Quasi-Equilibrium Temperature Differences between Radiating Ice Crystals and the Surrounding Air

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ABSTRACT

A previous attempt to calculate the temperature gradient around a growing ice crystal in clear air started with the radiation budget of the crystal plus the assumption that the measured frost point temperature (about 2°C below air temperature) represented the crystal temperature. When the conductivity equation for the air around the crystal is fully solved with the radiation budget as the boundary condition, however, it is found that less than 0.03°C temperature difference can be sustained by radiative cooling of the crystals. The most probable explanation of the difference between the two approaches is that the humidity measurements are in error, and that the error has been generally recognized only at measured humidities in excess of 100%.

1. Introduction

It has been suggested that measured humidities less than ice saturation in the presence of stable or growing suspended ice crystals could be accounted for by temperature differences between the crystals and the air (Gotaas and Benson, 1965). If the ambient air temperature is falling due to radiative cooling via ice crystals, conduction of heat to the crystals from the air requires that the crystals be colder than the air. Gotaas and Benson pointed out that measured humidities of the order of 80% over ice were frequently observed on radiosonde and ground station measurements taken during ice crystal displays². Measurements by Thuman and Robinson (1954), using the Karl Fischer reagent method, showed similar undersaturation with respect to ice in the presence of ice fog. Since there was no evidence that the crystals were either growing or evaporating, Gotaas and Benson assumed that the air at the crystal boundary must be ice-saturated, i.e., that the crystals were at the frost point temperature. From this assumption and the measured humidities, they deduced that the crystals should be about 2°C colder than the surrounding air, and then used the radiation balance of the crystals to find out how far from the crystal the temperature anomaly extended (about 100–250 μm).

The mechanics of radiative cooling in the presence

of ice aerosols have recently been reexamined (Bowling, 1970). As a part of this study the quasi-equilibrium temperature difference between suspended ice crystals and the bulk of the surrounding air was computed on the basis of strictly thermal considerations. Thus, rather than assuming that the humidity measurements were accurate, we computed the possible humidity range from the computed radiation balance. The results imply that a temperature difference of 2°C cannot be maintained between an ice crystal in the size range considered (radius, 25 μm) and the air if a reasonable radiation balance is assumed. Therefore, the humidity measurements, which are notoriously difficult to make at low temperatures, are probably invalid.

2. Theory

Consider a suspension of n spherical ice crystals per unit volume, each of radius a . Assume that the average spacing R between crystals is large compared with the crystal dimensions. Let the thermal conductivity, density and specific heat of air be denoted by k , ρ and c_p , respectively. Numerical calculations indicate that one plane ice crystal per cubic centimeter of radius 25 μm will enhance atmospheric radiative cooling rates by about two orders of magnitude (Bowling, 1970). Therefore, in this case, about 99% of the radiative cooling of the air is due to the ice crystals contained in it, and to a good approximation we can assume that the energy flow H to each crystal balances the radiative flux divergence, dF/dz , divided by n , or

$$H = (1/n)dF/dz. \quad (1)$$

The energy flow H may come from two sources. If the crystal is growing at such a rate that the water

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² An ice crystal display consists of a layer of air with suspended or slowly falling small, well-formed ice crystals. The layer is at least several hundred meters in thickness, and in the case considered here its lower boundary is at the snow surface. These displays are common at high latitudes in the fall and winter, and give rise to well-marked optical effects.

vapor content of the surrounding air is held at ice saturation at the crystal temperature as the air and crystals cool, there will be a positive flux of latent heat equal to $-(L/n)(d\rho_i/dT_c)(dT_c/dt)$. Here L is the latent heat of sublimation, ρ_i the saturation vapor density over ice, T_c the crystal temperature, and t time. Spatial variations in vapor density do, of course, occur and, in fact, the atmospheric vapor density must be slightly above ice saturation at T_c for growth to occur. But fluctuations will be so small relative to ρ_i that the above equation is still valid over a suitable averaging time. In addition, the air itself is cooling at a rate dT/dt , and must provide by conduction an amount of energy $-(c_p\rho/n)(dT/dt)$ to each crystal. The air temperature T varies somewhat with position, but for a constant dF/dz major spatial variations in dT/dt must be small, and will be neglected. In particular, we assume $dT_c/dt = dT/dt$. In the type of crystal display considered, the ice crystals have a mass of the order of 2×10^{-8} gm cm $^{-3}$ compared with 10^{-3} gm cm $^{-3}$ for air, so the loss of energy due to the change of temperature of the ice crystal itself is several orders of magnitude smaller than either of the above terms.

For each crystal, (1) becomes $-[L(d\rho_i/dT_c) + c_p\rho] \times (dT/dt)/n = (dF/dz)/n$, or

$$\frac{dT}{dt} = -\frac{dF}{dz} / \left(L \frac{d\rho_i}{dT_c} + c_p\rho \right). \quad (2)$$

If we make the assumption that over the small distances involved the transfer of heat from the air is due to molecular conductivity k , the equation of thermal conductivity in the air is

$$\nabla^2 T = c_p\rho(dT/dt)/k. \quad (3)$$

Assuming spherical symmetry (the derivatives with respect to angle equal to zero), which implies that the influence of the other crystals on the isotherms near a given crystal is small, this may be written as

$$\frac{1}{r^2} \frac{d}{dr} \left(r^2 \frac{dT}{dr} \right) = -\frac{c_p\rho}{k} \frac{dF}{dz} / \left(L \frac{d\rho_i}{dT_c} + c_p\rho \right). \quad (4)$$

The variation of the right-hand side with r is negligible, and we may set $c_p\rho(dF/dz)/k[L(d\rho_i/dT_c) + c_p\rho] = B$. The appropriate boundary condition is that the temperature gradient at the crystal boundary must provide a total conductive heat flow of $c_p\rho(dT/dt)/n$ per crystal. Using (2), this boundary condition may be written as

$$4\pi a^2 k (dT/dr)|_a = (c_p\rho/n)(dF/dz) / [L(d\rho_i/dT_c) + c_p\rho] = kB/n. \quad (5)$$

Eq. (4) may then be solved to give

$$dT/dr = -Br/3 + Ba^3/3r^2 + B/4\pi r^2 n, \quad (6)$$

or, again integrating from a to r ,

$$T - T_c = B \left[-r^2/6 + a^2/2 - a^3/3r + (1/4\pi n)(1/a - 1/r) \right] \quad (7)$$

valid for $a < r < R/2$, where R is the average distance between crystals.

The first three terms of this equation arise from the right-hand side of (3), and result from the fact that the air very close to the crystal is contributing energy (from its own cooling) to the crystal in addition to conducting heat to the crystal from the more distant air. This would be expected to reduce dT/dr and thus $T - T_c$, as is observed. The remaining term is simply the solution to

$$\nabla^2 T = 0 \quad (8)$$

with the appropriate boundary conditions. Since (8) may readily be solved for non-spherical crystals by analogy with the electrostatic equation (Jeffreys, 1918), the relative magnitude of the terms in (7) is of some interest. It can, in fact, be shown that the first three terms are negligible so long as a is much less than R .

If the crystals are imagined as being arranged in a cubic grid, it becomes apparent that, with n crystals per unit volume, each crystal will be associated with a cube of volume $1/n$. The edge of this cube will have a length $R' = (1/n)^{1/3}$, where R' is not the same as the average distance between crystals R , but it is the same order of magnitude. Thus, n is of the order of $1/R^3$, and the last term of (7) is approximately

$$\frac{1}{4n} \left(\frac{1}{a} - \frac{1}{r} \right) = \frac{R^3}{4} \left(\frac{1}{a} - \frac{1}{r} \right) = \frac{R^3}{4ar} (r - a). \quad (9)$$

The first three terms may be combined to give

$$-\frac{r^2}{6} + \frac{a^2}{2} - \frac{a^3}{3r} = \frac{1}{ar} \left(-\frac{ar^3}{6} + \frac{a^3r}{2} - \frac{a^4}{3} \right). \quad (10)$$

For values of r close to a , the ratio of (10) to (9) is of the order of $4\pi a^3/R^3$. For larger values of r , the r^3 term dominates in (10), and the ratio is of the order of $4\pi r^2 a / (6R^3)$. Since the largest r of interest is $R/2$, this is at most $\pi a / (6R)$. As we are interested in a case with $a = 0.0025$ cm and $R = 0.5$ cm, it is clear that substitution of (8) for (3) will introduce a negligible error.

The ratio of a to $R/2$ makes it clear that the temperature at $r = R/2$ differs little from that of most of the air or from the theoretical temperature at infinity, which we will call T_0 . The temperature difference in which we are interested is $\Delta T = T_c - T_0$, or, from the last term of (7) with $r = \infty$,

$$\Delta T = -\frac{c_p\rho}{4\pi nka} \frac{dF}{dz} / \left(L \frac{d\rho_i}{dT_c} + c_p\rho \right). \quad (11)$$

As pointed out above, (8) is mathematically identical to the electrostatic equation, $\nabla^2 V = 0$, with the

boundary condition $V=V_e$ at a conducting surface which has the shape of the crystal. Here V is electrostatic potential. The solution for V_e is well known. If V_0 is the potential at infinity, Q the total charge on the crystal analog, and C the electrostatic capacitance, then $V_e=V_0+Q/C$, or $\Delta V=Q/C$. Since C depends only on the geometry of the situation, it is carried over unchanged into the solution for temperature, and a full solution may be obtained by finding the form of K , the geometry-independent thermal analog to Q . This may be most readily done by considering a case for which the solution for ΔT is easily obtained such as the spherical ice crystal considered above. From the capacitance of a sphere, $C=a$, we have $K=-c_p\rho \times (dF/dz)/(4\pi kn[L(d\rho_i/dT_e)+c_p\rho])$, which holds for a crystal of any shape. For a flat, thin, circular plate of radius a , the capacitance is $2a/\pi$ and we have

$$\Delta T = -\frac{c_p\rho}{8\pi k a} \frac{dF}{dz} \bigg/ \left(L \frac{d\rho_i}{dT_e} + c_p\rho \right). \tag{12}$$

In summary,

$$\Delta T = -B/(4\pi n a), \quad (\text{sphere, radius } a), \tag{12a}$$

$$\Delta T = -B/(8\pi n a), \quad (\text{flat circular plate, radius } a). \tag{12b}$$

3. Results

Table 1 gives the appropriate parameters for the displays considered by Gotaar and Benson. Substituting, we obtain $\Delta T=-0.027^{\circ}\text{C}$. This temperature difference is not sufficient to produce any measurable deviation from ice saturation. At the surfaces of a volume of air containing ice crystals, the flux divergence, dF/dz , will be roughly proportional to the surface area of the crystals in a unit volume, which in turn is proportional to a^2n . Since ΔT is proportional to $(dF/dz)/na$, ΔT is approximately proportional to the radius of the crystals. A snowfall (or rainfall) with particles of 1–2 mm radius might conceivably produce a 2C depression of the frost point (or dew point) near the boundaries of the air volume affected, but in the interior of the shower the outlying particles would provide a shield against radiative loss and no net energy loss from the interior droplets

TABLE 1. Parameters of an ice crystal display.

a	25×10^{-4} cm
shape	thin circular disc
n	1 cm^{-3}
$\frac{dF}{dz}$	3.5×10^{-8} cal $\text{cm}^{-2}\text{sec}^{-1}$
T_0	-20°C
k	5.45×10^{-5} cal $\text{cm}^{-1}\text{sec}^{-1} (^{\circ}\text{C})^{-1}$
c_p	0.24 cal $\text{gm}^{-1} (^{\circ}\text{C})^{-1}$
ρ_i	1.4×10^{-3} gm cm^{-3}
$L \frac{d\rho_i}{dT_e}$	5.5×10^{-6} cal $(^{\circ}\text{C})^{-1} \text{cm}^{-3}$

would be expected. Thus, measured humidities significantly below ice saturation in the presence of stable or growing ice crystals cannot be explained by radiatively produced temperature differences except in localized regions at the boundary of the affected volume.

Humidity measurements at low temperatures are notoriously difficult, and this fact almost certainly accounts for the low measurements of relative humidity in natural ice crystal aerosols, and probably for those in ice fogs as well. However, the possibility that impurities could affect the saturation vapor pressure over at least polluted ice fog crystals [as has been observed in polluted city fogs (e.g., Robinson, 1962)] should be investigated.

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