Atmos. Chem. Phys. Discuss., 7, S6109–S6113, 2007
www.atmos-chem-phys-discuss.net/7/S6109/2007/

Atmospheric Chemistry and Physics Discussions

Interactive comment on “Clouds and aerosols in Puerto Rico – a new evaluation” *by* J. D. Allan et al.

Anonymous Referee #1

Received and published: 18 October 2007

The manuscript presents a very rich dataset containing aerosol microphysical, cloud microphysical, and chemical properties measured at two sites in Puerto Rico. Four analyses are presented. First, a comparison between measured hygroscopic growth and cloud condensation nucleus activity, second a qualitative comparison between observed composition and measured hygroscopicity , third a comparison between CCN concentrations and observed cloud microphysical properties, and finally, a historical and airmass comparison of aerosol chemical composition. The manuscript is well written and the subject is appropriate for the audience of ACP. I recommend that this paper will be published subject to the following revisions.

While the authors present a lot of data the analysis remains qualitative only. In several places a more quantitative approach could be implemented with little effort while simultaneously strengthening the findings. Because of the mostly qualitative analysis the conclusions reported in the manuscript are overstated and need far more qualification than is currently presented in the manuscript.

In particular the consistency between hygroscopic growth and CCN activity (Figures 3 and 5) can now easily be quantified using one of a wide range of fairly similar methods available in the literature (e.g. Svenningsson et al., Tellus, 1992, Brechtel and Kreidenweis, JAS, 2000 Kreidenweis et al., ACP, 2005, Svenningsson et al., ACP, 2006, Petters and Kreidenweis, ACP, 2007, Wex et al., GRL, 2007, Vestin et al., JGR, 2007). These methods either extrapolate HTDMA growth factor data to predict the relationship between critical supersaturation and particle dry diameter shown in Figure 5. Alternatively effective hygroscopicity can be inferred from both the size resolved CCN data and the HTDMA data and expressed using a single parameter, which can then be compared in a scatter plot.

If the data are expressed as hygroscopicity or ion density it will also be easier to guide the discussion which compares chemical composition and hygroscopic properties. The plot could contain values for ammonium sulfate, letovicite, ammonium bisulfate and sulfuric acid so the reader can gauge to what extent the observations deviate from the ammonium sulfate results.

A more difficult closure is the link between CCN activity and cloud droplet number concentration. In the manuscript it is argued that "during this study the changes in cloud microphysical behavior noted here are entirely attributable to the influence of anthropogenic emissions". However, relevant parameter like updraft velocity, in-cloud processes, meteorological boundary conditions, aerosol size distribution, and aerosol chemistry are not considered either qualitatively or quantitatively in the framework of a parcel model. Little can be concluded about the relationship between aerosol and clouds by only contrasting CCN concentrations and droplet concentrations during different periods without constraining at least some of these influences.

Specific comments:

Pg. 12577 If CCN and nss-sulfate are correlated and CCN and droplet concentration are also correlated, then it seems a bit contradictory that droplet concentrations are insensitive to sulfate loadings.

"Following from these seminal and farsighted works" seems a bit overstated.

Pg. 12583 Although the CN concentrations are indeed similar between the CSJ and EP site there are notable spikes in concentration that were only seen at CSJ, possibly caused by a highly localized source. To my mind it would seem prudent to exclude these points from the Lagrangian analysis that is focusing on advected airmasses from the ocean.

Pg. 12584: Please define "growth factor spectra" and symbol "P". I interpret Figure 4 as normalized frequency distribution, but I am not entirely sure. A little more detailed explanation in the text is necessary.

Figure 5: What instrument are these from, the static CCN or the DMT CCNc?

Pg. 12558: The authors state that "The measured sulfate mass concentrations in the cloud residuals were higher by a factor of about 20. This indicates that almost all sulfate containing particles were activated as CCN". Although this interpretation is possible it is not plausible since droplet residuals do not necessarily relate to the droplet nucleation process. A significant source of sulfates from oxidation of SO2 by O3 and H2O2 via aqueous-phase reactions in cloud droplets likely contributes to this observation.

Pg. 12591: The CCN/CN ratios are not really shown and it is difficult to see the magnitude of this effect in Figure 3 (top panel). Perhaps these ratios should be added to this figure? The discussion that follows seems also to relate to Table 4, which is only introduced several pages later. If the inferences made about pollution changing microphysical properties are based on that data presented in Table 4 several clarifications/qualification must be added to the table and the text:

- It should be made clear that the inferences relate to the data from columns 6 and 8 in Table 4, in particular rows 3, 4, 5, and 6.

ACPD

7, S6109–S6113, 2007

Interactive Comment

- The supersaturation for the CCN/CN ratios needs to be included in the Table
- There should be some justification that the number of cloud droplets that was averaged through the periods can really be compared. For example, were the meteorological conditions similar? Were the cloud types similar (e.g. oreographic cloud vs. cumulus cloud, was the stability similar etc.)? Answers to these questions may help to constrain to what extent variations in updraft velocity can be excluded as the cause for this difference.
- Also where was cloud base and cloud top? Could entrainment and/or collision/coalescence have reduced cloud droplet number concentrations?

The authors proceed to conclude (pg. 12594) that "during this study the changes in cloud microphysical behavior noted here are entirely attributable to the influence of anthropogenic emissions"

I do not believe that this conclusion can be drawn from the limited analysis presented in this study.

The frequency distributions shown Figures 6 and 10 are very difficult to digest. It is unclear for which interval the frequencies were calculated. For example the frequency for the EBC (Figure 6d) for the three periods covers (probably due the juxtaposition) 40-70 ng m-3. Are all three periods evaluated in this interval? Or was period 1 evaluated from 40-50, period 2 from 50-60, and period 3 from 60-70? To clarify, I recommend separating the data into 3 histograms which should be normalized such that the integral evaluates to 1. This will allow unambiguous and quantitative comparisons between the periods. The three histograms can either be overlaid on one plot or spread over three panels for each of scalars.

Pg. 12591: It is argued that the low hygroscopicity particles during period 3 do not contribute to CCN concentrations (it is not clear at what supersaturation) since the

CCN/CN ratios were lower. Part of this may be due to the smaller size of those particles as mentioned by the authors. Figure 9 also suggests that some of the larger particles are not sufficiently hygroscopic to activate into cloud droplets since large particles are found in the interstitial aerosol. However, it is also argued that despite the low hygroscopicity the pollution from upwind has a measurable effect on the warm clouds. Perhaps the CCN/CN ratios were evaluated at the 'wrong' supersaturation, i.e. one that is not applicable to the conditions in the cloud? To resolve this apparent contradiction the authors need to show the following:

- What is the expected activation diameter (as a function of supersaturation) for the low hygroscopicity particles? Are these consistent with the residual signature for 200 nm particles shown in Figure 9? This can easily be answered if the quantitative comparison suggested earlier is implemented.
- Estimate the cloud supersaturation.

Without at least such semi-quantitative estimates the conclusion about the effect of the weakly hygroscopic particles on the warm cloud remains highly speculative.

Interactive comment on Atmos. Chem. Phys. Discuss., 7, 12573, 2007.