

A Reevaluation of the Role of Thermophoresis as a Mechanism of In- and Below-Cloud Scavenging¹

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ABSTRACT

A comparison is made of the roles played by thermophoresis, Brownian diffusion and diffusiophoresis in the precipitation scavenging of aerosol particles. It is shown, for a certain range of particles sizes, that thermophoresis dominates provided that the latent heat associated with the phase transition of water is transferred through the air, to or from the precipitation element by conduction. A plot of the resulting washout coefficient shows that thermophoresis enhances the below-cloud rain scavenging of aerosol particles whose radii are between 0.01 and 1.0 μ . The solution is presented for the appropriate convective diffusion equation which describes thermophoretic effects on the in-cloud scavenging problem.

1. Introduction

Scavenging of airborne particulate material through natural condensation and precipitation processes is known to occur by a number of mechanisms. Among the most important of those that have been recognized in the literature are Brownian diffusion, electrical effects, impaction, interception, diffusiophoresis and thermophoresis. Previous researchers have concluded, compared to other mechanisms of capture, that thermophoresis should play only a minor role as a process of in- and below-cloud scavenging. This conclusion has arisen largely from the fact that temperature gradients in the vicinity of atmospheric water droplets and crystals are small, suggesting that thermophoretic forces are insufficient to cause appreciable capture (Goldsmith and May, 1966). Deryaguin and Dukhin (1957) suggested that thermophoretic forces should be significant in promoting the coagulation of cloud droplets; however, these authors did not extend their analysis to the related problem of aerosol capture.

Contrary to the conclusions of previous authors, the present analysis indicates that situations exist wherein thermophoresis may predominate over the competing mechanisms of diffusiophoresis and Brownian motion (Slinn and Hales, 1970). This is based largely on the fact that condensation and evaporation processes must be accompanied by the transfer of substantial quantities of latent heat. For normal in- and below-cloud conditions a majority of this energy must be conducted through the surrounding medium, thereby providing sufficient impetus for appreciable thermophoretic transport.

The following section of this paper will illustrate the approach of the present analysis by considering a highly simplified model of the scavenging process. The treatment then will be repeated using more realistic models to describe the transport phenomena, and the results will be presented analytically and graphically in terms of pertinent variables. Relationships between these results and those of previous authors will be discussed in the concluding section of the paper.

2. Simplified analysis of phoretic and Brownian transport to a suspended droplet

To illustrate the contention that thermophoresis should be a rather significant mechanism of aerosol

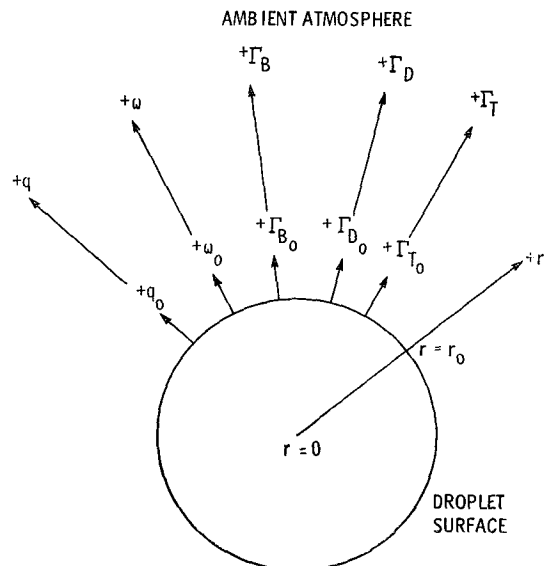


FIG. 1. Sign conventions.

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scavenging, we consider first the simplified case involving a suspended, uncharged, evaporating droplet. Sign conventions for the analysis, as shown in Fig. 1, will be such that all entities directed in the $+r$ direction are positive.

If all latent heat for the evaporation is supplied by conduction from the surroundings, one may write

$$q_0 = -\lambda w_0, \quad (1)$$

where λ is the latent heat of vaporization, and q_0 and w_0 the fluxes of heat and water vapor mass at the droplet surface.

As a simplified expression for thermophoretic behavior one may employ the equation suggested by Waldmann and Schmidt (1966) to obtain

$$\Gamma_t \approx -\frac{kN_\infty}{5p} \frac{dT}{dr}, \quad (2)$$

or, at the droplet surface,

$$\Gamma_{t0} \approx \frac{q_0 N_\infty}{5cRT_\infty} = -\frac{kN_\infty(T_\infty - T_0)}{5r_0cRT_\infty}. \quad (2a)$$

Here, Γ_t and Γ_{t0} are the thermophoretic fluxes of particles in the $+r$ direction, k the thermal conductivity, p the ambient pressure, and N_∞ the number-concentration of particles adjacent to the droplet surface. Subscripts 0 and ∞ pertain to surface and bulk conditions, respectively. The ideal gas law has been employed in the formulation of (2a), where c represents the total molar concentration in the surrounding medium and R the gas-law constant. Since (2) and (2a) are to be used for illustrative purposes only it is not necessary to discuss them further, except to state that they are known to describe physical behavior reasonably well for small aerosol particles.

A corresponding approximate expression for diffusiophoresis can be obtained by assuming the diffusiophoretic drift velocity is equal to the mass-average velocity of the surrounding fluid. This gives

$$\Gamma_{d0} \approx \frac{w_0 N_\infty}{\rho} = -\frac{q_0 N_\infty}{\rho \lambda}, \quad (3)$$

where ρ denotes the density of the ambient gas.

Finally, Brownian diffusion to the suspended droplet may be described by the classical diffusion equation, whose steady-state solution for this case is

$$\Gamma_{B0} \approx -\frac{D_p}{r_0} N_\infty, \quad (4)$$

provided that all aerosol particles approaching the droplet surface are captured irreversibly (total-retention boundary condition). In (4) D_p is the Brownian diffusion coefficient of an aerosol particle.

The total-retention boundary condition, i.e., $N=0$ at $r=r_0$, seems to conflict with the assumption of finite N adjacent to the interface used to formulate (2) and (3). This treatment is a valid approximation, however, whenever the particle-concentration gradients at the interface are large enough so that the bulk aerosol concentration is approached within very short distances from the droplet surface (Hales *et al.*, 1970). Such a requirement is satisfied under present conditions owing to the low values of D_p associated with the aerosol particles of interest. An additional assumption, the existence of a quasi-steady state, has been employed in the formulation of (2)–(4). This implies that for any given instant, changes with time are small compared with changes with radial position. We have assumed also that droplet size is quasi-constant, i.e., the motion of the droplet boundary by growth or shrinkage is small compared to the deposition velocities of the particles. The validity of both the quasi-steady state and the quasi-constant drop-size assumptions has been verified in this study by performing numerical solutions of the transient equations for particle transport.

Combining the above equations one can obtain rough estimates of the relative magnitudes of thermophoresis, diffusiophoresis and Brownian movement. For the phoretic transport ratio this gives

$$\frac{\Gamma_{t0}}{\Gamma_{d0}} \approx -\frac{M_a \lambda}{5RT_\infty}, \quad (5)$$

where M_a is the "molecular weight" of air, 29.0. The minus sign in (5) indicates, as expected, that diffusiophoresis and thermophoresis act counter to one another under the conditions of this analysis. For small droplets suspended in air, (5) predicts that thermophoretic transport should exceed diffusiophoretic transport by a factor of about 6 under expected atmospheric conditions.

From (2a) and (4) we obtain the thermophoretic-to-Brownian ratio,

$$\frac{\Gamma_{t0}}{\Gamma_{B0}} \approx \frac{k(T_\infty - T_0)}{5D_p c R T_\infty}. \quad (6)$$

Eq. (6) can be used to show that thermophoresis can exceed Brownian transport under appropriate atmospheric circumstances. For example, 0.1 μ particles in the vicinity of a cloud droplet evaporating to support the growth of neighboring -10°C ice crystals are characterized by a thermophoretic-to-Brownian transport ratio of about 24.

Eqs. (5) and (6) support the previously stated contention regarding the importance of thermophoretic effects. This contention can be justified even in view of the fact that temperature gradients are small by noting that, as predicted by (2), thermophoresis should be related directly to the net heat flux q_0 . This entity is proportional to the latent heat and water vapor

transport rates regardless of the magnitude of the temperature gradient.

3. Refined analysis

The analysis of the preceding section is somewhat limited because of the many simplifying assumptions employed therein. At best we would expect these results to be applicable for submicron particles only. We now attempt to refine and generalize our treatment by choosing more appropriate expressions for the mechanisms of particle transport. Although these expressions are more complex, the approach will be identical to that used in the previous section. One should note particularly that the previous assumptions of a quasi-steady state, total retention of particles with large interfacial particle gradients, and interfacial heat transport solely by conduction [Eq. (1)] are retained in this section. The primary result is the demonstration of the qualitative validity of the conclusions drawn earlier concerning submicron particles. The results also indicate that for larger particles the relative influence of thermophoresis depends upon the thermal conductivity of the aerosol.

The expression that will be used here to describe thermophoretic transport is that which has been derived by Brock (1962):

$$\Gamma_{t0} = \frac{q_0 N_\infty f(K, k, k_p)}{cRT_\infty}, \tag{7}$$

where

$$f = \frac{2 [1 + AK + BK \exp(-C/K)](k + C_t K k_p)}{5 (1 + 3C_m K)(2k + k_p + 2C_t K k_p)}, \tag{8}$$

and k and k_p denote thermal conductivities of the gaseous medium and the aerosol, respectively. The Knudsen number K is defined as the ratio of the mean-free path of the gas to the radius of the particles.

Waldmann and Schmidt (1966) have plotted (8) using the following values for the (dimensionless) constants: $A = 1.45$, $B = 0.40$, $C = 1.0$, $C_m = 1.0$, $C_t = 2.5$. In the range of interest of parameters, the values of f are between 0.16 and 0.01. One should note that for $f = 0.2$, (7) reduces to the simpler form (2) used in the preliminary analysis. Waldmann and Schmidt have shown (7) to provide a reasonable representation of physical behavior over the entire range of pertinent variables.

The expression to be used to describe diffusiophoresis is derived from the equation suggested by Waldmann and Schmidt for a particle moving in a binary, diffusing system wherein one of the gases is held stagnant. This may be expressed by giving the diffusiophoretic particle flux in terms of the mass flux of water vapor, w_0 , as

$$\Gamma_{i0} = N_\infty g(K) \left(\frac{M_w^{-\frac{1}{2}}}{c_w M_w^{\frac{1}{2}} + c_a M_a^{\frac{1}{2}}} \right) w_0, \tag{9}$$

where the subscripts w and a refer to water vapor and air. Since the numerical data of Waldmann and Schmidt show g to be only a weak function of K , we use

$$1.0 > g > 0.85. \tag{10}$$

Eq. (9) is known to agree with experimental findings reasonably accurately for all conditions in the range of interest. For present use it can be simplified, owing to the low concentration of water vapor molecules, c_w , relative to that of air, c_a . Combining (9) and (1), and simplifying, gives

$$\Gamma_{d0} = -N_\infty g q_0 / (M_a^{\frac{1}{2}} M_w^{\frac{1}{2}} \lambda c), \tag{11}$$

which reduces to the simpler form (3) upon replacing $g / (M_a^{\frac{1}{2}} M_w^{\frac{1}{2}} c)$ with the quantity $1/\rho$.

The expression that will be used to describe transport to the droplet surface by Brownian motion is

$$\Gamma_{B0} = -\frac{D_p N_\infty}{2r_0} (2 + 0.6 \text{Re}^{\frac{1}{2}} \text{Sc}_p^{\frac{1}{2}}), \tag{12}$$

where Re is the Reynolds number based upon the fall velocity and diameter of the droplet, and Sc_p is the Schmidt number for the particle, ν/D_p , ν denoting the kinematic viscosity of the gaseous medium. Eq. (12) can be viewed as an elaboration of (4) with an additional term to account for convective transport arising from the motion of the droplet. It is obtained by analogy from the similar equations for heat and mass transport to a falling sphere [Bird *et al.*, 1960], i.e.,

$$q_0 = -\frac{k(T_\infty - T_0)}{2r_0} (2 + 0.6 \text{Re}^{\frac{1}{2}} \text{Pr}^{\frac{1}{2}}), \tag{13}$$

$$w_0 = -\frac{M_w D_{AB} (c_{w\infty} - c_{w0})}{2r_0} (2 + 0.6 \text{Re}^{\frac{1}{2}} \text{Sc}^{\frac{1}{2}}), \tag{14}$$

where D_{AB} is the diffusion coefficient in the gaseous mixture, and Pr and Sc are the gas Prandtl and Schmidt numbers, respectively. Eq. (12) is expected to describe adequately the Brownian diffusional flux for particles of arbitrary size (Slinn and Shen, 1970), but it is to be emphasized that for particles $\gtrsim 1 \mu$ inertial terms must be incorporated into the analysis.

Revised estimates of the transport ratios can be obtained through combination of (7), (11) and (12). The resulting expression for the phoretic transport ratio is

$$\frac{\Gamma_{i0}}{\Gamma_{d0}} = \frac{f M_a^{\frac{1}{2}} M_w^{\frac{1}{2}} \lambda}{g R T_\infty}, \tag{15}$$

which can be compared with (5) from the previous section. It is important to note that not only is this ratio independent of droplet size, it is also independent of relative humidity of the air. Fig. 2 gives a plot of (15) based on values of f and g obtained from the paper by Waldmann and Schmidt (1966). This figure indicates

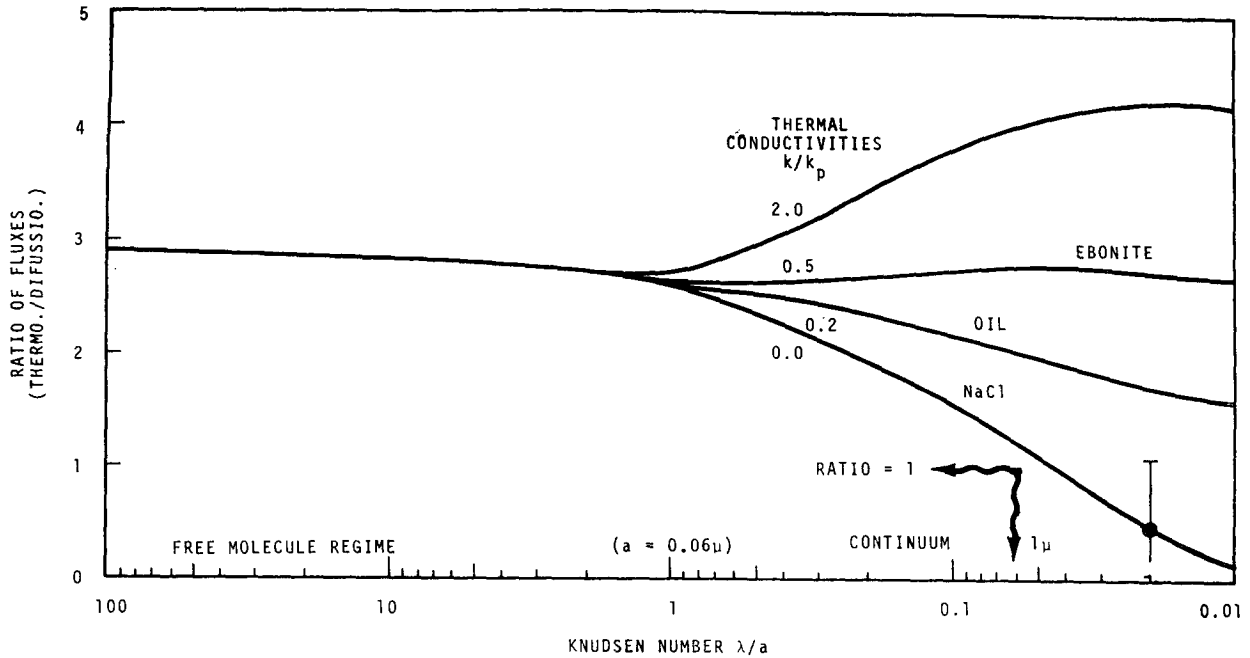


FIG. 2. Phoretic flux ratio as a function of Knudsen number.

again that thermophoresis should dominate diffusio-phoresis under most conditions of particle size and conductivity, although this dominance is somewhat less pronounced than that predicted on the basis of the simpler equation (5).

The thermophoretic-to-Brownian transport ratio, obtained through (11) and (12), is given by

$$\frac{\Gamma_{t0}}{\Gamma_{B0}} = \frac{2q_0 r_0 f}{D_p c R T_\infty (2 + 0.6 \text{Re}^{\frac{1}{2}} \text{Sc}_p^{\frac{1}{3}})}, \quad (16)$$

which, upon application of (13) appears as

$$\frac{\Gamma_{t0}}{\Gamma_{B0}} = \frac{fk(T_\infty - T_0)(2 + 0.6 \text{Re}^{\frac{1}{2}} \text{Pr}^{\frac{1}{3}})}{D_p c R T_\infty (2 + 0.6 \text{Re}^{\frac{1}{2}} \text{Sc}_p^{\frac{1}{3}})}. \quad (17)$$

Eq. (17) may be reduced into the two following limiting forms:

1) Small droplet fall velocity (in-cloud situation):

$$\frac{\Gamma_{t0}}{\Gamma_{B0}} \approx 2.4 \times 10^{-3} \frac{f(T_\infty - T_0)}{D_p}. \quad (18)$$

2) Large droplet fall velocities (below-cloud situation):

$$\frac{\Gamma_{t0}}{\Gamma_{B0}} \approx 4.2 \times 10^{-3} \frac{f(T_\infty - T_0)}{D_p^{\frac{2}{3}}}. \quad (19)$$

Numerical constants in (18) and (19) are based on transport and thermodynamic properties of air at 0C, but do not vary appreciably over moderate ranges of physical conditions. Particle diameters and tempera-

tures in (18) and (19) have units of centimeters and degrees Kelvin, respectively. From these equations the following generalizations are suggested:

1) For conditions where droplet velocities are low, thermophoresis becomes more important than Brownian motion whenever

$$D_p < 2.4 \times 10^{-3} f(T_\infty - T_0). \quad (20)$$

2) For situations involving high droplet velocities, thermophoresis becomes more important than Brownian motion whenever

$$D_p^{\frac{2}{3}} < 4.2 \times 10^{-3} f(T_\infty - T_0). \quad (21)$$

Situations satisfying inequalities (20) or (21) are not uncommon. For instance, a droplet falling through air of 90% relative humidity at 20C will drop to about 1C below the ambient temperature. A 0.1 μ aerosol characterized by a Brownian diffusivity of about 7 × 10⁻⁶ cm² sec⁻¹ and an *f* value of 0.12 provides values of 3.7 × 10⁻⁴ < 4.9 × 10⁻⁴ from (21), the inequality indicating that thermophoresis should indeed dominate Brownian motion under these circumstances.

4. Phoretic contribution to the overall washout coefficient

It is recognized widely that in-cloud scavenging, by virtue of its many interacting and competing mechanisms, is an extremely complex phenomenon. In view of such complexity the derivations of the previous sections should be viewed only as qualitative indications of the relative importance of the noted mechanisms.

Below-cloud scavenging, also complex, is recognized generally as being somewhat simpler than its in-cloud counterpart. This relative simplicity encourages an extension of the previous considerations in the interest of furthering the understanding of the total below-cloud process.

Specifically, these considerations can be employed to provide a theoretical estimate of the washout coefficient Λ defined by

$$R_w = \Lambda N_\infty, \tag{22}$$

where R_w is the rate of removal of particles of concentration N_∞ by washout in an element of unit volume.

On the basis of (7), (11) and (13), the net phoretic contribution to the particle flux is found to be

$$\Gamma_{ph0} = \frac{N_\infty \xi}{r_0} (2 + 0.6 Re^{1/2} Pr^3), \tag{23}$$

where

$$\xi = \frac{k(T_\infty - T_0)}{2c} \left(\frac{g}{M_a^{3/2} M_w^{1/2}} - \frac{f}{RT_\infty} \right). \tag{24}$$

To estimate the phoretic contribution to the washout coefficient we will use, for convenience, the formulas given by Zimin (1962) for raindrop size-distribution and terminal velocity:

$$N_d(r_0) dr_0 = \frac{10^{-4} J_0 r_0^2}{6 R_m^7} \exp\left(-\frac{2r_0}{R_m}\right) dr_0, \tag{25}$$

$$v = 8000 r_0 \text{ [length units sec}^{-1}\text{]}. \tag{26}$$

Here $N_d(r_0) dr_0$ is the number of raindrops per unit volume having sizes ranging between r_0 and $r_0 + dr_0$, J_0 the rainfall intensity (cm sec⁻¹), and R_m the radius of droplets in the maximum of the spectrum.

The net phoretic scavenging rate is determined by integrating the product of the phoretic flux and the surface area of the droplets over all possible raindrop sizes. This result may be combined with (22) to give

$$\Lambda_{ph0} = -\frac{10^{-4} J_0 \xi}{2 R_m^3} (1 + 0.6 \overline{Re}^{1/2} Pr^3), \tag{27}$$

where \overline{Re} is an intermediate Reynolds number, $2R_m v / \nu$.

Eq. (27) is plotted in Fig. 3 along with estimates of the effects of Brownian motion [cf. Eq. (12)], and of inertial contributions (Zimin, 1962). This figure demonstrates the enhancement of below-cloud washout of particles by thermophoresis; the effect is seen to be most important in the size range of 0.01–1 μ , where scavenging by diffusive and inertial mechanisms is ineffective. These findings may not be too significant quantitatively, since scavenging by electrical effects may play a major role under these circumstances. They are of qualitative significance, however, in that they indicate an enhancement of scavenging through phoretic

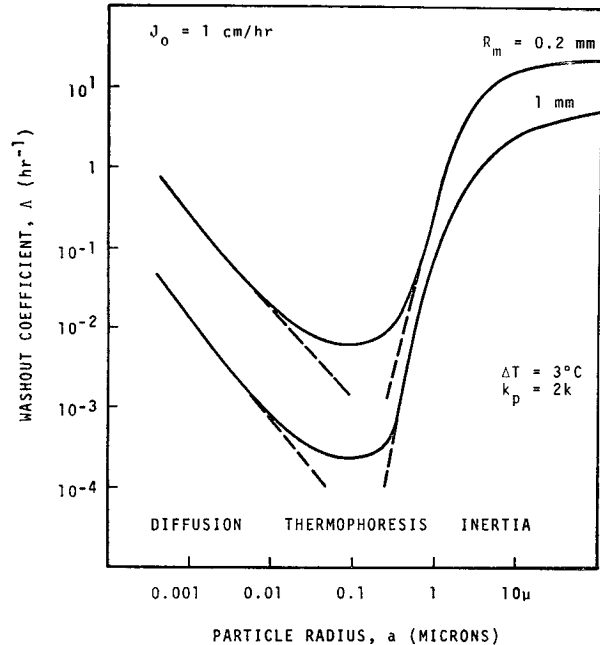


FIG. 3. Washout coefficient as a function of particle size illustrating the regimes of the various scavenging mechanisms.

phenomena—contrary to what has been accepted by most workers in this field up to the present time.

5. Discussion

Upon comparing the results of this investigation with those of previous authors, several questions arise. The most important of these concerns formation of the dust-free spaces that have been observed in the vicinity of evaporating surfaces during a number of laboratory investigations (cf. Goldsmith and May, 1966). Facy (1958) attributed this phenomenon to the sweeping away of particles by diffusiophoresis. This author based his conclusions upon his observations of the formation of dust-free spaces adjacent to evaporating droplets but not adjacent to those upon which condensation was in progress. Recently, the Facy effect has been incorporated into a composite model of in-cloud scavenging postulated by Vittori and Prodi (1967). This mechanism focuses upon the in-cloud growth of ice crystals at the expense of droplets through the Bergeron-Findeisen process. Provided that the Facy effect takes place, the formation of dust-free spaces around the droplets will serve to concentrate the aerosol elsewhere, thereby enhancing the opportunity for its capture by the growing ice crystals.

The indications of the present work, however, which state that under most conditions thermophoresis should oppose and dominate diffusiophoresis would seem to conflict with these findings. This apparent conflict can be examined more closely by reconsidering the simplified system of Section 2. At steady state the convective

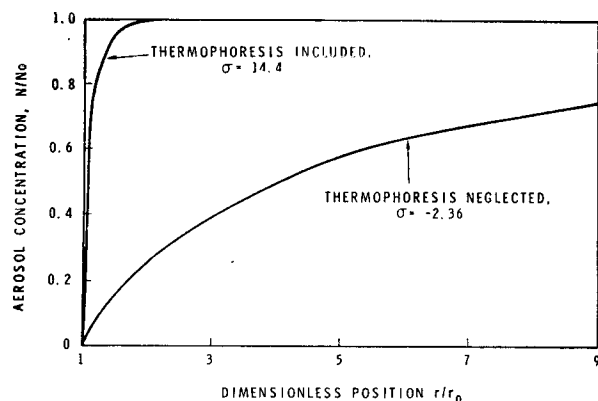


FIG. 4. Aerosol particle concentration profiles adjacent to an evaporating droplet.

diffusion equation describing this system reduces to

$$\frac{d^2N}{dr^2} = \left(-\frac{r_0\sigma}{r^2} - \frac{2}{r} \right) \frac{dN}{dr}, \quad (28)$$

where

$$\sigma = \frac{\Gamma_{i0}}{\Gamma_{B0}} + \frac{\Gamma_{d0}}{\Gamma_{B0}} = \frac{k(T_\infty - T_0)}{5D_p c R T_\infty} + \frac{k(T_\infty - T_0)}{M_a D_p c \lambda} \quad (29)$$

is the sum of the ratios of the phoretic transport rates to those by Brownian motion. For conditions corresponding to a single cloud droplet surrounded by 0.08μ aerosol particles and ice crystals at -10°C , σ is equal to 14.4 indicating, as expected, that the net phoretic motion is toward the evaporating droplet. If thermophoresis is neglected σ is found to be -2.4 , corresponding to movement in the outward direction.

The solution of (28) subject to the perfect-retention boundary condition $N(r_0) = 0$ is

$$\frac{N}{N_\infty} = \frac{\exp(\sigma r/r_0) - e^\sigma}{1 - e^\sigma}. \quad (30)$$

Plots of (30) corresponding to $\sigma = +14.4$ and $\sigma = -2.4$ are shown in Fig. 4. From these it is seen that the "dust-free space" should be rather constricted when thermophoresis is present, but becomes appreciable when its effect is deleted from the analysis. It should be noted that the σ values employed in Fig. 4 correspond to a rather fine aerosol. For larger particles (smaller Brownian transport) the magnitudes of σ would be greater and differences in the sizes of the dust-free areas would be even more pronounced. It should also be noted that a corresponding treatment for a planar evaporating surface must consider transient effects, since no steady-state solution exists for this geometry. For the case of a planar surface, the predicted dust-free space will increase indefinitely with time, regardless of the magnitude or sign of σ .

Much of the apparent discrepancy between the prediction of (30) and the earlier results possibly can be explained by reexamination of (1), which was funda-

mental to our derivation. This condition, which states that all latent heat must be transferred to or from the surroundings by conduction/convection, would seem to be fairly valid under normal in- and below-cloud conditions. This, however, may not have been applicable during the majority of the previous laboratory tests; indeed, some of these experiments actually involved evaporation from intentionally heated surfaces (Watson, 1936; Aitken, 1923), obviously voiding (1). It is somewhat doubtful whether other experiments satisfied (1), either, even though they did not involve intentionally heated (or cooled) surfaces. Energy input from other sources (e.g., by transient conduction from support structures, radiation from illuminators) may have been present in these experiments, thereby perturbing the system to characterize behavior more like that shown by the curve with $\sigma = -2.4$ of Fig. 4. It is suggested, therefore, that while these experiments undoubtedly demonstrated a valid phenomenon, they may not have provided an adequate representation of droplet and crystal behavior under normal atmospheric conditions.

The results of the previous sections suggest that a number of processes contributing to in-cloud scavenging may be related directly to thermophoretic effects. Owing to the complex nature of in-cloud behavior, most of these are highly speculative; nevertheless, they should be recognized as possible phenomena. Some of the more obvious of these are listed below.

1) Thermophoresis in itself emerges as a more important mechanism of in-cloud scavenging than had been supposed previously. For the simplified in-cloud condition described in this paper it is approximately three times more effective than diffusiophoresis. Goldsmith and May (1966) calculate that a cloud droplet would have to grow and evaporate at least 100 times to account for observed scavenging rates, if diffusiophoresis were the primary mechanism. Including thermophoresis in the analysis reduces the number of cycles by approximately three and also indicates that the majority of submicron-size aerosol particle collection occurs during the evaporation phase, the opposite of what might be expected from previous considerations. Junge (1964) estimates for average precipitation rates of 6, 30, 60, 120 and 180 cm year^{-1} that the number of such cycles on each cloud condensation nuclei is 60, 30, 16, 9 and 7, respectively.

2) The mechanism postulated by Vittori and Prodi may be significant, although the findings of the present study would seem to reverse the visualization of the process. Because of the complexities of crystalization and energy absorption in a forming ice particle, it is extremely difficult to predict how important this mechanism might be.

3) Thermophoretic collection of particles by an evaporating droplet should be expected to combine small particles into larger ones, thereby changing their

properties as condensation nuclei and as particles in subsequent scavenging processes. Generally this should enhance scavenging.

4) Growth of large droplets at the expense of small ones (generally $<1\mu$) by virtue of the Kelvin effect may be significant in the promotion of thermophoretic deposition in much the same way as it is during occurrence of the Bergeron-Findeisen process and normal evaporation-condensation cycles.

5) Capture of aerosol particles by evaporating droplets during occurrence of the Bergeron-Findeisen process may result in contact ice nucleation of the droplet, which could alter significantly the nature of subsequent in-cloud behavior.

In conclusion it should be reemphasized that the underlying assumptions of this analysis, particularly with regard to the existence of isolated droplets in uniform aerosol-concentration fields, preclude its valid extension to predict quantitatively the amounts of scavenging that occur during a typical in-cloud situation. This weakness has been overcome to a certain extent by presenting the results as ratios of fluxes, thereby tending to cancel errors arising from these assumptions. In this regard we can conclude with some confidence that the predictions of appreciable thermophoretic scavenging under certain atmospheric conditions is a valid one, and that thermophoresis should receive more direct attention in future investigations of in- and below-cloud scavenging phenomena.

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