Atmos. Chem. Phys. Discuss., 10, C2692–C2697, 2010
www.atmos-chem-phys-discuss.net/10/C2692/2010/

Atmospheric Chemistry and Physics Discussions

Interactive comment on "Empirical predictions of CCN from aerosol optical properties at four remote sites" *by* A. Jefferson

A. Jefferson

anne.jefferson@noaa.gov

Received and published: 14 May 2010

Response to reviewer #1.

I thank the reviewer for his insightful and helpful comments.

Changes to the paper were made in accordance with suggestions on grammar, word use and references. Particularly the use of the word "prediction" was changed to "estimate" as a more accurate description of the analysis.

3. The ultimate goal of indirect forcing research is to predict cloud albedo from droplet concentrations and to understand the relationship between aerosols and cloud drops. Getting there from this work is a big step and I'm cautious to over state where I can go with this analysis. Ultimately I would like to explore if this method can be used

with remote sensing data. I added a few sentences to the introduction to explore the possibilities of where this method may lead in terms of further research. I will add more in the summary.

"The parameters derived from this empirical method can contribute to a regional climatology of CCN properties. While not a substitute for direct measurements of CCN, this method can be utilized as a proxy for CCN where measurements don't exist."

5. Dust from coastal North Africa can travel over the Azores (GRW) during the summer months. The usual path of dust from Africa is westward over the Atlantic to South America and the Caribbean via the West African mid-troposphere jet. Occasionally an anticyclone over the Azores will bring the dust further north. (Kaufman et al., JGR, 110, 2005) Dust is difficult to distinguish from sea salt with optical measurements as the two aerosol types are both large and have high single scatter albedos. The main difference is that dust will have a much lower hygroscopic growth. Lower hygroscopic growth factors were observed at GRW in August, however upon further examination of the data, the low fRH values were due to poor fits of the data from both low signal and from a low range of RH in the scans due to a high ambient dew point. The problem with detecting dust at GRW is not only these factors, but that dust would be highly diluted with sea salt, so that its effect on the aerosol properties would be small. The answer to the question about dust at GRW is that I don't have a clear way to distinguish it from sea salt, as its concentration may be too low or too rare of an occurrence to detect in the boundary layer. I can look at the lidar and satellite data to see if there were any clear dust events that made it north. I scanned the NAAPS model of surface dust for July and August of 2009 and didn't see any surface concentrations of dust over the Azores.

8. Dust doesn't work well with this method as the large size and high single scatter albedo place it in the same category as sea salt and other more soluble inorganics, and because of it's low solubility it doesn't activate as readily as sea salt of the same size, so it skews the data. There is also an instrument problem with dust that I encountered

at Niamey. The aerosol scattering and absorption measurements are size segregated via an impactor, the CCN accepts aerosol of all size. Most coarse mode particles are lost via deposition in the CCN sample line due to the low flow and narrow tubing, however some super micron particles make it through. The lowest size of the CCN is 1 micron, so the CCN can't distinguish between a 1 micron dust particle and a 1 micron droplet. I found that in the Sahel region during a dust storm the CCN at the lowest %SS was actually measuring the dust size distribution. I've enough data that I try to screen for dust as best as possible, but can't always do this effectively especially at marine sites or at places with fine dust particles as at HFE. The places where I run into trouble with dust are places like SGP, which has road and crop dust on only a few days. In this instance dust is an outlier in comparison to the other data. At HFE, dust was more common and wasn't an outlier, but a major component of the aerosol (I surmise). A better analysis would break down the data by season or aerosol type and single out days with high levels of dust or smoke during times with known crop burning, plowing or harvesting.

I added the following sentences to qualify the effect of dust. "In order to screen for large accumulation mode dust particles that may skew the fits, data with scattering Angstrom exponents below 1.0 were eliminated. Note that dust particles with Angstrom exponents greater than 1.0 or diameters less than about 0.5 μm were part of the analysis."

9-11. I rewrote the paragraph on page 9000 to discuss the role of particle size and composition on activation. I removed figure 2 from the paper, as it didn't add to the discussion on activation. Adding a graph of critical %SS as a function of dry diameter for different gRH isn't as straightforward as I'd hoped. Gasparini et al., 2006 has a nice study, which calculates the CCN spectra for various gRH at SGP. They empirically determine the particle surface tension and solute mole fraction from the size-dependent gRH values. I can't readily reproduce this information in a way that would help this paper. I will instead reference the Gasparini paper.

"Aerosol activation depends on both the Kelvin and solute terms of the Köhler equa-

tion. The critical super saturation for activation decreases with aerosol radius from an increase in surface tension and lower solute amount. Rose at al., 2010, found that the critical aerosol radius for activation at 0.87-0.07 %SS was about 20-100 nm, which covers a smaller aerosol size range than the aerosol optical measurements. Small particles tend to have a higher organic fraction that often dominates the composition of fine mode aerosol. Compared to an equivalent mass fraction of inorganic salt, the lower water activity of organics may result in a slightly higher particle critical super saturation (Koehler et al, 2006). The effect of a potentially high organic fraction on fine mode aerosol activation is mixed. Ervens et al., 2005 found reduced droplet number concentrations over a wide range of updraft velocities in their cloud parcel model, but only for organic species with a solubility less than 20 g L-1, much lower than most organic acids found in ambient aerosol. However this study did not account for nonequilibrium in clouds. Should the organic fraction form a soluble surfactant it could reduce the particle surface tension and lower the critical super saturation. Alternatively the organic fraction could form a hydrophobic layer that suppresses the uptake of water and raises the critical super saturation as was found by Kaku et al., 2006 in their study of marine aerosol. At SGP the gRH hygroscopic growth factors tend to decrease with particle size and indicate a high organic composition for particles less than 0.3 um diameter. These low growth factors in the subsaturated regime translate to critical super saturations in the range of 0.2 and 1.0 for sub 0.1 um diameter particles (Gasparini, 2006a,b), which is within %SS range of the CCN instrument. The comparison of CCN to aerosol optical properties assumes that the fine mode aerosol is essentially non-activating. The magnitude of this effect will increase at higher %SS and depend on the solubility of the organic compounds."

14. Figure 5 (now Figure 4) compares the CCN from about 0.2 to 0.8 %SS. I expect the correlation to be worse at higher %SS as particles below the size range of the optical measurements will activate to CCN. I inserted a figure to show the correlation for the low and high range of the %SS for the data from SGP. The graph is counter intuitive as the lower range %SS (<0.4%) has a smaller slope and under predicts the CCN more

than the higher range with %SS >0.8. I show SGP as this site has a high concentration of small particles (high CN number concentrations) that doesn't correlate well with the scattering. This type of plot suggests that more complex issues are at work and that more analysis is needed. Perhaps including the plot below doesn't help the paper as the uncertainty in the data may be too high or there are limitations of the method that need more detailed analysis to understand it.

15. My intention with this paper is to show the simple correlations, suggest a greater possibility for the work and then use the work to inspire more analysis in others or perhaps funding and collaborations to pursue it more. The greater implications of this work is not to replace CCN measurements, but to supply enough information about aerosol properties that models can approximate droplet concentration for different aerosol types in regions where there are limited measurements. I'd like to extend this work to compare it to the size distribution, gRH and composition data at SGP, segregate the data by aerosol type and season and even compare laboratory aerosol of know size and refractive index to understand the limitations of the method and also the possibilities. This analysis, as I present it here, may be too simplistic to tease out more information, hence the need for a more detailed analysis. The ultimate goal is to use this type of analysis in remote sensing, but it may take a few steps to get there. I'm hesitant to suggest a detailed plan as to how this could be applied to remote sensing, I think careful planning is needed on how to use remote sensing (e.g. lidar, cimel, mfrsr) and surface in-situ measurements in such a way to validate the method. We could use aircraft data of CCN at cloud base on days that the boundary layer was well mixed. The Racoro data set over SGP is appropriate for such a study as there were CCN and cloud droplet concentration measurements in and near cloud base, which could be combined with surface CCN and aerosol optical measurements for a validation of this method.

"This study is a first step in a process to better estimate CCN concentration as a function of %SS from the aerosol optical properties. Further analysis includes well-defined fit parameters for each aerosol type and region. This will entail analysis

of the long-term data from SGP for seasonal trends of the fits and comparison to size-dependent aerosol composition to better quantify the fit parameters with aerosol type. Validation work is needed to test the method with vertically resolved in-situ aircraft measurements near clouds, surface measurements and remote sensing to see if the method can be extended further to include lidar or Cimel data."

Please also note the supplement to this comment:
http://www.atmos-chem-phys-discuss.net/10/C2692/2010/acpd-10-C2692-2010-supplement.pdf

---

Interactive comment on Atmos. Chem. Phys. Discuss., 10, 8995, 2010.