Phloroglucinol Seeding of Undercooled Clouds[1]

ROSCOE R. BRAHAM, JR.

Cloud Physics Laboratory, The University of Chicago

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ABSTRACT

A series of twelve releases of phloroglucinol were made into stratus clouds at temperatures of −7C to −17C.

Showers produced by dry ice seeding were used to identify particular spots in the layer clouds from which the exact locations of the phloroglucinol releases could be obtained by simple navigation. Visual observations of the cloud behavior and Formvar replicas of cloud and precipitation particles provided a means for judging the effects of the phloroglucinol.

It is concluded that phloroglucinol will induce the formation of ice in undercooled clouds. However, in these experiments, it was not nearly as effective as the dry ice in causing shower formation.

1. Introduction

One of the exciting new developments in cloud physics is the realization that heterogeneous nucleation of ice from undercooled liquid water can be induced by organic substances. Several laboratories are carrying out, or have recently carried out, studies of this nature. Some of these studies have already appeared in the literature (Bashkirova, 1957; Head, 1961, 1962a and 1962b; Komabaysi, 1961; Krasikov, 1961; Langer and Rosinski, 1962; Power and Power, 1962). Other work known to to the author includes Fukuta (Imperial College, unpublished).

The interest of the author in this area was aroused by a personal contact with Langer and Rosinski, and through visits to their laboratory at Armour Research Foundation, Chicago, during the winter of 1961–1962. At that time, Langer and Rosinski were engaged in an Air Force-sponsored basic study of organic crystals as icing nuclei. Of more than 30 compounds studied in the laboratory the most promising was phloroglucinol, which repeatedly caused rapid nucleation of cold-box clouds at temperatures of −2C to −3C.

It was immediately evident that their laboratory findings should be subjected to field trial. The field facilities of Project Whitetop, including our instrumented airplane, appeared ideally suited for such a study. Accordingly, a sizeable quantity of phloroglucinol was prepared and taken to West Plains, Mo., at the start of operations of the 1962 summer season. It was our intention to seed undercooled cumulus clouds on days declared non-operational for Project Whitetop silver-iodide seeding operations. However, clouds suitable for a field check of phloroglucinol as an ice nucleant did not appear during the summer months and the tests had to be postponed.

During December 1962 and January 1963, a series of Arctic-airmass anticyclones moved across the Great Lakes area and gave rise to extensive undercooled stratus layers which enable us to carry out a group of experiments in phloroglucinol seeding. A summary of these experiments and an assessment of the results are given in the following sections.

2. Phloroglucinol as a seeding reagent

Phloroglucinol is an organic compound formed by replacing the chlorine atoms of 1, 3, 5 trichlorobenzene with OH radicals. It also goes under the name of 1, 3, 5 benzenetriol or sym. trihydroxybenzene [$C_6H_3(OH)_3$]. At room temperatures it exists in the form of brown rhombic crystals. It is moderately soluble in water (1.1 grams per 100 ml) and is very soluble in alcohol.

The laboratory experiments at Armour Research Foundation involved introducing either dry powder or an atomized solution containing phloroglucinol into a laboratory cold box after a cloud had been introduced by means of a steam generator. In the Langer-Rosinski experiments it was found that phloroglucinol induced rapid ice nucleation at cloud temperatures of −2C and colder. They estimated that 10 to 20 per cent of the particles nucleated ice crystals before falling out of the relatively small laboratory chamber. When dispersed in a water solution, phloroglucinol induced freezing of small droplets at similarly warm temperatures. This ability to promote freezing of solution drops was checked in the Cloud Physics Laboratory. We find that phloroglucinol will raise droplet freezing temperatures several degrees but in no case to temperatures as warm as −2C.

The phloroglucinol used in this series of field experiments was purchased from Morton Chemical Company, Ringwood, Ill. As received, it consisted of crystals ranging in size from about 0.5-mm diameter to less than

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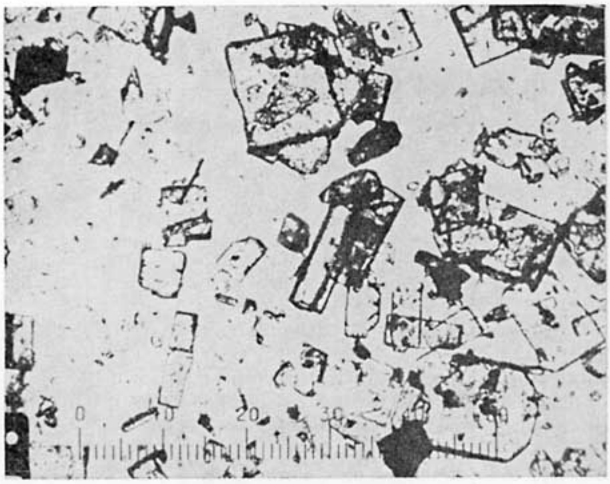

FIG. 1. Photomicrograph of phloroglucinol. Smallest scale division represents 20 microns.

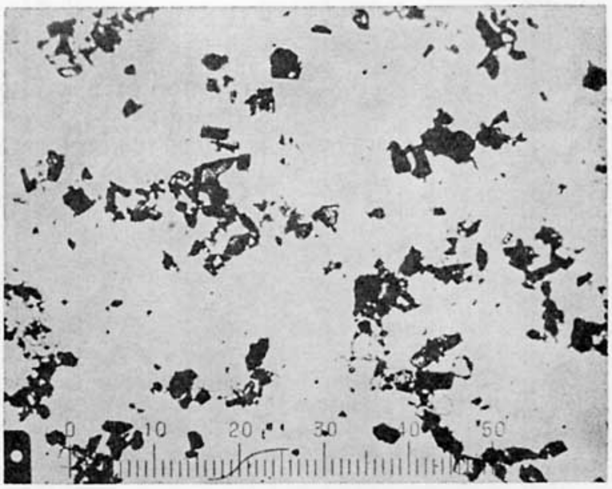

FIG. 2. Photomicrograph of ball-milled phloroglucinol. Smallest scale division represents 20 microns.

5-microns diameter. Bulk density of the original powder was 0.6. Since the Armour Research Foundation Laboratory studies had suggested that particles as small as 1 micron would be suitable for nucleation, one half of the material was ball milled in order to reduce its average particle size. This work was done for us by Mr. Langer in a manner which he describes as: "The powder was ball milled for 24 hours as a water slurry, then it was allowed to dry in air. The cake was broken up by hand and then sifted through a 325 screen with an electric sieve shaker. Two or three times the volume of Cabosil was added to this and milled. The resulting powder was free-flowing and was active at −2C." (Langer and Rosinski, 1962).

The result of the ball-milling was to reduce the largest particle to 50 microns. It apparently did not markedly increase the number smaller than 20 microns. The optical density of the milled particles is noticeably greater than that of the unmilled, presumably because they are coated with the Cabosil. The purpose of adding Cabosil was to reduce the tendency for clumping and indeed it may have been partially effective in doing this even though the microscope studies show that the milled material still has strong clumping tendencies. By the same token, the Cabosil probably made the individual particles harder to wet and as a consequence less effective as ice nucleants. Photomicrographs of the phloroglucinol material used in these experiments are given in Figs. 1 and 2.

3. Experimental procedure

The experiments described herein were carried out on 18 and 19 December 1962, and 4 and 5 January 1963, in the off-airways areas of northern Wisconsin and the peninsula of northern Michigan. Release 1 and release 2 of 18 December were made in a rather thick altostratus layer, while all other releases were in low stratus. Cloud liquid-water contents ranged from 0.1 gm m^{-3} to more than 0.3 gm m^{-3}, as measured by a Johnson-Williams hot-wire device. Temperatures at the various levels of flight were measured by a platinum resistance element mounted in a reverse-flow housing. Temperatures at the tops of the cloud layers, where seeding was done, is shown in Table 1.

A feature of the Project Whitetop airplane is a 4-inch diameter tube which permits cloud and precipitation particles to pass directly into the cabin. Formvar replicas of these particles were taken at the exhaust end of this tube. Microscopic examination of these replicas has assisted considerably in the assessment of these experiments.

As an aid in detecting ice crystals, Mr. John Kidney, who assisted in the experiments, rode in the co-pilot's seat and extended his gloved hand through the open cockpit window. Occasional ice pellets, too few to be seen otherwise, could be observed when they impacted upon his dark glove.

Four different phloroglucinol preparations were employed in these experiments:

1) Dry powder as received from the manufacturer;
2) Dry powder ball-milled and mixed with Cabosil;
3) The original dry powder dissolved in alcohol—both methanol and isopropynol;
4) Water slurry of the original dry powder.

In these releases the seeding substance was manually dumped directly into the airstream through a fuselage opening normally used with the automatic replicator or through the open, co-pilot's window. Release rates ranged from 42 to 2875 grams of phloroglucinol per mile of flight. In most cases the releases were made into the upper 100 feet of the visual cloud.

TABLE 1. Summary of phloroglucinal seeding experiments.

Type of release	Release number	Seeding rate gm mile^{-1} (particles per mile)	Seeding temp. deg C	Cloud depth ft	Cloud liquid-water content gm m^{-3}	Results
Alcohol solution	1	230 (1.5×10^8)	−17.2	1000	0.1–0.15	Moderate snow shower to ground, first evident below cloud 20 min after seeding; strong after 66 min.
	4	380 (1.5×10^8)	−7.5	2500	0.2	Weak snow shower to ground 36 min after seeding.
	13	525 (2.5×10^8)	−12.0	1200	0.2−0.3	Snow shower to ground 47 min after seeding; questionable origin because natural showers beyond immediate vicinity at end of experiment.
	19	1000 (2.2×10^8)	−11.0	1000	0.1	Ice crystals and snow pellets on replicas and on glove, too sparse to see against sky or surface background. Natural shower 40 mi away at end of experiment.
Dry, unmilled PHL	5	190 (1.1×10^9)	−9.8	1600	0.2	Snow shower to ground—heaviest PHL shower in entire set of experiments; shower reached maximum intensity 1 hour after seeding.
	9	1500 (9×10^9)	−11.2	800	>0.1	Negative (observed 57 min).
	15	2875 (1.7×10^{10})	−12.0	1200	0.2–0.3	Negative (observed 46 min).
	17	2875 (1.7×10^{10})	−11.8	1500	0.2–0.3	Negative (excellent test conditions observed 55 min).
Dry, milled PHL	2	42 (6×10^8)	−14.0	1100	0.1	Snow shower to ground, natural shower 20 mi away at end of experiment.
	7	210 (3×10^9)	−12.3	2100	0.2	Negative (observed 77 min).
	18	1600 (2.3×10^{10})	−12.0	1000	0.1	Snow pellets below cloud, ice crystals and frozen drops in cloud, too sparse to be seen against land or sky background but easily detected on replicas and glove. Few ice crystals found on control replicas outside seeding point. Natural shower sighted 30 mi from release point at end of experiment.
Water slurry	11	540 (1.9×10^8)	−9.0	1300	>0.1	Negative (observed 51 min).

The first two experiments indicated that the phloroglucinol seeding would not be followed by the marked visual effects which we have learned to associate with dry ice or silver-iodide seeding of undercooled stratus. These effects are:

1) Darkening of the upper cloud surface;
2) Development of an undersun, sundogs, and other optical phenomena when the upper surface is viewed toward the sunlight;
3) Substantial snow showers when viewed from below the cloud base.

Therefore, on the third and subsequent releases, it was the practice to include in each experiment a dry-ice release for the purpose of marking a particular spot from which the phloroglucinol releases could be located by simple navigation patterns. As the experiments proceeded we dropped smaller and smaller amounts of dry ice in an effort to produce a shower of visual intensity equal to that produced by the phloroglucinol. In this we did not succeed. Even very small amounts of dry ice produced snow showers much heavier than the largest phloroglucinol shower produced.

The use of dry ice showers as markers in the field testing of seeding materials is strongly recommended. It completely eliminates the necessity of worrying about wind drift, which at cloud level may differ from that below the cloud, and permits the experimenter to leave the seeded point and return to it with confidence tens of minutes later. In this series of experiments the release points were observed for periods up to one hour in order to check for possible delayed shower development.

4. Experimental results

A total of nineteen releases were made during this series of experiments. Twelve of these were phloroglucinol and seven were dry ice which provided the marker showers needed for navigational purposes. Pertinent data concerning the 12 phloroglucinol releases, clouds into which the releases were made, and the results thereof, are given in Table 1. In this table the releases have been grouped by type of release to facilitate comparison of the results. A complete report of all 19 releases has been given limited publication by Braham (1963).

In all four of the alcohol solution releases, showers formed at the release site. On releases 1 and 4 there were no other showers visible at any time during the experiments, nor were there any indications of natural ice crystals in the clouds. Natural showers were visible at the end of experiments 13 and 19, although in both cases they were at a considerable distance from the phloroglucinol release position. Only one of four releases of dry, unmilled phloroglucinol powder produced a snow shower. This particular shower was the heaviest of all those which might be attributable to phloroglucinol seeding. Ice pellets, ice crystals and frozen water drops accompanied two of the releases of the dry, ball-milled phloroglucinol but in both cases natural showers were visible at ranges of more than 20 miles from the seeding locations. The third release of the milled material and the water slurry release were both complete failures.

On the basis of these field trials we conclude that under favorable conditions phloroglucinol will induce the formation of ice crystals within undercooled clouds; however, it is obvious that it is not nearly as effective as dry ice for this purpose. Although our tests were somewhat crude, we were not able to release a small enough quantity of dry ice to produce a shower of visual equivalence to the heaviest phloroglucinol shower.

5. Discussion

The field experiments appear to stand in marked contrast to the laboratory findings of Langer and Rosinski. In the laboratory the materials nucleated with great rapidity and at relatively warm temperatures (−10C in the Bigg-Warner box and −2C or −3C in the laboratory cold box). In the field studies on natural clouds the first visual indications of nucleation occurred 12 min after seeding in the one case of a shower from the dry, unmilled powder (release 5) and 20 to 40 min after seeding in all of the alcohol releases. It is unlikely that more than about 10 min of this time interval can be attributed to the time required for ice crystals to grow large enough to appear at the cloud base. We note that the dry ice seeding produced virga from the same clouds in 10 to 15 min in spite of the fact that diffusive growth of the crystals must have been slowed considerably because of gross overseeding in the immediate vicinity of the dry ice release. The most likely explanation is that the slow response of the phloroglucinol was due to the fact that considerable time was required for the particles to pick up the first few layers of water. Both in the Bigg-Warner box and in the laboratory cold box the clouds were formed by condensation from warm vapor. It is likely that the size of the apparatus prevented the droplet-vapor system from coming into temperature-vapor pressure equilibrium. As a consequence, the vapor density and cloud droplet concentrations in the laboratory cold boxes were much larger than we encountered in the natural clouds. Thus the diffusion of water to the phloroglucinol particles and the chances of collision between these particles and cloud droplets would be much enhanced in the laboratory clouds.

At no time during the experiments were the phloroglucinol releases accompanied by visual manifestations of ice crystals when viewed from above the cloud; the concentrations of ice crystals were much less than that of the water droplets.

It is instructive to consider briefly the number of ice crystals one might expect from phloroglucinol as compared with dry ice. The number of crystals produced by dry ice is directly related to the amount of dry ice evaporated within the undercooled portions of the cloud. In a previous study (Braham and Neil, 1958) it was shown that dry ice particles larger than about 3-mm diameter could not be expected to evaporate completely before falling through these thin stratus layers. In these experiments the ice was pre-crushed into fragments ranging from about 2-cm to 2-mm diameter. For our purposes, it is sufficient to take 5 mm as an "average" size. This means that only about one half of the dry ice could have evaporated in falling through the cloud layers. Each gram of dry ice evaporated will produce about 10^{12} to 10^{13} crystals under typical conditions. Thus the lightest dry ice releases in these experiments would have produced an estimated 10^{15} ice crystals per mile. If the initial growth of some of these crystals were delayed because of the competition for vapor and if we could regard the duration of the shower as determined by a continuous diffusion of very small ice crystals into unseeded cloud where they grew without competition, we could estimate the number density of ice particles in the shower at the cloud base. Replicas made in these showers show that an average snow pellet diameter would be about 250 microns. Such a particle will fall at a speed of about 1 m $\sec^{-1}$. Making use of the visual observation that the shower widths were about 1 mile, we estimate that the snow pellet concentration below cloud base should have been about 3×10^4 m^{-3}. This compares resonably well with the visual shower density.

Turning now to the seeding with dry phloroglucinol, we assume that a single particle of phloroglucinol will produce, at most, a single ice crystal. Thus we can estimate the maximum number of ice crystals expected from the number of particles released.

Photomicrographs were prepared of the dry, milled

TABLE 2. Phloroglucinol size distributions.

Size class (microns)	Number particles per gram Unmilled	Milled
<25	3.6×10^6	7.7×10^6
25– 49	1.6×10^6	3.7×10^6
50– 74	0.5×10^6	2.6×10^6
75– 99	0.2×10^6	0.3×10^6
100–149	0.1×10^6	—
150–199	0.03×10^6	—
200–249	0.02×10^6	—

TABLE 3. Size distribution of particles from the alcohol releases.

Solution droplets Size microns	Number per gal	Phloroglucinol particles after evaporation (Equivalent cube—for 420 grams per gal)
0– 99	6.5×10^7	30 microns
100– 299	11.2×10^7	95 microns
300 499	2.5×10^7	160 microns
500– 699	0.7×10^7	235 microns
700– 899	0.2×10^7	315 microns
900–1099	0.03×10^7	400 microns

and unmilled powder (Fig. 1 and 2). Care was used to obtain samples which were as representative as possible. From the photomicrographs an estimate was made of the particle-size distributions and numbers of particles per gram of material. These results are given in Table 2. Combining these with the known dispersal rates, we arrive at seeding rates of 10^9 to 2×10^{10} phloroglucinol particles per mile. These estimates probably are too low in the smaller size classes, perhaps as much as an order of magnitude, because of the problem of obtaining a representative sample.

In the case of the alcohol solution releases, the number of phloroglucinol particles produced is probably governed principally by the number of solution drops created when the material was dumped into the airstream. This number is hard to estimate accurately but an attempt was made along the following lines. Several years ago we were engaged in water spray seeding of cumulus clouds. As a part of that research we measured the size spectrum resulting from dropping water through a 3-inch diameter opening in the belly of a B-17. That measurement was made by releasing the water from a flight altitude of 50 ft over several transverse rows of dye-impregnated filter papers. If we assume that the same distribution applies to the alcohol solution released through a 1-inch diameter opening in the belly of a Beech D-18, we obtain the drop distribution shown in Table 3. These data are probably correct to an order of magnitude. Translating these drop sizes into the phloroglucinol particles obtained when the alcohol evaporates, under the assumption that only one particle forms from each drop, we obtain values given in column 3 of Table 3. This exercise suggests that we might expect about 2×10^8 particles per mile of flight from the alcohol solution releases.

The length of time that a phloroglucinol particle remains in the cloud depends upon its falling speed, hence upon its initial size. The falling speeds of particles of milled and unmilled powder were measured in the Cloud Physics Laboratory. The results are shown in Fig. 3. It is interesting that the milled material has a slightly higher falling speed, presumably because of the somewhat greater clumping tendency. Based upon these measurements we find that particles larger than about 250 microns will fall through these thin stratus layers in times of 5 to 7 minutes, and we must therefore exclude them in our analysis of the seeding observations.

These computations suggest that the seeding rate was about 2×10^8 particles per mile for the alcohol and water slurry releases, and between 10^9 and 10^{10} particles per mile for the dry material. Translating these numbers into snow pellet concentrations in the sub-cloud shower leads to estimates in the order of 1 to 0.01 m^{-3}. We see immediately why the phloroglucinol showers were visually thin and harder to detect. Numbers alone are not adequate to explain all of the observations, however. The alcohol solution releases gave the best results in spite of the fact that the computations suggest that they had the lowest seeding rates in particles per mile.

6. Conclusions

On the basis of 12 releases of phloroglucinol into undercooled stratus clouds it appears that this organic material will induce the formation of ice crystals. The

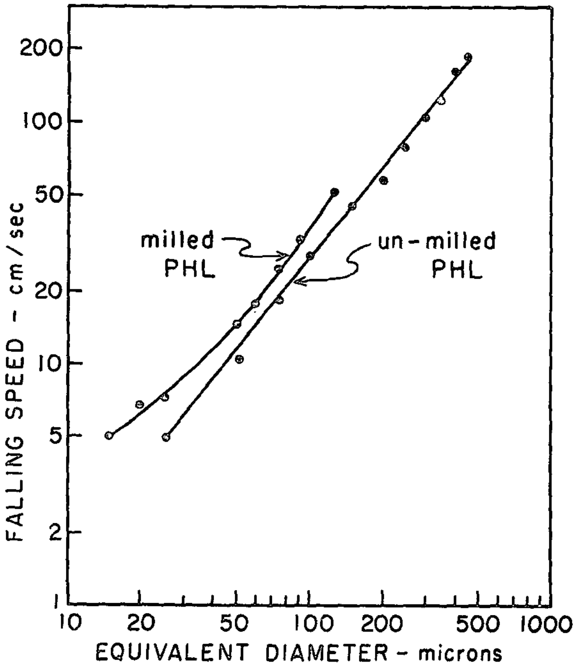

FIG. 3. Falling speeds of phloroglucinol particles.

methods of dispersal, in order of effectiveness, appear to be 1) alcohol solution, 2) dry milled powder, and 3) dry unmilled powder. A single water slurry release was ineffective. The nucleation time is much slower than suggested by earlier laboratory studies probably because of a difference in test conditions in the natural clouds as compared with clouds in laboratory cold boxes. The delayed action of the material in natural clouds is probably associated with the time required for the material to pick up water vapor, or alternatively to collide with a cloud droplet. Calculations of the number of particles released suggest that in the case of all four alcohol solution releases practically every droplet resulted in an ice crystal. The same probably also applies to three of seven dry-powder releases. The other four of seven dry-powder releases appeared to be complete failures.

The fact that an organic material, of any kind, has been found capable of initiating ice formation in field testing is very important. In the zest for developing an understanding of nucleation by inorganic materials, cloud physicists have overlooked what may be a rich field in the organic compounds.

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