Fog Modification with Long-Chain Alcohols

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ABSTRACT

The growth of water drops on condensation nuclei in a supersaturated environment can be greatly retarded by the presence of long-chain alcohols which form monomolecular layers on the liquid surface. The possibility that a fog could be significantly modified using this principle is examined quantitatively, and it is shown that by dispersing practicable amounts of long-chain alcohols it might be possible to produce useful changes of visibility in volumes of the order of 1 km^3 .

Field trials are then described in which about 200 kg of a hexadecanol-octadecanol mixture was released as a finely divided smoke into a valley where fog was expected. In each case, the results were consistent with a modification of the fog throughout a larger volume than expected, but it cannot be claimed that this was not due to natural causes. Assessment of the value of the method is likely to be as difficult as with other forms of cloud seeding.

1. Introduction

Justo (1964) has suggested that long-chain alcohols could be used to prevent fog by inhibiting the growth of a large proportion of the droplets. This would allow the remainder to grow larger, increasing their collection efficiencies and sweeping action on the fog. Concentration of the liquid water into fewer drops would also improve visibility. The growth-inhibiting property is due to the formation of a compressed monolayer of similarly oriented molecules which reduces the rate at which water molecules enter or leave the droplet surface.

In laboratory studies he showed that a mixture of hexadecanol, $\text{CH}_3(\text{CH}_2)_{15}\text{OH}$, and octadecanol, $\text{CH}_3(\text{CH}_2)_{17}\text{OH}$, reduced the rate of growth of water drops by a factor of 100 to 10,000. He also found that the dissolution of sodium chloride crystals in a rising humidity could be retarded by the presence of these alcohols, an observation which has been confirmed by Pilié (1966).

The hypothesis that we adopt is that long-chain alcohols dispersed in particulate form evaporate to give a saturated environment. Under the influence of falling temperature a supersaturation is then produced sufficient to cause re-condensation of the alcohols on the nuclei active in water condensation. We suppose that the resulting inhibition of growth of many of the potential fog droplets leads to more rapid growth of the remainder, contributing to the instability of the fog. First the amounts of material necessary, and the times for evaporation or condensation, are shown to be reasonable, and then some field experiments are described.

2. Design considerations

a. Experimental volume

In any form of practical fog modification a fundamental limitation is the quantity of seeding material required to produce the desired effect. Probably the greatest interest lies in clearing fog from airport runways, which suggests an experimental volume of the order of 1 km^3 . It would be unnecessary and undesirable, even if it were possible, to prevent all drop formation in this volume. Thus, 1 km^3 , completely treated, is to be regarded as an upper limit of what may be required.

b. Mass necessary to form monolayers

The first problem is how much of the material is required to form a compressed monolayer round the fog drops or on the condensation nuclei on which they form. Surface concentrations for hexadecanol at an equilibrium pressure of 40 dyn cm^{-1} on a plane surface have been given by Mansfield (1959) to be $2 \cdot 10^{-7} \text{ gm cm}^{-2}$. Approximate amounts required at various stages in cloud formation for a 1-km^3 volume, assuming each droplet or nucleus to have the most suitable amount and assuming 1000 cm^{-3} active condensation nuclei, are given in Table 1. The figures given take no account

TABLE 1. Amount of hexadecanol (kg) and diameter of equivalent particle required to form a compressed monolayer on various size droplets in a 1 km^3 fog having 1000 cm^{-3} active condensation nuclei.

Droplet diameter (μ)	0.2	2	20
Total amount of hexadecanol	0.25	25	2500
Diameter of each equivalent hexadecanol particle (μ)	0.1	0.4	2

of the curvature of the droplets, which will alter the surface concentration required for a given surface pressure.

Clearly, it will be more economical to seed before the fog forms or in its early stages of formation.

c. Quantity required to saturate 1 km³

Unless the experimental volume is saturated with alcohol vapor, the alcohol particles required for inhibiting fog drop formation will evaporate. It is therefore essential to consider how much mass is involved in producing saturation vapor pressure.

Since hexadecanol has a considerably greater vapor pressure than octadecanol at the temperatures of interest, say 0–15°C, it is necessary to consider only the former.

The constants A and B for hexadecanol in the equation

$$\log P = \frac{-A}{T} + B, \quad (1)$$

where P is the saturation vapor pressure (mm Hg) and T the temperature (°K), have been determined by Sherwood and Johannes (1962); a different pair is given in Landolt-Börnstein (1960). The two pairs of resulting values of P at 0 and 15°C, were then used to estimate corresponding concentrations of hexadecanol needed to produce saturation vapor pressure in a 1 km³ volume; the results are given in Table 2.

TABLE 2. Hexadecanol concentrations (kg) needed to produce saturation vapor pressure in a 1 km³ volume.

	Sherwood and Johannes	Landolt- Börnstein
15°C	30	70
0°C	14	7

Thus, the amounts required to saturate 1 km³ are not prohibitively large, the main problem being how to disperse the alcohol through the volume. Since there is a large rate of decrease of vapor pressure with decreasing temperature, cooling of the air can produce supersaturation and may cause the larger particles in the atmosphere to become coated with the alcohol.

d. Time taken to produce coating in a slightly supersaturated vapor

While it may be possible to deposit sufficient alcohol from the vapor to form a monolayer on all the nuclei or water drops present, it must be shown that the time taken for small particles of the alcohol to evaporate under subsaturation conditions, or to form under conditions of supersaturation, is not prohibitively long.

Fuchs (1959) has shown that for very small particles the rate of evaporation per unit area at atmospheric pressure is very nearly equal to the rate in a vacuum.

For this case, the rate of decrease of mass, dm/dt , per unit area has been stated by Sherwood and Johannes to be

$$\frac{dm}{dt} = 0.06P_s \left(\frac{M}{T_s} \right)^{\frac{1}{2}}, \quad (2)$$

where M is the molecular weight of hexadecanol, P_s its vapor pressure (mm Hg), and T_s the temperature of the particle's surface (°K). Using their values of P_s we find

$$\left. \begin{aligned} \left(\frac{dm}{dt} \right)_{0^\circ\text{C}} &= 5.6 \times 10^{-8} \text{ gm sec}^{-1} \text{ cm}^{-2} \\ \left(\frac{dm}{dt} \right)_{15^\circ\text{C}} &= 1.2 \times 10^{-7} \text{ gm sec}^{-1} \text{ cm}^{-2} \end{aligned} \right\} \quad (3)$$

Thus, particles of diameter 0.6 μ evaporate in times of the order of 10 min at 0°C. One of the most common condensation nuclei in the air is sodium chloride, which has often been used successfully as a nucleus for the formation of monodisperse aerosols of substances resembling hexadecanol. It is therefore reasonable to suppose that the supersaturated vapor of hexadecanol will condense on such particles. Since such a small amount of hexadecanol is required on each to form a compressed monolayer, the time taken should be small compared with 10 min, provided the condensation nucleus is large enough.

The times required for this process of monolayer formation therefore seem quite reasonable.

e. Prevention of fog evaporation

From the point of view of fog dispersal it could be disastrous if the hexadecanol aerosol smoke concentration was such that when it invaded part of the fog which had already formed, the droplets were prevented from evaporating. From Table 1 we see that a 20 μ diameter fog drop will require the equivalent of a 2 μ diameter hexadecanol particle to retard its evaporation. Capture by droplets of this mass of hexadecanol is very slow by diffusion and direct capture of the submicron particles due to differential fall velocities is very improbable unless the drops or particles carry electrical charges or the concentration of hexadecanol is extremely high.

It therefore seems unlikely that the presence of the hexadecanol could help to preserve the fog.

f. Quantities released by cooling a saturated vapor

It therefore seems that the most suitable way of using the growth-inhibiting property of the higher alcohols for fog modification would be to dispense as large a volume of smoke of as small particle size as possible and allow diffusion to distribute it through a large volume well before a fog begins to form. It is then necessary to have a temperature fall, presumably through radiative

cooling, sufficient to produce the supersaturation needed to coat the natural aerosol particles with the alcohol, in order to reduce their ability to act as condensation nuclei.

On the assumption that the alcohols will tend to condense only on the condensation nuclei if fog has not already formed, the amounts required to inhibit growth to a particular size can be estimated from Table 1. For example, to inhibit growth to diameters of $1\ \mu$, $5\ \text{kg km}^{-3}$ must be released. This could be achieved by a temperature fall of the order of 3°C from 10°C , using a compromise between the values in Table 2. These amounts must be increased by a factor depending on how many other particles of the natural aerosol are actually involved in the condensation. For temperatures $<0^\circ\text{C}$, there would be a strong case for using tetradecanol, since sufficient amounts of it to inhibit growth would be released by relatively slight cooling.

g. Effects of retarding growth of all nuclei

Will indiscriminate coating of all the condensation nuclei with alcohol lead to the desired effect?

In an untreated aerosol, cloud formation is preceded by a swelling of the hygroscopic condensation nuclei as the humidity rises. The largest nuclei are the first to pass the critical supersaturation beyond which unlimited growth can occur, and when sufficient of them have passed it, they deplete the available water vapor to the extent where the remaining nuclei can grow no further.

If all the swelling condensation nuclei are covered with monolayers of alcohol, retarding their growth, much greater supersaturation should occur. The growth of the first droplets to break their monolayers should therefore be spectacular, allowing them to deplete the humidity rapidly and slow the growth of their still retarded neighbors even further. The droplet sizes should therefore be larger and their concentrations lower than in the untreated atmosphere.

3. Experimental considerations

a. The material used for fog modification

The requirements listed above suggest that octadecanol or high alcohols may be useful because large volumes of air can be saturated with their vapor without too great a mass of it being needed, while hexadecanol or lower alcohols are desirable because of the greater amounts available in saturated air for coating particles when the temperature falls.

The material actually used was a compromise, with the following composition:

$\text{C}_{20}\text{H}_{41}\text{OH}$	(eicosanol)	3%
$\text{C}_{18}\text{H}_{37}\text{OH}$	(octadecanol)	48%
$\text{C}_{16}\text{H}_{33}\text{OH}$	(hexadecanol)	42%
$\text{C}_{14}\text{H}_{29}\text{OH}$	(tetradecanol)	4%
	hydrocarbons	1.5%
	iodine	0.9%
	esters	0.4%

FIG. 1. The smoke generator used in fog modification experiments.

Its melting point was about 50°C and its boiling point about 350°C .

a. The smoke generator

In our first smoke generator, the liquid alcohol mixture was allowed to run down the walls of a vertical tube against a stream of hot air. Evaporation only became rapid above the boiling point, at which temperature the alcohol ignited easily. It was used in only one operation in fogs in July 1966 and was then discarded because of its erratic operation. A more efficient design was then used in which a spray of fine drops of the liquid alcohol mixture was injected into a vertical wind tunnel, which carried $2\ \text{m}^3\ \text{sec}^{-1}$ of air heated to 350°C by a propane burner. This produced complete vaporization of the alcohol which recondensed as a blue cloud of submicron particles as the air emerged from the tunnel and cooled. The input of alcohol was about $25\ \text{kg hr}^{-1}$ under typical conditions. The buoyancy of the hot air produced very vigorous mixing. Fig. 1 shows it in a trial operation.

b. The experimental site

Within 100 km of Sydney there is a group of connecting valleys which seemed to be ideal for experiments on fog modification. They are virtually uninhabited, about 500 m deep, steep-sided, narrow and long, with a deep lake (Sydney's largest reservoir) at the bottom. It is a

fact observed by anyone who often flies southwest from Sydney that they are frequently completely filled with fog.

Access to the valley is restricted in the interests of keeping the water pure, but its guardians (the Metropolitan Water Sewerage and Drainage Board) were most cooperative in allowing us to enter and to use their facilities.

Although the junction of the valleys (shown in Fig. 2) was selected for the experiment primarily because of accessibility, local information suggested that the densest fog was often found nearby. A convenient overlook (at L in Fig. 2) provided good visibility down three arms of the valley, and cameras were located there to study the formation of the fog during the night.

At sunrise a light aircraft flew overhead and photographs were taken from it of the cloud cover in the whole length of the valley.

The smoke generator was located at one of the locations 1, 2, 3 or 4 of Fig. 2 in different experiments, and measurements were made from one other of these places.

FIG. 2. Topographic map of experimental site and generator site locations.

FIG. 3. Illuminated balloon ascending from near the smoke generator at 2200, 19 June 1967.

c. Air motions

Air circulations were studied by releasing low-lift 30-gm hydrogen-filled balloons carrying a string of "sparklers" (popular fireworks for holding in the hand). These burned slowly, uniformly and brightly for about 15 min. Two-minute time-markers were added by tying pieces of magnesium ribbon at appropriate distances along the chain of sparklers. Examples of the type of record obtained from cameras at L with open shutters during the balloon ascent can be seen in Figs. 3, 6a and 6b.

d. Measurements

Qualitative measurements of condensation nuclei were made using a diffusion chamber or an expansion device. The alcohol smoke contains a certain number of active ice nuclei, probably because of its free iodine, so that ice nucleus concentrations were also measured to try to determine whether smoke from the burner was reaching the observing site. To make this more definite, on two occasions silver iodide nuclei were released near the smoke generator.

Droplet sizes were measured by exposing papers treated with "water blue" dye, the size of the stains as a function of drop size being calibrated in the laboratory. When the fog did not reach the ground, the filters were raised on a tethered balloon.

Another useful device, on the one occasion when large droplets were present, was the acoustic counter of Langer (1965). Air was drawn through a specially shaped tube at a known rate and drops in the air whose diameters exceeded a certain critical value made an audible pop as they went through it. This was tape-recorded at high speed, and played back at low speed, allowing a good estimate of concentration to be made.

FIG. 4. Fog conditions at sunrise on 20 June 1967: A, near the junction of the three valleys; B, the southeast valley; C, the southwest valley; D, the north valley. The arrow shows the direction toward the smoke generator.

4. Results

One satisfactory night's operation was obtained in the winter of 1966 and three in 1967 with the improved smoke generator. On each occasion in 1967 about 200 kg of alcohol was used over a period of about 8 hr, commencing when the temperature fell to within 0.5°C of the dew point and continuing until sunrise.

a. Summary of individual experiments

1) 31 July to 1 August 1966: smoke generator at 1 in Fig. 2, observers at 2, inspection from L at 0400 and 0600; smoke released from 2200 to 0600 at 5 kg hr⁻¹.

Fog started to form along the valley walls at 0100 and thickened quite rapidly, the minimum temperature being 6°C. At 0400 it appeared to be continuous in all directions except at the junction of the valleys, where the burner was located. At 0500 a local deck of stratocumulus suddenly formed over the region and the fog failed to develop further. By the time the aircraft arrived at 0700 to photograph the valley, there was an extensive clear area near the junction and the fog elsewhere was thinning.

2) 19–20 June 1967: burner at 2 in Fig. 2, observers at 1, photography from L; smoke released from 1930 to 0630 at average rate of 25 kg hr⁻¹.

Patches of fog commenced to form about 2100, dissipating and reforming until about 2300, when the

cover became almost continuous in all three parts of the valleys. The temperature was then 9°C. Near the burner the fog did not come closer to the water than about 100 m, but at point 1 it reached the water level at intervals between 0100 and 0200. Its top was at about 500 m, so that its thickness was 400 m. At 0100 a steady rain of small drops started to fall at 1 and continued for several hours, diminishing at 0400 and ceasing at 0500. There was a progressive thinning and lifting of the fog after 0400, and by 0600 patches of clear sky could be seen from the burner with virga beneath the cloud patches.

Drop sizes in the precipitation reached about 0.2 mm diameter and their concentration was quite uniform at about 10 liter⁻¹.

Ice nucleus and condensation nucleus concentrations both diminished considerably throughout the night, but it is uncertain whether this was due to the smoke. They commenced to increase again at sunrise.

The air circulation was obviously complex. The smoke drifted toward the north from the burner, while the fog (initially at about 300 m above the water) advanced toward the south. As the fog thickened, the balloons only revealed that the net drift below cloud top was zero, emerging almost directly over the burner. An example is shown in Fig. 3. Above the walls of the valley a light south breeze was blowing. This double reversal of air motions makes very difficult the task of

deciding in which direction an effect of the smoke is to be sought. Fig. 4 shows the fog as it was seen from the air between 0700 and 0730. The photograph C shows the almost cloud-free southwest valley (see Fig. 2); in A we are near the junction looking toward the southeast fog-filled valley, a closer view of which is shown in B. The dense continuous fog in the north arm and the clear junction area are seen in D. The direction in

FIG. 5. Fog conditions on 7 July 1967: A, near the junction of the three valleys, looking northeast; B, the southeast valley; C, the smoke generator (bright spot of light, lower center) and fog at 0600.

which the smoke generator was located is shown with an arrow on each photograph.

3) 6-7 July 1967: smoke generator at 3 in Fig. 2, observers at 1, photography from L; smoke released from 2050 to 0645 at average rate of 23 kg hr^{-1} .

The first patches of fog appeared at 2320 over the lake and did not become continuous in the junction area at all. Consequently, no precipitation was observed or drop sizes measured. Air motions appeared to be mainly toward the junction from the smoke generator, fog patches drifting northward along the west wall of the north arm and southward around the junction and to the northeast of the east bank of the southwest arm. Silver iodide released from 3 was detected soon afterward at 1 at 2200, but subsequent releases were not detected, and ice nucleus and condensation nucleus concentrations fell steadily during the night as on the previous occasions. The temperature fell to 3C at 1.

In spite of the clear skies at the junction, continuous fog formed in the north arm (Fig. 5a) and southeast arm (Fig. 5b). At 0600 fog reached the water in places in the southwest arm, beyond the smoke generator, which is the bright spot surrounded by smoke on the left bank in Fig. 5c.

Cloud in the southwest arm had diminished considerably by the time aerial photographs such as Fig. 5a were taken at 0730, but did not disappear from the other valleys until after 1000.

4) 3-4 August 1967: smoke generator at 4, observers at 1 and 5, photography from a tower above L, smoke released from 2150 to 0710 at rate of 23 kg hr^{-1} .

The smoke generator was relocated in the southeast arm for this experiment, since dense fog had appeared there on each of the preceding occasions.

The fog was late in forming because of a layer of stratocumulus which persisted above the site until after midnight. Temperatures then fell quickly by about 5C , to 6C at point L and -1C at point 5 (see Fig. 2), accompanied by a very rapid formation of fog.

Fig. 6a is a photograph taken near midnight showing the position of the smoke generator, from which a balloon had been released, revealing the wind structure. The bright patch of light on the right marks the position of a coal mine. Taken a few hours later, the photograph of Fig. 6b shows the then uniform sheet of fog in its early stages of formation. Although it appears to extend to the smoke generator, it did not actually come closer to 4 than about 2 km, and the valleys in the direction of 5 remained clear except for isolated wisps of fog on their sides. The extent to which it had thickened by daybreak can be seen in Fig. 6c. Beyond 2 in a northerly direction, the fog was much thicker, as seen in Fig. 6d. The clear stretch of valley in this photograph is the outlet of the complex of valleys and was fog-free in all the experiments. Toward the southwest, beyond 3, the fog was patchy.

The wind structure on this occasion appeared slightly

FIG. 6. Stages in fog formation on 4 August 1967: A, B, wind profiles shown by illuminated balloons released from near generator, at 0000 and 0300; C, same scene at 0600; D, fog in the north arm at sunrise.

simpler than on the previous occasions. From the generator the smoke travelled eastward up a valley just off the map on Fig. 2, and was presumably returned at a higher level by the slightly stronger easterly breeze which can be seen to exist from Figs. 6a and 6b. It would then possibly have travelled up to 3 and westward along the southwest arm.

b. Comparison with days on which no smoke was released

Mr. P. Cross, who arrived in the valley each day at dawn, mapped the extent of the fog each morning for a month, starting in July 1967. In Table 3 only the number of mornings (control days) on which continuous fog formed in at least one arm (and therefore comparable to our experimental days) are listed together with the type of fog (continuous, patchy or none) found in each part of the valley during these days. These eight control days were 14, 20, 21, 24, 29 and 31 July and 4 and 15 August. Comparable entries are also given for the four experimental days.

The sample is a small one, but either we have achieved clearing around the burner or we have been lucky. To find no occasions in four trials when fog was continuous near the burner, and only one out of eight of the control days when this would have been true, suggests that the smoke was effective in dispersing the fog.

Nevertheless, more doubt than the statistics would suggest to be reasonable remains in the minds of those

who were present, because: 1) the drift of the air on at least two occasions appeared to be in the wrong direction for the clearing observed; 2) the entire southwest arm, a distance of over 20 km with a width of 1–3 km, cleared to a large extent on three of the four occasions which seems rather a lot to expect from the burner's output of about 25 kg hr⁻¹; and 3) the one control day on which fog was patchy at the junction was photo-

TABLE 3. Number of mornings (control days) on which continuous fog formed in at least one arm of the valley during a one month period starting in July 1967. See text for further explanation.

Fog	Part of valley			
	Southeast	Southwest	North	Junction
Control days				
Continuous	7	6	8	7
Patchy	1	1	0	1
None	0	1	0	0
Experimental (generator near junction)				
Continuous	3	1	3	0
Patchy	0	1	0	2
None	0	1	0	1
Experimental (generator in southeast arm)				
Continuous	0	0	1	1
Patchy	1*	1	0	0
None	0	0	0	0

* Continuous to within 2 km of burner, patchy beyond.

graphed from the air, and the result was strongly reminiscent of that shown in Fig. 4a when the clearing was attributed to the fog modification.

The circulation within the valley is undoubtedly complex. When the fog is patchy, thin shafts can sometimes be seen gliding in opposite directions on the opposite walls of the valley and the drift of the smoke near the ground can sometimes be opposite to the apparent motion of the fog. Stratification seems to be intense and the smoke may remain undiluted for long distances, so that it is possible that 1) and 2) are not real objections. However, the impact of observation 3) is strong.

The steady, persistent, light precipitation on 20 June from a cloud only 400 m thick or less after it had formed for only about 2 hr is perhaps the best evidence of the required effect. Some mechanism was at work which produced large drops (for a fog) relatively quickly, in concentrations of about 10 liter^{-1} for 4 hr.

We have found no reports in the literature of droplet diameters in fog as large as the 0.2 mm ones observed on 20 June, diameters <0.1 mm being usually quoted as the maximum. However, each fog is unique in its temperature, mode of formation, liquid water content and duration, and the location of this experiment introduces some unusual features. Perhaps conditions might be more like those in the nocturnal stratus clouds on the windward side of a mountain described by Okita (1962). He reported concentrations of droplets of 0.1 mm diameter or larger which were comparable to those that we have found.

Until we can find enough similar "control" situations the significance of the observation must remain unknown.

5. Conclusions

The results that we have obtained are consistent with our having prevented fog throughout a large volume of air but provide no certain evidence that this would not have happened naturally.

Before starting the experiment we were led to believe that the usual situation was either one of complete fog or no fog. It is now clear that partial fogs can occur, and the problem of determining whether the smoke which we introduced had any effect is entirely analogous to those of other cloud modification experiments. To go further into it would require a much more detailed

study of air circulations, humidities and temperatures as a function of height, and the associated patterns of fog formation with and without the introduction of smoke.

The experiment is relatively expensive, the large quantities of propane being burnt and the alcohols dispersed in one night amounting to about \$200, Australian. Suitable occasions which could be predicted far enough in advance to allow us to assemble the smoke generator, cameras and measuring instruments, and to arrange photography from an aircraft, were also rare. We are presenting our results and methods at this stage, not to demonstrate that we have an effective method of fog prevention, but more in the hope that they may be a useful guide to others with more urgent fog problems and with more frequent opportunities for experiment.

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