

A New Theory of Heterogeneous Ice Nucleation for Application in Cloud and Climate Models

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Abstract. A new formulation is presented of the thermodynamical theory of heterogeneous ice crystal nucleation in clouds by freezing. This theory unifies and explains the empirical ice nuclei dependence on temperature and supersaturation, predicts crystal formation via condensation-freezing at a subsaturation over water. The theory also explains observations of high nucleation rates and crystal concentrations at warm ($-5 > -12$ °C) temperatures when the splintering mechanism may be not effective. This theory can be applied to parameterizations for use in cloud and climate models.

1. Introduction

The heterogeneous nucleation of ice crystals on ice nuclei (IN) governs the phase transformation of clouds and precipitation formation, and thereby influences the life times and optical properties of clouds. Heterogeneous nucleation of ice has also been studied in the context of cloud seeding and weather modification efforts.

As summarized by *Pruppacher and Klett* [1997, hereafter PK97], two main functional relationships between IN and the heterogeneous nucleation of ice have been found: 1) temperature dependence [*Fletcher*, 1962], $N_{IN}(T)$; and 2) dependence on ice saturation ratio S_i or ice supersaturation, $\delta_i = S_i - 1$, [*Huffman*, 1973; *Meyers et al.*, 1992, hereafter MDC92]. MDC92 showed that Fletcher's T -dependence underpredicts the ice needle concentration by 10^1 - 10^2 at $-5^\circ\text{C} < T < -25^\circ\text{C}$, but may overpredict N_{IN} by 10^3 - 10^5 at $T < -40^\circ\text{C}$.

Dependence of IN concentration on supersaturation has been invoked to explain the phenomenon of "ice multiplication" in clouds [e.g., *Rangno and Hobbs*, 1991, hereafter RH91] and could not be explained with *Fletcher's* T -dependence or the *Hallet-Mossop* splintering mechanism. RH91 hypothesized that high water supersaturation $\delta_w \sim 5$ -10% is required to explain the ice multiplication.

The above T - and δ -relations for IN concentration have been derived empirically. *Fletcher* [1962, 1969] formulated the classical theory of heterogeneous nucleation of an ice germ on the surface of an insoluble substrate for the freezing and deposition modes. *Fukuta and Schaller* [1982] extended Fletcher's theory by assuming that condensation and freezing proceeds in two steps. In all of the classical formulations, the T - and δ -effects remain independent, and condensation-freezing may occur only at water supersaturation.

In this paper, we examine heterogeneous ice nucleation via condensation-freezing (CF) and derive a general expression for the critical radius of an ice germ and free energy that depend simultaneously on both T and S_w and allows CF at $S_w < 1$.

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Paper number 1999GL011211.
0094-8276/00/1999GL011211\$05.00

The rate of CF is compared also with the deposition mode of ice nucleation. This theory explains many observed features of heterogeneous nucleation, and creates a basis for more physically justifiable parameterizations of heterogeneous nucleation in cloud and climate models.

2. Critical Radius, Energy, Nucleation Rates

The classical theory of heterogeneous ice nucleation considers two main mechanisms of ice formation: 1) ice germ formation on the surface of the solid insoluble substrate (SIS) which is in contact with a supercooled liquid droplet (hereafter referred to as the "freezing" mode) that may occur in condensation-freezing, contact, and immersion modes; and 2) ice germ formation from the vapor on the surface of the insoluble curved substrate (hereafter referred to as the "deposition" mode). We consider models for both freezing and deposition nucleation, whereby [following *Fletcher*, 1962] a droplet and an ice germ are approximated by a spherical cap on the surface of a solid aerosol particle.

2.1 Freezing Mode

Mixed aerosol particles usually contain a partially soluble portion (cloud condensation nucleus, CCN) along with an insoluble portion. After deliquescence of the CCN at $S_w < 0$, the SIS may initiate ice nucleation in the solution. The rate of germ formation in a supercooled droplet of water or solution per unit time per particle, $J_{s,fr}$ [s^{-1}], can be calculated following PK97:

$$J_{s,fr}(T, r_N) = Z_s N_c \Omega_{g,s} \frac{kT}{h} 4\pi r_N^2 c_{l,s} \exp\left[-\frac{\Delta F_{act}}{kT} - \frac{\Delta F_{g,s}}{kT}\right] \quad (2.1)$$

where k and h are Boltzmann's and Planck's constants, Z_s is *Zeldovich* factor, $N_c = 5.85 \times 10^{12} \text{ cm}^{-2}$ is the number of water molecules contacting unit area of ice germ, $\Omega_{g,s}$ is the ice cap surface area, ΔF_{act} is the activation energy of water molecule transition from liquid to ice at the solution-ice interface, $\Delta F_{g,s}$ is the critical energy of germ formation, r_N the radius of an insoluble fraction of an aerosol particle (IN), and $c_{l,s} = 10^{28} \text{ cm}^{-2}$ is the concentration of water molecules adsorbed on 1 cm^{-2} of a surface. Following PK97 (p.342), $Z_s N_c \Omega_{g,s} \sim 1$. In the classical theory, the expression for $\Delta F_{g,s}$ is related to the critical radius r_g of an ice germ [*Fletcher*, 1962; PK97].

The classical expressions for r_g and $\Delta F_{g,s}$ do not account for solution effects in the droplet, the temperature dependence of the latent heat of fusion, and the supersaturation (humidity) dependence. The generalized expressions for r_g and energy $\Delta F_{g,s}$ that account for both dependencies can be derived in a manner similar to *Khvorostyanov and Sassen* [1998, KS98], using some methods from *Khvorostyanov and Curry* [1999], and incorporating here the effects of the insoluble substance. We start with the general thermodynamical condition of equilibrium for an aqueous solution droplet with radius r_d , containing an ice germ of radius r_g located on a curved insoluble

substrate with radius r_N [PK97, (6-52)], generalized to account for the elastic misfit strain of the ice lattice by the insoluble substrate following *Turnbull* and *Vonnegut* [1952].

$$-\frac{L_m(T)}{T}dT + d\left(\frac{C\varepsilon^2}{\rho_i}\right) + \left(\frac{2}{\rho_w(T)} - \frac{2}{\rho_i(T)}\right)d\left(\frac{\sigma_{sa}(T)}{r_d}\right) - \frac{2}{\rho_i(T)}d\left(\frac{\sigma_{is}(T)}{r_g}\right) + \frac{RT}{M_w}d[\ln a_w] = 0 \quad (2.2)$$

Here L_m is the latent heat of fusion, ρ_i and ρ_w the ice and water densities, σ_{sa} and σ_{is} are the surface tensions at the solution-air and solution-ice interfaces, M_w is the molecular weight of water, R is the universal gas constant, ε is the elastic strain produced in ice embryo by the insoluble substrate, and C is the constant estimated by *Turnbull* and *Vonnegut* as 1.7×10^{11} dyn cm^{-2} . The values of ε are discussed in section 3. The activity of water a_w in solution is defined by

$$\ln a_w = -\frac{\nu\Phi_s \varepsilon_m M_w \rho_N r_N^3}{M_s \rho_w (r_d^3 - r_N^3)} \quad (2.3)$$

where Φ_s is the osmotic coefficient, ν is the number of ions in solution, ε_m and M_s are the mass fraction and the molecular weight of soluble material of a nucleus, and ρ_N is the density of the insoluble fraction.

Using a polynomial fit for $L_m(T)$ [PK97], we can integrate (2.2) with the usual boundary conditions ($a_w = 1$, $r_g = \infty$, and $\varepsilon = 0$ at $T = T_0$), and obtain:

$$L_m^{ef}(T) \ln \frac{T_0}{T} + \frac{C\varepsilon^2}{\rho_i} = \frac{2}{\rho_i} \left(\frac{\sigma_{sa}}{r_d} + \frac{\sigma_{is}}{r_g} \right) - \frac{RT}{M_w} \left[\frac{2\sigma_{sa}}{r_d \rho_w} - \frac{\nu\Phi_s \varepsilon_m M_w \rho_N r_N^3}{M_s \rho_w (r_d^3 - r_N^3)} \right] \quad (2.4)$$

Here $B_{sa} = (2\sigma_{sa}M_w/\rho_w RT)$ is Kelvin's curvature parameter for the air-solution interface, R the universal gas constant and we introduce an "effective" latent heat of fusion following KS98.

A droplet is much larger than an ice germ, $r_d \gg r_g$, so we can neglect the first term in the first bracket on the RHS of (2.4). The expression (2.4) can be simplified using the Kohler equation, which relates the saturation ratio over water S_w to the curvature and solution [PK97, (6-33)]:

$$\ln S_w = \frac{B_{sa}}{r_d} - \frac{\nu\Phi_s \varepsilon_m M_w \rho_N r_N^3}{M_s \rho_w (r_d^3 - r_N^3)} \quad (2.5)$$

Using (2.5), the ice germ radius can be found from (2.4):

$$r_g = \frac{2\sigma_{is}}{\rho_i L_m^{ef}(T) \ln \left(\frac{T_0}{T} S_w^G \right) - C\varepsilon^2} \quad (2.6)$$

where we have introduced a new dimensionless parameter

$$G(T) = RT/M_w L_m^{ef}(T) \quad (2.7)$$

The critical energy ΔF_{gS} can then be expressed as

$$\Delta F_{g,S} = \frac{4\pi}{3} \sigma_{iv} r_g^2 f(m_{iv}, x) = \frac{(16\pi/3) \sigma_{is}^3 f(m_{is}, x)}{\left[\rho_i L_m^{ef} \ln \left(\frac{T_0}{T} S_w^G \right) - C\varepsilon^2 \right]^2} \quad (2.8)$$

where $f(m_{iv}, x)$ is the shape factor caused by the geometry of the spherical cap and an aerosol particle with radius r_N , so that $x = r_N/r_g$. The term $m_{is} = \cos\theta_{is} = [(\sigma_{Ns} - \sigma_{Ni})/\sigma_{is}]$ is the cosine of the contact angle or "wettability parameter" at the so-

lution-ice interface and $f(x, m_{is})$ is defined by [Fletcher, 1962]:

$$2f(m, x) = 1 + [(1 - mx)/\phi]^3 + x^3(2 - 3\psi + \psi^3) + 3mx^2(\psi - 1), \\ \psi = (x - m)/\phi, \quad \phi = (1 - 2mx + x^2)^{1/2} \quad (2.9)$$

The expression for ΔF_{gS} can be generalized following *Fletcher* [1969] to account for the "active sites" with the relative area α , so that an IN of radius r_N contains the area $\alpha r_N^2 \ll 4\pi r_N^2$, with $m_{is} = 1$, and the rest of the surface with $m_{is} < 1$. Then ΔF_{gS} is

$$\Delta F_{gS} = \frac{4}{3} \pi \sigma_{is} r_g^2 f(m_{is}, x) - \alpha r_N^2 (1 - m_{is}) \quad (2.10)$$

Eqs. (2.6)-(2.10) along with (2.1) for $J_{s,fr}$ provide the solution to the problem of heterogeneous nucleation by freezing mode on the surface of a single aerosol particle (IN). For the condensation-freezing mode, the crystal concentration N_{fr} and crystal nucleation rate $dN_{fr}/dt \equiv R_{fr}$ ($\text{cm}^{-3} \text{s}^{-1}$) in a polydisperse aerosol with uniform size and surface properties can be calculated by integrating the probability of freezing P_{fr} of an individual droplet over aerosol size spectrum $f(r_N)$, normalized to the aerosol concentration N_a :

$$N_{fr}(t) = \int_{r_{\min}}^{r_{\max}} P_{fr}(r_N, t) f(r_N) dr_N, \quad R_{fr} = dN_{fr}/dt, \\ P_{fr}(r_N, t) = 1 - \exp \left(- \int_0^{\infty} J'_{s,fr}(t') dt' \right) \quad (2.11)$$

For the immersion mode, the probability of freezing of an aerosol particle $P_{fr}(r_N)$ embedded in a drop with radius r_d should be averaged over $f(r_N)$ (the activated aerosol embedded in drops), and integrated over the drop size spectrum $f(r_d)$. The calculation for the contact mode is a little more complicated and can be based on the evaluation of the Brownian, diffusive and thermoforetic fluxes of interstitial unactivated aerosol to a drop [e.g., *Young*, 1993].

The previous considerations show that the same aerosol particles may act both as IN and CCN under various conditions; thus N_a is of the order of N_{CCN} . The classical formulation for condensation-freezing mode [PK97, (9-38)] allowed nucleation only for the case of water saturation. This new formulation (2.6)-(2.8) allows nucleation both for supersaturation in liquid clouds and at subsaturation over water, $S_w < 1$, but supersaturated over ice, $S_i > 1$.

2.2. Deposition Mode

The work of an ice germ formation from the vapor is [following *Young*, 1993, PK97]:

$$\Delta F_{g,S} = \frac{4\pi}{3} \sigma_{iv} r_g^2 f(m_{iv}, x) = \frac{(16\pi/3) \sigma_{iv}^3 f(m_{iv}, x)}{\left[R_v T \ln S_i - C\varepsilon^2 \right]^2}, \quad (2.12)$$

where M_w is the wettability parameter at the ice-vapor interface. Then the nucleation rate $J_{s,dep}$ can be written as [PK97]

$$J'_{s,dep}(r_N, T) = \frac{4\pi r_N^2 r_g^2 Z S e}{(2\pi m_w k T)^{1/2}} c_{1,s} \exp[-\Delta F_{g,S}/kT] \quad (2.13)$$

where e is the vapor pressure, m_w the mass of water molecule, $c_{1,s}$ concentration of vapor molecules in the monolayer on the surface. The preexponential factor (kinetic coefficient) is about $10^{26} \cdot r_N^2 \text{ cm}^{-2}$ [Fletcher, 1962]. The crystal nucleation rate

$R_{dep}=dN_{dep}/dt$ for polydisperse aerosol is calculated by integrating over $f(r_N)$ as in (2.11).

3. Discussion

Equation (2.6) for r_g contains two particular cases of classical nucleation theory. At $S_w=1$, $\epsilon=0$, (2.6) transforms into *J.J. Thompson's* (1882, as cited by *PK97*) expression $r_g=2\sigma_{is}/(L_m\rho_i \ln(T_0/T))$, for nucleation by freezing of water. In the second case $T \rightarrow T_0$ and $\epsilon=0$, (2.6) simplifies to *Kelvin's* (1870, see *PK97*) expression $r_g=2\sigma_{is}/(R_v T \rho_i \ln S_w)$ for the nucleation of a crystal from the vapor. In general, (2.6)-(2.8) unify and generalize these two cases and provide a quantitative description of nucleation that accounts for the both temperature and supersaturation dependencies.

Eq. (2.8) for ΔF_{gs} differs from the case of homogeneous nucleation by the term $f(m,x)$. If the insoluble substance is completely wettable, then $\theta=0$, $m_{is}=1$, $f=0$, and $\Delta F_{gs}=0$. If the substrate is completely non-wettable ($\theta=180^\circ$, $m=-1$, $f=1$), $\alpha=0$, and misfit $\epsilon=0$, then (2.8) transforms into the energy of homogeneous nucleation [*KS98*] which is therefore a particular case of heterogeneous nucleation. The exact values of m_{is} for the substances that may constitute the SIS have not yet been accurately measured [*PK97*], but they can be estimated by comparison with the parameter m_{wa} for water: $\theta=43-68^\circ$, $m=0.36-0.73$ for surface soil, quartz, sand; and $\theta=9-17^\circ$, $m=0.95-0.99$ for *AgI* (which explains its high efficiency in cloud seeding). If the crystal lattice of the insoluble substrate satisfies the condition of epitaxy, i.e., have hexagonal structure with the parameters close to ice, then ϵ can be $\sim 1-5\%$, and if the substrate lattice is dissimilar to ice, then the germ lattice is not accommodated to it and $\epsilon=0$ [*Young, 1993; PK97*].

We can introduce the "effective temperature" and the "effective" supercooling ΔT_{ef} caused by supersaturation δ_w . T_{ef} can be defined using (2.6) from the relation $\ln(T_0/T_{ef}) = \ln[(T_0/T) S_w^G]$. Then $T_{ef} = T S_w^{-G} = T(1+\delta_w)^{-G} \approx T + \Delta T_{ef}$, where $\Delta T_{ef} = -T_0 G \delta_w \approx -100\delta_w$, i.e., ΔT_{ef} in $^\circ\text{C}$ is approximately equal to water supersaturation in % with the opposite sign. Table 1 shows G and ΔT_{ef} for $\delta_w=0.1$ (10 %), corresponding to an effective shift in T of 10-12 $^\circ\text{C}$ to the colder T_g accelerating freezing; at subsaturation $\delta_w = -0.1$ (-10 %), the T_{ef} is by $\sim 10^\circ\text{C}$ warmer decelerating the freezing rate. Note that according to this theory, the crystals may form at high δ very close to 0°C or even slightly above. This explains the success of many cloud seeding experiments at $T \sim 0^\circ\text{C}$.

Since r_g should be positive, we have from (2.6) the condition $(T_0/T) \cdot S_w^G \geq \exp[C\epsilon^2/(\rho_i L_m \epsilon^f)]$, or a condition of minimum threshold $S_{w,th}$ for nucleation

$$S_{w,th} = [(T/T_0)\exp(C\epsilon^2/L_m \epsilon^f)]^{1/G} \tag{3.1a}$$

which, for $\epsilon=0$, is the same as for homogeneous nucleation:

$$S_{w,th} = (T/T_0)^{1/G} \tag{3.1b}$$

Figure 1 shows the temperature dependence of the threshold humidity ($100 \cdot S_{w,th}$) of heterogeneous nucleation in com-

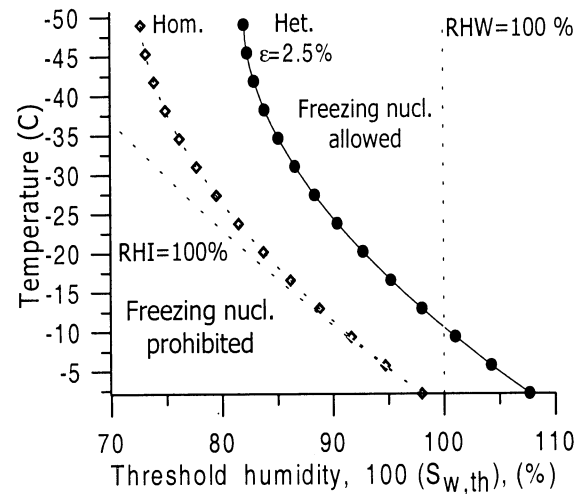


Figure 1. Temperature dependence of the threshold humidity ($100 S_{w,th}$) of heterogeneous nucleation, calculated with the misfit strain $\epsilon=1\%$ (asterisks), 2.5% (circles), and $\epsilon=0$ (diamonds, same as for homogeneous nucleation). The curve $RHI=100\%$ (crosses) denotes saturation over ice.

parison with the homogeneous nucleation threshold. The values of $S_{w,th}$ in Figure 1 decrease with T . In contrast to the classical theory for pure water where freezing nucleation is prohibited at $S_w < 1$ (to the left of the vertical line $RHW=100\%$), it is allowed by the new theory in the much wider region between this line and the threshold curves on the left down to $RHW=0.75-0.85$ at $T=-35$ to -50°C . Here, crystals may form from the solution droplets (haze particles) in the clear sky and survive since the humidity is higher than saturation over ice, $RHI > 100\%$ (line on the left). This explains the phenomenon of "diamond dust" which is often observed in the Arctic and shows that cirrus clouds may also form via heterogeneous freezing of haze at $S_w < 1$. This lowering of the threshold humidity for freezing mode below 100% is valid only for mixed aerosols while the threshold $RHW=100\%$ is valid for the insoluble aerosols

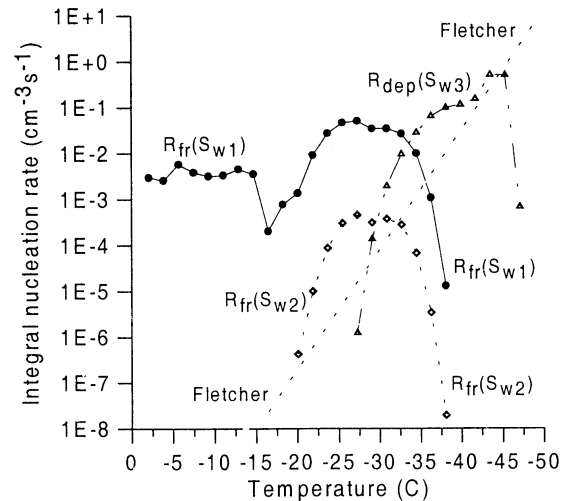


Figure 2. Heterogeneous nucleation rates: freezing, $R_{fr}(S_{w1})$, $R_{fr}(S_{w2})$, for two saturation ratios S_{w1} and S_{w2} , shown in Table 1, integrated according to (2.10) over the size spectrum of the aerosol with $N_a=200 \text{ cm}^{-3}$; deposition rate, $R_{dep}(S_{w3})$, and Fletcher's rate calculated with a vertical velocity of $w=5 \text{ cm s}^{-1}$, $m_{is}=0.5$ for freezing mode and $m_{iv}=0.95$ for deposition mode.

Table 1. Temperature variations of G , ΔT_{ef} ($\delta_w=0.1$), S_{w1} , S_{w2} , S_{w3}

$T, ^\circ\text{C}$	-5	-10	-20	-30	-40	-50
$G(T)$	0.39	0.40	0.43	0.47	0.53	0.64
$\Delta T_{ef}, ^\circ\text{C}$	9.7	9.8	10.2	10.6	11.6	13.0
S_{w1}	1.08	1.03	0.94	0.87	0.81	0.74
S_{w2}	1.03	1.004	0.93	0.86	0.809	0.74
S_{w3}	1.11	1.07	1.016	0.946	0.889	0.820

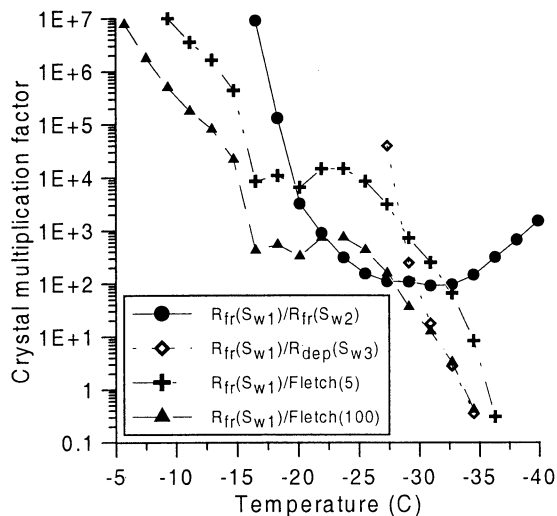


Figure 3. Possible “ice multiplication factor”, determined from the ratio of the rates shown in Fig. 2: $P_{fr}(S_{w1})/P_{fr}(S_{w2})$, $P_{fr}(S_{w1})/P_{dep}(S_{w3})$, $P_{fr}(S_{w1})/(\text{Fletcher's rate at } w=5 \text{ cm s}^{-1})$, $P_{fr}(S_{w1})/(\text{Fletcher's rate at } w=100 \text{ cm s}^{-1})$.

3.2. Results

To calculate J_S' from (2.1), we use the temperature parameterizations of ρ_i , ρ_w , L_m , σ_{as} , σ_{is} , $\Delta F_{act}(T)$ from [PK97, ch. 3, 5], and a new fit for $\Delta F_{act}(T)$ at $T_c < -30^\circ\text{C}$ that matches the measured homogeneous nucleation rates [cf. KS98]: $\Delta F_{act}(T) = 0.694 \cdot 10^{-12} \{1 + 0.027 \cdot (T_c + 30) \cdot \exp[0.01 \cdot (T_c + 30)]\}$ with T_c in $^\circ\text{C}$. Figure 2 shows results of the comparison of freezing nucleation rates R_{fr} (for two saturation ratios S_{w1} , S_{w2} given in Table 1) with the deposition rate at $S_{w3} = 0.97$ and with the Fletcher's rate.

The integration in (2.11) was performed over $r_N = 0.02\text{--}4 \mu\text{m}$ with the Junge aerosol size distribution with index $\mu = 4.5$ normalized to the total concentration $N_a = 200 \text{ cm}^{-3}$. The curve $R_{fr}(S_{w1})$ with $\delta_w = 2\text{--}8\%$ may be identified with conditions in a cloud at the stage of intensive coagulation and high water supersaturation [RH91]. This curve quantitatively explains the results from RH91, since $N_c \sim 300\text{--}600 \text{ l}^{-1}$ may form in a period as short as 100–200 s. A small decrease in δ_w by 3–4% in $R_{fr}(S_{w2})$ can be identified with conditions when high supersaturations are absent, leading to a sharp decrease in nucleation rates and N_{fr} by several orders of magnitude. The deposition nucleation rate calculated with (2.12)–(2.13) at $S_{w3} = 0.97$ is much less than the freezing nucleation rate at warm temperatures, becomes comparable at -30 to -35°C , then exceeds freezing rates at lower temperatures since $S_{w3} \gg S_{w1,2}$. Both freezing and deposition modes exceed Fletcher's rate in the temperature range -15 to -35°C by 2–4 orders but are much lower at $T < -40$ to -45°C , in agreement with the experimental results described in MDC92.

Figure 3 shows possible “ice multiplication factors” (N_c/N_N) as described in [RH91]. Specifically we present various relations of the nucleation rates by different modes and different humidities, and compare them to Fletcher's rates with $w = 5 \text{ cm s}^{-1}$ (St clouds) and $w = 100 \text{ cm s}^{-1}$ (Cu clouds). As we can see from Figure 3, even the moderate δ_w in clouds (2–8%) may cause substantial enhancement of the nucleation rates. Note that in our interpretation, no actual “ice multiplication” occurs, and this factor simply reflects the increase in the number of crystals nucleated in this theory at $S_w > 1$.

4. Conclusions

The heterogeneous nucleation theory described above explains the empirical dependencies of ice nuclei behavior on temperature and supersaturation, allows crystal formation via condensation-freezing at subsaturation over water (down to $\text{RH} = 75\text{--}85\%$), and explains observations of high crystal concentrations ($200\text{--}600 \text{ L}^{-1}$ in 300–600 s) at warm (-5 to -12°C) temperatures when the splintering mechanism is not effective. This theory can serve as an alternative to the empirical methods of ice nucleation parameterizations used now and can be recommended for cloud and climate models, in particular, for simulation of cirrus clouds and diamond dust.

However, many basic parameters and assumptions of the theory still require further measurement and verification: the temperature dependence of all thermodynamical parameters (latent heat, surface tension, free energy); values of the contact angle for many substances that may serve as IN; and the chemical composition of IN. The irregularities of the surface of solid insoluble substrate may also strongly influence the nucleation rate. Incorporation of this nucleation theory into numerical models of various complexity, especially with explicit microphysics [e.g., Khvorostyanov et al., 2000] may help to test its validity and improve it further.

Acknowledgments. This research has been supported by the Department of Energy Atmospheric Radiation Measurement Program. We would like to thank the anonymous reviewers for their very helpful comments.

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(Received: November 9, 1999, revised: August 23, 2000; accepted: August 24, 2000)