TREES, AEROSOLS, AND CLOUDS

Colby J. Fronk[1], Gwenore F. Pokrifka[2], Jerry Y. Harrington[3], and Jose D. Fuentes[4]
1. Rochester Institute of Technology, Rochester, NY
2, 3, 4. Pennsylvania State University, University Park, PA

1. INTRODUCTION

For about the last 20 years, the aerosol composition of the eastern United States has been changing as a result of two major factors: 1.) anthropogenic aerosol emissions have steadily been decreasing [Doughty, 2014] and 2.) biogenic secondary organic aerosols (SOAs) emissions originating from plants have steadily increased as a result of an increase in vegetation brought about by reforestation [Barr et al., 2003].

The current impact of the changing aerosol composition defined by an increase in SOAs and a decrease in anthropogenic aerosols on the formation and growth of cloud droplets is currently unknown. In order to get a preliminary understanding, its impact on cloud microphysics will be studied by looking at Köhler theory and observing how these aerosols behave in an air parcel model.

2. MODIFIED KÖHLER THEORY

Köhler theory is the traditional method for calculating the equilibrium size of an aqueous solution droplet based on the surrounding vapor pressure, droplet curvature of a spherical droplet, and solute concentration within the droplet:

$$S_{eq} = \exp\left(\frac{4M_w\sigma}{RT\rho_w D_p} - \frac{6v_s m_s M_w}{\pi\rho_w M_s D_p^3}\right) \tag{1}$$

where D_p is the diameter of the droplet, σ is the surface tension, M_w is the molecular weight of water, R is the universal ideal gas constant, T is the absolute temperature, ρ_w is the density of water, v_s is the vant Hoff factor, m_s is the total mass of the solute, and M_s is the molecular weight of the solute [Lamb and Verlinde, 2011].

Equation (1) relies heavily on the assumption that the solute concentration is dilute and soluble, which works well for inorganic aerosols components but not so well with generally insoluble organic components. So that it can be applied to organic species such as secondary organic aerosols (SOAs) and the chemical species produced through their oxidation processes, traditional Köhler theory will be modified to include the effects of an insoluble core and dissolution kinetics of a slowly dissolving core with some level of solubility.

If the aerosol has an insoluble part, it can be treated as a solid core within the water droplet such that:

$$S_{eq} = \exp\left(\frac{4M_w\sigma_{sol}}{RT\rho_{sol}D_p}\right) * \frac{(D_p^3 - d_s^3)\gamma_w M_s\rho_w}{D_p^3 M_s\rho_w + d_s^3(\epsilon_s v_s M_w\rho_s - M_s\rho_s)} \tag{2}$$

where σ_{sol} is the surface tension of the droplet, d_s is the dry diameter of the aerosol, ρ_{sol} is the density of the solution droplet, γ_w is the water activity coefficient, and ϵ_s is the dissolved mass fraction of the aerosol [Raymond and Pandis, 2002].

If the aerosol is treated as a slowly dissolving core at the center of the droplet, then the saturation ratio is:

$$S_{eq} = \exp\left(\frac{4M_w\sigma}{RT\rho_w D_p} - \frac{6M_w v_s}{\rho_w}C_i\right) \tag{3}$$

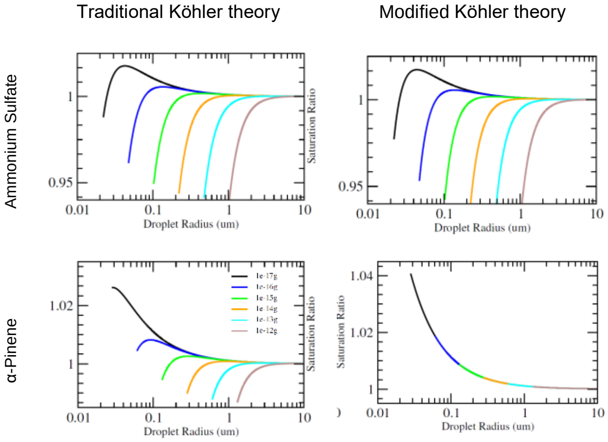
$$C_i = \max\left(0.5 * C_{sat}, \frac{C_{sat}}{1+\delta(1-\delta)\frac{w(S-S_{eq})}{4GDw}}\right) \tag{4}$$

where C_i is the concentration of the solute at the surface of the droplet, C_{sat} is the solute saturation concentration, D_c is the diameter of the non-dissolved core, D_s is the aerosol dry diameter, S is the ambient saturation ratio, G is the kinetic growth parameter, D is the diffusivity of solute in water, $\delta = 1 - \frac{D_c}{D_s}$, $w = \frac{D_p}{D_c}$ [Asa-Awuku and Nenes, 2007]. After complete dissolution, Equation (1) is used.

Equations (1), (2), and (3) can be combined to obtain the following expression for a slowly dissolving organic aerosol:

$$S_{eq} = \exp\left(\frac{4M_w\sigma_{sol}}{RT\rho_{sol}D_p}\right) * \frac{\gamma_w}{1+\frac{M_w}{\rho_w}C_i v_i}. \tag{5}$$

Köhler curves were created using Equations (1) and (4) for ammonium sulfate (anthropogenic aerosols), α-pinene (SOA), and pinic acid (oxidized form of SOA).



---

[1] Corresponding author address: Colby J. Fronk, Rochester Institute of Technology, Dept. of Chemical Engineering, Rochester, NY 14623; e-mail: cjf8874@rit.edu

[2] Corresponding author address: Gwenore F. Pokrifka, Pennsylvania State University, Dept. of Meteorology, University Park, PA 16802; e-mail: gfp5025@psu.edu

[3] Corresponding author address: Jerry Y. Harrington, Pennsylvania State University, Dept. of Meteorology, University Park, PA 16802; e-mail: jyh10@psu.edu

[4] Corresponding author address: Jose D. Fuentes, Pennsylvania State University, Dept. of Meteorology, University Park, PA 16802; e-mail: jdfuentes@psu.edu

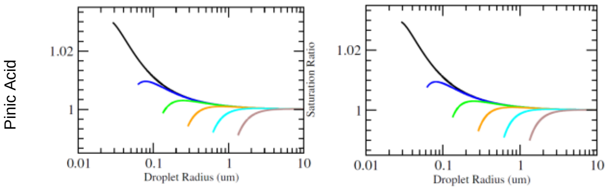


Fig. 1. Köhler curves for ammonium sulfate, α-pinene, and pinic acid calculated based on traditional Köhler theory and modified Köhler theory for varying solute amounts.

2. AIR PARCEL MODEL

An adiabatic air parcel model containing an initial distribution of dry aerosol particles was used to understand the impact that modified Köhler theory and CCN composition have under different cloud scenarios. The air parcel model described in Lebo et al. [2008] was used with some modifications made to the manner that the saturation ratio was calculated for the swelling routine and growth routine. The model is initialized by calculating the equilibrium swelled size of a lognormal distribution of hygroscopic aerosol. The distribution is divided into 200 Lagrangian bins, and the growth of each solution droplet is tracked in time.

Various updraft speeds were chosen ranging from 0.25 m/s to 10 m/s to account for cloud formation processes of cirrus to cumulonimbus cloud types. The parcel model was ran completely and the size distribution of particles at the time just after all cloud droplets have finished activating was plotted (see Figure 2).

The inorganic species grew the largest as indicated by the average diameter followed by the monoterpene derived acid compounds and the monoterpenes respectively. A maximum updraft speed of 5 m/s was needed to activate roughly 50% of the monoterpene particles whereas only 0.25 m/s was needed for the monoterpene derived acid compounds. Even at low updraft speeds, some of the larger organic based aerosols were able to activate and become cloud droplets. The data shows that organic aerosols derived from plants actively play a role in cloud formation processes.

3. FUTURE WORK

This set of simulations is for aerosols of one component, but actual aerosols are a mixture of several different components. The current simulations give a good idea of what to expect, but for a better prediction the parcel model needs to be modified to handle internally and externally aerosols. Additionally, a series of scenarios will need to be ran in the air parcel model for a few snapshots of the aerosol composition over a series of years.

4. ACKNOWLEDGEMENTS

The authors are grateful for the support from The National Science Foundation (NSF), grant No. AGS-1560339. This research was part of the NSF's REU program and was conducted at The Pennsylvania State University within the Department of Meteorology and Atmospheric science's REU program during Summer 2018.

5. REFERENCES

Asa-Awuku, A. and Nenes, A. (2007). Effect of solute dissolution kinetics on cloud droplet formation: Extended köhler theory. *Journal of Geophysical Research: Atmospheres*, 112(D22).

Doughty, D.C. (2014). Particulate matter and trace-gas changes at beltsville, md, and influences on cloud condensation nuclei.

Lebo, Z., Johnson, N., and Harrington, J. (2008). Radiative influences on ice crystal and droplet growth within mixed-phase stratus clouds. *Journal of Geophysical Research: Atmospheres*, 113(D9).

Raymond, T.M. and Pandis, S.N. (2002). Cloud activation of single component organic aerosol particles. *Journal of Geophysical Research: Atmospheres*, 107(D24), AAC-16.

Lamb, D. and Verlinde, J. (2011). *Physics and chemistry of clouds.* Cambridge University Press.

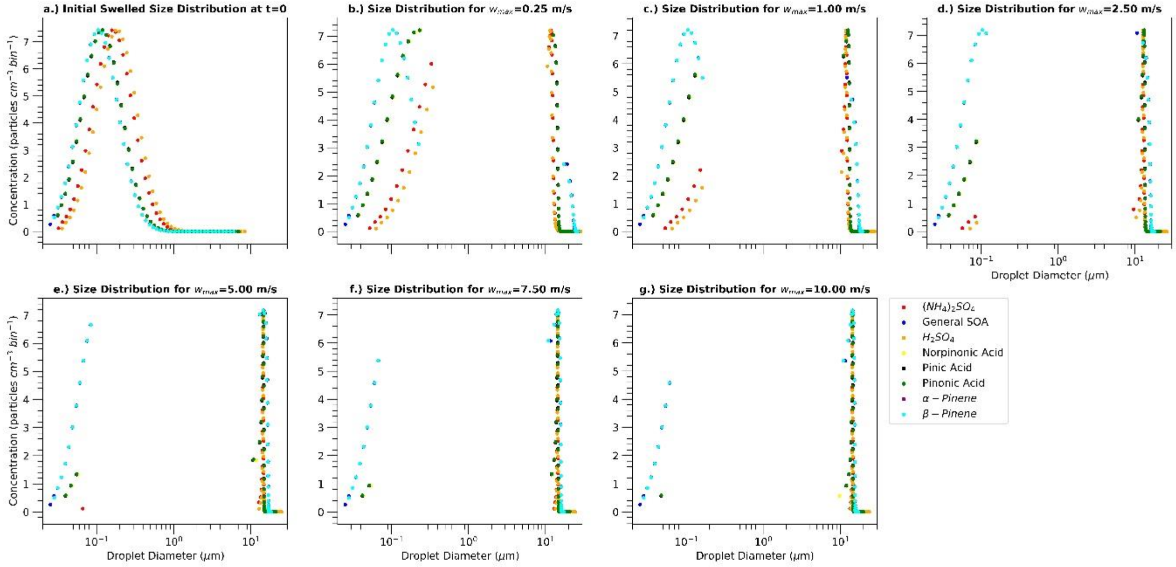


Fig. 2. The parcel model was ran for all of the chemical species with varying maximum updraft speeds, wmax. This 7 panel figure shows the size distribution profile of the particles in the air parcel for a.) the initial swelled sizes at t=0, b.) the sizes for wmax=0.25 m/s at t=1388 sec, c.) the sizes for wmax=1.00 m/s at t=360 sec, d.) the sizes for wmax=2.50 m/s at t=152 sec, e.) the sizes for wmax=5.00 m/s at t=83 sec, f.) ths sizes for wmax=7.50 m/s at t=57 sec, and g.) the sizes for wmax=10.00 m/s at t=44 sec. With the exception of the first panel, the distribution is shown at the time immediately after all of the particles are finished activating. In all simulations, the initial conditions were: RH_0=0.80, z_0=300 m, T_0 = 15 deg C, and P_0=919 hPa. The CCN size distribution was kept constant with the following lognormal distribution: geometric standard deviation=1.8 , NCCN=100 particles cm^{-3}, and median=0.05 um.