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THE UNIVERSITY OF MICHIGAN

COLLEGE OF ENGINEERING
Department of Meteorology and Oceanography

MASTER

Progress Report No. 7

RAIN SCAVENGING STUDIES

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TABLE OF CONTENTS

Section

LIST OF TABLES

LIST OF FIGURES

Figure Page

ABSTRACT

In 1970 analytical work on samples collected in 1969 was completed, and some areas of analytical technique for the neutron-activation measurement of indium were worked out, and the procedures published.

Field experiments were conducted during the period 15 May - 30 June in the area of Clinton, Illinois, using the Illinois State Water Survey rain-gauging network augmented by our own samplers to a sampling density of about 1 per 3 sq. mi. Sequential sampling was done at the University of Michigan base station, and, by means of automatic samplers supplied by Argonne National Laboratory, at three additional stations. In addition to the aircraft services of Atmospherics, Inc., an airplane and crew were supplied to the project by the National Center for Atmospheric Research.

Six tracer experiments were conducted and samples were collected from a seventh storm system that passed the area at night. Although analysis has begun no case study is ready for presentation at this time. Preliminary results for the case of 1 June 1970 are presented.

I. 1970 FIELD PROGRAM

A. Schedule and Personnel

The agreed period for field operations under Project ITREX was 15 May - 30 June. UM personnel, vehicles and instrumentation arrived at the field site on 11 May for the preparatory phase. Personnel were as follows:

Professor A.N. Dingle, director
Dr. K.S. Bhatki, research radiochemist
Mr. R.E. Crabtree, research engineer
Mr. Y. Lee, graduate research assistant
Mr. J. Fairobent, undergraduate ass't in research
Mr. J. Goll, undergraduate ass't in research
Mr. L. Krupnak, undergraduate ass't in research
Mr. R. Osburn, undergraduate ass't in research

Vehicles leased from UM car pool were:

2 Ford club-wagons
1 Ford 4-door sedan

The crew and equipment were transported to the field program headquarters at Clinton, Illinois by means of these vehicles on 11 May. Headquarters was established at

Town and Country Motel
Highway 54 West
Clinton, Illinois 61727

and the field research station was located as in 1969 at the Country 4-H Fairgrounds, Highway 51 North.

Repairs to the station structure, installation of equipment, calibration, testing, preliminary layout of sampling networks, and preparation of sampling equipment were completed by 15 May. Following this date, as weather and other circumstances allowed, meetings with the other ITREX crews were convened to complete our collaborative arrangements, work out procedural details, etc.

It was agreed that ISWS crews would set and retrieve samples at 53 stations near Clinton, whereas the UM crews would supplement that network, having a density of about 1 station per 9 square miles, by setting an interspersed network of about 1 station per 9 square miles, by setting an interspersed network of about 100 stations so as to increase the sampling density to 1 per 3 square miles or so. The resulting network of stations is shown on the network master map for 1970 (Figure 1 a and b).

Field operations were conducted as previously outlined (see Report No. COO-1407-33).

B. Instrumentation

The complement of instruments for the project has been discussed in prior reports, e.g., COO-1407-33, Dec., 1969. For the 1970 field program a few modifications were introduced

1. Hi-volume air sampler, supplied by Argonne National Laboratory (ANL) for use at the base station. This sampler is shown in Figure 2. Air is drawn through the filter (papers supplied by ANL) at the intake (A), at the rate of 40 ± 3 liters per min. Samples were taken for 1 to 3 hours depending upon circumstances.
2. Andersen sampler using 7 impaction stages with an exhaust line filter (stage 8) and Gelman pump. Experience showed that the Andersen pump would not operate continuously for 1 hour or more, nor would it maintain the required 1 cfm flow rate. The larger Gelman pump maintained an adequate flow rate and enabled the use of Whatman #41 filter paper in the eighth sampling stage. This sampler [shown in Figure 3] was usually operated in coordination with the hi-vol sampler so that the size-distribution data gained could be regarded as complementary to the analytical results from the hi-vol

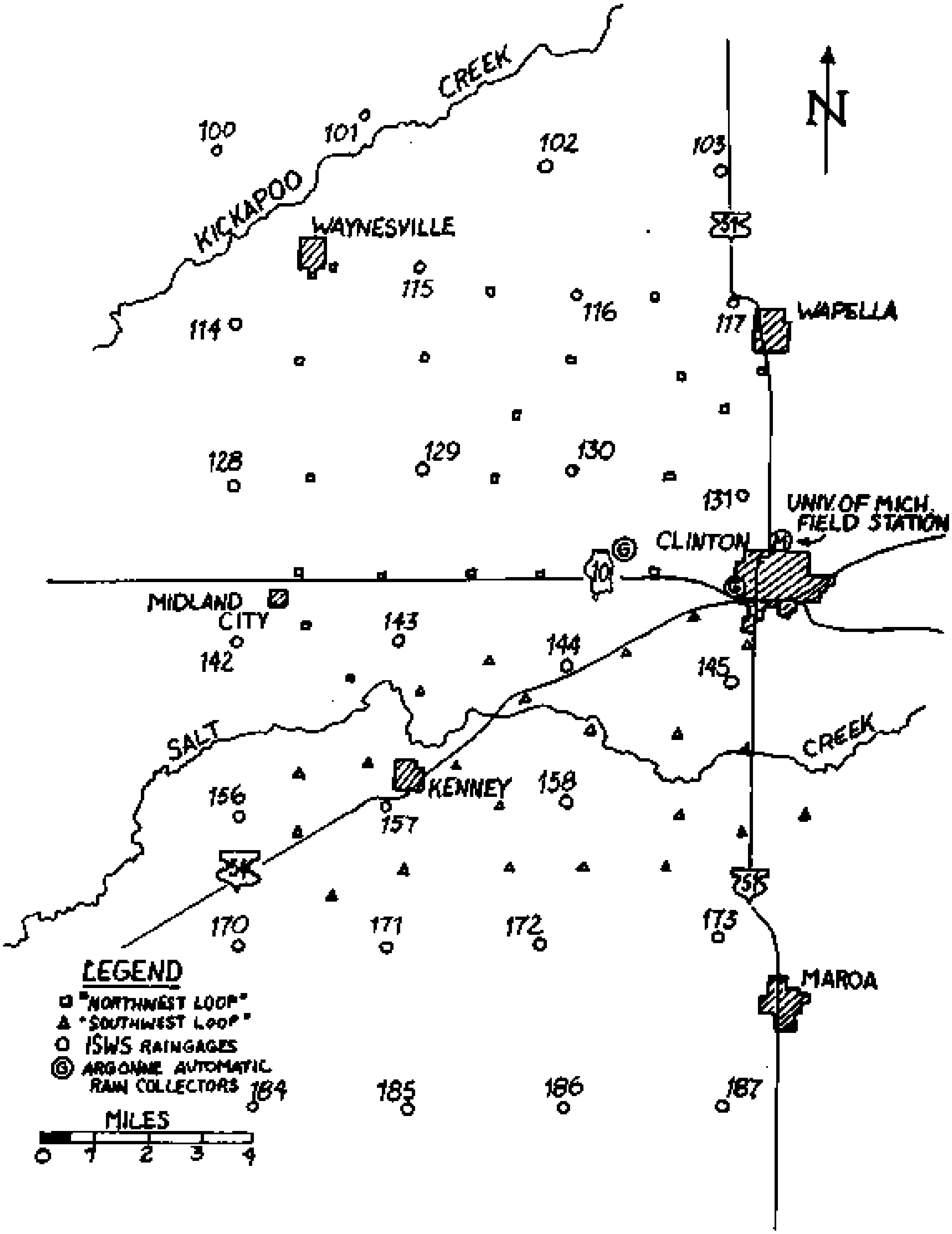

Figure 1a. Network master map of Clinton, Illinois area in the spring of 1970 showing the Northwest and Southwest "loops".

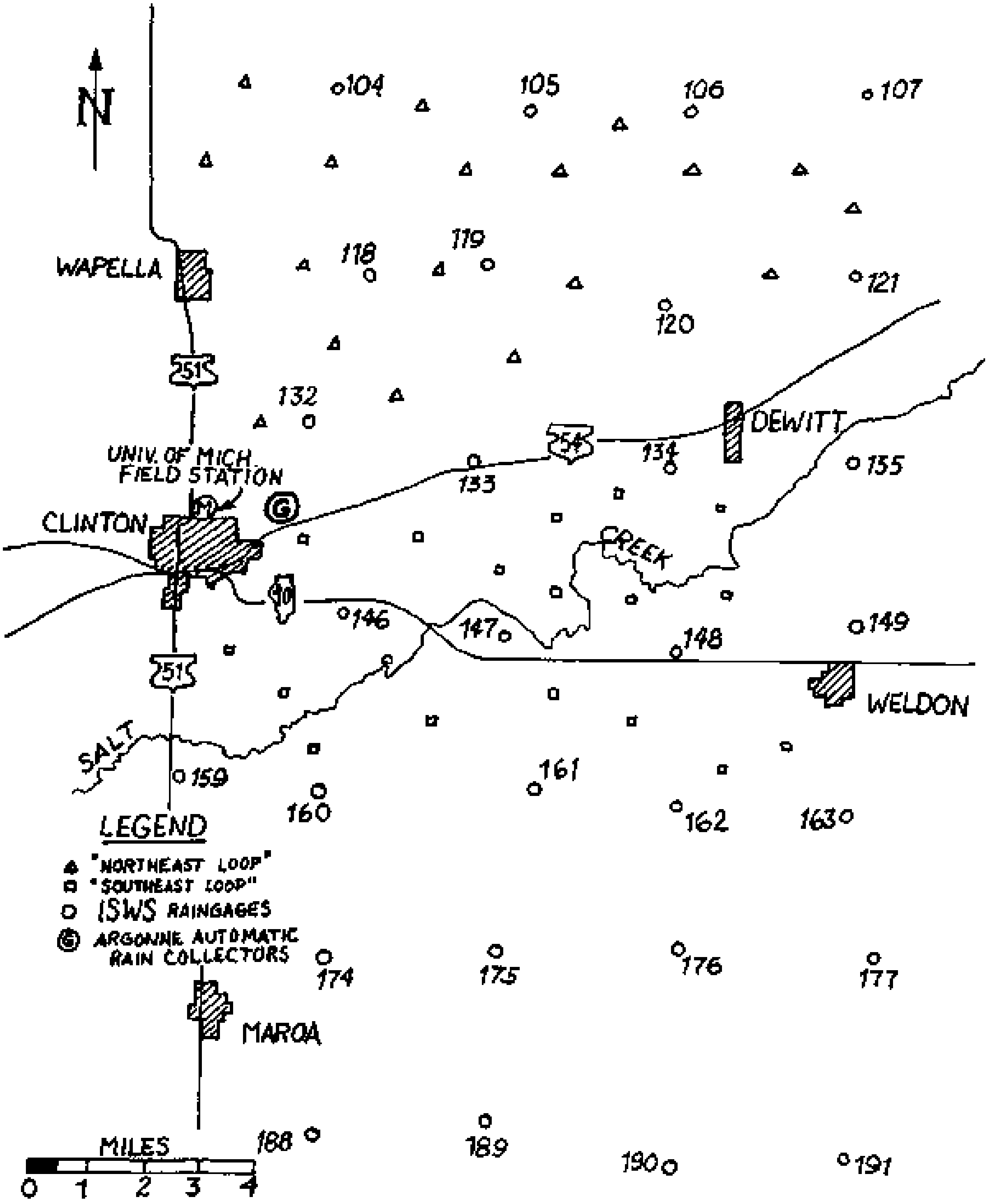

Figure 1b. Network master map of Clinton, Illinois area in the spring of 1970 showing the Northeast and Southeast "loops".

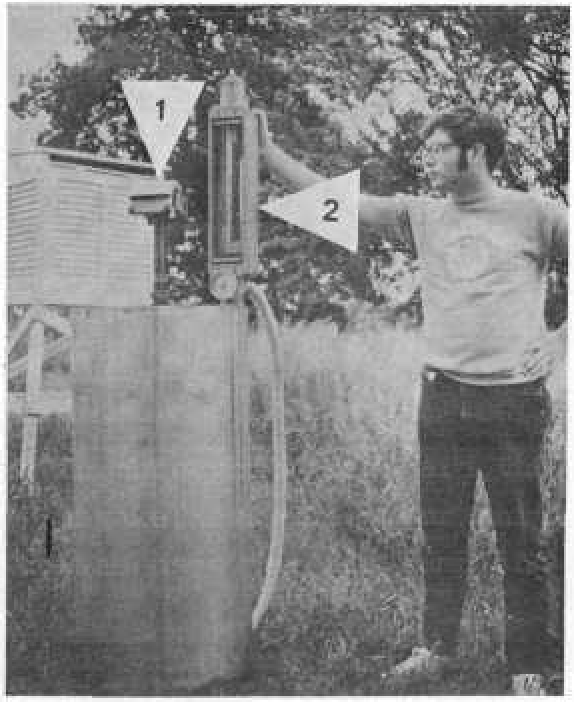

Figure 2. Hi-volume air sampler, supplied by Argonne National Laboratory for use at the base station. Indicated in the photograph are the (1) intake and (2) flow rate meter.

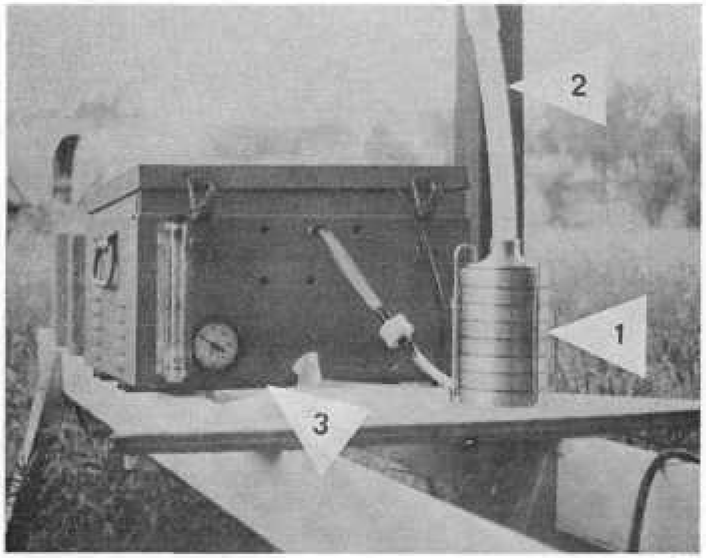

Figure 3. Shown in this photograph are the (1) Anderson sampler, its (2) intake, and the power unit a (3) Gilman pump. This setup was used for air sampling at the base station.

sampler.

3. Three automatic sequential rain samplers, developed by Dr. Gatz and supplied by ANL for three stations, west, north and east of the base station (see map, Figure 1 a and b). These samplers are described by Gatz elsewhere.

C. Aircraft

As agreed prior to the launch of Project ITREX, aircraft support for the project has been funded via ISWS. This funding provided for one contractor-supplied and operated airplane, as follows:

1. In 1969 a Cessna 206 was supplied with instrumentation, pilot and observer by Weather Science Inc., Norman, Oklahoma. The flare-capacity of this unit was limited to 14, and its single engine represented a further limitation on its ability to serve the project needs;
2. In 1970 a Piper Aztec plane was supplied with instrumentation, pilot and observer by Atmospherics, Inc., Fresno, California. The flare-capacity of this unit was 24, and its twin engines provided an improved safety factor and flexibility for the operations.

Reports on the operations of these aircraft have been submitted by ISWS. From our viewpoint, the service was more adequate in 1970 than in 1969, the contractor collaboration closer.

In view of the fact that experiments requiring two aircraft were developed in our designs, and the budget provision was not sufficient to hire two contract aircraft with crews, etc., the writer approached the Aviation Facility of NCAR in 1968 to see whether aircraft support from this quarter might be obtained. The result of that inquiry was that the NCAR plane was made available to the project but not during our planned field program in 1969. It was therefore used by the project to support Dr.

Gatz's work in July-August.

Our request for the 1970 expedition therefore received some degree of preferred treatment, and NCAR provided the Queen Air 304D to the project for the period 15 May to 5 June. Unfortunately, the ISWS attempt to field Li flares led to problems: the Li flares popped, denting the skin of 304D and alarming the NCAR ground and air crews. Thus 304D arrived late (17 May) and reluctant to burn Li. This reluctance was quite reasonable under the circumstances, but we were nonetheless able to place Li into a few rain-producing systems.

D. Field Data Acquired

The sampling program was continued in basically the same design as that of 1969 except for minor changes in the network sampler locations. The complement of instruments and samplers was augmented somewhat as noted above. The tabulation of rain and air samples reflects these changes. Table 1 gives a summary of the numbers of rain samples collected, and Table 2 a summary of the air samples.

Additional field data include all those from the recorders operated at the base station, verbal comments, data acquired by the two project aircraft, and data recorded at the ISWS operations base in terms of predictions, alerts, etc., and weather radar records. Of these, the data obtained at the U of M station are tabulated in Table 3.

Table 1. Summary of Rain samples collected by University of Michigan during Project ITREX 1970.

Date	Type of Sample	DISTRIBUTION				
		University of Michigan			ISWS †	ANL°
		In*	Pb*	Ra-P*	atom. absrpt.	Activation
24 May	Whole storm	50	22	6	27	
	Sequential	15				
29-30 May	Whole storm	14	14	9	14	
30 May	Whole storm	37	17		17	
	Sequential	16	10	7		
1 June	Whole storm	124	123	77	128	
	Sequential	70	76			60
14 June	Whole storm	103	80	13	82	
	Sequential	12	6			
15 June	Whole storm	93	93	20	93	
	Sequential	72	143	15		121
20 June	Whole storm	59	60		60	
	Sequential	9	4			
TOTALS		674	638	147	421	181

* U of M analytical procedures give indium (In), lead (Pb), radioactivity and pollen (Ra-P) amounts

† ISWS analysis gives a number of elements by atomic absorption spectrometry

° ANL analysis uses neutron activation for a number of elements

Table 2: Summary of Air Samples collected by University of Michigan during Project ITREX 1970

Date 1970	Rotobar	Dry fallout	Hi-Vol (Brar)	Impactor 8-stages
May 19	2	✓		
20	2	✓		
21	6	✓		
22	6	✓		
23	4	✓		
24	6	✓		
25	2	✓		
26	2	✓		
27	2	✓		
28	1	✓		
29	2		2	1 set
30	6		4	2 sets
31	2		2	1 set
June 1	2		2	1 set
3	2			
4	2			
5	2			
6	2			
8	2			
9	2			
10	2			1 set
11	2		2	1 set
12	2		2	1 set
13			2	1 set
14	2		2	1 set
15	2			
24			2	1 set

Table 3: Summary of field data acquired by University of Michigan during Project ITREX 1970.

A. Magnetic tape record dates and times (CDT):

date	time tape on	time tape off
23 May	15:04	16:32
24 May	18:04	19:34
29 May	11:15	13:32
29 May	17:15	17:19
30 May	16:48	18:25
31 May	15:58	16:36
1 June	15:58	18:47
14 June	13:37	16:49
15 June	14:59	16:21

1. Event Channel, AM #1
   Tipping bucket raingauge
   Wind speed at 12 ft level
   Sequential sample timing and identification
2. Audio channel, AM #2
   Radio transmission and reception notes
   Verbal observations during operations
   Time checks, WWV
3. Raindrop-size and count, FM #1
4. Wind-direction, 12-ft level, FM #2

B. Other Data

1. Sample weights, distribution to collaborators, identification, etc.
2. Maps of network with identification of stations
3. Weighing rain gauge charts
4. Barograph records
5. Hygrothermograph records (Stevenson screen)
6. Mobile unit records and comments
7. Project journal and log book
8. Records of air sample dates, periods, volumes, etc.
9. Observations of pollens in season, specimens, counts identification.

Compilation of synoptic weather information, preparation of soundings, and preparation of sectional weather maps from Service A teletype data are under way. Radar data operational details from the project weather monitoring center and from the airplane records remain to be procured. These data are not presented here, but will be prepared for publication together with the individual case studies.

The eminently successful season, as indicated by the above tabulations, has provided us an enormous inventory of samples to analyze. We have established priorities for the In analysis so that the data for each case will be complete before we turn to analysis of samples for the next one. Top priority was given to the experiment of 1 June 1970 because this was the best, although not perfect, two-tracer experiment.

Following, in order of analysis priority are brief descriptions of the most promising experiments.

<u>1 June 1970</u>

The synoptic situation is shown in Fig. 4 a and b. The day was overcast with intermittent light shower activity all

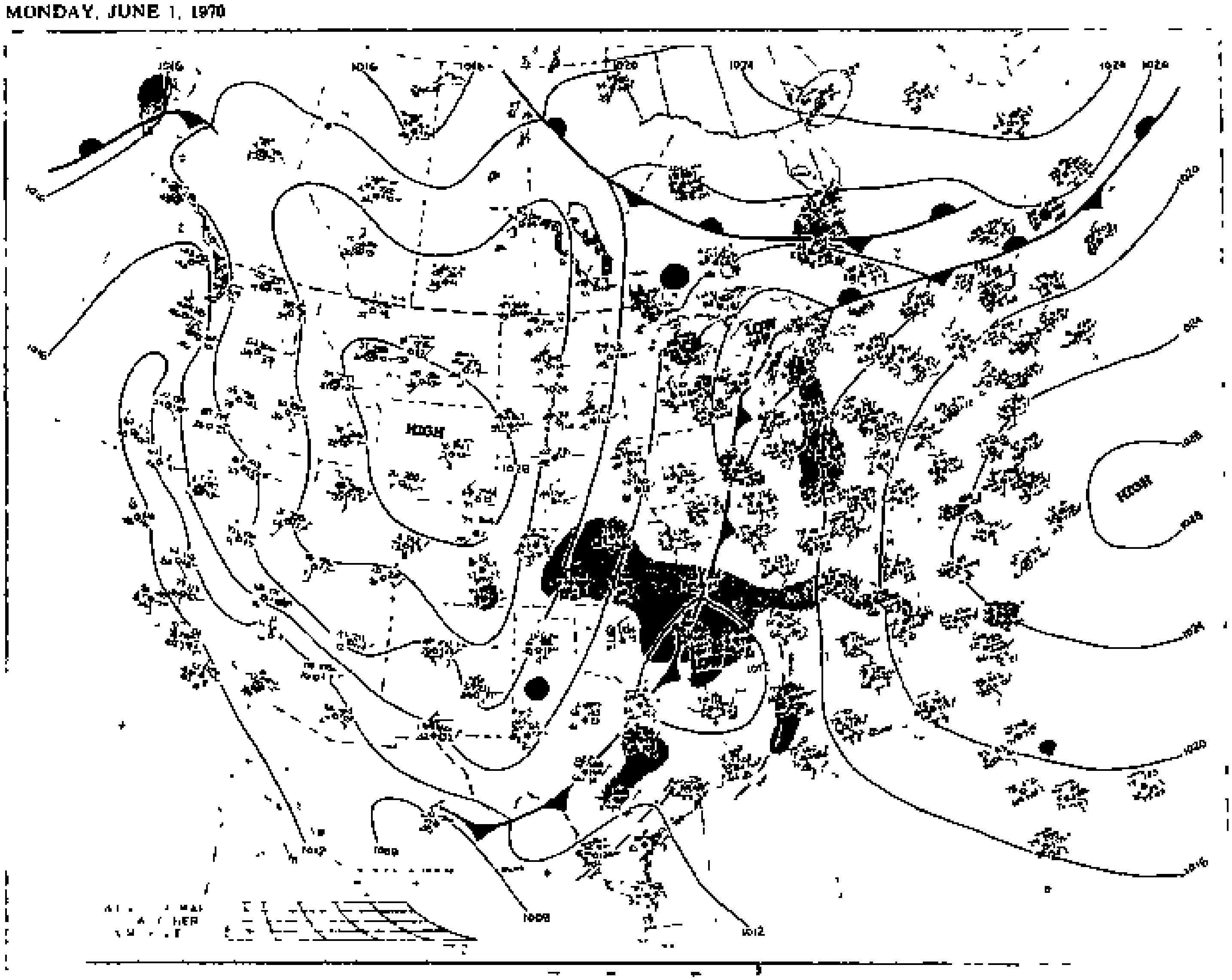

Figure 4a. Weather map at 0700 CDT. Surface map for 1 June 1970.

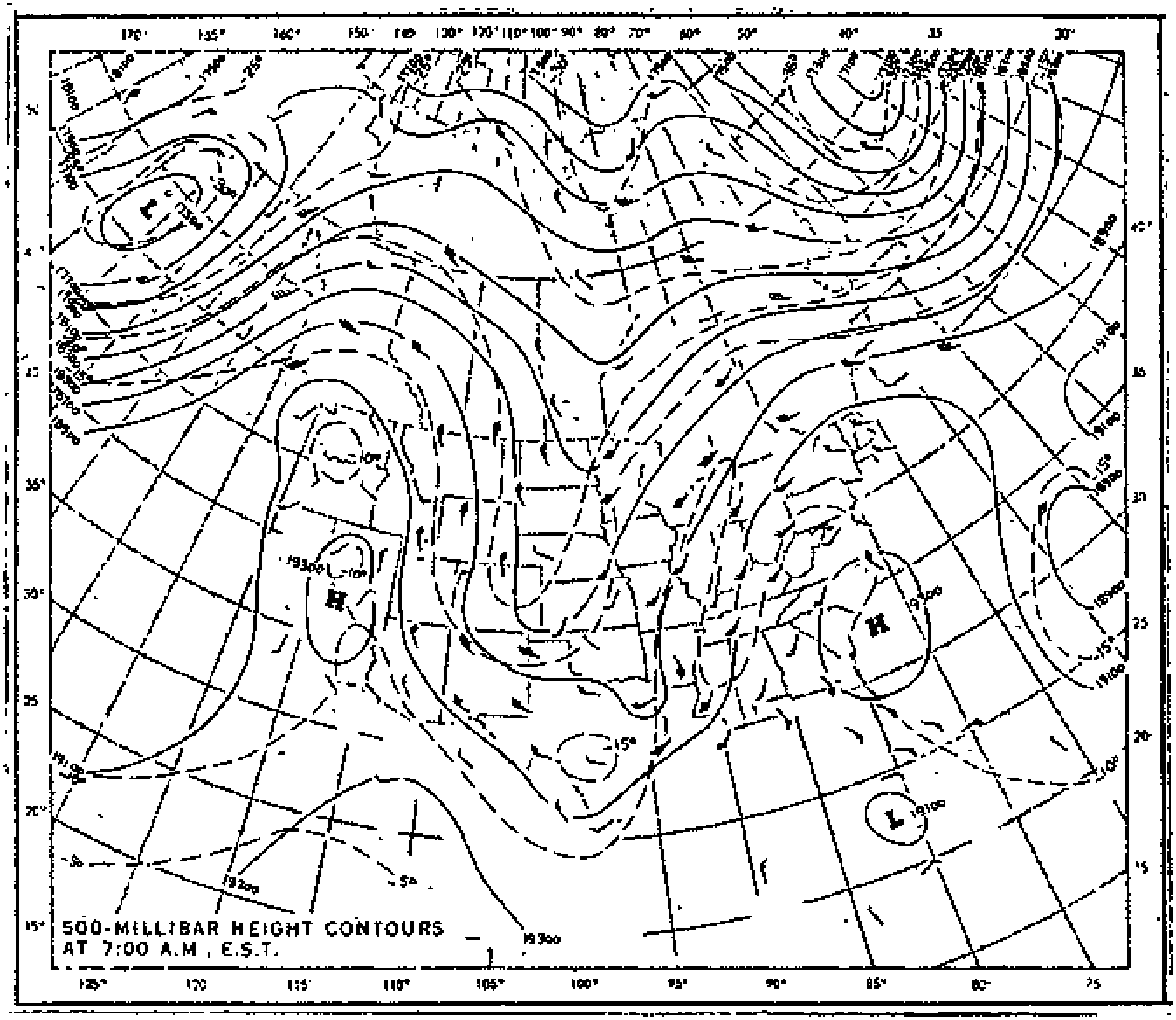

Figure 4b. Weather map at 0700 CDT. 500 mb map for 1 June 1970.

day. This activity reached a climax late in the day as the cold front approached, and radar echoes showed some promise of more organized convective cells. Both aircraft were directed to operational "boxes" at 1414 but suitable cells were not located until much later.

In Burn:
At 1639 the Aztec began an indium burn (12 flares) in an updraft of 300-500 ft per min at about 1900 ft altitude. A track of about 10 n. miles was flown (see Fig. 12) until the flares were consumed at 1644.

Li Burn:
At 1637:50 the Queen Air ignited four Li flares in the tops of confused clouds near 16,000 ft MSL. Immediately after ignition, Chicago FAA traffic control ordered the aircraft to descend to 8,000 ft. This order could not be ignored. Shortly after the descent began, the Li flares "popped".

Upon inspection afterward several dents were observed in the lower surfaces of the rear fuselage, tail and elevator of the aircraft from impacts of large pieces of fragmented flare material.

The two airplanes did not introduce their tracers into the same cloud because of the circumstances. The cloud field was simply too confused for good aircraft coordination to be feasible. Preliminary patterns of Li deposition, and results of the In analyses indicate some interesting points for investigation nonetheless (see below).

The ground-based operations obtained a full complement of data and a comprehensive sample array (Table I, and below).

<u>15 June 1970</u>

Scattered showers by noon, organizing into lines in the afternoon, were forecast for this day (weather maps, Fig. 5 a and b). Strong cells developed throughout the area by 1300. Aztec 178A carried both In and Li flares and at 1520 2,000 ft MSL, ignited 8 In and 2 Li flares, followed by 2 more Li flares ignited at 1522, the entire burn ending at 1526. The navigation situation was difficult because of heavy overcast, strong updrafts, and considerable electric activity.

This storm produced 4.36 in. of rain at UM base station in about 45 minutes.

Circumstances eliminated both drop-size measuring instruments from activity, but otherwise good data were procured at ground level. We shall be especially alert to dissimilarities in the In and Li deposition patterns.

20 June 1970

A relatively diffuse rain situation provided the opportunity to do a "below-cloud-scavenging" experiment. Weather maps are given in Fig. 6 a and b. Here the airplane (Aztec 178A) carried 4 Li and 1 In flares at a level of about 4,000 ft in stable air. The aircraft flew one straight W-E track, approximately normal to the air flow, and sampling was conducted along a N-S line. A direct observation of washout was sought. The results should prove most interesting. In the flare burn sequences, the In flare burned for the 5-min period, and the Li flares were ignited in sequence. Three of the Li flares popped.

14 June 1970

The indices indicated widespread instability in our area on this date, so alert status was declared. Weather maps are shown in Figure 7 a and b. Aztec 178A carrying 12 In flares ignited them at 1416 in generalized cloud and fog with no clear resolution of cells possible from the aircraft. Upon

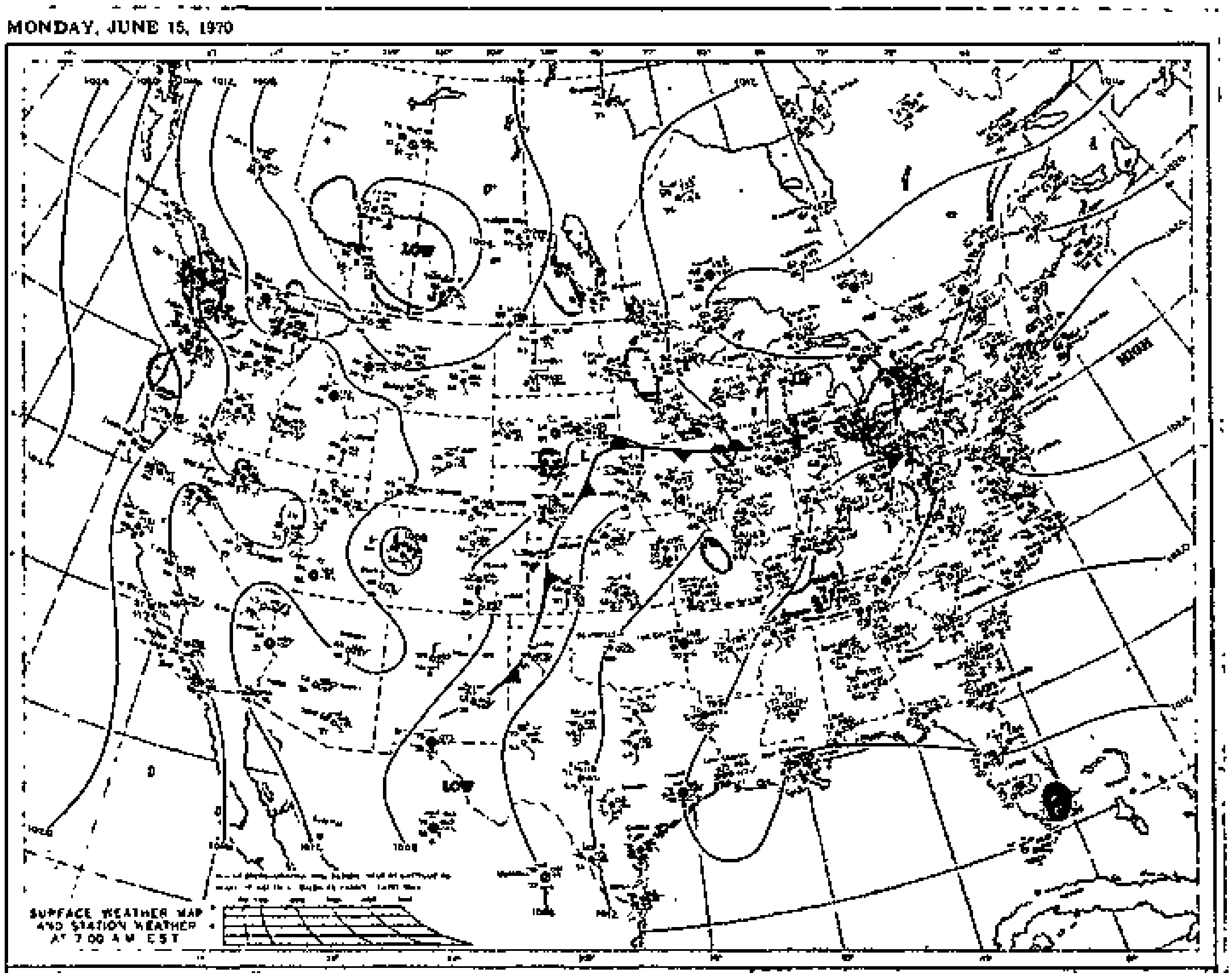

Figure 5a. Weather map at 0700 CDT. Surface map for 15 June 1970.

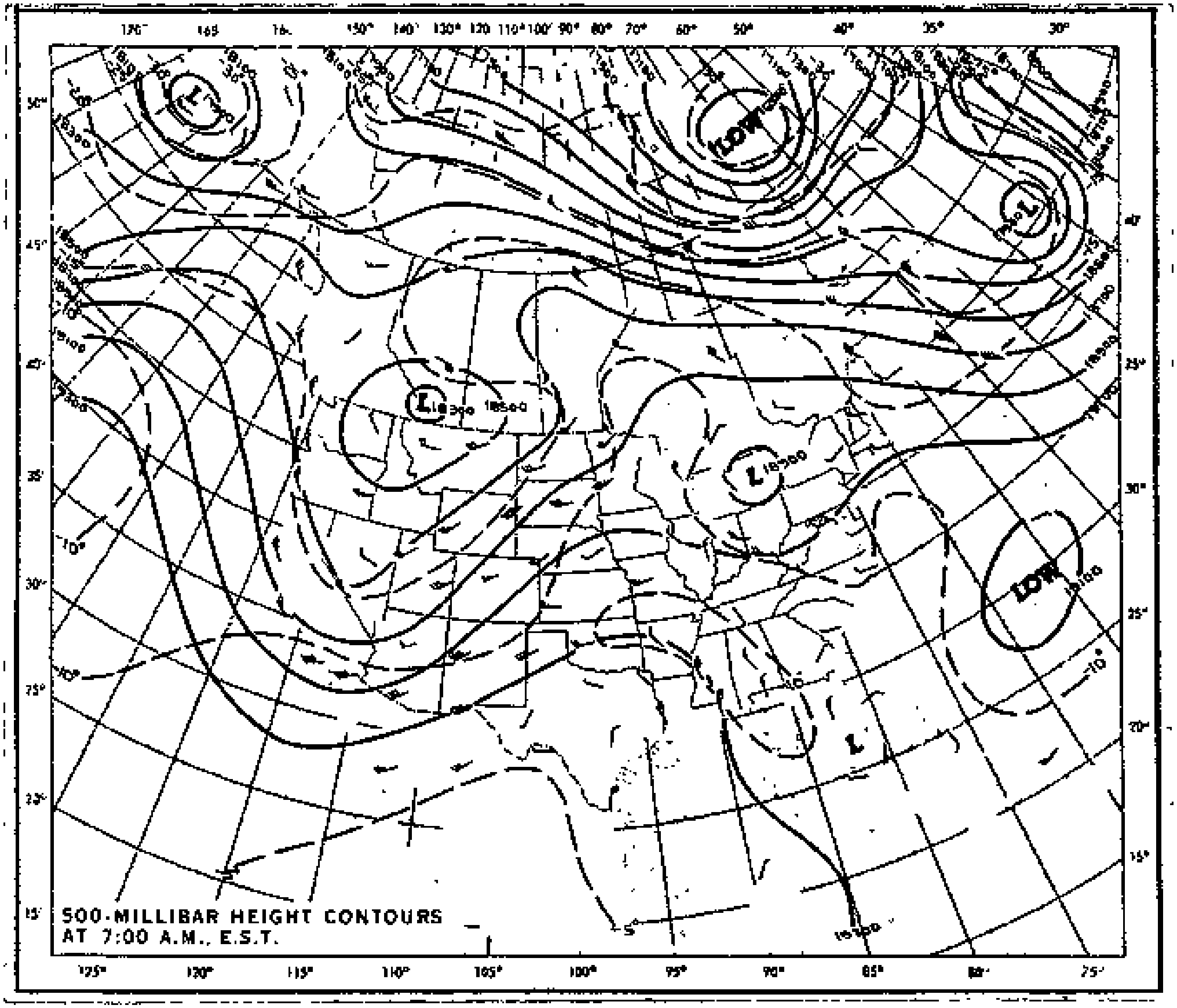

Figure 5b. Weather map at 0700 CDT. 500 mb map for 15 June 1970.

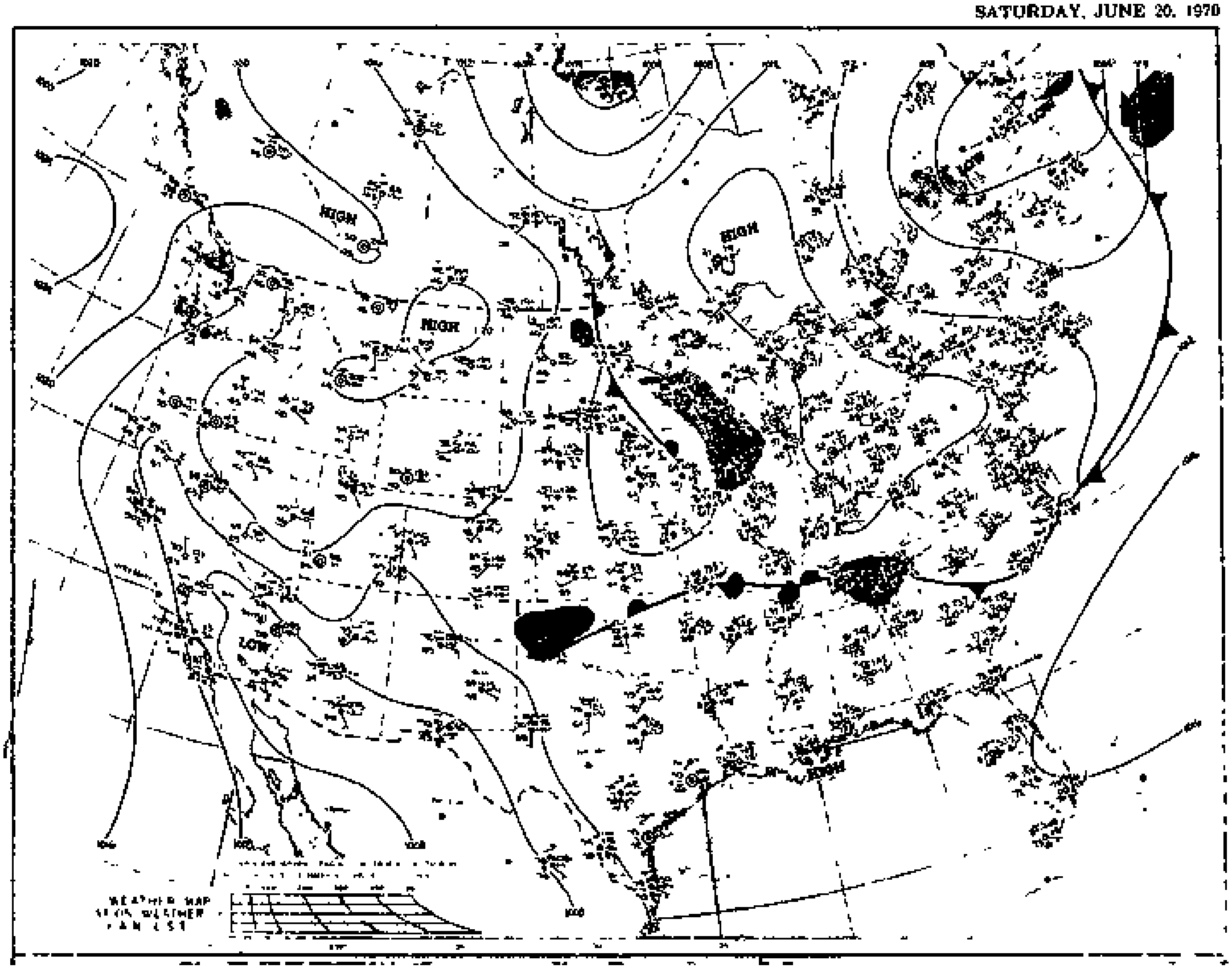

Figure 6a. Weather map at 0700 CDT. Surface map for 20 June 1970.

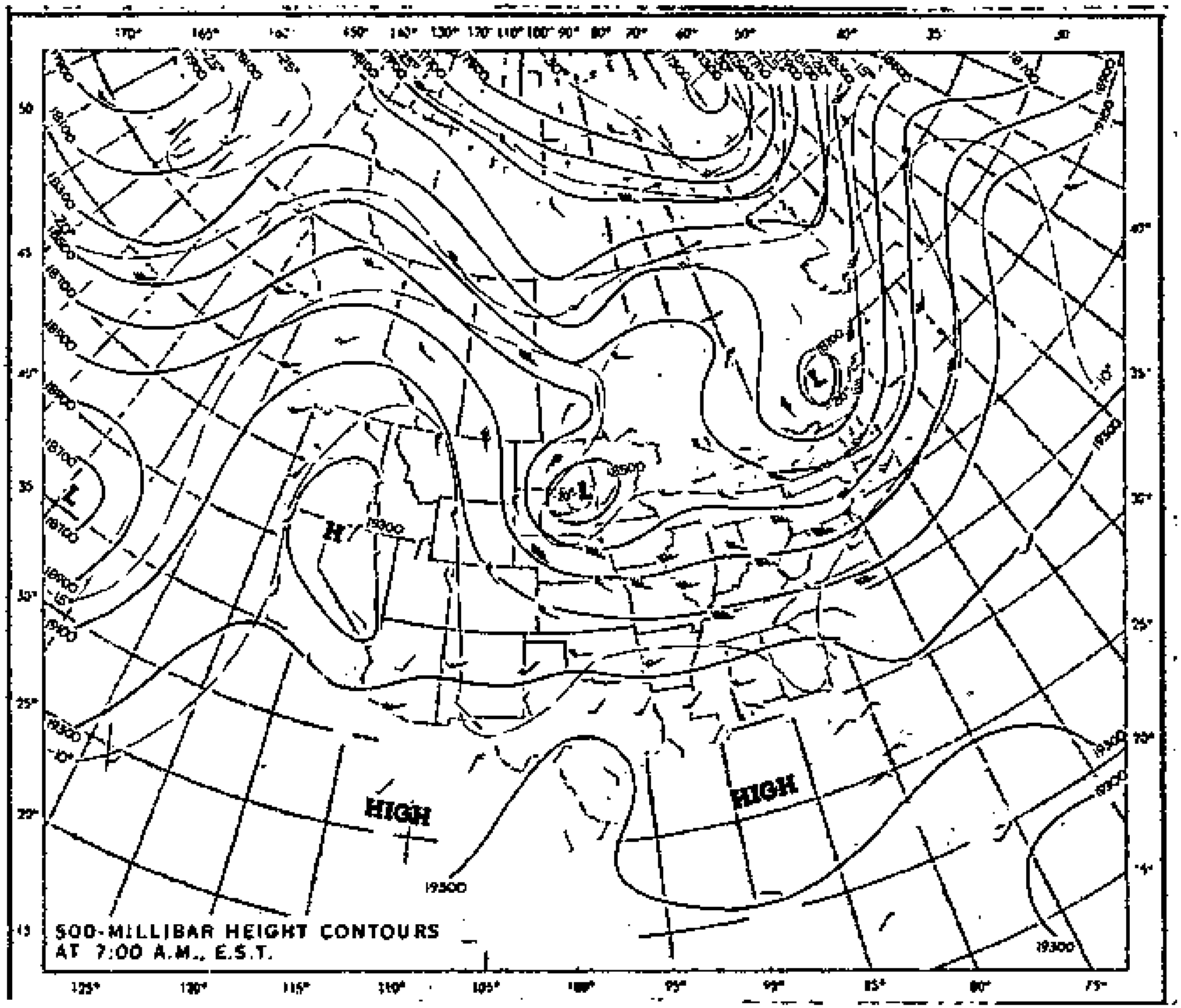

Figure 6b. Weather map at 0700 CDT. 500 mb map for 20 June 1970.

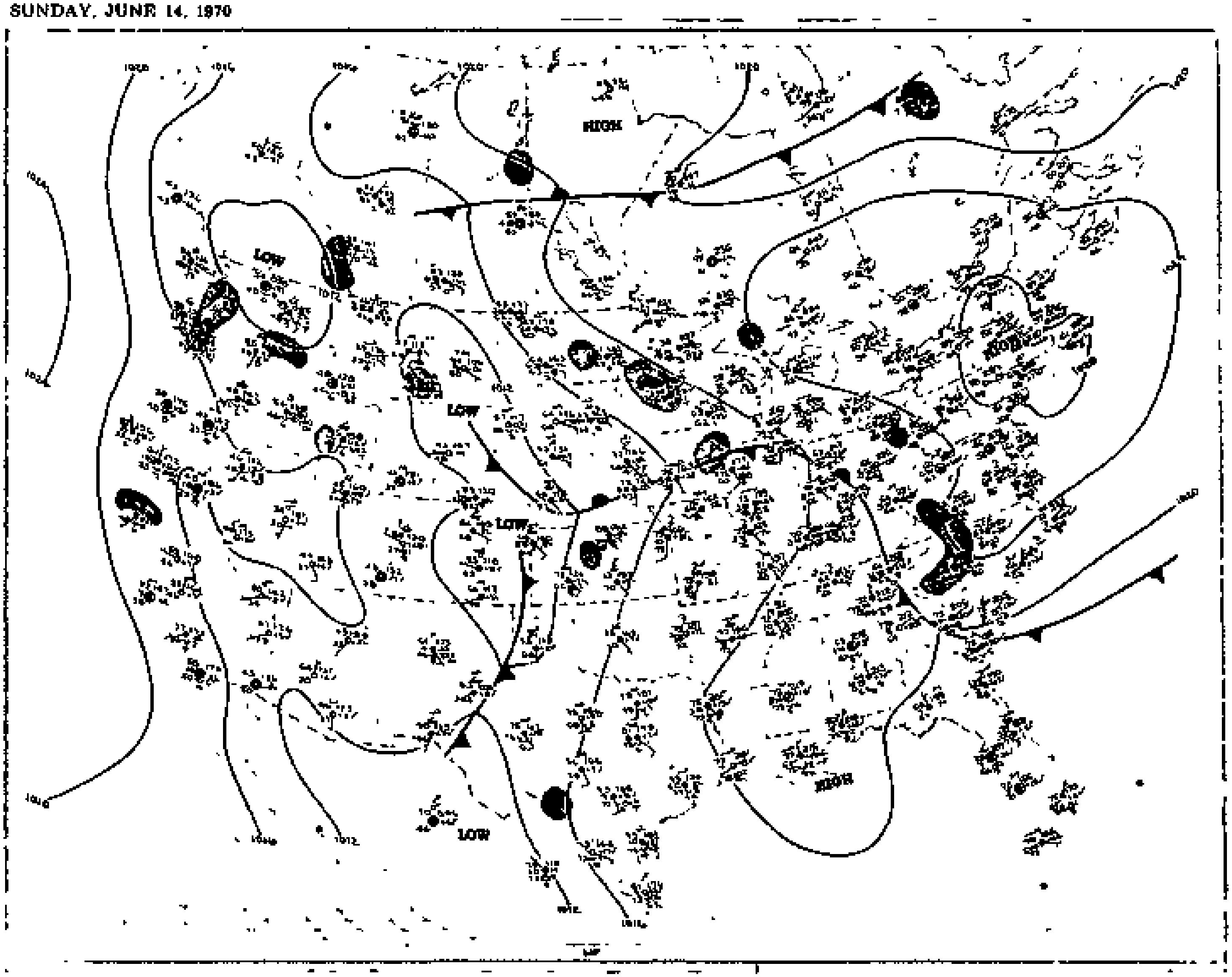

Figure 7a. Weather map at 0700 CDT. Surface map for 14 June 1970.

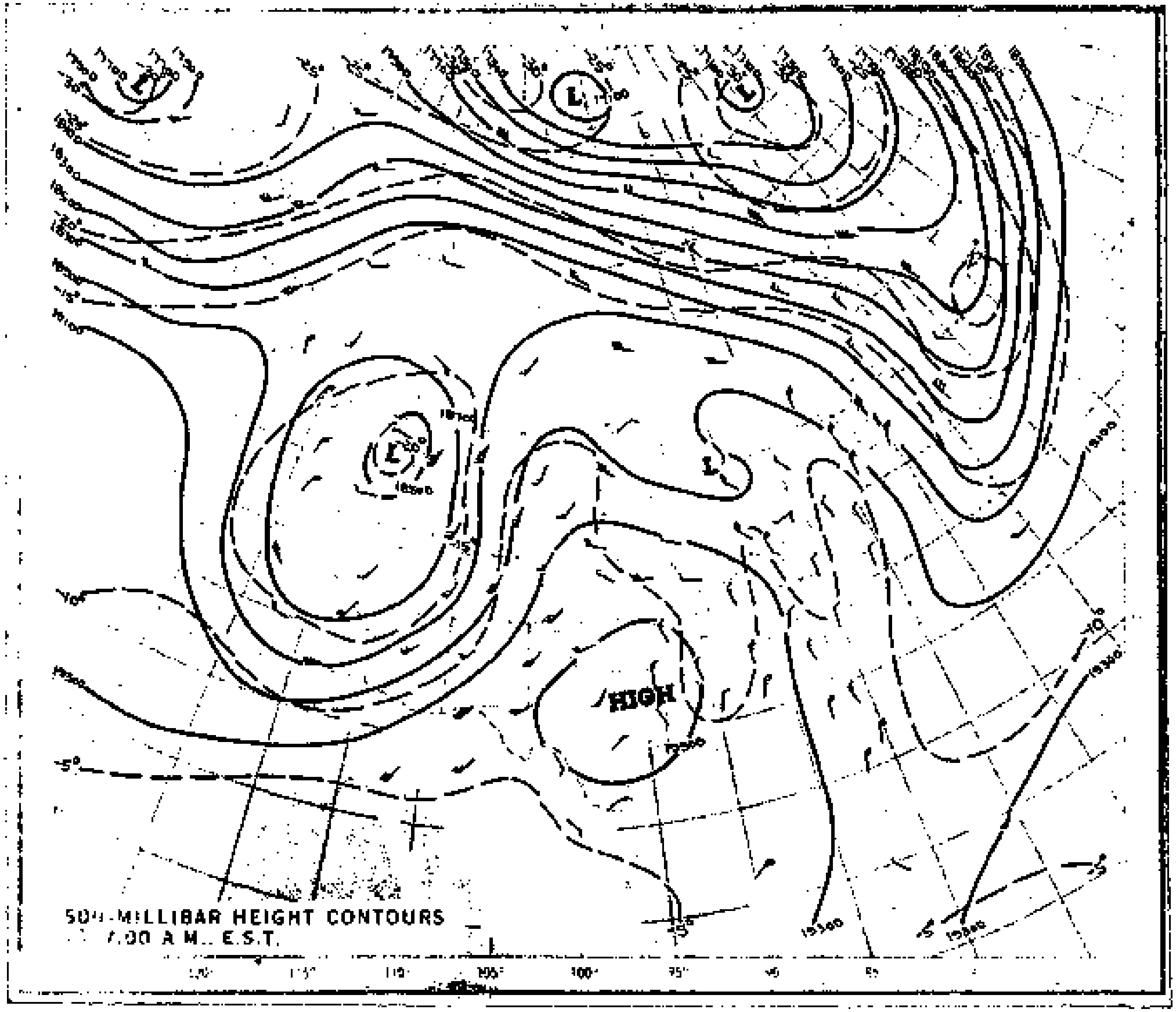

Figure 7b. Weather map at 0700 CDT. 500 mb map for 14 June 1970.

completion of the burn at 1421, 178A returned to base without attempting further observations. A great deal of water was collected by the ground crews, and the case looks as if it may be relatively well-suited to an In-budget study.

24 May 1970

The first test case of the season occurred this date. Numerous Cu and Cb clouds were present, and Aztec 178A selected one well-developed cell for an In burn starting at 1804 and ending at 1809. Updrafts of 500 to 1000 ft per min were measured. Cell moved rapidly toward ENE across the south edge of the network. The storm developed strongly, but because of its path, the sampling of the rain at the ground was not well-distributed. Weather maps are shown in Figure 8 a and b.

30 May 1970

This day was declared a No Go day at the morning weather briefing. Weather maps (Figure 9 a and b) indicate the situation. Thunderstorm activity began, however, about 1600, and UM personnel alerted ISWS radar to Go situation by telephone. NCAR air crew was available, so Queen Air carrying In flares came to storm area about 1800. Indium burn began at 1813 and concluded at 1819. Difficulties of logistics and storm trajectory cause us to suspect that most of the tracer moved about NNE-ward across the NW corner of the network. We anticipate inadequate resolution of the deposition streak for most of our central purposes.

29-30 May 1970

Weather maps, for the 29th (Figure 10 a and b) and the 30th (Figure 9 a and b) show the situation. A strong squall crossed Clinton about 2015, too late for aircraft operations. Samplers in place on the network, and one Gatz sequential

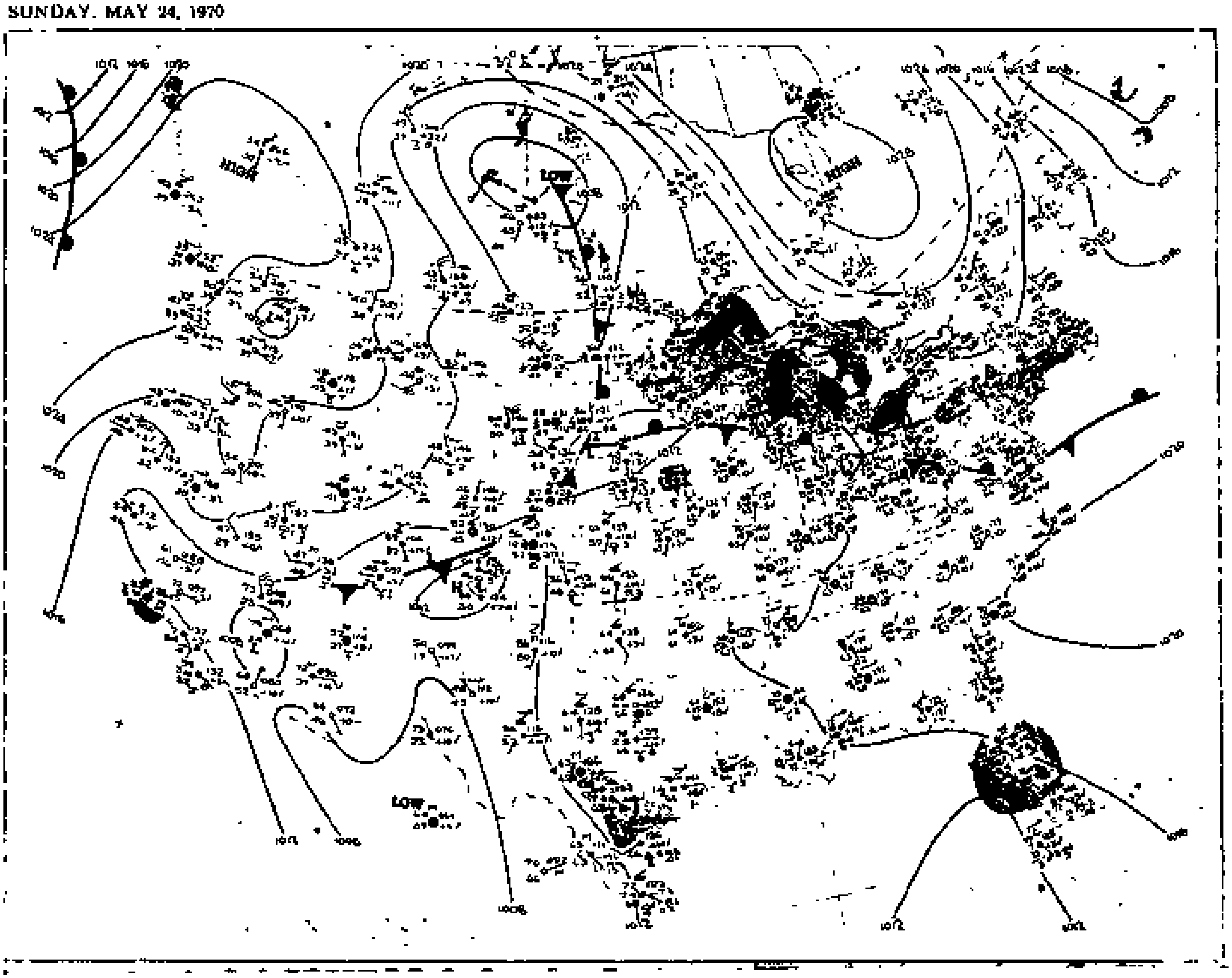

Figure 8a. Weather map at 0700 CDT. Surface map for 24 May 1970.

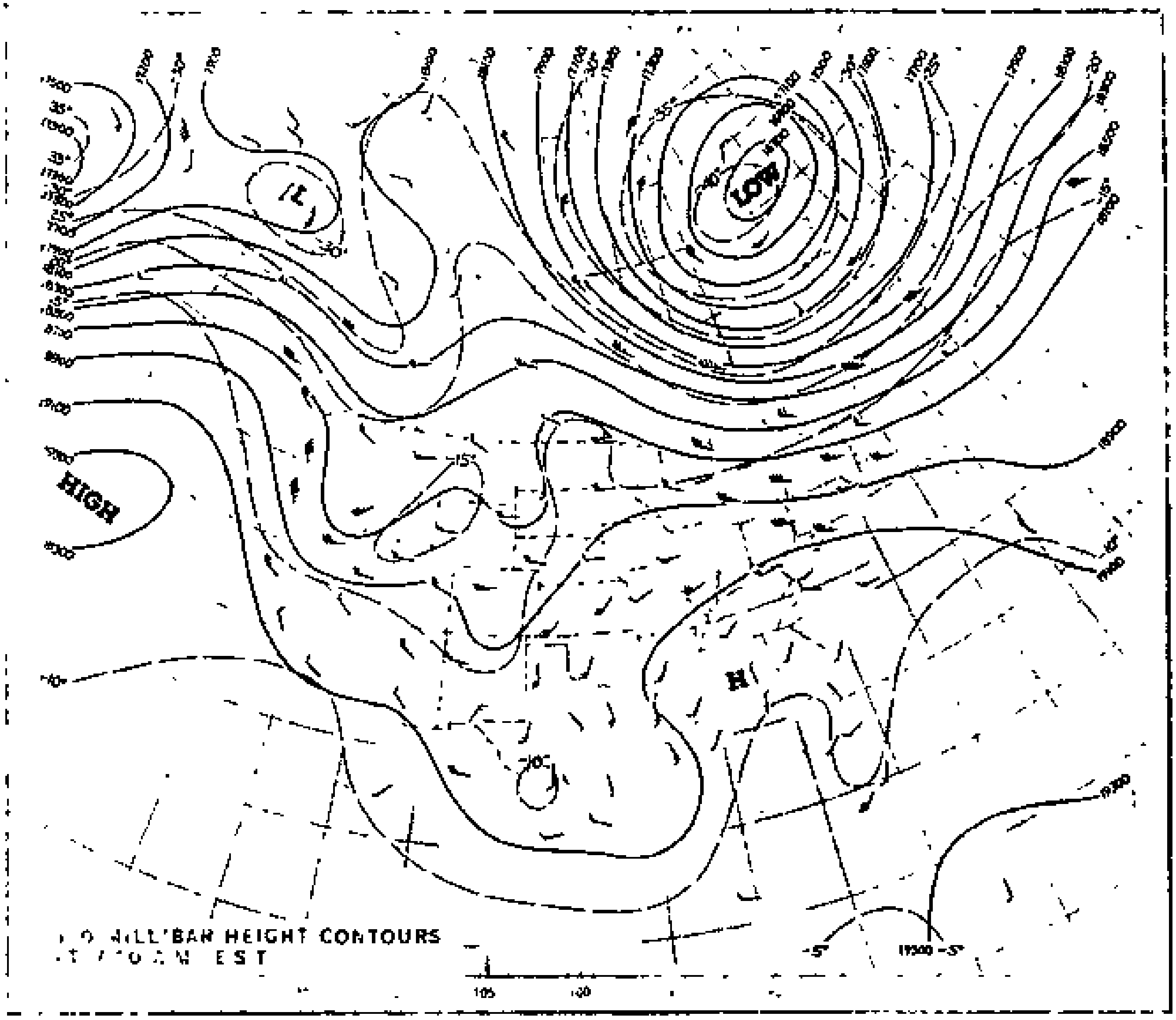

Figure 8b. Weather map at 0700 CDT. 500 mb map for 24 May 1970.

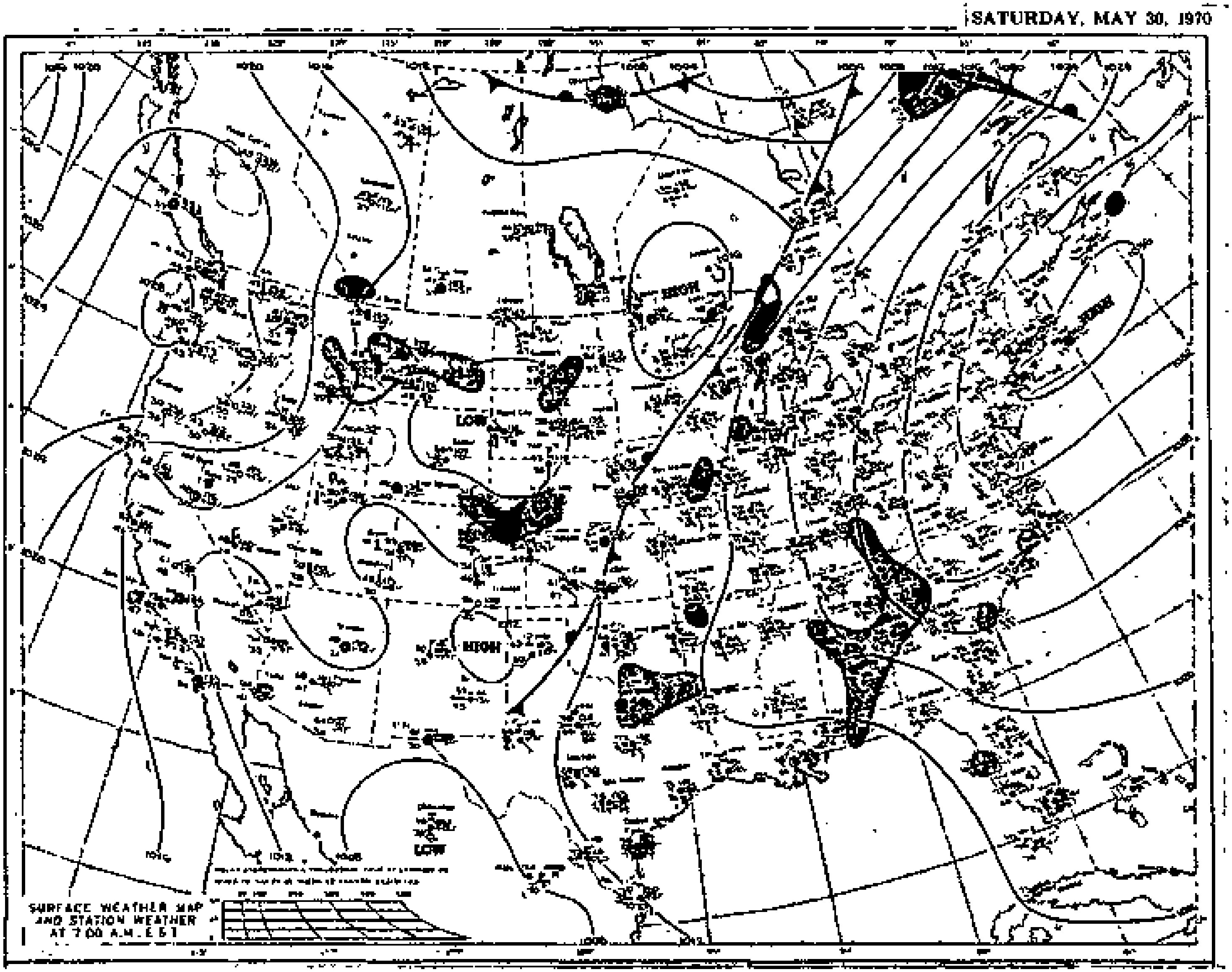

Figure 9a. Weather map at 0700 CDT. Surface map for 30 May 1970.

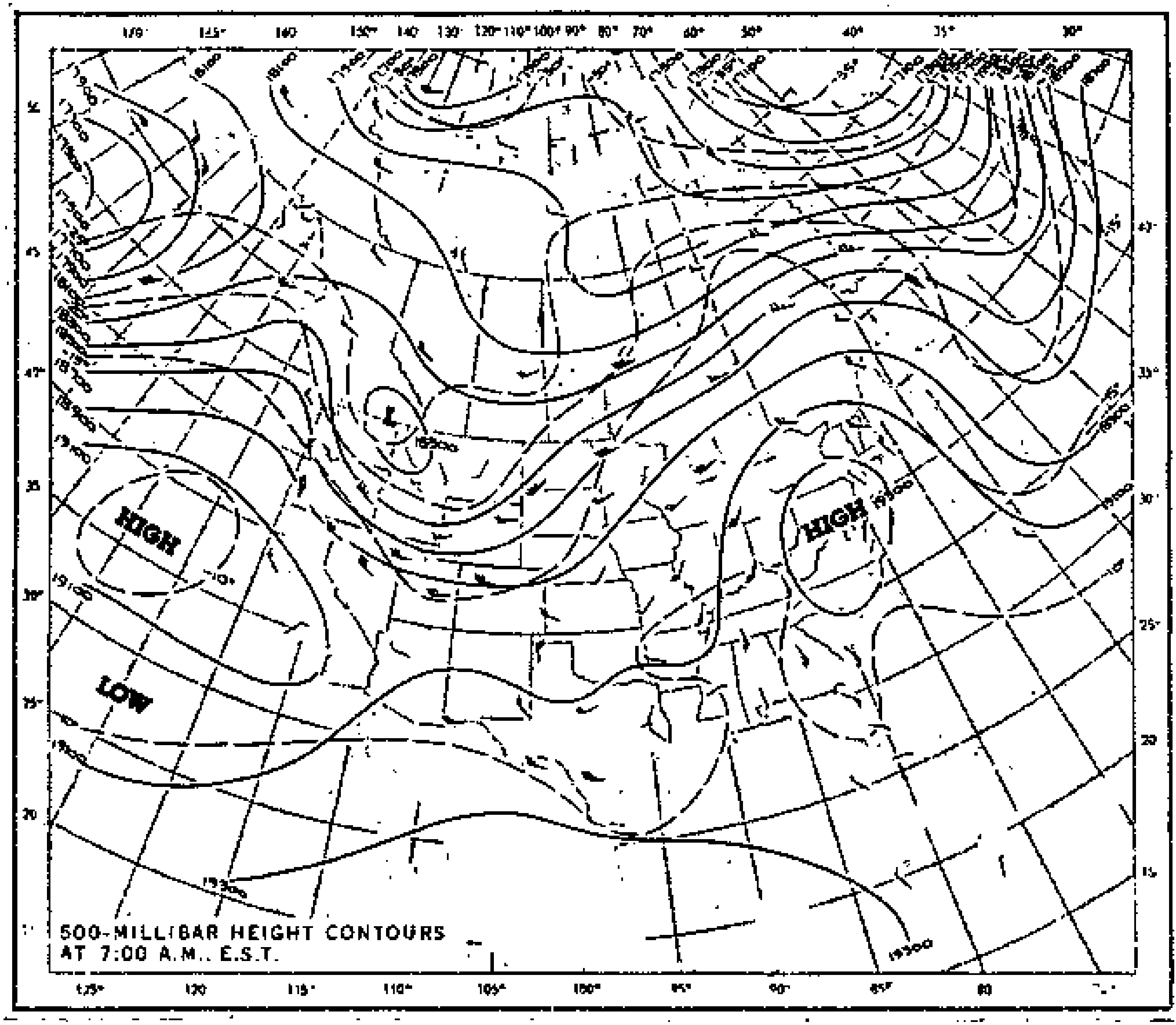

Figure 9b. Weather map at 0700 CDT. 500 mb map for 30 May 1970.

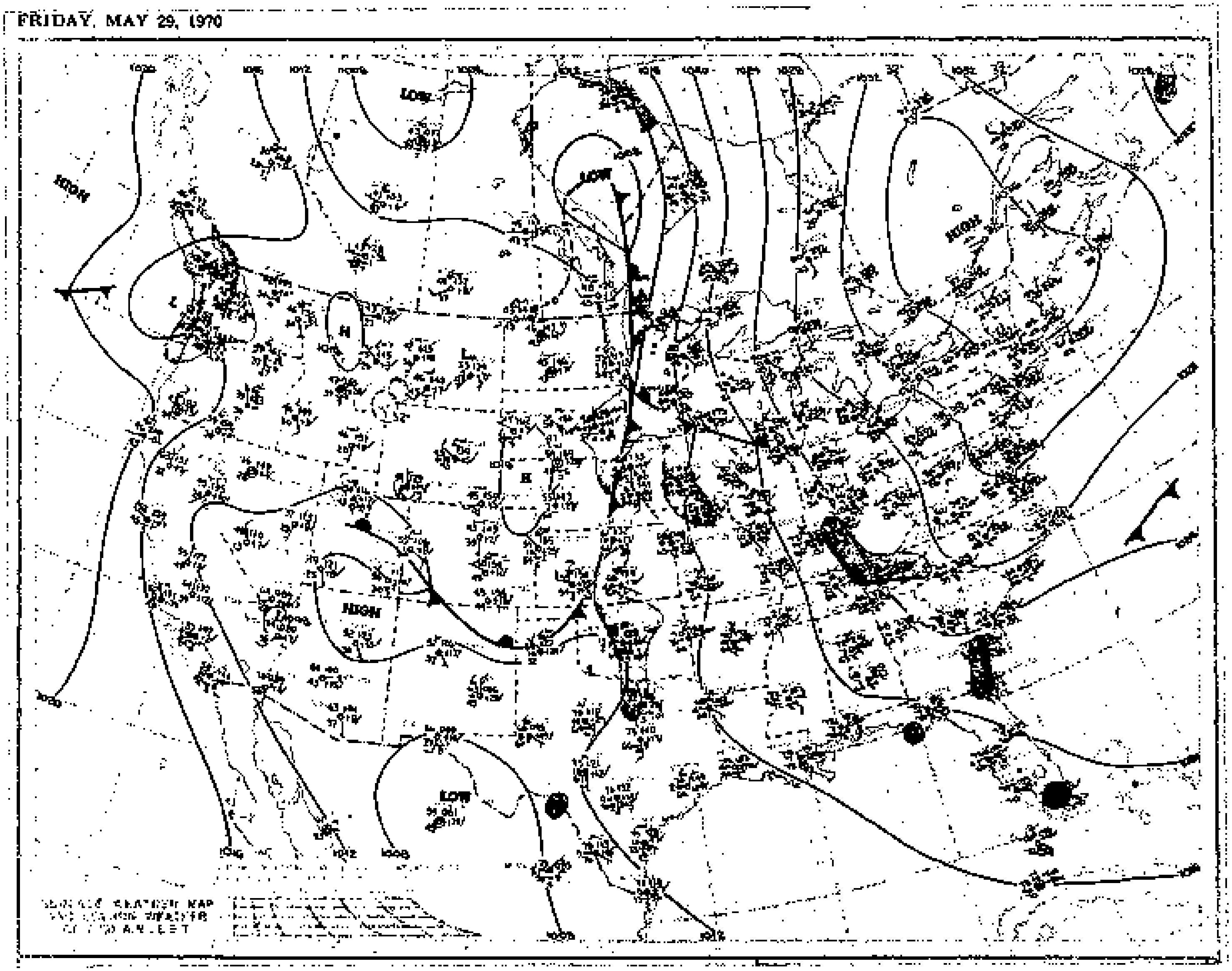

Figure 10a. Weather map at 0700 CDT. Surface map for 29 May 1970.

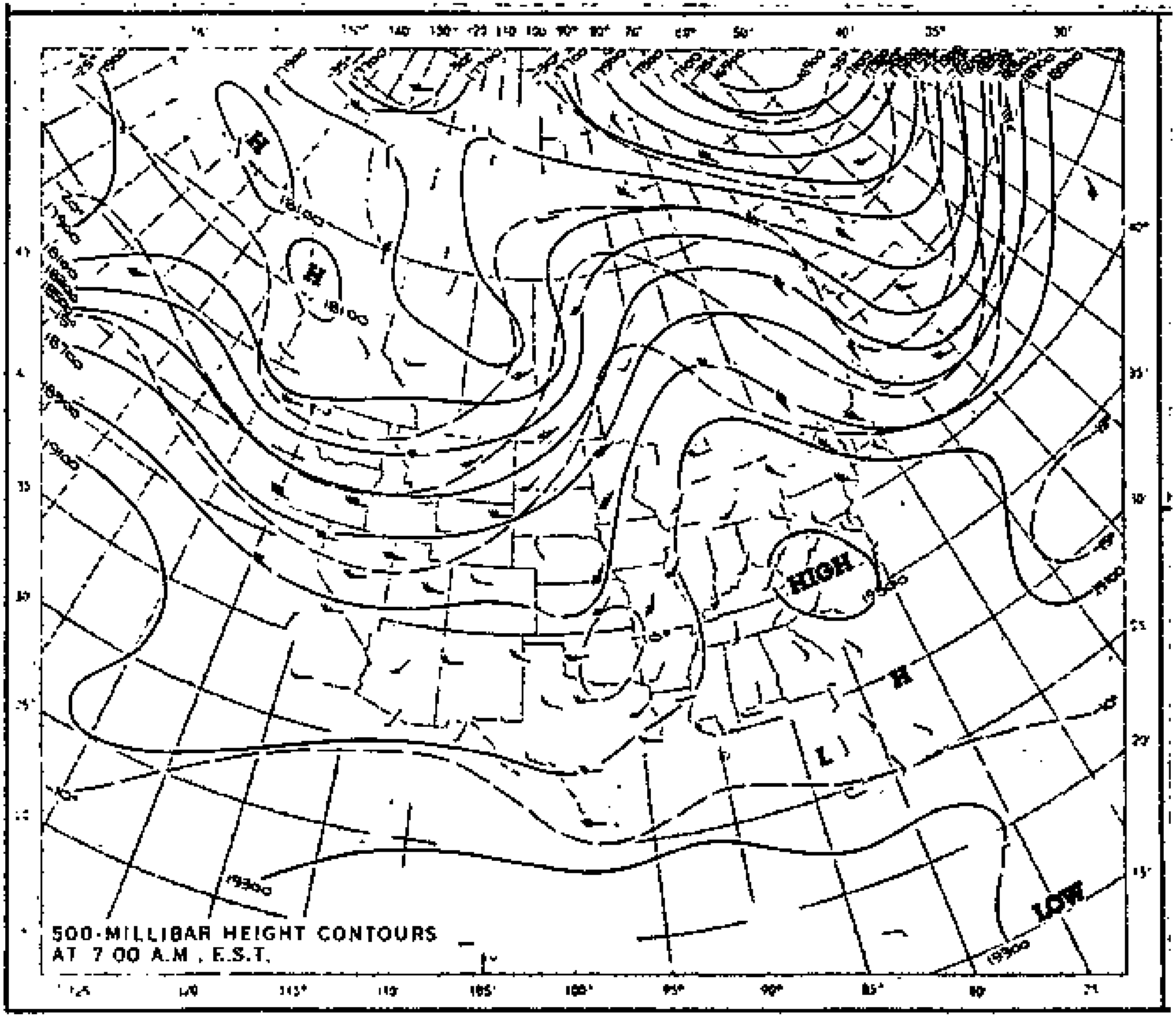

Figure 10b. Weather map at 0700 CDT. 500 mb map for 29 May 1970.

sampler were in operation. A selection of samples from those caught was made and these were processed to serve as background level indication for In and Li especially.

5 June 1970

An attempt to determine the particle-size distribution of the In and Li plumes was made this date. Weather maps are shown in Figure 11 a and b. The combination of circumstances, most severe of which was that the trained observer for the Queen Air was not available for the mission, was such that this experiment must be repeated.

II. Analysis and Collation of 1970 Data

In as much as Project ITREX is composed of many independent parts, each having its own schedule and focus, it is still too early to put together a complete case study from the 1970 expedition. Some data are obtained by each of the participating units, some analyses, and some collations also by each unit. We do not have the contributions of the other participants at this time, so we can report only upon our own progress with analysis and collation.

Our analytical effort has centered upon

(1) indium determinations by neutron activation analysis
(2) reduction of samples collected for atmospheric and pollen counting, and
    (a) β-counting, and
    (b) pollen counting and classifying for these samples

Some of the resulting data are reported below without interpretation.

At the same time we have spent a great deal of effort in collecting, sorting, and plotting the basic synoptic weather information required for the interpretive phase of our work. Having been forced to go this route, we note here for the bene-

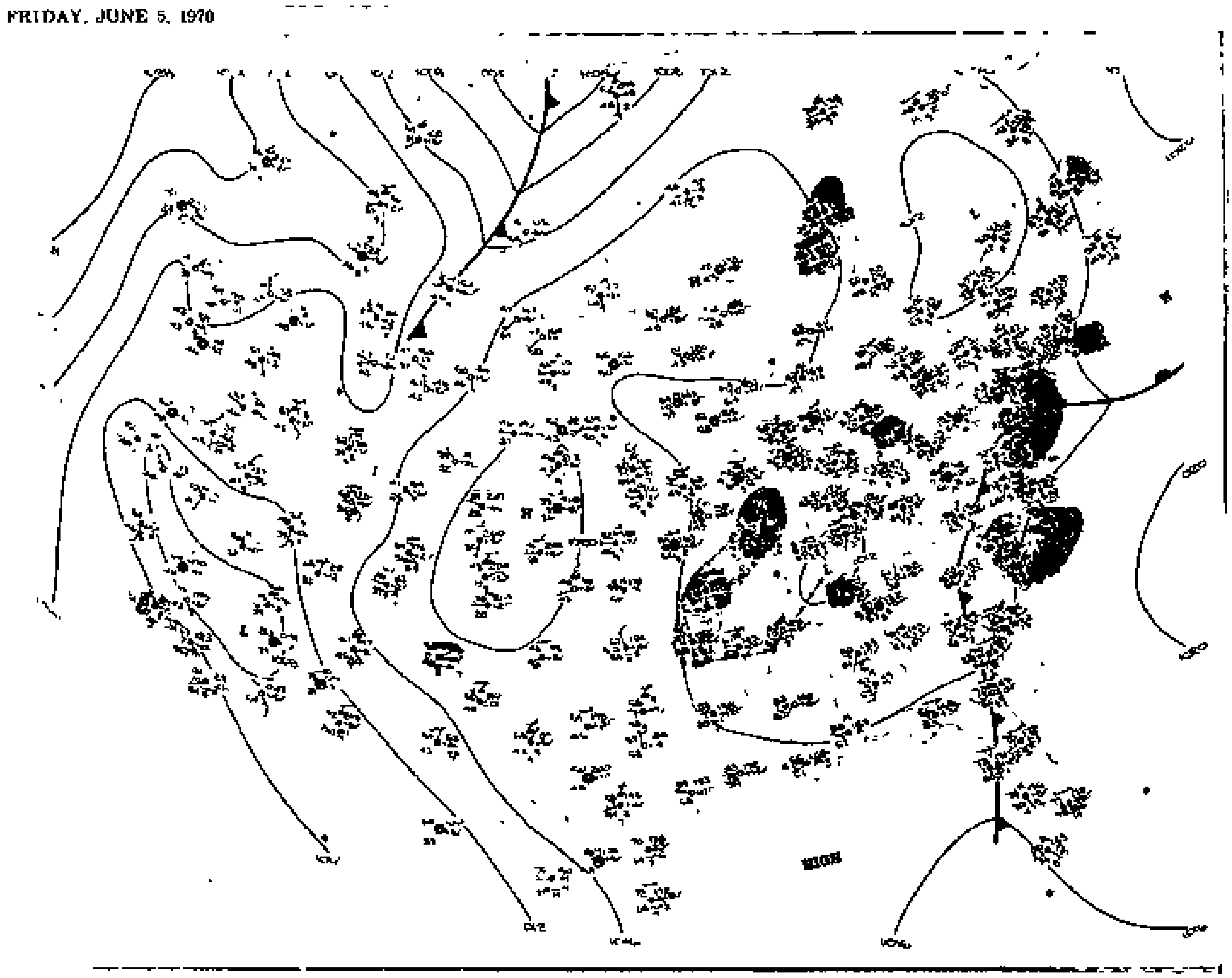

Figure 11a. Weather map at 0700 CDT. Surface map for 5 June 1970.

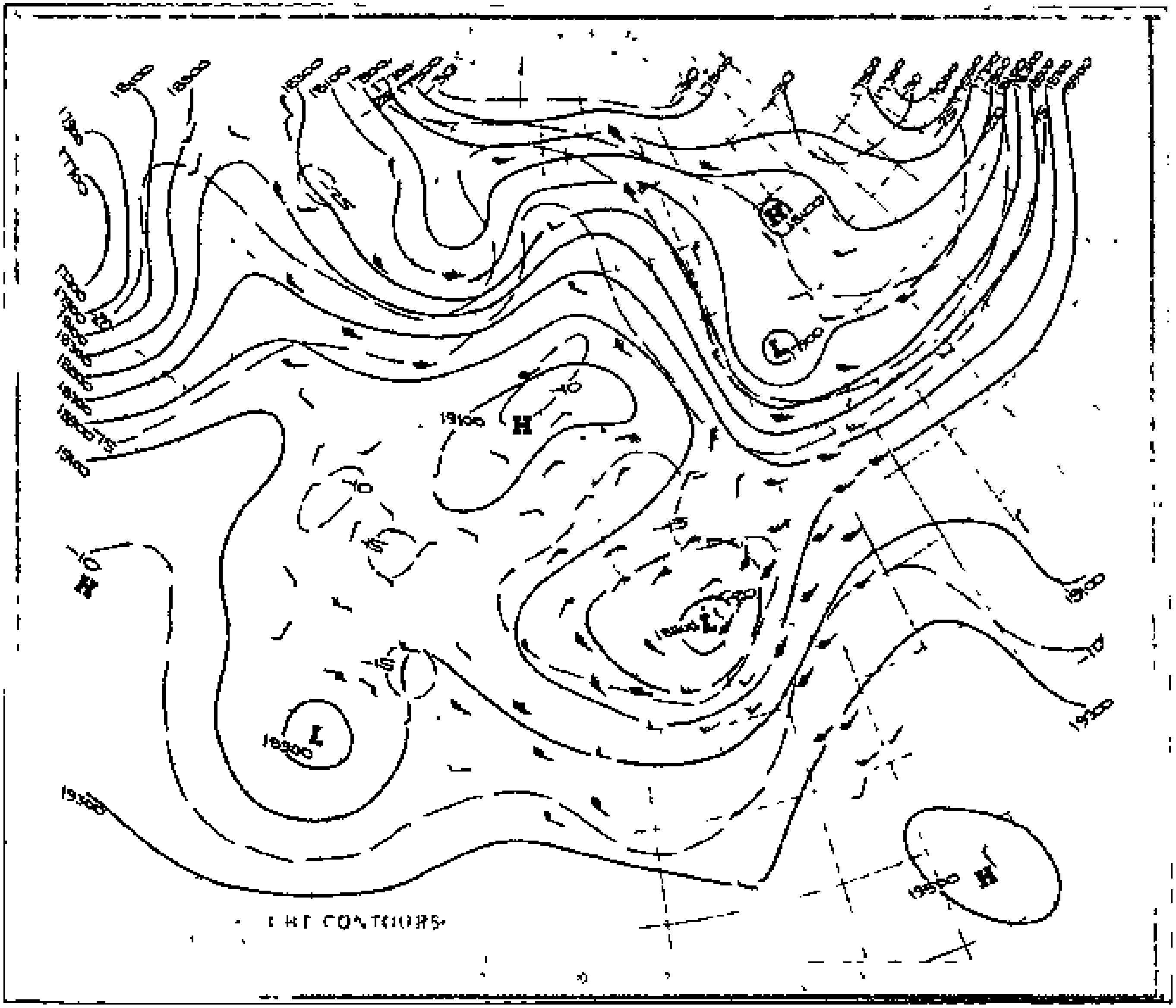

Figure 11b. Weather map at 0700 CDT. 500 mb map for 5 June 1970.

fit of future field projects that it would be an economy, to have at least Service A teletype facilities in association with any field operation. This statement is reinforced by the current ruling that Service A data are not to be stored by the National Weather Records Center. The hourly sectional weather maps obtained from these reports are most relevant to our interpretations, but after the fact, the data are hard to obtain. They should be obtained and stored specifically by and for any such project as ITREX, and they should be plotted and preliminarily analyzed during the slack (dry) periods of the field operation as far as possible. This is one of the very useful supporting services of NSSL for operations in its network area.

In addition, the analysis of air samples taken by means of the rotobar sampler, the Andersen impactor and the Brar (ANL) hi-volume sampler remains to be done. The rotobar counting was begun in the field, but in the pressure of that activity was done mostly piecemeal to give indications of the airborne microscopic material. Comprehensive counting and evaluation in terms of particle size spectra, etc., remains to be done in our particle laboratory.

The Brar and Andersen samples have been transmitted to Dr. Gatz and ANL for trace metal analysis by Dr. Brar's method. At this point, because of reactor problems at ANL, it is uncertain when the work may be done, and we are contemplating the advantages of analyzing them ourselves using the Dams, Rahn, Winchester multi-element analysis procedures (neutron-activation). This we can and will do, as in fact we have already done for Dr. Gatz on his airborne samples taken in 1969, provided that our funding is sufficient and our analytical program moves along satisfactorily.

Additional areas of our trace element analysis program include the following:

(1) the determination of Pb, Zn, Cd, Cu, and a few other metals by anodic stripping voltammetry (ASV), and

(2) the measurement of a number of elements by atomic absorption spectrometry (AAS),

each to be used for the analysis of samples of rainwater and of airborne particles (Andersen) that were collected at Clinton in 1969 and 1970. Whereas both ASV and AAS are physico-chemical techniques that do not require radioactivity, they are considered complementary to the neutron activation (NA) technique in that they give access to some elements that are not as readily measurable by NA. ASV, for example, is both fast and sensitive, and is unique in its ability to indicate and measure Pb. In the present context of air pollution by automotive exhausts, and the study of rain scavenging of the specific associated contaminants, Pb is of considerable importance. AAS is less sensitive, but is relatively easy and quick to use for the alkali metals especially, but also for Pb (with special accessories) and others. We have the requisite basic facilities in our laboratory for these analyses, and we are confident that the information to be gained for our case studies will be well worth having.

III. Results from 1970 Field Expedition

As indicated above, the samples for 1 June 1970 were given first priority in the In-analysis schedule. As a result, we can present Figure 12 showing the distribution of In concentrations in the whole-storm rain samples collected on the network. The north-south pattern found here is to be compared to the NE-SW pattern of Li concentrations shown by the preliminary ISWS results shown in Figure 13.

The Li was placed in the height range 12,000 to 16,000

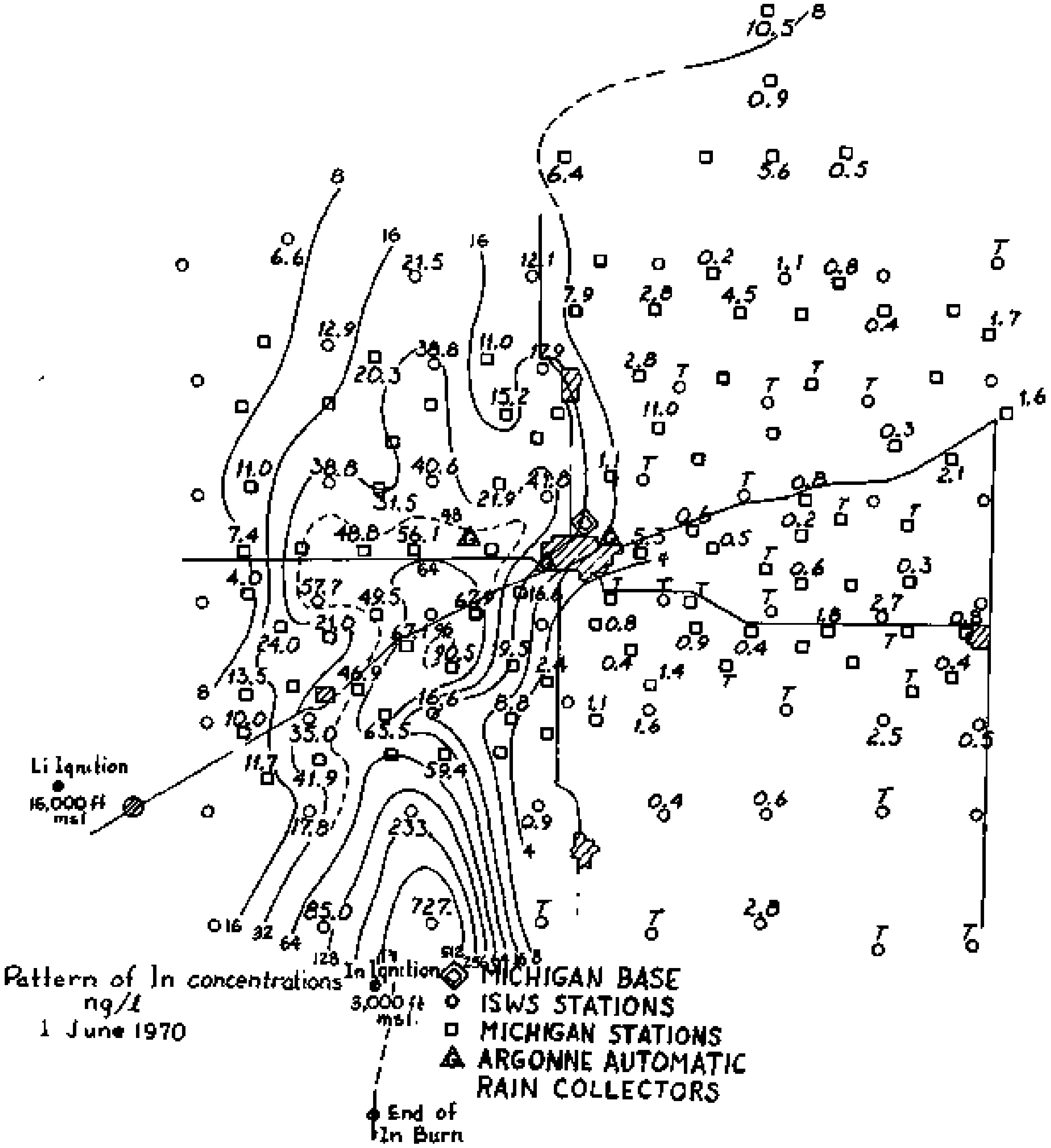

Figure 12. Map showing the distribution of Indium concentrations in the whole storm rain samples collected in the network on 1 June 1970. Also indicated is the aircraft path followed during the indium burn.

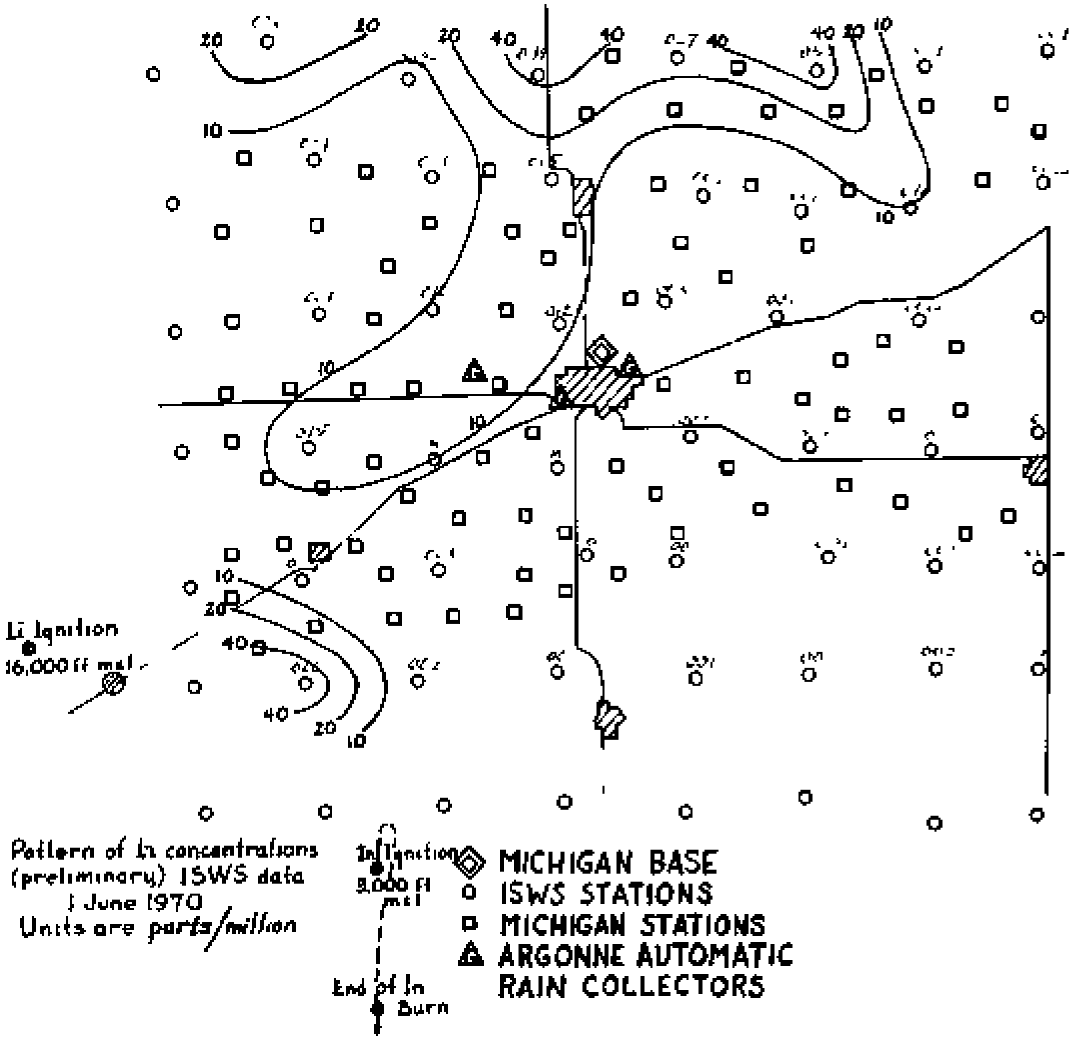

Figure 13. Map showing the distribution of lithium concentrations in the whole storm rain samples collected in the network on 1 June, 1970. Along with these preliminary ISWS results is indicated the aircraft path followed during the lithium burn.

ft as nearly as we can tell, in the midst of the general cloud mass, along a path some 11 to 12 mi NW of the track on which the In was placed at about 1,900 ft msl.

The patterns show a sharp contrast in the directions of translation of the tracer plumes and in the extent of lateral diffusion at each level. The concentration maxima are not at all coordinated, although they must be affected by the rainfall pattern to some extent. The composite picture appears to show results of low level versus high level rain-generating and scavenging processes. The plumes appear to have labeled totally different populations of raindrops initially, but there also appears the possibility that they converged near the northern limit of the sampling array. Resolution of these relationships and the processes themselves will be contributed by the addition of other data such as the sequential samples from the ANL automatic stations, the UM station and the UM mobile stations. These will provide temporal relationships between the high In and high Li rainfall samples and the rainfall intensity.

Inasmuch as the Li flares "popped:, thus making pellet-sized fragments, it is not possible to make a definitive Li-budget study of this situation. The In pattern, on the other hand, is probably sufficient basis upon which to attempt such a study for In.

The cellular character of the In-pattern suggests the possibility that the respective distinct scavenging processes played different proportionate roles in producing the separate maxima. The mechanics of the system are obviously complex, but it seems quite reasonable to postulate a dominance of large-particle nucleation and washout processes for the primary maximum, versus mainly nucleation for the secondary maximum and perhaps nucleation and diffusion for the tertiary maxima. Our theoretical studies and our sequential data will help to resolve these problems of interpretation.

Rainfall intensity variations throughout the period from 1709 to 1837 CDT at UM station are shown in Figure 14 together with the atmospheric β-radioactivity concentration and pollen counts. No attempt is made here to draw interpretations from these data, but we do note that the most intense shower activity in the period 1825 to 1833 CDT was probably too late to be labeled by either tracer.

Rainfall intensity and β-radioactivity for the rains of 30 May and 14 June are shown in Figures 15 and 16. The rainfall intensities are shown for 15 and 20 June in Figures 17 and 18.

IV. New Results from 1969 Field Expedition

Most of the results from our 1969 field program were presented in our last report. Only one indium tracer experiment was carried out, on 29 May 1969, and that was marginal by virtue of the leap-frogging behavior of the storm as it approached and skipped over our field station. This one case deserves further study, and will be given #4 or #5 priority in our schedule. The other cases have provided indium background data, sequential sets of β-radioactivity and pollen data, and dry fallout rates. All were reported previously.

By virtue of the request for NCAR aircraft support that was made by UM, Dr. Gatz was enabled to procure a series of air samples using the NCAR Queen Air 304D and the Shedlovsky air-sampler on 1 and 6 August. We were asked to analyze some of these samples using the multielement non-destructive neutron activation procedure (Rahn, Dams, Robbins and Winchester, 1970). Twelve elements were measured by this method, and the results are presented in Table 4. Description of the location and conditions for the samples are given in Table 5.

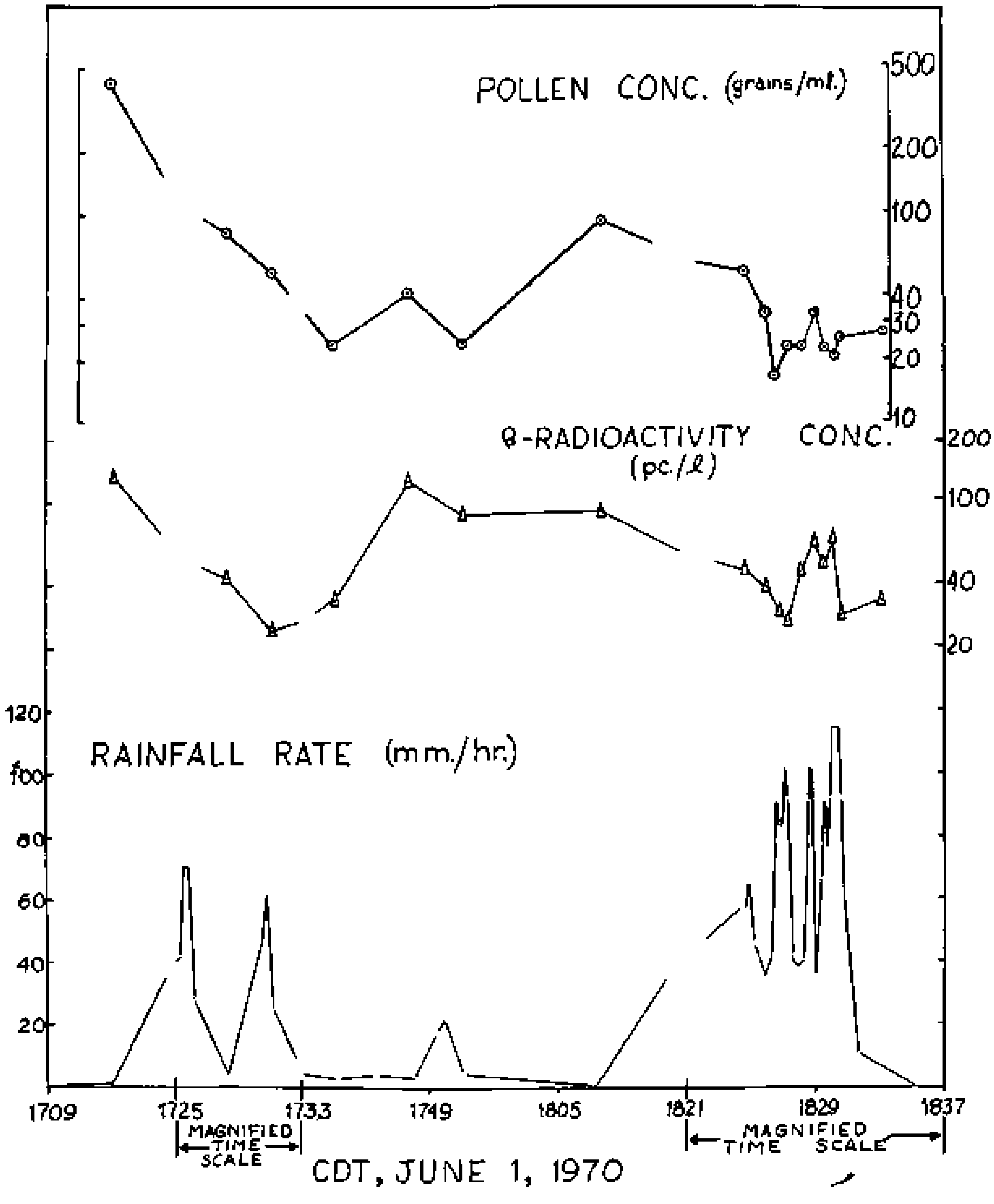

Figure 14. Rainfall intensity variations and pollen and β-radioactivity concentration for rain samples collected on 1 June, 1970, at Clinton, Illinois.

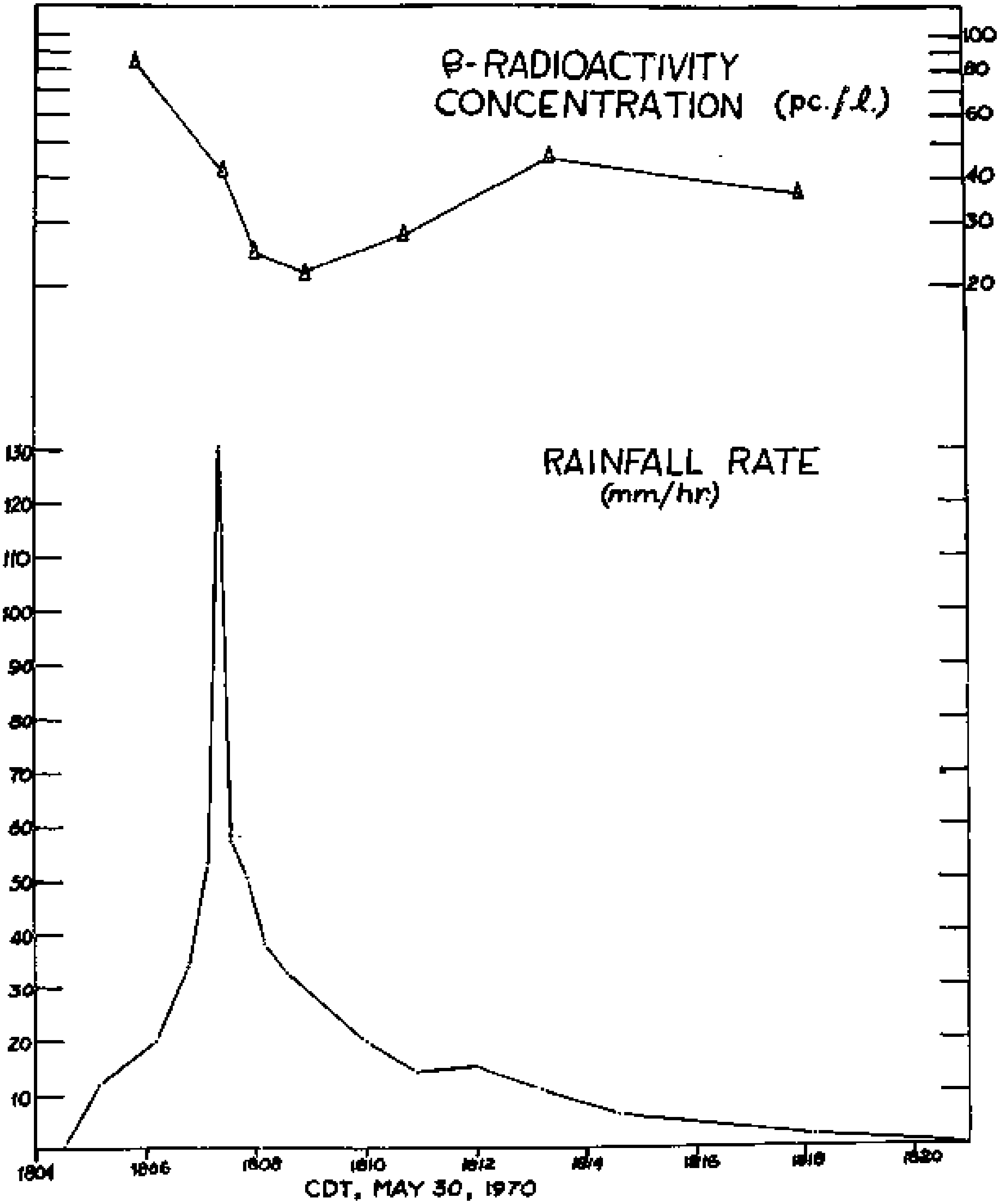

Figure 15. Rainfall intensity and β-radioactivity concentration for rain samples collected on 30 May 1970 at Clinton, Illinois.

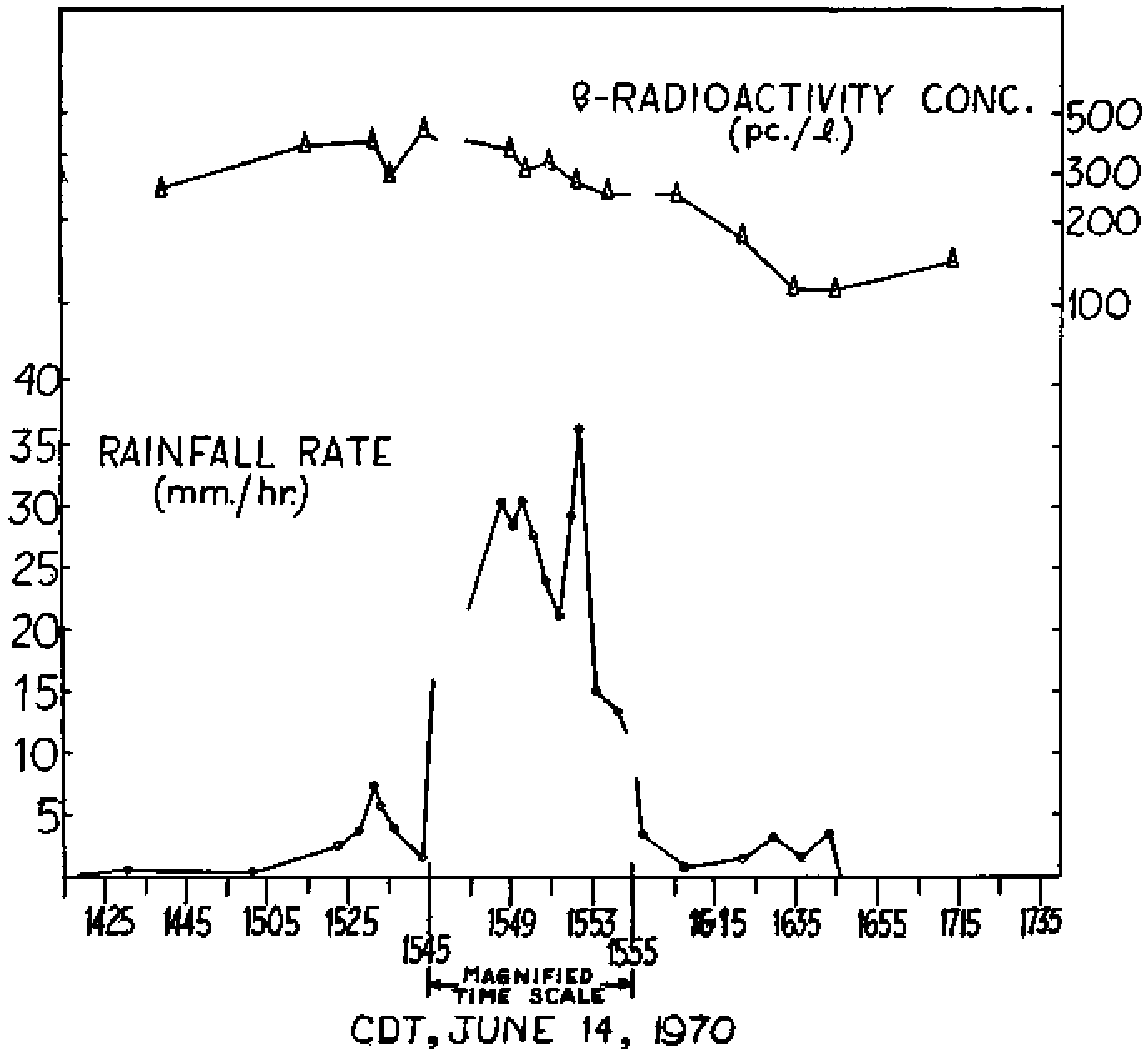

Figure 16. Rainfall intensity and β-radioactivity concentration for rain samples collected on 14 June 1970, at Clinton, Illinois.

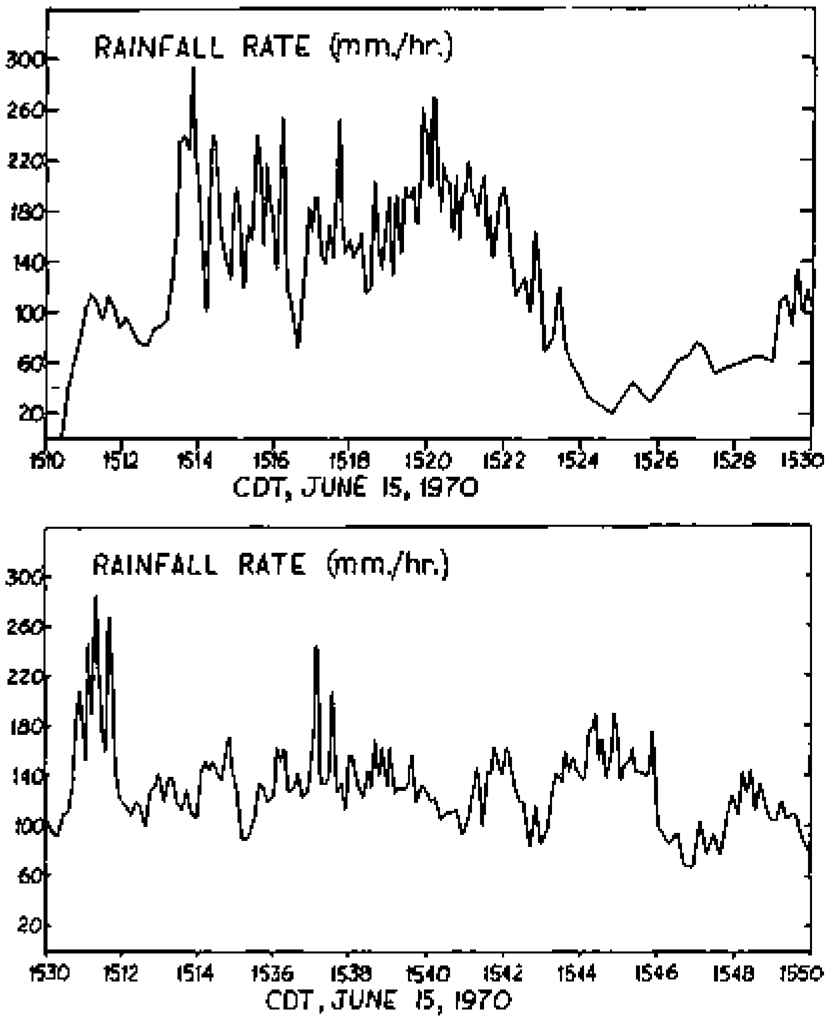

Figure 17. Rainfall intensity for rain of 15 June 1970, at Clinton, Illinois.

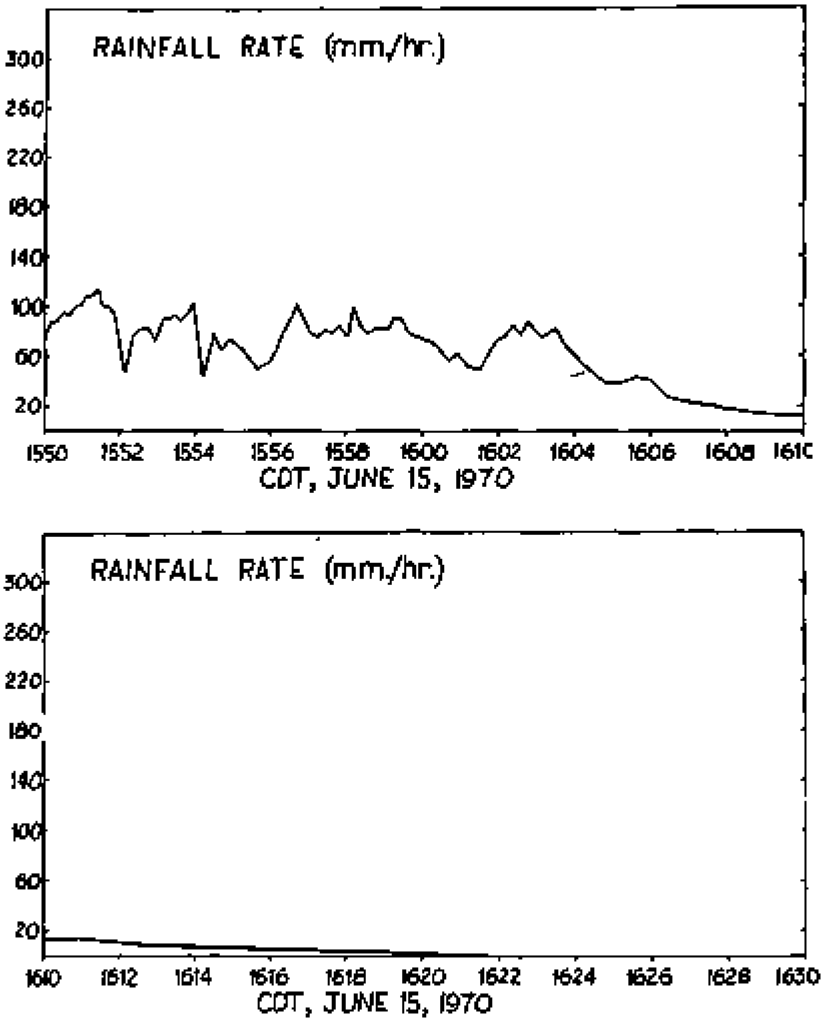

Figure 17 continued.

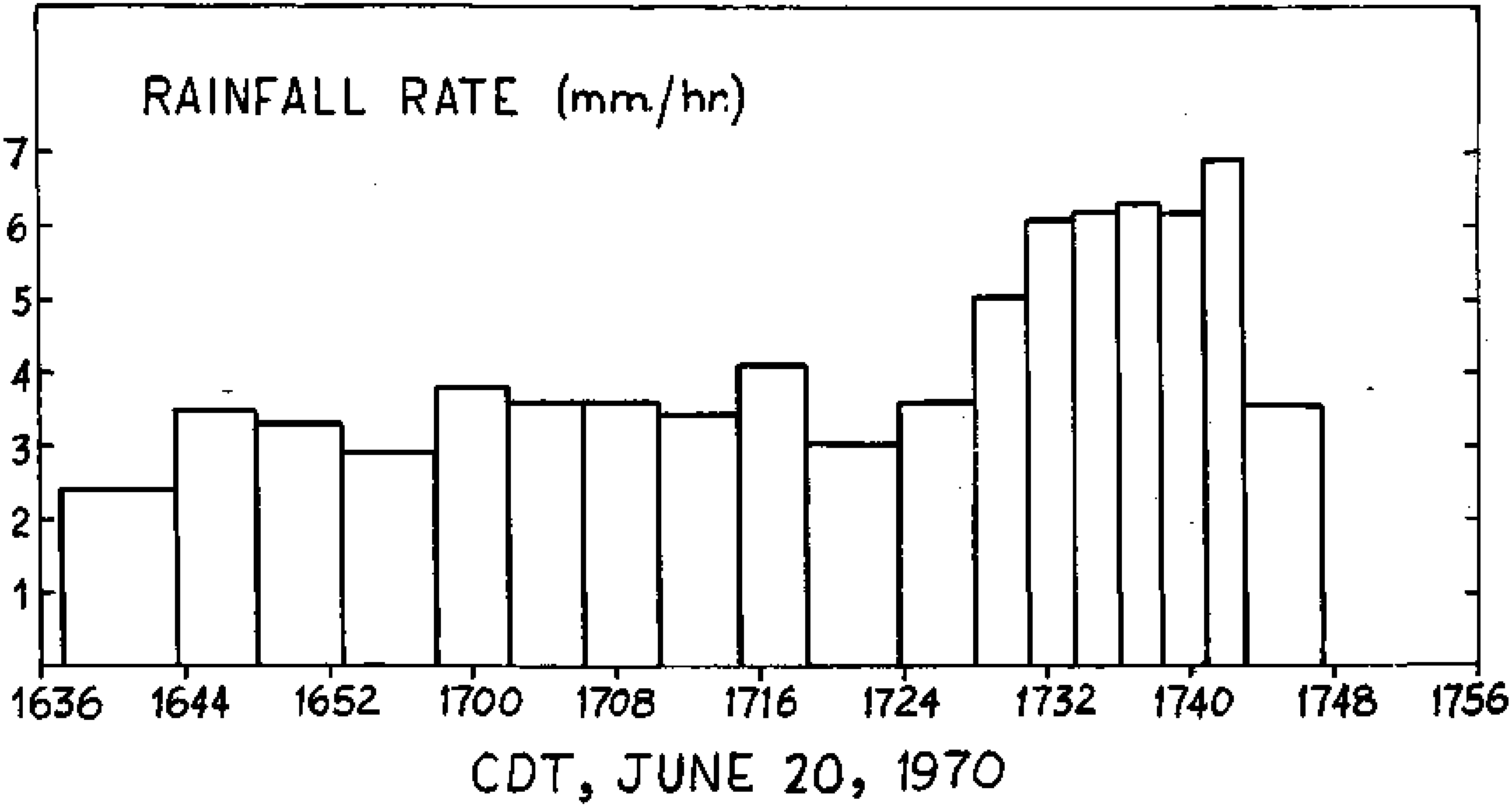

Figure 18. Rainfall intensity for rain of 20 June 1970, at Clinton, Illinois.

Table 4: Amounts of 12 Elements found on Shedlovsky Filters. Samples taken 1 & 6 August 1969 by D.F. Gatz using NCAR Queen Air 304D. Amounts are given in ng per gm of filter plus sample (see text).

Sample No:	#623	#619		#621		#624		#627	
Total wt,gm:	0.47375	0.642		0.306		0.513		0.252	
ELEMENT		GROSS	NET	GROSS	NET	GROSS	NET	GROSS	NET
Aluminum	21910	18512	---	29569	7659	32660	10750	18258	---
Bromine	199.6	---	---	245	45.4	611.5	411.9	313.0	113.4
Chlorine	15900	17384	1484	9801	---	15130	---	19154	1770
Calcium	25240	583.2	---	962.1	---	26320	1080	24762	---
Copper	4195	583.2	---	962.1	---	---	---	1816	---
Indium	1.33	---	---	---	---	.78	---	0.46	---
Iodine	---	70.57	70.57	399.0	399.0	---	---	866	766
Manganese	344.9	184	---	160.0	---	---	---	222.0	---
Magnesium	---	---	---	---	---	---	---	---	---
Sodium	5941	6374	433	4170	---	7597	1651	4238	---
Titanium	3071	863.2	---	788.9	---	2754	---	2280	---
Vanadium	39.68	26.9	---	47.81	8.13	72.06	32.38	34.9	---

Table 5: Dates; Locations and Conditions for Samples listed in Table 4.

Sample No.	Date	Altitude ft, MSL	Location	Remarks
619	1 Aug	9,500	Watseka, Ill. to Crown Pt., Ind.	Clear air above clouds
621	1 Aug	4,700 - 6,600	Crown Pt., Ind.	In and out of cloud
623	6 Aug	12,600	En route to St. Louis, Mo.	CONTROL
624	6 Aug	4,800	Champaign, Ill.	In haze, occasionally in Cu cloud
627	6 Aug	12,650	NE of Troy, Ill. to St. Louis, Mo.	Above Cloud

V. Subsidiary Studies

In anticipation of the need to make quantitative interpretations of our field data, leading to numerical models of the processes of rain generation and scavenging, we have engaged in supportive studies of limited scope. Among these studies are (a) a study of the pollen grain size spectra in relation to time and rainfall intensity changes, and (b) a study of the terminal speed of freely falling raindrops.

A. Variations of the Size Spectra of Scavenged Pollen Grains

An integral part of our rain scavenging observations has been that of the numbers and species of pollen grains found in our sequential rain samples (see previous reports and publications, e.g., Dingle & Gatz, 1966; Dingle, 1966). Whereas the general classification of all pollens among the largest airborne particles indicates that they are especially subject to scavenging by the impact-collection mechanism, the possibility that some pollens may serve as highly favored super-giant condensation nuclei also exists, and it is appropriate to investigate our data with this possibility in mind.

Mr. Steven Jermaine, graduate assistant, has now worked with this project for several years, initially as undergraduate assistant in research. In this capacity, he undertook special training in palynological technique, and has since had principal responsibility for our pollen analysis. For the present samples, he adopted the procedure of counting pollens by diameter categories instead of using the biological classification by species which was used heretofore. Using 3-μ intervals of diameter through the gamut, from 9μ to 66μ, of observed pollens, he has produced an interesting set of data which affords a much improved resolution of the large-particle portion of the contaminant spectrum.

Examination of the changes of the pollen size-spectrum through the course of the rain of 1 June 1970 leads to some interesting observations relating most directly to the impact-collection process. A specific feature is the tendency toward bimodalism of the pollen-size spectrum. The two modes are associated with two dominant varieties of grass pollinating on 1 June. Inasmuch as these particles most consistently approximate the spherical shape assumed by theoretical analyses of impact collection, we anticipate that this process can be modeled quite successfully using established collision efficiencies in conjunction with the pollen - and raindrop-size spectra. This sub-project will be reported more fully in our next report and/or a published paper.

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B. A Study of Terminal Velocity

1) Introduction

The terminal velocity of falling raindrops has been studied by a large number of investigators (Lenard, 1904; Schmidt, 1909; Liznar, 1914; Flower, 1928; Laws, 1941; Gunn and Kinzer, 1949; Imai, 1950; Kumai and Itagaki, 1954; Beard and Pruppacher, 1969). Lenard measured drops in the interval from 1,000 to 130,000 g. Schmidt measured the terminal velocity for drops in the interval from 35 to 4,000 g. Laurs investigated drops having masses from 1,000 to 125,000 g. Terminal velocity measurements for water droplets from 0.2 g to 100,000 g, falling in air of 50% relative Humidity at 20°C and 1013 mb, were made by Gunn and Kinzer. Because of the crude experimental techniques used in early studies to determine the terminal velocity and the inaccuracies involved in the determination of the masses and equivalent radii of the drops, it is believed that the falling drops either had not reached terminal speed or were evaporating in unsaturated air. The results of individual investigations of the terminal velocity of falling drops as summarized in Figure 19 differ significantly. However, Gunn and Kinzer's data so far have benn regarded as the most complete and are used almost universally by meteorologists today.

2) Experimental Results

Recently Beard and Pruppacher (1969) measured the drag on small water drops falling in water-saturated air at terminal velocity for Reynolds number, Re, between 0.2 and 200. The radius, a, of a drop was determined by comparison with a calibrated glass scale and the terminal velocity, v, was determined from the air speed in the wind tunnel necessary to suspend the drop. From the experimental values of v and a, the ratio of drag, D, of a drop to the Stokes drag,

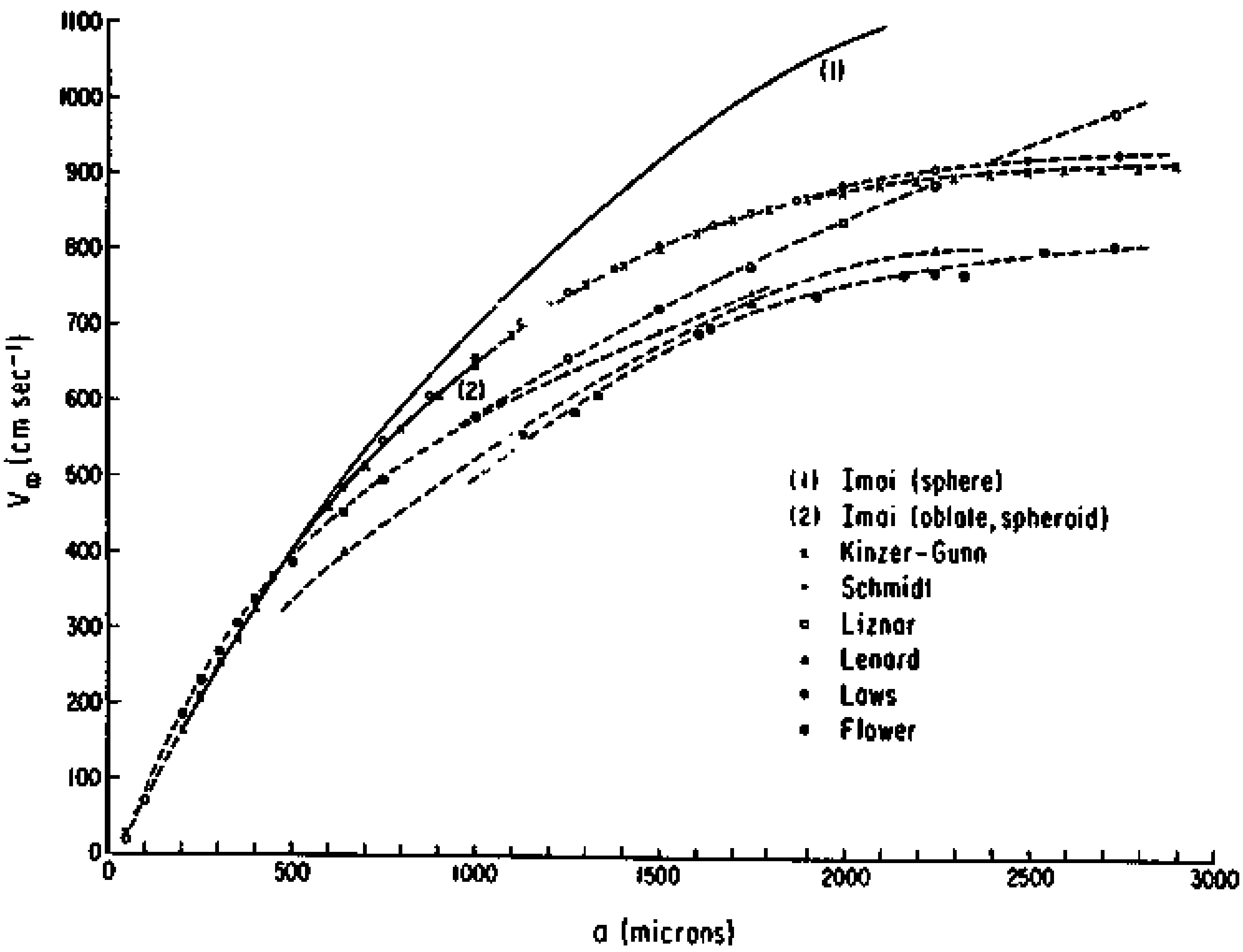

Figure 15 Terminal velocity of water and rain drops as a function of their equivalent spherical radius a. (From results cited in the literature. After Beard and Pruppacher, 1969)

D_s, was computed using

$$\frac{D}{D_s} = \frac{2}{9}\,\frac{a^2 g(\rho_s - \rho_m)}{\eta v} \qquad (1)$$

where ρ_s, ρ_m are the densities of the drop and of the viscous medium, respectively, and η is the dynamic viscosity. The least-square method gave the following empirical formulae

$$(\frac{D}{D_s}) = 1 + 0.102\mathrm{Re}^{0.955}, \quad 0.2 \leq \mathrm{Re} \leq 2$$

$$(\frac{D}{D_s}) = 1 + 0.115\mathrm{Re}^{0.802}, \quad 2 \leq \mathrm{Re} \leq 21$$

$$(\frac{D}{D_s}) = 1 + (0.189 \pm 0.006)\mathrm{Re}^{(0.632 \pm 0.007)}, \quad 21 \leq \mathrm{Re} \leq 200$$

From these empirical relations for $\frac{D}{D_s}$ vs Re the drag coefficient C_D was computed as a function of Re from

$$C_D = (\frac{24}{\mathrm{Re}})\ (\frac{D}{D_s}) \qquad (2)$$

The radius, a, of a spherical water drop falling in saturated air at various pressures and temperatures was computed as a function of Re from

$$a^3 = (\frac{9}{4})\ [\eta^2/\rho_m\ (\rho_s - \rho_m)g]\ \mathrm{Re}(\frac{D}{D_s}) \qquad (3)$$

The terminal velocity, v, of these drops was then calculated as a function of a from

$$v = \mathrm{Re}\eta/2\rho_m a \qquad (4)$$

The variation of v with a for drops of radius between 10 and 475μ falling at 400, 500, 700, and 1013 mb, and at temperatures -16, -8, 14 and 20°C, respectively, is shown in Figure 20.

It is seen from this figure that Beard and Pruppacher's data are slightly but consistently lower than Gunn and Kinzer's.

3) Graphical Method

A graphical method originally described by Langmuir (1948) can be used for the computation of terminal velocities of spherical drops. Solving

$$\frac{1}{2}\rho_m v^2 C_D \pi a^2 = \frac{4}{3}\pi a^3(\rho_s - \rho_m)g \qquad \text{gives}$$

$$v^2 = \frac{8}{3}\left(\frac{\rho_s}{\rho_m} - 1\right)\frac{ag}{C_D} \tag{5}$$

This expression is not particularly helpful since C_D is itself a function of v through its dependence on the Reynolds number. But the C_D-Re dependence is only an empirical relation not expressible in terms of any elementary functions. However, the expression $C_D Re^2$ is known from the empirical relations between D/D_s and Re; and equation (2), from which $C_D Re^2$ may be plotted for any value of Re. On the other hand $C_D Re^2$ may be written as

$$C_D Re^2 = \left(\frac{32}{3}\right)a^3(\rho_s - \rho_m)(\rho_m g/\eta^2)$$

Given the drop size a and the atmospheric parameters ρ_m and η, $C_D Re^2$ can be computed, the corresponding value of Re read from the graph, and v calculated from Equation (4).

4) Empirical Expression for Terminal Velocity

In raindrop-size distribution studies it is frequently necessary to predict terminal velocities of slightly changed

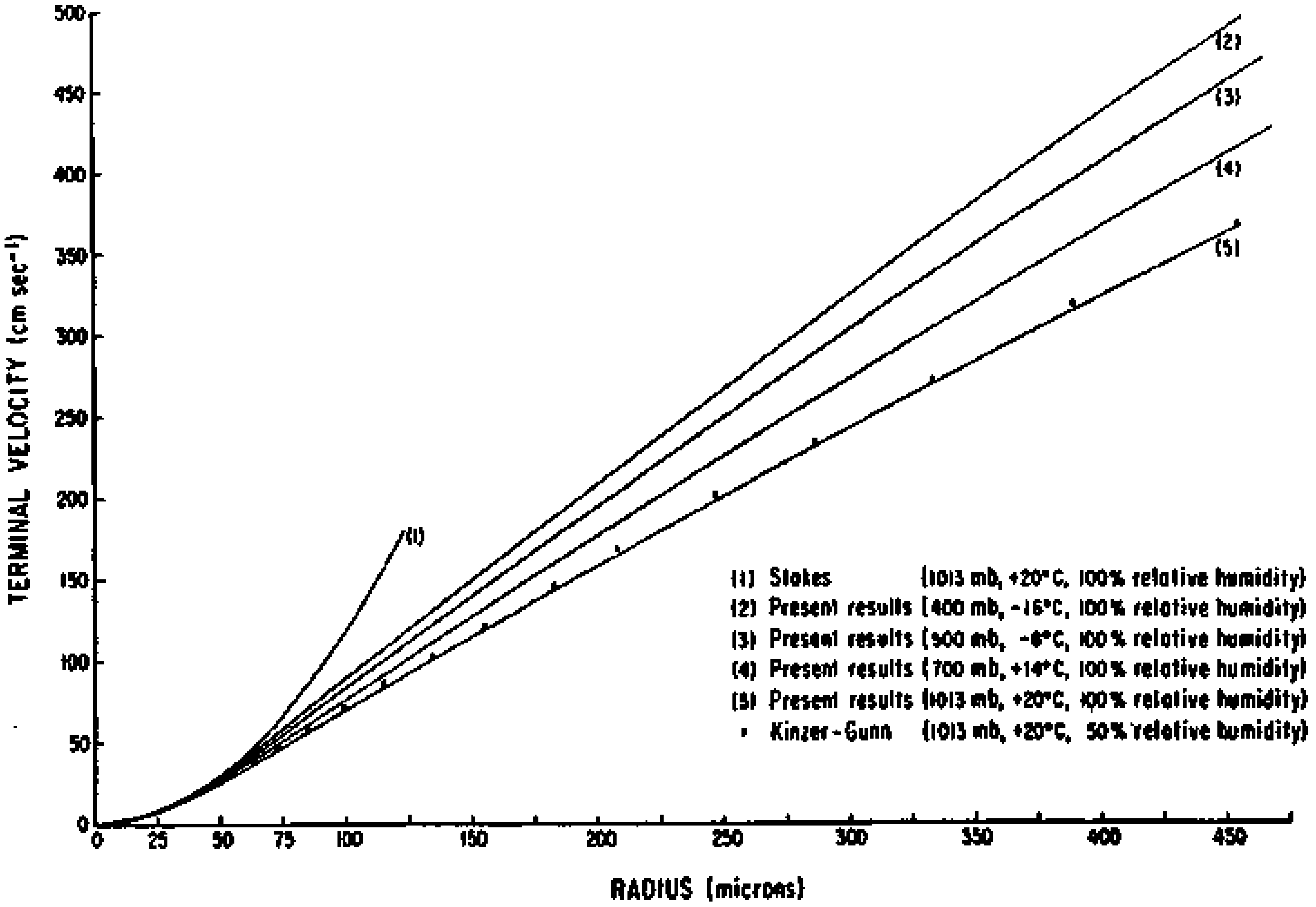

Figure 20. Terminal velocity of water drops of various sizes and under various environmental conditions. (After Beard and Pruppacher, 1969.)

drop-size caused by the processes of coalescence or evaporation. The indirect graphical method was found very inadequate and inconvenient for such computations. It is desirable to have reliable estimates of the experimental values. Hence an accurate empirical expression is required. Gunn and Kinzer's data at present are regarded as the most complete, covering a much wider range more accurately than any other investigator's results.

As a preliminary study, statistical techniques were used to see whether or not a second degree regression equation would fit the data. It was soon found that no such relation existed, but a third degree regression equation was found to fit the data better[1]. An equation higher than third degree may give an even better fit, but it would use too much computer time to be justified. Unfortunately the third degree equation, fitting the data ranging from drop diameter 0.1 mm to 5.8 mm, is still unsatisfactory. The maximum residual, about 11 cm sec^{-1}, occurred at drop diameter 1 mm, and the maximum percentage residual was found at small drop sizes (see Figure 21, Table 6).

It is known that for $Re \leq 200$, corresponding to maximum drop diameter about 0.9 mm, the drag on a water drop can be well represented by the drag on a sphere. Water drops having these Reynolds numbers do not greatly deviate from sphericity. For $200 \leq Re \leq 500$, the drag of water drops is larger than that of spheres. This is caused by the flattening of larger drops with an increase in horizontal cross sections. The empirical curve indeed shows this variation beginning

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[1] No published solution was known at the time the regression equation was found. However, subsequently to our solving this problem we came across a paper (G.B. Foote and P.S. du Toit, 1969) which agreed with our results.

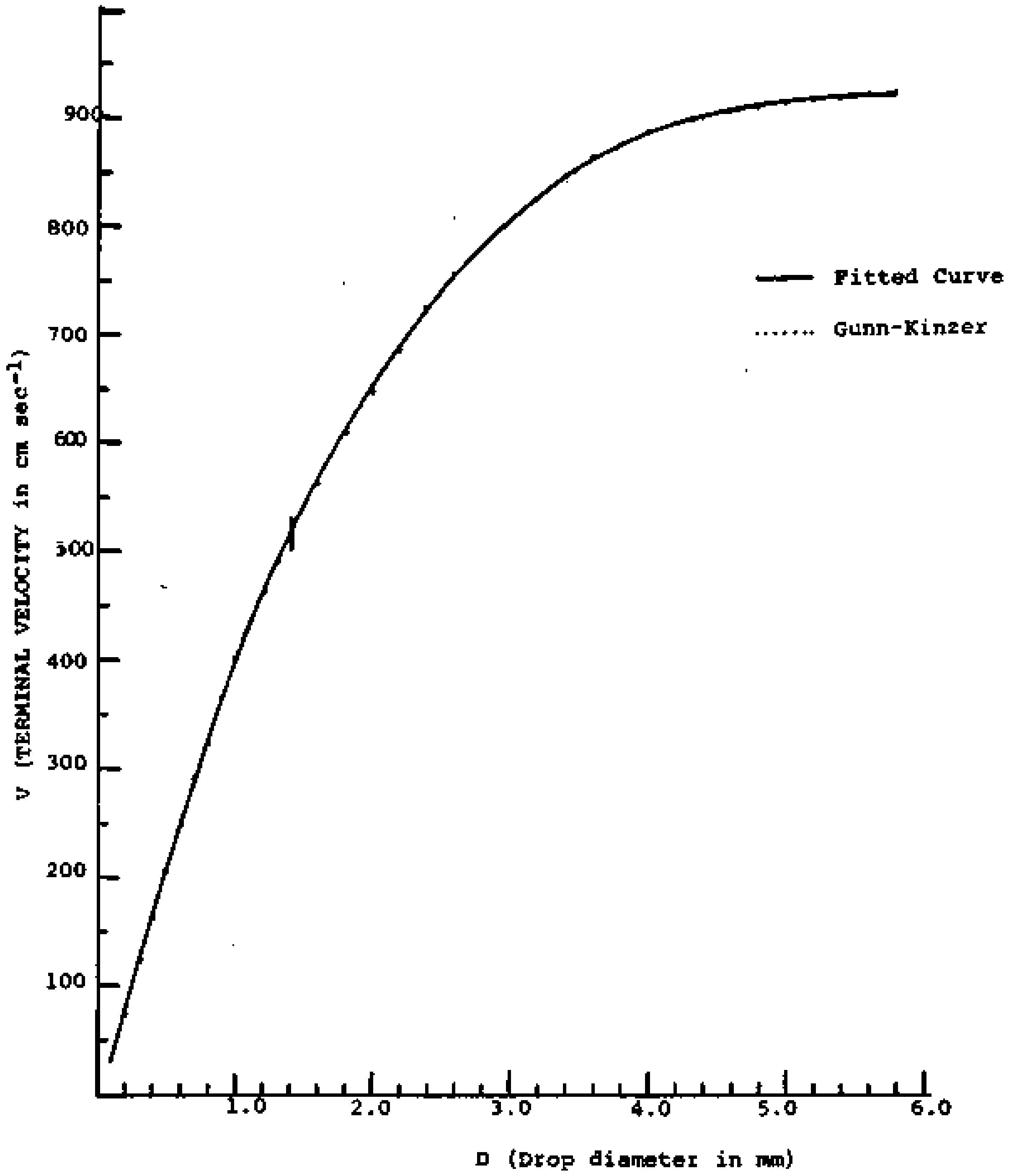

Figure 21. Results of third degree least squares fit to terminal fall speed data of Gunn and Kinzer (1949) for the diameter range 0.1 to 5.8 mm (see also Table 6).

Table 6. Comparison of terminal velocity data computed from third degree regression formula against the data of Gunn and Kinzer (1949).

Drop Dia. (mm)	(Gunn-Kinzer) Terminal Velocity (cm sec^{-1})	Estimated (cm sec^{-1})	Residual %	Drop Dia. (mm)	(Gunn-Kinzer) Terminal Velocity (cm sec^{-1})	Estimated (cm sec^{-1})	Residual 1%
0.1	27	31.69	-15.0				
0.2	72	78.26	-8.7	3.2	826	827.83	-0.2
0.4	162	166.28	-2.6	3.4	844	844.85	0.0
0.6	247	247.72	-0.3	3.6	860	859.25	0.0
0.8	327	322.85	1.3	3.8	872	871.29	0.0
1.0	403	391.93	2.8	4.0	883	881.23	0.2
1.2	464	455.22	1.9	4.2	892	889.34	0.3
1.4	517	512.99	0.8	4.4	898	895.87	0.2
1.6	565	565.50	0.0	4.6	903	901.10	0.2
1.8	609	613.00	-0.6	4.8	907	905.28	0.2
2.0	649	655.79	-1.0	5.0	909	908.68	0.0
2.2	690	694.10	-0.6	5.2	9.2	911.56	0.0
2.4	727	728.20	-0.2	5.4	914	914.19	0.0
2.6	757	758.36	-0.2	5.6	916	916.83	-0.1
2.8	782	784.85	-0.3	5.8	917	919.74	-0.3
3.0	806	807.91	-0.2				

(1) Regression equation of degree 3
$V = -16.603 + 491.844D - 88.801D^2 + 5.488D^3$
Where V in cm sec^{-1}, D in mm are terminal velocity and drop dia. respectively.

(2) Minus sign indicates that the experimental value is less than the estimated value.

at drop diameters in the range from 1 mm to 1.4 mm. Upon further study it was found that, if the data are divided into 2 regimes, one from 0.1 mm to 1.4 mm, the other from 1.4 mm to 5.8 mm (as shown in Figure 22, Tables 7a and 7b), the third degree regression equation gives very good agreement with the values given by Gunn and Kinzer. The maximum percentage residual is reduced to less that 1% by this device, and the physical change appears to justify it.

5) Conclusion

Although we have by the described procedure obtained a satisfactory empirical equation for terminal fall speed in terms of drop diameter, we have also opened a series of questions with respect to the physical causes underlying the results. It appears, in fact, that at low Re the drops actually fall more rapidly than solid spheres would, apparently because of internal circulations in the drops. Other phenomena are anticipated: (a) the onset of flow separation, (b) the flattening of drops, (c) the oscillation of large drops. As a result of these, it appears that the concept of a unique terminal fall speed is scarcely valid for medium to large raindrops, because each size of vibrating drop must have a range of fall speeds that might better be expressed by an amplitude and a frequency. These facts are of fundamental interest, and are undoubtedly significant in relation to the scavenging action of rain. We plan to extend these investigations.

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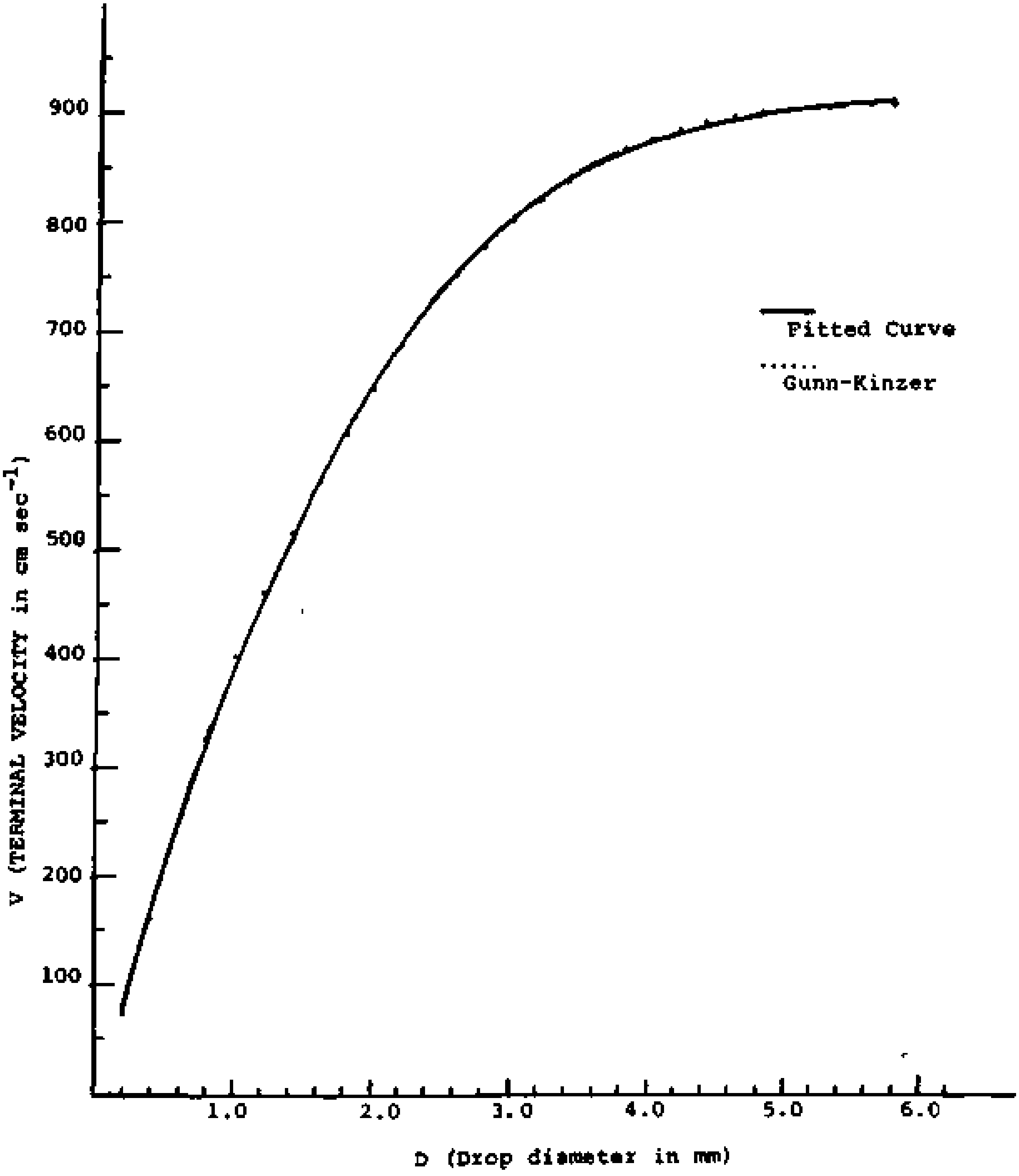

Figure 22. Results of third degree least squares fit to terminal fall speed data of Gunn and Kinzer (1949) for the diameter range 0.1 to 1.4 mm and 1.4 to 5.8 mm (see also Tables 7a & 7b).

Table 7a. Comparison of terminal velocity data computed from third degree regression formulae for the ranges $0.1 \leq D \leq 1.4$ mm . against the data of Gunn and Kinzer (1949).

Drop dia. (mm)	(Gunn-Kinzer) Terminal velocity (cm sec^{-1})	Estimate (cm sec^{-1})	Residual %
0.1	27	27.12	-0.4
0.2	72	72.18	-0.3
0.3	117	117.02	0.0
0.4	162	161.36	0.4
0.5	206	204.93	0.5
0.6	247	247.44	-0.2
0.7	287	288.63	-0.5
0.8	327	328.22	-0.4
0.9	367	365.92	0.3
1.0	403	401.47	0.4
1.2	464	465.02	-0.2
1.4	517	516.63	0.1

(1) Regression equation of degree 3

$$V \quad -17.895 + 448.949D + 16.371D^2 - 45.951D^3$$

where V in cm sec^{-1}, D in mm are terminal velocity and drop diameter respectively.

(2) Minus sign indicates that the original value is less than the estimated value.

Table 7b. Comparison of terminal velocity data computed from third degree regression formulae for the ranges $1.4\ mm \le D \le 5.8\ mm$ against the data of Gunn and Kinzer (1949).

Drop Dia. (mm)	(Gunn-Kinzer) Terminal Velocity (cm sec^{-1})	Estimated (cm sec^{-1})	Residual %	Drop Dia. (mm)	(Gunn-Kinzer) Terminal Velocity (cm sec^{-1})	Estimated (cm sec^{-1})	Residual %
1.4	517	516.01	0.2	3.8	872	871.76	0.0
1.6	565	566.16	-0.2	4.0	883	882.49	0.1
1.8	609	611.92	-0.5	4.2	892	891.27	0.1
2.0	649	653.45	-0.7	4.4	898	898.29	0.0
2.2	690	690.99	-0.1	4.6	903	903.77	-0.1
2.4	727	724.73	0.3	4.8	907	907.90	-0.1
2.6	757	754.88	0.3	5.0	909	910.91	-0.2
2.8	782	781.63	0.0	5.2	912	912.98	-0.1
3.0	806	805.21	0.1	5.4	914	914.32	0.0
3.2	826	825.80	0.0	5.6	916	915.14	0.1
3.4	844	843.62	0.0	5.8	917	915.65	0.1
3.6	860	858.88	0.1				

(1) Regression equation of degree 3
$V = 24.166 + 448.833D - 75.626D^2 + 4.265D^3$
Where V in cm sec^{-1}, D in mmare terminal velocity and drop diameter respectively.
(2) Minus sign indicates that the original value is less than the estimated value.

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VI Publications

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COO-1407-31. Trace substances in rain water: Concentration variations during convective rains and their interpretation, by D.F. Gatz and A.N. Dingle. Scheduled to appear in Tellus, 23, no. 2, May, 1971.

COO-1407-32. Tracer indium determination in rain samples by neutron activation and radiochemical analysis, by K.S. Bhatki and A.N. Dingle. Radiochem. and Radio anal. Letters, 3, 71-79, 1970.

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COO-1407-35. Miscellaneous Report No. 2, Rain Scavenging Studies. Report on travel of 29 August - 29 September, 1969, to attend three technical conferences in Europe, by A.N. Dingle, January, 1970.

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3. Mr. Yean Lee, graduate research assistant.
5. Mr. David Curtin, graduate research assistant.
6. Mr. James Fairobent, assistant in research.
7. Mr. Robert Osburn, assistant in research.
8. Mr. John Goll, assistant in research, field phase only
9. Mr. Lawrence Krupnak, assistant in research, field phase only.

A number of personnel changes are anticipated. Dr. Bhatki is required to return to Tata Institute early in 1971; he will be replaced by B.M. Joshi, also of Tata Institute, who will join the project as radioanalytical chemist, and who will undertake graduate studies in Meteorology. Mess'rs Curtin and Fairobent have been admitted to graduate school, but both are under imminent threat of military draft. Mr. Jermaine is obligated to begin four years of U.S. Air Force service in May 1971. Mess'rs Krupnak and Goll have left the area to take employment elsewhere.

Mr. Duane Harding returns from his military service in November, 1970, and will be associated with our work upon his return as graduate research assistant.

Mr. Osburn will continue as undergraduate assistant in research.