A PARAMETERIZATION OF CLOUD DROPLET NUCLEATION

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1. INTRODUCTION

Droplet nucleation is a fundamental cloud process. The number of aerosols activated to form cloud droplets influences not only the number of aerosols scavenged by clouds but also the size of the cloud droplets (Twomey, 1977). Cloud droplet size influences the cloud albedo (Charlson et al., 1992) and the conversion of cloud water to precipitation (Albrecht, 1989; Radke et al., 1989). Global aerosol models are presently being developed with the intention of coupling with global atmospheric circulation models to evaluate the influence of aerosols and aerosol-cloud interactions on climate (Erickson et al., 1991; Langner and Rodhe, 1991; Charlson et al., 1991; Penner et al., 1991). If these and other coupled models are to address issues of aerosol-cloud interactions, the droplet nucleation process must be adequately represented. Here we introduce a droplet nucleation parameterization that offers certain advantages over the popular Twomey (1959) parameterization.

2. PARAMETERIZATION

The parameterization relies on several simplifying approximations. First, the aerosol size distribution is represented as a log-normal size distribution,

$$\frac{dn}{d\ln a} = \frac{N_a}{\sqrt{2\pi}s} \exp\left[-\frac{(\ln(a/a_m))^2}{2s^2}\right] \quad (1)$$

where N_a is the total aerosol number concentration, a_m is the number mode radius, and $s = \ln\sigma$ is the natural logarithm of the standard deviation σ. The number nucleated can be expressed in terms of N_a, a_m, s, and the radius of the small-

est aerosol activated, a_c, by integrating (1) over all aerosols larger than a_c, which yields

$$N = N_a\frac{1}{2}[1 - \mathrm{erf}(x_c)]. \quad (2)$$

where

$$x_c \equiv \frac{\ln(a_c/a_m)}{\sqrt{2}s} \quad (3)$$

and erf(x) is the error function. Noting that erf(x) can be approximated to within 5% by the hyperbolic tangent $\tanh(2x/\sqrt{\pi})$, the expression for the number nucleated reduces to

$$N = \frac{N_a}{1 + \left(\frac{a_c}{a_m}\right)^b} \quad (4)$$

where $b \equiv 4/(\sqrt{2\pi}s)$. Because the number nucleated depends only on the known total aerosol number, s and a_m, and the unknown a_c, the problem reduces to that of solving for a_c.

The radius of the smallest aerosol activated is determined from the balance at maximum supersaturation, $\psi_1 w = \psi_2 C$, between supersaturation forcing by upward motion and supersaturation reduction by condensation, where ψ_1 and ψ_2 can be considered constant during the nucleation process. The condensation rate is more readily expressed in terms of the droplet size distribution rather than aerosol size distribution,

$$C = \frac{4\pi\rho_w}{\rho} \int_0^\infty \frac{dn}{dr} r^2 \frac{dr}{dt} dr \quad (5)$$

where droplet growth is assumed to be due entirely to diffusion of water vapor,

$$r\frac{dr}{dt} = G\left(S - \frac{A}{r} + \frac{Ba^3}{r^3}\right). \quad (6)$$

Here $S \equiv q_v/q_s - 1$ is the supersaturation with respect to a plane surface of liquid water, and the parameters G, A, and B depend on the material properties of water vapor, liquid water, and aerosol (Pruppacher and Klett, 1978).

The condensation rate is expressed in terms of the aerosol size distribution by assuming the droplet radius r at maximum supersaturation is given by the critical radius for activation of the wet aerosol as a cloud condensation nuclei, i.e., at the maximum of the Köhler curve for the aerosol. Then

$$r = Ha^{3/2} \qquad (7)$$

where H depends on the aerosol composition.

Equation (7) is the most significant approximation involved in deriving the nucleation parameterization. By neglecting droplet growth between the time of activation and the time of maximum supersaturation, the droplet size is underestimated. Figure 1 shows the droplet radius at maximum supersaturation as a function of aerosol dry radius as predicted by (7) and as simulated by the detailed size-resolving nucleation model of Edwards and Penner (1988). As expected, the droplet radius is underestimated for aerosols somewhat larger than the smallest aerosol activated (those aerosols with dry radius smaller than a_c are not plotted). However, the size of droplets formed on aerosols much larger than the smallest activated aerosol is actually overestimated. Such large particles fail to grow to the maximum of the Köhler curve by the time maximum supersaturation is achieved. Although droplet size is overestimated by (7) in this case, the number of droplets involved will be very small unless the aerosol number mode radius a_m is much larger than a_c. But if $a_m \gg a_c$ then according to (4) most of the aerosols are activated anyway, so the error in droplet size will have little impact on the number of droplets nucleated. Thus, we expect the most serious errors in the droplet size to occur when $a_m \sim a_c$ and droplet size is underestimated by (7). By underestimating the droplet size, we underestimate the droplet growth rate and hence the condensation rate. The maximum supersaturation is consequently overestimated, which implies too many aerosols activated. In addition, the approximation also leads to the incorrect result that the number nucleated is independent of both the aerosol composition and the mode radius of the aerosol size distribution. Yet the parameterization performs
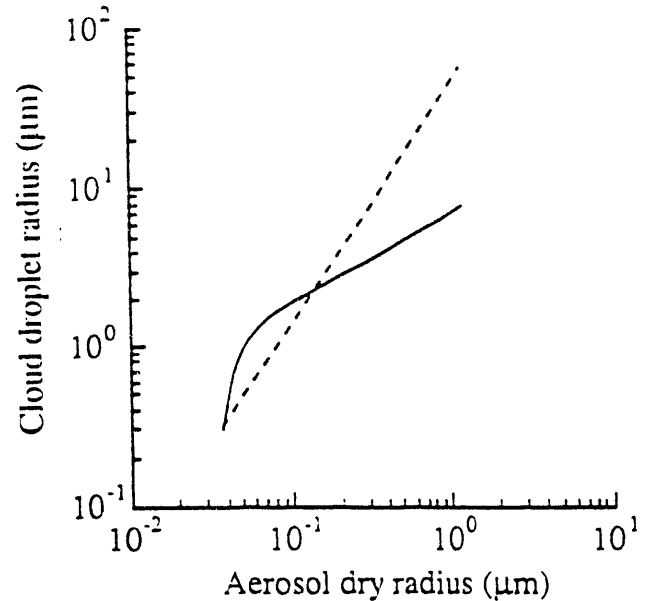

Figure 1: Cloud droplet radius at maximum supersaturation plotted as a function of aerosol dry radius, as parameterized (dashed line) and as simulated by a detailed size-resolving nucleation model (solid line). In the simulations the dry aerosol size distribution is assumed to be log-normal with a number mode radius of 0.05 microns and a standard deviation of 2. The aerosol is assumed to be completely soluble with a total number concentration of 1000 cm^{-3}. The updraft velocity is prescribed at 50 cm s^{-1}.

remarkably well, as we shall demonstrate.

The smallest aerosol that can be activated for a given supersaturation is determined by substituting (7) in (6) with droplet growth set to zero, which yields

$$r_c = Ha_c^{3/2} = \frac{2A}{3S} \qquad (8)$$

Note that (8) is not an approximation, as the smallest particles activated have no time to grow beyond their critical size.

As a brief digression, note that the activation spectrum for a log-normal aerosol size distribution can be determined from (4) and (8), yielding

$$N = \frac{N_a}{1 + \left(\frac{2A}{3Ha_m^{3/2}S}\right)^{2b/3}} \qquad (9)$$

If the fraction of aerosol activated is small then (9) is consistent with the form of Twomey's sim-

ple activation spectrum, $N = cS^k$ with $k = 2b/3$. However, for large S the two activation spectrums diverge significantly, with Twomey's expression yielding nucleation numbers exceeding the total aerosol number concentration.

Applying (7) to the expression for the condensation rate in terms of droplet growth, (5), the condensation rate can be expressed in terms of the aerosol size distribution and a_c,

$$C = U \int_{a_c}^{\infty} \left[\left(\frac{a}{a_c} \right)^{3/2} - 1 \right] \frac{dn}{da} da \quad (10)$$

where

$$U \equiv \frac{8\pi \rho_w G A}{3\rho}. \quad (11)$$

Again assuming a log-normal aerosol size distribution, and equating the reduction in supersaturation by condensation to the increase in supersaturation by adiabatic cooling at maximum supersaturation, a complicated function of a_c results. One final approximation then yields the parameterization for the number nucleated,

$$\frac{N}{N_a} = \frac{1}{1 + cN_a/w} \quad (12)$$

where the coefficient c depends on numerous known factors, including the standard deviation of the aerosol size distribution but not the mode radius a_m nor the aerosol composition.

To account for the dependence of the number nucleated on aerosol mode radius and composition we resort to detailed numerical solutions of the nucleation process. The coefficient c is then scaled so that the difference between the parameterization and the detailed solution is minimized. The values of the scaling factor are listed in Table 1 for aerosol mode radius ranging from 0.01 to 0.2 microns and soluble mass fraction ϵ ranging from 0.01 to 1. The values of the scaling factor are all larger than one and are insensitive to the aerosol mode radius and solubility for aerosol mode radii larger than 0.05 microns and soluble mass fractions larger than 10%. For smaller or less soluble aerosols, the scaling factor can be quite large, which suggests that a single value for the scaling factor for all mode radii and compositions is inappropriate. Variations in the scaling factor can be accounted for in a model by using Table 1 as a look-up table.

Table 1. Scaling factor for coefficient c.

ϵ	a_m (μm)					
	0.01	0.02	0.05	0.1	0.15	0.2
0.01	220.1	66.1	13.3	4.4	2.6	2.0
0.1	67.9	20.9	5.0	2.0	1.3	1.3
0.2	47.8	13.2	3.4	1.5	1.3	1.3
0.5	24.5	7.5	2.3	1.3	1.3	1.2
1.0	15.3	4.9	1.5	1.3	1.2	1.2

Having scaled c to account for aerosol solubility and mode radius, the parameterization of the number nucleated can be compared with the detailed simulations for different vertical velocities and total aerosol numbers. Figure 2 shows a scatter diagram of the number nucleated as parameterized versus the number nucleated as simulated, for aerosol numbers ranging from 50 to 5000 cm^{-3} and vertical velocities ranging from 1 to 500 cm s^{-1}. The errors are 50% or less, which is remarkable considering the range in aerosol number and vertical velocity.

Absolute errors as large as 50% are somewhat worrisome. Fortunately, relative errors are substantially smaller. Figure 3 shows the sensitivity of the number nucleated to changes in aerosol number, $\frac{\partial N}{\partial N_a}$, as simulated against that parameterized. The sensitivity ranges from one for low aerosol concentrations and high vertical velocities, to zero for high aerosol concentrations and low vertical velocities. The parameterization is highly accurate at both of these limits. Errors for intermediate cases are typically 10% and never exceed 20%.

3. COMPARISON WITH TWOMEY'S PARAMETERIZATION

Twomey's (1959) droplet nucleation parameterization is widely used, and hence must be considered a standard against which other parameterizations should be compared. To do so we consider again a log-normal aerosol size distribution. The activation spectrum for such a distribution is given by (9), which differs from Twomey's logarithmic spectrum,

$$N = cS^k \quad (13)$$

unless the fraction nucleated is small. When the fraction nucleated is small the parameters c and k can be determined by comparing (9) and (13). The number nucleated then follows from Twomey's ex-
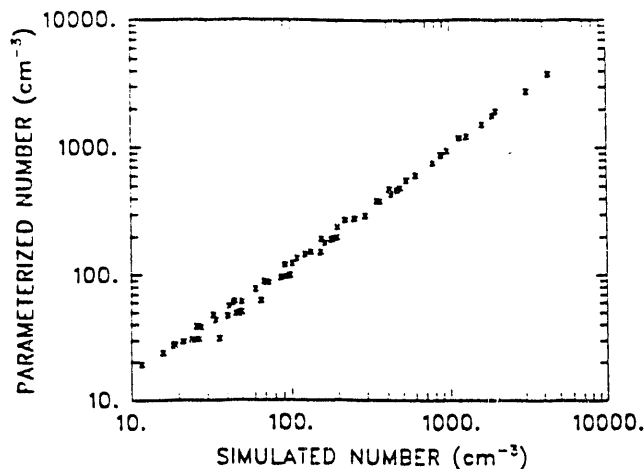
Figure 2: Scatter diagram of the number nucleated as parameterized versus the number nucleated as simulated for aerosol numbers ranging from 50 to 5000 cm^{-3} and vertical velocities ranging from 1 to 500 cm s^{-1}. The parameterized number includes scaling of the coefficient c by a factor dependent only on the aerosol mode radius and composition. The aerosol is assumed to be completely soluble and log-normally distributed with a mode radius of 0.05 μm and a standard deviation of 2. The scaled value of the coefficient c is 0.032 cm^4 s^{-1}.

pression for the maximum supersaturation.

Figure 4 shows a scatter diagram of the number nucleated as parameterized according to Twomey (1959) and as simulated for a log-normal aerosol size distribution (a_m = 0.05 microns, σ = 2) and the same ranges in aerosol number and vertical velocity as above. For cases in which the vertical velocity is large or the aerosol number is small the logarithmic approximation for the activation spectrum breaks down, and Twomey's parameterization predicts droplet numbers exceeding the total aerosol number. To correct this obvious weakness, the parameterized droplet number has been bounded by the total aerosol number. Thus, some of the points in figure 4 that lie on the diagonal (i.e., those for which the parameterized number equals the simulated number) are a result of the bound on droplet number and do not represent skill in the parameterization. The other points indicate errors as large as a factor of three. Com-
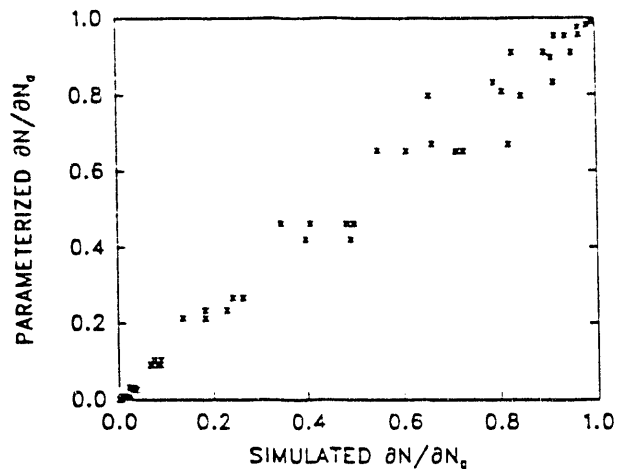
Figure 3: As in Figure 2, but for the sensitivity of the number nucleated to changes in aerosol number, $\frac{\partial N}{\partial N_a}$

paring figures 2 and 4, it is apparent that the new parameterization yields generally somewhat smaller errors than Twomey's parameterization. It is noteworthy that the new parameterization requires no bound on the number nucleated.

4. REALISTIC SIZE DISTRIBUTIONS

One limitation of the parameterization (12) is its reliance on the log-normal approximation for the aerosol size distribution. Observed aerosol size distributions are often multimodal or skewed. To evaluate the applicability of the nucleation parameterization to more general aerosol size distributions, we employ a realistic bimodal aerosol size distribution and compare the number nucleated as parameterized with that calculated by the detailed droplet nucleation model.

Our test of the parameterization utilizes the "group 3" aerosol size distribution of Quinn et al.(1990), illustrated in Figure 5. This distribution is markedly bimodal, with modes at radii of 0.02 and 0.1 μm. The standard deviation of the distribution is 2.0 and mean number radius is 0.05 μm. The parameter values are used for σ and a_m in the parameterization.

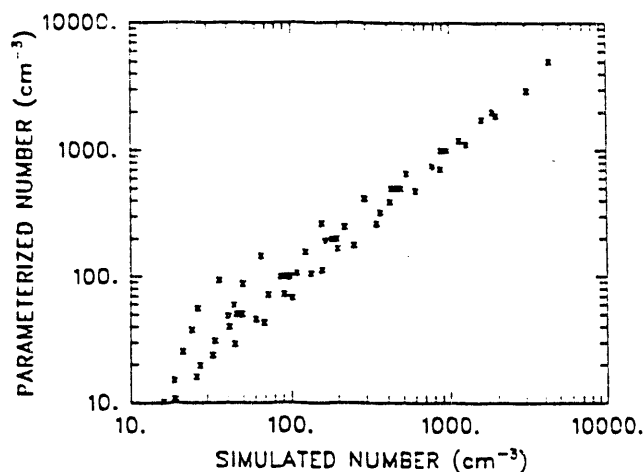Figure 6 shows a scatter diagram for the pa-

Figure 4: As in Figure 2, but for the Twomey (1959) parameterization.

Figure 5: Size distribution of aerosol number for Quinn et al.(1990) "group 3" aerosol.

rameterized versus detailed solutions using the realistic aerosol size distribution. The errors appear to be comparable to those for the log-normal aerosol size distribution. We conclude that moderate bimodality in the aerosol size distribution does not degrade the performance of the nucleation parameterization.

5. CONCLUSIONS

The droplet nucleation parameterization presented here offers several advantages over the popular Twomey scheme. First, it does not require an artificial bound on the droplet number under conditions of high updraft velocity or low aerosol concentration. The sensitivity of droplet number to changes in aerosol number under such conditions is consequently more accurate in the new parameterization than in the Twomey scheme. Second, it relates the number nucleated to more fundamental aerosol variables, such as aerosol number, composition, and the standard deviation and mode radius of the size distribution. Although Twomey's parameterization is more closely related to observations of aerosol activation spectra, equation (9) provides a means of relating such observations to the parameters of the new scheme. Finally, the new parameterization provides an intuitively appealing and simple explanation for the dependence
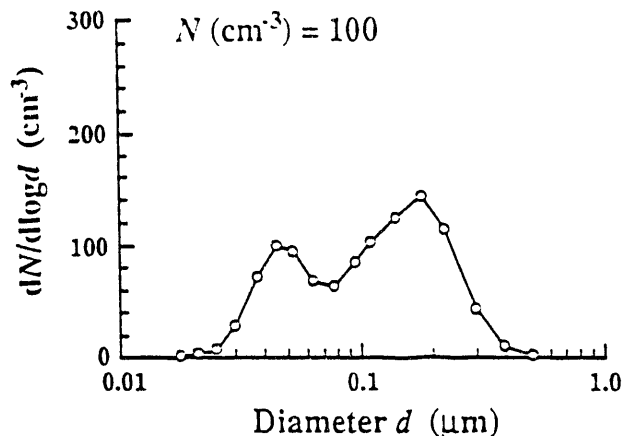
of the fraction nucleated on vertical velocity and total aerosol number concentration.

The treatment of droplet nucleation in this paper has been restricted to the case of a single aerosol species that can be characterized by a single log-normal size distribution. The resulting parameterization was found to be independent of the aerosol composition and mode radius of the size distribution unless a correction factor is applied. The treatment of the more realistic case of an external mixture of aerosol species, in which species of different composition and size distribution compete with each other in the nucleation process, will be addressed in future work. In the meantime, in a paper prepared for submission to Atmospheric Research we provide a more complete derivation of the parameterization for single aerosol species.
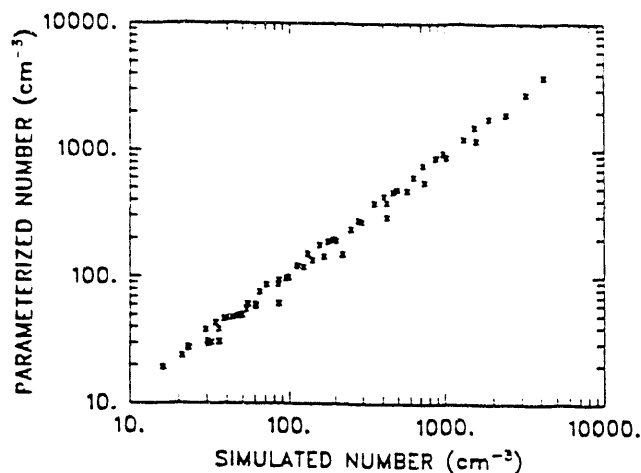
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Figure 6: As in Figure 2 but for the aerosol size distribution illustrated in Figure 5.

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