# Evaporation of Water Droplets by Radiation: Effect of Absorbing Inclusions

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# Abstract

Numerical solutions are obtained for transient response of water droplets to radiative heating. Results are presented for evaporation of polydispersed droplets in the solar radiation field. The evolution of the temperature and size of the droplet are determined. The results are discussed based on cloud droplet evaporation in the presence of absorbing inclusions.

Keywords: Cloud droplets; Evaporation; Radiation.

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# **INTRODUCTION**

The interaction between radiation and water droplets results in the attenuation of radiative energy by scattering and absorption. The absorbed energy consequently increases droplet temperature. If the temperature increase is significant, the equilibrium of water vapor with a droplet will be perturbed and the droplet begins to vaporize. At the same time, the change in the droplet size further alters the radiation transfer parameters. These processes are governed by the energy and vapor transport between droplets and the environment. This heating and subsequent evaporation of droplets in a radiation field are important in many areas, such as fog clearance and cloud processes (Ackerman *et al.*, 2000; Koren *et al.*, 2004). In this paper, radiative evaporation of water droplets containing soluble salt (NaCl) and a black-carbon core is presented. The effect of radiation absorption on the size distribution of polydispersed droplet is examined.

## METHODOLOGY

Mass and energy balance of a single water droplet in gas irradiated by a uniform radiation field is considered here. Single droplet equations are then expanded to obtain the mass and energy balance of polydispersed droplets. For an incompressible droplet of density  $\rho_D$  and constant heat capacity  $C_D$  the energy conservation equation (Davis and Brock, 1987) is:

$$C_{D} \frac{d(\pi D_{p}^{3} \rho_{D} T_{D} / 6)}{dt} = \pi D_{p}^{2} \left[ K_{g} \left(\frac{\partial T_{g}}{\partial t}\right)_{D_{p}} \right] + L \frac{dM}{dt} + \frac{\pi D_{p}^{2} Q_{abs} I_{0}}{4}$$
(1)

where  $T_D$  is the uniform droplet temperature,  $D_p$  is the droplet diameter,  $K_g$  is the heat conductivity,  $T_g$  gas temperature, t is the time, L is the heat of evaporation, M is the droplet mass,  $Q_{abs}$  is the radiation absorption efficiency and  $I_0$  is the uniform beam intensity. On the right hand side of Eq. (1), the first term is the energy loss due to conduction, while the second term is due to latent heat loss. The last term in Eq. (1) represents the heat gain by radiation absorption. In the above formulation the convective energy transfer is neglected. Assuming steady state conditions, flux and gradient terms in the Eq. (1) are given by (Williams, 1965; Sageev and Seinfeld, 1984)

$$\frac{dM}{dt} = 2\pi D_p \rho_g D_v \ln\left(\frac{1 - Y_\infty}{1 - Y_s(D_p, T_D)}\right) = J\pi D_p^2$$
(2a)

$$K_{g} \left(\frac{\partial T_{g}}{\partial t}\right)_{D_{p}} = \frac{J(T_{D} - T_{\infty})}{\exp\left[\frac{D_{p}JC_{a}}{2K_{g}}\right] - 1}$$
(2b)

where  $\rho_g$  is the density of the host gas,  $D_v$  is the diffusivity of the water vapor in the gas,  $C_a$  is the heat capacity of the host gas, J is the mass flux,  $Y_{\infty}$  and  $T_{\infty}$  are the water vapor mass fraction and temperature far away from the droplet, and  $Y_s$  is the saturation mass fraction at the droplet surface.

Neglecting the receding radius term, for low-rate heating Eq. (1) can be rearranged as:

$$\frac{dT_D}{dt} = \frac{6}{D_p \rho_D C_D} \left[ K_g \frac{\partial T_g}{\partial t} + LJ \right] + \frac{3Q_{abs} I_0}{2D_p \rho_D C_D}$$
(2c)

For a dilute solution droplet containing a non soluble core, the saturation mass fraction at the surface of droplet  $Y_s$  ( $D_p$ ) is (Pruppacher and Klett, 1978):

$$\ln\left(\frac{Y_s(D_p)}{Y_s}\right) = \frac{A}{D_p} - \frac{B}{\left(D_p^3 - D_u^3\right)}$$
(3a)

where

$$A = \frac{4M_{w}\sigma}{RT\rho_{w}}$$
(3b)

$$B = \frac{6S_n M_w}{\pi \rho_w}$$
(3c)

where  $\sigma$  is the droplet surface tension,  $\rho_w$  water density,  $S_n$  moles of solute dissolved in the droplet and  $D_u$  is the equivalent diameter of the insoluble fraction of the particle. The temperature dependence of  $Y_s(T_D)$  can be related to  $Y_s(T_{\infty})$  by using the Clausius-Clapeyron equation. Approximations for dilute solutions are used in the calculation of solute effect. The approximation of ideal solution, pertinent to Rault's law, deviates significantly from actual data on the behavior of concentrated solutions (Pruppacher and Klett, 1978). This is of concern for the solution droplets found in the final evaporation phase of solution droplets. Eq. (3a) neglects the non-continuum effects that may induce error in the small droplet (Knudsen number >> 1) growth estimation. The continuum equation is corrected by using Fuchs-Stutgin interpolation formula (Seinfeld and Pandis, 1998) for the transition regime.

The single droplet interaction with radiation and evaporation is extended to polydispersed droplets. The heating rate of the host gas which contains the droplet matrix per unit volume can be expressed as:

$$\frac{\partial T_{\infty}}{\partial t} = \frac{1}{\rho_g C_a} \left[ q_a + \int_0^\infty N(D_p) \pi D_p^2 K_g \left( \frac{\partial T}{\partial r} \right)_{D_p} dD_p \right]$$
(4)

where  $N(D_p)$  is the droplet size distribution,  $q_a$  is the absorption per unit volume by the host gas,  $\rho_g$  is the density of the host gas. Similarly, the water vapor mass fraction in the host gas:

$$\frac{\partial Y_{\infty}}{\partial t} = \frac{\rho_D \pi}{\rho_g 2} \int_0^\infty D_p^2 N(D_p) \frac{dD_p}{dt}$$
(5)

A sectional method with moving grid size structure was used to solve Eqs. (2) to (5) over droplet size distributions (Varghese and Gangamma, 2006). The solutions are determined by a public domain solver DVODE (Hindmarsh, 1983).

# NUMERICAL RESULTS AND DISCUSSIONS

#### Heating of single droplet

Intense irradiation of a pure water droplet in suspended air is considered here. Irradiation of polydispersed droplets by solar radiation is discussed in the next section. Irradiation is assumed uniform and constant. For this purpose, the last term of Eq. (2c) is held constant, and calculations are presented for  $3Q_{abs}I_0/2D_p$  equal to  $1 \times 10^5$  W/cm<sup>3</sup>. The physical parameters used in the calculation are given in Table 1. The droplet is assumed to be at equilibrium with the environment, initially. The time scales involved in the droplet evaporation under intense radiation is very small. Therefore, the variation of environmental temperature and vapor concentration are not considered for this calculation. The behavior of the droplet temperature and radius with time is given in Figs. 1 and 2. The evaporation behavior of a droplet comprising aqueous solution of salt (NaCl) is also presented in the figures. Initially, the temperature of the droplet is increased due to the absorption of radiation. This rise in temperature perturbs the equilibrium of the droplet with the environment, then evaporation of the droplet starts. At the same time, the loss of heat by evaporation and conduction reduces droplet temperature. These two effects oppose each other, resulting in the shape of the temperature curve. For pure droplet, the vaporization continues to reduce the size of the droplet and the process is finally driven by Kelvin effect. The presence of

salt in the droplet and the reduction in the size of the droplet increase the concentration of the solution, and thus reduce the saturation vapor pressure over the droplet. This effect opposes further reduction in the size of the droplet, and the droplet may find a new state of equilibrium in which the conductive heat flux compensates the radiative absorption. The normalized conductive and latent heat term of Eq. (2c) for an evaporating droplet with 0.0003 mass fraction (equivalent to 0.1  $\mu$ m dry diameter) of solute is given in Fig. 3. Initially, the latent heat term dominates and reaches pseudo-equilibrium with the heating rate. But the salt content in the droplet decreases the reduction of the size, thus the latent heat term. To reach equilibrium, the temperature of the droplet has to increase such that the conductive loss compensates the radiation absorption. This explains the upward limb of the temperature of droplets containing solute in the final stage of evaporation (Fig. 2).

Symbol	Value (unit)
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$T_{\infty}$	298.15 (K)
$Y^0$	0.02
L	584.0 (cal/g)
$C_a$	0.25 (cal/g/K)
$D_{v}$	$0.24 \ (cm^2/sec)$
$K_{g}$	$6.2 \times 10^{-5}$ (cal/cm/sec/K)
$ ho_{ m g}$	$1.2 \times 10^{-3} (g/cm^3)$
$\rho_D$	$1.0 (g/cm^3)$
σ	72.0 (dyne/cm)

Table 1. Physical constants for water droplet-Air system.

Absorption of solar radiation by polydispersed droplets suspended in air with absorbing inclusions is examined here. Although the transient part of Eq. (2c) may be neglected for a slow heating rate, the full formulation is used in the present calculations. The radiation absorption coefficient is obtained for a layered sphere, with absorbing material in the core (black carbon, BC) and in the water shell outside (Toon and Ackerman 1981). The refractive index is assumed constant over the wavelength. This may induce error in the absorption efficiency of BC. However, for simplicity, a constant refractive index of (1.96, 0.66) is assumed for BC. A constant intensity of solar radiation 600 W/m<sup>2</sup> is assumed for the calculation. The absorption efficiency is calculated for a wavelength of 16 grids ranging from 0.2 to 2.2  $\mu$ m, and accounting for a solar constant of 1307 W/m<sup>2</sup>. The absorption efficiency is weighted averaged over the solar insolation spectrum (Seinfeld and Pandis, 1998). The initial size distribution of the droplets is assumed as lognormal and has a number mean diameter (NMD) of 7.0  $\mu$ m and a GSD of 1.4. For present calculations, the size distribution is divided into five diameter bins. The number concentration of the droplet is assumed as 50 /cm<sup>3</sup>. In each size bin, the droplets contain a constant absorbing core

of diameter of 0.2  $\mu$ m of BC. The salt (NaCl) is a variable in the size bin. The largest bin is assumed to contain a salt mass equivalent to 0.2  $\mu$ m in diameter of the NaCl particle. The saturation vapor pressure of water for this droplet is calculated. By assuming that the droplets are in equilibrium with the above-calculated water vapor pressure, the salt content of the other droplets are determined from Eq. (3). This ensures that all the droplets are in equilibrium initially.



**Fig. 1.** Droplet diameter variation with time due to intense radiative heating. The initial droplet diameter is 1  $\mu$ m. The radiative absorption is 10<sup>5</sup> W/cm<sup>3</sup>. Solid line represents pure water droplet. Dotted line has an insoluble core diameter of 0.2  $\mu$ m and salt mass fraction of 3 × 10<sup>-4</sup>. The dashed line represents salt fraction of 2.5 × 10<sup>-3</sup> with 0.2  $\mu$ m core.



**Fig. 2.** Droplet temperature behavior with radiative heating. The initial droplet diameter is 1  $\mu$ m. The radiative absorption is 10<sup>5</sup> W/cm<sup>3</sup>. The solid line represents pure water droplet. The dotted line has an insoluble core diameter of 0.2  $\mu$ m and salt mass fraction of 3 × 10<sup>-4</sup>. The dashed line represents salt fraction of 2.5 × 10<sup>-3</sup> with 0.2  $\mu$ m core.



Fig. 3. Relative energy flux from the evaporating droplet. The droplet has an insoluble core diameter of 0.2 mm and soluble salt mass fraction of  $3 \times 10^{-4}$ .

#### Evolution of droplet spectra by radiative heating

The droplet size distribution evolution is shown in Figs. 4 and 5. The lognormal function is fitted over the size distribution at each time step. The figures are plotted using fitted lognormal distribution parameters. Initially, the droplet distribution widens (high GSD) and finally the polydispersity of the droplets decreases (low GSD) considerably. The NMD of the droplet is decreased from 7 to  $\sim 3 \,\mu m$ , due to evaporative loss by heating. The variation of the size distribution can be explained as follows. Small droplets heat up faster (see inverse relation to D<sub>p</sub> in Eq. 2c) than bigger droplets. Therefore, small droplets experience rapid evaporation, which results in decreased size. This initial evaporation of smaller droplets can cause initial increased polydispersity. But, later, the decrease in size of the bigger droplets decreases polydispersity. The evaporation of the smaller droplets stops due to two effects. One is the solute effect which reduces the water vapor pressure over the droplet surface (discussed in previous section). The second is an increase in ambient mass fraction of water vapor due to evaporation of bigger droplets. It is also possible that some of the small droplets can grow at the expense of bigger droplets due to the increase in the ambient vapor concentration. Finally, all the droplets tend to attain a conductive equilibrium with the ambient environment, which reduces the rate of droplet evaporation considerably.



Droplet diameter (µm)

**Fig. 4.** Evolution of water droplet size distribution in the presence of absorbing inclusion and soluble salt.



**Fig. 5.** Variation of water droplet number mean diameter (NMD) and geometric standard deviation (GSD) NMD with time.

#### CONCLUSIONS

The radiative evaporation of water droplets which contains absorbing material is presented. Results are presented for droplets with soluble salts interacting with the solar radiation spectrum. The absorption of solar energy by the droplets with absorbing inclusions significantly affects the equilibrium between water vapor and droplet. The results show that the droplet size spectrum is modified by the evaporation induced by absorbed energy. The initial mean diameter of the droplet distribution reduced to half of the initial value under a representative solar radiative heating. The polydispersity of the droplets are also found reduced with evaporation.

These results are important for cloud droplet size distribution evolution. Cloud albedo is a strong function of the polydispersity of cloud droplets. The dispersion of cloud droplet size has significant effect on earth's radiation balance (Liu and Daum, 2002; Peng and Lohmann, 2003). Even though the center core assumption of the inclusion may not accurately represent the absorption (Jacobson, 2006), the preliminary results show that the size and polydispersity of the droplets can be modified in the presence of absorbing material. Earlier studies on cloud droplet evaporation were made for cloud droplets immersed in high concentration of the absorbing aerosols (Ackerman *et al.*, 2000). Aerosols were treated as externally mixing with the droplets. These studies showed the effect on cloud lifetime due to increased heating in the presence of absorbing aerosols and consequently affect radiation balance of earth. Similar magnitude of

radiative forcing may be possible if the cloud droplets contain radiation absorbing compounds. There is also a possibility that the cloud droplet distribution can vary diurnally, heating and decreased polydispersity during the day, and cooling and increased polydispersity at night. Such calculations require inclusion of long wavelength radiation, accurate radiation transfer in the droplet field (multiple scattering), and other processes controlling the environmental variables. Such calculations are beyond the scope of this present investigation.

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