SIO 217A: Cloud Droplet Growth

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1 Introduction

Atmospheric thermodynamics focuses on water and its transformations. Advanced topics are usually focused on phase transitions of water, homogeneous and heterogeneous nucleation, and the role of supersaturation on the formation of ice crystals and cloud droplets. Among those sub-topics, cloud nucleation has been a large area of research throughout the twentieth century.

Nucleation process is an energy balance between surface tension and latent heat. When the latent heat associated with condensation is enough to counteract the surface tension on the drop, the droplet is nucleated. However, the surface tension of a small water droplet can be very large. In order to break the high energy barrier on the surface for homegeneous nucleation, high values of supersaturation ($>> 100\%$) are required.

In the Earth's atmosphere supersaturation typically only slightly exceeds 100%, e.g., 100.05%. Therefore cloud droplets are formed via hetereogeneous nucleation, which involes cloud condensation nuclei (CCN), a subset of aerosol particles. The presence of CCN, such as sodium chloride (NaCl), lower the equilibrium vapor pressure of the water relative to pure water, thereby reducing the effects of surface tension and enabling the formation of cloud droplets.

In this study we evaluate the effects of CCN on droplet growth within warm clouds, which form due to vapor-to-liquid condensation. Water droplets are assumed to be isolated and spherical, and form due to the diffusion of water vapor to the surface in a steady state diffusion field.

2 Cloud Droplet Growth

2.1 Model Description

In order to model the cloud droplet growth as a function of time, we have used Equation (3) from Curry and Webster [1999]. The approximation assumes that a droplet has the shape of a sphere that develops around a saturated nucleus core. Furthermore, the environment conditions (temperature, pressure, and supersaturation) are assumed to be constant, and the solute and curvature effects are neglected, which will be explained in more detail. The initial approximation is obtained by applying diffusion equations to the droplet, as shown in Equation (1).

$$\frac{dm}{dt} = 4\pi r D_v \left(\rho_v(\infty) - \rho_v(r)\right) \tag{1}$$

Adding the latent heat by condensation, shown in Equation (2), gives the basis of the droplet growth rate model.

$$\frac{dQ}{dt} = -L_{lv}\frac{dm}{dt} = -4\pi r\kappa \left(T(\infty) - T(r)\right) \tag{2}$$

These equations are combined to approximate the growth rate of a droplet by diffusion as

$$r\frac{dr}{dt} = (S-1)\left[\frac{L_{lv}^2\rho_l}{\kappa R_v T^2} + \frac{\rho_l R_v T}{e_s(T) D_v}\right]^{-1} = \frac{S-1}{K+D} \tag{3}$$

where K and D are the thermodynamic terms associated with heat conduction and diffusion of water vapor, respectively. This differential equation is the same as Equation 5.26 in Mason [1971]. S is the saturation ratio, ρ_l the liquid density, e_s the saturation pressure in the droplet and T is the environment temperature.

Assuming the atmospheric ambient conditions are constant (i.e. T, S, K, and D are constant), then Equation (3) can be integrated to get

$$r(t) = \left[r_0^2 + \frac{2(S-1)}{K+D}(t-t_0)\right]^{1/2}, \tag{4}$$

which can then be rearranged to find t

$$t = (r^2 - r_0^2)\frac{K+D}{2(S-1)} \tag{5}$$

Using the values from Table 1, the main results in Table 2 can be approximated. But to include the effects of initial mass, curvature and the solute, we must modify our model.

One of the main properties of Equation (5) is that it does not depend on the nucleus mass, which directly contradicts with Table 2. This led us to the first modification on the approximation Equation (5). Rather than starting from $r = r_0 = 0.75\ 10^{-6}\mu m$ and $t_0 = 0$ as given in the conditions of Table 2, we have altered the definition of the starting radius as the radius of the nucleus without any water. We haven't changed the value of r_0, as this was given as one of the constants. But, we have defined t_0 as the time the radius of the droplet develops to r_0, which is definitely not 0.

To calculate the initial radius, we have assumed that the nucleus is a perfect sphere. Thus, the initial radius can calculated as

$$\rho_{NaCl} \frac{4}{3}\pi r_i^3 = m_{NaCl}. \tag{6}$$

We have used ρ_{NaCl}=2160 kg m^{-3}. This adjustment introduced an effect of the nucleus mass and decreased the overall error of the first given approximation.

The secondary improvement was on e_s. The approximation assumed e_s to be a constant, but normally it is not. Especially for small r values, Best [1951] states that e_s deviates from its average value significantly. Since our starting point involves small values of r, we have applied two modifications on e_s. The first adjustment is the usage of Raoult's law to incorporate the effect of surface tension, introducing an additional factor depending on both r and the mass of the nucleus. This factor is shown in Equation (7). i is a constant depending on the molecular structure of the nucleus core and $i = 2$ for NaCl. $e_{s\infty}$ is the saturation pressure of the environment (at a virtual radius of $r = \infty$).

$$\frac{e_s}{e_{s\infty}} = \frac{n_{H2O}}{in_{solt} + n_{H2O}} = \frac{\frac{4}{3}\pi r^3 \rho_{H2O}}{M_{H2O}} \left[i\frac{m_{solt}}{M_{solt}} + \frac{\frac{4}{3}\pi r^3 \rho_{H2O}}{M_{H2O}} \right] \tag{7}$$

The second modification is the introduction of the effect of curvature:

$$\frac{e_s}{e_{s\infty}} = \exp\left(\frac{2\sigma_{lv}}{\rho_l R_v T r}\right). \tag{8}$$

The combined effect of surface tension and the curvature is then

$$e_s(r,T) = e_{s\infty} \left[\frac{\frac{4}{3}\pi r^3 \rho_{H2O}}{M_{H2O}} \right] \left[i\frac{m_{solt}}{M_{solt}} + \frac{\frac{4}{3}\pi r^3 \rho_{H2O}}{M_{H2O}} \right]^{-1} \exp\left(\frac{2\sigma_{lv}}{\rho_l R_v T r}\right) \tag{9}$$

Throughout our calculations and simulations, we assume T to be constant as its deviations from its initial value is negligible. This modification in e_s directly affects our D value in Equation (3).

A final modification has been applied on the numerator $(S - 1)$ of Equation (3) as recommended by Mason [1971]. By adding the curvature effects we find

$$(S-1) \to (S-1) + \frac{2\sigma_{lv}}{\rho_l R_v T r} - \frac{in_{solt}}{in_{solt} + n_{H2O}} \tag{10}$$

where n represents the mole number of the material in the subscript.

Although this equation increases the accuracy of the results, it depends itself on r. This means that the simple quadratic solution in Equation (5) can not be used any more and we need a numerical integration method to solve the differential equation in Equation (3). Using the explicit Euler method we are able to calculate r iteratively as

$$r_{k+1} = \left[\frac{S - 1 + \frac{2\sigma_{lv}}{\rho_l R_v T r} - \frac{n_{solt}}{in_{solt} + n_{H2O}}}{K(T) + D(r_k, T)} \right] \frac{\Delta t}{r_k} \tag{11}$$

The initial condition at $k = 0$ is $r = r_0$ which was calculated as the radius of the nucleus core itself.

Although this approximation is close to the values in Table 2, it has its drawbacks with respect to reality. The first limitation is that we have assumed the temperature to be constant throughout the droplet and same as the environment at all times. The second drawback is that we have assumed that the environment properties like κ, D_v, L_{lv} stay constant, which introduces an extra error into our approximation.

2.2 Model Parameters

K is the thermodynamic term related to the latent heat release due to condensation and diffusion of heat away from the droplet. D is the vapor diffusion term related to the diffusion of water vapor onto the growing droplet. Plus, S is saturation ratio. If $S < 1$, then $dr/dt < 0$, which describes the evaporation of a cloud drop. And if $S > 1$, then $dr/dt > 0$ and the droplet grows by condensation.

Water vapor is transferred to the drop by molecular diffusion as long as the vapor pressure surrounding the drop exceeds the saturation vapor pressure of the drop. As water condenses on the drop, latent heat release, the drop becomes warmer than the environment, and heat is diffused away from the drop. Condensation can thus be considered as a double diffusive process, with water vapor diffused towards the drop and heat diffused away from the drop. Therefore cloud droplet growth is a balance between diffusion of water vapor and convection.

3 Results of Modeling Study

Figure 1 shows a visual comparison between the droplet growth rate modeled by Equation (4) with $r_0 = 0.75\mu m$, $T = 273K$, $p = 900mb$, and $(S - 1) = 0.05\%$. A major assumption of Equation (4) is that the curvature and solute effects are neglible once the droplet grows beyond a few microns. The figure shows general agreement between the model and values from Table 5.5 of Curry and Webster [1999]. But by considering curvature and solute effects, Equation (11) is able to recreate the values from Table 5.5 of Curry and Webster [1999] (see Figure 2).

To study the sensitivity of the model, we varied T and $(S - 1)$, separately, while holding all other parameters constant. Figure 3 and 4 show the results. The increased values of T correspond physically to increasing the heat diffused away from the droplet (decreased K) and decreasing the amount of water vapor diffused into the droplet (increased D). Similarily, increased values of $S - 1$ increase the droplet growth rate due to an increased amount of water vapor, which lowers the amount of energy needed to achieve activation.

4 Conclusion

We have evaluated two droplet growth models for clouds that form due to vapor-to-liquid condensation. The first model, taken from Curry and Webster [1999], is a simplified model that ignores curvature and solute effects, but captures the general behavior of the droplet growth. The second model includes curvature and solute effects, and is able to recreate the values from Table 5.5 of Curry and Webster [1999]. A sensitivity analysis shows a positive correlation between droplet growth, and both temperature and supersaturation.

Both models describe droplet growth by diffusion only and therefore do not take into account the effect of collisions and coalescence between droplets, which become important once $r > 20\mu m$. Additionally, the models do not consider changes in S over time as less water vapor is available. A more robust version of the models should incorporate the effects of both coalesence and changing S.

References

A.C. Best. The size of cloud droplets in layer-type cloud. *Royal Meteorological Society*, 77:241–248, 1951.

J.A. Curry and P.J. Webster. *Thermodynamics of atmospheres and oceans*. Academic Press, San Diego, 1999.

B.J. Mason. *Physics of clouds*. Clarendon Press, Oxford, 2nd edition, 1971.

Table 1: Parameters for droplet growth rate.

Parameter	Value	Units	Notes
$S-1$	0.05	%	
p	900	mb	
T	273	K	
r_0	0.75	μm	
L_{lv}	2.5x10^6	$J\ kg^{-1}$	pure water at $T = 273K$
ρ_l	1000	$kg\ m^{-3}$	pure water at $T = 273K$
R_v	461	$J\ kg^{-1}K^{-1}$	
κ	2.4x10^{-2}	$J\ m^{-1}s^{-1}K^{-1}$	$T = 273K$
D_v	2.21x10^{-5}	m^2s^{-1}	$T = 273K$, $p = 1000mb$
	2.46x10^{-5}	m^2s^{-1}	$T = 273K$, $p = 900mb$
$e_s(T)$	6.15	mb	$T = 273K$

Table 2: Growth rate of droplets with nuclei of NaCl, $(S-1)$=0.05%, p=900mb, T=273K, and r_0=0.75μm recreated from Table 5.5 of Curry and Webster [1999].

m [g]	10^{-14}	10^{-13}	10^{-12}
r [μm]		t [s]	
1	2.4	0.15	0.013
2	130	7.0	0.61
4	1,000	320	62
10	2,700	1,800	870
20	8,500	7,400	5,900
30	17,500	16,000	14,500
50	44,500	43,500	41,500

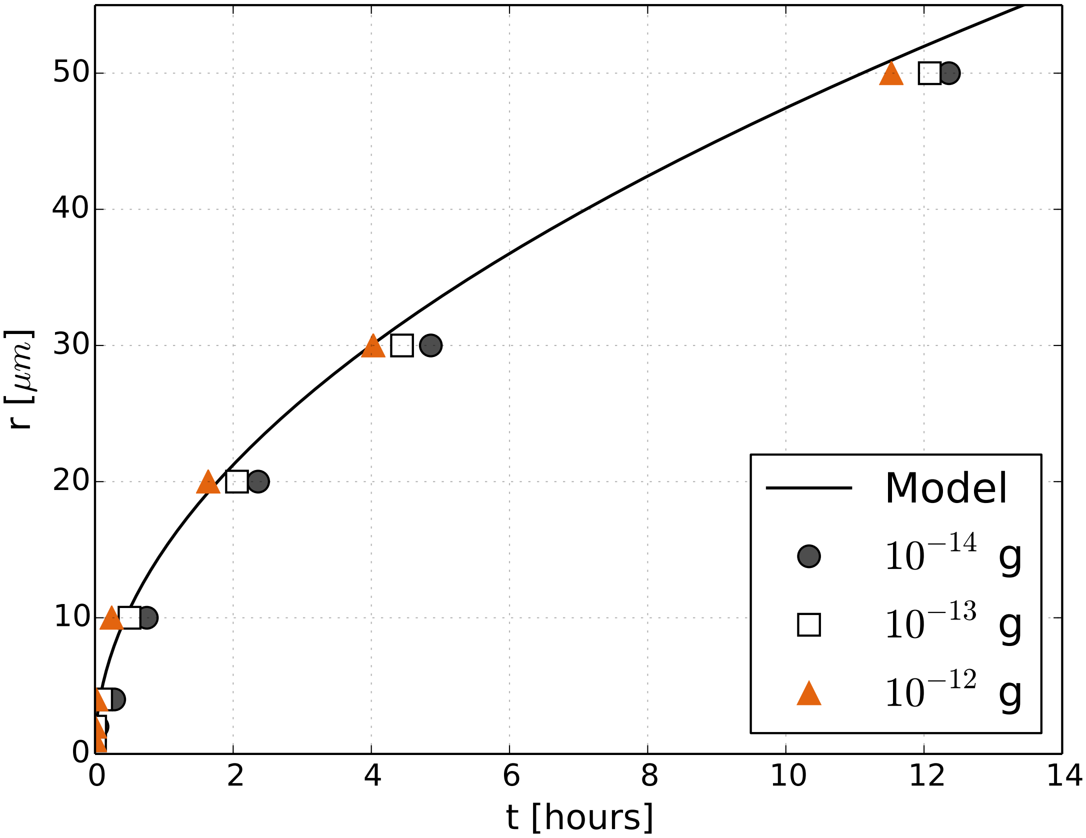

Figure 1: Droplet growth rate of the simplified model (Equation (4)) against the values from Table 5.5 of Curry and Webster [1999].

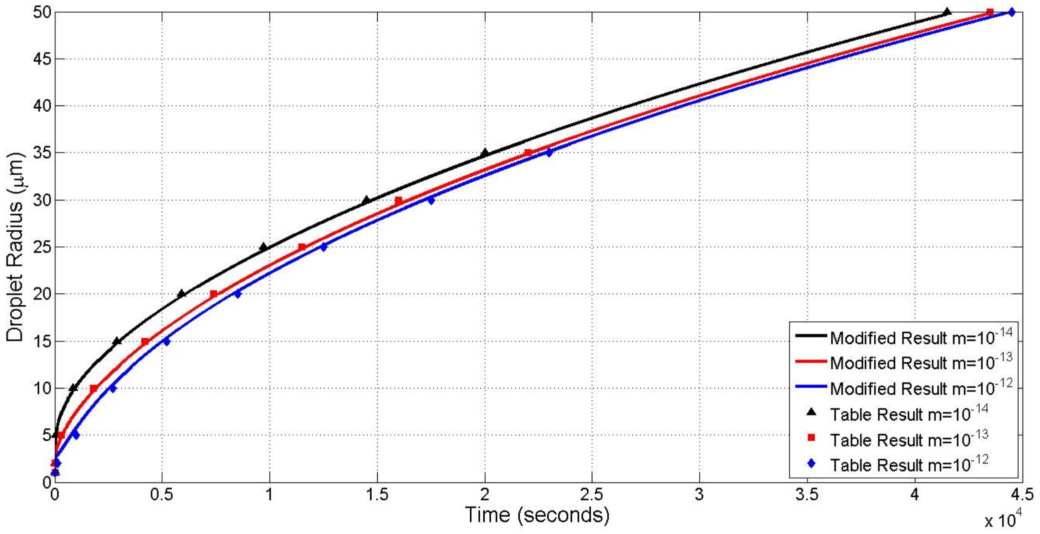

Figure 2: Droplet growth rate with a nuclei of NaCl, and curvature and solute effects are included. The values are calculated numerically using Equation (11) with $\Delta t = 0.1$, $T = 273K$, $p = 900mb$, and $S - 1 = 0.05\%$.

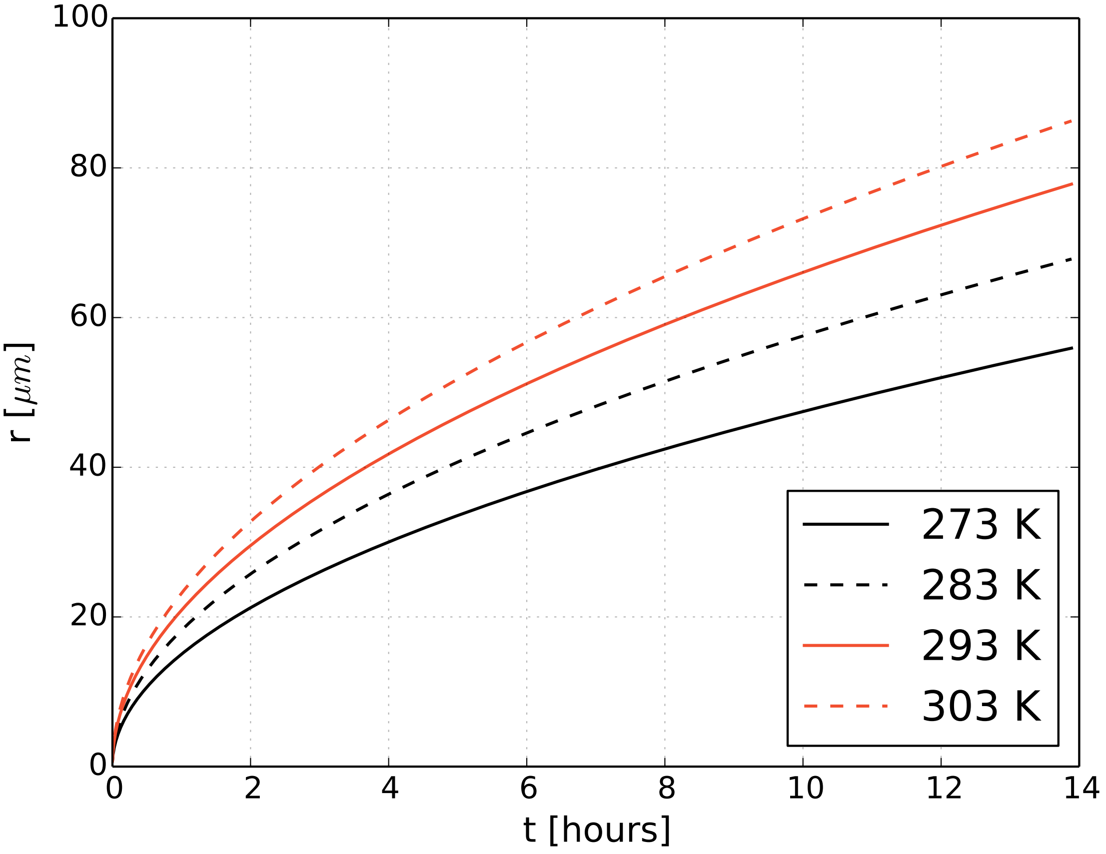

Figure 3: Droplet growth rate at $p = 900mb$ and $S - 1 = 0.05\%$ as T is increased.

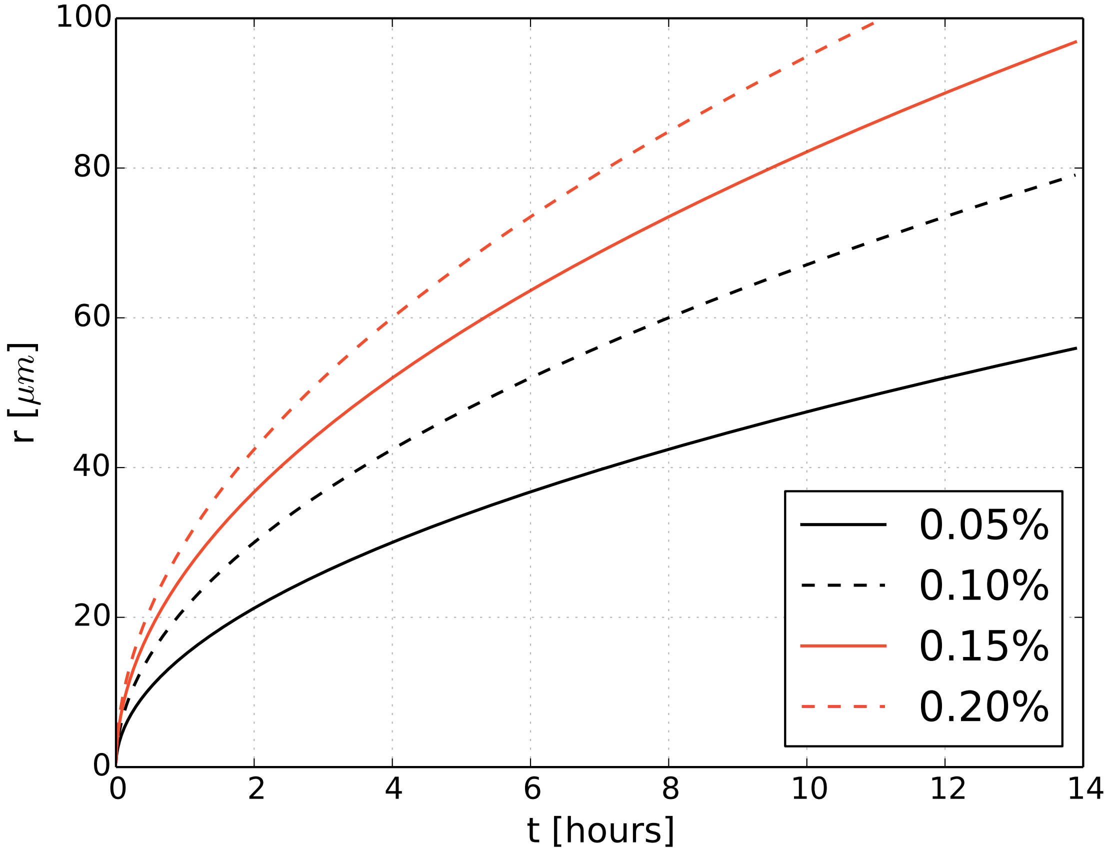

Figure 4: Droplet growth rate at $p = 900mb$ and $T = 273K$ as the $S - 1$ is increased.