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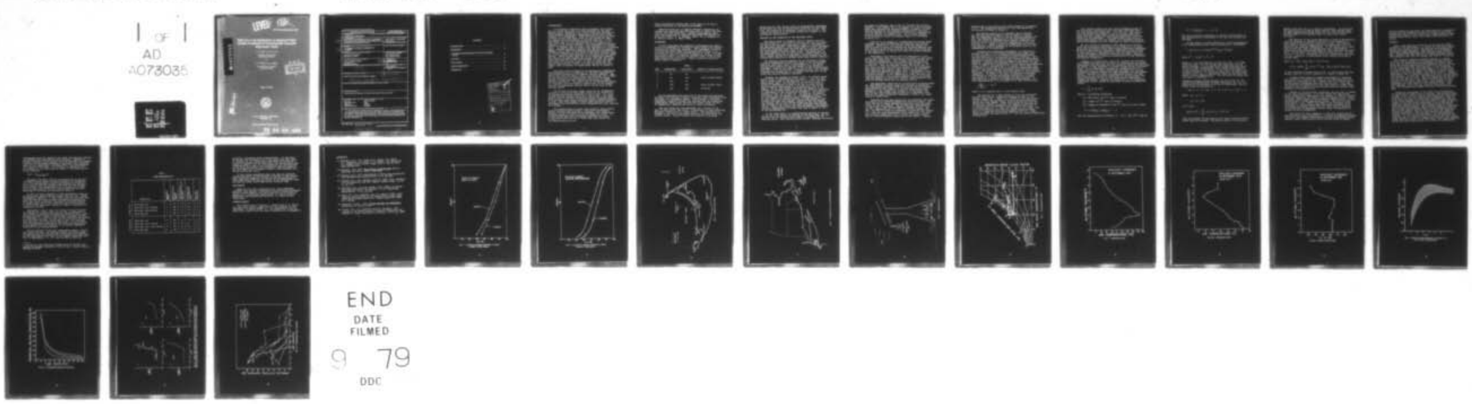

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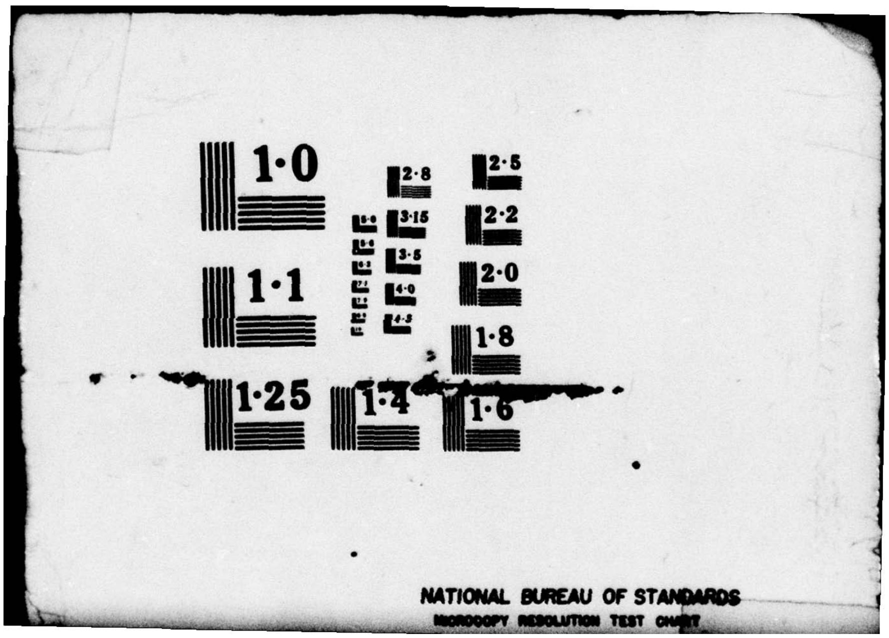
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NRL Memorandum Report 4059

Field Test of the Stabilization of Supersized Water Droplets Condensed on Pyrotechnically Generated Hygroscopic Nuclei

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A field experiment is described which was undertaken to determine if water droplets grown on pyrotechnically generated hygroscopic nuclei at high relative humidities can be stabilized in size when exposed to lower ambient relative humidities. Droplets coated with surface active materials were observed to persist longer than untreated droplets in identical experiments performed off of Nantucket, Massachusetts.

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INTRODUCTION

A pyrotechnic formulation which throughout this report will be called Salty Dog, has been developed by the Naval Weapons Center (Blomerth et al, 1970) which upon burning produces abundant hygroscopic condensation nuclei with a mean diameter of 2 micrometers. These particles provide the basis of water droplet fogs which occur in calm stable air when the ambient relative humidity is greater than 75%. It has been shown that the size distribution of the droplets is a function of the relative humidity and fogs formed at relative humidities greater than 90% have a mean diameter of 8 micrometers. It appears that in order to be considered as a screening agent, relative humidities greater than 80% must be assured. Figures 1 and 2 show the cumulative probability distributions of relative humidity at ship level in the North Atlantic Ocean and in the Eastern Mediterranean for the winter and the summer seasons. These figures show that if an effective screening fog requires relative humidities greater than 90% that these occur less than 20% of the time at least in these two important parts of the world ocean. In fact in areas like the Eastern Mediterranean during the winter months we can only expect these occurances to happen about one out of every 10 days.

Fog water drops residing on the hygroscopic condensation nuclei produced by the combustion of the Salty Dog material are affected strongly by the ambient relative humidity. Generally they obey the Köhler curves (see Fletcher, N.H. (1962)) for salt particles in subsaturated conditions and become smaller at lower relative humidities and larger at higher relative humidities. The actual size of an individual droplet at a specific relative humidity depends both on the composition of the hygroscopic nucleus and on the dry size of the nucleus itself.

The ability to produce large quantities of condensation nuclei in a controlled environment opens up the possibility of engineering desired properties of water fogs. If a population of water droplets which have been grown to the appropriate sizes to contain the desired optical screening properties can have their sizes stabilized against further reduction in size, then large plumes of these water droplets could be introduced into a relatively drier atmosphere and still function as a screen.

The stabilization of super large droplets could provide useful screening applications in the marine environment. Screens of this type would have advantages over conventional military smokes in fulfilling the following two needs: 1) A need for an agent that is neither corrosive nor toxic. 2) The logistical need for a "water fog" type screen for

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Naval applications whereby most of the mass of the fog is readily available in the marine environment.

Materials which act as highly efficient evaporation retardants are available and have been used to reduce the evaporation loss from reservoirs. Several investigators have used these materials to stabilize fog water with various degrees of success. (W. D. Garrett, 1971), (W. C. Kocmond, W. D. Garrett, and E. J. Mack, 1972).

EXPERIMENT

A field experiment was devised to investigate the feasibility of extending the lifetime of super large water droplets stabilized by surfactant material. The experiment consisted of producing finite plumes of droplets and allowing it to blow with the wind over a uniform stretch of ocean. Although the droplets were all formed by condensation of water on the hygroscopic nuclei, one of five distinct types of water fogs were produced for each plume. Table 1 shows the five different types of droplets.

TABLE 1

TYPE	SUPERSIZED	STABILIZED	METHOD OF STABILIZATION
1	no	no	
2	no	yes	cetyl alcohol spray
3	yes	no	
4	yes	yes	cetyl alcohol spray
5	yes	yes	"frostop"

In the experiment the evolution of the plume was monitored by means of an aircraft flying a racetrack course and penetrating the plume in a systematic manner. The aircraft, a Bellanca atmospheric research aircraft was equipped with an integrating nephelometer (Meteorology Research Inc. Model 1550b) as well as temperature, dewpoint and electric field instruments.

The site chosen for the experiment was a narrow (75 m) spit of sand called "The Galls", a projection of land connecting Nantucket Island with its "Great Point". This peninsula is bounded on the west by Nantucket Sound and on the east by the Atlantic Ocean. Figure 3 shows the location of the plume generation site and clearly shows the geographical advantages of the Galls in that no matter which direction the wind might blow, the chances of an overwater plume are very great. Fig. 4 shows the location of Nantucket Island with respect to the continental United States.

ANALYSIS OF THE OPERATION OF THE NANTUCKET STOVE

The device shown in schematic form in figure 5 was constructed directly on the sand of the spit and used to produce any one of the fog types described in Table 1, depending on the experiment desired. This device named by us the "Nantucket Stove" was orientated with the wind vector in such a way that the wind entered the lower inlet of the top pipe while the fog plume emanated from the upper level. The lower part of the Nantucket Stove was in the form of a pyramid which acted like a funnel mixing the condensation nuclei produced by the burning Salty Dog with or without water vapor and/or chemicals.

The measured relative humidity in the vertically rising column of condensation nuclei and vapors is not particularly high even when water vapor is being added to it by rapidly boiling water because of the heat added by the combustion processes. However the dewpoint of this column is much higher than that of the outside air. Consequently when this effluent is mixed with outside air in the upward slanting part of the stack the relative humidity increases.

Consider the diagram in figure 6 where the ambient atmospheric state is represented by the point A. When the Salty Dog is burned alone in the Nantucket Stove without the additional water vapor, the effluent in the vertical stack is raised in temperature to point B but not in water content. This process is represented by the horizontal line in the figure. During this process the absolute humidity of the effluent is the same (lower if the dry gas from the combustion is considered) as the surrounding atmosphere. The relative humidity of this air is very low. As this air is mixed in the mixing tube with ambient air, intermediate points along the line segment AB describes these intermediate states. At some point, the relative humidity is high enough to form droplets and when this happens, radiation from the aerosol and droplets add to the cooling process so that the effluent just outside of the stove is not much different from ambient values. Using this process the relative humidity surrounding the Salty Dog aerosols is always less than or equal to that of the surrounding air.

On the other hand, if a humidification process is carried out in the stove during the burning of the Salty Dog, the temperature and the absolute water vapor is increased in the vertical stack. This humidification process, although increasing the absolute humidity, may or may not increase the relative humidity. Line segment AM_1 is an example of a mixing process which does increase the relative humidity slightly. In fact various mixtures of this effluent represented by state M_1 with the ambient air represented by state A can increase the relative humidity by mixing alone. The relative humidity of the mixture at state E is greater than it is in either state M_1 or A.

Point M_2 is an example of a humidification process which increases the absolute humidity but decreases the relative humidity. Mixtures of this effluent with the ambient air such as shown by point D increases the relative humidity over that which we would obtain from the dry Salty Dog itself but at the same time it is always less than the relative humidity in the ambient air.

Experiments show however that the temperature in the plume, (one meter from the outlet of the stove) is very nearly the ambient temperature. The effluent passing through the ducting of the Nantucket Stove conducts and radiates heat to the walls which in turn conduct and radiate heat to the outside air. Once the effluent reaches the outlet part of the stove, aerosols and particles radiate to the environment. This radiation will cause a rapid cooling of the effluent which does not decrease the vapor load. Consequently a process which proceeds along segment CD is a description of the radiative process. At all temperatures lower than that represented by point F the relative humidity values are greater than the ambient air which will produce larger droplets than would be produced using the dry process line AB.

The experiment consisted of 8 artificially produced plumes. The time history of these plumes was obtained by measuring the scattering coefficient from the aircraft during repeated penetrations of the plume as it drifted downwind from the generator. Four plumes were studied on 21 September 1978 and the last four plumes were studied on 23 September 1978. Each of the types of droplets described in Table 1 are represented in these tests.

Although the two days chosen for the experiment had cloudless skies, they differed dramatically from each other. The first day had a strong temperature inversion starting essentially at the sea surface. The surface wind speed measured 24 knots and it came from a magnetic heading of 243°. The dry air temperature was 20°C and the relative humidity was 88%. The PRT-10 radiation thermometer calibrated with respect to a black box gave measurements which showed that the sand had a surface temperature of 23 C while the water surface was 14 C on the Atlantic Ocean side of the "Galls". Bucket temperatures of the Atlantic surf water showed it to measure 17.5 C. The temperature profile for that day measured by the aircraft is shown in figure 7.

The weather picture on 23 September was as follows: The wind was from 83° magnetic with a speed of 11 knots. The air temperature was 16.7 C and the relative humidity was 69%. The radiation temperature of the sand measured 18 C while the waters in Nantucket Sound showed a radiation temperature of 13 C while its bucket temperature of the surf zone was 18 C. Figure 8 shows the temperature and the relative humidity profile obtained by the aircraft prior to the commencement of the experiment.

Although the integrating nephelometer is sensitive to aerosols in the 0.1 to 2 microns diameter size range there is some question as to its accuracy in measurements where the size spectrum of the measured aerosols is unknown. More specifically we want to know how the integrating nephelometer responds to droplet population changes as the plumes are blown downwind. Obviously the measured values obtained by the aircraft is suppose to be proportional to the scattering coefficient. A paper by R. A. Rabinoff and B. M. Herman (1973) shows that two types of errors can occur in using the integrating nephelometer. They also indicate correction terms for these errors which are a function of the size distribution of the scatterers themselves. In figure 9 the ratio of indicated scattering coefficient to the true scattering coefficient is plotted as a function ν^* where the envelope includes the relative efficiency for a variety of integrating nephelometers. In this representation ν^* is the power of an input distribution:

$$\frac{dN}{d \log r} = Cr^{-\nu^*}$$

where C is a constant and r is the droplet radii.

The question is at what values of ν^* do our artificial aerosols best match? Laboratory experiments done in the Calspan chamber (E. J. Mack et al (1978)) on the size distribution of Salty Dog aerosols at various relative humidities should give this answer. These tests indicate that the conditions most likely to simulate the droplet distribution as it eminates from the Nantucket Stove (RH≈97%) show that ν^* is on the order of 1.5. This also appears to be a worst case type of distribution done with an initial amount of Salty Dog of 0.1 grams and at relative humidities of 97%. Other cases that they ran seem to give $\nu^* \approx 2$ for low relative humidities of about 59%.

This analysis shows then that typical measured values of the scattering coefficient tend to be about 30% lower than the true scattering coefficient. Thus the fact that the aircraft obtained by measurement large scattering coefficient shows that even larger true scattering coefficients were the results of the experiment. Thus measurements with the airborne nephelometer will give answers to a reasonable accuracy and will be adequate for the investigation under consideration.

We can, within an accuracy of 10%, measure with the aircraft the scattering coefficient of the plume as it is being acted upon by natural forces while being transported downstream. We need to know how the scattering coefficient can be related to the stability of the droplets as they are affected by the two main processes tending to dissipate the plume. These processes are reduction in droplet concentration by eddy diffusion and droplet evaporation.

Changes in concentration, while the relative size distribution remains the same will be active in reducing the scattering coefficient from the plume even for perfectly stabilized particles. The effect of eddy diffusion on droplet concentration is modeled below. In our experiments we make the assumption that for a particular day in which observations are made that the eddy diffusions processes remains the same throughout the measurement period. Therefore if there are differences between the time histories of the scattering coefficient of various plumes then these differences are due to size distribution differences between the plumes. Consider the definition of the scattering coefficient from N classes of scatterers as given by Middleton, (1952):

$$b = \sum_{i=1}^{N} N_i K_i \Pi a_i^2$$

where b = scattering coefficient

K_i = Mie factor for i^{th} class of droplet

a_i = radius of i^{th} class of droplet

N_i = number of droplets in the i^{th} class in a unit volume

$$b = \Pi (N_1 K_1 a_1 + N_2 K_2 a_2 + ...)$$

Let the concentration be doubled i.e.: $N_1^{1} = 2N_1$, $Na^1 = 2N_2$ etc.

$$b^1 = 2\Pi(N_1K_1a_1 + ...) = 2b$$

Thus the scattering coefficient is directly proportional to the concentration of scatterers if all the size factors are the same.

We may model the plume diffusion by using the assumption of an instantaneous point source diffusing in 3 dimensions.

$$\chi\ (x,y,z,t) = Q\ (2\Pi\sigma_y{}^2)^{-3/2}\ \exp\ (-r^2/2\sigma_y{}^2)$$

$$\text{where } r^2 = (x-\bar{u}t)^2 + y^2 + z^2$$

where χ is the concentration in space and time, σ_y an eddy diffusion term, $\bar{u}$ is the average wind speed and Q is the source strength. In our case we will eventually sum the effects of N such instantaneous point sources operating over a time interval of one minute. If we look at the effect of one point source at all locations which have the same concentration, ie $X = X_o$, a 3 dimensional sphere is described the radius of which depends on $\sigma_y{}^2 \simeq 2Kt$ but which is moving with the wind at a velocity of $\bar{u}$.

Likewise a second similar equation can be written for the second instantaneous puff of smoke occuring at $t_2 = t_1 + \Delta t$. Since we are now looking at the sum of these puffs, the net effect would be described as:

$$\chi(x,\ y,\ z,\ t) = \chi(x,\ y,\ z,\ t_1) + \chi(x,\ y,\ z,\ t_2) + ...$$

where $t_2 = t_1 + \Delta t$

$$t_3 = t_1 = 2\Delta t$$

at $T = N\Delta t$

$$\chi_T(x,y,z,T) = \sum_{i=1}^{N} \chi(x,y,z,t_1 + (i-1)\ \Delta t)$$

This now becomes the description of a finite duration source which lasts $N\Delta t$ seconds. The problem then is to be able to express this function χ_T in terms of observables. If we plotted all space where $\chi_T > \chi_o$ then we would observe a pear shaped plume. As time progresses the plume moves with the wind and eventually diffuses down to the condition where no point exist which has $\chi_T > \chi_o$.

The behavior of this plume expressed in terms of measureables could be obtained by looking at the plot of the maximum χ/χ_o value found in the plume as a function of time. This type of plot could be directly compared with the experimental plots obtained by the airbourne nephelometer. If the source was in operation for NΔt sec, then the maximum peak would approximately follow the point moving across space on the x axis with the speed of $\overline{u}$. At the time t, the point has an x value of $\overline{u}$ $(t - N\,\Delta t/2)$ for all $t > N\,\Delta t/2$. We can now write the concentration function, χ^1/Q as a series of N instantaneous point sources occurring Δt seconds apart

where $\sigma_y{}^2 = 2Kt$, $y=z=0$ and $x = \overline{u}(t-N\,\Delta t/2)$

$$\chi^1/Q = \text{Const}^* \sum_{i=0}^{N} (t-i\Delta t)^{-3/2} \exp \quad (\overline{u}\Delta t(i-N/2))^2/4K(t-i\Delta t)$$

In this equation we assume values of K, Δt, and $\overline{u}$ and N and then obtain a plot of χ^1/Q as a function of t shown in Figure 10.

A simple BASIC computer program was designed to calculate this function to simulate the decay of the plume from eddy diffusions as a function of time. Several sets of realistic values of the physical parameters were used for the plot. Note that the functions are normalized to give 100% values at t=2min. These curves are very similar to those obtained from the airborne nephelometer and discussed below.

Changes in the scattering coefficient are also due to changes in the size distribution. It is well known that hygroscopic nuclei change sizes dramatically with changes in relative humidity therefore changes in the ambient relative humidity about a cloud of nuclei cause changes in the scattering coefficient for a fixed concentration of these hygroscopic nuclei. Data of this form (see Covert, et al. (1972)) are shown in the following curves of Figure 11. These figures show that large decreases in the scattering coefficient occur when the lower relative humidity of a population of hygroscopic nuclei causes the population to change its size distribution.

The result of these arguments is that both stabilized and unstabilized droplet populations in our Salty Dog generated plumes, will have their number concentration reduced by turbulent mixing. However, super sized droplets which are both grown at higher relative humidities and which are stabilized with a surfactant coating will not evaporate but will have higher scattering coefficients which will be visible for longer periods of time than one would expect from a dry normal sized population of nuclei.

ANALYSIS

Table 2 describes the production of the various plumes produced in the experiments. It should be noted that approximately equal amount of Salty Dog material were prepared for burning for each experiment. The weights of this material had a 10% standard deviation about the mean value of 436.5 gms. Likewise the burn times of all eight experiments were approximately equal with an average burn time of 1.3 minutes.

The external conditions differed markedly in thermal stability and the height of the inversion between the first group and the second group of experiments but within each group the external meteorological parameters were as constant as any field experiment could provide. At the beginning of each experiment, the Salty Dog element (cut in the shape of a piece of cake) was ignited by means of a propane torch heating a corner of the wedge. The timing of the experiment began when a self sustaining burning of the Salty Dog material was achieved. At time zero, the Nantucket Stove was positioned over the burning Salty Dog. When the composition of the fog required the addition of two rapidly boiling pots of water setting on a two burner camp stove. This assembly is also underneath the lower funnel of the Nantucket Stove. When the experiment called for the "Frostop" stabilizer to be used, the substance was dropped into one of the boiling water pots.

Sequential photographs of the burns were obtained with a Polaroid SX-70 camera every 15 seconds. The aircraft being in radio contact with the ground party marked the analog data recorder as to the start time of every experiment. The aircraft then proceeded to periodically penetrate the plume as it was blown downwind over the ocean water in order to monitor changes in the scattering coefficient as the plume evolved under the influence of both eddy diffusion and of droplet size distribution changes. The airplane flying in a racetrack pattern made repeated penetrations flying downwind along the axis of the plume as seen by the pilot until the plume disappeared altogether. Each of these penetrations produced a pulse above the stable background in the nephelometer recording. As the pilot was not always able to penetrate the plume through its center in exactly the same way, the recorded pulses were not always of the same width. The longest duration pulses had pulse widths which at the aircraft flying speed corresponded with the length of the plume calculated by knowing the surface wind speed and the burn duration. However on several occasions it was obvious that the aircraft only skimmed a portion of the plume producing an abnormally short duration pulse on the nephelometer recording which also had abnormally low values at their peak. In an effort to compensate for this variation, the data shown in Figure 12 has been plotted as the function:

$$f(t) = (b_{scat})_{max}/\Delta t$$

Obviously the exact time of the penetration of the last "invisible" pulse cannot be known precisely but estimates as to the time required to make the complete racetrack pattern is known from the visible pulses and thus an estimate of the time of the disappearance of the plume can be made. Both the exact age of the last visible pulse and the estimated age of the plume are recorded in Table 2.

In both of these categories, three experiments stand out as having ages several minutes longer than the rest. These experiments, numbers 4, 9, and 10 are precisely the same fogs in which care was taken to grow the droplets formed on the Salty Dog nuclei in greater than ambient relative humidity to larger sizes and to then stabilize these droplets with a cetyl alcohol monolayer coating in order to slow down the evaporation process.

Experiments 4 and 9 used the boiling "Frostop"* method for coating the droplets where as the fog in experiment 10 was treated by spraying the supersized droplets with a cetyl alcohol-isopropyl alcohol solution from a pressure sprayer. In experiment 5, a similar cetyl alcohol-isopropyl was sprayed on a dry Salty Dog fog. As the age of this case was even less than that of the dry Salty Dog itself, it is possible that the monolayer coated nuclei had their potential growth in the ambient relative humidity of the marine atmosphere inhibited by the coating process.

The two shortest lived fogs (experiments number 3 and 8) were those two fogs, one from each group, which were grown to supersize but not treated with a surfactant for stability. One can hypothesize that some of the largest supersized droplets in experiments 3, 4, 8, 9 and 10 tend to fall out

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* Frostop is a fog stabilizer manufactured exclusively for Applied Technology Corporation, 6361 1st Ave., South, Seattle, Washington 98108.

TABLE 2

PLUME CHARACTERISTICS

Date	Composition	Experiment Number	Amount of Salty Dog used (grams)	Elapsed time to last plume detection (minutes)	Estimated time to background level	Burn time (minutes)
21 Sept 1978	Salty Dog + H_2O	3	441	3.0	5.2	1.5
	Salty Dog + H_2O + Frostop	4	446	8.5	10.5	1.3
	Salty Dog + Cetyl Alcohol	5	342	4.5	6.0	1.4
	Salty Dog (dry)	6	445	7.0	8.5	1.4
23 Sept 1978	Salty Dog + H_2O	8	482	2.4	4.0	1.4
	Salty Dog + H_2O + Frostop	9	427	9.3	12.	1.2
	Salty Dog + H_2O + Cetyl Alcohol	10	436	9.6	11.3	1.6
	Salty Dog (dry)	11	452	7.0	7.5	1.4

initially, decreasing their concentrations. As time goes on however the increased size of the stabilized fogs causes an increase in b_{scat} which more than makes up for the decrease in numbers. On the other hand, the unstabilized fogs evaporate down in time to the equalibrium size distribution of the Salty Dog aerosol for the ambient relative humidity and thus their loss in numbers causes a lower measured scattering coefficient value.

If this latter hypothesis were true then the observed stabilizing effect of the monolayer coatings is even greater than originally suggested in that the net concentration of these fogs should be less than those of the equivalently aged dry fogs yet their scattering ability outlasts them.

CONCLUSIONS

Based on this very limited data set, this experiment indicates that droplets condensed on pyrotechnically generated nuclei at high humidity and stabilized with a monolayer can persist for a measurably longer period of time in a field experiment than unstabilized droplets, all other factors being equal.

ACKNOWLEDGMENTS

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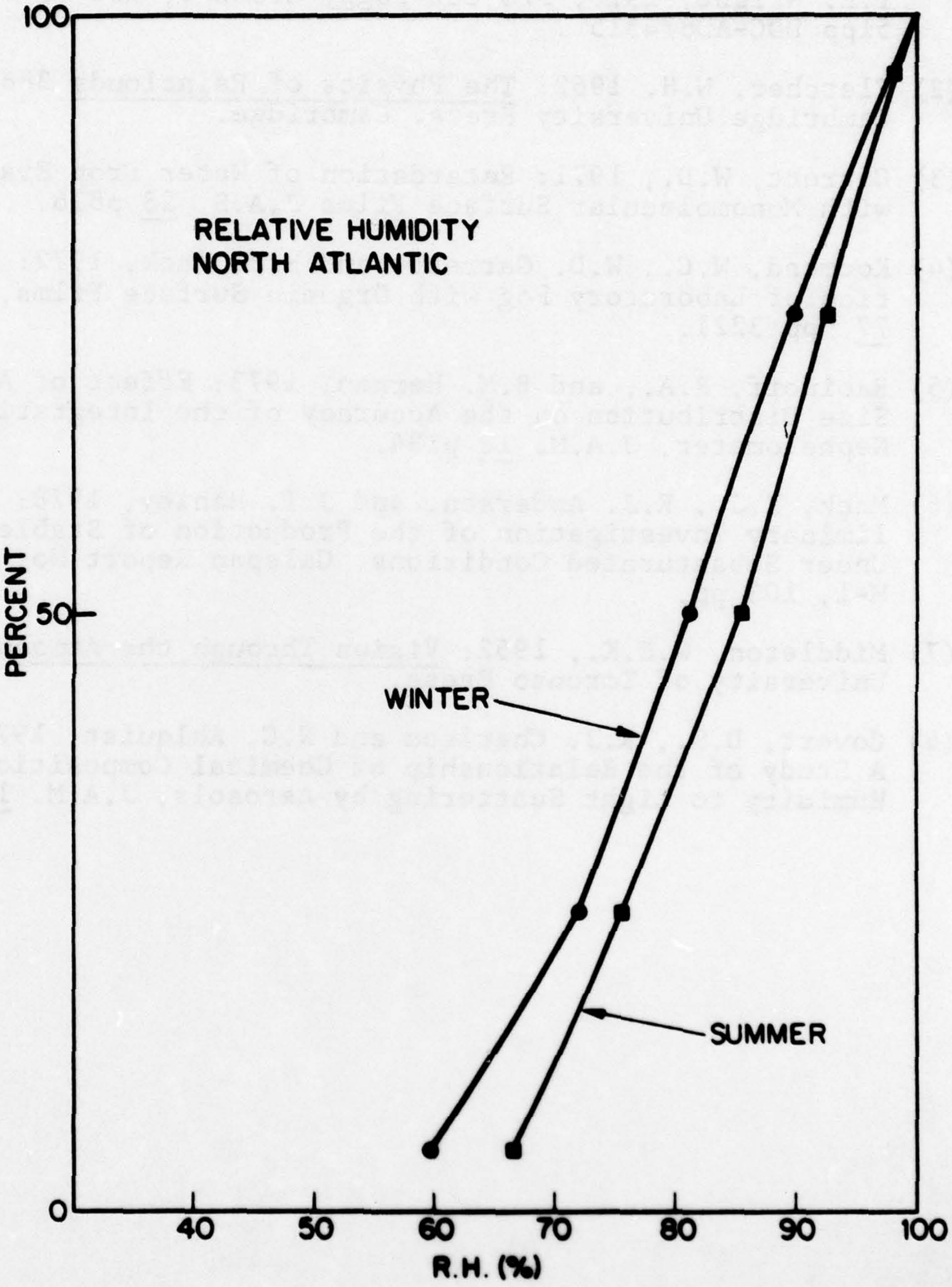

Fig. 1 — Cumulative probability distribution of relative humidity in North Atlantic

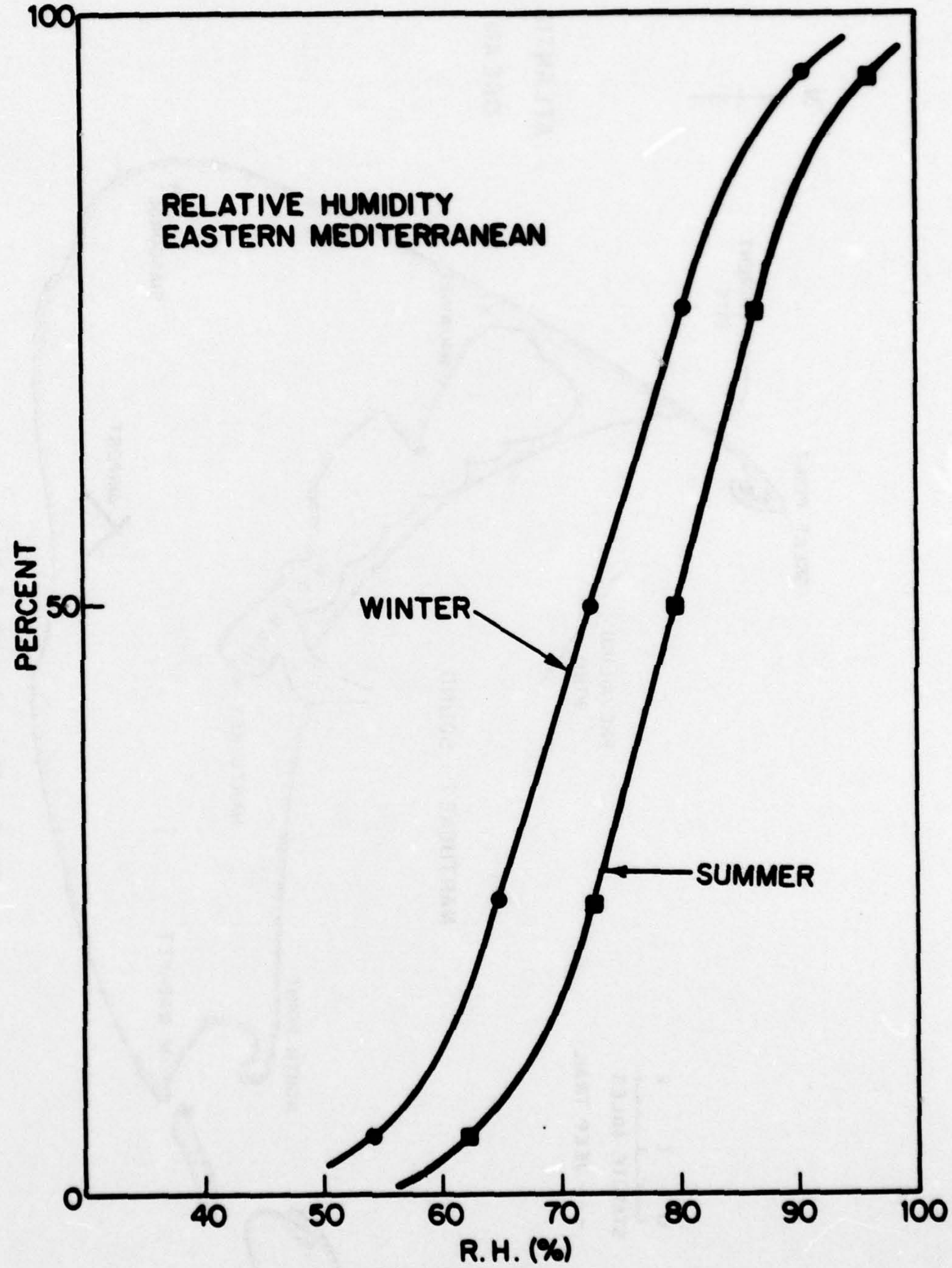

Fig. 2 — Cumulative probability distribution of relative humidity in Mediterranean

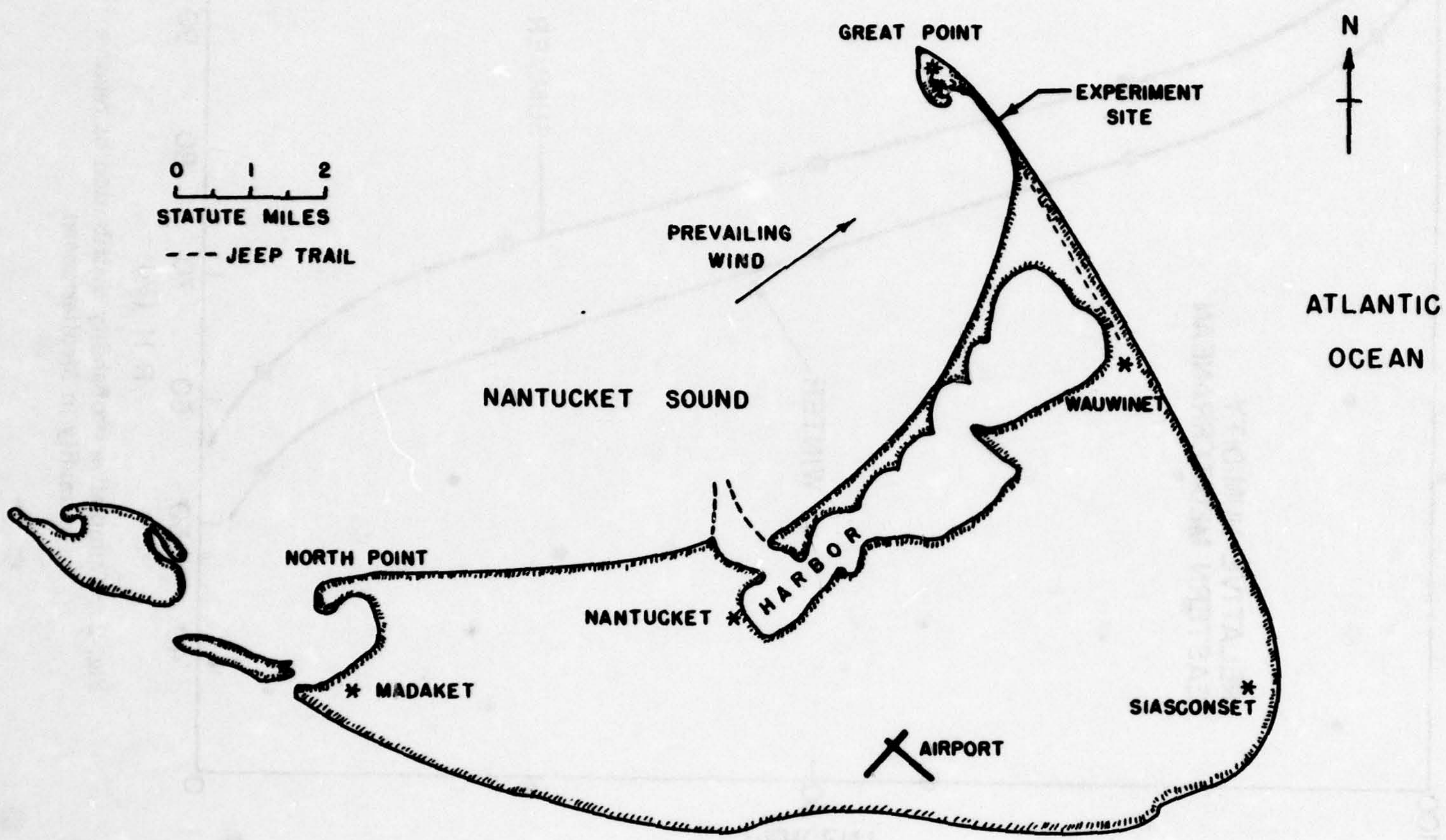

Fig. 3 — Nantucket Island, Mass.

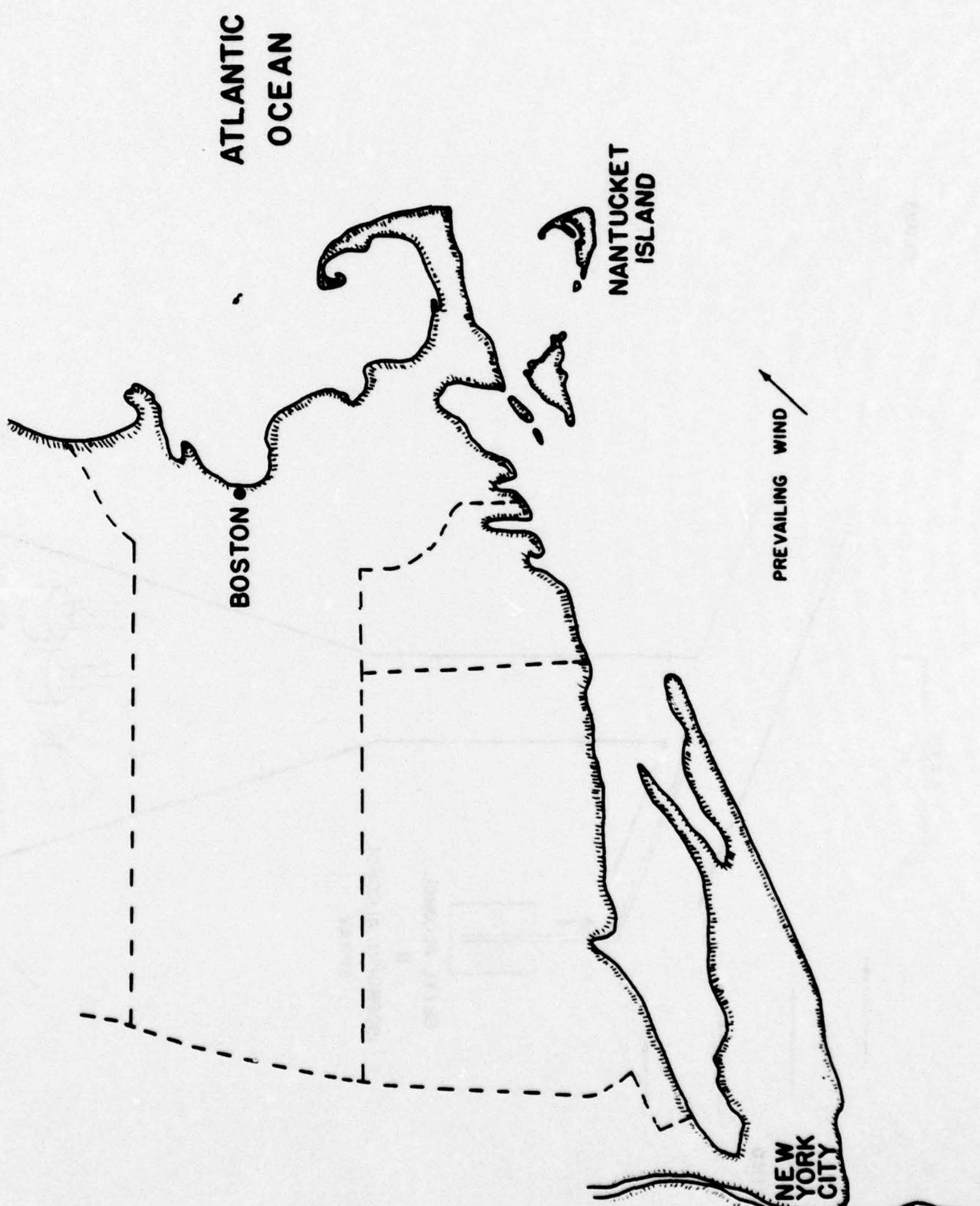

Fig. 4 — Continental U.S.A. near Nantucket Island

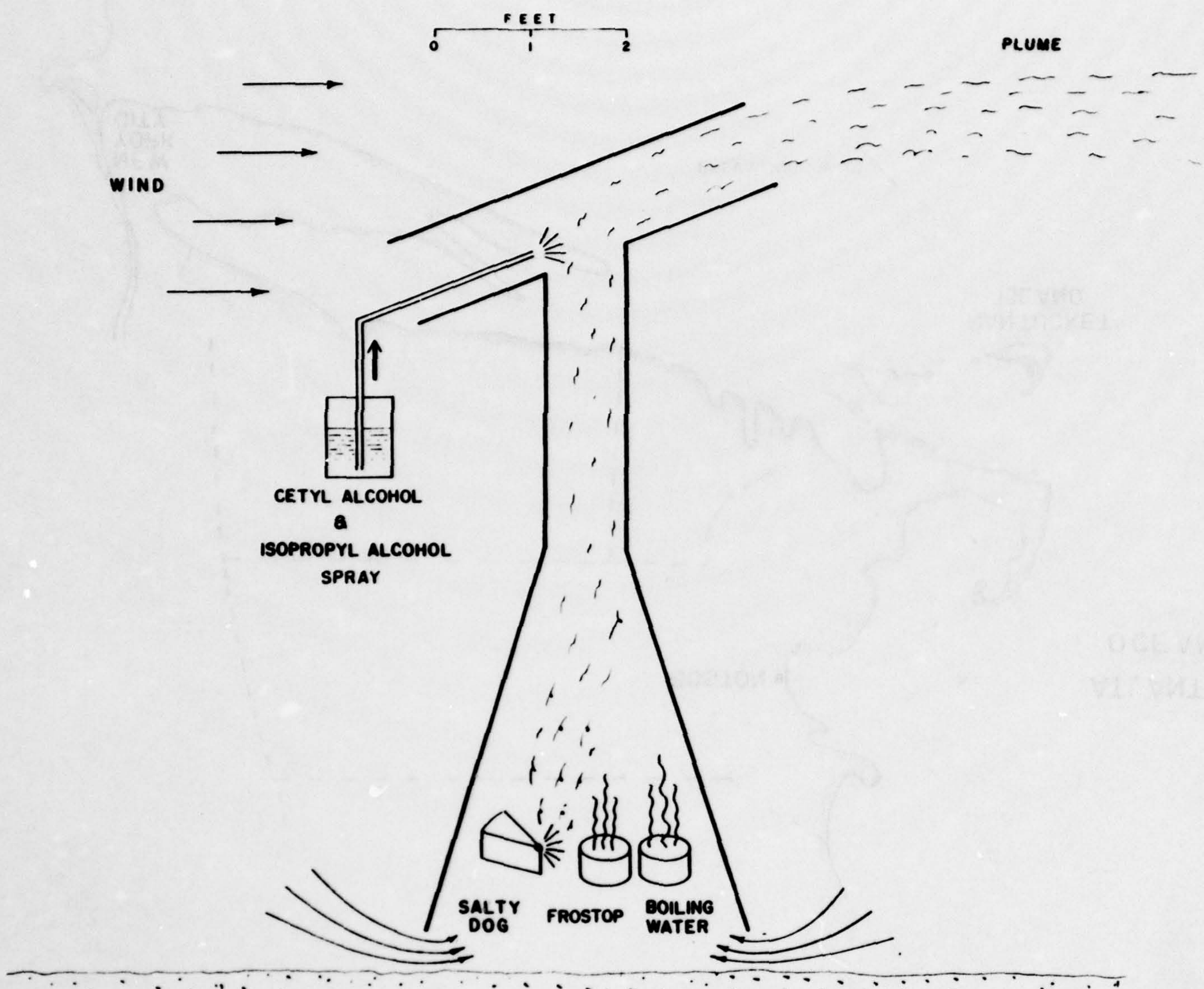

Fig. 5 — "Nantucket Stove"

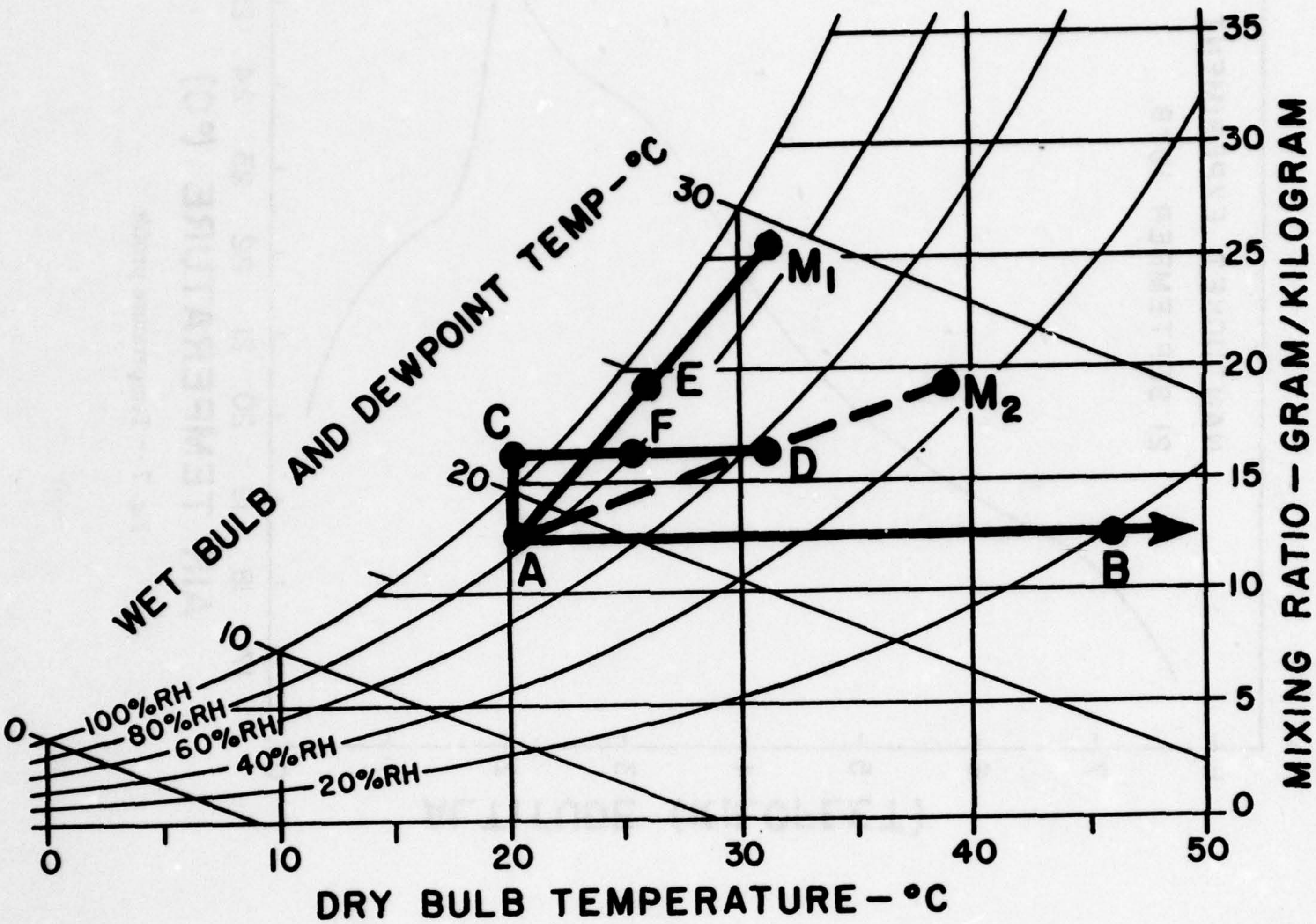

Fig. 6 — Thermodynamic processes

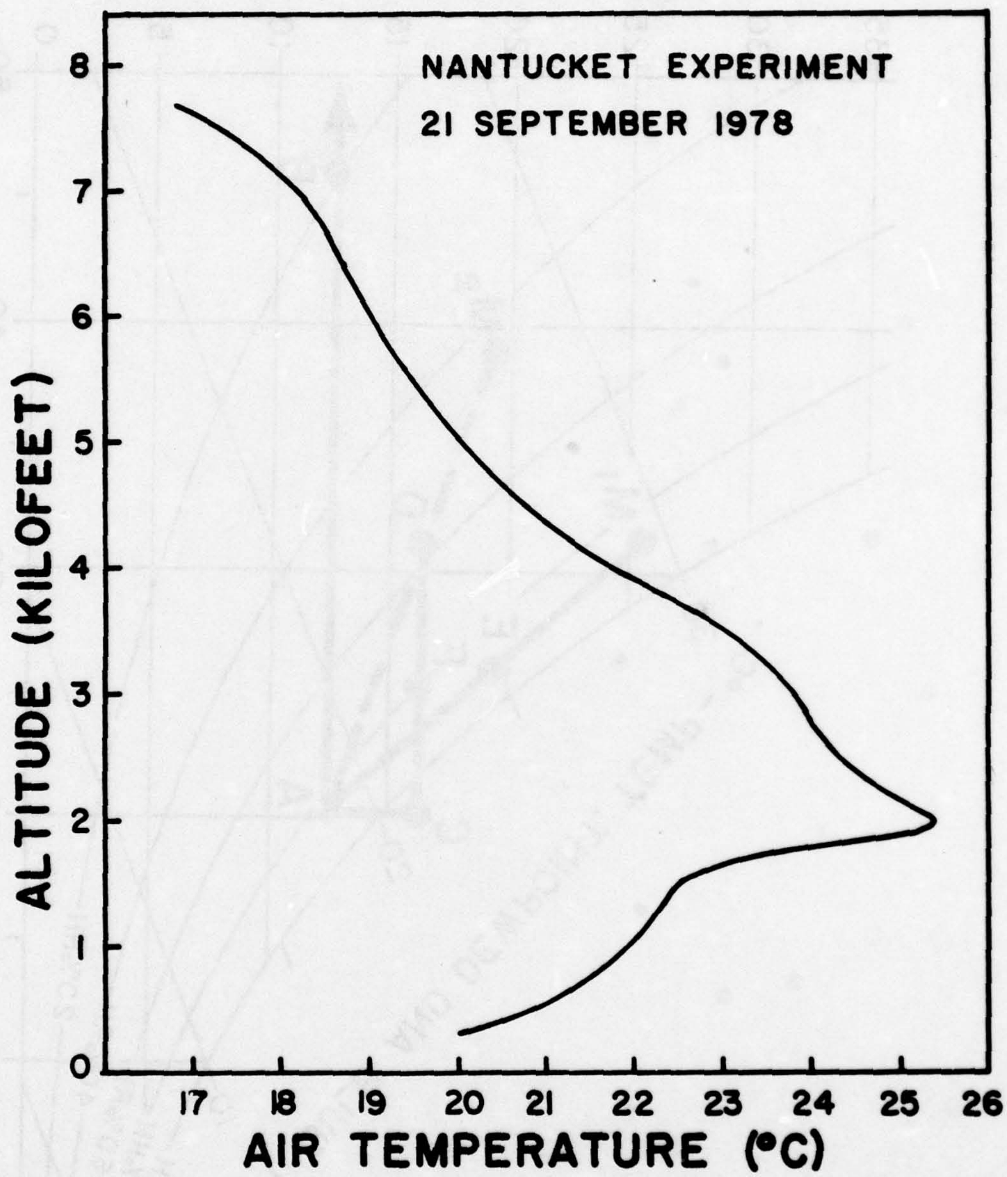

Fig. 7 — Temperature profile

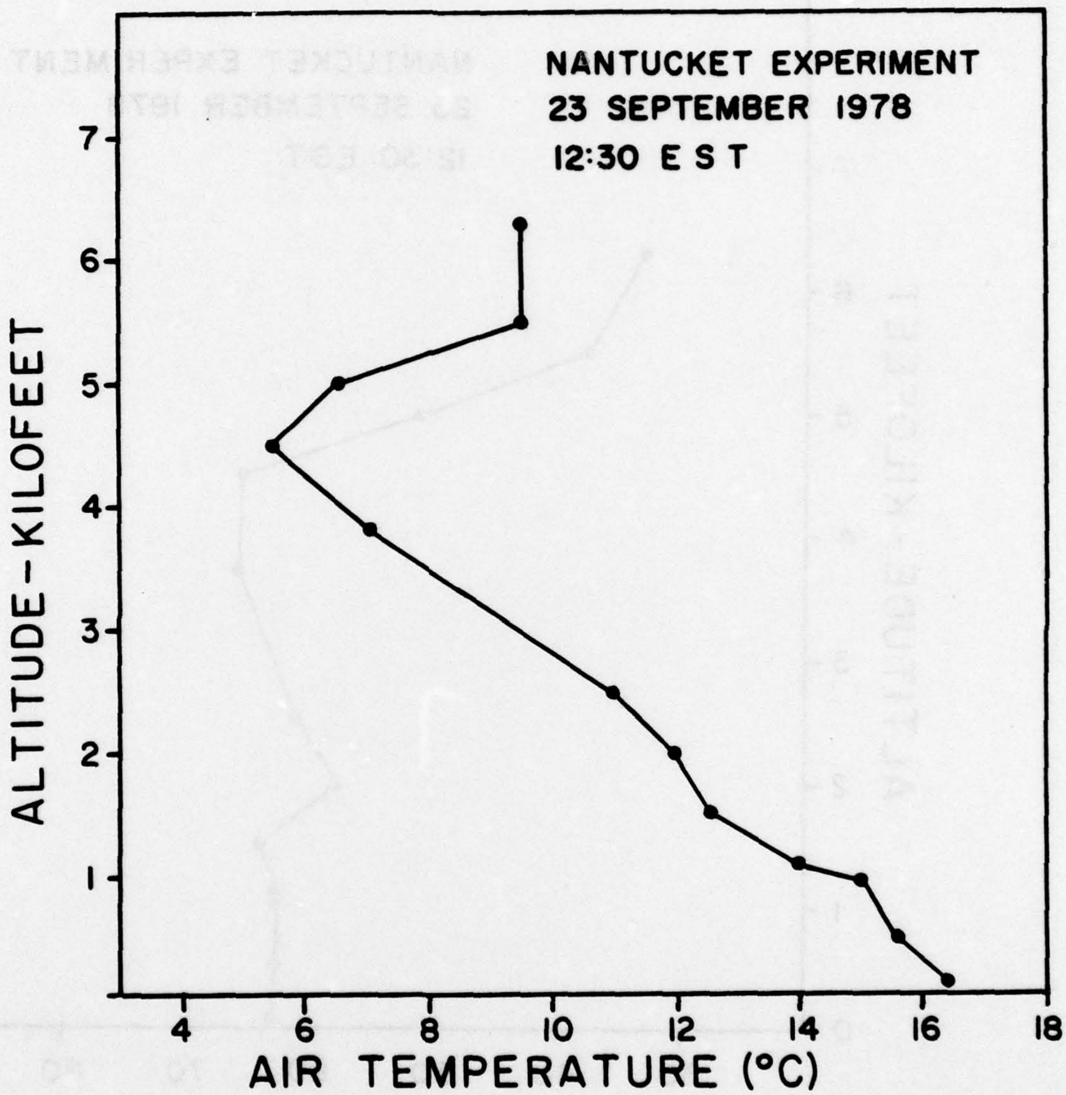

Fig. 8(a) — Temperature profile

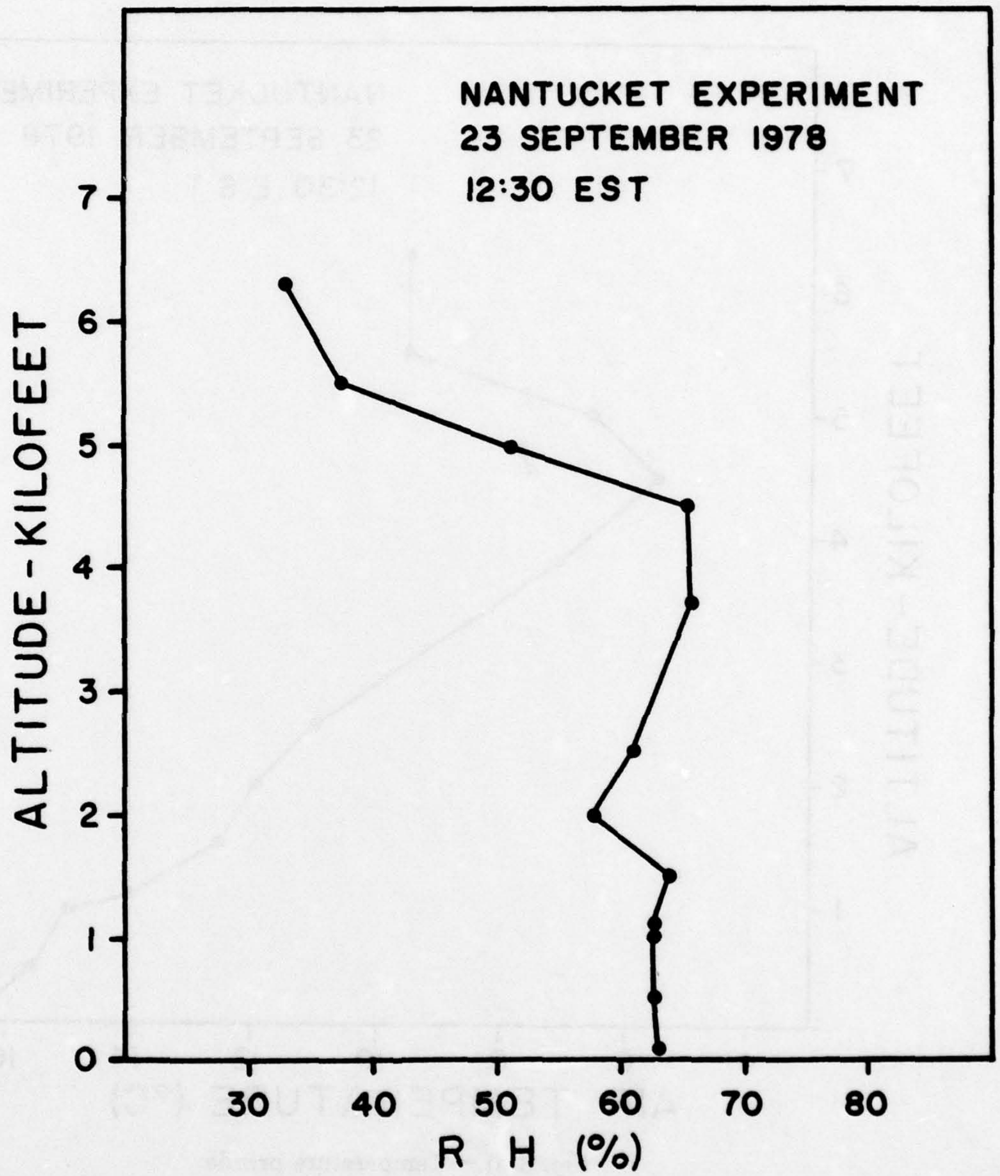

Fig. 8(b) — Relative humidity profile

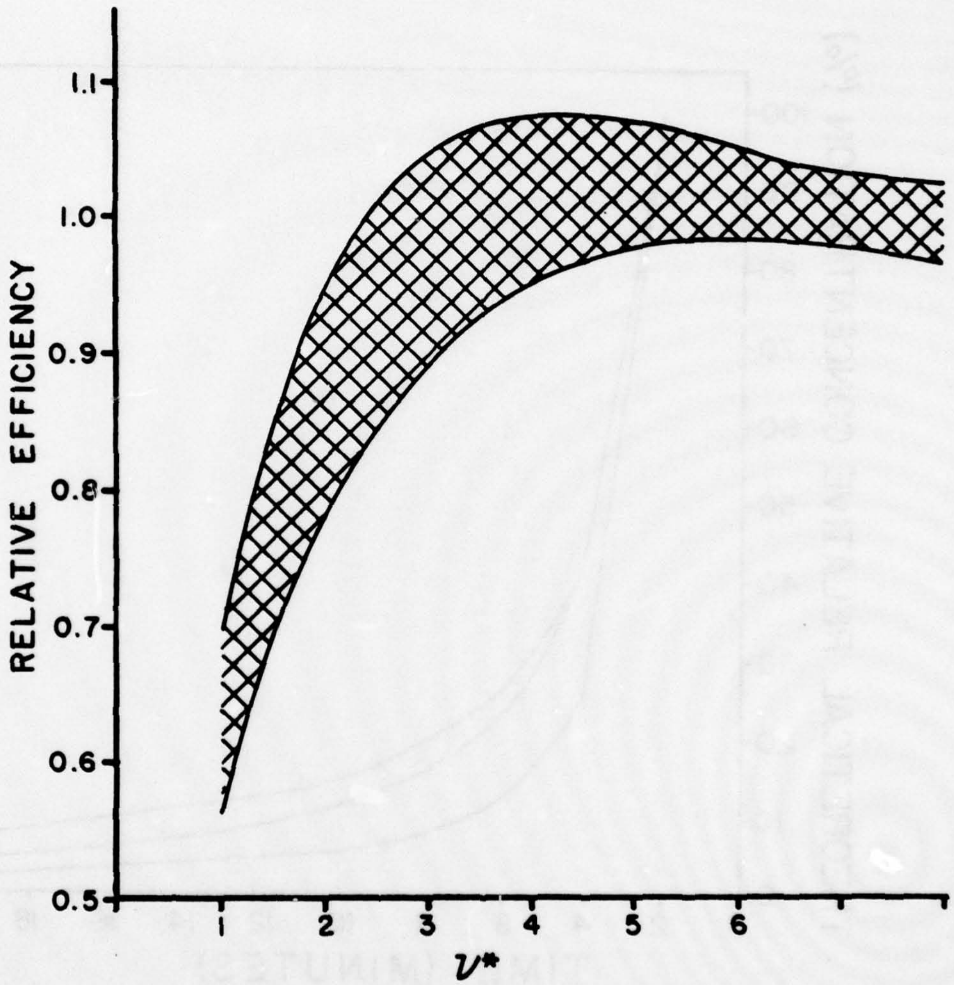

Fig. 9 — Envelope of relative efficiency as a function of ν^* for four types of nephelometers

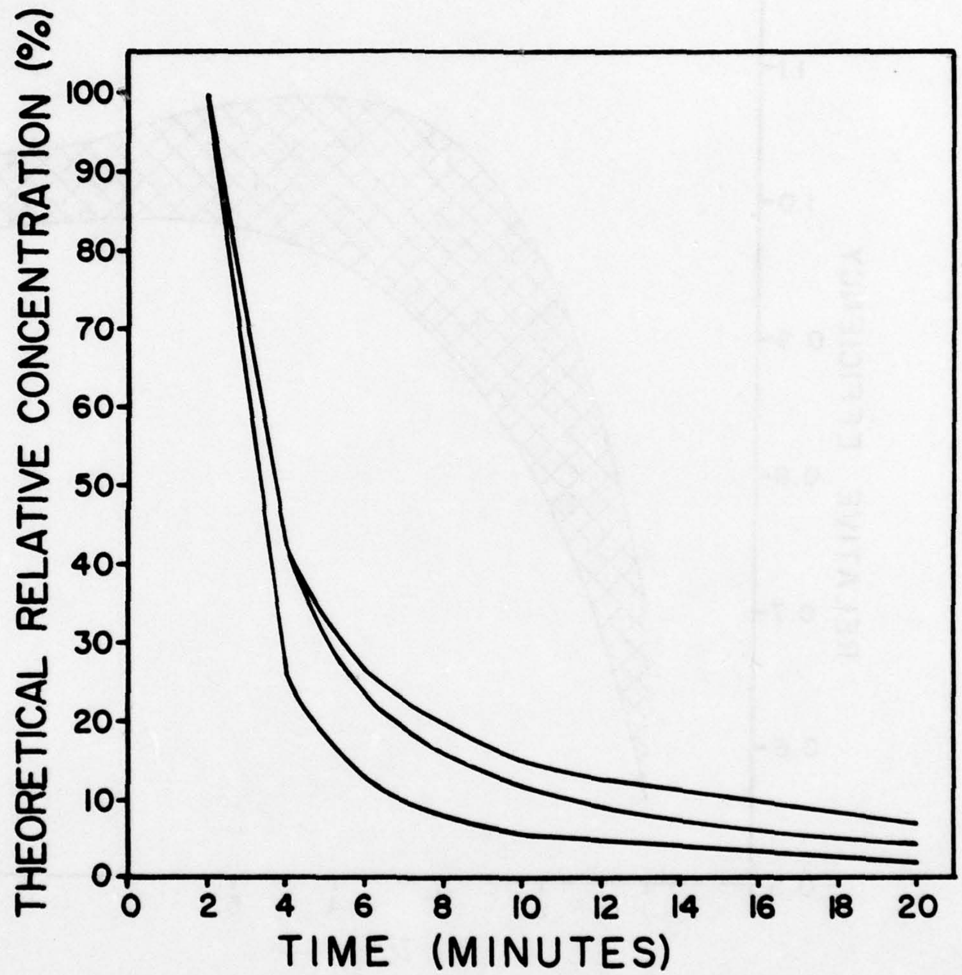

Fig. 10 — Concentration reduction by turbulence

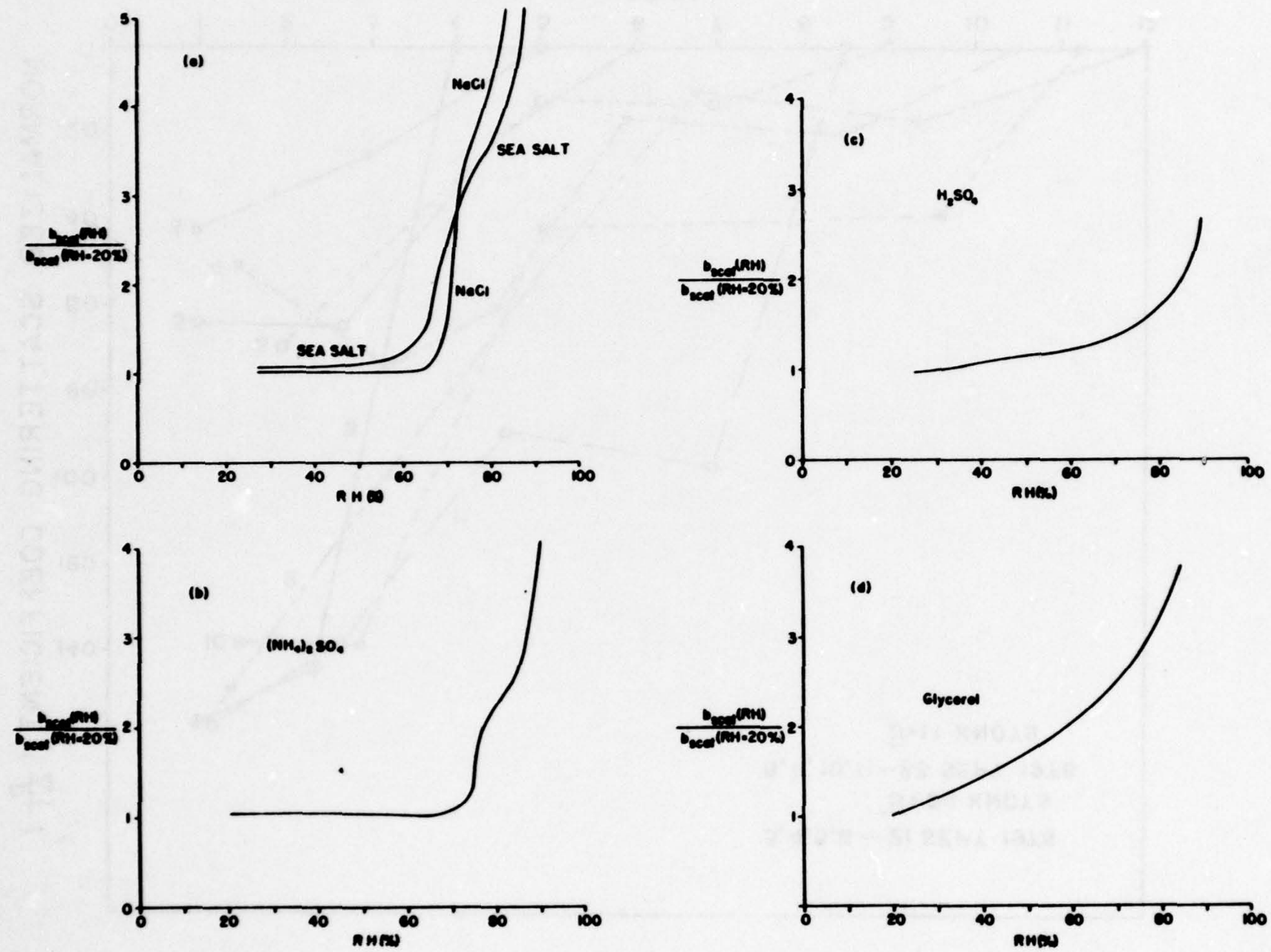

Fig. 11 — Relative light scattering coefficient for laboratory aerosols generated in a bursting-bubble generator (*Covert*, 1972). [From S.S. Butcher & R.J. Charlson(1972)].

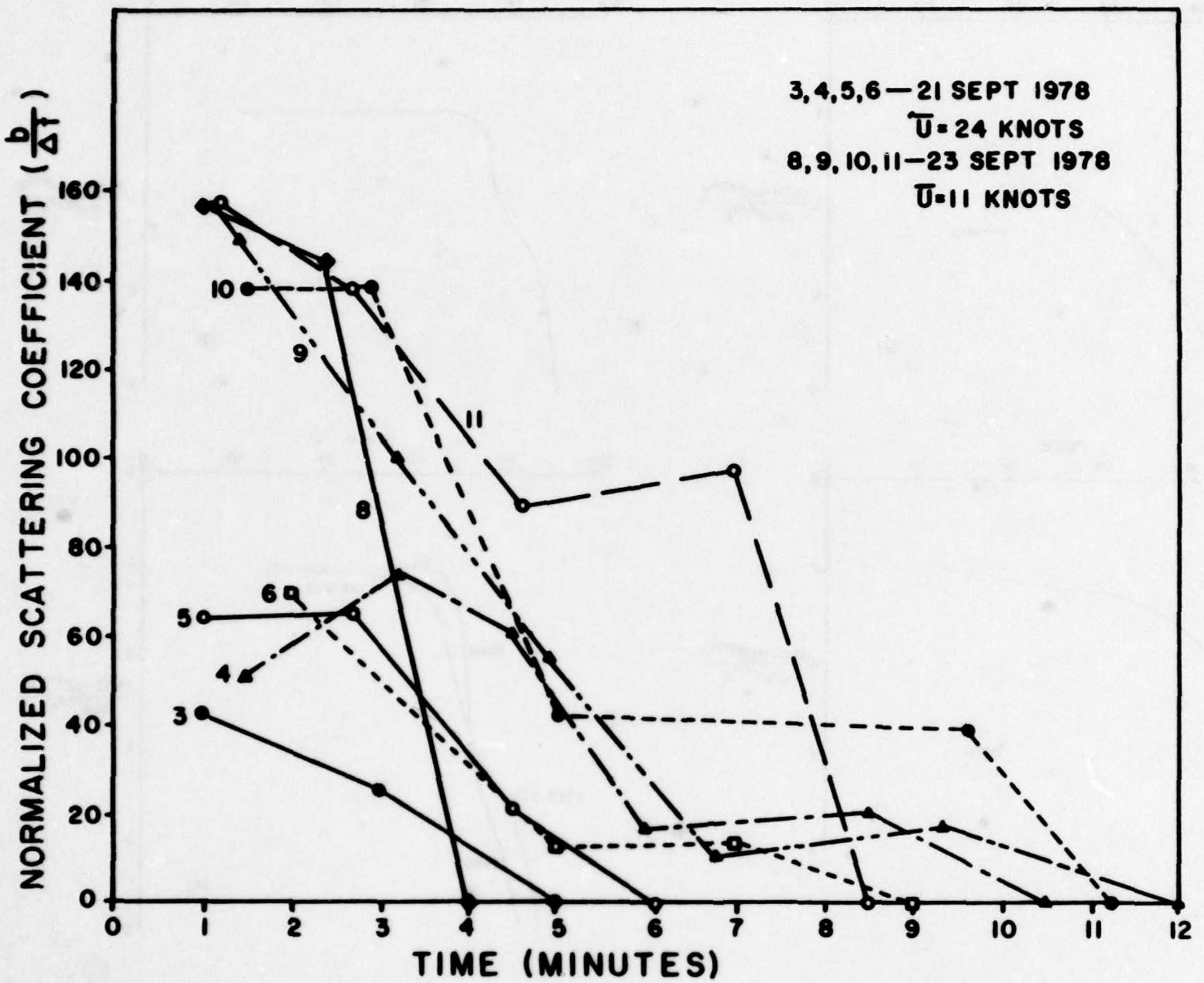

Fig. 12 — Experimental plume observation