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THE UNIVERSITY OF MICHIGAN
COLLEGE OF ENGINEERING
Department of Meteorology and Oceanography

Progress Report No. 4

RAIN SCAVENGING STUDIES

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A BRIEF SUMMARY OF PROGRESS SINCE MAY, 1967

OBJECTIVES

The objectives of the work for the Fourth Year grew out of the findings of previous years and may be summarized as follows:

1. To pursue the study of the scavenging of contaminants from the air by rain with a view toward defining the physical processes that are most effective in cleaning the air of contamination.

2. To devise experiments and techniques that may be applied to natural convective storms for the purpose of resolving the details of their circulational systems, in other words, to develop and perform experimental tests of existing and proposed convective storm models.

We proposed to work toward these objectives both by introducing new experimental efforts, and by continuing our previously developed data-gathering program and intensifying our analyses of the data in hand. In particular, in 1967, it was our aim to field a pilot experiment designed to place a controlled tracer (indium) into a convective storm, using the criterion that the clear identification of the tracer in rain samples collected at the ground should indicate feasibility of the basic idea.

Needless to say, each of the above broad objectives implies numerous subobjectives that must be attained by the developmental, experimental, and analytical efforts of the project. It is not apprrpriate here to detail these. Many are to be found in the following sections of this report, and others will be specifically enunciated as our work progresses.

DATA COLLECTION

In pursuing these objectives, we have based our program upon the observation and analysis of natural rain and the construction of a rationale of the physical processes that must produce the observed scavenging effects. In 1967 as in the previous contract years, we participated in the spring field observational program of the National Severe Storms Laboratory during the period 1 May to 15 June. In view of the above noted special objective to perform a pilot experiment using indium tracer, the emphasis of our field program was different from that of earlier efforts. It was necessary, in preparation for the tracer experiment, to develop a mobile rain-sampling procedure to complement that at our fixed station. In addition to the collection of sequential rain samples at our fixed station for β-radioactivity and pollen analysis as in former years,

it was necessary for us to collect a number of samples of natural rain for tracer background analysis. A number of additional desiderata became evident during and following the field expedition. A concise summary of data collected at our Chickasha, Oklahoma, field station is given in Tables 1, 2, and 3. Table 1 lists the samples collected for indium background determinations in untagged rain; Table 2 lists those collected for long-lived β-radioactivity and pollen measurement; and Table 3 lists samples collected for the purpose of determining their halogen content. This last set of samples has had to await the completion of our analytical laboratory. Additional collections were attempted by our mobile units in connection with the tracer experiment. Two such attempts aborted because of the vagaries of transient convective activity. The successful pilot experiment of 30 May 1967, is discussed in detail in Sections I and II of this report.

Complementary data taken at our station include surface-wind direction and speed, airborne pollen concentrations, barometric pressure, and raindrop-size distributions. As in previous years, the radiosondes, radar and synoptic weather data, the mesonetwork data, and the Agricultural Research network data are available to us, but are not presented here.

The field operational phase for 1968 was limited to the month of May. Significant data collections were made on 6, 7, 9, 13, and 31 May, and the reduction of these data has proceeded since our return from the field. Among the special experiments attempted was an airborne sampling of pyrotechnic smokes containing indium and silver iodide. The results of these efforts will be reported in a subsequent report.

ANALYSIS

Particularly because of our success during the 1967 data collection period in performing a pilot experiment with the indium tracer, a good deal of emphasis was placed upon the development, testing, and application of the required radiochemical techniques for analyzing the tracer-tagged rain samples. The results of this effort are presented in Section II of this report. Noteworthy here is the fact that our estimate of the natural background of indium in rain is based on some 29 samples. This represents a somewhat tenuous evaluation, which will be improved upon in future field and laboratory measurements.

Although in preliminary results (see letter report of 24 August, 1967, addressed to R. J. Engelmann) it was indicated that the ratio In/As would be a useful criterion, laboratory investigation showed that the arsenic had entered the system primarily as a contaminant of the hydrochloric acid. This led to further investigations of reagent contributions to the background statistics. The results of these efforts are presented in Sections I and II of this report.

One specific sub-study should be mentioned here. This concerns our effort

TABLE 1

TRACER BACKGROUND SAMPLES COLLECTED
AT THE UNIVERSITY OF MICHIGAN CHICKASHA STATION

Sample No.	Sample Volume, ml	Rainfall Rate, mm/hr	Time, CDT			Equivalent Indium, ng/ℓ
			Begin	End	Mid	
			5 May 1967			
1	932	---	a	---	---	11.7[b]
2	964	---	a	---	---	11.5[b]
3	920	---	a	---	---	9.1
4	874	---	a	---	---	7.7
5	802	---	a	---	---	3.6
6	1049	---	a	---	---	1.7[c]
7	980	1.25	1958.45	2017.26	2007.85	0.5[c]
8	950	19.83	17.26	18.41	17.83	5.3
9	962	28.16	18.41	19.23	18.82	2.4[c]
10	953	25.41	19.23	20.13	19.68	5.1
11	1018	42.12	20.13	20.71	20.42	0.9[c]
12	995	43.42	20.71	21.26	20.98	2.27[c]
13	967	27.30	21.26	22.11	21.68	0.9[c]
14	940	16.71	22.11	23.46	22.78	2.27[c]
15	948	3.35	23.46	30.26	26.86	1.6[c]
16	81	0.34	30.26	36.00	33.13	24.4[d]
17	962	1.94	42.45	54.38	48.41	1.17[c]
18	910	9.29	54.38	56.73	55.55	1.15[c]
19	950	21.31	56.73	57.80	57.26	3.0
20	915	7.96	57.80	00.56	59.18	0.605[c]
21	975	6.70	00.56	04.05	02.30	18.97
22	966	14.05	04.05	05.70	04.87	7.39
23	945	23.14	05.70	06.68	06.19	0.9[c]
24	964	24.35	06.68	07.63	07.15	3.9
25	970	36.95	07.63	08.26	07.94	3.81
26	958	57.48	08.26	08.66	08.46	4.8
27	1013	62.34	08.66	09.05	08.85	3.2
28	992	76.80	09.05	09.36	09.20	4.85
29	1001	68.64	09.36	09.71	09.53	2.8
30	995	70.24	09.71	10.05	09.88	2.2
31	1018	61.08	10.05	10.45	10.25	1.75
32	e	---	10.45	12.00	11.22	---
33	e	---	12.00	13.88	12.94	---
34	921	22.10	13.88	14.88	14.38	2.03
35	947	11.84	14.88	16.80	15.84	3.08
36	966	7.29	2116.80	2119.98	2118.39	3.44

TABLE 1 (Concluded)

Sample No.	Sample Volume, ml	Rainfall Rate mm/hr	Time, CDT			Equivalent Indium, ng/ℓ
			Begin	End	Mid	
			30 May 1967			
1	967	17.58	1234.03	1235.35	1234.69	5.7
2	951	16.30	37.31	38.71	38.01	7.7
3	958	8.61	40.88	43.55	42.21	13.5
4	856	5.17	46.01	49.98	47.99	7.9
5	925	1.05	1253.55	1314.78	1304.16	8.6
6	933	7.06	1409.16	1412.33	1410.74	2.4
7	926	0.95	1709.21	1732.59	1720.90	4.0
8	906	1.03	1733.39	1754.59	1743.99	6.5

[a]Malfunction of tape recorder, time not recorded.
[b]Internal standard correction approximate only.
[c]No internal standard correction possible, hence these In amounts are not reliable. Not used for background statistics.
[d]Fictitiously high value because of small sample volume. Sample content of In only 1.98 ng which is close to the reagent blank value.
[e]Incorrect collection procedure, hence samples discarded.

TABLE 2

RADIOACTIVITY/POLLEN SAMPLES COLLECTED
AT THE UNIVERSITY OF MICHIGAN CHICKASHA STATION

Sample No.	Volume, ml	Rainfall Rate, mm/hr	Time, CDT			β-Conc., pCi/ℓ	Residue, mg/ℓ
			Begin	End	Mid		
			5 May 1967				
1	3780	---	*	---	---	23.0	194
2	3910	---	*	---	---	18.6	132
3	3600	---	*	---	---	34.4	64.2
4	4100	5.20	2009.21	2018.68	2013.95	23.5	107.7
5	4030	27.95	18.68	20.41	19.54	25.2	90.8
6	4140	45.58	20.41	21.50	20.96	14.9	81.4
7	4110	18.27	21.50	24.20	22.85	16.5	40.1
8	1570	1.60	24 20	36.01	30.11	25.9	91.1
9	4030	3.28	42.20	56.93	49.56	14.1	60.4
10	3870	10.16	56.93	2101.50	59.21	9.4	39.3
11	3900	10.22	2101.50	06.08	03.79	12.7	76.1
12	3835	24.88	06.08	07.93	07.00	8.0	88.0
13	4010	49.10	07.93	08.91	08.42	7.9	46.6
14	4200	66.31	08.91	09.67	09.29	11.7	37.6
15	4250	65.38	09.67	10.45	10.06	10.1	32.7
16	4075	65.20	10.45	11.20	10.86	7.64	25.5
17	4180	62.70	11.20	12.00	11.60	13.5	31.8
18	4105	51.31	12.00	12.96	12.48	6.05	36.0
19	4020	30.34	12.96	14.55	13.75	5.1	30.8
20	3860	13.43	14.55	18.00	16.27	14.3	28.2
21	1290	2.09	2118.00	2125.41	2121.70	14.3	74.4
			20 May 1967				
1	4160	15.60	1044.30	1047.50	1045.90	21.0	130.4
2	4310	103.4	47.50	48.00	47.75	18.25	84.2
3	4250	51.00	48.00	49.00	48.50	15.1	43.8
4	4320	62.46	49.00	49.83	49.41	11.7	33.0
5	4255	76.21	49.83	50.50	50.16	10.05	53.4
6	4290	88.76	50.50	51.08	50.79	9.45	27.7
7	4360	88.68	51.08	51.67	51.37	9.63	23.95
8	4100	72.35	51.67	52.35	52.01	8.4	28.0
9	4280	70.36	52.35	53.08	52.71	8.87	24.7
10	4300	36.34	53.08	54.50	53.79	8.96	21.3
11	4260	34.08	54.50	56.00	55.25	8.35	23.8

TABLE 2 (Continued)

Sample No.	Volume, ml	Rainfall Rate, mm/hr	Time, CDT			β-Conc., pCi/ℓ	Residue, mg/ℓ
			Begin	End	Mid		
			20 May 1967 (Concluded)				
12	4180	35.83	1056.00	1057.40	1056.70	8.34	20.9
13	4225	33.36	57.40	58.92	58.16	7.8	19.3
14	4190	33.97	58.92	1100.40	59.66	6.8	12.9
15	4360	26.83	1100.40	02.35	1101.37	6.86	18.6
16	4170	35.74	02.35	03.75	03.05	8.0	14.9
17	4280	29.35	02.75	05.50	04.63	8.0	14.6
18	4220	14.15	05.50	09.08	07.29	8.69	14.3
19	4320	7.41	09.08	16.08	12.58	6.39	13.65
20	4170	10.72	16.08	20.75	18.41	3.9	10.9
21	1520	1.03	20.75	1138.50	1129.63	5.57	37.8
22	4320	34.56	38.50	40.00	39.25	8.22	15.15
23	4290	22.88	40.00	42.25	41.13	5.84	13.7
24	4360	52.32	42.25	43.25	42.75	3.12	10.2
25	4240	50.88	43.25	44.25	43.75	6.27	17.9
26	4090	25.56	44.25	46.17	45.21	8.21	36.1
27	3900	8.25	46.17	51.84	49.01	8.49	18.6
28	730	0.38	1151.84	1214.67	1203.26	11.7	64.0
29	1735	1.56	1214.67	1228.00	1221.34	20.4	71.0
			29 May 1967				
1	4200	3.52	0847.45	0901.78	0854.62	3.92	15.25
2	4200	2.77	0901.78	0919.95	0910.87	9.78	16.15
3	4200	3.05	19.95	36.48	28.21	5.84	23.2
4	4100	3.06	36.48	52.55	44.51	4.7	18.4
5	4230	2.83	52.55	1010.51	1001.53	5.39	30.2
6	3980	1.49	1010.51	1042.53	1026.52	4.21	20.6

TABLE 2 (Concluded)

Sample No.	Volume, ml	Rainfall Rate, mm/hr	Time, CDT			β-Conc., pCi/ℓ	Residue, mg/ℓ
			Begin	End	Mid		
			30 May 1967				
1	4250	7.42	1228.96	1235.83	1232.39	28.2	136.5
2	4220	18.90	35.83	-38.51	37.17	20.6	64.2
3	4035	10.83	38.51	42.98	40.74	21.8	56.9
4	4010	10.51	42.98	47.56	45.26	19.6	46.9
5	4160	1.60	47.56	1318.85	1303.20	24.0	38.9
6	2820	1.99	1318.85	35.88	27.36	12.1	29.5
7	3900	2.27	35.88	1356.55	46.21	9.94	26.6
8	4090	3.43	56.55	1410.85	1403.70	7.85	26.1
9	4110	5.55	1410.85	19.73	15.29	6.67	29.1
10	1960	0.84	1419.73	1447.88	1433.80	14.3	35.1
11	4030	1.02	1706.25	1753.59	1729.92	3.58	61.4

*Malfunction of tape recorder, time not recorded

TABLE 3

SAMPLES COLLECTED AT THE UNIVERSITY OF MICHIGAN CHICKASHA STATION FOR HALOGEN ANALYSIS

Sample No.	Time, CDT Begin	Time, CDT End
	20 May 1967	
1	1108.00	1108.20
2	09.20	09.30
3	12.00	12.15
4	13.45	14.15
5	15.45	15.55
6	1146.10	1146.20
	29 May 1967	
1	0849.26	0850.00
2	52.90	53.78
3	56.85	57.50
4	59.80	0900.45
5	0907.85	08.55
6	13.55	14.81
7	20.48	21.70
8	25.70	26.88
9	33.45	34.45
10	39.51	40.58
11	45.53	46.53
12	58.81	1000.06
13	1008.00	09.18
14	17.78	19.03
15	24.78	26.46
16	32.55	37.63

Sample No.	Time, CDT Begin	Time, CDT End
	30 May 1967	
1	1233.00	1233.60
2	36.05	36.21
3	39.51	39.73
4	44.50	44.75
5	50.81	51.65
6	1332.95	1333.91
7	36.90	37.31
8	38.86	39.38
9	41.38	42.13
10	43.88	34.71
11	46.11	47.11
12	50.85	51.61
13	1412.75	1412.93
14	14.06	14.26
15	15.86	16.30
16	17.55	18.03
17	26.45	27.46
18	29.50	33.40

to determine the particle-size distribution of the indium-bearing plume generated from pyrotechnic flares. For this purpose, we submitted samples of the pyrotechnic material to Professor B.Y.H. Liu, University of Minnesota, for analysis in the Particle Technology Laboratory. The results of this study are summarized in Figure 1.

Because of the circumstances of sampling the pyrotechnic smoke in the laboratory, however, Professor Liu expressed doubt as to the relation of this measurement to the conditions of the actual airborne experiment (flares burned aft of aircraft wing at speeds of 120-130 mph). Figures 2 and 3 are electron microphotographs of some of the smoke particles generated by burning the flare material in a small chamber and sampling the smoke through a dilution chamber. Most of the aggregation shown in Figure 2 probably takes place within a few milliseconds of ignition of the material. The rapid ventilation and dilution of the flare smoke at flying speeds could conceivably prevent this process from going so far.

We are continuing our effort to determine this particle-size distribution by means of an airborne experiment in which a sampling airplane pursues an emitting airplane and procures samples from the flare plume actually generated under field experimental conditions.

PUBLICATIONS

The following publications have resulted from the work under our contract during the past year:

1. COO-1407-13. Rainwater impurities: implications of concentration changes during convective rains, by D. F. Gatz. (Abstract) Bull. Am. Meteorol. Soc. 48 (1), 24, 1967.

2. COO-1407-14. Low-altitude input of artificial radioactivity to a convective storm—comparison with deposition, by D. F. Gatz. J. Appl. Meteorol. 6 (3), 530-535, 1967.

3. COO-1407-15. Progress Report No. 3, Rain Scavenging Studies, by A. N. Dingle and D. F. Gatz. May, 1967.

4. COO-1407-16. Air cleansing by rain, by A. N. Dingle. Submitted to J. Atmos. Sci., 1967.

5. COO-1407-17. Tracers for the study of scavenging by rain, by A. N. Dingle. Proc., USAEC Meteorological Conference, 11-15 September, 1967, Chalk River, Ontario.

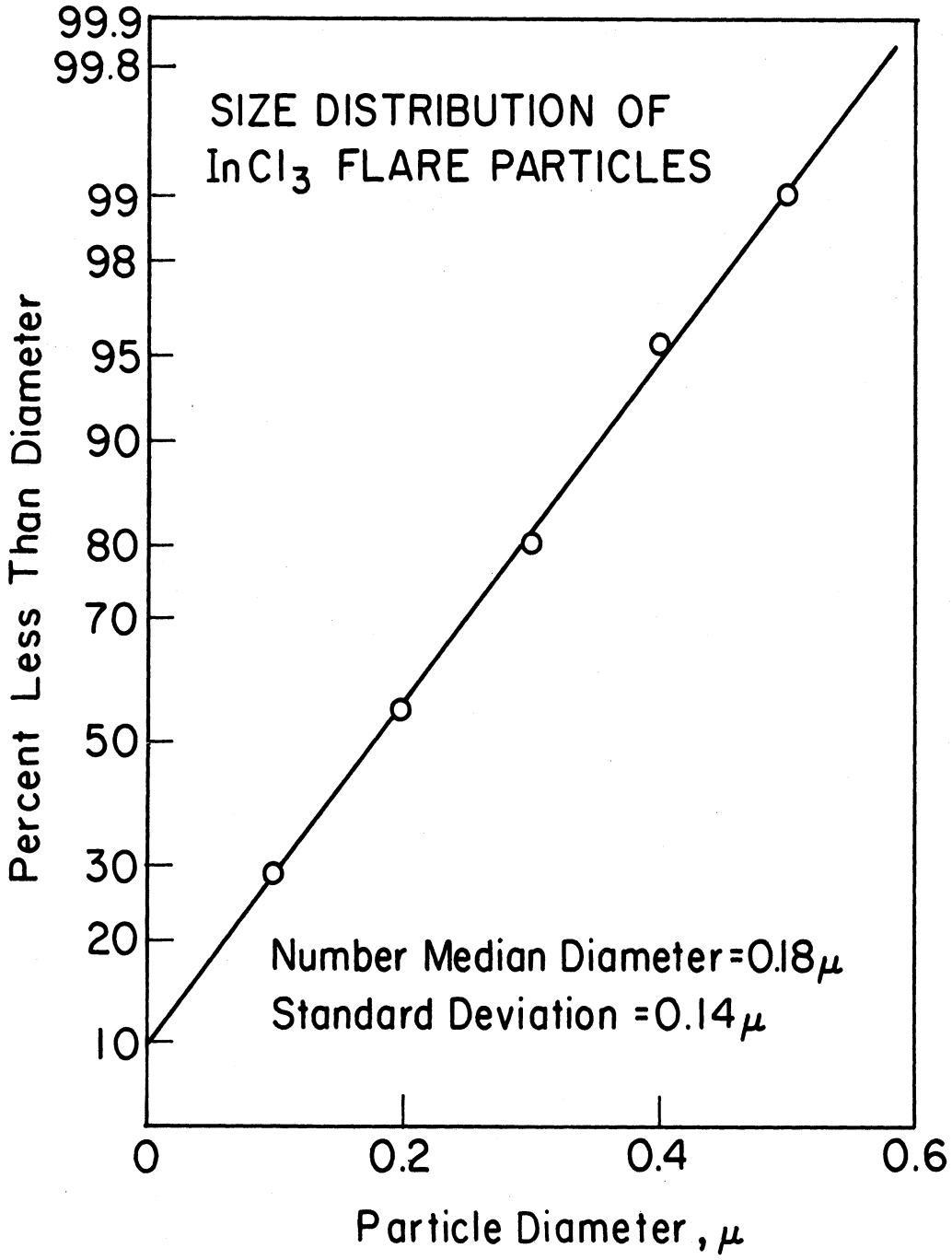

Figure 1. Size distribution of particles formed by ignition of pyrotechnic flare material containing indium. Smoke was drawn through a dilution chamber and sampled after precipitation of large aggregates (see Figure 2).

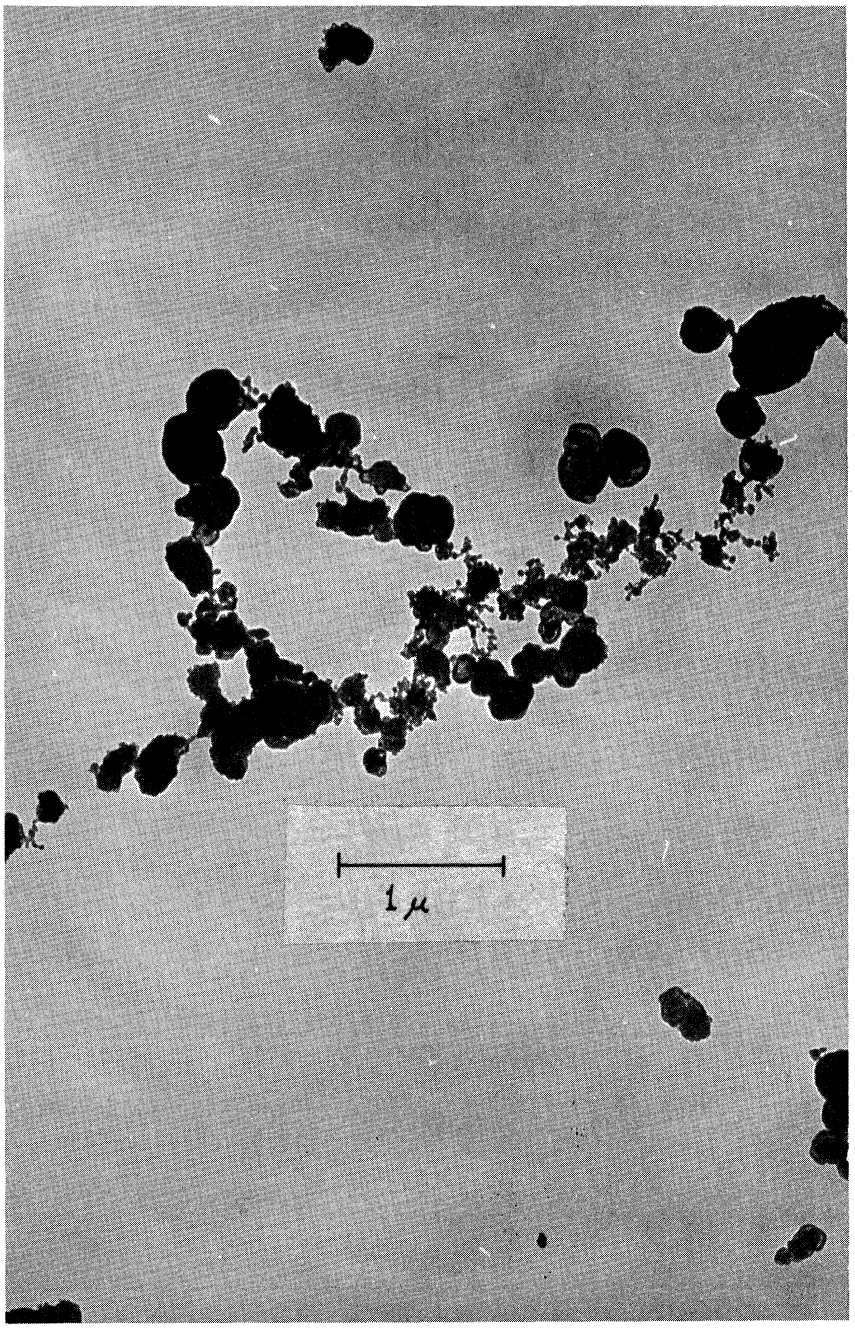

Figure 2. Aggregates formed in vapor from pyrotechnic flares burning in a small chamber with no ventilation. It is not yet certain to what extent aggregation is permitted when the flares are ventilated at normal flying speeds (120-130 mph).

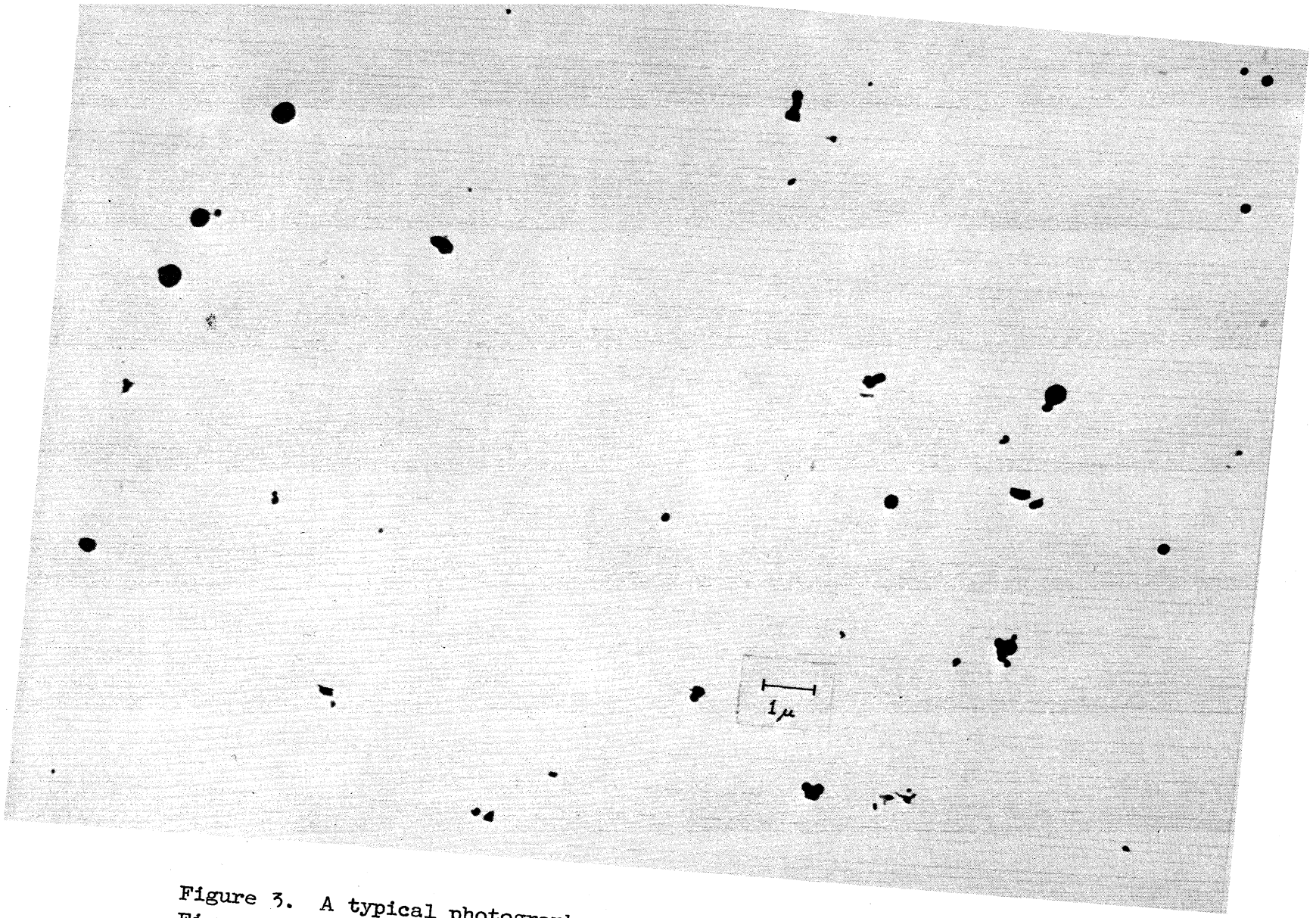

Figure 3. A typical photograph of the particles counted to construct Figure 1. The ventilation present at 120-130 mph under field operating conditions may be sufficient to yield a large proportion of the mass of the flare smoke in particles of these sizes and shapes.

6. COO-1407-18. A pilot experiment using indium as tracer in a convective storm, by A. N. Dingle, D. F. Gatz, and J. W. Winchester. (Abstract) Bull. Am. Meteorol. Soc. 48 (11), 835, 1967.

7. COO-1407-19. Detection of indium as an atmospheric tracer by neutron activation, by D. F. Gatz, J. W. Winchester, and A. N. Dingle. (Abstract) Bull. Am. Meteorol. Soc. 48 (11), 835, 1967.

The last two of these have been prepared as companion technical papers for submission to the Journal of Applied Meteorology, and are included as Sections I and II, respectively, of this report. In addition, a technical paper entitled "Trace Substances in Rainwater: Physical Processes Implied by Concentration Changes During Convective Rains" has been prepared for submission to Tellus. This paper is included as Section III of the present report.

PERSONNEL

As of 21 March 1968 Dr. Gatz terminated his association with the project. It is perhaps appropriate to note that he and the writer together developed this line of research through his doctoral research and thesis and an additional period of nearly two years of post-doctoral work. We hope to find further opportunity for collaboration with him in his new post at Argonne National Laboratory.

For the 1968 field program, Messrs. Tom Grayson, David Curtin, James Fairobent, and Stephen Jermaine were employed as research assistants. Mr. Grayson has returned to this University as a graduate student having completed four years of service as an U. S. Naval officer. The other three men have finished two years of our undergraduate program in meteorology. Each has indicated interest in continuing his connection with the project, and each has contributed effectively in both data collection and analysis at the time of this writing.

Section I

A Pilot Experiment Using Indium as Tracer in a Convective Storm

by
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ABSTRACT

In an effort to determine whether it is feasible to use tracer techniques in the study of circulations and rain scavenging processes in severe convective storms, a pilot experiment using indium as tracer was conducted. A total of 200 gm of indium was released over a period of 21 min into the updraft feeding a relatively small convective system. The tracer was released by means of pyrotechnic flares from an airplane flying at about 3200 ft. altitude. The rainfall from the system was sampled at the ground by means of an array of samplers placed and recovered by two mobile units. Analysis of the samples compared against analyses of untagged rain samples and reagent backgrounds of indium, distinctly indicates the presence of tracer indium in a reasonable distribution pattern.

I. Introduction.

The experiment described here is a remarkably simple one, but one for which the chances of success seem rather small. As an outgrowth of several years' studies of the removal of contaminants from the air by rain (Gatz and Dingle, 1963; Dingle and Gatz, 1966; Gatz, 1967), we came to the decision to try to place our own tracer into a convective rain-generating system to see whether we could recover it in measurable quantities in rain samples collected at the ground downstream.

Experiments such as this are necessary for some and significant for all of the following areas of current interest:

a) the natural cleansing of the air by rain

b) the details and the effects of the circulations within convective storms, and

c) the possible beneficial application of modification techniques to violent convective storms.

A more complete discussion of each of these areas is in preparation (Dingle, 1968) although basic information on the first two can be found in Junge (1963), Browning (1964), Dingle (1965), and Newton (1966) for example.

II. Objective.

The objective of this initial experiment was solely to determine whether such a tracer experiment is feasible. In placing tracer material into the updraft of a strong convective

storm, one may have considerable doubt. The principal criterion for success at this stage is therefore that the tracer be clearly identified and measured in rain samples collected so as to bear a reasonable relationship to the initial placement of tracer.

This overall criterion obviously encompasses others, such as the feasibility of striking a "target" area with tracer tagged rain, and that of coordinating adequately the tracer-releasing aircraft with the ground-level rain sampling system. It suggests but does not include the more comprehensive objective of sampling so as to determine the ground-level deposition pattern of the tracer, and thence estimating the overall rates of diffusion achieved by the diverse processes of circulation, condensation, scavenging, and precipitation.

III. Selection of Tracer and Emission System.

The factors which control the selection of a suitable tracer material include those properties that determine how it may be dispensed and how scavenged, and the technique that will be used to identify and measure it. Of these, the last is the *sine qua non*, so it was chosen first.

On the basis of sensitivity and demonstrated reliability in addition to the accessibility of the necessary neutron source and facilities at the Phoenix Memorial Laboratory, The

University of Michigan, we decided to use neutron activation analysis. Consideration of the nuclides and isotopes that might be used (Goldman and Stehn, 1962) led to the selection of indium as the element best suited to measurement by neutron activation. In this search we were aided materially by earlier work done by Duce, et al (1963), Gordon and Larson (1964), and Jones, et al (1967).

Stable In-115 is converted to In-116m by neutron capture. The latter is radioactive, emitting both beta-and gamma-rays with a half-life of 54 min. The neutron capture cross-section is 150 barns, so the activation is relatively efficient. Details of the analytical procedure are presented elsewhere (Gatz, et al., 1968). Suffice it here to say that the sensitivity limit for the detection of indium in our laboratory is 10^{-10} gm within ±5%.

Our selection of the technique for emitting tracer was influenced strongly by developments that had already taken place in silver iodide cloud seeding technology (Fuquay, 1960; St. Amand and Donnan, 1963; Finnegan, et al, 1967). Our needs appeared to be very similar to those of the cloud seeders: small particle-size, airborne emission with precise placement, and good control of the quantity of material emitted. For several reasons involving both procurement and operations, we chose to use pyrotechnic flares containing In-115 as $InCl_3$ as the source of our tracer. These burn at an estimated temperature of about 1200C, vaporizing the indium trichloride, and

probably producing chemical complexes of In with Cl, O, and some of the metallic constituents of the pyrotechnic compound (e.g., K and Mg). This chemical behavior is currently under study. The flares are carried at the trailing edge of the aircraft wing by means of supports designed for the purpose, and each flare is activated by electrical ignition from the cockpit.

IV. Experimental procedure.

The site and the season for the experiment were dictated by our collaboration with the National Severe Storms Laboratory during late spring. By this arrangement, mesonetwork data, special soundings, and comprehensive radar data of high quality are available to establish the meteorological setting of our various field experiments in substantial detail.

In particular, this pilot experiment was conducted on 30 May 1967 on a relatively small convective storm within a pre-cold-frontal squall line near the town of Holdenville, Oklahoma. The location is shown in Figure 1, the synoptic situation in Figure 2, and pertinent radar PPI echo contours are shown in relation to the sampling array in Figure 3.

Emission of indium tracer was initiated at 1511 CDT at an altitude of 3200 ft MSL in an updraft estimated at 1400 ft/min. The initial point of release was 6 mi. west of Holdenville, and 2 mi east of the rain curtain. (Figure 4).

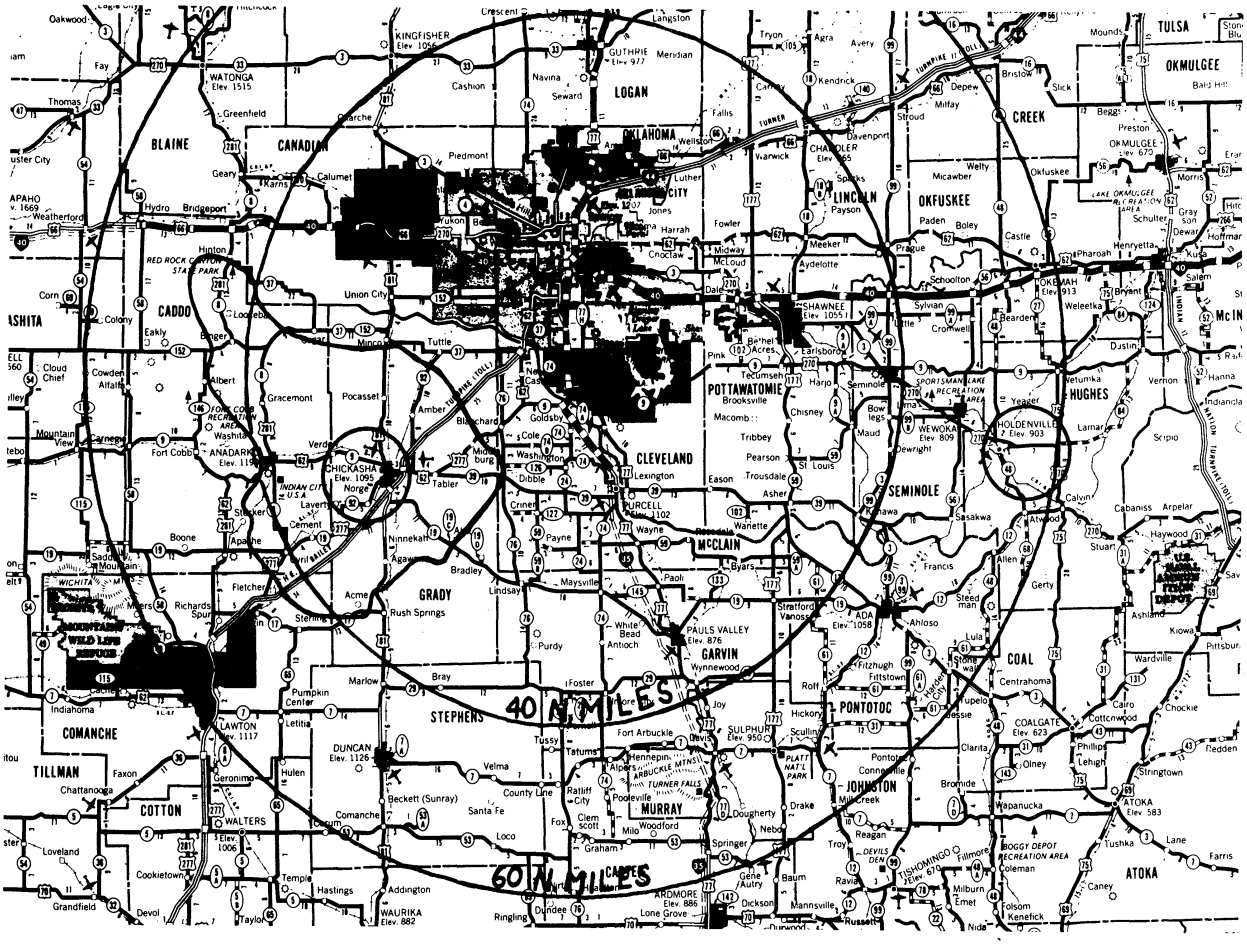

Figure 1.

Map showing location of the pilot tracer experiment near Holdenville, Oklahoma, in relation to the National Severe Storms Laboratory at Norman (40 and 60 n. mi range circles centered on NSSL radar), the University of Michigan field station at Chickasha, and the Agricultural Research Service Washita River Watershed study area (irregular figure which includes Chickasha). The entire area shown is within the mesonet of stations operated by NSSL.

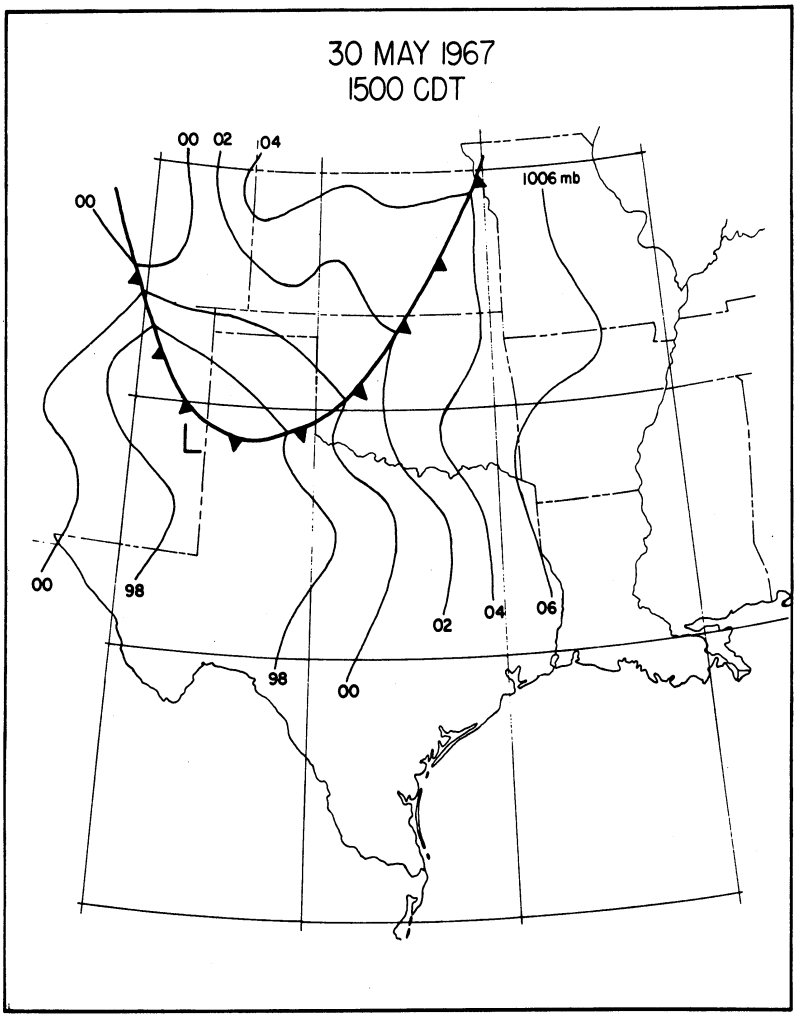

Figure 2.

Local synoptic weather map showing cold front position as of 1500 CDT on 30 May 1967. Squall activity which developed ahead of the cold front in the southerly current was not organized into a well-defined line at this time.

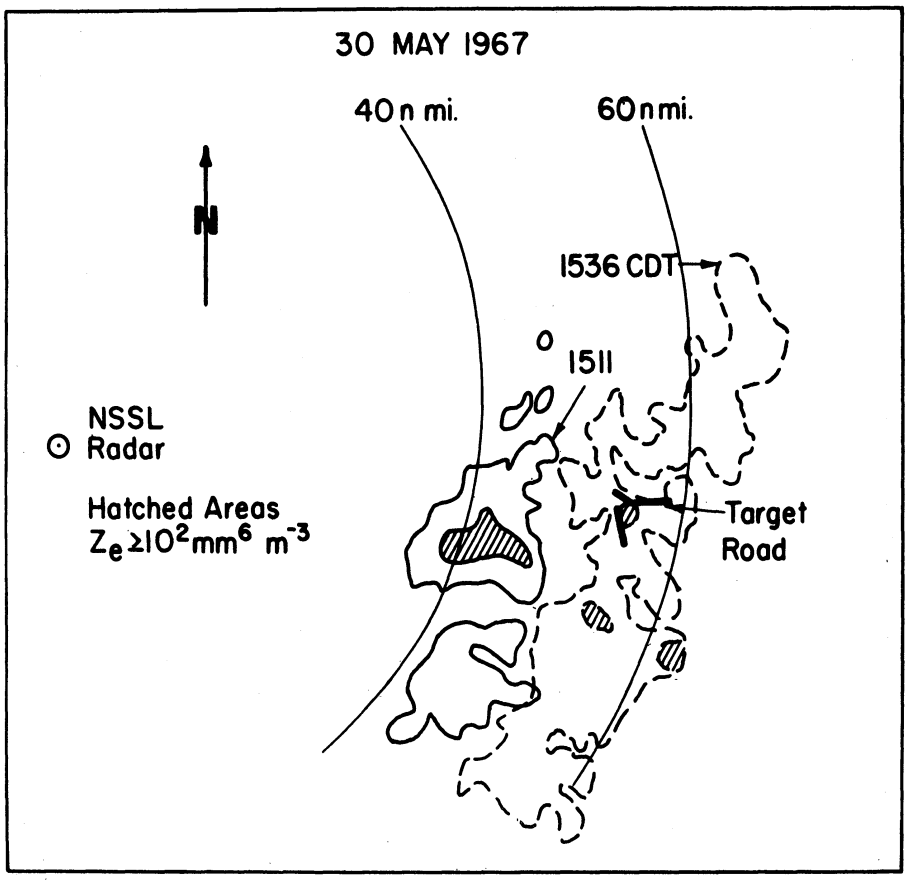

Figure 3.

PPI - scope photographs showing airborne precipitation at the time of the tracer experiment. Alternate shaded and unshaded areas indicate the ten fold increase of echo intensity, expressed as mm^6/m^3, thus the small round echo which appears to have been the tracer-innoculated shower is of 10^2 mm^6/m^3 nominal intensity.

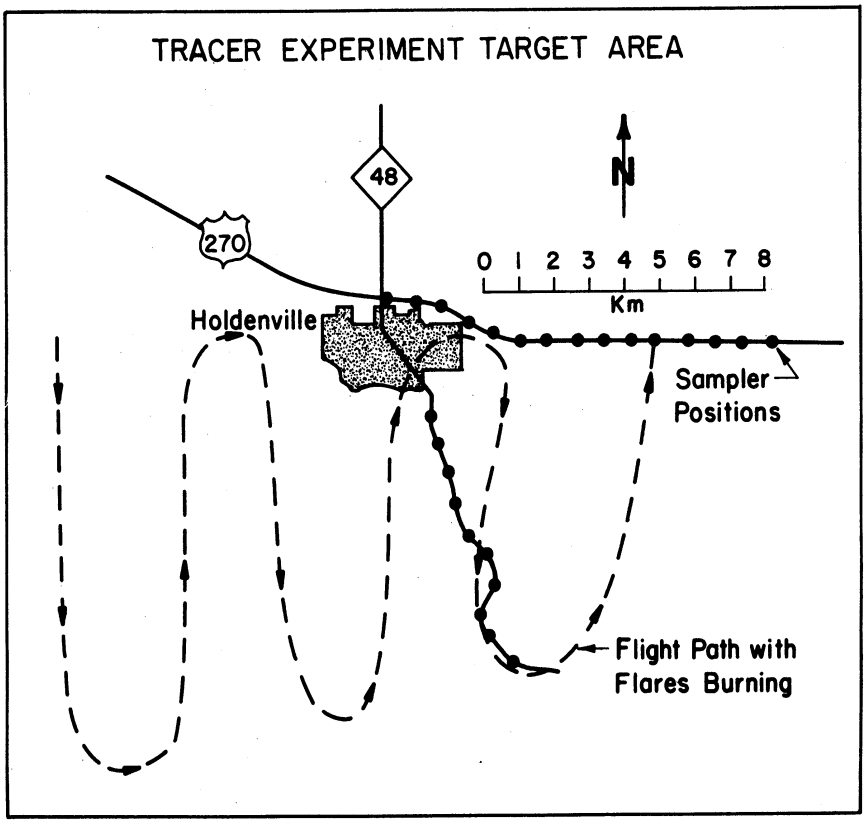

Figure 4.

Path traveled by the airplane during emission of indium tracer. Start of emission at 1511 CDT, end at 1532 CDT. Position of airplane was maintained in the major leading updraft about 2 mi east of the leading edge of the rain. Updraft speed decreased from about 1400 ft/min to 500 ft/min and width changed from about 5 mi to about 3 mi during the tracer emission period. Ground level samplers (Figure 5) were placed as shown by the dots along highways U.S. 270 and Oklahoma 48.

The airplane traversed the updraft on a track roughly perpendicular to the storm movement, made a 180° turn and retraversed the updraft and repeated this procedure, staying at a nearly constant distance ahead of the rain curtain. The emission continued for 21 min during which a total of 200 gm In was put out. The updraft decreased in width and intensity during this time from ~ 5.0 mi, 1400 ft/min to ~ 3.0 mi, 500 ft/min.

Rain samples were collected at ground level at the points indicated in Figure 4. This operation was done using carry-all trucks, each manned by a driver and an assistant, and supplied with 20 roadside samplers (Figure 5) previously prepared for rain collection. In setting the samplers, each is pre-innoculated with 50 ml of 6 N HCl and is then attached to a roadside fence for support against wind gusts.

As indicated by the map (Figure 1), the mobile units had to operate a long way from their home base in this case. The temporal coordination of the operations was therefore less than ideal, and most of the samplers were placed after the rain had begun at their respective stations. Samplers were placed along each road between 1528 and 1553 CDT, the sequences being from west to east and from north to south respectively. They were retrieved immediately following the rainshower, finishing by 1645 CDT.

Figure 5.

Samplers distributed by the mobile units are plastic yard baskets of 20-in. diameter at the mouth, lined with polyethylene bags. The liner is held by a rubber band which also serves to support the unit against a roadside fence. The samplers are pre-innoculated with 50 ml of 6 N HCl to assure acid solution of all tracer indium in the collected sample.

V. Results.

The chemical and radioactivation analysis procedures are presented in a companion paper by Gatz, et al (1968). Backgrounds of natural and reagent indium were determined by the analysis of samples procured from untagged rain showers (29 samples), and of reagent blank tests in the laboratory. The reagent blanks (16 samples) contributed 1.5 ± 1.5 ng In/liter according to these tests, and the untagged rain background turned out to be 6 ± 3 ng In/liter. These figures must then serve as the basic criterion for judging the significance of the In amounts found in the tracer-tagged rain samples.

Figure 6 shows how the indium concentrations found in the tagged rain compare with those in the untagged rain. All but two of the background (untagged) rain samples contained less than 12 ng In/l, the largest number of samples being at the 2 to 4 ng In/l level. Half of the tracer-tagged rain samples contained more than 12 ng In/l, the highest value being 40 ng In/l.

The In concentrations found in the tracer-rain samples are given as a function of position along the east-west road in Figure 7 and along the north-south road in Figure 8. The results of the background study are blocked in. The two distinct levels of In concentration shown in Figure 7 evidently show significant amounts of the In tracer near the

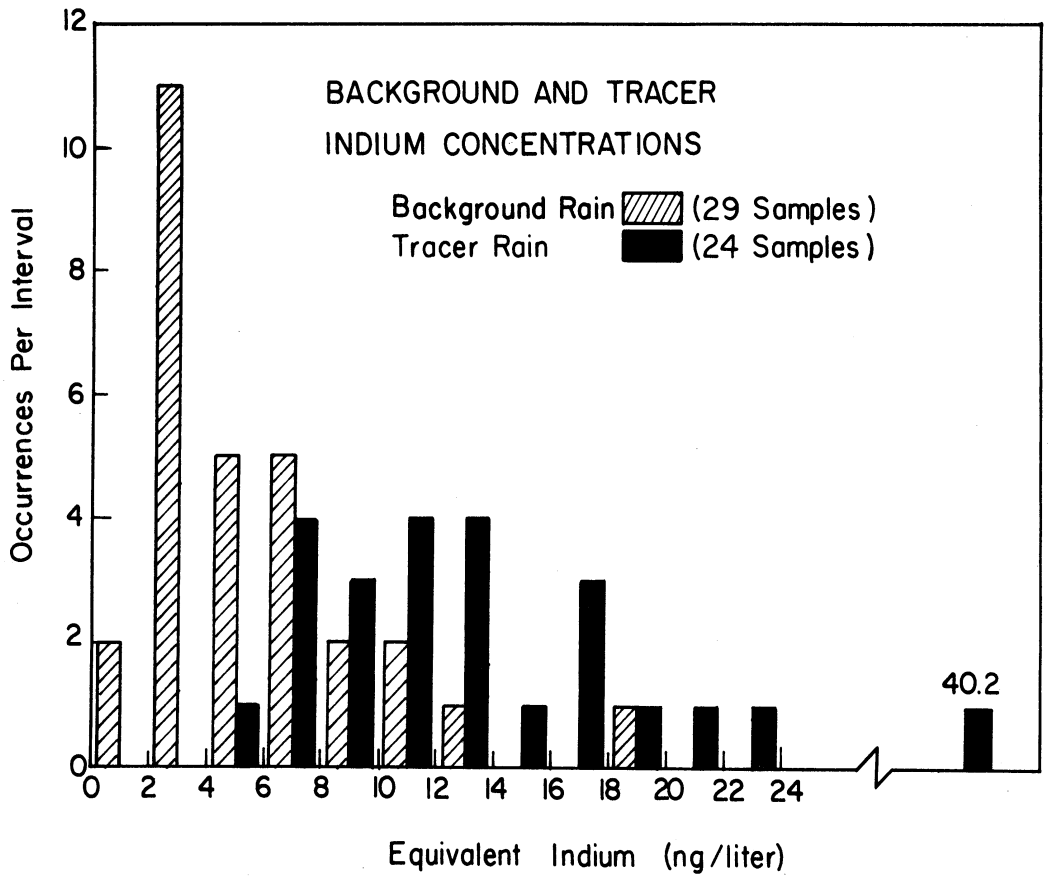

Figure 6.

Frequencies of occurrence of different concentrations of indium in samples from tracer-tagged and background (untagged) rain showers.

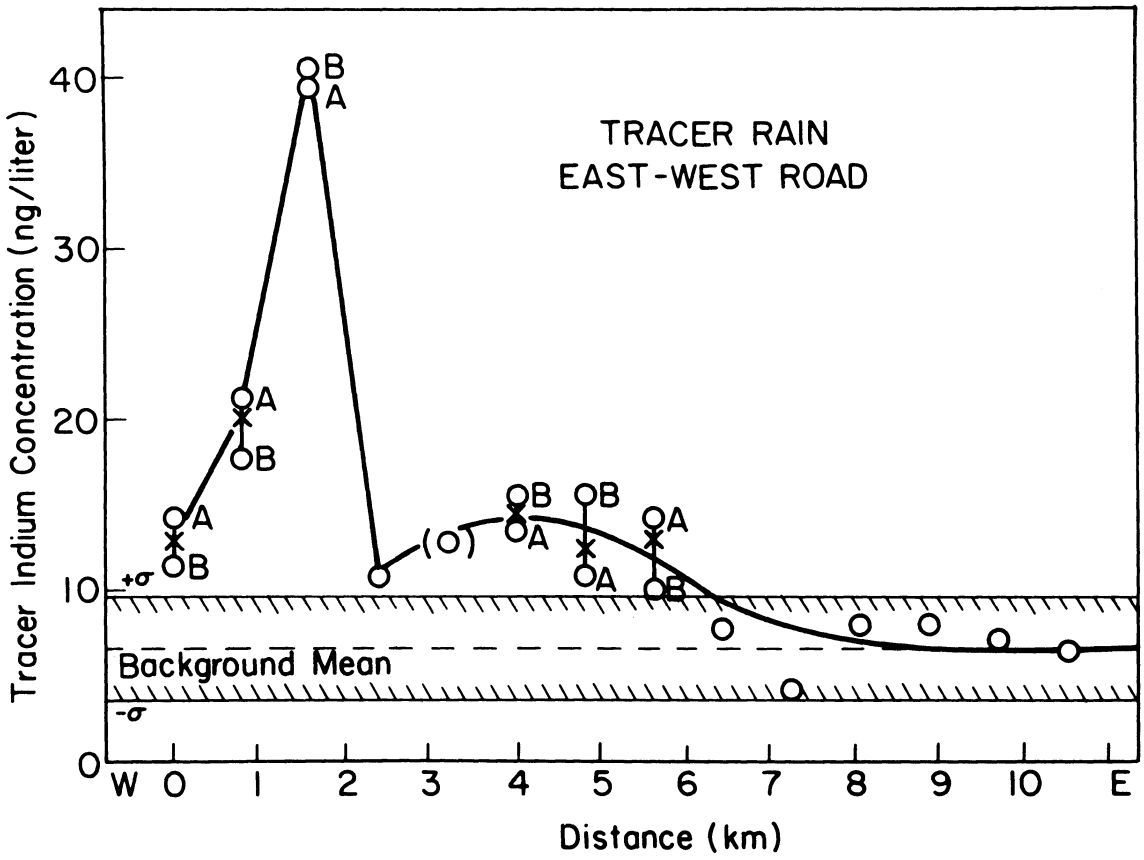

Figure 7.

Variation of indium concentration with distance along the east-west road (U.S. 270). Only the first three samplers were in position to sample throughout the rain shower. Those farther east were all placed after the rain had begun, and they were placed increasingly later with respect to the rain core as the array was extended eastward.

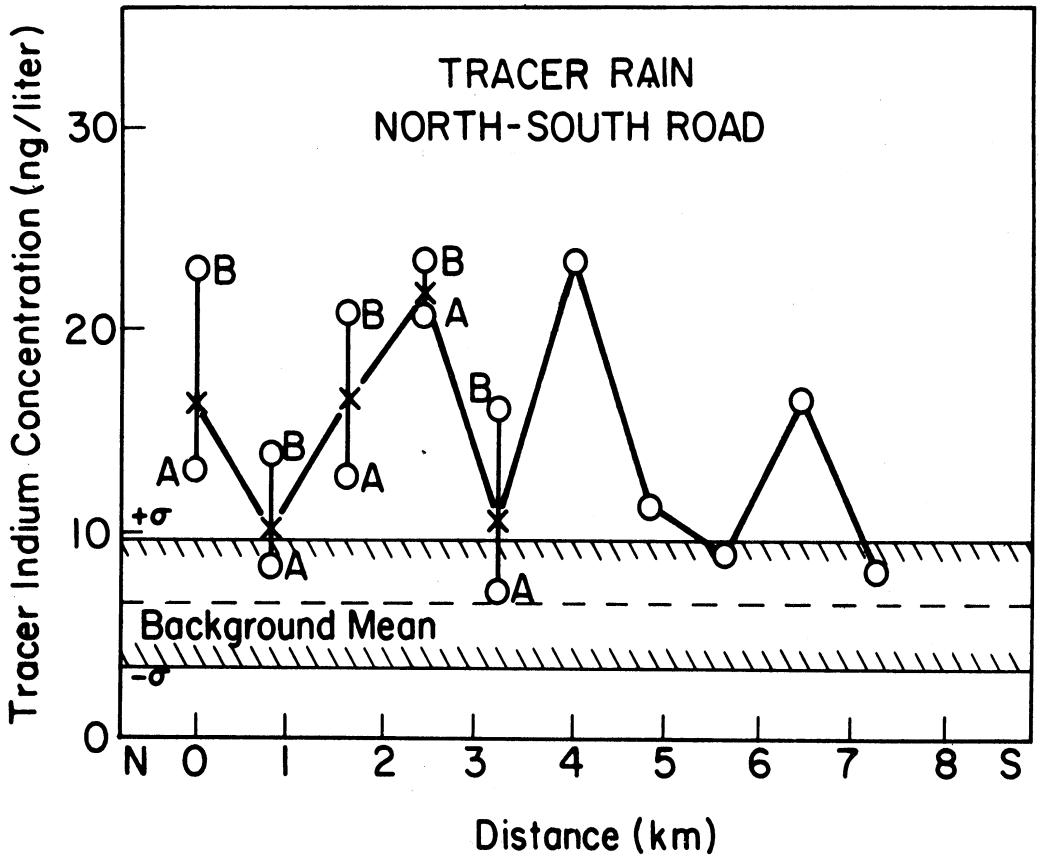

Figure 8.

Variation of indium concentration with distance along the north-south road (Oklahoma 48). Rain began just prior to setting the third sampler (at ~ 1.7 km).

west end of the array as contrasted to natural or background amounts at the east end.

Ordinarily, the concentrations of naturally occurring contaminants in convective showers are high in the initial light rain, decrease to a minimum at or near the rainfall rate maximum or core of the shower, and then increase again in the more stable light rain at the end. This sequence of concentration changes is also reported by Gatz, et al. (1968) for In in one shower. In the present instance, we find evidence of the tracer In plume in the samples taken along the east-west road by virtue of a strong deviation from the usual scavenging pattern. Various considerations enter. One is that only the first three samples (at $\sim$ 0, 1, and 1.7 km) in Figure 7 represent the whole shower. All of the other samplers were set out after the rain had begun at the respective stations, and that at $\sim$ 10.5 km was set definitely after the rainfall climax. Naturally distributed contaminants should occur in moderate to high concentrations in the easternmost samples because these represent the light rain at the end of the shower. The indium concentrations in these approach the background mean. The samples for the interval from 2.5 to 6 km exceed the background mean by about 2 σ, but these contain water from the more intense part of the shower. If the contamination were natural, it should be lower in concentration for these samples than for those farther east. This, in addition to the significant concentration maximum

at the third station ($\sim$ 1.7 km) tends to support the conclusion that indium from the tracer plume has indeed been found in these samples.

Although the results from the north-south road array (Figure 8) are less systematic than those of Figure 7, the evidence of these data also favors the hypothesis that tracer indium was collected in the samples and is observable considerably (3σ) above background.

VI. Conclusions.

We feel that this pilot experiment for the first time shows that it is feasible to place a finely divided tracer material into a convective rain-generating system and to identify and measure quantities of the tracer in rain samples collected at the ground.

Further, the usefulness of indium, specifically, as a trace element has been shown, and is largely attributable to (1) its properties favorable for neutron activation analysis (sensitivity routinely 10^{-10} gm $\pm$ 5%) and (2) its low natural atmospheric background.

VII. Acknowledgments.

The assistance of Mr. August H. Auer and the supporting personnel of Weather Science, Inc., of Norman, Oklahoma, who operated the aircraft, and the efforts of Messrs. Gerald W. Wambolt, Kenneth Vetter, and Joseph Sobel, and of Misses Sandra King and Joan Funk who aided in the field and laboratory work, are acknowledged with thanks. We are indebted to Mr. C.M. Gordon of the Naval Research Laboratory, Washington, D.C., for providing information on the neutron activation characteristics of indium. Support of the project was provided by the U.S. Atomic Energy Commission under contract no. At (11-1)-1407, and by the National Severe Storms Laboratory, ESSA, under contract no. E22-110-67(N).

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Section II

Detection of Indium as an Atmospheric Tracer by Neutron Activation

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ABSTRACT

In a test of the use of indium as a particulate atmospheric tracer, both tagged and untagged rain showers were sampled at ground level in Oklahoma during May 1967. In 29 samples of untagged rain 6±3 nanograms of In per liter were found, indicating a natural background somewhat above the reagent blank of about 2 ng In/ℓ. After aircraft injection at cloud base of 200 grams of indium, as finely divided particles from pyrotechnic flares, a maximum of 40 ng In/ℓ was found in the rain, and a pattern of localization of the tracer indium was revealed in an array of 14 samplers spaced over 11 km. The procedure for analysis began in the field with (1) addition of HCl to the polyethylene samplers before rain was collected to ensure solubility of the tracer, (2) filtration to remove suspended solids, (3) addition of La internal standard, Fe^{+++}, and NH_3 to each 1 liter sample to precipitate In and La with $Fe(OH)_3$, and (4) membrane filtration of the $Fe(OH)_3$. At the University of Michigan the procedure continued with (5) reactor neutron activation of the filters, (6) separation of In from La by isopropyl ether extraction from HBr solution, and (7) assay of In and La radioactivities by β and γ counting, respectively. The sensitivity of the method s determined by the natural background of the indium found.

I. Introduction

In carrying out an atmospheric tracer experiment where discrete samples of the atmosphere are taken, only a very small fraction of the initial tagging material can be recovered in each sample. Suitable particulate materials for the tracer should contain a component which can be detected with great sensitivity and which is not present in large and variable amounts already in the atmosphere. Because the technique of neutron activation offers the high sensitivity needed, and the facilities at the University of Michigan make the method readily available, we chose the tracer material to be optimum for detection by this method. Indium on neutron irradiation gives rise to 54-minute In-116m, a beta and gamma emitter of convenient half-life, with a cross-section of about 150 barns per atom of indium. A beta counting rate of about 500 counts per minute was observed in samples containing 1 ng In, and a conservative estimate of the practical sensitivity limit in our laboratory is 10^{-10} gram or 0.1 ng In. Indium is thinly dispersed in geochemical materials and is not known to be added to the atmosphere as a contaminant. Fine particles bearing indium can be generated by a pyrotechnic flare of conventional design. To the extent that these particles are scavenged by cloud development and precipitation processes, the indium is detected and measured in collected samples of the rain. Some

experience has already been gained with indium as a tracer in cloud physics, where a procedure for collection and neutron activation analysis different from ours was employed (Jones, et al., 1967).

Three types of samples were analyzed to show the feasibility of the tracer method: tagged rain, untagged rain, and reagent blanks. The untagged rain and reagent blank samples were used to establish a basic background criterion. The tagged rain samples were then evaluated in relation to this basic criterion.

2. Experimental

About 200 grams of indium, associated with fine particles produced by burning seven pyrotechnic flares, were released over a period of 20 minutes from an aircraft flying at cloud base level in the storm updraft (see Dingle et al., 1968). Rain sampling bags of polyethylene supported by widemouthed baskets were deployed by a ground crew along roads where rain was expected to fall. Each of these was pre-innoculated with 50 ml of 6N HCl to assure dissolution of all captured In. After the rain the bags were recovered and taken to the field laboratory for pre-irradiation chemical processing.

Each rain sample was rough-filtered to remove large particles of suspended solids from the solution containing dissolved In. One or two measured volumes were then taken from each sample for further processing. To each volume 1 μg of La internal standard and 1 mg of Fe scavenging agent were added using nitric acid solutions. To precipitate $FE(OH)_3$ carrying La and In, NH_4OH was added, and the solid was separated using a membrane filter. This filter was put into a polyethylene capsule for later neutron activation analysis. An automatic correction for variations in sample volume and neutron flux is built into this procedure by means of the La internal standard.

Samples were irradiated in groups of 4 for 20 minutes in the pneumatic tube facility at the Ford Reactor at the University of Michigan in a slow neutron flux nominally of 2×10^{12} n/cm^2 sec.

After irradiation, the samples were processed further to separate the In-116m from the La-140 prior to counting. The $FE(OH)_3$ was dissolved from the filters in HCl, reprecipitated in NH_4OH and filtered for purification from Na, Cl, and other water-soluble radioactive species, and dissolved in 6N HBr. From this solution In was extracted using isopropyl ether,, leaving La in the aqueous phase. Back extraction of In into water, co-precipitation with $Fe(OH)_3$, and filtering gave a solid source suitable for proportional counting of 54-minute In-116m beta particles.

Beta decay was followed for several hours to establish the presence of 54-minute activity and then over several days to observe a longer-lived component identified as mostly 26.5-hour As-76. This component, observed in all decay curves of untagged and tagged rain water as well as in reagent blanks, is largely, if not completely, due to reagent impurity

The aqueous solution containing La was counted in this form for the characteristic 1.6 Mev gamma ray of 40-hour La-140 in a single channle NaI(Tl) scintillation spectrometer. Gamma activity was measured usually twice on successive days to check for the expected 40-hour decay.

The net In component of the beta decay curve was resolved graphically and compared to the gamma counting rate of the La fraction. This result was compared to counting rates obtained from analysis of a standard solution containing known amounts

of In and La, and the analytical result was calculated from the relation:

$$\frac{\text{Wt. In}_2}{\text{Wt. In}_1} = \frac{(\text{In/La})_2}{(\text{In/La})_1}$$

where subscripts 2 and 1 distinguish between sample and standard and the quantities on the right are counting rate ratios of In to identical amounts of La. The reliability of the overall procedure was established by means of replication and gamma-ray spectrometry of selected fractions using a multichannel analyzer.

3. Results

The analytical results are presented below in two sections, the first dealing with the tracer-tagged rain samples collected near Holdenville, Oklahoma, on 30 May 1967 (Dingle, et al., 1968), and the second dealing with our investigation of the background level of indium.

a. Rain samples from tracer-tagged storm. The results of analyses of rain water collected from a tracer-innoculated convective storm are given in Table 1, and Table 2 for the north-south road. The sample numbers increase toward east and south, respectively. Samples larger than one liter in volume were split for duplicate analysis, and each part is reported separately in the table.

Analytical results are given in terms of equivalent In. This means that the net short-lived component, ($t_{1/2} \sim 1$ to 1.5 hr) after subtraction of long-lived ($t_{1/2} \gtrsim 1$ day) activity, was all read as In. We found from gamma-ray spectrometry of selected samples that 54-min In-116m was the dominant, although not the sole component of the short-lived activity A relative measure of the non-In contribution to the short-lived activity is found by comparing the observed half-life values for each sample given in Tables 1 and 2 with the 54 min half-life of pure In-116m. Both deposition and concentration of In are reported for each aliquot in the case of split samples. For these samples total deposition and over-all concentration are also given.

On the east-west road (Table 1) total sample volumes ranged from 337 ml (1.7 mm of rain) up to 2081 ml (10.3 mm) Total deposition ranged from 2.4 to 55.3 ng In, and concentrations from 4.3 to 40.2 ng In/liter On the north-south road (Table 2) total sample volumes ranged from 656 ml (3.2 mm) to 1499 ml (7.4 mm), deposition from 5.4 to 24.8 ng In/sample and concentration from 8.3 to 23.5 ng In/liter. To tell whether some of the In in these

TABLE 1

Indium Tracer Rain Samples, East-West Road

Sample Number	Aliquot volume (ml)	Collected rainfall[1] (mm)	Equivalent Indium[2,3] ng/aliquot		ng/liter		$t_{1/2}$[4] (min)
1A	1065	10.3	15.2	26.9	14.3	12.9	65
1B	1016		11.7		11.5		58
2A	966	7.7	20.6	31.3	21.3	20.0	63
2B	602		10.7		17.8		59
3A	873	6.8	34.5	55.3	39.5		62
3B	513		20.8		40.6		62
4	1120	5.4	12.1		10.8		55
5[5]	440	2.2	5.7		12.9		78
6A[6]	789	7.3	10.7	21.5	13.5	14.5	79
6B[6]	693		10.8		15.6		84
7A	819	5.9	8.9	14.9	10.9	12.4	62
7B	383		6.0		15.6		57
8A	795	6.0	11.4	15.7	14.3	12.9	76
8B	423		4.3		10.1		63
9	901	4.4	7.0		7.8		67
10	674	3.3	2.9		4.3		65
11	680	3.3	5.5		8.0		70
12	538	2.7	4.4		8.1		73
13	337	1.7	2.4		7.2		57
14	439	2.2	2.9		6.6		64
15[7]	650	3.2	-		-		-

(1) Rain began at most sites before collectors were exposed. Collector area = $0.20 m^2$.

(2) Net short-lived radioactivity assumed to be all In.

(3) When the sample was analyzed in two parts, results for each part and the whole sample are given.

(4) For pure In-116m, $t_{1/2}$ = 54 min.

(5) This collector was found tilted at a steep angle after the rain. Rainfall and In deposition are too low, but In concentration may be representative.

(6) Plastic bag ripped, contamination by dust possible.

(7) Sample lost in analysis.

TABLE 2

Indium Tracer Rain Samples, North-South Road

Sample Number	Aliquot volume (ml)	Collected Rainfall[1] (mm)	Equivalent Indium[2,3] ng/aliquot		ng/liter		$t_{1/2}$[4] (min)
1A	882		11.6		13.2		60
1B	403	6.4	9.3	20.9	23.1	16.3	71
2A	767		6.5		8.5		75
2B	374	5.6	5.2	11.7	13.9	10.3	6
3A	823		10.6		12.8		74
3B	676	7.4	14.2	24.8	21.0	16.5	70
4A	692		14.4		20.8		60
4B	344	5.1	8.1	22.5	23.7	21.8	77
5A	599		4.4		7.3		64
5B	364	4.8	5.9	10.3	16.2	10.6	74
6	904	4.5	21.2		23.2		62
7	785	3.9	9.0		11.4		76
8	802	4.0	7.3		9.1		70
9	660	3.2	11.0		16.7		96
10	656	3.2	5.4		8.3		74

(1), (2), (3), (4) See footnotes, Table 1.

samples was recovered tracer, we must know the level of natural In in non-tagged rain samples.

b. Background determination. The problem of background determination is to assess as well as possible all sources of indium other than the artificially introduced tracer material. Sources that need to be evaluated are (1) the "natural", or uncontrolled, atmospheric content of indium, and (2) the indium content of reagents used in chemical processing of the rain samples. The sum of the "natural" and reagent contributions of indium, therefore, is considered to form a basic background upon which the tracer indium is superimposed. In evaluating the tracer samples, this background must be subtracted from the amounts detected in the tracer rain samples, and its magnitude must be compared to that of the net tracer indium found.

Background samples were collected at our Chickasha, Oklahoma, station on two days during May 1967. On 5 May 1967 a series of samples was collected during the passage of a pre-frontal squall line. Two separate showers were sampled Results are given in Table 3 On 30 May 1967 eight samples were collected during the passage of a developing squall line. Results are given in Table 4. This squall line produced the subject storm of the tracer experiment about 4 hours later and about 140 km to the east near Holdenville.

The values given in Tables 3 and 4 show the temporal variation of indium concentrations in rain samples collected serially at a fixed station throughout each storm. It is important to note immediately that this collection procedure was not used in procuring the tracer samples (Tables 1 and 2). These latter were collected at different places in samplers which remained exposed for the duration of the rainfall. Thus care must be used in applying the data of Tables 3 and 4 to the evaluation of the tracer results in Tables 1 and 2.

TABLE 3

Background Rain Samples Without Indium Tracer

5 May 1967

Sample Number	Sample volume (ml)	Collected rainfall[1] (mm)	Equivalent indium[2] ng/Sample	ng/liter	$t_{1/2}$[3] (min)
1[4]	932	0.32	10.9	11.7	80
2[4]	964	0.38	11.1	11.5	90
3	920	0.36	8.4	9.1	66
4	874	0.34	6.7	7.7	86
5	802	0.32	2.9	3.6	74
8	950	0.38	5.0	5.3	97
10	953	0.37	4.9	5.1	88
19	950	0.37	2.8	3.0	75
21	975	0.39	18.5	19.0	119
22	966	0.39	7.4	7.6	127
24	964	0.38	3.8	3.9	89
25	970	0.39	3.7	3.8	97
26	958	0.38	4.6	4.8	99
27	1013	0.40	3.3	3.2	79
28	992	0.40	4.8	4.8	113
29	1001	0.40	2.8	2.8	97
30	995	0.40	2.2	2.2	88
31	1018	0.41	1.8	1.8	70
34	921	0.37	1.9	2.0	88
35	947	0.38	2.9	3.1	92
36	966	0.38	3.3	3.4	82

Average Concentration, $\bar{X}$ = 6±3

(1) Collector area = 2.5 m^2.

(2) Net short-lived radioactivity assumed to be all In.

(3) For pure In-116m, t 1/2 = 54 min.

(4) Correction for internal standard counting rate, approximate only.

TABLE 4

Background Rain Samples Without Indium Tracer

30 May 1967

Sample Number	Sample volume (ml)	Collected rainfall[1] (mm)	Equivalent indium[2] ng/sample	ng/liter	$t_{1/2}$[3] (min)
1	967	0.38	5.5	5.7	69
2	951	0.37	7.3	7.7	66
3	958	0.38	12.9	13.5	87
4	856	0.34	6.8	7.9	56
5	925	0.36	8.0	8.6	65
6	933	0.37	2.2	2.4	65
7	926	0.36	3.7	4.0	73
8	906	0.36	5.9	6.5	168

Average Concentration, $\bar{X} = 7 \pm 3$

(1) Collector area = $2.5 m^2$.

(2) Net short-lived radioactivity assumed to be all In.

(3) For pure In-116m, $t_{1/2}$ = 54 min.

In earlier work, we have collected and analyzed sequences of samples at our Chickasha station and at Willow Run, Michigan, in a number of storms (Dingle and Gatz, 1966). For these cases we evaluated long-lived β-radioactivity and pollen content of the samples, both of which may be considered, at the scale we are concerned with, as more or less uniformly distributed contaminants in the lower troposphere. The findings show a nominally inverse relationship during the course of a storm between rainfall intensity and contaminant concentrations in the rain samples. This relationship has also been noted by others (Bleichrodt, et al., 1959; Salter, et al., 1962; Hinzpeter, 1958; Bleeker, et al., 1966).

To obtain an estimate of whole-storm average concentration, these can be averaged in an appropriate manner (i.e., for Table 3, 5.7 ng/l; for Table 4, 7.0 ng/l). We should expect the background indium concentrations to vary similarly among serial samples, unless the indium for some reason is not evenly distributed at the scale of the storm systems we are dealing with.

A second consideration, in using the background data obtained on 5 May 1967 (Table 3) to evaluate tracer data obtained on 30 May 1967 (Tables 1 and 2), is that of the constancy of the background from one storm system to another. At this point in the development of the indium tracer technique we do not have data adequate to demonstrate such a constancy. Georgii and Weber (1960) report data showing that the storm-to-storm variability tends to decrease with increasing rainfall amounts; that is, for 1-min rains, the ratio of maximum to minimum concentrations is more than 20, whereas

for 10-min rains it is about 10. The rains we are dealing with are in the 10-min class, and we may therefore expect about a tenfold variation from the least to the most contaminated whole storm rain samples. It is noteworthy that the variation within the 5 May 1967 storm (among the serial samples, Table 3) is about tenfold from the minimum to the maximum concentration, although this fact is not readily interpreted in the present context.

Inasmuch as the data of Table 4 were acquired from the same storm system (squall line) that provided the conditions for our tracer experiment some 4 hours later and some 140 km eastward, the additional question of the constancy of the background level for a given system from one place and time to another arises. A comparison of the concentrations tabulated in Tables 1 and 2 against those in Table 4 shows that the lowest concentrations observed in the rain samples from the tracer-tagged storm are comparable to those that appeared in the samples collected upstream prior to the tracer experiment. This may then be accepted as evidence of the constancy of the background over this range of distance and time.

c. Reagent blanks. A number of reagent blanks were prepared in the field on days when rain samples were processed (Table 5). Four of these contained, in addition to the reagents, ~1-liter samples of the deionized water used in reagent preparation. Comparison of the results for the samples containing water with those for reagents alone shows that the water itself had no In content; therefore all 16 samples can be considered reagent blanks. A mean and standard deviation of 1.5 ± 1.5 ng/sample were found. With this information, we can compare In levels in the three kinds of samples - tracer samples, background rain samples, and reagent blanks.

The reagent blanks contain smaller amounts of indium than the rain samples, and this observation is quite consistent throughout the data we have so far compiled as is shown by the graphic presentation of Figure 1. All but two of the rain samples (both tracer and background specimens) contained more than 2 ng In per sample, whereas the reagent blanks contained 2 ng or less In per sample.

In comparing the tracer rain samples against the background rain samples, it is convenient to refer to In concentrations because the sample sizes vary. Making comparisons on this basis, the average background concentration of 6 ± 3 ng/liter determined above, is seen to be well below the concentrations found in many of the tracer rain samples (Tables 1 and 2). The additional distribution of In concentrations within the east-west sampler array,

TABLE 5

Blank Analyses

Sample Number	preparation date, 1967	sample type	equivalent indium (ng/sample) [1]	$t_{1/2}$ (min) [2]
1	6 May	reagents	0.3	121
2	6 May	reagents	0.7	117
3	17 May	reagents + water	1.6	78
4	2 June	reagents	7.5	82
5	3 June	reagents	0.8	75
6	3 June	reagents	1.5	58
7	3 June	reagents	1.2	101
8	3 June	reagents	2.2	134
9	3 June	reagents	0.8	58
10	6 June	reagents	0.6	123
11	9 June	reagents	2.9	114
12	9 June	reagents	1.1	92
13	9 June	reagents	0.9	145
14	9 June	reagents + water	0.6	93
15	9 June	reagents + water	0.8	64
16	9 June	reagents + water	0.8	113

Average indium content, $\bar{X}$ = 1.5 + 1.5

(1) Net short-lived radioactivity assumed to be all In.

(2) For pure in-116m, $t_{1/2}$ = 54 min.

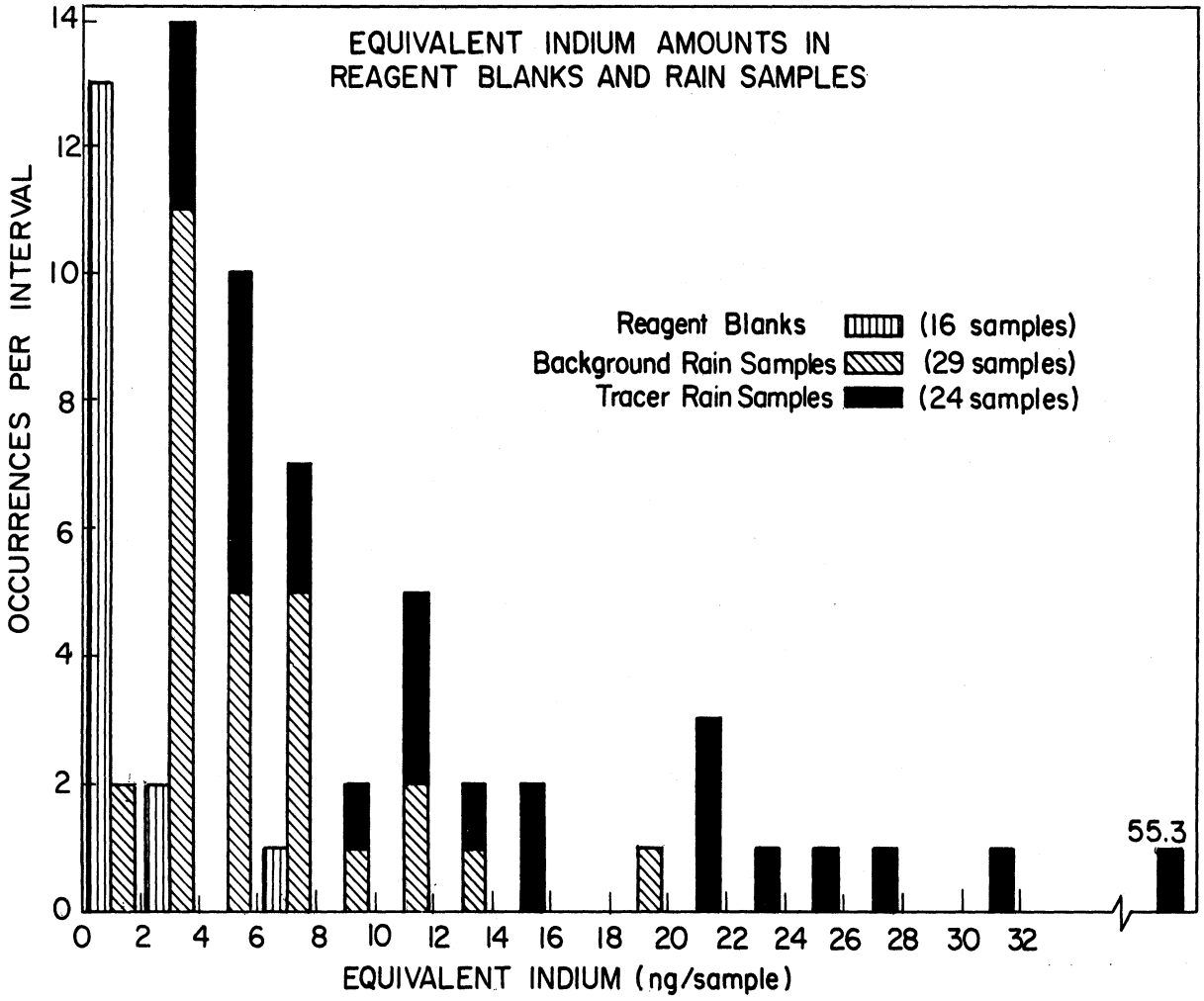

Figure 1. Frequency of occurence of equivalent indium amounts in reagent blanks, background rain samples, and tracer rain samples.

grading from the highest values near the western end (low sample numbers, Table 1) to values within the range of 6 ± 3 ng/l at the eastern end adds evidence that some of the tracer In was indeed recovered. Again, inasmuch as the highest In concentrations are associated with the largest rain samples (highest rainfall amount), in opposition to the usual trend of concentration with rainfall rate observed for uniformly distributed contaminants (see above) we feel that the inference that tracer In was collected is further reinforced.

4. Discussion

a. Analytical aspects. The radiochemical procedure adopted for this exploratory study was designed to give effective results with a minimum of analyst time under the limitations imposed by the simple radioactivity counting equipment which was available. We chose activation analysis because of its great sensitivity and relative freedom from analytical errors.

The internal standard La was selected to provide a means of normalizing the final indium radioactivity level. With this technique, a sample innoculated with the La internal standard is evaluated by making only a relative measurement of In to La. Actual sample volumes selected for each analysis, the neutron flux in the reactor on the day of irradiation, and the counter sensitivity for the radioactivity measurement need not be known absolutely. A relative measurement of radioactivity can be made quickly, easily, and with precision.

The technique of concentrating the indium chemically in the collected rain samples proved to be helpful as a means of reducing the bulk of material to be stored and transported from the field in Oklahoma to the Michigan laboratory. The field station processing, by preliminary coarse filtration and precipitation of $Fe(OH)_3$, was readily set up and done during periods of dry weather.

The post-irradiation procedure of separating the indium from other species in the sample that were also activated, was a necessary step to clarify the ultimate interpretation of the radio-

activity counts. This separation took about 15 minutes per sample for an analyst and technician working together, from the end of irradiation until counting began. The counting procedure and calculations in this work were done using manually-operated proportional and scintillation counters and manual data processing. Because selective counting techniques for indium were not employed, the chemical separation procedure was conducted so as to give good radiochemical purity, and the time devoted to counting extended over several days. About 3 manhours were required for the analysis of each sample.

With automatic counting equipment and data processing we believe the task of chemical analysis will be greatly reduced. An analyst and technician should be able to prepare a sample for counting in less than 15 minutes, and automatic gamma ray spectroscopy with magnetic tape recording can be performed without constant attention. Several hundred samples per month may then be analyzed by the investigator and a co-worker, and the effort of chemical analysis should be small compared to that of preparation and execution of the field experiment.

b. Analytical errors. Random errors in activation analysis as carried out here customarily are 10% to 20%, including chemical errors in the separations and statistical errors in radioactivity measurement. In addition, a systematic error appears in the presence of some radioactive impurities in the indium fraction; these were usually small, however, and the technique of comparing tagged and background rains partially corrects for this source of

error. An ultimate test of the significance of the analytical data is the reproductibility of duplicate analyses of the same water samples. In Table 1 most of the duplicates agree to within 20%, although Table 2 shows poorer agreement. Errors due to adsorption of In onto clay particles in the rain sample are guarded against by pre-innoculation of the samplers with 50 ml of 6 N HCl, but future experimentation should include a re-evaluation of this procedure. It must be emphasized that nanogram amounts of indium are very small and their measurement requires great care.

c. Time-variability of natural In. The time profile of background In concentrations observed during the convective shower on 5 May 1967 is plotted in Fig. 2, along with similar profiles of gross non-volatile residue, artificial beta radioactivity, and the rainfall rate. The profiles of In and gross residue, and to a lesser extent, beta radioactivity, exhibit the familiar quasi-inverse relationship with rainfall rate. Thus, although the data are rather sparse, natural In appears to behave very much like other constitutents of rain. This parallel behavior suggests that another trace constitutent might prove useful as an indicator of the natural In in rain. It is clear that, if two constituents vary in a parallel manner, the ratio of these two must exhibit less variability, both <u>within</u> and <u>between</u> storms, than their individual absolute concentrations. Thus, we suggest that future rain-tracer work include the indentification of a specific rainwater constituent to use as an index of the background level

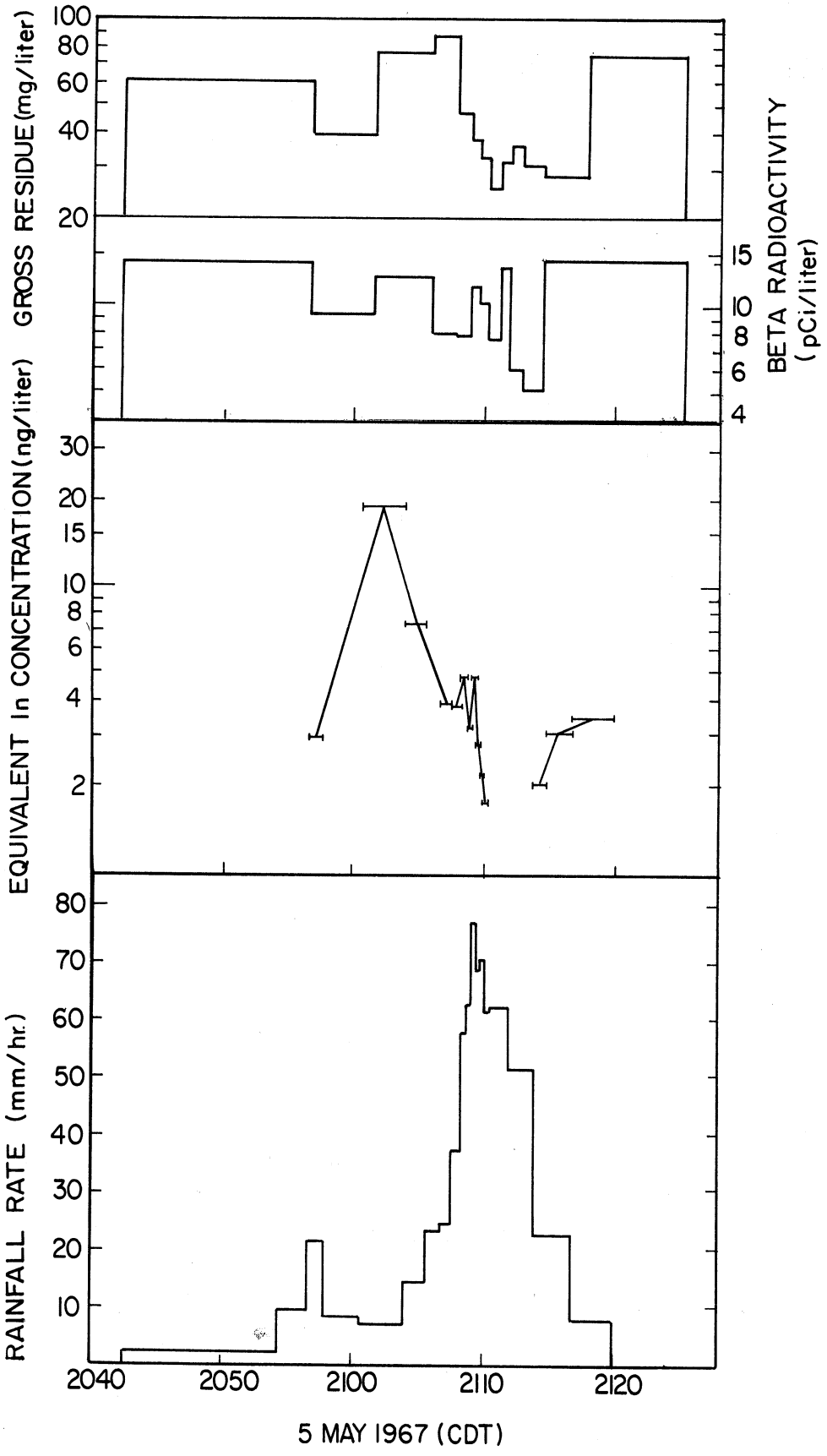

Figure 2. Temporal variation of background indium, beta radioactivity, and gross residue during a convective shower. Horizontal bars indicate duration of In background samples.

of the tracer in each sample.

5. Summary and conclusions

To test the use of In as a particulate tracer for cloud processes, one tagged and two untagged rains were sampled in Oklahoma during May 1967. In 29 rain samples collected serially from the two untagged rains, 6 ± 3 nanograms of In per liter were found on the average indicating a natural background somewhat above the reagent blank of about 2 ng In/liter.

Chemical processing of the rain samples was done in two steps. In the pre-irradiation field phase, In and a La internal standard were concentrated by coprecipitation with $Fe(OH)_3$ on a membrane filter. In the post-irradiation laboratory phase, done at the University of Michigan, the neutron-irradiated In and La were separated by a solvent extraction of In from HBr solution into isopropyl ether. The In and La radioactivities were measured with simple beta and gamma counting devices.

More than a chance number of tracer samples contained indium in concentrations above background. The localization of high In concentrations at the western end of one of the sampler arrays is additional evidence that tracer In was recovered. From this evidence and others presented more fully by Dingle et al., (1968) it is apparent that some tracer In was recovered.

This demonstrates the feasibility of the In tracer method for convective precipitation studies. That is, it is possible to release from an aircraft enough In tracer to be clearly

detectable above measured backgrounds in rain.

The amount of the tracer which must be released to be detectable in 1-liter rain samples is governed by natural backgrounds in rain, and by reagent blanks, but not by analytical sensitivity when neutron activation is employed. Whereas the In tracer experiment reported here was done using a release of 200 gm of In, it is felt that the signal-to-noise-ratio (tracer In/background In) would be greatly improved by increasing the release rate 5- to 10- fold. Although some development is required to do this, present indications are that it is feasible.

Future development of the method should include more extensive measurements of the natural In background and perhaps the selection of another rainwater constituent to serve as an index of the tracer background.

6. Acknowledgements

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Section III

Trace Substances in Rain Water: Physical Processes Implied by Concentration Changes During Convective Rains

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ABSTRACT

Sharp variations in the concentration of trace substances in rain water samples are observed during the passage of convective rain showers over a fixed collecting station. These most frequently have an approximately inverse relationship to the changes of rainfall rate observed at the same station, but detailed examination of many showers shows that the shift of phase between the concentration and rainfall-rate profiles varies from 0° to 180° (0 to ~ 10 min.). The continuous nature of the phase shift function suggests a continuously changing behavior of the physical processes that produce it. These processes appear to be controlled by the vertical penetration of the convective updraft and by the height of origin of the low level downdraft. Several cases, including one of the persistent or "steady state" severe storm category are presented.

I. Introduction

Many researchers have followed trace substance concentrations, either continuously or in discrete samples, through individual rains. These observations commonly reveal certain relationships between trace substance concentrations and rainfall rate, particularly in convective rains. Inverse relationships (low concentration at high rainfall rate) are observed most often but direct relationships also appear from time to time.

A basic premise is that both rainfall rate and level of contamination of the rain are products of the convective generation of precipitation, and hence that their correlation, direct, inverse or otherwise, may therefore be a key to the structure of the convective system. Because rainfall rate is an indicator of shower structure, we make the best resolved rainfall rate measurements that we can, and use these to define the showers that we sample.

Rainfall rates having the same resolution as the concentration data, namely those computed from the circumstances of sample collection though usually better than average are not

adequate to show the very small details of convective shower rainfall rates.

These details become clearer as the resolution of the rainfall rate improves; i.e., as the rain depth per rate measurement decreases. Tipping-bucket rain gauges commonly provide one rate measurement every 0.25 mm (0.01 in.). This is usually better resolution than can be obtained by using sample collection data unless very large collectors and small samples are involved.

It is our purpose here to report on relationships between concentration and rainfall rate which have been observed when concentrations from high-resolution rain sampling were compared with tipping-bucket rainfall rates.

A. Previous observations A partial list of literature references to observations of concentration-rainfall rate relationships is given in Table 1.

The substances investigated cover a broad diameter range, from 100-μ pollen grains to individual lead-214 atoms. Most of the observations, however, are of artificial beta radioactivity. Two other materials which have received moderate attention are inorganic ions (by conductivity measurements) (Georgii, 1965) and natural radioactivity (Jacobi, 1962).

Table 1 shows that regardless of the material selected as a tracer, almost every experimenter finds that tracer concentration

TABLE 1 Summary of observed relationships between trace substance concentrations and rainfall rate in individual convective rains

Trace Substance	Author	No. of Cases	Predominant Relationship Reported	Rainfall per Sample (mm)	Method of R(1) Measurement	Rainfall per R(1) Measurement (mm)
pollen	Dingle/Gatz (1966a)	3	inverse	0.73-1.46	t-b gauge(2)	0.25
	Gatz(1966)	2	inverse	0.25-3.00	t-b gauge	0.25
artificial beta radio-activity	Bleichrodt, et al. (1959)	11	inverse	0.20	sample collection	0.20
	Huff/Stout (1964)	87	inverse	1.03-1.52	weighing gauge	1.03-1.52
	Huff (1965)	190	inverse	1.03-1.52	weighing gauge	1.03-1.52
	Dingle/Gatz (1966a)	5	inverse	0.73-1.46	t-b gauge	0.25
	(1966b)	3	inverse	0.25	t-b gauge	0.25
	Gatz (1966)	2	inverse	0.25-3.00	t-b gauge	0.25
	Hall/Nelson (1964)	11	inverse	1.27	weighing gauge	1.27
	Hall (1965a)	29	inverse	1.27	weighing gauge	1.27
	Hall (1965b)	46	inverse	1.27	weighing gauge	1.27
inorganic ions	Georgii (1965)	not reported	inverse(single showers)	continuous sampling	not reported	(3)
			direct (series of showers)	continuous sampling	not reported	(3)
deuterium	Ehhalt, et al. (1963)	1	inverse	not reported	not reported	-
	Bleeker, et al. (1966)	2	inverse	0.19	sample collection	0.19
oxygen-18	Bleeker, et al. (1966)	2	inverse	0.19	sample collection	0.19
lead-214 (RaB)	Jacobi (1962)	not reported	direct	continuous sampling	not reported	(3)

(1) rainfall rate
(2) tipping-bucket rain gauge
(3) rainfall rate computed over 15-min intervals.

is usually inversely related to rainfall rate. Most workers reported some direct relationships, but these were definitely not the rule.

Huff and Stout, (1964) and Huff (1965) performed beta radioactivity analyses on sequential rain samples collected in a network of automatic sampling stations. They analyzed a large number of cases and found an inverse relation most often. Direct relations were not uncommon, however, and often both types occurred within the same storm system.

Hall (1965a) operated a sampling program similar to that of Stout and Huff and reported much the same experience. In addition, he found a tendency for inverse relationships in storms which did not penetrate into the stratosphere and direct relationships in dissipating storms. Hall (1965a) reported a direct relationship and Gatz (1966) an inverse relationship for the same shower sampled at stations about 0.4 km apart on 9 May 1964. If these observations are correct, it appears that both types of relationships can occur in the same storm, at times within short distances compared to the horizontal dimensions of the storm.

B. Data quality. The question of data quality becomes important in view of the different methods of rain sampling and rainfall rate computation listed in Table 1. First of all, one may ask whether the sampling resolution is adequate in each case to distinguish concentration and rainfall-rate peaks which occur

within a few minutes of each other.

This question may be analyzed by studying high-resolution data to see at what point, as the resolution is reduced, the discrimination of the important maxima and minima becomes questionable. The argument, which need not be repeated here, leads to the obvious conclusion that discriminations that are made possible by highly resolved data cannot be made equally clearly by data of lesser resolution. Of the discrete sample data sets presented in Table 1, those of Bleeker, et al. (1966) have the finest resolution in terms of rainfall amount (0.19 mm per sample and per rainfall-rate determination). Our rainfall rates (Dingle, Gatz) are all computed from tipping-bucket rain-gauge records, each tip giving a rainfall-rate figure for 0.25 mm of accumulated rain. Our sample resolution varies from 0.25 to 3.00 mm per sample depending upon the circumstances of the sample collection. Bleichrodt's data also are of high resolution, comparable to Bleeker's. Georgii and Jacobi have by far the finest resolution of samples by virtue of their continuous monitoring technique, but their observations of rainfall-rate give only the average for each 15-min. period. Generally speaking, the rainfall rates in the United States (Huff, Stout, Dingle, Gatz, Hall) run to considerably higher values at their maxima than those in Europe (Bleichrodt, Georgii, Bleeker, Jacobi), and this may tend to justify acceptance of the 15-min. average

rainfall-rate figures by Georgii and Jacobi. However, in determining the relationship of trace concentration in rain samples to rainfall intensity, even in relatively steady rains, the 15-min. average values of rainfall rate are clearly not adequate, and the relationship reported by Georgii (1965), and Jacobi (1962) must be considered nominal at best.

C. Proposed mechanisms The literature contains numerous suggestions of mechanisms that might explain both kinds of relationships.

1. Direct relationships. Kruger and Hosler (1963) suggested that radioactive debris would enter convective clouds by entrainment at the cloud top, and that the concentrations in precipitation would reflect the height at which the precipitation was formed. Heavy rain, falling from a mature convective storm, was expected to be formed at high altitudes, and so to contain relatively high concentrations. In other words, a direct relationship was expected by these authors to result from the input of radioactive debris at and near the tropopause.

Stout and Huff (1964) and Bleeker, et al. (1966) suggested that their cases of direct relationship could have resulted from the same mechanism.

Dingle (1968), on the other hand, has pointed out that such cloud top scavenging cannot be effective for bringing high altitude contaminants immediately to earth, because these tall clouds are

necessarily dominated by divergent flow at their tops. Such materials as may be collected by high-altitude cloud elements must therefore mainly return to the troposphere as somewhat larger aggregated particles as they lose moisture far downstream.

Hicks (1966) suggested that a direct relationship could result if large amounts of trace substance entered a severe convective storm at middle levels in the manner suggested by Browning (1964) and then were collected by the heavy rain falling in the downdraft. Because this downdraft is best developed in relatively well-organized and persistent convective storms, we should expect to see the direct relationship due to this process displayed best in the more severe convective storms of the Great Plains.

2. Inverse relationships. Huff and Stout (1964) found that inverse relationships were predominant in their data, and typically included a very high concentration in the first sample. This they attributed partly to evaporation and partly to excess contamination of the forward edge of the storm by particulate matter from the surface or low levels of the atmosphere.

Bleeker, et al. (1966) stated that concentration is a complicated function of

(a) the dominant scavenging mechanism,

(b) the lifetime of cloud droplets,

(c) the liquid water content of the cloud,

(d) the precipitation rate,

(e) the height of the cloud base, and

(f) evaporation of cloud and precipitation elements,

and that some of these are strongly intercorrelated. For example, they infer that large rainfall rates imply (1) large liquid water contents, (2) short droplet lifetimes, and (3) little evaporation, because of (4) low cloud bases. These factors lead to low concentrations associated with high rainfall rates. Conversely, they suggest that low rainfall rates involve higher concentrations because of (1) small liquid water contents, (2) long droplet lifetimes, and (3) much evaporation. In addition, Bleeker, et al. assume that Brownian diffusive capture is the principal scavenging mechanism and show theoretically that an inverse relationship is the natural consequence of that mechanism.

Storebø (1965), from statistical evidence, Hicks (1966), from physical and chemical reasoning, and Dingle (1968) on meteorological grounds, concluded that bomb debris particles become attached to precipitation elements chiefly because they tend to be attached to hygroscopic particles prior to their service as condensation nuclei. Gatz (1966) reached the same conclusion on the basis of (1) published size distributions of artificial radioactivity near the surface, (2) the theoretical inability of the Brownian capture and raindrop impaction processes to remove such particles, and (3) experimental results, which will be considered in more detail later.

Storebø (1966) suggested that in a convective situation, "...early rain from one cell would draw its water from the

first droplets formed during condensation and should therefore contain more radioactive particles than average rainwater." Hicks (1966) pointed out that the larger nuclei will be removed first and that therefore most of the large particles will be nucleated near the edge of the cloud. This may be viewed as the cloud microphysical equivalent of Storebø's statement. Both writers tend to justify the inverse relationship between rainfall rate and contaminant concentration in rain samples collected at a fixed point in terms of the physical processes of rain generation.

II. Results

Our earliest results (Dingle and Gatz, 1966a) showed a generally inverse relationship between trace substance concentrations and rainfall rate. Since 1964 we have collected rain samples and made other observations in the National Severe Storms Laboratory (NSSL) cooperative spring program in Oklahoma. The experimental and analytical procedures used there (see Gatz, 1966) were about the same as in the earlier work, except that a larger collector was used (7.5 m^2 as opposed to 5.2m^2) and both pollen grains and beta radioactivity were determined for each rain sample. These changes improved the resolution of the concentration data without any loss of precision of the results. Our results from eight convective rain systems sampled in Oklahoma also show a general inverse trend between concentration and rainfall rate. More interesting than this general feature are the more specific and highly

repeatable relationships that appear upon closer and more critical study of the data.

A. A rain consisting of a single moderate shower.

The results from the 11 May 1966 rain presented in Figure 1 are fairly typical for a shower having a single maximum rainfall rate. The trace substance concentration expressed as beta radioactivity is high at the start of the rain, decreases rapidly as the rainfall rate increases, and reaches a minimum just prior to the rainfall rate minimum. The later small shower shows a nearly inverse relationship Figure 1(b), but its departure from the strictly inverse rule is shown in Figure 1(a).

B. A rain having two nearly equal intense shower peaks.

Figure 2 shows concentration variations during the rain of 9 May 1964 in which two separate but intense rainfall rate maxima occurred. This rain occurred during a severe squall line situation north of a stationary front at Chickasha, Oklahoma. The individual showers produced heavy rain, but as shown by the NSSL WSR-57 radar data, the pertinent rain cells were highly transient in nature, and could be traced individually for no longer than a few minutes.

The trends of pollen and radioactivity concentrations shown in Figure 2 are quite typical. In particular, the trends shown in the light rain periods before and after the principal showers suggest that evaporation may be a significant factor in producing the inverse relationship for light rain. The second

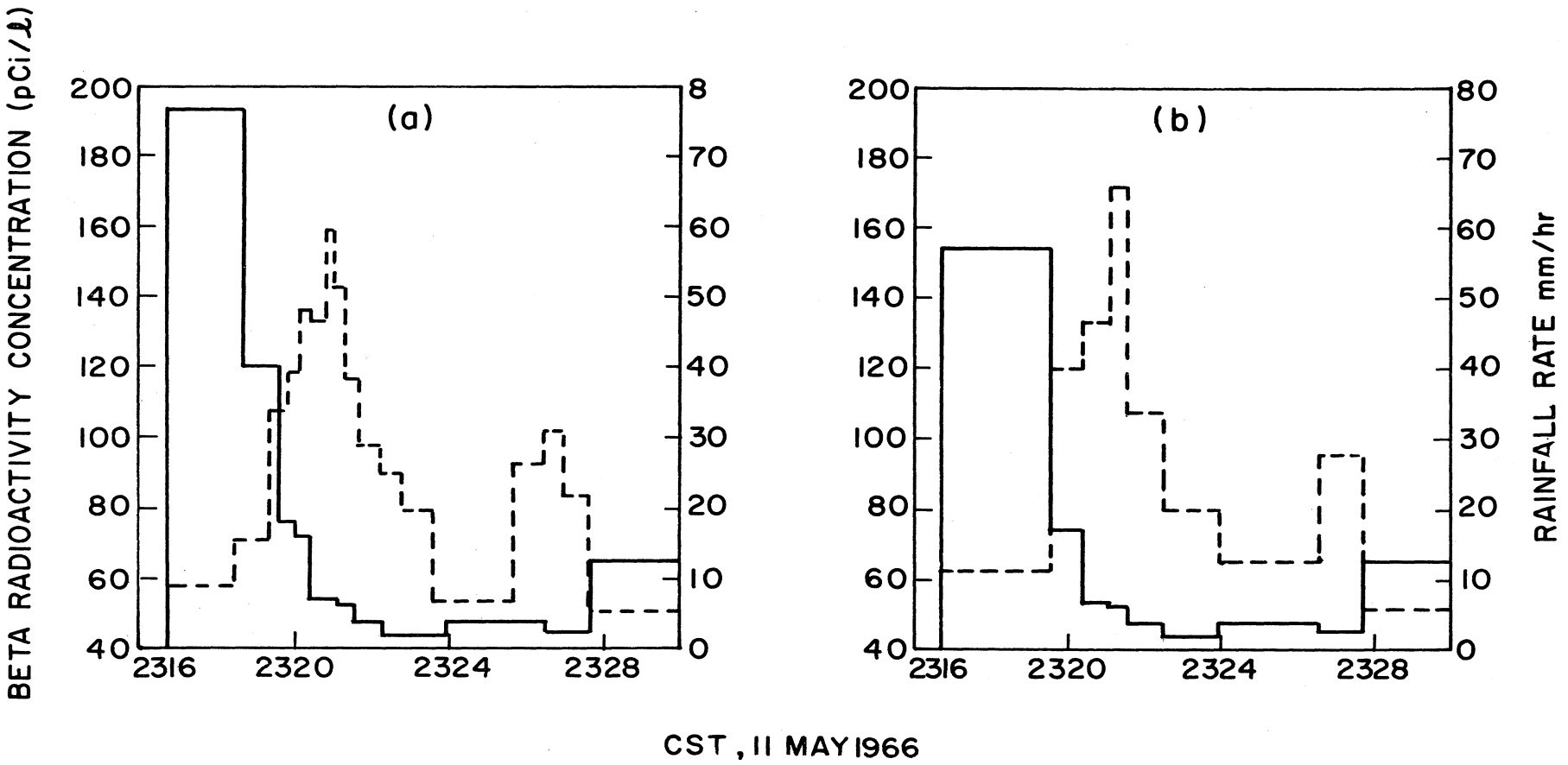

Figure 1.

(a) Rainfall rate in mm/hr (dashed line), and β-radioactivity concentration in pico curies per liter (full line) plotted against time for a rain shower of 11 May 1966 at Chickasha, Oklahoma.

(b) The same data collated so that the rainfall rate values are given for the same time periods as the concentration data.

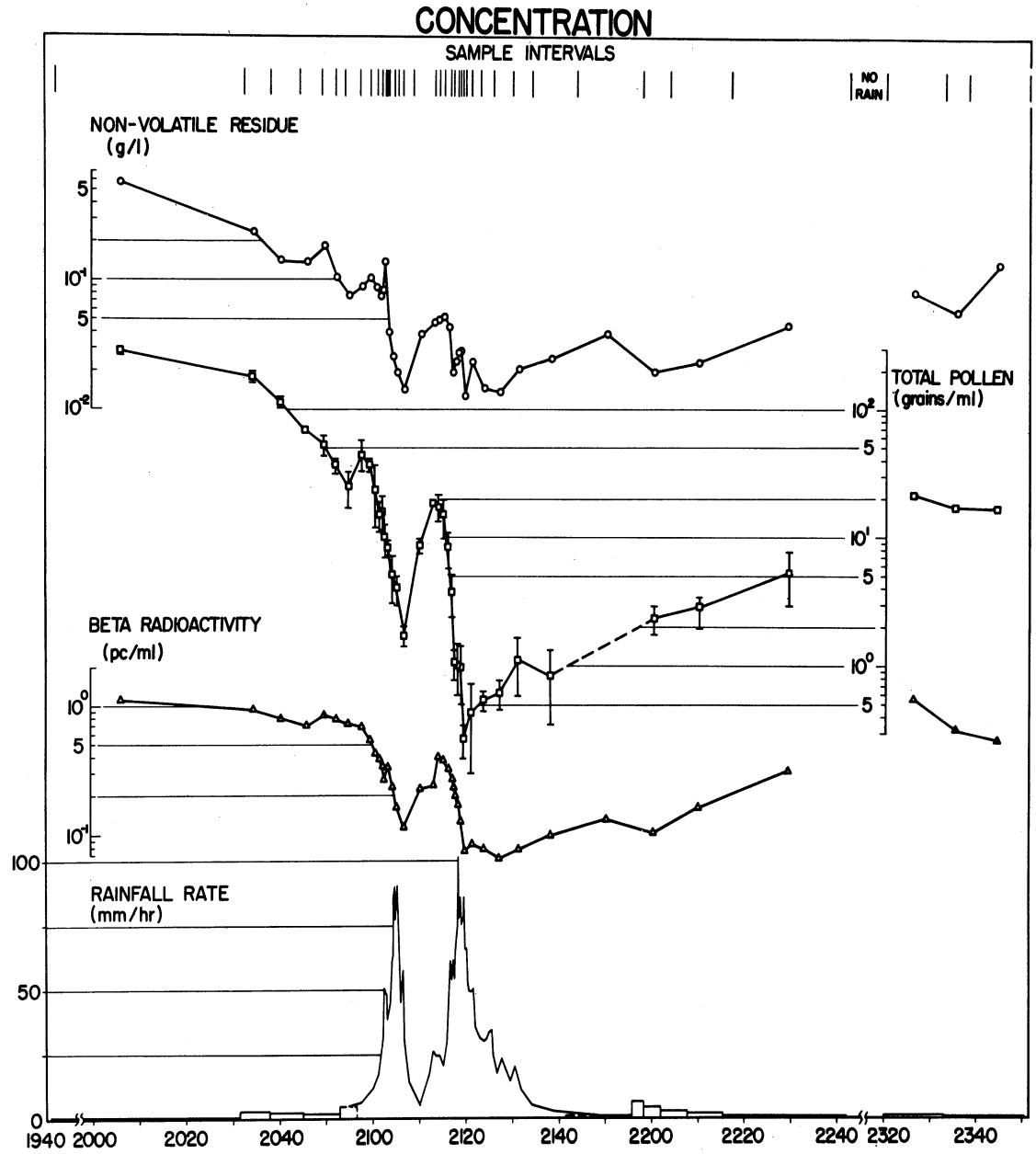

Figure 2.

Rainfall rate, pollen, and β-radioactivity profiles for rain of 9 May 1964 at Chickasha, Oklahoma.

maximum on 9 May 1964, however, is a good example of a shower that occurred after considerable rain had already fallen, and thus may be examined to see what happens when evaporation is not an important influence. Both pollens and radioactivity reached their relative minumum concentrations in the same sample which was entirely collected after the first rainfall rate maximum. Their concentrations both increased, from this minimum to a maximum during the onset of the second shower.

If it is assumed that the evaporative effects are neglible for the period between the two showers, the observed changes of contaminant concentrations must be attributed to other mechanisms. The process of elevating low level contaminated air by convection to form rain necessarily imposes a temporal pattern upon both the rainfall rate and the rain sample contamination experienced at a fixed station. The above assumption, therefore, allows us to consider the rainfall and scavenging patterns for the second shower as a model of the results produced directly by the convective rain-generating processes. This model has the following outstanding features;

(1) contaminant concentrations reach their maximum during shower onset;

(2) They decrease rapidly as rainfall rate increases to and somewhat beyond its maximum.

(3) They reach a minimum soon after the rainfall rate maximum; and

(4) they remain constant or increase gradually in the light rain after the shower.

C. A rain having three closely spaced shower peaks.

Fig. 3 shows concentration variations during a rapid succession of three showers on 9 May 1965. The beta radioactivity curve shows a peak during onset of the first shower, followed by a minimum at the first rainfall rate peak. The concentration increased temporarily as the second shower began, then dropped to another minimum at the second rainfall rate peak. The concentration then held rather steady for about three minutes before dropping sharply during the third shower. The concentration pattern typical of individual cells is somewhat obscured by the overlapping of the showers in this last instance.

The pollen concentrations, having a larger error, (Gatz, 1966) give a less clear pattern of change; yet the typical variations can be seen for each shower. It is interesting that the significant peak in the pollen concentration which occurred just before the third shower peak was not accompanied by a similar peak in the radioactivity concentration. Such relatively minor deviations appear to offer clues to differences in input and attachment mechanisms which may be simulated by modeling techniques.

D. A rain having two closely spaced moderate rainfall-rate peaks.

Fig. 4 is a portion of the rain of 22 April 1966. The two principal rainfall rate peaks were 5 min apart. The radioactivity

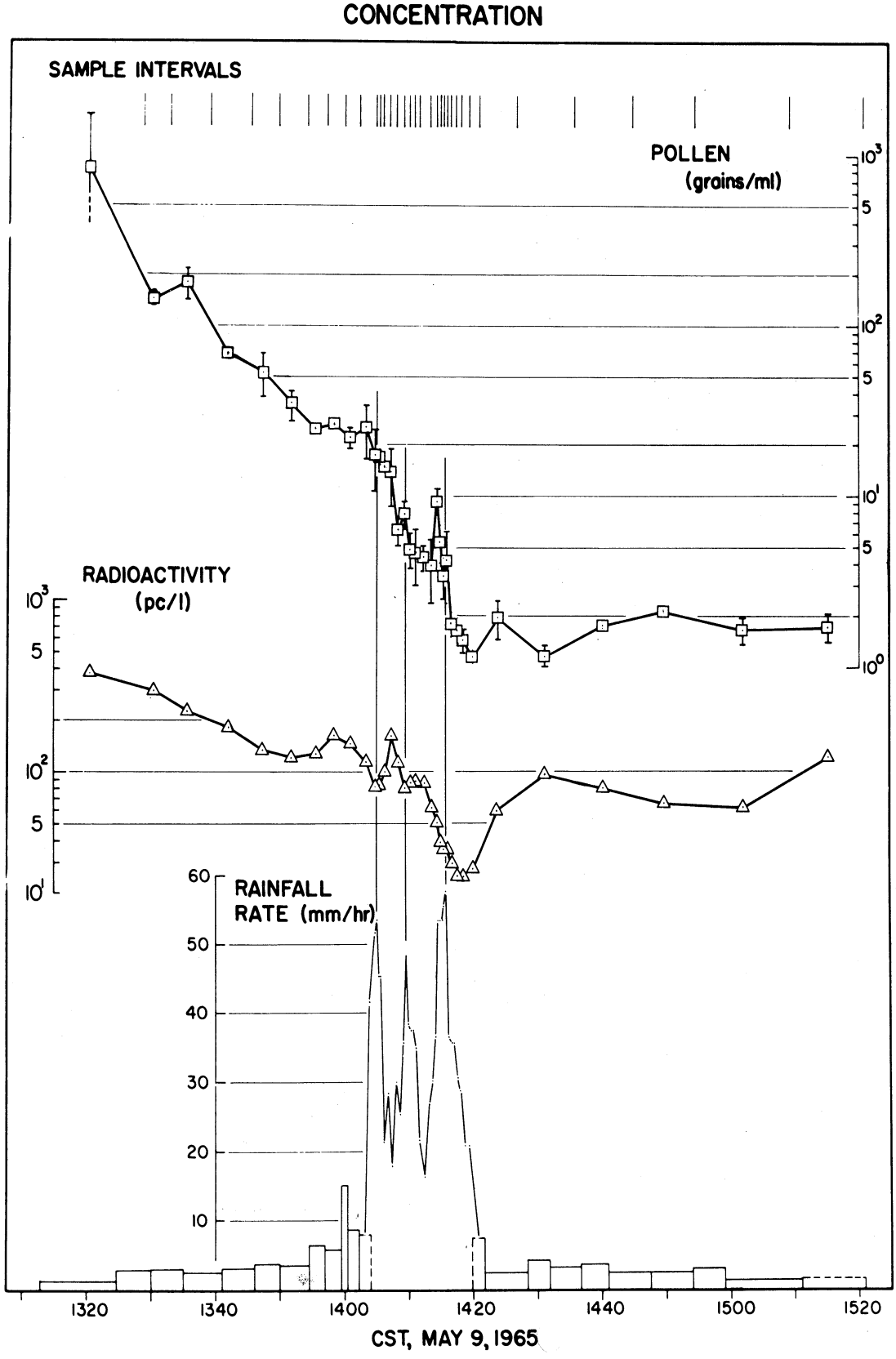

Figure 3.

Rainfall rate, pollen and β-radioactivity concentration profiles for rain of 9 May 1965 at Chickasha, Oklahoma.

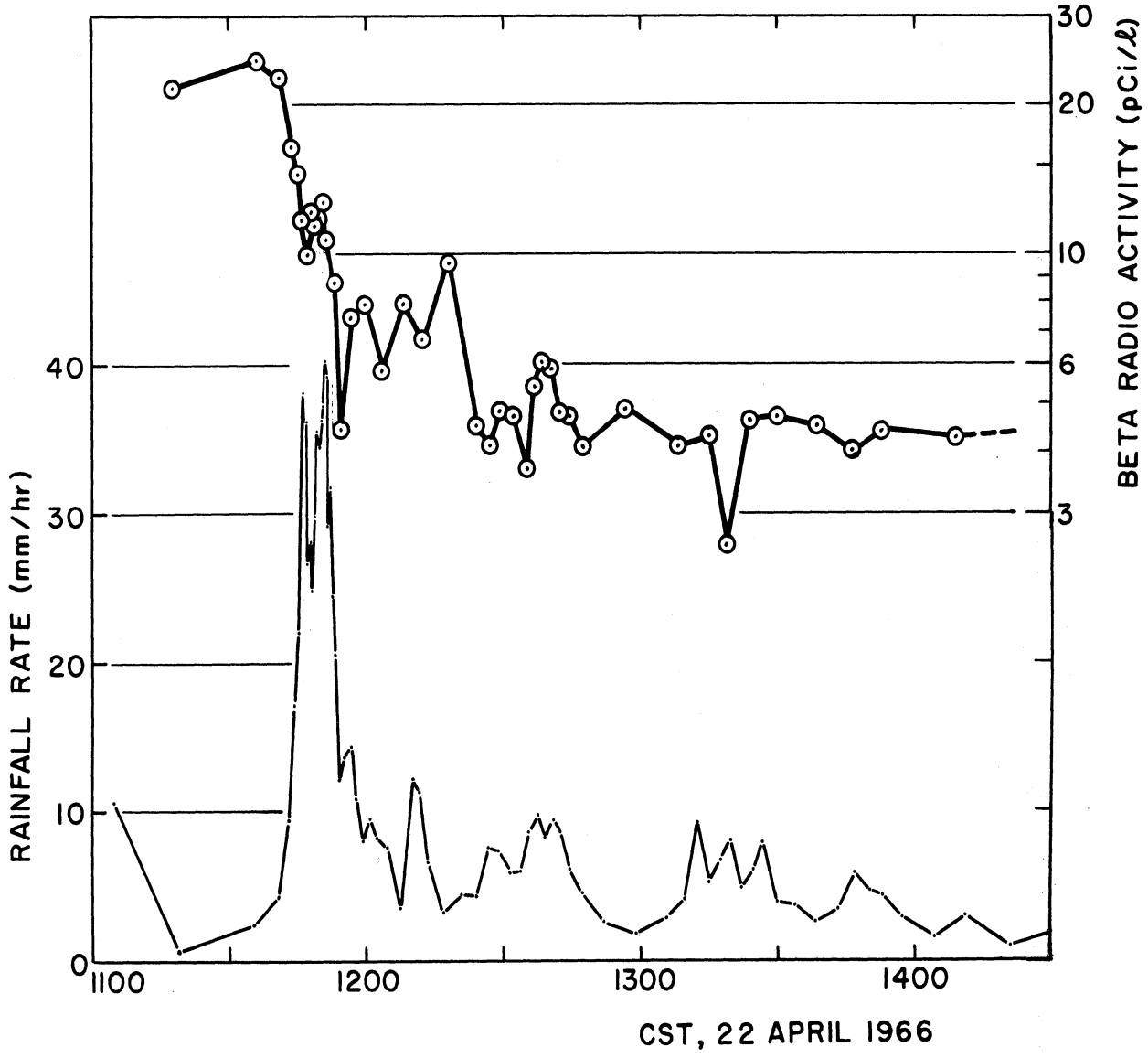

Figure 4.

Rainfall intensity and β-radioactivity concentration profiles for rain of 22 April 1966 at Chickasha, Oklahoma.

concentration reached a peak as the first shower began, then dropped sharply to a minimum at about the same time as the first rainfall rate minimum. In a clearly direct relationship, the second concentration peak and the second rainfall rate peak occurred together. Here is a case of two showers, very similar in maximum rainfall rate and occurring in rapid succession at the same place, having different relationships. Notice that the shapes of both concentration profiles are typical. There is nothing to suggest that radically different attachment mechanisms were involved. Perhaps in accord with Storebø's (1966) idea this may be a case where first precipitation from a cell is also the heaviest. The close association and obvious overlapping of the two showers may have contributed to the direct relationship also. Certainly the climax of the second shower is so abrupt as to preclude any "onset" such as we have observed in the showers previously discussed.

E. An intense rain from a persistent severe storm.

On 10 May 1964 a well-organized storm, associated with a sharp squall line, passed over our field station at Chickasha, Oklahoma. Considerable hail and a number of tornadoes were reported associated with this system. NSSL radar data show that the center of the most intense echo in the area passed over our station, and that the same echo was identifiable for several hours. These and other data indicate that this storm was similar to that modeled by Browning (1964).

Concentration and rainfall-rate profiles are shown in Figure 5. There was a slight decrease in both pollen and radioactivity concentrations during the heaviest rain, but the amount of the decrease was much less than that produced by the shower maxima discussed earlier. The ratio of the maximum to the minimum radioactivity concentration was 3, whereas the usual ratio is at least 10, and often 20. It is clear that the patterns of change observed here are markedly different from those in the other showers.

III. Interpretation

It is clear from the results presented here and others in the literature, that large fluctuations of contaminant concentrations are directly associated with convective precipitation cells. The usual pattern of change of these concentrations appears to be nominally the inverse of the pattern of rainfall rate change, for ordinary moderate to strong convective showers. Occasionally, however, these two patterns are directly related, and in studying a number of cases one finds that the time interval from the concentration maximum to the rainfall rate climax varies from zero to about 10 min. A strong departure from this behavior is found in the one well-organized severe storm that we have sampled to date. This departure is principally shown in a reduction of the amplitude of the concentration profile through the storm, which occurs despite the extremes of rainfall rate that were achieved (rain of 10 May 1964, Fig. 5)

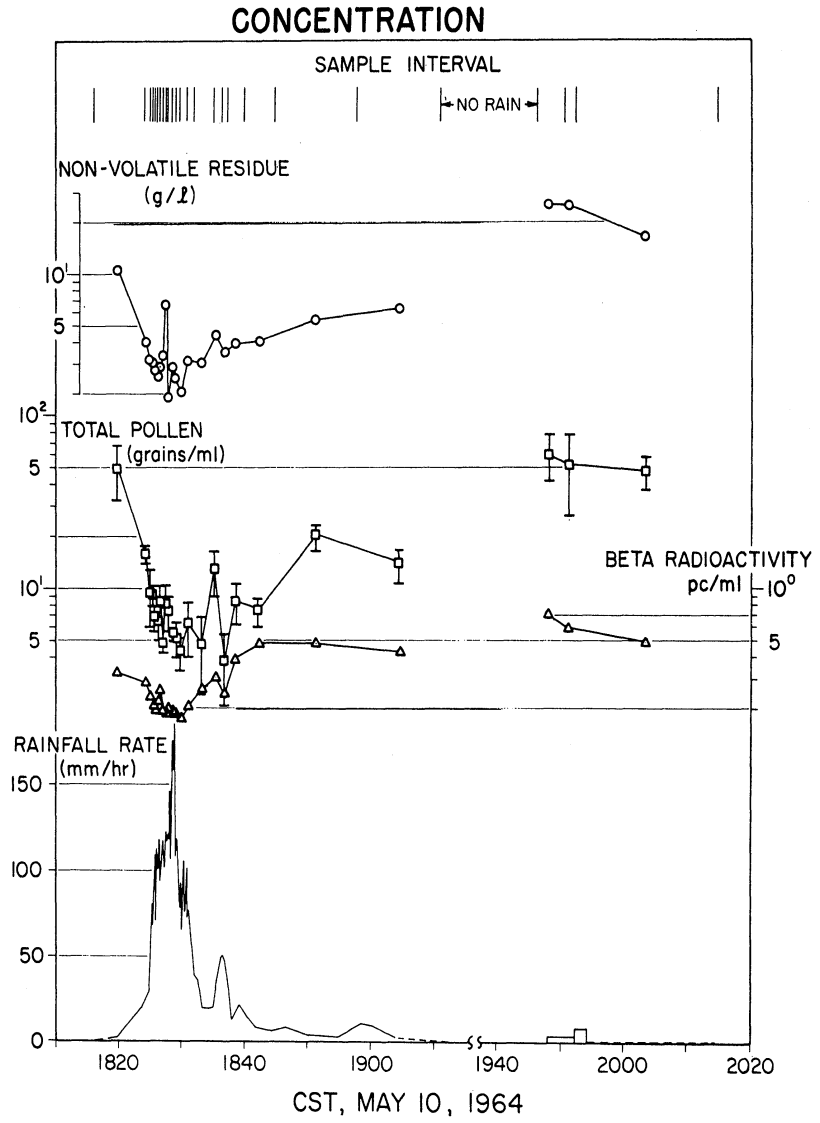

Figure 5.

Rainfall-rate, pollen, and radioactivity concentration profiles for rain of 10 May 1964 at Chickasha, Oklahoma.

To interpret the concentration variations, it is necessary to examine the mechanisms that produce them as well as those that produce the rainfall rate changes. Dingle (1968) has considered the physical requirements for the rain scavenging of particulate materials in relation to the several scales of atmospheric circulations and to the zones of horizontal convergence and divergence that are involved in convective shower activity. The overall conclusions from these considerations are (1) that the predominant rain-scavenging mechanism for radioactive materials is the nucleation of condensation by hygroscopic materials characteristic of the tropospheric aerosol; (2) that the radioactive particles become associated with tropospheric aerosol particles by aggregation which proceeds over a period of 3 to 7 days or more after the stratospheric (radioactive) aerosol has been extruded into the troposphere (Danielsen, 1964); and (3) that pollens are scavenged by impact-collection, and possibly in part by nucleation.

Gatz (1966) concluded that bomb-debris radioactivity must be predominantly scavenged by the nucleation process. His reasoning is based upon consideration of (a) published size distributions of the particles bearing radioactivity in low level air (Shalmon, 1961; Shleien, et al, 1965; Lockhart, et al, 1965), and (b) the inability of Brownian diffusion and raindrop impaction to remove such particles.

Storebø (1965) also inferred on statistical grounds, and Hicks (1966) on the basis of physical and chemical evidence, that predominantly the radioactive particles from bomb debris must be scavenged as components of mixed or aggregated condensation nuclei.

Whereas Greenfield (1957) held that the bomb debris particles must be scavenged mainly by Brownian diffusion to cloud particles, the cumulative evidence set forth by the above authors is that the Brownian mechanism has its principal significance in the dry aggregation processes that determine the size, structure and content of the condensation nuclei.

Because pollen grains are born large, and some appear to have hygroscopic or wettable surfaces for a short time at least, they may serve as very large condensation nuclei (Dingle, 1967), but they must certainly also be subject to impact collection by falling raindrops. This latter mechanism is considered completely negligible in the scavenging of radioactive materials.

It is well known that condensation nuclei are activated selectively, the largest and most hygroscopic forming the first droplets in rising air, and the smaller and less hygroscopic particles becoming activated later. Study of the growth and trajectories of these particles in rising air has been pursued by Bowen (1950), Ludlam (1951), and East (1957). These authors agree that the largest droplets in a given rising parcel must fall out first, whereas the small ones must rise higher, grow longer, and fall out later, usually as larger drops. Thus, one should

expect that the first raindrops that fall from a shower should contain the highest concentrations of nucleating material. On the other hand, the raindrops that arrive with the heaviest rain should usually have formed on smaller nuclei and have moved through a higher trajectory accumulating a larger percentage of pure water from the vapor. This model is indeed supported by the data from moderate convective storms which show maximum concentrations during the shower onset and minima near the rainfall rate climax. The data from the severe storm of 10 May 1964, however, appears to depart strongly from this model.

Persistent severe storms, of the type presented in model form by Browning and Ludlam (1962), and called "steady state" storms by Newton (1967), appear to attain their persistence by achieving better organized circulations than are found in the lesser convective storms. In these, the updraft transports low level air to the upper troposphere, where it spreads out laterally forming the extensive anvil; the low level downdraft then becomes an organized flow of dry mid-tropospheric air capable of considerable cooling by evaporation, which contributes additional energy to the storm circulation (Normand, 1946). Two basic differences between storms of this type and the less severe, more transient convective storms are found in the level to which the updraft ascends, and the organization and level of origin of the mid-tropospheric downdraft. These two characteristics affect directly the parameters we have measured.

Firstly, the maximum height of ascent obviously controls the maximum dilution of the water solution in the cloud droplets, that is, the amount of water accumulated upon a given condensation nucleus increases as the cloud element formed upon that nucleus ascends to greater height. Thus the purest water should be found in the topmost reaches of the updraft. This selfsame water should be found in rain at the ground associated with the highest rainfall rates because in falling these particles traverse the greatest depth of cloud and so form the maximum intensity rain core.

The organization and level of origin of the downdraft, however, produce effects that modify the above relationship. Apparently (Newton, 1967), the most severe storms are characterized by the best organized downdrafts originating at levels up to 25,000 ft or so. Inasmuch as the rain core falls out of the updraft and descends with the downdraft in these storms (loc. cit.), water must be supplied to the dry downdraft air from the falling rain mass. This has the effect of increasing the solute concentrations in the falling water, at the same time that it contributes to the cooling and downward acceleration of the downdraft. It also obviously, decreases the rainfall intensity at the ground well below the values that would otherwise be achieved.

The effect of evaporation has been evaluated by Fujita (1959) from his own and Thunderstorm Project data. His curves (Figure 6) show that the amount of rain evaporated, R_e, in a downdraft originating at 15,000 ft., is about double the amount of rain that remains and reaches the ground, R_s.

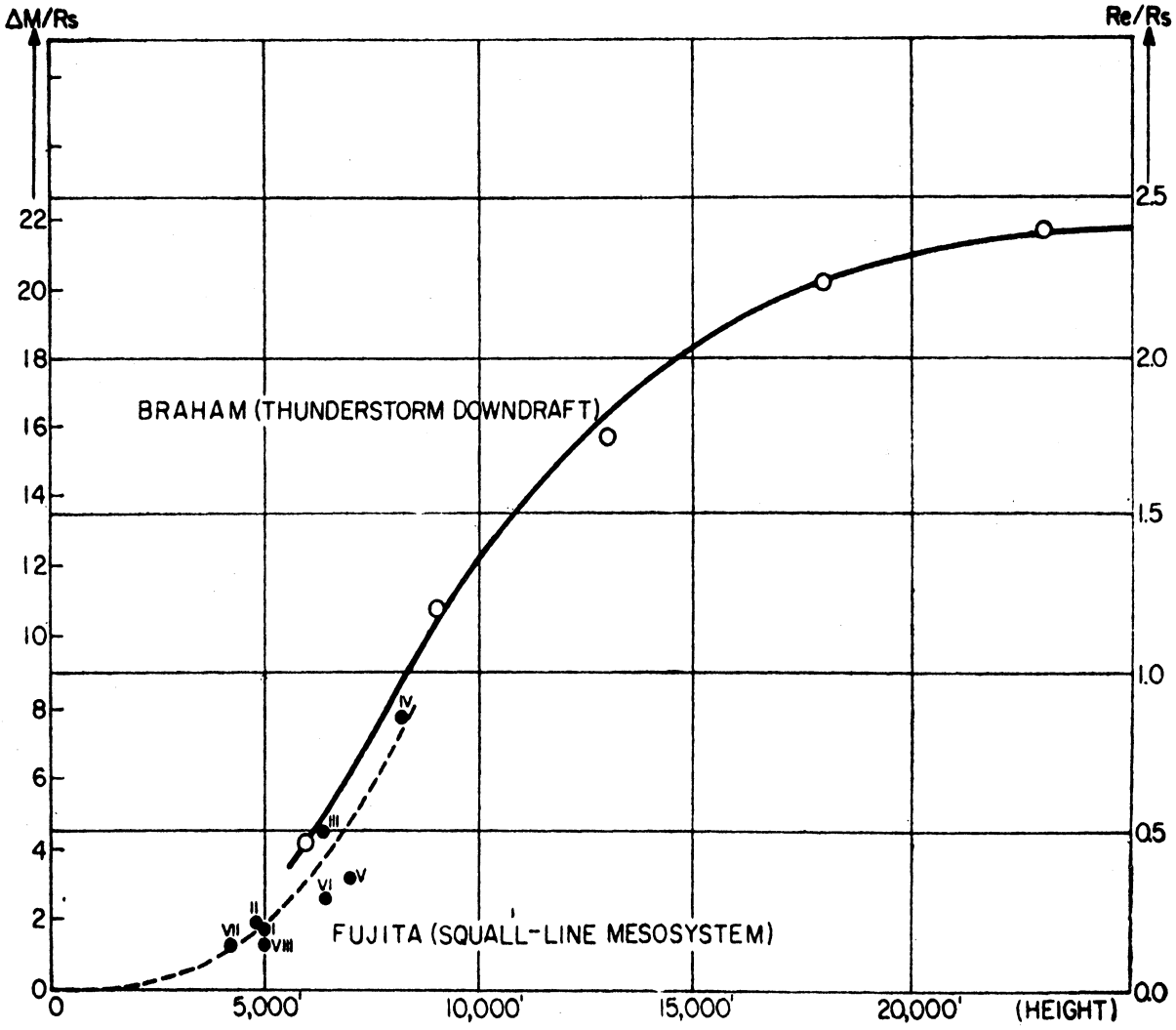

Figure 6.

The ratio of evaporated rain, R_e, to rain reaching the surface, R_s, in downdrafts of various depths, after Fujita (1959).

To estimate the height of origin of the downdraft, the wet-bulb potential temperature, θ_w, may be used as a criterion. Table 2 gives θ_w values for the downdraft air at our Chickasha station during the 9 and 10 May 1964 storms, and θ_w profiles from the soundings taken at Fort Sill (FSI) and Oklahoma City (OKC). The apparent source of the downdraft was below 700 mb on 9 May, whereas it was between 700 and 500 mb on 10 May. Using a nominal height of origin of 7,000 ft for the May 9 situation, and referring to Figure 6, the evaporated rain is estimated to be 0.5 to 0.6 of the rain received at the surface. This means that the solute concentration in the rain samples should have been increased 1.5- to 1.6-fold over that in the cloud water. For May 10 situation, using 12,000 ft as the height of origin, $R_e/R_s \simeq 1.6$, and the solute concentration enhancement factor is estimated at 2.6.

These preliminary findings suggest that reasonably quantitative models of the rainfall-rate and solute concentration profiles of convective rains may be constructed on the basis of the two characteristics, (1) height of ascent of updraft and (2) level of origin of low level downdraft, using the primary assumption that condensation on tropospheric nuclei is by far the principal solute scavenging mechanism.[7]

Further implications for the study of the structures of convective rain-producing systems by means of the techniques set forth here are suggested by the multiple-shower incidents reported above. These relate to the circulational systems and dimensions of the spectrum of convective storms ranging downward in size,

TABLE 2

HEIGHTS OF ORIGIN OF DOWNDRAFT USING θ_w

Observed at Chickasha				1800 CST Sounding				1700 CST Sounding	
				FSI		OKC		FSI	
Time (CST)	Temp. (C)	R.H. (%)	θ_w (C)	Pressure (mb)	θ_w (C)	Pressure (mb)	θ_w (C)	Pressure (mb)	θ_w (C)
				(a) May 9, 1964					
2100	21.1	96	21.7	970 (sfc)	21.8	969 (sfc)	20.8		
2105	20.0	94	20.4	889	20.4	850	17.9		
2110	18.9	91	19.0	850	20.4	824	16.0		
2115	18.9	92	19.1	700	15.8	700	13.9		
2120	18.3	99	19.4	633	16.5	696	13.1		
2125	18.3	100	19.4	592	14.2	628	15.6		
2130	18.3	100	19.4	517	15.3	538	15.2		
				500	15.8	500	15.3		
				(b) May 10, 1964					
1800	22.8	86	22.4					962 (sfc)	23.4
1805	21.1	76	19.7					900	21.9
1810	18.9	78	17.8					869	21.6
1815	18.3	82	17.9					850	21.8
1820	17.8	84	17.3					834	21.0
1825	17.2	92	17.9					750	19.7
1830	15.0	99	16.0					700	16.9
								650	16.6
								600	17.5
								550	16.4
								500	17.1
								430	19.0

intensity, and organization from the persistent, "steady state" class described by Newton (1966), Browning (1964), and Browning and Ludlam (1962).

IV. Conclusions

Trace analysis of rainwater is useful in many aspects of cloud and precipitation physics, precipitation scavenging, and convective precipitation processes. The technique is in its infancy, but has already produced a number of interesting results and holds promise of special applicability to the study of precipitation processes and severe storms.

The rain from individual shower cells shows marked variations in the concentration of contaminants produced by individual shower cells. In most cases, time profiles of concentrations are related to rainfall rate profiles in a particular way. Their relationship might loosely be termed "inverse," but close examination reveals that this is not strictly true, because opposing maxima and minima most frequently are not coincident.

The concentration time profile is quite distinctive. Most commonly the concentration peak occurs prior to the shower maximum, but there is a continuous range of values which the phase shift may take. This suggests that the causes of "direct" and "inverse" relationships are not different in kind (e.g., high-altitude vs low-altitude input), but rather of degree (e.g., up-draft penetration).

The data generally support the hypothesis that early precipitation of the largest nuclei produces high solute concentrations early in showers and low concentrations in the heavest rain.

The one persistent severe storm sampled showed a smaller concentration decrease during the heavy rain than that characteristic of non-persistent storms. The cause of the difference is probably that the persistent storm had a much deeper downdraft and hence evaporated much more water into the downdraft, thereby producing unusually high solute concentrations in the heavy rain.

The data collected in this effort to date suggest strongly that the spectrum of convective storms of lesser intensity and smaller dimensions than the most severe ones is accessible to study by these techniques. In addition to the fact that these lesser storms have a much higher frequency of occurrence than the severe storms, and are therefore significant on that ground, it is worthy of note that these lesser storms are also more subject to modification by artificial means.

Acknowledgments.

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