

Evaporation and Condensation of Large Droplets in the Presence of Inert Admixtures Containing Soluble Gas

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ABSTRACT

In this study the mutual influence of heat and mass transfer during gas absorption and evaporation or condensation on the surface of a stagnant droplet in the presence of inert admixtures containing noncondensable soluble gas is investigated numerically. The performed analysis is pertinent to slow droplet evaporation or condensation. The system of transient conjugate nonlinear energy and mass conservation equations was solved using anelastic approximation. Using the material balance at the droplet surface the authors obtained equations for Stefan velocity and the rate of change of the droplet radius taking into account the effect of soluble gas absorption at the gas–liquid interface. The authors also derived boundary conditions at gas–liquid interface taking into account the effect of nonisothermal gas absorption. It is demonstrated that the average concentration of the dissolved species in a droplet strongly depends on the relative humidity (RH) for highly soluble and for slightly soluble gaseous atmospheric pollutants. Therewith the difference between the average concentration of the dissolved species in water droplets attains tens of percent for different values of RH.

1. Introduction

Wet removal of atmospheric polluted gases by cloud droplets is involved in many atmospheric processes such as polluted gases scavenging, cloud microphysics, etc. Heat and mass transfer during gas absorption by liquid droplets and during droplets evaporation and vapor condensation on the surface of liquid droplets is important in various fields of modern environmental engineering and atmospheric science. Clouds represent an important element in self-cleansing process of the atmosphere (Flossmann 1998). The consequence for the aerosol climate forcing is that the cooling can be intensified with increasing atmospheric amount of water-soluble trace gases such as NH_3 and SO_2 , counteracting the warming effect of the greenhouse gases (Krämer et al. 2000). All these phenomena involve evaporation of droplets suspended in a multicomponent gaseous mixture. Scavenging of atmospheric pol-

luted gases by cloud droplets is a result of gas absorption mechanism (Pruppacher and Klett 1997; Flossmann 1998). Sources of soluble gases presented in the atmosphere are briefly reviewed by Macdonald et al. (2004), Sutton et al. (1995), Fraser and Cass (1998), Van der Hoek (1998), and Elperin and Fominykh (2005).

Gas scavenging by atmospheric water droplets includes absorption of SO_2 , CO_2 , and NH_3 , and some other gases. Presence of soluble gas in the atmosphere can affect the dynamics of evaporation and condensation of water droplets in an atmospheric cloud. Average concentrations of CO_2 , NH_3 , and SO_2 in the atmosphere can be found, for example, in Seinfeld (1986) and Liu and Lipták (1999). Comprehensive study of coupled heat and mass transfer during gas absorption by liquid droplets and droplets evaporation and growth is a necessary step in an adequate predicting of atmospheric changes under the influence of hazardous gases.

Different facets of the problem of evaporation of droplets in the flowing or stagnant gases were discussed in numerous theoretical and experimental studies, and several comprehensive reviews are available (see Sirignano 1993; Chiu 2000).

Soluble gas absorption by noncirculating droplets

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was investigated experimentally by Hixson and Scott (1935), Bosworth (1946), and Taniguchi and Asano (1992). Conditions of noncirculation for falling liquid droplets were achieved by using high-viscosity liquids in experiments of Hixson and Scott (1935) and Bosworth (1946) or by using small water droplets in the experiments of Taniguchi and Asano (1992). In these experimental studies droplet diameter was larger than 1 mm. Taniguchi and Asano (1992) used water droplets with Sauter mean diameter equal to 0.185, 0.148, and 0.137 mm in experiments with CO_2 absorption. Bosworth (1946) compared experimental results with error function solution of nonstationary equation of diffusion written in spherical coordinates for a liquid droplet. Taniguchi and Asano (1992) correlated experimental results with Newman's solution (see Newman 1931).

Dispersed-phase controlled isothermal absorption of a pure gas by stagnant nonevaporating liquid droplet was investigated analytically by Newman (1931), and gas absorption in the presence of inert admixtures when both phases affect mass transfer was analyzed by Plocker and Schmidt-Traub (1972). Liquid and gaseous phase controlled mass transfer during soluble gas absorption in the presence of inert admixtures by a stagnant nonevaporating liquid droplet was studied also by Chen (2002) by solving the coupled time-dependent diffusion equations for gas and liquid phases. Vesala et al. (2001) solved the problem of trace gas uptake by droplets under nonequilibrium conditions numerically and analytically and derived simple formulas for the gas uptake coefficient. State of the art in gas absorption by spheroidal water droplets is presented by Amokrane and Caussade (1999).

Effect of vapor condensation at the surface of stagnant droplets on the rate of mass transfer during gas absorption by growing droplets was investigated theoretically by Karamchandani et al. (1984), Ray et al. (1987), and Huckaby and Ray (1989). In particular the effect of vapor saturation ratio on sulfur S(IV) accumulation during sulfur dioxide absorption by evaporating (growing) droplets was specified by Huckaby and Ray (1989). Liquid-phase controlled mass transfer during absorption was investigated by Karamchandani et al. (1984) and Ray et al. (1987) in the case when the system consists of liquid droplet, its vapor, and soluble gas. Droplet growth was assumed to be solely controlled by vapor condensation, and influence of gas absorption on the rate of vapor condensation was neglected. The assumption of uniform temperature distribution in both phases was used by Karamchandani et al. (1984). The results of Karamchandani et al. (1984) indicate that for fast droplet growth rates, induced by relatively high degrees of supersaturation, the absorp-

tion rate is significantly enhanced, leading to higher solute concentration near the surface than that observed for nongrowing droplets. Ray et al. (1987) analyzed gas absorption by a stationary growing droplet in a stagnant supersaturated medium by solving the coupled nonstationary mass and energy balance equations for the gaseous and liquid phases. The obtained results showed that in the case when heats of condensation are low, the droplet growth can increase significantly the rate of absorption. For high heats of condensation, the rapid increase of surface temperature induced by high growth rates can significantly reduce absorption rates for those gases whose solubility decreases with temperature increase. Huckaby and Ray (1989) employed the approach suggested by Ray et al. (1987) to investigate a system consisting of a liquid droplet, its vapor, soluble and inert gases. Diffusion resistances in both phases during gas absorption were taken into account. Numerical calculations were performed for water droplet evaporating (growing) in gaseous mixture containing sulfur dioxide (SO_2). It should be emphasized that in the above mentioned studies (see Karamchandani et al. 1984; Ray et al. 1987; Huckaby and Ray 1989), only the influence of condensation on the rate of gas absorption by stagnant liquid droplets was investigated.

In the present study we investigate the interrelated effects of gas absorption and evaporation (condensation) at the surface of a droplet on transient heat and mass transfer in the presence of inert gases. We also took into account the following effects that were neglected in all previous studies: (i) effect of gas absorption on Stefan velocity, and (ii) thermal effect of absorption on droplet evaporation or condensation. In contrast to previous studies (see, e.g., Karamchandani et al. 1984; Ray et al. 1987; Huckaby and Ray 1989) in our calculations we considered the molecular transport coefficients of the gaseous phase as functions of temperature and concentrations of gaseous species. Consequently the suggested model can be used over a wide range of parameters such as temperature, relative humidity, etc.

2. Mathematical model

a. Governing equations

Consider a spherical droplet with the initial radius R_0 immersed in a stagnant gaseous mixture with temperature $T_{e,\infty}$. The gaseous mixture containing K components includes noncondensable soluble species that is absorbed into the liquid. The first component of the gaseous mixture is formed by the molecules of the volatile species of a droplet. The soluble species is absorbed

into the liquid, and other $K - 2$ components do not undergo phase transition at the droplet surface. The droplet is heated by a conduction heat flux from the high temperature surroundings and begins to evaporate. In the further analysis we assume spherical symmetry and neglect effects of buoyancy and thermal diffusion. Under these assumptions, the spherically symmetric system of mass and energy conservation equations for the liquid phase $0 < r < R(t)$ reads

$$r^2 \frac{\partial T^{(L)}}{\partial t} = \alpha_L \frac{\partial}{\partial r} \left(r^2 \frac{\partial T^{(L)}}{\partial r} \right), \quad (1)$$

$$r^2 \frac{\partial Y_A^{(L)}}{\partial t} = D_L \frac{\partial}{\partial r} \left(r^2 \frac{\partial Y_A^{(L)}}{\partial r} \right). \quad (2)$$

In the surrounding gaseous medium, $r > R(t)$, the mass, species, and energy conservation equations read

$$r^2 \frac{\partial \rho}{\partial t} + \frac{\partial}{\partial r} (r^2 \rho v_r) = 0, \quad (3)$$

$$r^2 \frac{\partial}{\partial t} (\rho Y_j) + \frac{\partial}{\partial r} (\rho v_r r^2 Y_j) = \frac{\partial}{\partial r} \left(\rho D_j r^2 \frac{\partial Y_j}{\partial r} \right), \quad (4)$$

$$r^2 \frac{\partial (\rho c_p T_e)}{\partial t} + \frac{\partial}{\partial r} (\rho v_r r^2 c_p T_e) = \frac{\partial}{\partial r} \left(k_e r^2 \frac{\partial T_e}{\partial r} \right). \quad (5)$$

In Eqs. (1)–(5) ρ is the gas density, j is the number of gaseous phase species (subscript 1 denotes the volatile species, $j = 1, \dots, K - 1, j \neq A^1$), Y_j and M_j are the mass fraction and the molar mass of the j th gaseous species, $Y_A^{(L)}$ is the mass fraction of the absorbate in the liquid, D_j is the diffusion coefficient, $D_j = (1 - X_j)/\sum_{k \neq j} (X_k/D_{jk})$, X_j is the mole fraction of the j th species, and D_{jk} is the binary diffusion coefficient for species j and k .

Estimation of the characteristic values of the terms $\partial \rho / \partial t$ and $\nabla(\rho \mathbf{v})$ in the continuity Eq. (3) shows that in the considered problem $v^2/c^2 \ll 1$, where v is the gas velocity and c is the speed of sound. Therefore for the solution of the system of energy and mass conservation equations instead of Eq. (3) we can use anelastic approximation:

$$\frac{\partial}{\partial r} (r^2 \rho v_r) = 0. \quad (6)$$

The radial flow velocity v_r can be obtained by integrating Eq. (6)

$$\rho v_r r^2 = \text{const}. \quad (7)$$

¹ In general, Eqs. (4) are written for all $j = 1, \dots, K$. However, only $K - 1$ of the values D_j can be specified independently. Therefore the K th species can be treated differently from the others and may be found consistently using the identity $\sum_{j=1}^K Y_j = 1$.

The system of energy and mass conservation Eqs. (3)–(5) must be supplemented with the momentum conservation equation. However, in the case of small flow velocities the pressure gradient is negligibly small ($\Delta p \sim \rho v^2$), and the pressure can be assumed constant. The gaseous phase properties can be related through the ideal gas equation of state:

$$p = p_\infty = \rho R_g T_e \sum_{j=1}^K \left(\frac{Y_j}{M_j} \right), \quad (8)$$

where R_g is the universal gas constant, p_∞ is the gaseous mixture pressure far from the droplet.

b. Stefan velocity and droplet vaporization rate

Equations (1)–(5) must be supplemented with equation for determining gas flow velocity. Consider the case when effect of droplet volumetric expansion is negligible. The continuity condition for the radial flux of the absorbate at the droplet surface reads

$$\begin{aligned} j_A|_{r=R} &= \rho Y_A v_s - D_{A\rho} \frac{\partial Y_A}{\partial r} \Big|_{r=R_+} \\ &= -D_L \rho_L \frac{\partial Y_A^{(L)}}{\partial r} \Big|_{r=R_-}, \end{aligned} \quad (9)$$

where the signs “+” and “−” denote values at the external and internal surfaces, respectively. Since other $K - 2$ nonsoluble components of the inert admixtures are not absorbed in the liquid the integral fluxes vanish; that is, $J_j = 4\pi R^2 j_j = 0$, ($j \neq 1, j \neq A$), and

$$j_j = \rho Y_j v_s - D_j \rho \frac{\partial Y_j}{\partial r} \Big|_{r=R_+} = 0, \quad (j \neq 1, j \neq A). \quad (10)$$

Taking into account this condition and using Eq. (7) we can obtain the expression for Stefan velocity:

$$v_s = - \frac{D_L \rho_L}{\rho(1 - Y_1)} \frac{\partial Y_A^{(L)}}{\partial r} \Big|_{r=R_-} - \frac{D_1}{(1 - Y_1)} \frac{\partial Y_1}{\partial r} \Big|_{r=R_+}. \quad (11)$$

The material balance at the gas–liquid interface reads

$$\frac{dm_L}{dt} = -4\pi R^2 \rho_s [v(R, t) - \dot{R}]. \quad (12)$$

Equation (12) yields

$$v_s = \frac{dR}{dt} \left(1 - \frac{\rho_L}{\rho_s} \right). \quad (13)$$

Substituting Eq. (13) into Eq. (11) and assuming that $\rho_L \gg \rho$ we obtain the following expression for the rate of change of droplet's radius:

$$\dot{R} = \frac{D_L}{(1 - Y_1)} \frac{\partial Y_A^{(L)}}{\partial r} \Big|_{r=R_-} + \frac{\rho D_1}{\rho_L (1 - Y_1)} \frac{\partial Y_1}{\partial r} \Big|_{r=R_+}. \quad (14)$$

As can be seen from Eqs. (11) and (14) in the case when all inert admixtures are not absorbed in the liquid, we obtain the following expressions for Stefan velocity and rate of change of droplet's radius:

$$v_s = - \frac{D_1}{(1 - Y_1)} \frac{\partial Y_1}{\partial r} \Big|_{r=R_+}, \quad (15)$$

$$\dot{R} = \frac{\rho D_1}{\rho_L (1 - Y_1)} \frac{\partial Y_1}{\partial r} \Big|_{r=R_+}. \quad (16)$$

Expressions (11), (14) imply that the absorption of soluble admixture decreases Stefan's velocity and evaporation rate. However, since absorption is accompanied by thermal effect and solubility of different gases in a liquid strongly varies with temperature, the influence of gas absorption on the rate of evaporation is quite involved.

c. Initial and boundary conditions

The system of conservation Eqs. (1)–(5) must be supplemented by initial conditions and the boundary conditions at the droplet surface. The initial conditions for the system of Eqs. (1)–(5) read

$$\text{At } t = 0, \quad 0 \leq r \leq R_0, \quad T^{(L)} = T_0^{(L)}, \quad Y_A^{(L)} = Y_{A,0}^{(L)}, \quad (17)$$

$$\text{At } t = 0, \quad r \geq R_0, \quad T_e = T_{e,0}(r), \quad Y_j = Y_{j,0}(r).$$

At the droplet surface the continuity conditions for the radial flux of nonsoluble gaseous species yield

$$D_j \rho \frac{\partial Y_j}{\partial r} \Big|_{r=R_+} = \rho Y_j v|_{r=R_+}. \quad (18)$$

For the absorbate this condition assumes the form of Eq. (9). The vapor concentration at the droplet surface $Y_{1,s}(R, t)$ is the function of temperature $T_s(t)$ and can be determined as follows:

$$Y_{1,s}(R, t) = Y_{1,s}(T_s) = \frac{\rho_{1,s}}{\rho} = \frac{p_{1,s}(T_s)M_1}{p_\infty M}, \quad (19)$$

where M_1 and M are the molar mass of volatile species and gaseous mixture, respectively. Partial pressure at

TABLE 1. The coefficients a_i used in Eq. (20).

Coefficient	a_1	a_2	a_3	a_4
Value	-7.764 51	1.458 38	-2.775 80	-1.233 03

the droplet surface $p_{1,s}$ is determined by the following equation (see Reid et al. 1987):

$$\ln(p_{1,s}/p_c) = (1 - \Theta)^{-1}(a_1\Theta + a_2\Theta^{1.5} + a_3\Theta^3 + a_4\Theta^6), \quad (20)$$

where $\Theta = 1 - T_s/T_c$ is the critical temperature, and p_c is the critical pressure, and the values of the coefficients a_i are presented in Table 1.

The droplet temperature can be found from the following equation:

$$k_e \frac{\partial T_e}{\partial r} \Big|_{r=R_+} + \rho_L L_v \frac{dR}{dt} = k_L \frac{\partial T^{(L)}}{\partial r} \Big|_{r=R_-} - L_a \rho_L D_L \frac{\partial Y_A^{(L)}}{\partial r} \Big|_{r=R_-}. \quad (21)$$

The last term in the right-hand side of Eq. (21) arises due to a heat released at the gas–liquid interface during gas absorption in liquid (see, e.g., Elperin and Fominykh 2003). The equilibrium between soluble gaseous and dissolved in liquid species can be expressed using the Henry's law. Consequently the boundary condition for the Eq. (2) reads

$$C_A = H_A p_A, \quad (22)$$

where C_A is the molar concentration of the species dissolved in liquid, H_A is the Henry's law coefficient, and p_A is the partial pressure of species A in the gas. The functional dependence of the Henry's law constant versus temperature reads

$$\ln \frac{H_A(T_0)}{H_A(T)} = \frac{\Delta H}{R_G} \left(\frac{1}{T} - \frac{1}{T_0} \right), \quad (23)$$

where ΔH is the enthalpy change due to transfer of soluble species A from the gaseous phase to liquid, R_G is the gas constant. The dependencies of Henry's law constant on temperature for aqueous solutions of different soluble gases (ammonia, carbon dioxide, and sulfur dioxide) are shown in Fig. 1. Inspection of Fig. 1 shows that Henry's law constant strongly varies with temperature and hence the dependence of Henry's constant on temperature must be taken into account.

At the gas–liquid interface

$$T_e = T^{(L)}. \quad (24)$$

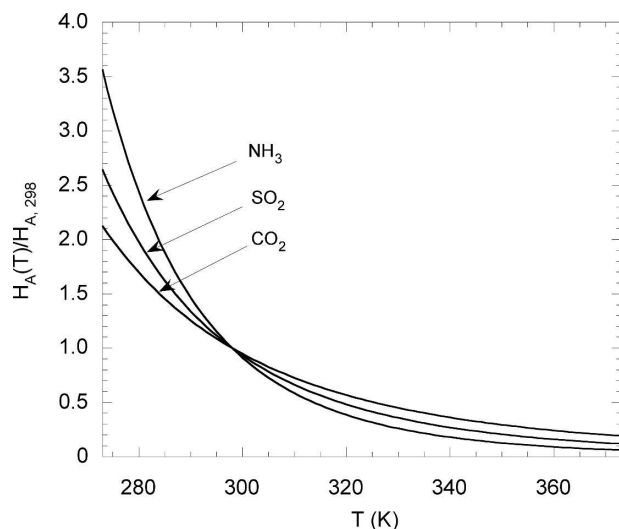


FIG. 1. Henry's law constant for aqueous solutions of different soluble gases vs temperature.

In the center of the droplet the symmetry conditions yields

$$\left. \frac{\partial Y_A^{(L)}}{\partial r} \right|_{r=0} = 0, \quad \left. \frac{\partial T^{(L)}}{\partial r} \right|_{r=0} = 0. \quad (25)$$

At $r \rightarrow \infty$ and $t > 0$ the soft boundary conditions at infinity are imposed:

$$\left. \frac{\partial T_e}{\partial r} \right|_{r \rightarrow \infty} = 0, \quad \left. \frac{\partial Y_j}{\partial r} \right|_{r \rightarrow \infty} = 0. \quad (26)$$

3. Method of numerical solution

The presence of two computational domains [$0 \leq r \leq R(t)$ for the liquid phase, and $r \geq R(t)$ for the gaseous phase] for the system of Eqs. (1)–(5), which are separated by a moving boundary $r = R(t)$ complicates the numerical solution. Moreover the characteristic times of the heat and mass transfer in the gaseous phase and heat transfer in the liquid phase are much less than the characteristic time of the diffusion of soluble component in liquid. To overcome these problems we introduced the following dimensionless time variable:

$$\tau = \frac{D_L t}{R_0^2}. \quad (27)$$

Then the governing equations can be rewritten for time variable τ using the following coordinate transformations (see, e.g., Ray et al. 1987):

$$x = 1 - \frac{r}{R(t)}, \quad (28)$$

for the domain $0 \leq r \leq R(t)$, and

$$w = \frac{1}{\sigma} \left[\frac{r}{R(t)} - 1 \right] \quad (29)$$

for the domain $r \geq R(t)$. The parameter σ was chosen such that the concentrations of the gaseous species and the gas phase temperature achieved their initial values at the distance $\sigma R(t)$.

In the transformed computational domains the coordinates $x \in [0, 1]$, $w \in [0, 1]$, and can be treated identically in numerical calculations. The gas–liquid interface is located at $x = w = 0$. The system of nonlinear parabolic partial differential Eqs. (1)–(5) was solved using the method of lines developed by Sincovec and Madsen (1975). The spatial discretization on a three-point stencil was used in order to reduce the system of the time-dependent partial differential equations to a semidiscrete approximating system of coupled ordinary differential equations. Thus the system of partial parabolic differential equations is approximated by a system of ordinary differential equations in time for functions T_e , $T^{(L)}$, $Y_A^{(L)}$, and Y_j at the mesh points. The mesh points were spaced adaptively using the following formula:

$$x_i = \left(\frac{i-1}{N} \right)^n, \quad i = 1, \dots, N+1, \quad (30)$$

so that they cluster near the left boundary where the gradients are steep. In Eq. (30) N is the chosen number of mesh points, n is an integer coefficient (in our calculations n is chosen equal to 3). The resulting system of ordinary differential equations was solved using a backward differentiation method. Generally, in the numerical solution, 151 mesh points and an error tolerance $\sim 10^{-5}$ in time integration were employed. Variable time steps were used to improve the computing accuracy and efficiency.

During numerical solution of the system of Eqs. (9)–(10) the properties ρ , c_p , D_j , and k_e were evaluated simultaneously at each grid point at each time step. The compilation of the formulas for calculating these properties is presented by Reid et al. (1987), Ben-Dor et al. (2003a,b). Properties of the liquid and the gaseous phases were adopted from Reid et al. (1987). Calculations were terminated when the condition $R/R_0 \leq 0.1$ was fulfilled.

4. Results and discussion

The above model of droplet evaporation in the presence of inert admixtures containing noncondensable

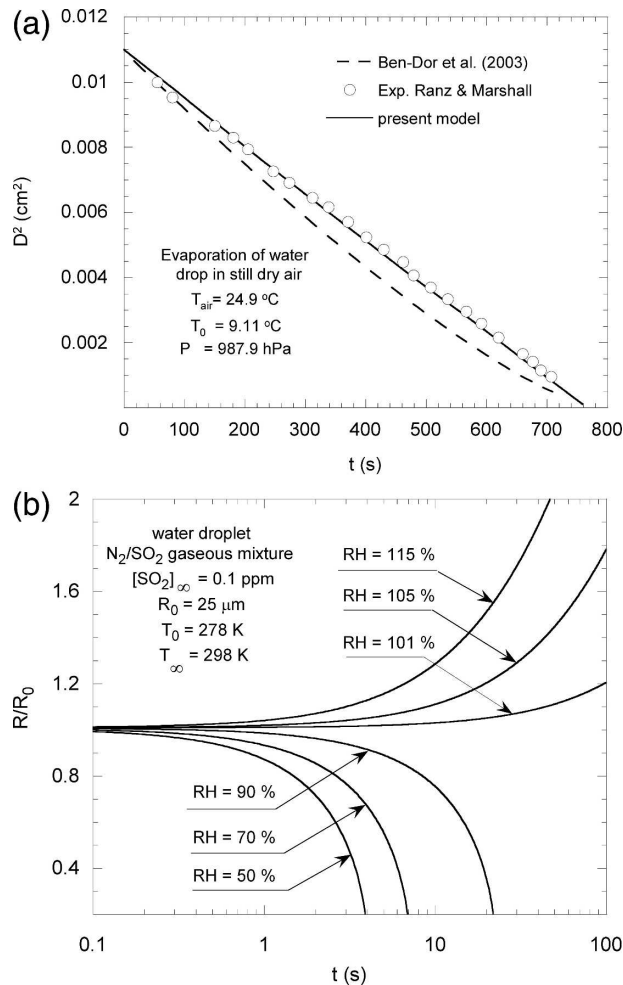


FIG. 2. (a) Temporal evolution of the radius of the evaporating water droplet in dry still air. Solid line is the present model, dashed line is the nonconjugate model (Ben-Dor et al. 2003a), circles are experimental data (Ranz and Marshall 1952). (b) Temporal evolution of droplet radius for a droplet suspended in $N_2/SO_2/H_2O$ gaseous mixture with $T_0 = 278$ K, $T_{\infty} = 298$ K, $R_0 = 25$ μm , $[SO_2]_{\infty} = 0.1$ ppm.

soluble gas was applied to study evaporation of water droplet immersed in a stagnant gaseous mixture composed of the ternary combinations of nitrogen (N_2), soluble ammonia (NH_3), soluble carbon dioxide (CO_2), sulfur dioxide (SO_2), and vapor of the water droplet.

For the purpose of validation of a computer code the present theoretical model was compared with the experimental results obtained by Ranz and Marshall (1952), and with our previous model developed for moderately large droplets which assumed uniform temperature distribution inside the droplet (Ben-Dor et al. 2003a). The dependence of droplet-squared diameter versus time for stagnant water droplet evaporating in a still dry air is shown in Fig. 2a. In these calculations the

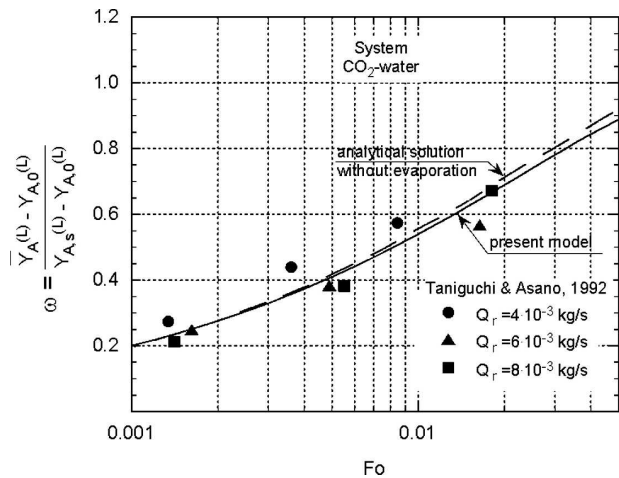


FIG. 3. Comparison of the calculated dimensionless average aqueous concentration with the experimental data (Taniguchi and Asano 1992) and analytical solution for stagnant nonevaporating droplet.

effect of gas absorption was excluded. In this plot the solid line indicates theoretical results obtained using the present model, the dots indicate experimental results and the dashed line present the results obtained in Ben-Dor et al. (2003a). The model suggested by Ben-Dor et al. (2003a) was developed for the case of small droplet Biot number when the temperature distribution inside the liquid droplet can be assumed to be uniform. As can be seen from this plot the present model demonstrates better agreement with experimental results of Ranz and Marshall (1952).

Using the present model and taking into account gas absorption we calculated the dependence of dimensionless droplet radius R/R_0 versus time for evaporating as well as growing water droplets with the initial radii 25 μm immersed into a gaseous mixture containing soluble gas (sulfur dioxide; see Fig. 2b) and different concentrations of water vapor. The curves were plotted for water droplet with the initial temperature 278 K suspended in $N_2/SO_2/H_2O$ gaseous mixture with ambient temperature equal to 298 K and concentration of sulfur dioxide of 0.1 ppm. The value of relative humidity varied in the range from 50% to 115%. The concentration of sulfur dioxide equal to 0.1 ppm is pertinent to highly polluted atmosphere.

In the following calculations we accounted for the effect of gas absorption during droplet evaporation. The results obtained using the above described theoretical model were compared with the experimental results by Taniguchi and Asano (1992). The dependence of the relative dissolved carbon dioxide concentration in a droplet versus Fourier number is shown in Fig. 3.

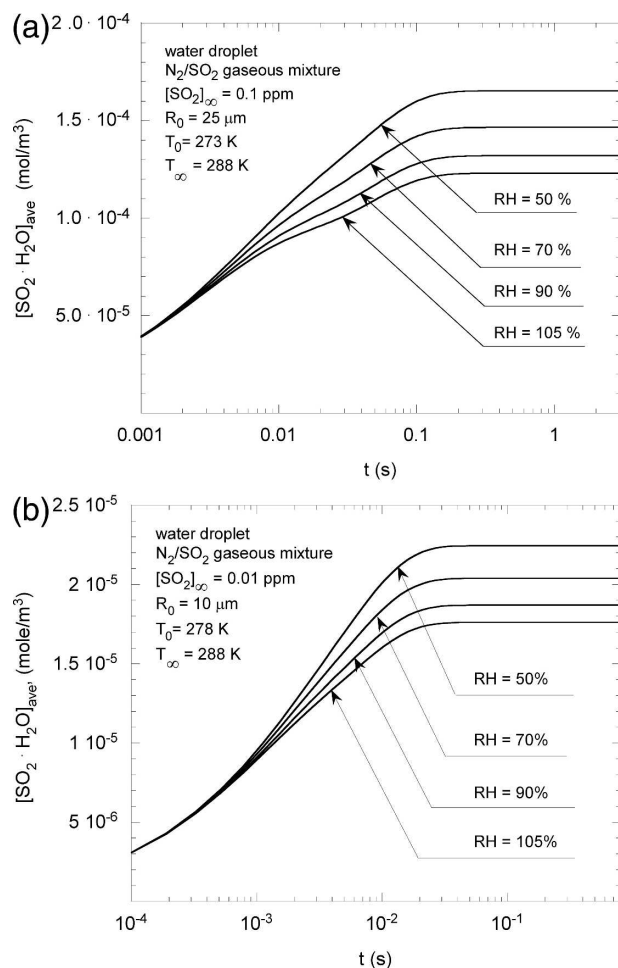


FIG. 4. (a) Temporal evolution of the average aqueous SO_2 molar concentration for various values of relative humidity ($[\text{SO}_2]_{\infty} = 0.1 \text{ ppm}$). (b) Temporal evolution of the average aqueous SO_2 molar concentration for various values of relative humidity ($[\text{SO}_2]_{\infty} = 0.01 \text{ ppm}$).

The relative absorbate concentration is determined as follows:

$$\omega = \frac{\bar{Y}_A^{(L)} - Y_{A,0}^{(L)}}{Y_{A,s}^{(L)} - Y_{A,0}^{(L)}}, \quad (31)$$

where $\bar{Y}_A^{(L)}$ is the average concentration of the absorbed CO_2 in the droplet:

$$\bar{Y}_A^{(L)} = \frac{1}{V_d} \int Y_A^{(L)}(r) r^2 \sin \vartheta \, dr \, d\vartheta \, d\varphi. \quad (32)$$

In Eq. (32) r , θ , and φ are spherical coordinates. Taniguchi and Asano (1992) performed measurements for the CO_2 -water spray system with the local mass flow rates of liquid in the range from $4 \times 10^{-3} \text{ kg s}^{-1}$ to $8 \times 10^{-3} \text{ kg s}^{-1}$. The dashed line in the figure repre-

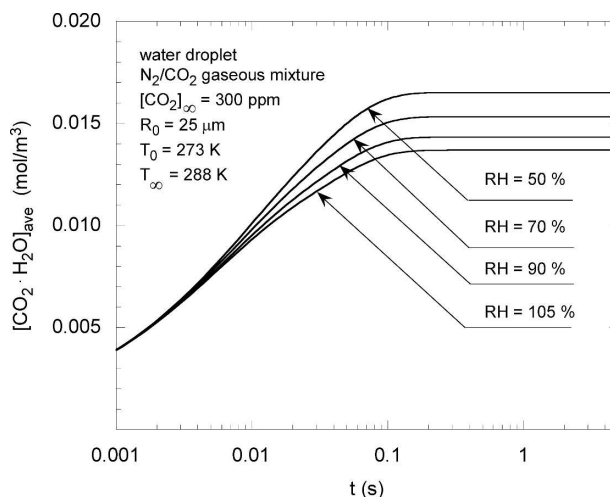


FIG. 5. Temporal evolution of the average aqueous CO_2 molar concentration for various values of relative humidity.

sents the analytical solution in the case of aqueous-phase controlled diffusion of carbon dioxide in a stagnant nonevaporating droplet (see, e.g., Seinfeld 1986):

$$\omega = 1 - \frac{6}{\pi^2} \sum_{n=1}^{\infty} \frac{1}{n^2} \exp(-4\pi^2 n^2 \text{Fo}). \quad (33)$$

As can be seen from Fig. 3 for small Fourier numbers the obtained results show good agreement with the experimental data obtained by Taniguchi and Asano (1992) and analytical solution (33).

The typical concentration of carbon dioxide in atmosphere varies in the range from 300 to 360 ppm (Liu and Lipták 1997) while concentration of sulfur dioxide varies from 0.01 ppb at the ground level in clean air to 0.2 ppm and higher in a polluted air (Liu and Lipták 1997; Seinfeld 1986). The dependence of average aqueous SO_2 and CO_2 molar concentration versus time is shown in Figs. 4a,b and 5. The curves were plotted for different values of the relative humidity (RH). The calculations presented in Fig. 4a were conducted for a large droplet immersed into a highly polluted atmosphere with the concentration of SO_2 equal to 0.1 ppm (Seinfeld 1986). Numerical results shown in Fig. 4b are typical for cleaner atmosphere (10 ppb of SO_2). As can be seen from these plots the average concentration of the dissolved species increases with decrease of RH. Since the solubility and Henry's constant of the dissolved admixture increase with temperature decrease, the evaporating droplet with lower surface temperature can absorb a higher amount of the dissolved species.

The dependence of the relative concentrations of the dissolved carbon dioxide and sulfur dioxide, ω , in a droplet versus time is shown in Figs. 6–7. The curves are plotted for droplets with different radii evaporating

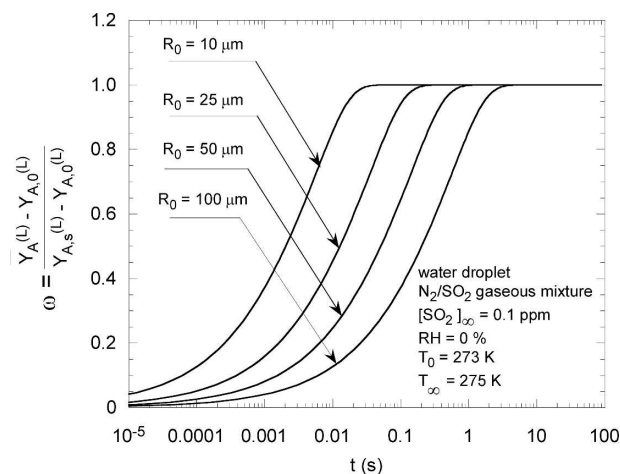


FIG. 6. Temporal evolution of dimensionless average aqueous SO_2 concentration (dry ambient gaseous mixture) for various initial sizes of evaporating droplet R_0 .

into the ambient N_2/SO_2 and N_2/CO_2 gaseous mixtures with $\text{RH} = 0$. In calculations the concentrations of sulfur dioxide and carbon dioxide were set equal to 0.1 and 300 ppm, respectively (see, e.g., Pruppacher and Klett 1997; Gravenhorst et al. 1978). These values of SO_2 and CO_2 concentrations are typical for atmospheric conditions. Inspection of these plots shows that for a droplet with the radius 10 μm the saturation is reached after ~ 0.01 – 0.02 s while several seconds are required for the droplet with the radius 100 μm . We performed calculations for evaporating and growing droplets. The dependencies of the normalized concentration of the dissolved CO_2 versus time for the case of droplet evaporation into the dry gaseous mixture and for the case of

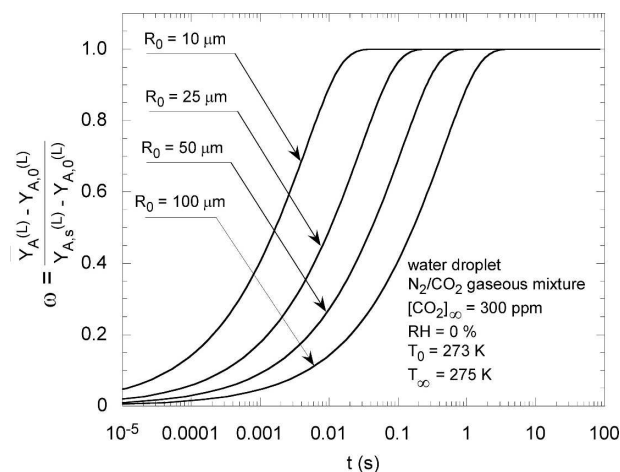


FIG. 7. Temporal evolution of the dimensionless average aqueous CO_2 concentration (dry ambient gaseous mixture) for various initial sizes of evaporating droplet R_0 .

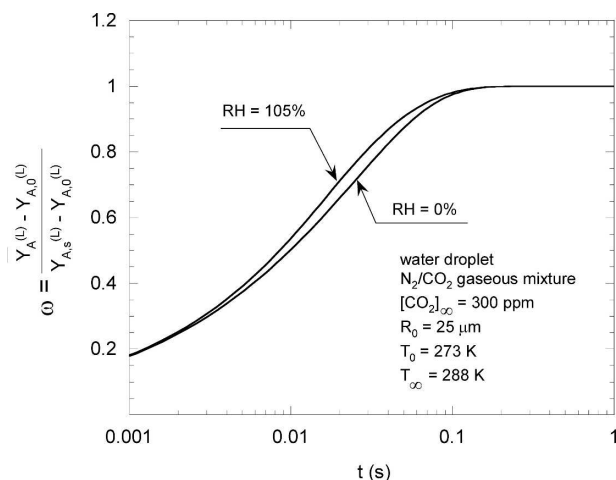


FIG. 8. Temporal evolution of the dimensionless average aqueous CO_2 concentration ($R_0 = 25$ μm).

droplet condensation in gaseous mixture with $\text{RH} = 105\%$ are shown in Fig. 8. The calculations were performed for a water droplet with the radius of 25 μm evaporating in N_2/CO_2 gaseous mixture with a given carbon dioxide concentration (300 ppm) at infinity. The obtained results show that saturation is reached more rapidly in the case of droplet condensation. The latter phenomenon is associated with the magnitude of droplet surface temperature. During condensation droplet temperature increases while during evaporation droplet surface temperature decreases. Since Henry's law constant depends exponentially on temperature, the increase of the surface temperature strongly affects equilibrium aqueous phase concentration. Therefore in the case of droplet condensation the aqueous phase saturation is reached faster although the droplet volume increases.

It was emphasized above that Henry's law constant and, consequently, concentration of the dissolved species at the droplet surface strongly depend on temperature. Therefore droplet surface temperature is an important parameter for investigating gas absorption by evaporating (growing) droplets. Figure 9 shows the droplet surface temperature as a function of time. The calculations were performed for water droplet with the initial radius 100 μm evaporating in N_2/CO_2 gaseous mixture with the different initial values of carbon dioxide mass fraction (from 0.1 to 1.0) far from the droplet. Water vapor appears in the gaseous phase due to droplet evaporation, and it constitutes the third species of the ambient gaseous mixture. It is assumed that initially the mass fraction of water vapor outside the droplet is equal to zero. Inspection of Fig. 9 shows that in the case of large concentrations of carbon dioxide during the

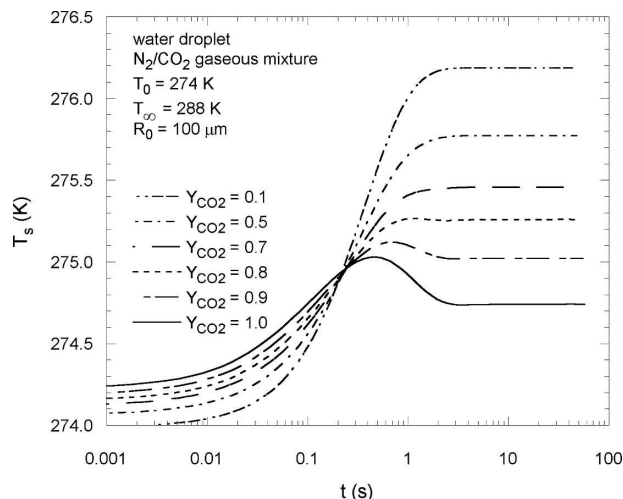


FIG. 9. Temporal evolution of the surface temperature for a water droplet evaporating in $N_2/CO_2/H_2O$ gaseous mixture, $R_0 = 10 \mu m$, $T_0 = 274 K$, $T_{e,\infty} = 288 K$ for various ambient concentrations of CO_2 .

transient period of droplet evaporation the droplet surface temperature as a function of time passes through the maximum. This well-pronounced nonlinear behavior of the droplet surface temperature stems from the interaction of several different phenomena. In particular, thermal effect of dissolution and Stefan flow are the cause of the maximum of droplet surface temperature during the transient period of droplet evaporation (see Fig. 10). As can be seen from Fig. 10 neglecting these effects we obtain the result that is typical for droplet evaporation in gaseous mixture containing nonsoluble gaseous species. Calculations were also performed for water droplets evaporating in N_2/SO_2 and N_2/NH_3 gaseous mixture (Figs. 11 and 12). As can be seen from Figs. 10–12 in the case of CO_2 dissolution thermal effect of dissolution is essential for large concentrations of carbon dioxide in the ambient gaseous mixture while in the case of SO_2 and NH_3 dissolution this effect is essential for significantly smaller concentrations of SO_2 in the gaseous phase. Inspection of Fig. 11 shows that in the case of NH_3 dissolution the thermal effect is detectable when ammonia concentration is of the order of 10^3 ppm. The essential differences in surface temperature at steady-state stage in the case of water droplet evaporation into N_2/CO_2 gaseous mixture (see Fig. 9) are associated with large differences between heat and mass transport coefficients for carbon dioxide and nitrogen. Figure 13 shows the droplet surface temperature as a function of time for droplets with the radius $100 \mu m$ growing in $N_2/CO_2/H_2O$ gaseous mixture ($Y_{H_2O,\infty} = 0.011$). The curves were plotted for water droplets growing in $N_2/CO_2/H_2O$ gaseous mixture with

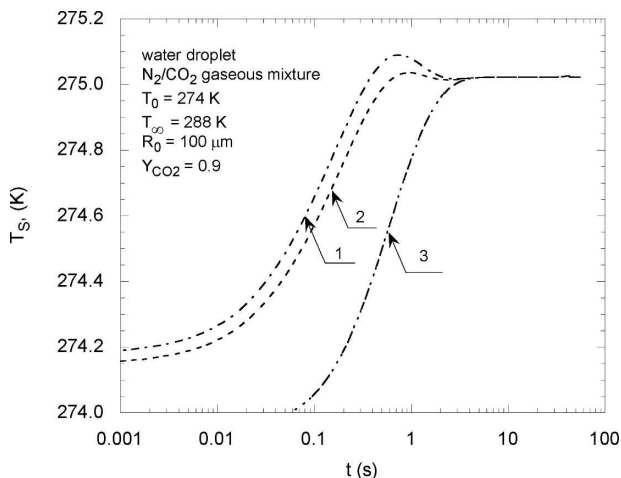


FIG. 10. Effect of Stefan flow and thermal effect of absorption on droplet surface temperature for a water droplet evaporating in $N_2/CO_2/H_2O$ gaseous mixture, $Y_{CO_2} = 0.9$, $R_0 = 100 \mu m$, $T_0 = 274 K$, $T_{e,\infty} = 288 K$. Line 1 includes effects of Stefan flow and thermal effect of absorption, for line 2 the thermal effect of absorption is neglected, and for line 3 the effects of Stefan flow and the thermal effect of absorption are neglected.

different initial concentrations of CO_2 at infinity. As can be seen from Fig. 13 the droplet surface temperature rises above the ambient temperature. Similar to the case of droplet evaporation the essential differences in surface temperature at the steady-state stage during droplet condensation are associated with large differences between heat and mass transport coefficients for carbon dioxide and nitrogen.

Summarizing the above findings it can be concluded

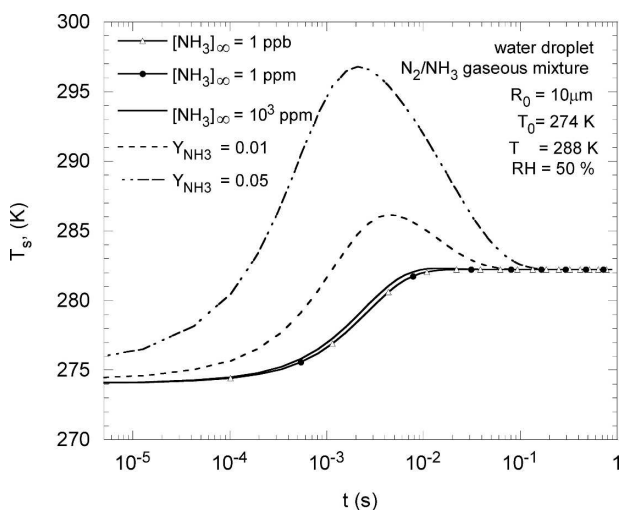


FIG. 11. Temporal evolution of the surface temperature for a water droplet evaporating in $N_2/NH_3/H_2O$ gaseous mixture, $R_0 = 100 \mu m$, $T_0 = 274 K$, $T_{e,\infty} = 288 K$.

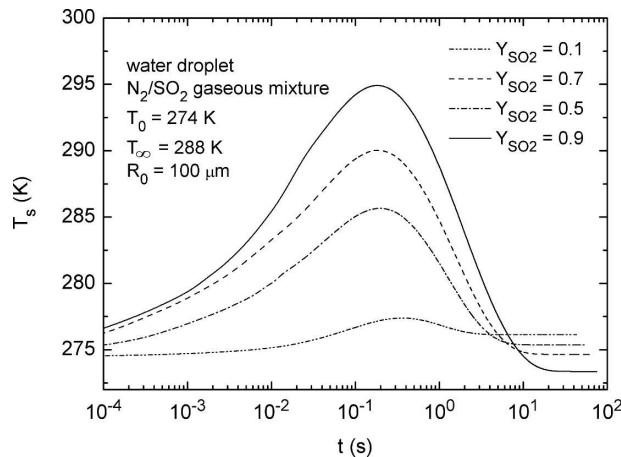


FIG. 12. Temporal evolution of the surface temperature for a water droplet evaporating in $N_2/SO_2/H_2O$ gaseous mixture, $R_0 = 100 \mu m$, $T_0 = 274 K$, $T_{e,\infty} = 288 K$.

that droplet temperature, interfacial absorbate concentration, and the rate of droplet evaporation (condensation) during gas absorption are highly interdependent. Gas absorption by evaporating (growing) droplets is affected by simultaneous heat and mass transfer processes: diffusion of soluble gas into the droplet and heat effect of absorption. A number of physical parameters, such as, Stefan velocity, Henry's law constant and droplet surface temperature, have a strong effect on increasing (decreasing) evaporation (condensation) rate, interfacial absorbate concentration, etc. The scheme of the interrelation between heat and mass transport during transient period of droplet evaporation with gas ab-

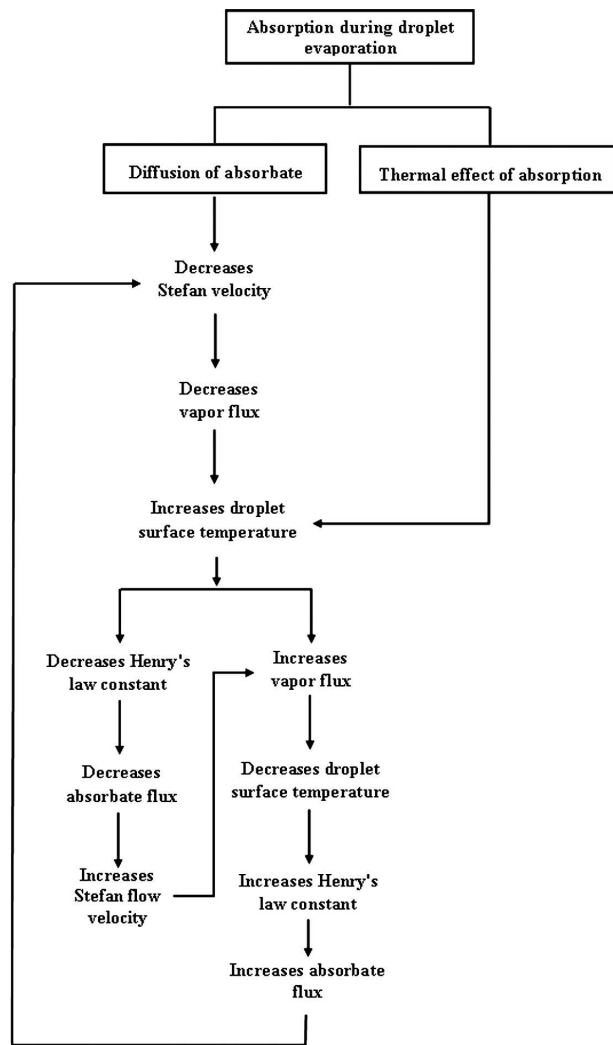


FIG. 14. Schematic description of the interrelation between heat and mass transport during the transient period of droplet evaporation with gas absorption.

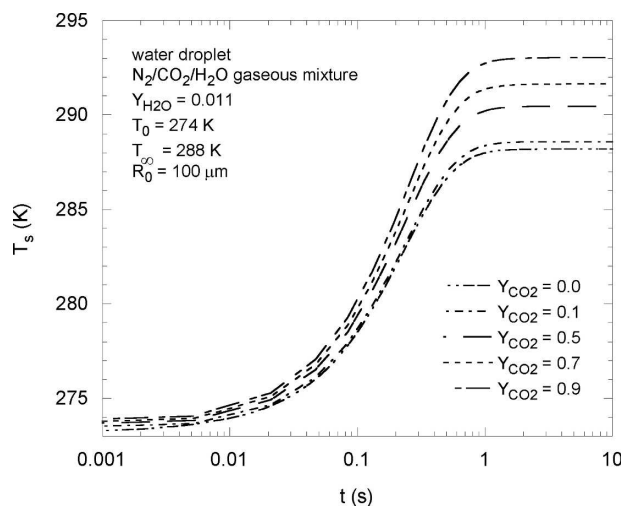


FIG. 13. Temporal evolution of the surface temperature for a water droplet condensation in $N_2/CO_2/H_2O$ gaseous mixture with $Y_{H_2O} = 0.011$, $R_0 = 100 \mu m$, $T_0 = 274 K$, $T_{e,\infty} = 288 K$.

sorption is shown in Fig. 14. The weak effects associated with the dependence of heat of absorption and diffusion coefficient of soluble gas in a liquid phase on temperature were neglected in our model. In contrast to droplet evaporation during droplet condensation the vapor flux and the hydrodynamic flow (Stefan flow) are directed toward the droplet surface. Vanishing of the surface temperature maximum during transient period of condensation (see Fig. 13) is a result of these phenomena. Nevertheless the above analysis demonstrates that in the case of absorption by evaporating droplets the heat and mass transfer processes are also highly interdependent.

The results of numerical calculations (see Figs. 9–13) show that small amounts of soluble gases in a gaseous mixture, corresponding to their typical concentrations

in a clean atmosphere, do not have essential influence on evaporation or condensation of liquid droplets. At the same time soluble gases with higher concentrations affect droplet evaporation or condensation. Consequently, in order to analyze this effect we performed numerical calculations of droplets evaporation or condensation for the concentrations of soluble gases, higher than those for typical atmospheric conditions. It should be emphasized that even for small concentrations of soluble species taking into account the mutual influence of absorption and condensation (evaporation) allows us to analyze correctly the influence of condensation (evaporation) on gas absorption. The latter stems from the nonlinear character of heat and mass transfer equations describing gas absorption and droplet evaporation (growth).

5. Conclusions

In this study we developed a model that takes into account the mutual effects of gas absorption and evaporation (condensation) of liquid droplet in the ambient atmosphere composed of liquid droplet vapor and inert noncondensable and nonabsorbable gas or noncondensable and soluble gas. The results obtained in this study can be summarized as follows.

- 1) The suggested model of droplet evaporation (condensation) in the presence of soluble trace gases takes into account a number of effects that were neglected in all previous studies, such as effect of gas absorption on Stefan velocity and thermal effect of absorption on droplet evaporation (condensation). It is demonstrated that droplet evaporation or condensation rate, droplet temperature, interfacial absorbate concentration, and the rate of mass transfer during gas absorption are strongly interdependent. We performed the detailed analysis of the interrelation between heat and mass transport during transient period of droplet evaporation (condensation) with gas absorption. The scheme of interrelation between droplet evaporation (condensation) and gas absorption is presented in Fig. 14.
- 2) During droplet evaporation the interfacial temperature as a function of time shows a maximum that increases with the increase of the ambient concentration of absorbate. For CO_2 dissolution the interfacial temperature as a function of time shows a maximum for large concentration of carbon dioxide in the ambient gaseous mixture while in the case of SO_2 and NH_3 dissolution the maximum of temperature arises even for small concentration of absorbate in the gaseous phase. In contrast to droplet evapo-

ration during droplet condensation vapor flux and the hydrodynamic flow (Stefan flow) are directed toward the droplet surface. It is shown that vanishing of the surface temperature maximum during the transient period of condensation is a result of these phenomena.

- 3) The results obtained using the suggested model for CO_2 absorption by water droplets agree with the experimental data by Taniguchi and Asano (1992) for CO_2 absorption by falling noncirculating water droplets and with analytical solution obtained for the case of aqueous-phase controlled mass transfer inside a stagnant nonevaporating droplet.
- 4) It is demonstrated that the time of saturation during gas absorption by liquid droplets only weakly depends on the value of the RH.
- 5) It is shown that the average concentration of the dissolved species strongly depends on RH for highly soluble and for slightly soluble gaseous atmospheric pollutants. Therewith the difference between the average concentrations of the dissolved species in water droplets attains tens of percent for different values of RH.

The performed analysis of gas absorption by liquid droplets accompanied by droplets evaporation and vapor condensation on the surface of liquid droplets can be used in calculations of scavenging of hazardous gases in atmosphere by rain and atmospheric clouds evolution.

APPENDIX A

Symbol Definitions

c_p	Specific heat at a constant pressure, $\text{J kg}^{-1} \text{K}^{-1}$
D_j	Diffusion coefficient of j th species in the gaseous phase, $\text{m}^2 \text{s}^{-1}$
D_{jk}	Binary diffusion coefficient in the gaseous phase, $\text{m}^2 \text{s}^{-1}$
$\text{Fo} = D_L t / 4R^2$	Fourier number
H_A	Henry's law constant, $\text{mole m}^{-3} \text{Pa}^{-1}$
k	Thermal conductivity, $\text{W m}^{-1} \text{K}^{-1}$
L_v	Latent heat of evaporation, J kg^{-1}
L_a	Heat of dissolution, J kg^{-1}
M_j	Molar mass of j th species, kg mole^{-1}
m_L	Mass of the droplet, kg
p	Pressure, Pa
r	Radial coordinate, m
R	Radius of the droplet, m
R_g	Universal gas constant, $\text{J mole}^{-1} \text{K}^{-1}$
R_G	Gas constant, $\text{J kg}^{-1} \text{K}^{-1}$
RH	Relative humidity

T	Temperature, K
t	Time, s
v, u	Velocity, m s ⁻¹
V_d	Droplet volume, m ³
Y	Mass fraction

Greek symbols

α	Thermal diffusivity, m ² s ⁻¹
ρ	Density, kg m ⁻³

Subscripts and superscripts

0	Initial value
1	Volatile species
A	Absorbate
e	Value outside a droplet
j	Number of a species
$L, (L)$	Liquid
s	Value at the droplet surface
∞	Value at infinity

APPENDIX B

Transformed System of Eqs. (17)–(5) and Appropriate Initial and Boundary Conditions

After the transformation of the spatial coordinate system with the aid of dimensionless independent variables (27)–(29) and the following dimensionless variables:

$$\xi = \frac{R(t)}{R_0}, \quad \vartheta_e = \frac{T_e}{T_{e,\infty}}, \quad \vartheta^{(L)} = \frac{T^{(L)}}{T_{e,\infty}}, \quad (\text{B1})$$

the Eqs. (1)–(5) can be rewritten in the following dimensionless form:

$$\frac{D_L \xi^2}{\alpha_L} \frac{\partial \vartheta^{(L)}}{\partial \tau} + \frac{\partial \vartheta^{(L)}}{\partial x} \left[\frac{2}{(1-x)} + \frac{D_L \xi}{\alpha_L} (1-x) \dot{\xi} \right] = \frac{\partial^2 \vartheta^{(L)}}{\partial x^2}, \quad (\text{B2})$$

$$\xi^2 \frac{\partial Y_A^{(L)}}{\partial \tau} + \frac{\partial Y_A^{(L)}}{\partial x} \left[\frac{2}{(1-x)} + (1-x) \xi \dot{\xi} \right] = \frac{\partial^2 Y_A^{(L)}}{\partial x^2}, \quad (\text{B3})$$

$$\begin{aligned} \xi^2 \sigma^2 \frac{\partial \vartheta_e}{\partial \tau} + \frac{\partial \vartheta_e}{\partial w} \sigma \left[\frac{u R_0 \xi}{D_L} - \xi \dot{\xi} (\sigma w + 1) - \frac{2k_e}{\chi (\sigma w + 1)} \right] \\ = \frac{1}{\chi} \frac{\partial}{\partial w} \left(k_e \frac{\partial \vartheta_e}{\partial w} \right), \end{aligned} \quad (\text{B4})$$

$$\begin{aligned} \xi^2 \sigma^2 \frac{\partial Y_j}{\partial \tau} + \frac{\partial Y_j}{\partial w} \sigma \left[\frac{u R_0 \xi}{D_L} - \xi \dot{\xi} (\sigma w + 1) - \frac{2\rho D_j}{\phi (\sigma w + 1)} \right] \\ = \frac{1}{\phi} \frac{\partial}{\partial w} \left(\rho D_j \frac{\partial Y_j}{\partial w} \right), \end{aligned} \quad (\text{B5})$$

where $\chi = \rho c_p D_L$, $\phi = \rho D_L$, and $u = u(w, \tau)$ is the function of dimensionless time τ and dimensionless coordinate w .

Equations (1)–(5) must be supplemented with two equations for determining gas velocity and for the rate of change of droplet's radius

$$u = \frac{\rho_s u_s}{\rho (\sigma w + 1)^2}, \quad (\text{B6})$$

where

$$u_s = \frac{\rho_L D_L}{\rho R_0 \xi (1 - Y_1)} \frac{\partial Y_A^{(L)}}{\partial x} \Big|_{x=0} - \frac{D_1}{\sigma R_0 \xi (1 - Y_1)} \frac{\partial Y_1}{\partial w} \Big|_{w=0}$$

and

$$\xi \dot{\xi} = - \frac{1}{(1 - Y_1)} \frac{\partial Y_A^{(L)}}{\partial x} \Big|_{x=0} + \frac{\rho D_1}{\rho_L D_L \sigma (1 - Y_1)} \frac{\partial Y_1}{\partial w} \Big|_{w=0}. \quad (\text{B7})$$

Dimensionless boundary conditions for $x = 0$, $w = 0$ read

$$\frac{\partial Y_j}{\partial w} \Big|_{w=0} = \frac{\sigma \xi R_0}{D_j} u_s Y_{j,s}, \quad j = 2, \dots, K-1, \quad j \neq A, \quad (\text{B8})$$

$$Y_{1,s}(0, \tau) = Y_{1,s}(\vartheta_s) = \frac{\rho_{1,s}}{\rho} = \frac{p_{1,s}(\vartheta_s) M_1}{p_\infty M}, \quad (\text{B9})$$

$$\vartheta^{(L)} = \vartheta_e, \quad (\text{B10})$$

$$\frac{\partial Y_A}{\partial w} \Big|_{w=0} = \frac{\sigma \xi R_0 u_s}{D_A} Y_{A,s} - \frac{D_L \rho_L \sigma}{D_A \rho} \frac{\partial Y_A^{(L)}}{\partial x} \Big|_{x=0}, \quad (\text{B11})$$

$$\begin{aligned} - \frac{\alpha_L}{D_L} \frac{\partial \vartheta^{(L)}}{\partial x} \Big|_{x=0} + \frac{L_a}{c_L T_\infty} \frac{\partial Y_A^{(L)}}{\partial x} \Big|_{x=0} \\ = \frac{k_e}{\rho_L D_L c_L \sigma} \frac{\partial \vartheta_e}{\partial w} \Big|_{w=0} + \frac{L_v}{c_L T_\infty} \xi \frac{d\xi}{d\tau}, \end{aligned} \quad (\text{B12})$$

and $Y_{A,s}$ can be found using Eq. (22).

Dimensionless boundary conditions for $x = 1$ and $w = 1$ read

$$\frac{\partial Y_j}{\partial w} = 0, \quad (\text{B13})$$

$$\frac{\partial \vartheta_e}{\partial w} = 0, \quad (\text{B14})$$

$$\frac{\partial \vartheta^{(L)}}{\partial x} = 0, \quad (\text{B15})$$

$$\frac{\partial Y_A^{(L)}}{\partial x} = 0. \quad (\text{B16})$$

Initial conditions for $\tau = 0$, and $0 < x < 1$, $0 < w < 1$, are as follows:

$$\vartheta^{(L)} = \vartheta_0^{(L)}, \quad Y_A^{(L)} = Y_{A,0}^{(L)}, \quad (\text{B17})$$

$$\vartheta_e = \vartheta_{e,0}(w), \quad Y_j = Y_{j,0}(w). \quad (\text{B18})$$

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