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RESULTS OF IN CLOUD TRACER RELEASES IN FRONTAL STORMS

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Six frontal storms have been injected with a variety of tracers. The examination to date of the tracers in rainwater has revealed that measurable concentrations have occurred in many of the samples. Maximum tracer deposition has occurred 20-40 nautical miles downwind from the area where the vertical wind profile, coupled with fall velocities of $1-5 \text{ m sec}^{-1}$, would indicate where initial ground deposition should have occurred. Some possible explanations are presented as well as a discussion of tracers used.

INTRODUCTION

In-cloud scavenging appears to play a major role in cleansing the atmosphere of pollutants (see Precipitation Scavenging (1970)).¹ The question then arises: "How effective is in-cloud scavenging in removing pollutants?" In order to answer this question, there is a need for field experiments. In the past, several successful field experiments have been carried out in which the injection of tracers has been made into convective systems (see Burtsev, et al.,² Dingle, et al.,³ Semonin,⁴ Skopauskas, et al.,⁵ Summers,⁶ Warburton,⁷ and Young, et al.⁸). The work presented in this paper differs in that it deals with in-cloud scavenging in prefrontal clouds.

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This report will cover experiments carried out in six storms. Also, the report includes the experimental design as well as description of the individual experiments, the results of analysis for tracers in samples taken during the experiments, and a section on recommendations for future field studies.

RESEARCH PLAN

The following problem areas were decided as necessary areas of research in in-cloud scavenging..

- (1) The testing of tracers to find those that would be deposited in sufficient concentrations to be measured.
- (2) Varying the physical and chemical character of tracers in order to see if there are differences in removal, eg., varying the size of particles, using tracers with differing solubilities, and comparing tracers that will nucleate with inert tracers.
- (3) Determining the variation of washout under differing meteorological conditions. Some are:
 - a. stable or unstable lapse rates in cloud,
 - b. temperature, $>0^{\circ}\text{C}$, $<-5^{\circ}\text{C}$, 0°C
 - c. liquid water content.

As seen above and in Table 1, which gives the plan on the basis of individual experiments, we have attempted to look at a number of differing meteorological conditions in frontal storms in order to estimate both the scavenging effects in storms where nucleation effects have been enhanced by AgI, as well as the scavenging effects in storms injected with inert tracer. The following is a discussion of the individual experiments with the results of analysis to date of some of the samples.

Experiments

Initially, part of the research plan (see (1) above) was aimed at finding a suitable tracer or tracers. Since Dr. Dingle² had prior success with In, it was selected as a tracer in the initial experiments. Li, and Ag were also selected to be released the first year.

March 11, 1971

On March 11, 1971, a test was carried out at Quillayute, Washington with In flares and Li stearate released into prefrontal clouds. The results which appear in Ackinson, et al.⁸ 1972, will only be briefly touched on. As indicated in Table 1, the 450 grams on In were released at 0° C, in a stable layer at 5000 feet within the storm on two legs each 5 nautical miles. Using the radiosonde winds and a fall velocity of 1.5 m/sec, the position for the release was determined such that the deposition of the tracer would occur in the center of the array. The release was made using the University of Washington's B-23. The deposition pattern is shown in Figure 1. From the pattern a calculation of the amount deposited was made. About 30% of the tracer was found to be deposited in 20 minutes or a total removal rate of about 100%/1hr.

It is interesting to note that the maximum concentrations in the deposition pattern did not occur in the area where the tracer was targeted to arrive initially, but at a position downstream on the last arc. If we were to use below cloud scavenging with a constant washout coefficient, the maximum deposition would be expected to occur at the initial depositing position with deposition decreasing with time. Since this doesn't appear to be the case, two possibilities have been put forth: (1) that the material injected is gradually picked up by the cloud water which is more easily scavenged (Davis, 1971)⁹ and

(2) that the rainfall rate must increase causing the scavenging coefficient to increase which will in turn cause the deposition to increase. The effect could also be a combination of the two. Looking at Figure 2, we have analysed for the rainfall rate using the volume/time to denote a mean rate for the time the individual buckets were out. Comparing this to the deposition we can see some correlation. (This should be kept in mind when the results of the December 5, 1973 test are discussed.)

Approximately 60 grams of Li was dispersed in the form of lithium stearate. The analysis for Li, to date, has not indicated any of the samples concentrations to be above background.⁹

The main result of the March 11 test was that analysis indicated positive In deposition. Therefore In could be used as a tracer in future tests. Also from analysis the decision was made for the 1972 test series that an expanded arc system was necessary to attempt to contain the maximum deposition.

April 16, 1971

The second experiment was carried out on April 16, 1971, when a release of 500 g of AgI and 450 g of In was made at -15°C at 12,000 feet on two legs of 5 nautical miles (see Table 1). Again, details are presented in Ackinson, et al. 1972.⁹ A photograph taken after release from the University of Washington aircraft indicated a line of convective development along the speeding path.⁹ The release, unfortunately, was made in a lighter wind speed than the previous storm. Using the same size grid for the lighter wind speed resulted in loss of detail in the deposition pattern. Figure 3 shows the deposition of In.

If we use the winds at 12,000 feet the time of arrival on the first arc near Quillayute (UIL) airport would be 30 minutes after initial deposition using a 1.5 m/sec full velocity. The deposition path should have moved

northward as indicated by the 12,000 foot winds, crossing the northern arc at 75 minutes after initial deposition. There is some organization to the pattern in the western edge of the arcs as seen in Figure 3. However, there are significant measurements of In that indicated deposition further to the east. These measurements could not be explained from the UIL radiosonde taken at the time of release. This indicated that in future test better definition of the wind flow pattern aloft would be necessary to accurately predict the deposition pattern of the injected tracer. The wind directions computed from the 1935Z sounding, 1 hour and 44 minutes before release, would explain the eastern deposition (see dashed lines on Figure 3). When Ag was analysed for the samples, little correlation was found to exist between measured Ag and In. Three samples had both In and Ag, whereas 36 samples had measurable quantities in either Ag or In, but not both. Also, the maximum concentration for both tracers appear independently. The main result was that both inert tracer and the Ag which were injected into a heavy seeded area were measured at the ground.

March 21, 1972

On March 21, a tracer release was made in prefrontal clouds with 450 grams of In and 2 kg of AgI released over 2 legs of 8 nautical miles.¹² The release on this occasion was made over an expanded arc which is shown in Figure 4. The reason for the expanded arc was the result of the previous year's analysis of a March 11 release indicated maximum deposition on the last arc (Ackinson, et al.,⁹ Davis¹⁰). By increasing the downstream size of the arc, it was hoped that a falloff from a maximum deposition would be seen. Also an auxiliary radiosonde was established south of the arc system at Hoquian, Washington in order to better define the upper wind flow pattern.

The injection of both tracers was made at 6800 feet at -5° C in a stable layer by the University of Washington aircraft.

The resulting deposition as determined from analysis of both Ag and In indicated that only three samples showed slightly above background concentrations of In (see Figure 4) and no samples showed concentrations above background of Ag.¹³

In an attempt to understand why the concentrations were near or below detection, radar and pilot reports were examined. Problems existed with the RHI at Neah Bay, thus no photos or readings were available during the test. Some readings off an auxiliary scope 14 minutes before the test indicated returns to 10,000 feet in the area of the release. The PPI photos taken during the test period failed to indicate any echo returns from the area of release.

A review was made of pilot reports. The aircraft during the flight to UIL to the release point reported at one point breaking out of clouds into the sun at 6300 feet. A review of tape recordings taken aboard the release aircraft indicated that visual ground contact occurred occasionally during the release, as well as a reported bright spot due to sun overhead that was visible at times. They did report that the release was made in rain although no indication of intensity. From their description of the clouds, the rain was probably sporadic along the release path. Also, rain gages on the ground indicated only light rainfall during the period when tracer deposition should have occurred. Thus the initial conclusion reached was that although the release was made in cloud, the rate of removal from the cloud was insufficient for organized measurable surface concentrations.

March 31, 1972

Two tracer experiments were carried out during the flight (see Slinn, 1973¹¹). Rhodamine had been previously used in below cloud scavenging experiments with success (Dana¹⁴). Thus it was decided that a test be made to determine the suitability of Rhodamine as an in-cloud tracer. Rough calculations indicated that deposition above the detection limit of 1 ng should be possible. Based on these calculations 600 grams were released at 6200 feet at 0° C on two flight legs of 9 nautical miles¹⁰ by the University of Washington B-23.

A fluorimetric analysis of precipitation samples in the area of expected deposition revealed none above background. Samples further to the west indicated positive results plus some organization (see Figure 5).⁹ Further analysis of samples this year by a spectral fluorimeter indicated that no fingerprint for Rhodamine in the samples could be detected (Lee, 1974¹⁵). In fact, when samples were spiked with 20 ng of Rhodamine, interference was still too great to obtain a fingerprint. From these results, our conclusion is that Rhodamine is not suitable as an in-cloud tracer.

The second tracer release in the same storm was made at 12,000 feet at -15°C in a stable layer with significant liquid water. The result from the release was a hole in the cloud (see Slinn 1973¹²). The release was made with a stable lapse rate and significant liquid water to see if the nucleation due to AgI would occur and whether deposition of precipitate could be targeted. Again using 1.5 m/sec full velocity and the wind profile from VIL the targeted area was Forks, Washington (see Figure 6). One sample, as indicated in Figure 6, was analysed by anodic ion stripping voltameter which gave a reading equivalent to 2 ng of In.

February 28, 1973

A second attempt was made at a release which would cover the extended arc system. A storm on February 28, 1973, was selected for a tracer injection. The storm was not as intense as desired, but a decision was made for an attempted release. Using the winds from radiosonde data at Quillayute, Wa., an area for release was selected for a tracer injection as seen in Figure 7. Radar indicated echos in that region; however, they did not appear to be precipitating. Instructions were given the aircraft to orbit the area of release and if they hit a precipitating cell to release. These instructions were carried out and a cell was injected with tracer by the Battelle Cessna-411. Lithium flares with 216 grams of Li and ~400 grams of Indium* from burners were released at 5000 feet at 0° C.

The precipitation records show little rain falling (<0.1 in/45 min) on the first arc crossed using 15 min sequential and rain gage measurements during predicted passage. No rainfall was recorded by Lake Pleasant rain gage measurements during time of predicted deposition. A rain gage further to the north and west of the deposition pattern indicated .01 in/15 min during time of predicted deposition. The sequential samples east of Lake Pleasant indicated a minimum slightly before the time estimated for deposition, but during the time when deposition should have occurred showed a slight increase to .001 in/15 min.

The average pattern of rainfall during the test was calculated using the amount collected by the time exposed for each sampler. The result of this calculation indicated a maximum along Burnt Mountain Road (see Figure 8).

*837 grams were planned for release, however due to failure in the release system, only ~1/2 were released.

The initial analysis of samples for Li is shown in Figure 7. As can be seen, the maximum occurs in the area of maximum precipitation. The concentration of Li in ppb water varies widely with values running from .07 ppb to 10.2 ppb. The concentration in the samples along Burnt Mountain Road and a sample at the junction of Highway 101 and the Hoh River Road. An In analysis has not been carried on this test yet. The only conclusion that can be drawn so far is that Li values varied considerably with some high deposition values.

December 5, 1973

A series of tracer release was made by the Battelle aircraft at 5000 feet in a stable layer at 2° C (see Davis, Young and Thorp, 1974¹⁶) on December 5, 1973. The plan for the release of the tracer differed from past plans. Previously control on the area of burn was determined by both the rain trajectory and the wind direction at the release height with the aircraft releasing into a precipitation cell. The updated plan was made because the analysis of the March 21, 1972, test indicated little or no deposition of tracer. The new plan required a cell visible on the PPI and indication that precipitation is occurring on the RHI and further that the cell is located in a general area that will allow deposition to occur over the arcs.

The tracer portion of the experiment included two tracers previously used, In and Li, and the addition of four new tracers for use at UIL: Eu, Os, Re, and Ir. The effect of particle solubility was to be studied by the simultaneous release of a soluble tracer such as lithium and a nonwetable tracer such as iridium or indium. A water-soluble tracer should act as a much better CCN than a nonwetable tracer and therefore might be scavenged more rapidly.

In addition to the above experiment, it was planned to conduct other experiments. A burn of a rhenium-europium mixture in the acetone generators was to be carried out to determine whether the two tracers would behave identically, showing the same deposition patterns. Also planned was a release of osmium into a precipitating cell to determine the suitability of osmium as a tracer.

On December 5, 1973, the PPI radar at Makah AFB near Neah Bay, Washington, showed a cell imbedded in a frontal storm moving toward the planned release area near Quillayute. The decision was made to release tracers into this cell, and the Battelle Cessna-411, equipped with indium flares on one wing, and two aerosol generators on the other, was directed to this cell. With the transponder signal from the Cessna was displayed on this radar scope, positioning was made possible of the aircraft rather accurately within the precipitating cell.

When the Cessna reached the proper position, an attempt was made to burn the europium-rhenium solution in the aerosol generators. But the generators refused to light, probably because the burner nozzels had been clogged by a precipitate which later was found to have formed in the tracer solution. The indium flares were then burned from 0950 to 0956, and a second attempt was made to light the aerosol generators. This time because one of the generators fired and the other did not, lithium, iridium, osmium, and the remaining rhenium-europium solution were released sequentially with only one burner firing. The burn of the last tracers was completed at 1025. The tracers were all released at 5000 feet at an ambient temperature of 2° C and in a stable layer (see Table 1). Pictures of the scope displays of the RHI and PPI radars at Makah AFB were taken before, during, and after the tracer

release (see Figure 9). Following the passage of the cell, samples of rain were collected from the 190 rain collectors of the rain sampling network.

The result of the analysis of the samples to date is shown in Figures 10, 11, 12 and 13. In the case of In in Figure 10, an organized pattern can be seen. This may be since it is the only tracer for which the analysis is nearly complete. As can be seen in this test, the expanded arc system was used with the north-south extent of the pattern over km.

The resulting deposition pattern is surprising in that we expected from winds from the UIL radiosonde the pattern would come down oriented $\sim 200^\circ$, rather than 180° . Right now we do not have any explanation for this except that in looking at the cell track on radar it appears to have moved on a 180° heading. Thus the radiosonde winds at UIL do not appear to be reliable for release to the south. (You might note that the previously mentioned rhodamine, March 31, 1972, test had possible tracer in an area not expected.) The December 5 analysis for all tracers was based on the 200° wind direction for aligning the surface deposition pattern. That may be one reason the analysis of the other tracers does not show organization that In does, in that the first analysis should have been of samples further to the west.

When considering the deposition rate for In as a function of time (see Figure 15) we see a factor of 3 increase at ~ 3500 seconds. Unfortunately, our analysis is still not complete, but it appears that the maximum deposition occurs on the last arc so we are not able to see the decrease in deposition rate. At this time there has not been developed a totally satisfactory answer. One possibility which was investigated was the rainfall rate which

could have caused the increased deposition. The volume of water collected in the samplers was used to compute a mean rate of rainfall (see Figure 16). As you will recall in the March 11, 1971, storm, the maximum rate of rainfall did correlate with the maximum in deposition. Again, the maximum deposition occurs with the rainfall maximum.

From the incomplete analysis, the indication is that the amount deposited was ~10-15%/hour. Hopefully in the near future the analysis will be completed.

CONCLUSIONS AND RECOMMENDATIONS

Organized tracer deposition patterns have been observed in at least three of six storms injected with tracer. Maximas in deposition have occurred on the last arc on March 11, 1971 and December 5, 1973. Two possible causes have been suggested:

- a) increased rainfall caused increased deposition and/or
- b) attachment coefficient of the tracer to the cloud water would cause a delay in the maximum deposition time.

The success of seeing inert tracer deposited from injection into a stable layer has been tied closely to the insertion of tracer into a cell precipitating heavily enough to be seen by RHI and also be noted on the PPI. This was borne out by the comparison of surface deposition and rainfall between the March 21, 1972, storm and the December 5, 1973, storm. Future test of this type will be carried out with a close link between the radar and aircraft.

In two tests where nucleation was expected by overseeding by AgI, tracer has also been observed on the ground (April 16, 1971, and in the only sample analyzed for March 31, 1972).

Problems have existed in predicting the deposition pattern because of differences between the radiosonde winds at UIL and the observed deposition pattern. Aircraft winds, increased radiosonde coverage as well as the use of dual doppler radar system are recommended to better define the air flow.

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March 21, 1972
In and Ag Release
6800 feet at 0°C
2 kg AgI and 450 g In

(fig 3 map)

Fig 4

NEAH BAY
RADAR

MARCH 31, 1972
RHODAMINE

FEB. 28, 1973
Release @ 5000' 0°C

1523 - 1541 PST
(2323Z - 2341Z)

Li Flares 216 grams
In Burners ~400 grams
(~837 - 1/2 released)
AgI Burners - 3000 g

5000' winds UIL
180/14.5 m/sec
— Predicted Cloud Trajectory
— Predicted Ground Trajectory
— Revised Ground Trajectory

Sequential rain

Fig 1

STRAIT OF JUAN DE FUCA

MAKAH
RADAR

UIL

PACIFIC
OCEAN

INDIUM
RELEASE
AREA

0.5 1 2 3 4 5 6 6 5 4 3 2 1 0.5 0

$\frac{2}{3} / 10$
73. ~ 80
3/220

DEPOSITION PATTERN
ng/0.03m² (PER BUOY)

DEC. 5, 1973
INDIUM RELEASE
700 GRAMS
@ 5000' IN CELL
2°C - STABLE

5 10

NAUTICAL MILES

66 = 110

Fig 3

Europium Deposition Pattern
ng/bucket (.2 meter dia)
Dec. 5, 1973
Eu release (half raw, half
burned)
73 grams, 5000', in cell,
Stable, 2°C.

Dec 5 1973 In Release (700grams Released ~ 72 grams d