.5	115.6	45.1	-35.8	2.6	1.5	17.2	110.0	19.3	167.2	167.1
6/20/60	11	2.8	68.4	21.9	-38.0	1.3	0.5	7.2	42.8	16.6	93.1	89.8
6/20/60	12	2.3	70.1	25.8	-48.2	1.7	0.5	19.4	34.4	11.2	98.2	104.2

encountered in these analyses, that even a qualitative correlation was found between rainwater chloride analysis and particulate concentration.

Table 2 summarizes the data collected during the program. The mineral constituents of these analyses support the relationships established by Larson and Hettick (1956). It should be noted, however, that more acidity was found in these samples than in the samples taken by Larson.

The data collection was discontinued because of the complex data reduction problem and the desire to place more emphasis on the air-borne sampling.

4.5 General Discussion

The spatial and temporal variations of the chloride particulate content of the atmosphere over Illinois do not suggest a very strong relationship to the precipitation process. In general, the chloride content is quite low over Illinois with periodic counts of several thousand per cubic meter. However, the fluctuations in the number concentration cannot easily be related to specific synoptic parameters. The local sources of chloride nuclei are nearly as potent as the maritime sources. Therefore, the importance of these nuclei in the precipitation process would be detected most easily by examining the rainfall distributions near industrial areas. Work is currently underway within the organization to determine the possible effect of urban areas on rainfall patterns.

Radar observations in 1960 indicate that there were many first echoes below the freezing level observed by a CPS-9 radar located at Champaign, Illinois. These observations suggest that a coalescence mechanism is active in Illinois. On two of the days when warm first echoes were observed, chloride nuclei counts were obtained; and on each of the days, the giant nuclei numbered approximately 25 per cubic meter just below the cloud base level. Thus, It seems likely that the coalescence process in Illinois is a result of a something other than giant chloride nuclei originating over the Gulf of Mexico.

During dry periods of five days or longer over Illinois, Semonin (1960) found that in many cases the cloudiness and instability were insufficient to produce precipitation. The data from this program further suggest that there may be insufficient giant-nuclei to initiate the coalescence mechanism in Illinois. Therefore, further work is being directed toward examining in greater detail additional precipitation mechanisms in Midwest clouds.

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PUBLICATIONS

"A Semiautomatic Air-borne Aerosol Sampler," R. G. Semonin and R. E. McCrady, accepted by the Journal of Meteorology for publication.

"On the Influx of Chloride Nuclei over Illinois," R. G. Semonin and D. D. Watson, in preparation for publication.

"The Distribution of Chloride Nuclei over Illinois," R. G. Semonin, in preparation for publication.

"Local Sources of Giant Chloride Nuclei," R. G. Semonin and D. D. Watson, in preparation for publication.

"Observation of Cloud Formations Relative to an Industrial Site," J. W., Bullock, in preparation for publication.

Master's Thesis, R. E., McCrady, in preparation for submission as partial fulfillment of the degree of Master of Science from the Agricultural and Mechanical College of Texas.

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