Detection of Indium as an Atmospheric Tracer by Neutron Activation[1]

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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 liter^{-1}. After aircraft injection at cloud base of 200 gm of indium, as finely divided particles from pyrotechnic flares, a maximum of 40 ng liter^{-1} 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 lanthanum 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 is determined by the natural background of the indium found.

1. 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 already present in large and variable amounts in the atmosphere. Because neutron activation analysis offers the high sensitivity needed, and the required facilities are available at The University of Michigan, we chose the tracer material to be optimum for detection by this method. Indium on neutron irradiation gives rise to the convenient 54-min half-life isotope In-116*m*, a beta and gamma emitter with a cross section of about 150 barns per atom of indium. A beta counting rate of about 500 counts min^{-1} was observed in samples containing 1 ng (10^{-9} gm) In, and a conservative estimate of the practical sensitivity limit in our laboratory is 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 emission, 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 program

About 200 gm of indium, associated with fine particles produced by burning seven pyrotechnic flares, were released over a period of 21 min from an aircraft flying at cloud base level in the storm updraft (Dingle *et al.*, 1969). Rain sampling bags of polyethylene supported by wide-mouthed baskets were deployed by a ground crew along roads where rain was expected to fall. Each of these was pre-inoculated with 50 ml of 6*N* 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 lanthanum 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 collected on 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

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neutron flux is built into this procedure by means of the La internal standard.

Samples were irradiated in groups of 4 for 20 min 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^{-1}.

After irradiation, the samples were processed further to separate the In-116*m* 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 6*N* 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-min In-116*m* beta particles.

Beta decay was followed for several hours to establish the presence of 54-min activity and then over several days to observe a longer-lived component identified as mostly 26.5-hr As-76. This component, observed in all decay curves of untagged and tagged rain water as well as in reagent blanks, was 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-hr La-140 in a single channel NaI(Tl) scintillation spectrometer. Gamma activity was measured usually twice on successive days to check for the expected 40-hr 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

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

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.

TABLE 1. Indium tracer rain samples, east-west road.

Sample no.	Aliquot volume (ml)	Collected rainfall[a] (mm)	Equivalent Indium[b,c] (ng aliquot^{-1}))		(ng liter^{-1})		$t_{\frac{1}{2}}$[d] (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	40.2	62
3B	513		20.8		40.6		62
4	1120	5.4	12.1		10.8		55
5[e]	440	2.2	5.7		12.9		78
6A[f]	789	7.3	10.7	21.5	13.5	14.5	79
6B[f]	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[g]	650	3.2	—		—		—

[a] Rain began at most sites before collectors were exposed: collector area, 0.20 m^2.
[b] Net short-lived radioactivity assumed to be all In.
[c] When the sample was analyzed in two parts, results for each part and the whole sample are given.
[d] For pure In-116*m*, $t_{\frac{1}{2}}=54$ min.
[e] 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.
[f] Plastic bag ripped, contamination by dust possible.
[g] Sample lost in analysis.

3. Results

The analytical results are presented below in two sections, the first dealing with the tracer-tagged rain samples collected near Holdenville, Okla., on 30 May 1967 (Dingle *et al.*, 1969), 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-inoculated convective storm are given in Table 1 for the east-west road, 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 tables.

Analytical results are given in terms of equivalent In. This means that the net short-lived component ($t_{1/2}\sim$1–1.5 hr), after subtraction of long-lived ($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-116*m* 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-116*m*. Both deposition and concentration of In are reported for each aliquot in the case of split samples. For these samples total deposition and overall 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 2.4–55.3 ng In per sample and concentrations from 4.3–40.6 ng In liter^{-1}. On the north-south road (Table 2) total sample volumes ranged from 656 (3.2 mm) to 1499 ml (7.4 mm), deposition from 5.4–24.8 ng In per sample and concentration from 8.3–23.7 ng In liter^{-1}. To tell whether some of the In in these 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 the "natural" or uncontrolled atmospheric content of indium, and 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, Okla., 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 hr 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

TABLE 2. Indium tracer rain samples, north-south road.

Sample no.	Aliquot volume (ml)	Collected rainfall[a] (mm)	Equivalent Indium[b,c] (ng aliquot^{-1})		Equivalent Indium[b,c] (ng liter^{-1})		$t_{\frac{1}{2}}$[d] (min)
1A	882	6.4	11.6	20.9	13.2	16.3	60
1B	403		9.3		23.1		71
2A	767	5.6	6.5	11.7	8.5	10.3	75
2B	374		5.2		13.9		68
3A	823	7.4	10.6	24.8	12.8	16.5	74
3B	676		14.2		21.0		70
4A	692	5.1	14.4	22.5	20.8	21.8	60
4B	344		8.1		23.7		77
5A	599	4.8	4.4	10.3	7.3	10.6	64
5B	364		5.9		16.2		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

[a,b,c,d] See footnotes, Table 1.

TABLE 3. Background rain samples without indium tracer, 5 May 1967.

Sample no.	Sample volume (ml)	Collected rainfall[a] (mm)	Equivalent indium[b] (ng sample^{-1})	(ng liter^{-1})	$t_{\frac{1}{2}}$[c] (min)
1[d]	932	0.32	10.9	11.7	80
2[d]	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, $\bar{X} = 6 \pm 3$ ng liter^{-1}

[a] Collector area, 2.5 m^2.
[b] Net short-lived radioactivity assumed to be all In.
[c] For pure In-116m, $t_{\frac{1}{2}} = 54$ min.
[d] Correction for internal standard counting rate, approximate only.

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.

In earlier work, we have collected and analyzed sequences of samples at our Chickasha station and at Willow Run, Mich., 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 liter^{-1}; for Table 4, 7.0 ng liter^{-1}). 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.

TABLE 4. Background rain samples without indium tracer, 30 May 1967.

Sample no.	Sample volume (ml)	Collected rainfall[a] (mm)	Equivalent indium[b] (ng sample^{-1})	(ng liter^{-1})	$t_{\frac{1}{2}}$[c] (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, $\bar{X} = 7 \pm 3$ ng liter^{-1}

[a,b,c] See footnotes to Table 3.

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. Junge (1963) points out that the data of Georgii and Weber (1960) show that the storm-to-storm variability tends to decrease with increasing rainfall amounts; that is, for 1-mm rains, the ratio of maximum to minimum concentrations is more than 20, whereas for 10-mm rains it is about 10. The rains we are dealing with are in the 10-mm 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, 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.

A number of reagent blanks were prepared in the field on days when rain samples were processed (Table 5). In addition to the reagents, four of these contained ~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 contained no In; therefore, all 16 samples can be considered reagent blanks. A mean and standard deviation of 1.5 ± 1.5 ng per 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 background rain samples, and this ob-

TABLE 5. Blank analyses.

Sample no.	Preparation date (1967)	Sample type	Equivalent indium[a] (ng sample^{-1})	$t_{\frac{1}{2}}$[b] (min)
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, $\bar{X}=1.5+1.5$ ng (sample)$^{-1}$				

[a] Net short-lived radioactivity assumed to be all In.
[b] For pure in-116m, $t_{\frac{1}{2}}=54$ min.

servation is quite consistent throughout the data we have so far compiled as is shown by the graphic presentation of Fig. 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 generally contained 2 ng or less In per sample.

In comparing the tracer rain samples with the background rain samples, however, it is more convenient to refer to unit volume In concentrations because the sample sizes vary. Making comparisons on this basis, the average background concentration of 6 ± 3 ng liter^{-1} 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, grading from the highest values near the western end (low sample numbers, Table 1) to values within the range of 6 ± 3 ng liter^{-1} 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 inoculated with the

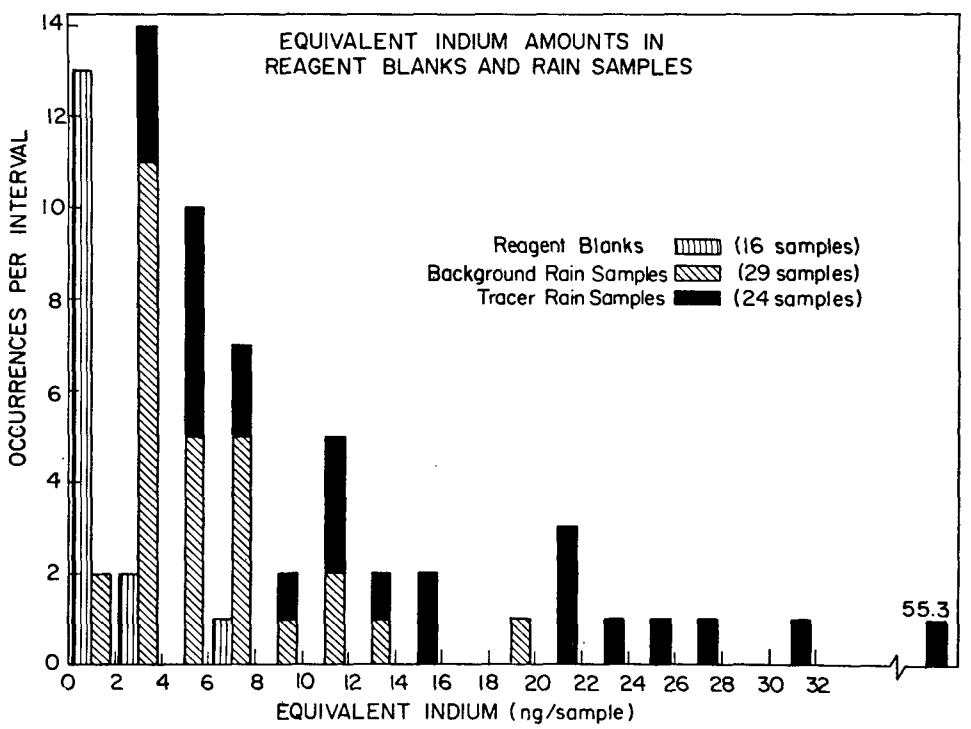

FIG. 1. Frequency of occurrence of equivalent indium amounts in reagent blanks, background rain samples, and tracer rain samples.

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 through each sample, and the counter sensitivity for the radioactivity measurement need not be known absolutely. A relative measurement of radioactivity can be made quickly, easily and precisely.

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 radioactivity counts. Four samples were irradiated simultaneously in each "rabbit." The chemical separation then required about one hour for an analyst and a technician working together, from the end of irradiation until counting began. The counting procedure and calculations in this work were done by 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 three 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 four samples for counting in less than an hour, 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 coworker, 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 10–20%, 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 reproducibility 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-inoculation 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 nonvolatile 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

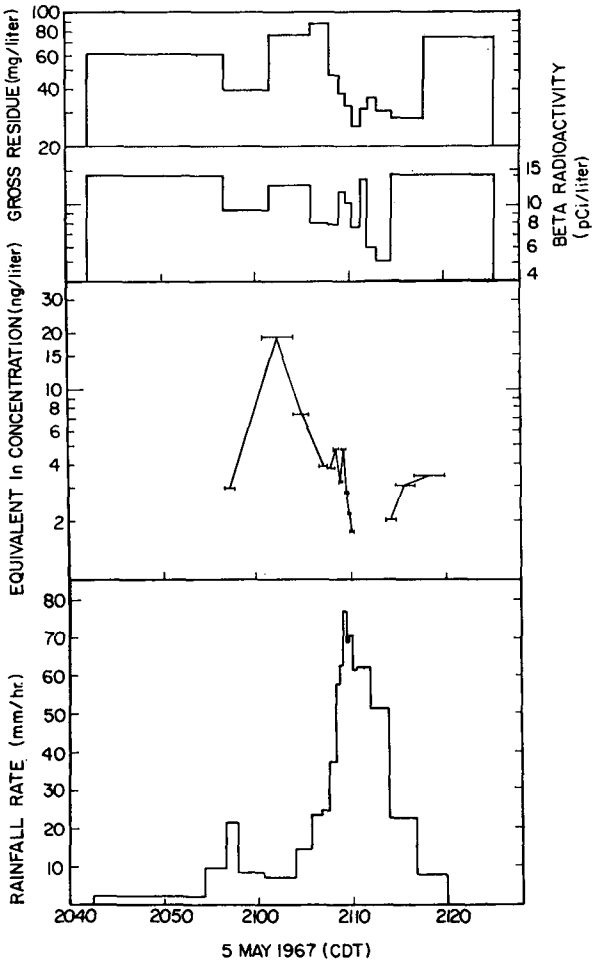

FIG. 2. Temporal variation of background indium, beta radioactivity, and gross residue during a convective shower. Horizontal bars indicate duration of In background samples.

exhibit less variability, both within and between 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 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 ng In liter^{-1} were found on the average, indicating a natural background somewhat above the reagent blank of about 2 ng liter^{-1}.

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 co-precipitation 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.* (1969) 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 not by reagent blanks nor 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 amount of indium released by five-to tenfold for each experiment. 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.

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