

# Sensitivity of cloud condensation nuclei activation processes to kinetic parameters

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Received 25 July 2005; revised 5 November 2005; accepted 4 January 2006; published 5 May 2006.

[1] The activation of cloud condensation nuclei to form cloud drops can be a kinetically limited process under some circumstances. Here the sensitivity of particle activation to parameters that are related to kinetic processes, in particular condensational growth and ambient supersaturation, is examined. Specifically, the parameters studied are the thermal and mass accommodation coefficients, the rate of solute dissolution for a particle, and the updraft velocity. Results are based on the equation for condensational growth and simulations using adiabatic parcel models. We conclude that activation is not very sensitive to the thermal accommodation coefficient but is sensitive to the mass accommodation coefficient below some critical value of  $\sim 0.1$  to  $0.001$ . Dissolution rates (defined for 100 nm dry particles) in the range of 10 min to 1 day can significantly affect activation. Sensitivity to updraft velocity uncertainties can be significant and are likely to be more important for polluted rather than clean conditions.

**Citation:** Chuang, P. Y. (2006), Sensitivity of cloud condensation nuclei activation processes to kinetic parameters, *J. Geophys. Res.*, *111*, D09201, doi:10.1029/2005JD006529.

## 1. Introduction

[2] Clouds are an important component of the atmosphere, representing a key uncertainty in our understanding of both weather and climate. The activation of cloud condensation nuclei (CCN) to form cloud droplets is one of the first steps in cloud formation, and therefore is one of the processes that determines the microphysical properties and evolution of a cloud. CCN activation is not always well described by equilibrium theory [Chuang *et al.*, 1997; Nenes *et al.*, 2001]. Model calculations have shown that changes in droplet growth rate can potentially lead to significant changes in cloud properties, in particular cloud effective radius and albedo [Feingold and Chuang, 2002].

[3] Here, the sensitivity of activation-related processes to a variety of “kinetic parameters,” i.e., parameters that are related to droplet growth in a cloud updraft, is studied. Specifically, we focus on: (1) mass and thermal accommodation coefficients, which are relevant to the rate of condensational growth; (2) the timescale of dissolution of solutes, which leads to a time dependence in the equilibrium size of a droplet; and (3) the updraft velocity, an important parameter for determining cloud drop number concentration and cloud supersaturation, both of which are coupled to the total sink of water vapor by droplets growing by condensation.

[4] We define *sensitivity* of some process  $P(x)$  to an independent variable  $x$  as:

$$\text{Sensitivity} = \frac{d\tilde{P}}{d\tilde{x}} \Big|_{x_0} = \frac{d(P/P_0)}{d(x/x_0)} = \frac{d(\ln P)}{d(\ln x)} \Big|_{x_0} = \frac{x_0}{P_0} \frac{dP}{dx} \quad (1)$$

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where  $\tilde{P}$  and  $\tilde{x}$  are dimensionless, referenced to values  $P_0$  and  $x_0$ . According to this definition, a sensitivity of 1 means that a 10% change (or uncertainty) in  $x$  leads to a 10% change (or uncertainty) in  $P(x)$ . A sensitivity of 0.2 means that a 10% change in  $x$  leads to only a 2% change in  $P(x)$ , whereas a sensitivity of 5 means that a 10% change in  $x$  leads to a 50% change in  $P(x)$ .

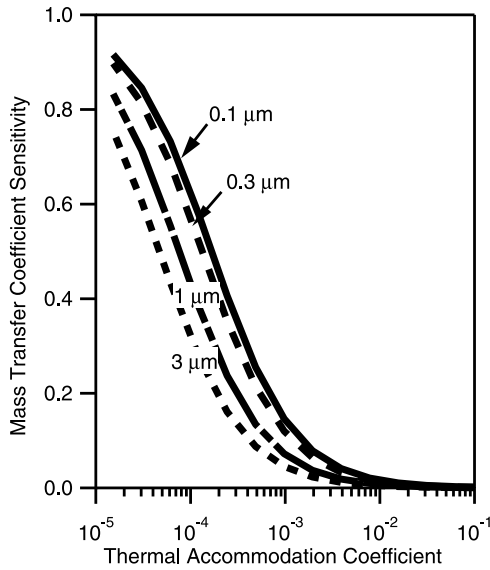
## 2. Sensitivity of the Condensational Growth Rate

[5] We first examine the sensitivity of condensational growth to two parameters, the mass ( $\alpha_c$ ) and thermal ( $\alpha_T$ ) accommodation coefficients. Condensational growth rate is given by the equation [Fukuta and Walter, 1970]:

$$a \frac{da}{dt} \approx \frac{S_\infty - S_{eq}}{\frac{\rho_w RT_\infty}{p_{sat}(T_\infty) D^2 M_w} + \frac{L \rho_w}{k_a T_\infty} \left( \frac{LM_w}{RT_\infty} - 1 \right)} \quad (2)$$

where some of the multiplicative terms have been removed for clarity. See Appendix A for all nomenclature. The numerator represents the driving force for condensation, i.e., the difference between the partial pressure of water far from the droplet ( $S_\infty$ ) and that at equilibrium with the droplet surface ( $S_{eq}$ ). We will examine sensitivity of activation to maximum  $S_\infty$  in section 4. The first term in the denominator represents growth due to diffusion of water vapor and the second term in the denominator represents the change in mass transfer due to latent heat release from condensation, with both terms corrected for noncontinuum effects. We rewrite the growth equation as:

$$a \frac{da}{dt} \approx K (S_\infty - S_{eq}) \quad (3)$$



**Figure 1.** Sensitivity of condensational growth rate to the thermal accommodation coefficient for droplets of different diameters as shown. Both axes are dimensionless.

where we will now call  $K$  the mass transfer coefficient which then has the form

$$K = \left[ \frac{\rho_w RT_\infty}{p_{sat}(T_\infty) D_v^* M_w} + \frac{L \rho_w}{k_a^* T_\infty} \left( \frac{LM_w}{RT_\infty} - 1 \right) \right]^{-1} \quad (4)$$

The modified mass diffusivity is in turn given by [Fukuta and Walter, 1970]:

$$D_v^* = \frac{D_v}{\left[ \frac{a}{a+\Delta} + \frac{D_v}{a\alpha_c} \left( \frac{2\pi M_w}{RT_a} \right)^{1/2} \right]} \quad (5)$$

where now the mass accommodation coefficient,  $\alpha_c$ , is explicitly written. Similarly, the modified thermal diffusivity is given by [Fukuta and Walter, 1970]:

$$k_a^* = \frac{k_a}{\left[ \frac{a}{a+\Delta} + \frac{k_a}{a\alpha_T \rho_a c_{pa}} \left( \frac{2\pi M_a}{RT_\infty} \right)^{1/2} \right]} \quad (6)$$

where the thermal accommodation coefficient,  $\alpha_T$ , is explicitly written.

[6] We show here the calculated sensitivities of the mass transfer coefficient  $K$  to the two accommodation coefficients  $\alpha_T$  and  $\alpha_c$  based on equations (2) to (6). Figure 1 shows the results for  $\alpha_T$ . The sensitivity is a function of droplet size  $a$ , but in all cases it is clear that the condensational growth of droplets 0.3  $\mu\text{m}$  in radius and above exhibit very little sensitivity to  $\alpha_T$  down to  $\alpha_T \sim 10^{-2}$ , and only grow above 0.1 for  $\alpha_T$  values  $10^{-3}$  and smaller. It is typically argued that  $\alpha_T$  is close to unity [Rogers and Yau, 1989], an assertion that has some experimental support [Shaw and Lamb, 1999]. These results show that it is not important to know accurately the value of  $\alpha_T$  in order to predict accurately the condensational growth rate because the sensitivity of  $K$  to  $\alpha_T$  is very small in this range of  $\alpha_T$ . The explanation of this result is that the heat transfer-related term in equation (2) (i.e., the second term in the denominator)

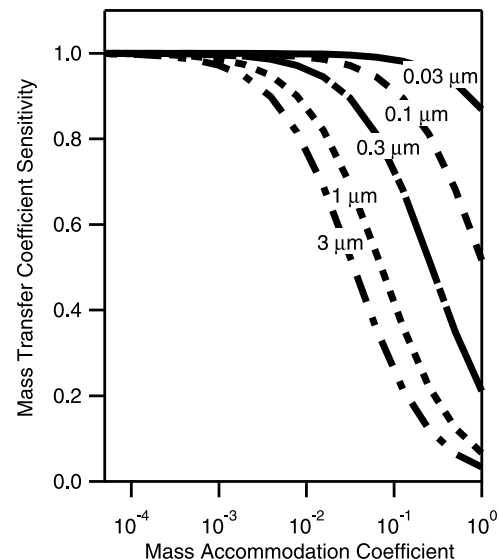
does not become significant relative to the other (diffusion) term except for these small values of  $\alpha_T$ . Physically, the interpretation is that the latent heat released does not substantially change the surface temperature of the drop. The lack of sensitivity of growth rates to  $\alpha_T$  has been understood in the community for a long time; they are presented here partly to show how small  $\alpha_T$  must be to be significant, but primarily for completeness and to serve as a contrast with the results for  $\alpha_c$ . The results in Figure 1 are weakly sensitive to the choice of  $T$  and  $\alpha_c$  used during this calculation, but the main conclusions remain the same.

[7] Figure 2 shows the sensitivity of  $K$  to  $\alpha_c$ . Here, we see that depending on droplet size, the sensitivity asymptotes at a value of 1 given sufficiently small  $\alpha_c$ , with values on the order of  $10^{-1}$  to  $10^{-3}$  depending on droplet size. In contrast with  $\alpha_T$ , however, laboratory measurements of  $\alpha_c$  show that these values can be as low as  $\sim 10^{-5}$  in the presence of organic films on the surface of the droplets [Rubel and Gentry, 1984; Seaver et al., 1992], and evidence that such particles may exist in the atmosphere has been presented [Chuang, 2003]. See Barnes [1986] and Chuang [2003] for more comprehensive reviews of this subject. Note that not all studies seeking to observe such effects yielded positive results [e.g., Cruz and Pandis, 1998]. These results show that the uncertainty in droplet growth rate is equal to the uncertainty in  $\alpha_c$  (i.e., sensitivity = 1) if  $\alpha_c$  is below  $10^{-1}$  to  $10^{-3}$ , depending on droplet size. To what extent such values of  $\alpha_c$  exist for real CCN is not known, but these results show that it is important that future work addresses this issue.

### 3. Sensitivity to Dissolution Rate

[8] The activation of cloud droplets for simple salts can be described by classic Köhler theory [e.g., Pruppacher and Klett, 1997]:

$$\ln S = \frac{4M_w \sigma_w}{RT \rho_w D_p} - \frac{6n_s M_w}{\pi \rho_w D_p^3} = \frac{A}{D_p} - \frac{B}{D_p^3} \quad (7)$$



**Figure 2.** Sensitivity of condensational growth rate to the mass accommodation coefficient for droplets of different diameters as shown. Both axes are dimensionless.

From this equation, the critical supersaturation  $S_c$  of a particle can be shown to be:

$$\ln S_c = \left( \frac{4A^3}{27B} \right)^{1/2} \quad (8)$$

Normally, it is assumed that the number of moles in solution,  $n_s$ , is a constant, which implies that  $B$  is constant. In this section, we explore what happens if dissolution is not instantaneous, but instead is associated with some finite timescale.

[9] It has been hypothesized that the dissolution of some solutes can require long timescales despite the fact that complete dissolution is the thermodynamically favorable (i.e., equilibrium) state [Hegg *et al.*, 2001; Shantz *et al.*, 2003]. If the solute in a growing droplet is unable to dissolve sufficiently rapidly to maintain a saturated solution (i.e., is kinetically limited from maintaining equilibrium), then this decreases the amount of solute present  $n_s$ , which increases the particle critical supersaturation  $S_c$  and can therefore prevent the particle from activating even if an equilibrium calculation says it would. Here, we explore the parameter space for dissolution rates in order to study what range of dissolution timescales can cause appreciable changes in activation. We note that because this is an exploratory study, we seek an order-of-magnitude result for the range of dissolution timescales that are significant for this problem. Also, it is typically believed that a larger fraction of particles are present in their deliquesced state (either stable or metastable) rather than their crystallized state. Dissolution timescales are not relevant to the activation of such particles because they have no solid phase.

[10] We first define dissolution timescale in a very simple manner:  $t_{sol}$  is the time required for a dry 100 nm solute particle to fully dissolve. We also assume that the rate limiting step for dissolution is a solid-phase surface process, which implies that the rate of dissolution is proportional to surface area and independent of the solute concentration in solution. Given these assumptions, the rate of dissolution of a solid solute particle can be expressed as:

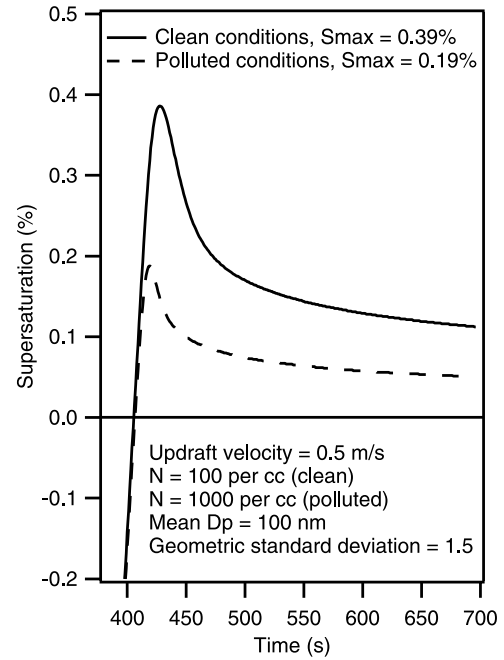
$$\frac{dV_{sol}}{dt} = kA_{sol} \quad (9)$$

Assuming a spherical solute inclusion, it is easy to show that equation (9) can be expressed as:

$$\frac{dD_{sol}}{dt} = k \quad (10)$$

where  $k$  has absorbed more factors but remains a constant. Therefore equation (10) shows that the decrease in particle diameter due to dissolution proceeds at a constant rate, and thus the rate constant for dissolution,  $k$ , is inversely proportional to  $t_{sol}$ , which we have defined as the time for a dry 100 nm particle to dissolve. We will express dissolution rate using  $t_{sol}$  since it is more physically intuitive.

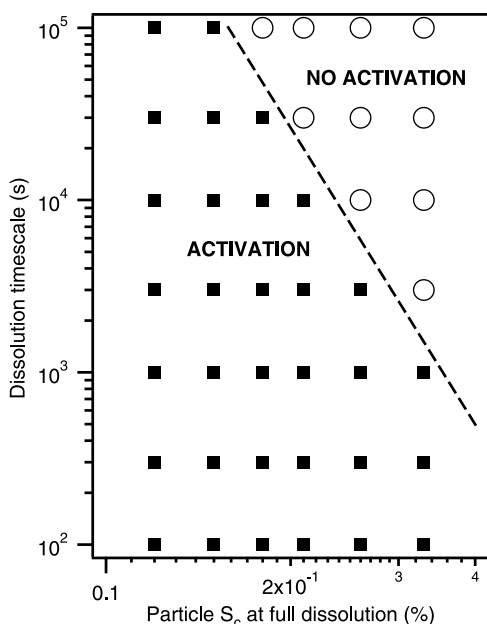
[11] To study the effect of dissolution rate, we use results from an adiabatic 1-D cloud parcel model (model adapted from Feingold and Heymsfield [1992]). Two different cases



**Figure 3.** Time profile of relative humidity for two conditions, clean and polluted, used for calculating relevant dissolution timescales  $t_{sol}$ . For times less than 400 s, the RH increases linearly with time, with initial condition of RH = 90% at  $t = 0$  s.

(which we will term “clean” and “polluted”) are used to calculate the relative humidity as a function of time in a rising air parcel, as shown in Figure 3. The “clean” case has fewer particles and therefore a higher maximum supersaturation. We then consider whether or not a single particle in this environment will activate as a function of the particle dry size (expressed as particle equilibrium critical supersaturation,  $S_c$ ), and  $t_{sol}$  for that particle. We assume that the model solute has similar properties (molecular mass, density) to NaCl, but the results here do not depend significantly on the solute properties chosen.

[12] A simple model was used to determine whether or not a particular particle activated. Particles are initially assumed to be at equilibrium at a RH of 90%. Using the desired time profile of RH (either clean or polluted), the particle solute content  $n_s$  in the next time step is determined by the dissolution rate  $t_{sol}$ , with the constraint that it cannot exceed the equilibrium  $n_s$  at that RH. The calculation proceeds until (1) activation is achieved, defined as the ambient supersaturation  $S$  exceeding the particle supersaturation, or (2) the simulated cloud updraft exceeds 270 s after peak supersaturation. In principle, (1) is not a perfect criterion for activation. It is possible that a particle that satisfies (1) at some moment could fail that same criterion a short period of time later, thus de-activating. This could occur if a particle activates according to (1) when ambient  $S$  is decreasing. Once activated, the particle grows very quickly by condensation, and this dilution reduces the particle equilibrium  $S$ . However, if the ambient  $S$  decreases even more quickly, this would lead to net evaporation and eventually de-activation (such behavior was observed in simulations by Nenes *et al.* [2001]). For realistic adiabatic



**Figure 4.** Grid showing those combinations of dissolution timescale  $t_{sol}$  and particle equilibrium critical supersaturation ( $S_c$ ) that are kinetically inhibited from activating. Results are for clean aerosol conditions (see Figure 3).

situations, however, this seems unlikely, though updrafts with significant entrainment to dilute ambient  $S$  might see such an effect. Also, the criterion chosen in (2) is somewhat arbitrary, but will not significantly affect the model predictions. It should also be noted that these simulations represent a single particle in this updraft environment, surrounded by other particles that do not exhibit dissolution kinetics. If, instead, all particles exhibited some finite  $t_{sol}$ , the total water uptake would decrease at any given time, and therefore  $S_{max}$  for that updraft would increase, rather than being a constant as assumed here.

[13] Figure 4 shows the results for the clean case, which exhibits a maximum ambient supersaturation,  $S_{max}$ , of 0.39%. The shortest  $t_{sol}$  where any inhibition of activation is observed is  $3 \times 10^3$  s ( $\sim 1$  hr), and this occurs for particles whose  $S_c$  is 0.33%, which is very close to  $S_{max}$  and therefore require only a small decrease in solute concentration to be unable to activate. The discrepancy between  $S_{max}$  of 0.39% and  $S_c$  of 0.33% is due to the limited resolution of the calculations. The range of particles that are inhibited from activating because of dissolution kinetics increases as  $t_{sol}$  increases, with particles in the range of  $S_c$  from 0.18% to 0.39% inhibited for the largest  $t_{sol}$  considered here,  $10^5$  s ( $\sim 1$  day). This represents a range of dry diameters from 60 to 90 nm.

[14] Figure 5 shows the results for the polluted case, which has  $S_{max} = 0.19\%$ . In general, the lower supersaturation environment leads to a broader range of particles that are kinetically inhibited from activating. The lowest value of  $t_{sol}$  that prevents activation is  $10^3$  s ( $\sim 15$  min). At the largest  $t_{sol}$  of  $10^5$  s, particles with  $S_c$  between 0.09% and 0.19% do not activate, corresponding to particles in the diameter size range of 180 to 280 nm.

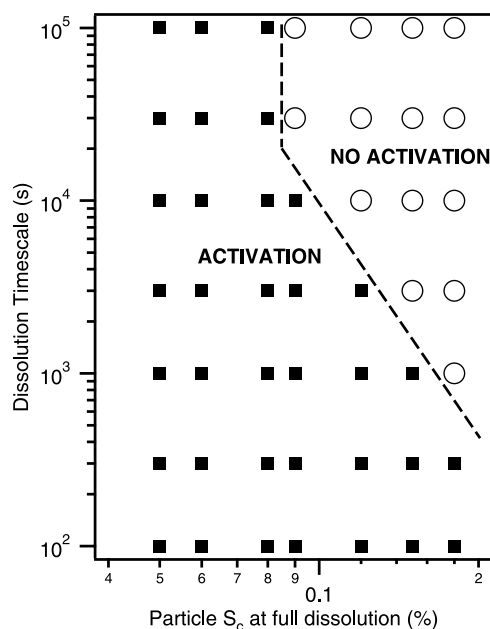
[15] Clearly, these results depend on the choice of parameters used for these specific calculations (e.g., updraft

velocity, aerosol number and size distribution), and many more calculations could be performed to better map out the parameter space. However, the purpose here is to get some order-of-magnitude estimate of the kinetic dissolution timescales that may be relevant to the activation of real aerosol under cloud conditions, and perhaps guide future experiments. These results suggest that the minimum  $t_{sol}$  that is relevant is on the order of 10 min, while values of  $\sim 1$  day would lead to significant changes in activation behavior over a range of particle size. Intuitively, these values do not seem out of the realm of possibility, but whether or not atmospheric aerosol contains solutes that exhibit such timescales remains unknown. These results serve as a useful guide for future experiments.

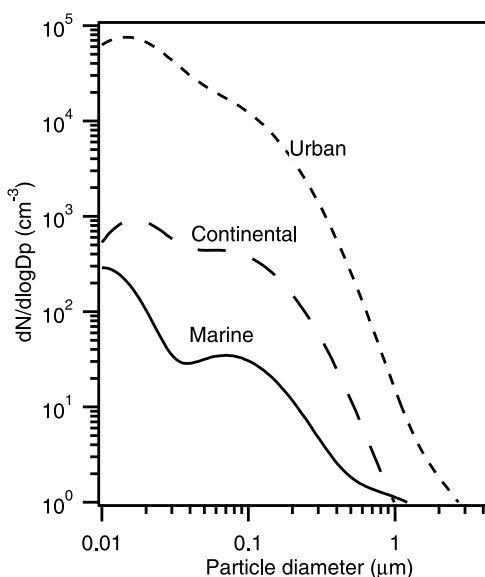
#### 4. Sensitivity to Uncertainties in Updraft Velocity ( $w$ )

[16] Cloud droplet closure experiments compare measured cloud droplet number (CDN) concentrations with that predicted from measured aerosol size distribution and chemical composition along with measured cloud updraft velocity ( $w$ ). The adiabatic CDN cannot be predicted using an equilibrium model since it depends on the balance of two time-dependent processes: the time evolution of supersaturation, which depends primarily on  $w$  and temperature; and the rate of water vapor uptake by condensation onto existing particles.

[17] Accurately measuring  $w$  from aircraft is a nontrivial exercise. A typical estimated  $w$  uncertainty is  $\sim 0.1$  m/s [Khelif *et al.*, 1999; Kalogiros and Wang, 2002]. In this section, we study the sensitivity of  $S_{max}$  and CDN to the uncertainty in measured  $w$  using an adiabatic parcel model (we refer the reader to Lance *et al.* [2004] and Rissman *et*



**Figure 5.** Grid showing those combinations of dissolution timescale  $t_{sol}$  and particle equilibrium critical supersaturation ( $S_c$ ) that are kinetically inhibited from activating. Results are for polluted aerosol conditions (see Figure 3).

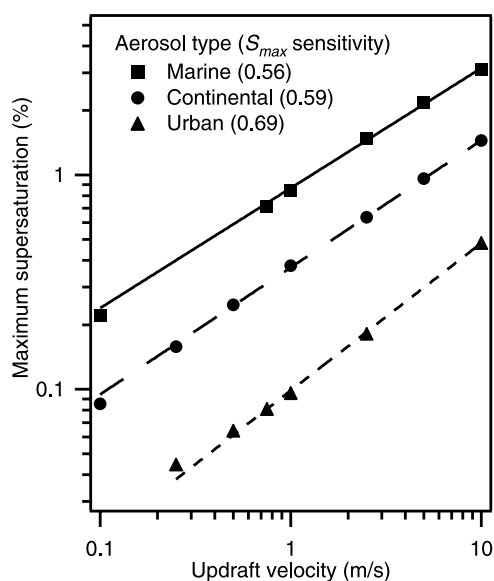


**Figure 6.** Aerosol size distributions [Whitby, 1978] used for calculating sensitivities to updraft velocity (results shown in Figures 7 and 8).

*al.* [2004] for alternate approaches to this topic). The simulations are performed using three different aerosol size distributions, which are the model marine, clean continental, and urban distributions proposed by Whitby [1978]. Each aerosol is assumed to be composed only of ammonium sulfate. The aerosol size distributions are plotted in Figure 6. A range of  $w$  values between 0.1 to 10 m/s are modeled. Activation of CCN to form cloud droplets is determined kinetically, but under these conditions, any deviation from the predicted equilibrium CDN is very small.

[18] Cloud parcel model predictions of  $S_{\max}$  are calculated for a given updraft velocity and aerosol size distribution, as shown in Figure 7. The slope of the curve of  $\log S_{\max}$  versus  $\log w$  is a measure of the sensitivity of  $S_{\max}$  on  $w$  (see equation (1)). Figure 7 shows that such a curve fits the model results very well ( $R^2 > 0.99$ ) for all three aerosol cases. The modeled sensitivity of  $S_{\max}$  to  $w$  is 0.56, 0.59, and 0.69 for the marine, continental and urban aerosol over the range of  $w$  studied. These values are fairly consistent with each other despite the large differences in the aerosol size distributions, suggesting that the sensitivity of  $S_{\max}$  to  $w$  is not a strong function of aerosol type. A careful study in order to understand exactly why these cases differ is not presented here because, again, the goal is to estimate the sensitivities, and therefore deviations on the order of 10 to 20% are not significant.

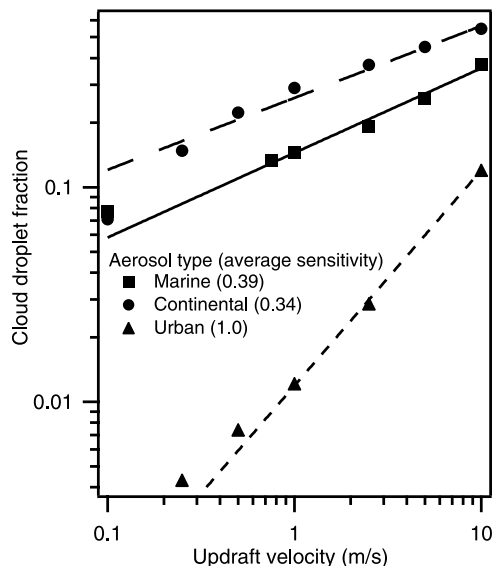
[19] These sensitivity results suggest that the uncertainty in predicting  $S_{\max}$  can be very significant under weak updraft conditions, such as stratiform clouds, where mean  $w$  can be in the range of 0.1 to 0.3 m/s. The uncertainty in measured  $w$  of 0.1 m/s would then represent a  $w$  uncertainty of 100% to 33%, which translates into uncertainties in  $S_{\max}$  of 60% to 20%, assuming a sensitivity of 0.6. Understanding the microphysical properties of stratiform clouds is climatically important because their albedo is sensitive to the presence of anthropogenic aerosol [Charlson *et al.*, 1992]. For more strongly convective clouds, where  $w$  is 1 m/s or larger, these results predict that the uncertainty in measured



**Figure 7.** Maximum supersaturation as a function of updraft velocity for different aerosol size distributions. The slope of the curve represents the sensitivity of  $S_{\max}$  to  $w$  and is given in parentheses for each aerosol type.

$w$  will lead to uncertainties in  $S_{\max}$  of less than 6% in regions that are well represented by a single updraft.

[20] The sensitivity of CDN to the uncertainty in  $w$  is also studied. The results are presented as the fraction of particles that form cloud drops (rather than absolute cloud drop concentrations), as shown in Figure 8. The average sensitivity of cloud drop fraction for the marine, continental and



**Figure 8.** Fraction of particles that activate to form cloud drops as a function of updraft velocity for different aerosol size distributions. The slope of the curve represents the sensitivity of the cloud drop fraction to  $w$  and is given in parentheses for each aerosol type.

urban aerosol cases are 0.39, 0.34 and 1.0 based on the best fit curves in Figure 8. The much larger sensitivity for the urban case is a striking result. The likely explanation for this is that  $S_{\max}$  is much smaller in the urban case (Figure 7), and this means that the minimum particle size for activation is much larger (since the aerosol is assumed to be internally mixed). For the urban case, the slope of the aerosol size distribution is fairly steep in the size range of 0.05 to 0.5  $\mu\text{m}$ . As  $w$  increases,  $S_{\max}$  increases, and therefore the minimum size for activation decreases, but because of this steep slope, the number of particles which can activate increases very quickly, leading to a large change in CDN, and therefore in cloud drop fraction. For the other two aerosol cases, the values of  $S_{\max}$  are larger, which means that the minimum size is smaller, and the slope of the aerosol size distribution at these sizes is generally flat or sloping in the opposite direction, leading to a much smaller sensitivity. For the continental case, the fitted curve does not seem to be the best representation of the model results, as there is a clear concavity to the results, i.e., sensitivity is higher for low  $w$  than high  $w$ . The reason this occurs is likely the same explanation as for the increased sensitivity in the urban case discussed above.

[21] Given these variations, the general conclusion is that CDN is reasonably sensitive to  $w$  (sensitivity  $\sim 0.3$  to  $0.4$ ) for the cleaner aerosol cases, with increased sensitivity ( $\sim 1$ ) for the polluted case. These results are consistent with Feingold [2003], who showed that drop effective radius exhibits stronger sensitivity to  $w$  for higher aerosol extinctions. The exact value of the sensitivity appears to depend quite strongly on the shape of the aerosol size distribution, which is in contrast to the sensitivity results for  $S_{\max}$ , which appear to be fairly insensitive to the aerosol size distribution.

## 5. Summary

[22] A number of processes related to cloud droplet activation that involve time dependence are studied. The focus is on estimating a reasonable value for the sensitivity of these processes to parameters that are not well characterized for real atmospheric aerosols. Because many of the parameters are not well constrained, a much more detailed analysis of the sensitivity is not attempted here. The main conclusions of this work are:

[23] 1. Droplet growth rate is not very sensitive to the thermal accommodation coefficient for the range of values that are believed to be realistic. Improved constraints of  $\alpha_T$  are therefore not of any significant value for understanding droplet activation.

[24] 2. Droplet growth rate is sensitive (sensitivity  $\sim 1$ ) to the mass accommodation coefficient below some critical value whose range is from  $10^{-1}$  to  $10^{-3}$ . Whether these values are exhibited for atmospheric aerosols remains an open question.

[25] 3. If solute dissolution exhibits timescales on the order of 10 min to 1 day, then this can significantly affect whether or not a nondeliquesced particle composed of this solute will activate.

[26] 4. The sensitivity of maximum parcel supersaturation to uncertainties in measured updraft velocity  $w$  are  $\sim 0.6$  to  $0.7$ , and appears not to be strongly dependent on the aerosol selected.

[27] 5. The sensitivity of the fraction of particles that activate to  $w$  is in the range of  $\sim 0.3$  to  $1$ , depending on the aerosol size distribution chosen.

## Notation

$A$	Köhler equation term
$A_{sol}$	surface area of solute
$a$	droplet radius
$B$	Köhler equation term
CDN	cloud droplet number
$c_{pa}$	heat capacity of air at constant pressure
$D_p$	particle diameter
$D_{sol}$	solid solute particle diameter
$D_v$	water vapor diffusivity
$k$	dissolution rate constant
$K$	mass transfer coefficient for condensational growth
$k_a$	thermal diffusivity
$L$	latent heat of evaporation
$M_w$	molecular weight of water
$n_s$	moles of solute
$P$	some process that depends on independent variable $x$
$p_{sat}$	saturation water vapor pressure
$r$	particle radius
$R$	ideal gas law constant
$S$	supersaturation
$S_c$	critical supersaturation for a particle
$S_{\max}$	maximum supersaturation achieved in a rising adiabatic air parcel
$T$	temperature
$t$	time
$t_{sol}$	time constant for dissolution
$V_{sol}$	volume of solute
$w$	updraft velocity
$x$	some independent variable
$\Delta$	flux correction length scale
$\alpha_c$	mass accommodation coefficient
$\alpha_T$	thermal accommodation coefficient
$\rho_a$	density of air
$\rho_w$	density of liquid water
$\sigma_w$	surface tension of water

## Subscripts

$\infty$	infinity, a distance far away from the droplet
$a$	air
$eq$	at equilibrium with the droplet surface
$sat$	at saturation
$sol$	solute
$v$	vapor
$w$	water

## Superscripts

*	modified diffusivities
$\sim$	denotes a normalized quantity

[28] **Acknowledgments.** This work was partly funded by NASA under grant NNG04GE16G. The author also thanks G. Feingold (NOAA ETL) and A. Nenes (Georgia Tech) for their generous contributions of model output and useful discussion, B. Ervens (Colorado State Univ./NOAA), and one anonymous reviewer for useful comments.

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