AN EXPERIMENTAL STUDY OF THE TRANSIENT RESPONSE OF WATER DROPLETS TO RAPIDLY CHANGING CONDITIONS AT LOW TEMPERATURES
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1. INTRODUCTION

Cloud droplets continually respond to the time-varying supersaturations found in atmospheric clouds by exchanging mass (water vapor) and energy (thermal and latent forms) with the surrounding air. Thus, supersaturations caused, say, by adiabatic expansion of rising air currents drive fluxes of vapor molecules and latent heat toward the cloud particles, leading to the condensational growth of droplets that are slightly warmer than the ambient air. Conversely, during periods of undersaturation, these mass and latent heat fluxes are reversed, leading to evaporating droplets that are cooler than the air. How effective the water droplets are in responding to the environmental conditions influences a number of phenomena, including activation of cloud condensation nuclei, the breadth of initial drop-size distributions and possibly freezing nucleation, so it is important that we can quantify the rates of mass and energy exchange.

At temperatures below 0 °C, the cooling effect that accompanies droplet evaporation can in principle enhance the likelihood of freezing by increasing the supercooling and thereby the thermodynamic driving force for nucleation. Observations in wave clouds (Cooper 1995) and cumuliform clouds (Hobbs and Rangno 1985) indeed show enhanced ice formation in association with droplet evaporation. Beard (1992) contrasts several possible mechanisms of ice initiation and concludes that organic residues from the evaporating droplets could be responsible for such enhanced ice formation. Some mechanisms were dismissed from further consideration in his assessment in part because the magnitude of cooling due to evaporation is traditionally expected to be small. Cooper (1995), on the other hand, suggested that the cooling could be substantially larger for droplets in the kinetic regime than that calculated from conventional wet-bulb theory if the thermal accommodation coefficient is small. One cannot distinguish the actions of such contrasting hypotheses without further investigation.

Laboratory experimentation offers an excellent opportunity to gain the additional information needed to distinguish alternative mechanisms of action in a case like this. Whereas investigation of the "evaporation ice nuclei" hypothesized by Beard is made difficult by the uncertain chemical history of droplets in real clouds, the kinetically based cooling mechanism of Cooper is amenable to testing. In this paper we attempt to define an appropriate experimental procedure for doing so.

2. EXPERIMENTAL APPROACH

The basic idea underlying our investigation of the kinetically driven cooling of evaporating droplets is to expose individual water droplets to subsaturated air or N₂ at various low temperatures and measure their rates of evaporation as the radii become comparable with the mean free path in the gaseous environment. The composition of the droplets is not a directly relevant factor driving the physics in Cooper's hypothesis, so we simply use distilled, deionized water as the parent solution. The rate of evaporation is then dependent on the combined rates of vapor and heat transfer through the air (by molecular diffusion and conduction, respectively) and the kinetic processes occurring at the liquid-air interface. To the extent that the non-condensable gas molecules are able to exchange thermal energy with the liquid, a process traditionally parameterized in terms of the thermal accommodation coefficient $\alpha$, the droplet gains the enthalpy needed to vaporize the water. Comparisons of the measured evaporation rates with those computed from theoretical models incorporating the surface-kinetic processes (e.g., Fukuta and Walter 1970; Carstens 1979; Robinson and Scott 1981) then lead to conclusions about the likely magnitude of $\alpha$ for...
The actual temperature of an evaporating droplet is of course not directly measurable, so we must invariably depend on model calculations to gain this information and estimate the probabilities of ice nucleation. However, because the homogeneous nucleation of ice in pure water is so strongly dependent upon the water temperature, we use the empirical appearance of a phase change to "anchor" the calculated temperature trends to this relatively well-defined point. As shown in Fig. 1, this approach also enables us to distinguish qualitatively between low- and large- \( \alpha \) possibilities. The decrease in drop freezing rate with decreasing radius that appears toward the right-hand side of the graph reflects the volume-dependence inherent in calculating the fractional rate of freezing as the product \( JV \), where \( J \) is the temperature-dependent nucleation rate \( \left[ \text{m}^3 \text{s}^{-1} \right] \) and \( V \) is the volume \( \left[ \text{m}^3 \right] \) of a droplet at a given instant. At a given temperature, smaller droplets offer fewer opportunities for critical embryos to form. As a droplet shrinks in size, however, the surface-kinetic processes begin to dominate the resistance to mass and energy transport, so the curves separate, depending on the magnitudes of the thermal and mass accommodation coefficients. For sufficiently low values of \( \alpha \) (for \( \beta = 1 \)) and saturation ratio, we see that the nucleation probability becomes very large in response to the strong cooling that occurs when the noncondensible gas is restricted in its ability to transfer thermal energy to the liquid to compensate for the evaporating water. Large droplets introduced into the cold gas may not freeze initially (freezing rate below the chosen threshold of \( JV = 1 \left[ \text{s}^{-1} \right] \)), but they could as they evaporate if the magnitude of \( \alpha \) is relatively small (allowing \( JV \) to exceed the threshold rate).

The experimental apparatus used to measure the droplet evaporation rates and detect freezing events is a low-pressure chamber housed in a special vacuum chamber used to isolate the system from the laboratory environment. A cubic electrodepressive levitator (Lamb et al. 1996) mounted within the inner chamber permits a charged droplet to remain isolated at a given location in a gas stream while it is interrogated optically from outside the apparatus. A gentle flow of gas, pre-conditioned to the desired temperature and humidity, is made to pass through the cubic cell in order to provide a well-defined environment for the droplet.

Once a slightly charged droplet is introduced into this environment and trapped in the cell, it is observed with two video cameras, one for imaging the particle, the other for obtaining the interference patterns generated by the elastic scattering of the incident laser light. The scattering angle detectable with this camera ranges from about 30° to 60°. The optical information is recorded on magnetic tape until the droplet mass-to-charge ratio falls below the criterion for stable levitation (Allison and Kendall 1996). Operation of the experiment at reduced gas pressure extends the time that the droplets reside in the kinetic-transition regime. The pattern of interference fringes generated by the particle are highly regular for the spherical droplets, but they become obviously irregular once the liquid freezes and the particle becomes nonspherical. The regularity of the fringe spacing associated with scattering from spheres allows us to use Mie theory (Bohren and Huffman, 1983, Chapt. 4) to determine the size of the droplet and its evaporation rate with high accuracy and precision. The time at which the interference patterns becomes irregular defines the freezing transition.

3. PRELIMINARY FINDINGS

The procedure outlined in the previous section has been used to obtain preliminary data on the evaporation of individual droplets in subsaturated air. By way of example, Fig. 2 shows the change in radius of a water droplet, initially 15 \( \mu \text{m} \) in radius, following injection into a relatively warm environment \( (T = 258 \text{K}) \) at atmospheric pressure. At this temperature, droplets remain supercooled indefinitely and evaporate relatively rapidly. The
Fig. 2. Results from a water droplet evaporating in air at about 258 K. Points: Radii derived from observed scattering patterns; Curve: Radii calculated from continuum theory.

data (points) indicate that the droplet evaporated at rates that increased as the droplet became smaller and the gradients of vapor and temperature in the air over the surface of the droplet increased. For the first 10 s or so, the radii followed a trend consistent with theory based on a traditional continuum model (see, for instance, Rogers and Yau, 1989, Chapt. 7), as given by the curve in Fig. 2. Toward the end of this experimental run, however, the rate of evaporation lagged behind that calculated from such a model, giving hints that something other than bulk-phase transport properties may have limited the evaporation of the droplet.

4. CONCLUSIONS

Based on preliminary data, the experimental approach outlined here is reasonable and appropriate for testing the hypothesis that evaporating cloud droplets cool substantially, because of a kinetic limitation to the transfer of heat, and thereby enhance the probability of freezing. However, it should be noted that the change is drop volume during evaporation needs to be accounted for in order to gain an accurate prediction of ice enhancement. Because the diminishing volume during evaporation limits the opportunity for ice embryos to reach critical size, one should expect that this evaporation mechanism can be effective only if the thermal accommodation coefficient is very low (α less than about 0.03), while the condensation coefficient β is close to unity. It is not likely that weak or moderate restrictions to energy exchange are sufficient to compensate for the substantial volume effect inherent in freezing nucleation.

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