

CONDENSATION ON DROPLETS IN MOIST AIR BY HETEROGENEOUS NUCLEATION

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Abstract

Condensational growth of micro droplets that dissolve soluble aerosols in atmospheric air is investigated theoretically. An adequate theoretical procedure is proposed for calculating condensation rate of vapor to a droplet. The theoretical treatment contains the effects of heat transfer with condensation, binary diffusion of vapor/noncondensable gas system, and solution of soluble aerosols. Two theoretical approaches, kinetic theory and diffusion theory, are used to derive effective equations on growth rate of a binary component droplet respectively, and are compared with an experimental result [Majerowicz and Wagner, Lecture Notes in Physics **309** (1988), 27]. Two equations derived from these approaches are used according to a droplet radius. The equation derived from kinetic theory is useful when the radius is smaller than 40 nm, and the equation derived from diffusion theory is better for the other cases. The thermal effect is dominant to the growth rate regardless of the droplet radius. The equilibrium vapor pressure on the droplet surface, which affects the condensation rate, depends on the concentration of the dissolved materials in the droplet. An equation that is useful for the estimation of vapor pressure on a droplet surface was derived. In consideration of solubility of dissolved materials, the equation can estimate the vapor pressure adequately even if the droplet consists of high concentration solution. Finally, distributions of droplet radii in atmospheric air are computed based on the present theoretical treatment. By using the procedure, distributions of droplet radii, temperature and humidity changes were calculated, and these were affected by aerosol materials and aerosol concentrations. The insoluble aerosols do not act as nuclei for droplets when the humidity approaches 100%.

Key Words: Condensation, Nucleation, Droplet growth rate, Aerosol, Two-phase flow

1 INTRODUCTION

Atmospheric air is made up of not only noncondensable gas such as N₂ and O₂ but also vapor and aerosols, which are soluble and insoluble micro particles. From a macroscopic viewpoint, fog and mist occur in atmospheric air when vapor pressure becomes greater than saturated pressure. There are a lot of

droplets that grow from aerosols as nuclei by heterogeneous nucleation in fog and mist.

Droplets formation in fog and mist is a fundamental subject in meteorology and aerosol science. It is also important on computational fluid dynamics in engineering field. Airplane track behind airplanes is an example of the droplets formation in the field of aeronautics[1].

Condensational growth of droplets is a key phenomenon to understand the formation of droplets in fog, mist, and airplane track. Condensation, a kind of phase change from gas to liquid, is considered as accumulating process of gas molecules. The mass flow rate from vapor to a droplet is determined on the basis of molecular kinetics. The simplest theoretical formula from the molecular kinetics basis may be the Hertz-

Received on.

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Knudsen equation

$$I_v = -4\pi r^2 m_w \alpha \frac{P_{v\infty} - P_{vd}}{(2\pi m_w kT)^{\frac{1}{2}}}, \quad (1)$$

where I_v is the mass flow rate of water vapor leaving from a droplet of radius r , m_w is the mass of a water molecule, α is the mass accommodation coefficient for water vapor, $P_{v\infty}$ is the vapor pressure in air remote from the droplet, P_{vd} is the vapor pressure on the surface of the droplet, k is the Boltzmann constant and T is the temperature of the air. The effect of droplet surface curvature should be considered to estimate P_{vd} . The Kelvin equation is written in the form

$$\frac{P'_{vd}}{P_{v0}} = \exp\left(\frac{2\gamma_w}{\rho_w RT r}\right), \quad (2)$$

where P_{v0} is the saturated vapor pressure of pure water, γ_w is the surface tension of gas and liquid water interface, ρ_w is the density of liquid water and R is the gas constant. When the droplet consists of pure water, P'_{vd} is equal to P_{vd} . Equation (2) shows that vapor pressure on a micro droplet surface is higher than the pressure on a flat surface.

Another theoretical approach is based on continuum mechanics. The mass flow rate of water vapor leaving from a droplet and passing through a sphere with a radius x ($x \geq r$, in the case of $x = r$ the sphere corresponds to the surface of the droplet) is estimated by a simple diffusion equation

$$I_v = -4\pi x^2 \frac{D_{gv} M_w}{RT} \frac{dP_v(x)}{dx}, \quad (3)$$

where $P_v(x)$ and D_{gv} denote vapor pressure on the sphere and binary diffusion coefficient of water vapor in air, M_w is molecular weight of water.

Which is better treatment?

The system described in eqs.(1) and (2) is too simplified to describe the real situation in atmospheric air. Vesala et al.[2] reviewed that heat transfer with condensation and binary diffusion of vapor/noncondensable gas in air should be taken into consideration to evaluate the droplet growth rate. Neither heat transfer nor binary diffusion is taken into account in eq.(1).

On the other hand, the continuum approximation may be violated if the size of droplets is less than the mean free path of gas molecules in atmospheric air. Thereto heat transfer is not taken into account in eq.(3), too.

In addition, the effect of soluble nuclei is also important. The atmospheric air contains a lot of soluble aerosols such as NaCl. Because these aerosols act

as nuclei for heterogeneous nucleation, most droplets dissolve soluble aerosols. The vapor pressure on the surface of these droplets is decreased by the effect of solution of soluble aerosols.

The present investigation deals with the effects of 'heat transfer with condensation', 'binary diffusion of vapor/noncondensable gas system', and 'solution of soluble aerosols' on estimation of the mass flow rate of vapor from a droplet in atmospheric air. An adequate theoretical procedure is developed for calculating the mass flow rate of vapor from a droplet. These effects are considered in the procedure.

Then, varieties of droplet radii distribution and trends of the temperature and humidity in the air were computed for some cases by using the procedure. The effects of aerosol materials and concentrations in the air on the condensation rate were clarified.

These derivations and results are described in detail in this article.

2 EFFECT OF HEAT TRANSFER WITH CONDENSATION

In order to estimate effect of heat transfer with condensation on the mass flow rate of vapor from a droplet, three equations were shown. One of these equations was based on the kinetic theory of gas without consideration of the heat transfer, one of the others was derived based on the kinetic theory of gas with consideration of the heat transfer. The other equation does not include mass accommodation coefficient. By comparing of results given by these equations, effect of the heat transfer with condensation on the mass flow rate of vapor from a droplet was discussed.

2.1 Kinetic Theory

The Hertz-Knudsen equation (1) that was derived based on the kinetic theory of gas was generally used to estimate condensation rate. However the equation does not have parameters for consideration of chemical potential changes caused by such as the effect of solution of soluble aerosols when a vapor molecule put on the droplet surface from air.

On the other hand, the theory of rate process that is usually used for estimation of rate of chemical process advocated by Eyring[3]. Because the kinetic theory of gas was used for the derivation of the theory of rate process, the theory of rate process can be classified as more advanced theory of the kinetic theory. Eyring[3], Fujikawa and Maerefat[4] tried to apply the theory to condensation rate. Based on these research, an equation for estimation of the mass flow rate of vapor from

a droplet was derived[5] as

$$I_v = -4\pi r^2 m_w \alpha \tau \frac{P_{v\infty} - P_{vd}}{(2\pi m_w kT)^{\frac{1}{2}}} e^{-\frac{G_u^+ - G_{gu}}{kT_\infty}}, \quad (4)$$

where τ is the transmission coefficient, that is, the probability that a process actually takes place once the system in the activated state. G_u^+ and G_{gu} are chemical potentials for potential energy of a water molecule on the droplet surface and in air respectively. According to Fujikawa and Maerefat[4], $\tau = 1$ is given in this article.

Equation (4) is more precise than eq.(1), because it includes the effect of chemical potential change. Therefore, eq.(4) is used for the estimation of the mass flow rate of vapor from a droplet instead of eq.(1) hereafter.

2.2 Heat Transfer with Condensation

When a droplet is so small that its heat capacity can be negligible, all the enthalpy of water leaving from the droplet is equal to the heat leaving from the droplet as

$$h_l(T_d)I_v = Q_{rad} + h_v(T_d)I_v + 4\pi\kappa\beta r(T_d - T_\infty), \quad (5)$$

which is derived by Barrett and Clement[6] and corrected by using accommodation coefficient β according to Fukuta and Walter[7]. Where T_d is the temperature on the surface of a droplet, h_l and h_v are enthalpy of water and vapor for unit mass respectively, κ is the coefficient of thermal conductivity of air, and Q_{rad} is the rate of transferred heat by radiation from the droplet.

The mass flow rate of vapor from a droplet could be calculated with consideration of the heat transfer by the iteration method using eqs.(4) and (5) theoretically. However it was not easy to converge in fact, the theoretical equation to estimate the mass flow rate of vapor from a droplet with consideration of the heat transfer had to be derived even if it included approximation.

For the derivation, three parameters are defined as

$$\Delta h_{vl} \equiv h_v(T_d) - h_l(T_d), \quad (6)$$

$$S_\infty \equiv \frac{P_{v\infty}}{P_{v0}(T_\infty)}, \quad (7)$$

$$\eta \equiv \frac{M_w \Delta h_{vl}}{R} = \frac{\Delta H}{n_v R}. \quad (8)$$

Where M_w is molecular weight of water, Δh_{vl} means latent heat of evaporation for unit mass, S_∞ means saturation ratio and ΔH is given for latent heat of evaporation for n_v mol of water molecules. Because the volume V_v for n_v mol of vapor water molecules

is far more than the volume V_l for n_v mol of liquid water molecules, $\Delta V = V_v - V_l \approx V_v$ can be assumed. Therefore,

$$\eta = \frac{\Delta H T_\infty}{P_{v0}(T_\infty) V_v} = \frac{T_\infty}{P_{v0}(T_\infty)} \frac{\Delta H}{\Delta V}. \quad (9)$$

Clapeyron-Clausius relation is shown as

$$\frac{1}{T_\infty} \frac{\Delta H}{\Delta V} = \frac{dP}{dT} \approx \frac{P_{v0}(T_\infty) - P_{v0}(T_d)}{T_\infty - T_d}. \quad (10)$$

From eqs.(9) and (10),

$$\eta \approx \frac{T_\infty^2}{P_{v0}(T_\infty)} \frac{P_{v0}(T_\infty) - P_{v0}(T_d)}{T_\infty - T_d}, \quad (11)$$

was derived. Equation (5) was transformed as

$$\frac{Q_{rad}}{4\pi\kappa\beta r} = T_\infty - T_d - \frac{\Delta h_{vl} I_v}{4\pi\kappa\beta r},$$

and it was transformed again with using eq.(11),

$$\begin{aligned} \frac{Q_{rad}}{4\pi\kappa\beta r} \frac{\eta}{T_\infty^2} &= (T_\infty - T_d) \frac{\eta}{T_\infty^2} - \frac{\Delta h_{vl} I_v}{4\pi\kappa\beta r} \frac{\eta}{T_\infty^2} \\ &= 1 - \frac{P_{v0}(T_d)}{P_{v0}(T_\infty)} - \frac{\Delta h_{vl} I_v}{4\pi\kappa\beta r} \frac{\eta}{T_\infty^2}, \end{aligned} \quad (12)$$

hence

$$\frac{P_{v0}(T_\infty)}{P_{v0}(T_d)} = \left(1 - \frac{\Delta h_{vl} I_v}{4\pi\kappa\beta r} \frac{\eta}{T_\infty^2} - \frac{Q_{rad}}{4\pi\kappa\beta r} \frac{\eta}{T_\infty^2} \right)^{-1}. \quad (13)$$

On the other hand, equilibrium saturation ratio for vapor pressure on a droplet surface S_d was defined as

$$S_d \equiv \frac{P_{vd}}{P_{v0}(T_d)}. \quad (14)$$

When the droplet consists of pure water, S_d is given by the Kelvin equation (2). Then eq.(4) was transformed as

$$\begin{aligned} I_v &= -4\pi r^2 m_w \alpha \tau \frac{P_{v0}(T_\infty)}{(2\pi m_w kT)^{\frac{1}{2}}} e^{-\frac{G_u^+ - G_{gu}}{kT_\infty}} \\ &\quad \times \left(S_\infty - \frac{P_{v0}(T_d)}{P_{v0}(T_\infty)} S_d \right). \end{aligned} \quad (15)$$

Finally, the mass flow rate of vapor from a droplet with consideration of the heat transfer based on eq.(4) was derived from eqs.(13) and (15) as

$$I_v = -\frac{4\pi r (S_\infty - S_d) + S_d \frac{Q_{rad} \eta}{\kappa \beta T_\infty^2}}{S_d \frac{\Delta h_{vl} \eta}{\kappa \beta T_\infty^2} + \frac{4\pi r}{F_v}}, \quad (16)$$

where

$$F_v = \frac{4\pi r^2 m_w \alpha \tau P_{v0}(T_\infty)}{(2\pi m_w k T_\infty)^{\frac{1}{2}}} e^{-\frac{G_u^+ - G_{gu}}{k T_\infty}}. \quad (17)$$

When the droplet radius is enough large, $4\pi r/F_v$ can be negligible. Hence eq.(16) can be approximated as

$$I_v = -\frac{4\pi r(S_\infty - S_d) + S_d \frac{Q_{rad}\eta}{\kappa\beta T_\infty^2}}{S_d \frac{\Delta h_{vl}\eta}{\kappa\beta T_\infty^2}}. \quad (18)$$

Equation (18) does not have the mass accommodation coefficient. It shows that the effect of heat transfer with condensation is stronger than the effect of mass accommodation on mass flow rate of vapor, when the droplet is large enough.

Also Q_{rad} and T_d have to be given for calculation by using eq.(16) or eq.(18). Q_{rad} was defined as

$$\begin{aligned} Q_{rad} &\equiv 4\pi r^2 \epsilon \sigma (T_d^4 - T_\infty^4) \\ &= -4\pi r^2 \epsilon \sigma (T_\infty - T_d)(T_\infty + T_d)(T_\infty^2 + T_d^2), \end{aligned}$$

where σ is Stefan-Boltzmann constant and ϵ is the emittance on the droplet surface. When T_∞ closed to T_d , $T_\infty + T_d \approx 2T_\infty$ and $T_\infty^2 + T_d^2 \approx 2T_\infty^2$ are assumed. Therefore Q_{rad} is given as

$$Q_{rad} \approx -16\pi r^2 \epsilon \sigma T_\infty^3 (T_\infty - T_d), \quad (19)$$

and T_d was derived by using eqs.(5) and (19) as

$$\begin{aligned} T_d &\approx T_\infty + \frac{\Delta h_{vl}(T_d)I_v}{4\pi r(\kappa\beta - 4r\epsilon\sigma T_\infty^3)} \\ &\approx T_\infty + \frac{\Delta h_{vl}(T_\infty)I_v}{4\pi r(\kappa\beta - 4r\epsilon\sigma T_\infty^3)}. \end{aligned} \quad (20)$$

Therefore T_d is computed by using eq.(20) after given I_v by eq.(16) or eq.(18) for each calculation step.

2.3 Estimation of the Thermal Effect

According to the derivation of eq.(18), the mass flow rate of vapor from a droplet is governed by the effect of heat transfer with condensation when the droplet is enough large. For the estimation of the thermal effect in detail, calculations of droplet growth rate by using eqs.(4), (16) and (18) with actual values were needed. In the case of calculation by using eq.(4), it is assumed that the temperature of the vapor and the droplet surface are equal.

Three curves of droplet growth rate calculated by using eqs.(4), (16) and (18) where a droplet is growing are shown as Fig.1. The curve described by using eq.(18) is closer to the curve of eq.(16) than the curve

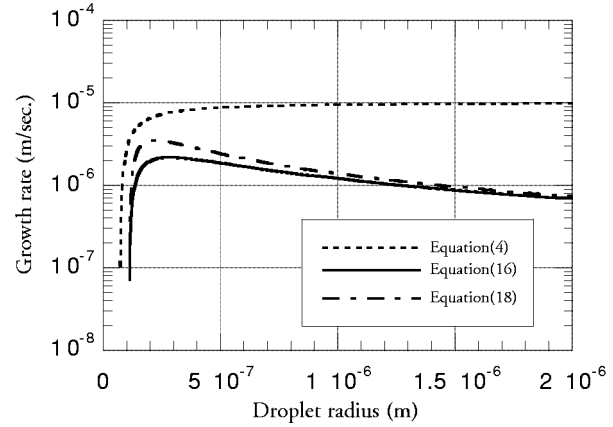


Fig.1: Droplet growth rate versus droplet radius calculated by using eqs.(4), (16) and (18) where a pure water droplet is growing with the humidity equal to 101.0%, $T_\infty = 293.15$ K.

of eq.(4) for the entire range of the droplet radius. Especially, it is important that the curve of eq.(4) starts at a smaller radius than the other curves. An extremely small droplet cannot grow by condensation due to temperature change. The temperature on the surface of a droplet increases during condensation by receiving latent heat, so that vapor pressure on the droplet surface surpasses the vapor pressure in the air remote from the droplet.

It is concluded that the effect of heat transfer with condensation is more important than mass accommodation in the estimation of the mass flow rate of vapor from a droplet and droplet growth rate. The effect of heat transfer with condensation should be considered for the estimation.

3 DROPLET GROWTH IN VAPOR/NONCONDENSABLE GAS SYSTEM

Because all of the equations shown above have no factors for the pressure of noncondensable gas, these equations should be only applied when there are no noncondensable gasses. There are far more noncondensable gas molecules than vapor molecules in atmospheric air however. In order to compute mass flow rate of vapor from a droplet in atmospheric air, the effect of binary diffusion of vapor/noncondensable gas system should be considered.

It is not easy to estimate the effect by using equations derived from kinetic theory. In order to estimate the effect, not only the transfer of noncondensable gas molecules but also the molecules collision of both vapor and noncondensable gas must be considered. In the case of a droplet's growth in atmospheric air, it is

not adequately to assume dilute gas for the consideration of molecular collision.

Conversely, diffusion theory is generally applied for problems such as binary diffusion. Growth rate of a droplet in atmospheric air is generally estimated by equations derived from diffusion theory in the field of meteorology and aerosols physics[6, 8, 9].

However diffusion theory is based on continuum hypothesis, droplet growth rate estimated by diffusion theory can not be precise when the droplet is small. Davis considered molecular kinetics in diffusion equation[10] and proposed that the diffusion coefficient should be given as a function of mean free path, though it is not generally used.

In order to adequately compute mass flow rate of vapor from a droplet in atmospheric air, the effect of binary diffusion of vapor/noncondensable gas system must be estimated. The thermal effect must be applied appropriately as well as the derivation of eq.(16). Thus an adequate equation based on diffusion theory considering of thermal effect was derived, and the equation and eq.(16) were verified by results of experiments conducted by Wagner and Majerowicz[11, 12].

3.1 Diffusion Theory

An equation for vapor flow from a micro droplet was derived based on the equation developed by Barrett and Clement[6]. Their equation considered the effects of heat transfer with condensation and the Stefan-flow. Barrett and Clement[6] showed an equation for the mass flow rate of vapor from a droplet passing through a sphere, where its radius was x , as

$$I_v = 4\pi x^2 \frac{D_{gv} M_w P}{RT_\infty} \frac{d}{dx} \ln(P - P_v(x)). \quad (21)$$

Where P is air pressure. Because I_v is constant for x of any value with assuming a steady-state

$$\frac{d}{dx} I_v = 0,$$

hence

$$\frac{d}{dx} \left\{ x^2 \frac{d}{dx} \ln(P - P_v(x)) \right\} = 0.$$

After variable separation, the equation was integrated as

$$\ln(P - P_v(x)) = \frac{K_1}{x} + K_2, \quad (22)$$

where K_1 and K_2 are integration constants. In order to decide K_1 and K_2 , $x = r$ and $P_v(x) = P_{vd}$ are given for the droplet surface, and $x = \infty$ and $P_v(x) = P_{v\infty}$

are assumed in air remote from the droplet surface. Then,

$$I_v = -4\pi r \frac{D_{gv} M_w P}{RT_\infty} \ln\left(\frac{P - P_{vd}}{P - P_{v\infty}}\right) \quad (23)$$

was derived.

The logarithm term of eq.(23) was approximated as

$$\begin{aligned} & \ln\left(\frac{P - P_{vd}}{P - P_{v\infty}}\right) \\ &= \ln\left(1 + \frac{S_\infty(P_{v0}(T_\infty) - P_{v0}(T_d)) + (S_\infty - S_d)P_{v0}(T_d)}{P - P_{v\infty}}\right) \\ &\approx \frac{S_\infty(P_{v0}(T_\infty) - P_{v0}(T_d)) + (S_\infty - S_d)P_{v0}(T_d)}{P - P_{v\infty}}, \end{aligned} \quad (24)$$

so eq.(23) was transformed as

$$\begin{aligned} I_v &= -4\pi r \frac{D_{gv} M_w P}{RT_\infty} \\ &\quad \times \frac{S_\infty(P_{v0}(T_\infty) - P_{v0}(T_d)) + (S_\infty - S_d)P_{v0}(T_d)}{P - P_{v\infty}}. \end{aligned} \quad (25)$$

When water vapor molecules act as an ideal gas, $P_{v0}(T_\infty) = \rho_v(T_\infty)RT_\infty/M_w$ is given. T_∞ was deleted as

$$I_v = -4\pi r \frac{(S_\infty - S_d) + s\left(\frac{P_{v0}(T_\infty)}{P_{v0}(T_d)} - 1\right)}{\frac{P - S_\infty P_{v0}(T_\infty)}{D_{gv} \rho_v(T_\infty) P} \frac{P_{v0}(T_\infty)}{P_{v0}(T_d)}}. \quad (26)$$

$P_{v0}(T_\infty)/P_{v0}(T_d)$ in eq.(26) was deleted by using eq.(13) as

$$I_v = -\frac{4\pi r(S_\infty - S_d) + S_d \frac{Q_{rad}\eta}{\kappa\beta T_\infty^2}}{S_d \frac{\Delta h_{vl}\eta}{\kappa\beta T_\infty^2} + \frac{P - S_\infty P_{v0}(T_\infty)}{D_{gv} \rho_v(T_\infty) P}}. \quad (27)$$

3.2 Verification of Equations (16) and (27)

For the verification of eqs.(16) and (27), it was best to compare the results of eqs.(16) and (27) to results of experiments that were conducted to measure particle diameter variation during periods of a droplet's growth. Constructing a control chamber for temperature and humidity in which particle diameter variation could be measured within the range of approximately 10 nm to 10 μ m, is extremely difficult, however.

Wagner developed an instrument for the experiments to measure the particle diameter variation during the period of growth of a nucleus to a droplet[13]. Wagner and Majerowicz[11, 12] conducted experiments for a DEHS nucleus that is an insoluble material in conditions of adiabatic expansion. The results

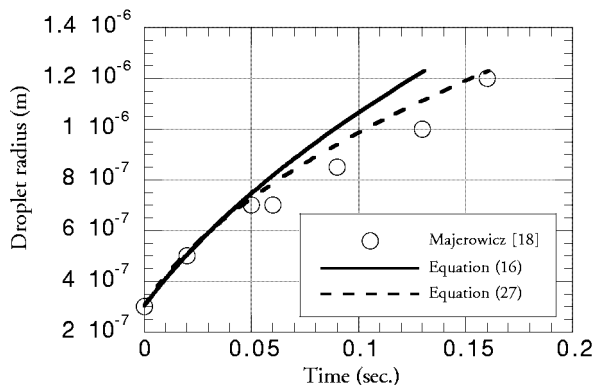


Fig.2: Trend of droplet radius at humidity equal to 106.1%, $T_{\infty} = 277$ K (average temperature during the experiment), $P = 95$ kPa, nucleus radius equal to 40 nm, and nucleus material is DEHS.

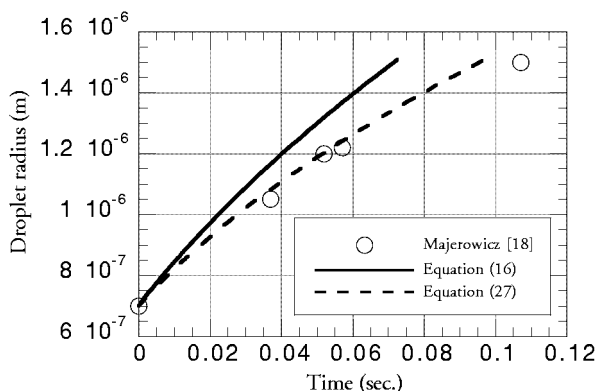


Fig.3: Trend of droplet radius at humidity equal to 112.2%, $T_{\infty} = 277$ K (average temperature during the experiment), $P = 95$ kPa, nucleus radius equal to 40 nm, and nucleus material is DEHS.

of their experiments were compared to computed results for the same conditions by using eqs.(16) and (27), shown as Figs.2, 3 and 4. It is noted that time 0 isn't the time that the experiment starts but the time of the first point of the droplet radius for each experiment in each figure, and the measured radius at that time was defined as the initial droplet radius for the calculation, respectively.

The results of eqs.(16) and (27) were in general agreement with the experimental results. The results of eq.(27) were better than the results of eq.(16). These results show that eq.(27) is more adequate than eq.(16) to calculate vapor flow from a micro droplet at almost all of the conditions in Figs.2, 3 and 4.

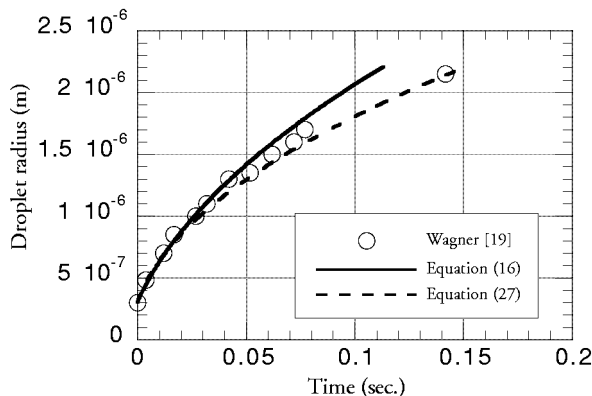


Fig.4: Trend of droplet radius at humidity equal to 120.2%, $T_{\infty} = 277$ K (average temperature during the experiment), $P = 95$ kPa, nucleus radius equal to 40 nm, and nucleus material is DEHS.

Although the diffusion theory is supported by the experimental data shown in Figs.2, 3 and 4, it is too simple to conclude that the diffusion theory is always valid. Because the continuum hypothesis, which is the basis of the diffusion theory, is violated when the size of droplet is comparable to the mean-free path of gas molecules. In fact, the mean free path of atmospheric air is 60 nm, while the droplet diameter in the experiments is almost larger than the mean free path.

Kinetics of gas molecules may be dominant for the growth rate of the droplet whose diameter is smaller than the mean free path. In order to verify this hypothesis, the growth rate calculated by the kinetic theory is compared with the diffusion theory. The results are shown in Figs.5 and 6. The surrounding air conditions are about 100 kPa in pressure, and 100% in relative humidity. Apparently, the kinetics of gas molecules is the most dominant factor to restrict the growth rate in the case where the droplet radius is less than 40 nm.

Because these verifications were given with consideration of conditions of the atmospheric air, the air pressures were given near atmospheric pressure, the humidity were given near 100%, and temperatures were given about 277 K - 293.15K. Therefore, it is concluded in this chapter that eq.(16) is useful to estimate the mass flow rate of vapor from a droplet in atmospheric air with the radius smaller than 40 nm, that is approximately the mean free path of molecules in atmospheric air. With droplet radius bigger than 40 nm, eq.(27) should be used in the estimation.

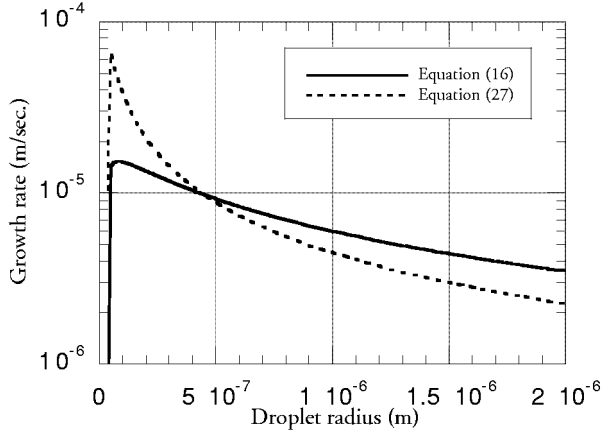


Fig.5: Droplet growth rate versus droplet radius calculated by using eqs.(16) and (27) where a droplet is growing in the case of the humidity equal to 106.1%, $T_\infty = 277$ K, $P = 95$ kPa, nucleus radius equal to 40 nm and nucleus material is DEHS.

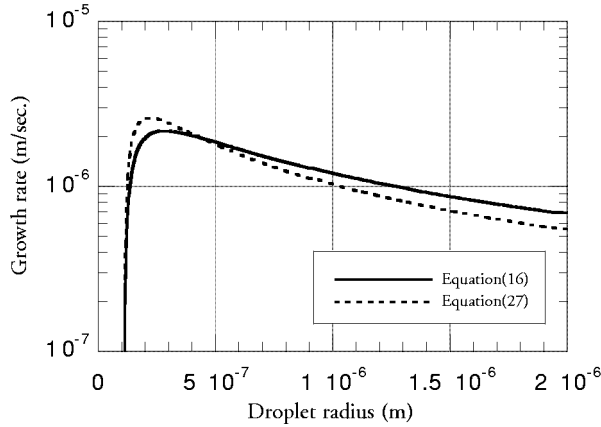


Fig.6: Droplet growth rate versus droplet radius calculated by using eqs.(16) and (27) where a droplet is growing in the case of the humidity equal to 101.0%, $T_\infty = 293.15$ K, $P = 101.3$ kPa, nucleus radius equal to 40 nm and nucleus material is DEHS.

4 VAPOR PRESSURE ON A SOLUTION DROPLET SURFACE

The mass flow rate of vapor from a droplet that dissolves a nucleus is strongly effected by vapor pressure on the droplet surface P_{vd} . In the eqs.(16) and (27), $S_d \equiv P_{vd}/P_{v0}(T_d)$ reflects the parameter P_{vd} . A droplet exists balanced in air when humidity is equal to $S_d \times 100$ %. Therefore a new equation to calculate S_d was derived, and the humidity that a droplet exists balanced in air was computed for some cases. For the computation, it was assumed that NaCl is the solute of the droplet, because the properties of NaCl as so-

lute are well-known and NaCl is a major component of aerosols over the sea.

4.1 Theory

Vapor pressure on the surface of a micro droplet that dissolves a nucleus is not the same as on a large flat surface of pure water due to the effects of droplet surface curvature and solution of soluble aerosols.

For describing the effect of droplet surface curvature, the Kelvin equation is usually used. Fisher and Israelachvili[14] conducted experiments and verified the equation when curvature radius is more than 4 nm. The radii of aerosols in atmospheric air are more than 1 nm in general[15]. It is difficult for small aerosols to nucleate because the surface vapor pressure of a small droplet must be far greater than the saturated vapor pressure. Therefore both aerosols and droplets with a radius less than 4 nm are not treated in this article. With droplet radius larger than 4 nm, the Kelvin equation is useful for describing vapor pressure on the surface of a pure water droplet.

For describing the effect of solution of soluble aerosols, Raoult's law as

$$\frac{P_s}{P_{v0}} = \frac{n_w}{n_w + i n_s} \quad (28)$$

was widely used to estimate vapor pressure on a surface of solution droplet. P_s is the vapor pressure on a flat surface of the solution, i is the van't Hoff factor of solute, n_w and n_s are the number of molecules of water and solute, respectively.

The vapor pressure on a micro droplet surface considering the effects of droplet surface curvature and solution used in eqs.(2) and (28) is shown by Manton[8] as

$$S_d = 1 + \frac{2\gamma}{RT\rho_w r} - \frac{i\rho_{sm}M_w}{\rho_w M_{sm}} \left(\frac{r_s}{r}\right)^3, \quad (29)$$

where γ is the surface tension of the liquid-vapor interface, ρ_{sm} is the density of solute, r_s is the equivalent radius of the nucleus, and M_{sm} is the weight of 1 mol of solute molecules.

However, eq.(29) is not adequate to estimate vapor pressure of non-dilute solutions, because Raoult's law is only valid for dilute solutions. Kuz proposed another formulation for the heterogeneous condensation based on his theoretical study of the variation around equilibrium of the Gibbs free energy[16]. Although his work is interesting, the formula was not verified through the experiments, and did not include the effect of solubility.

The most acceptable framework to estimate the

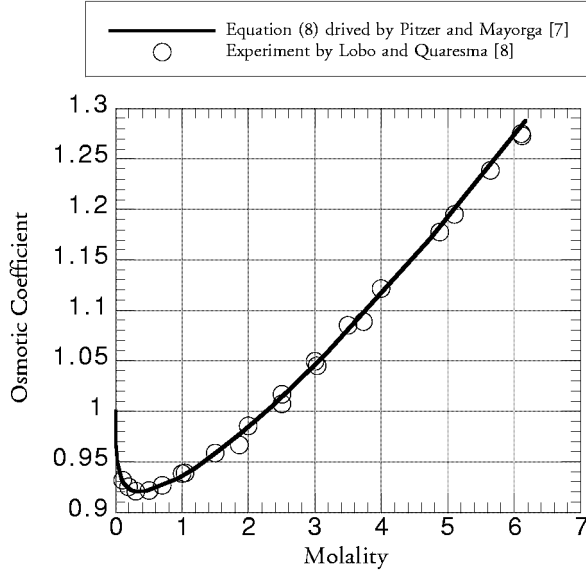


Fig.7: Comparison of the osmotic coefficient between experimental results[19] and the equation[18].

equilibrium vapor pressure of non-dilute solution is

$$\frac{P_s}{P_{v0}} = \lambda_w = \exp\left(-\phi_i \frac{a_i}{55.51}\right). \quad (30)$$

Where λ_w is water activity in solution, ϕ_i is the osmotic coefficient and a_i is the molality of the solute i .

Experimental studies and theoretical studies were conducted precisely by chemists in order to estimate ϕ_i [17]. In this article, an equation derived by Pitzer and Mayorga[18]

$$\phi_i = -A_\phi \frac{a_s^{\frac{1}{2}}}{1 + 1.2a_s^{\frac{1}{2}}} + a_s B_\phi + 0.00127a_s^2 + 1, \quad (31)$$

was used for the estimation of ϕ_i for aqueous NaCl solution. Where

$$A_\phi = 0.392 \times \left(\frac{273.15 + 25}{T}\right)^{\frac{3}{2}},$$

$$a_s = \frac{1000}{18} \frac{1 - X_w}{X_w},$$

$$B_\phi = 0.0765 + 0.2664 \exp(-2a_s^{\frac{1}{2}}),$$

X_w is mol fraction of water in the solution. Equation (31) is not only simple but also matches well the results of the experiments conducted by Lobo[19] as shown in Fig.7.

In order to estimate vapor pressure on the surface of a solution droplet, the parameters for pure water

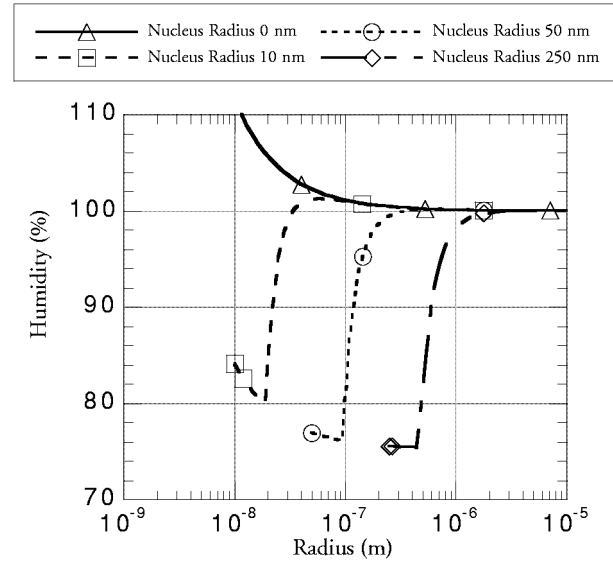


Fig.8: Köhler curves calculated by using eq.(32) at $T = 293.15$ K, the nucleus radius equal to 0 nm, 10 nm, 50 nm and 250 nm.

in eq.(2) such as γ_w and ρ_w should be substituted for the solution γ_s and ρ_s respectively. From eqs.(2) and (31),

$$S_d \equiv \frac{P_{vd}}{P_{v0}} = \exp\left(\frac{2\gamma_s}{\rho_s R T r} - \phi_i \frac{a_i}{55.51}\right) \quad (32)$$

was derived. Equation (32) should be useful to estimate vapor pressure on the surface of a solution droplet, even if the droplet consists of a high concentration solution.

4.2 Numerical Analysis

In the case of the heterogeneous condensation, initial droplet radius is the same as the nucleus radius. The water vapor molecules put on the particle surface, and the droplet radius varies with changing molality of the solute a_i and mol fraction of water X_w . a_i and X_w are functions of the droplet radius and the nucleus radius. Therefore vapor pressure on the surface of the droplet growing from the nucleus is determined by the droplet radius and temperature in eqs.(29) and (32).

The droplet radius increases more than 10 times its initial value during the droplet's growth. The temperature can only change a small percent of its initial value through condensation. Thus, results of the numerical analysis were shown in graphs as the relationship between the droplet radius and humidity $S_d \times 100\%$. Such graphs are called Köhler curves which are usually used in the field of aerosols studies.

Köhler curves were calculated by using eq.(32) as shown in Fig.8. With a nucleus radius equal to 0 nm, the humidity on the surface of the micro droplet is

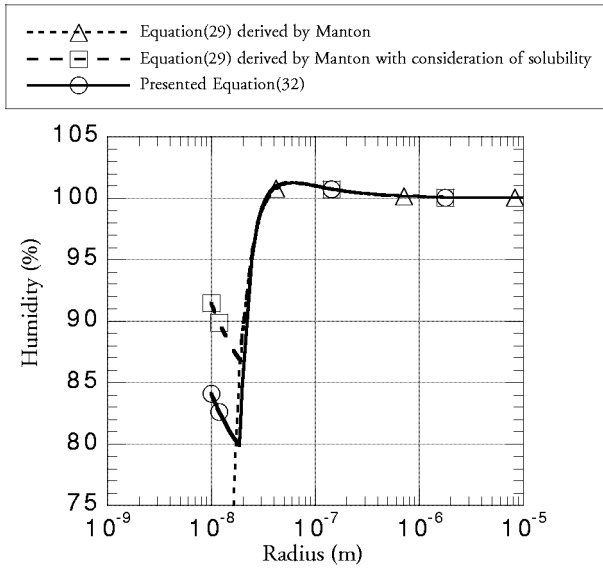


Fig.9: Köhler curves computed by using (29) and (32) at $T = 293.15$ K, nucleus radius equal to 10 nm.

so high that it is difficult to grow in atmospheric air. When there is a micro soluble aerosol such as a NaCl particle with radius 10 nm or 50 nm or 250 nm, it is easy to expand from the particle as a nucleus to a droplet. When an insoluble aerosol becomes a nucleus of a droplet, the humidity on the droplet should be given on the Köhler curve as for the nucleus radius equal to 0 nm. Because the solute is not dissolved in the droplet, the vapor pressure on the droplet surface should be considered as the only effect of droplet surface curvature in this case.

A Köhler curve computed by using eq.(29) is compared to a Köhler curve using eq.(32), as shown in Fig.9.

The curves computed by using eq.(32) have discontinuity, but the curves given by eq.(29) are continuous. The discontinuity is caused by the consideration of solubility. Hence, the curves computed by using eq.(29) with consideration to solubility have discontinuity. When the radius of the droplet is less than the discontinuity, the solution of the droplet should be a saturated solution. When the droplet radius exceeds the discontinuity, the concentration of the droplet solution decreases with the droplet's growth.

However, the curves computed by using eq.(29) with consideration of solubility do not correspond to the curves given by eq.(32), when the droplet is small. Because Raoult's law was used to derive eq.(29), it is not adequate for high concentration solutions.

Consequently, eq.(29) is not adequate for the estimation of vapor pressure on a solution droplet when the drop is too small. It is concluded in this chapter

that eq.(32) with consideration to solubility should be used for the estimation where nucleation process is included.

5 DISTRIBUTION OF DROPLET RADII

The mass flow rate of vapor from a droplet can be computed as mentioned above. However there are usually a lot of droplets in atmospheric air as in the case of fog or airplane track occurrence. Because each droplet needs water vapor molecules for growth, droplet growth causes humidity decrease in air. It follows that a droplet's growth affects other droplets' growth. The variety of vapor pressure in the air and the rate of change of sensible heat to latent heat are determined by the condensation rate. This is the sum of the mass flow rate of vapor to each droplet. Therefore a variety of droplet radii distribution is important for fluid simulation with consideration to vapor condensation.

In the area of cloud investigation, theoretical studies about distribution of droplet radii were conducted[8, 20]. The distribution of droplet radii was estimated with consideration of only droplet collisions in almost of these studies, because the droplet collisions govern the droplets' growth rate in the case of the droplets with radii larger than $10 \mu\text{m}$, such as the droplets in cloud[8]. However in the case of the droplet with radii smaller than $10 \mu\text{m}$, the effect of droplet collisions is negligible[8].

For the condensational growth of a droplet, the humidity in the air needs to be kept higher than a Köhler curve given for that condition. Because droplet growth needs vapor, vapor must be continuously supplied to keep a high humidity where droplets are growing. Therefore droplet growth by collisions is not considered for the estimation of the distribution of droplet radii, except in special circumstances such as in a cloud that is continuously supplied vapor.

Distributions of droplet radii were computed in the case a radius smaller than $10 \mu\text{m}$ with the assumption that the droplet growth is governed by condensation. At the same time, varieties of vapor pressure and temperature in air were calculated. The calculation method and the results were shown as follows.

5.1 Simulation Method

Fukuta and Walter[7] showed distribution of vapor density ρ_v and temperature T near a droplet as

$$\rho_v = \rho_{v\infty} - (\rho_{v\infty} - \rho_{vd}) \frac{r}{x}, \quad (33)$$

$$T = T_\infty - (T_\infty - T_d) \frac{r}{x}, \quad (34)$$

where $\rho_{v\infty}$ is the vapor density remote from the droplet surface and ρ_{vd} is the vapor density on the

droplet surface. x is a distance from the center of the droplet. In the case of $x = r$, $\rho_v = \rho_{vd}$ and $T = T_d$, and in the case of $x = \infty$, $\rho_v = \rho_{v\infty}$ and $T = T_\infty$. Therefore humidity and temperature at $100 \mu\text{m}$ away from a droplet center is little affected by the droplet existence, even if the droplet radius is $10 \mu\text{m}$.

Concentration of aerosols in atmospheric air is not stable. However aerosol concentration in room air were smaller than 10^4 cm^{-3} according to our own measurement by the Condensation Particle Counter. If aerosols are spread evenly, apart at a distance of $100 \mu\text{m}$ then the aerosol concentration is equal to 10^6 cm^{-3} . It was assumed that a droplet was not affected by the other droplets.

In order to compute the variety of the droplets and aerosols radii distribution in subject air, the mass flow rate of vapor from each droplet is calculated. The humidity and the temperature in subject air are computed by using the sum of the mass flow rate of vapor, the values of humidity and the temperature are used for the calculation of the droplets and aerosols radii distribution in the next time step. For each calculation for the mass flow rate of vapor from each droplet, eq.(16) was used to estimate in the case of radius smaller than 40 nm , and eq.(27) was used with a droplet radius larger than 40 nm . S_d was calculated by using eq.(32) for the computations of the mass flow rate of vapor from each droplet.

5.2 Results and Discussion

An example of the aerosols radii distribution in room air was given by Scanning Mobility Particle Sizer (SMPS), as shown in Fig.10. Thus, initial distribution of droplet radii was decided as in Fig.10. The initial temperature and humidity were 293.15 K and 105% in subject volume 1 cm^3 for the computation of the variety of the distribution.

With the assumption that all aerosols are made of NaCl, the variety of the droplet radii distribution was computed without a supply of water vapor or aerosols during the computation, as shown in Fig.11. Also, with the assumption that all aerosols are made of insoluble material, the variety of the droplet radii distribution was computed, as shown in Fig.12. Fig.12 shows that the aerosols made of insoluble material rarely can be the nuclei of droplets in such a condition. However in the case of Fig.11, most of the NaCl aerosols grow into large droplets and the distribution of droplet radii does not change after the droplets growth.

Also, trends of the humidity and temperature in subject air during the computation were shown in Fig.13. Because the droplets absorb water vapor molecules, the vapor pressure approaches saturated

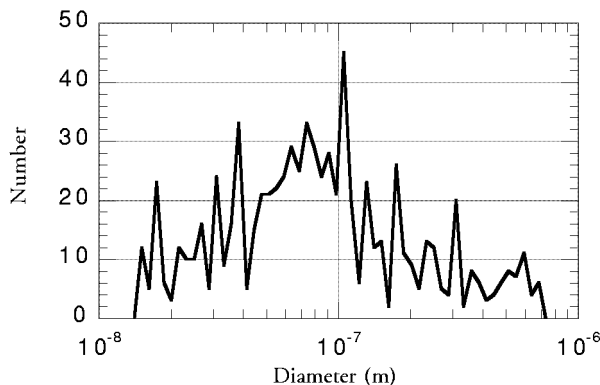


Fig.10: The initial droplet radii distribution for computation that was given by measurement of real aerosols in room air.

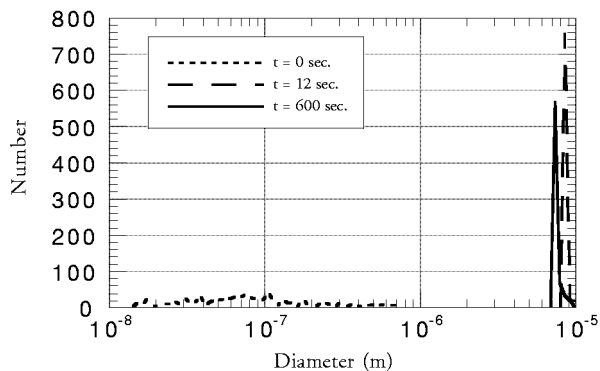


Fig.11: The variety of droplet radii distribution that was given by computation, growing from the NaCl aerosols.

vapor pressure. This inhibits droplet growth. At the same time, the temperature in subject air increased.

In order to calculate for higher concentrations of NaCl aerosols, a distribution of aerosols radii was given for the initial distribution by using a SMPS measurement in a chamber that aerosol experiments were conducted in. A variety of droplet radii distribution was computed without a supply of water vapor or aerosols during the computation, as shown in Fig.14.

Most of the NaCl aerosols grew into large droplets for 0.12 seconds after the droplets growth started, though the radii of almost droplets decreased and the others continued to grow for 600 seconds after the droplets growth started.

The trends of the humidity and temperature in the subject air during the computation are shown in

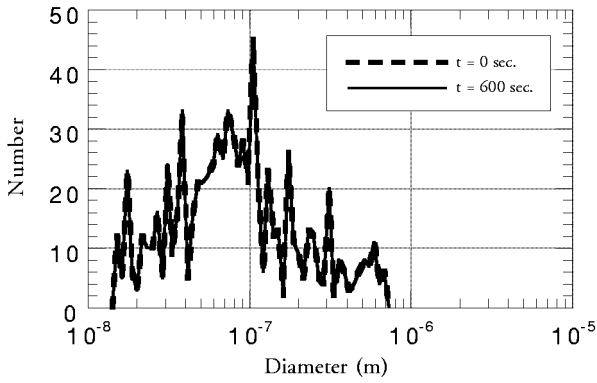


Fig.12: The variety of droplet radii distribution that was given by computation growing from the insoluble aerosols.

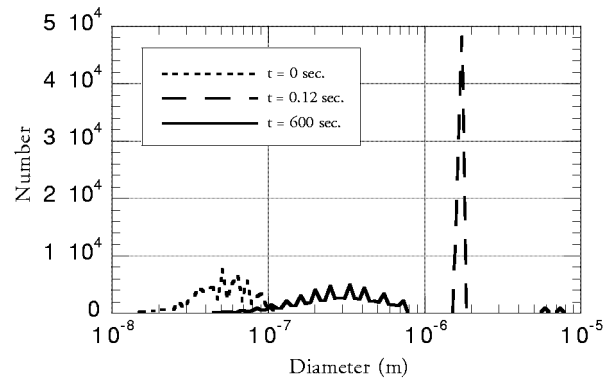


Fig.14: The variety of droplet radii distribution that was given by computation, growing from the high concentration NaCl aerosols.

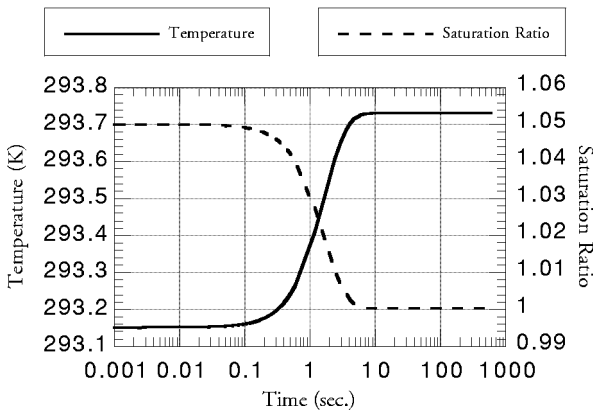


Fig.13: The trends of the humidity and temperature during the computation of Fig.11.

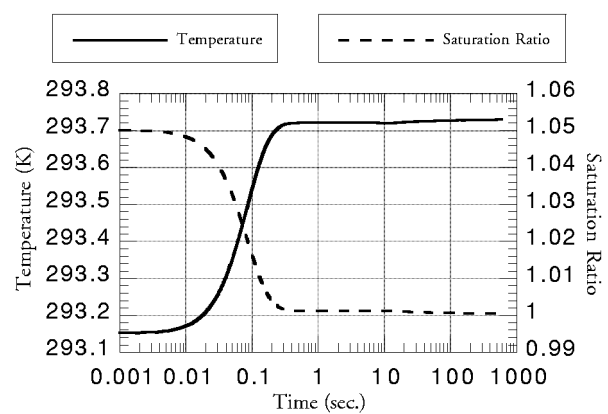


Fig.15: The trends of the humidity and temperature during the computation of Fig.14.

Fig.15. Figs.14 and 15 show that the humidity in the subject air decreased so instantaneously that most droplets can not keep large radii, and a part of the droplets can continue to grow.

On the other hand, Fig.15 shows that the humidity decrease and the temperature increase were more instantaneous in this case than in the case of lower concentration aerosols as shown in Fig.13. Therefore the rate of humidity and temperature changes is affected by the concentration of soluble aerosols.

Thus, the variety of droplet radii distribution is determined by aerosol materials, aerosol concentrations, humidity and vapor supply. It is especially difficult for insoluble aerosols to act as nuclei for droplets when the humidity approaches 100%. In addition, the aerosols concentration affects the temperature increase rate by the droplets growth.

6 CONCLUSIONS

A theoretical study on the effect of soluble aerosols on the condensation rate in atmospheric air was conducted. In order to estimate varieties of humidity and temperatures in atmospheric air that has aerosols and micro droplets, effects of ‘heat transfer with condensation’, ‘binary diffusion of vapor/noncondensable gas system’, and ‘solution of soluble aerosols’ on estimations of the mass flow rate of vapor from a droplet were studied. Based on these studies, a new adequate procedure for the estimation of the mass flow rate of vapor from a droplet was developed, and the variety of droplet radii distribution and trends of the temperature and humidity in the air were computed. It was concluded as follows.

In order to calculate the growth rate of a droplet in atmospheric air, the effect of heat transfer with con-

densation is more important than mass accommodation.

Equation (16) is useful to estimate the mass flow rate of vapor from a droplet in atmospheric air when the radius is smaller than 40 nm. When the droplet radius is bigger than 40 nm, eq.(27) should be used for the estimation.

Equation (32) with consideration to solubility should be used for estimations of vapor pressure on a droplet surface.

The variety of droplet radii distribution, and rates of temperature and humidity changes are determined by aerosol materials, aerosol concentration, humidity and vapor supply. It is especially difficult for insoluble aerosols to act as nuclei for droplets when the humidity approaches 100%.

APPENDIX: ESTIMATION OF THE PARAMETERS

Actual values for parameters in the equations had to be given for these calculations. However, equations for the estimation of some of these parameters are not generally accepted. These parameters are given from data already published.

1) Saturated Vapor Pressure

A lot of equations were proposed for the estimation of saturated vapor pressure. The equation that was given by Hyland and Wexler[21] is a well-known and useful equation for temperature range of 233.15 - 453.15 K. When the equation is applied for a temperature range of 233.15 - 323.15 K, it is shown as

$$P_{v0} = 610.78 \times 10^{\frac{7.5 \times (T-273.15)}{T-35.85}} \text{ [Pa]}. \quad (35)$$

2) Chemical Potential Variation

Chemical potential variation for the droplet's growth ΔG_{gl} and hydration ΔG_h where a water molecule is sticking to a droplet were derived by Kulmala, Laaksonen and Pirjola[22].

The free energy of surface tension of a droplet is given by

$$\Delta G_r = \frac{4}{3} \pi r^2 \gamma_d, \quad (36)$$

where γ_d is the surface tension of the liquid-vapor interface of the droplet. When a water molecule is applied to a droplet, r and γ_d change to r' and γ_d' respectively. Therefore ΔG_{gl} was given [22] as

$$\Delta G_{gl} = \frac{4}{3} \pi (r'^2 \gamma_d' - r^2 \gamma_d). \quad (37)$$

On the other hand, where a droplet consists of n_w mol of water molecules and n_s mol of solute molecules,

the chemical potential for hydration $\Delta G_w(n_w, n_s)$ is given[22] as

$$\Delta G_w(n_w, n_s) = kT_d \ln \frac{P_s}{P_{v\infty}}. \quad (38)$$

When a water molecule is applied to a droplet, $\Delta G_w(n_w, n_s)$ changes into $\Delta G_w(n_w + 1, n_s)$. Therefore ΔG_h is given by using (30) as

$$\begin{aligned} \Delta G_h &= \Delta G_w(n_w + 1, n_s) - \Delta G_w(n_w, n_s) \\ &= \frac{-kT_d}{55.51} (\phi_s(n_w + 1, n_s) a_s(n_w + 1, n_s) \\ &\quad - \phi_s(n_w, n_s) a_s(n_w, n_s)). \end{aligned} \quad (39)$$

In the case of a pure water droplet the chemical potential variation for hydration ΔG_h is equal to 0.

On the other hand, the chemical potential G is a function of partition function of a molecule Z as

$$G = -kT \ln Z + PV_m = -kT \ln (Z_t Z_r Z_v Z_u) + PV_m, \quad (40)$$

where Z_t , Z_r , Z_v and Z_u are partition function for translation, rotation, vibration and potential energy of a molecule respectively, V_m is volume for a molecule.

G_u^+ and G_{gu} are chemical potential for potential energy of a water molecule on the droplet surface and in air respectively. Therefore $G_u^+ - G_{gu}$ is not completely equal to $\Delta G_{gl} + \Delta G_h$.

Chemical potential for rotation and vibration of a molecule are the function of temperature, and chemical potential for parallel translation of a molecule is the function of temperature and the volume of subject system[23]. In the case of condensation, the effects of temperature change can be negligible in comparison with the effects of potential energy change that is determined by the change of the distance among molecules. Both ΔG_{gl} and ΔG_h are not functions of the volume of subject system. Therefore $\Delta G_{gl} + \Delta G_h$ can be approximately equal to $G_u^+ - G_{gu}$.

3) Diffusion Coefficient of Water Vapor in Air

The diffusion coefficient of water vapor in air is given as

$$D_{gv} = \frac{2.22915}{P} \times \left(\frac{T_\infty}{273.15} \right)^{1.75} \text{ [m}^2\text{s}^{-1}\text{]}, \quad (41)$$

according to the "International critical tables of numerical data"[24].

4) Density of Aqueous NaCl Solution

The density of aqueous NaCl solution should be related to NaCl concentrations and temperature. However, there was no general equation for describing the

relationship. Here an equation was given for calculations in this article based on the data[25], shown as

$$\rho_s = 998.6 \times \exp\left(\frac{40.65X_w - 40.65}{40.4X_w - 58.4}\right) [\text{kgm}^{-3}]. \quad (42)$$

Equation (42) has no variable for temperature. However, the effect of temperature change is less than 1% in the case of a temperature range of 283.15 - 303.15 K. The effect of temperature on the solution density is negligible in this article.

5) Surface Tension for Aqueous NaCl Solution

Surface Tension for aqueous NaCl solution should be related to NaCl concentrations and temperature, and an equation had to be given for the calculations in this article based on the data[25] as well as for the density, shown as

$$\begin{aligned} \gamma_s = & 7.408 \times 10^{-2} + 3.169 \times 10^{-4} \left(\frac{58.4X_w - 58.4}{40.4X_w - 58.4}\right) \\ & - 4.135 \times 10^{-6} \left(\frac{58.4X_w - 58.4}{40.4X_w - 58.4}\right)^2 \\ & + 7.163 \times 10^{-7} \left(\frac{58.4X_w - 58.4}{40.4X_w - 58.4}\right)^3 \\ & - 1.312 \times 10^{-8} \left(\frac{58.4X_w - 58.4}{40.4X_w - 58.4}\right)^4 [\text{Nm}^{-1}]. \quad (43) \end{aligned}$$

Because the effect of the temperature change is less than 5% in a temperature range of 283.15 - 303.15 K, the effect is ignored in this article.

6) Solubility

Solubility is a function of temperature. However the effect of the temperature change is negligible in a temperature range of 283.15 - 303.15 K, solubility of NaCl in water was given 26.3 ± 0.35 g/100g-water[25].

7) Others

The values of the other parameters were shown below.

$$\begin{aligned} R &= 8.314510 [\text{Jmol}^{-1}\text{K}^{-1}] [25] \\ \sigma &= 5.67051 \times 10^{-8} [\text{Wm}^{-2}\text{K}^{-4}] [25] \\ \epsilon &= 0.94 [26] \\ \kappa &= 2.41 \times 10^{-2} [\text{Wm}^{-1}\text{K}^{-1}] [26] \\ \Delta h_{vl} &= 2.5 \times 10^6 [\text{Jkg}^{-1}] [25] \\ \alpha &= 0.4 [4] \\ \beta &= 1 [7, 27] \end{aligned}$$

Acknowledgement

The authors would like to thank Professor Fumio Higashino for his insightful comments on this work and continuous encouragement to them.

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