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Atmospheric Chemistry and Physics Discussions



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

**J. D. Allan et al.**

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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.

Response: Agreed that this type of closure would be greatly desirable and an analysis using the model of Petters and Kreidenweis has been attempted. Unfortunately, this task was found to be more complex than anticipated, due to problems introduced by technical issues such as the transfer functions of the respective DMAs used. While not impossible, we concluded that this type of closure is not as straightforward as was hoped and would entail a lengthy technical discussion, so on balance, would be best left for a separate paper. A brief discussion of these types of closures is included in the revised conclusions section.

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.

Response: The predicted growth factors for ammonium sulphate, ammonium bisulphate, sulphuric acid and sodium chloride have been added to the hygroscopicity plot (figure 4).

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.

Response: In the original manuscript we have addressed these concerns by looking



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at the sounding from all the days and comparing the average cloud base pressure, as defined by the lifted condensation level (LCL) and the precipitable water (PW) for the three periods. In order to further address the reviewer's concern that we have not adequately argued that the cloud properties are a result in changes in aerosol characteristics, rather than meteorology, we have added additional evidence to support our conclusion. The new text is as follows:

"The 0:00 UTC and 12:00 UTC (19:00 and 7:00 local time) soundings from the San Juan International airport , launched by the US National Weather Service, were evaluated to compare the meteorological state of the atmosphere during the three periods. The droplet concentration and size distribution, in addition to being sensitive to the characteristics of the CCN at cloud base, will depend on the vertical profile of temperature and water vapor and the vertical velocity. The pressure of the lifted condensation level (LCL) is used as an indicator of expected cloud base and the precipitable water (PW) as a measure of potential rain amounts. The average LCLs for the three periods were 956 ś 8 mb, 962 ś 11 mb, and 963 ś 4 mb, respectively, indicating little difference in expected cloud bases for the three identified periods. The average PW for these same periods was 40 ś 6 mm, 44 ś 11 mm, and 50 ś 5 mm. There is a positive trend in PW during the nine days of the project; however, this is not reflected in either the precipitation rates or in situ liquid water contents that were measured (Table 2 and Fig. 10). The mountain site was approximately 300 m above cloud base as estimated from the pressure measured at the site and the photos that were recorded from the lighthouse every five minutes with a web camera. These photos showed that the daytime cloud bases and tops were not visibly different for the three periods, consistent with the sounding information that indicated fairly constant pressures and temperature at cloud base during the observational period.

The vertical velocity for the three time periods was not directly measured. In order to assess the effect of changes in vertical velocity at cloud base versus changes in the CCN concentrations, we ran a closed parcel model with constant updraft to estimate



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droplet activation, within 300m of cloud base. The model incorporates the parameterization introduced by Twomey and Wojciechowski (1969), of the CCN concentration (C) as a function of supersaturation, S, of the form C=C0Sk. The values of C0 and k were estimated from the CCN measurements made at the lighthouse site. Table 3 shows the results of the model for constant updraft velocities of 0.2 and 1 ms-1 and for the C0 and k values derived for the clean and polluted periods. It can be seen in the Table that the droplet concentration, ND, predicted for an altitude of 300 m above cloud base increases by less than 20% when the vertical velocity is increased by a factor of five. The observed increase in the CCN concentration (represented in this case only by the change in C0) increases the droplet concentration by more than a factor of two when C0 is increased by a factor of three. This increase in concentration is similar to what was observed in the measured droplet concentrations. These results suggest that the observed differences in the LWC, droplet concentration and median volume diameter are the result of changes in the aerosol between the periods, rather that due to changes in the type of convection or other mesoscale forcing."

Additionally, a new table 3 is added with the model outputs described in the text as follows:

Table 3. Results of a closed parcel model with constant updraft velocity (W), based on the observed CCN characteristics at the lighthouse, for the three periods identified in the study. Note that periods 1 and 3 had similar characteristics, so are grouped for these purposes.

Period, C0, k, ND (W = 0.2 ms-1), ND (W = 0.2 ms-1)

1 & 3, 700, 0.8, 373, 397

2, 250, 0.8, 142, 172

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



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concentrations are insensitive to sulfate loadings.

Response: The text has been clarified as follows: "While sulphate constituted a large fraction of CCN mass, the researchers concluded that this had been added to the particles through processing after activation and therefore did not affect the initial CCN number concentrations, which were more dependent on the sea spray fraction."

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

Response: The words 'seminal and farsighted' have been removed from the revised manuscript.

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.

Response: These points were indeed excluded from the analysis. This has been stated more clearly in the revised version.

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.

Response: These are indeed normalised frequency distributions. This is explicitly stated in the revised version.

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

Response: These are taken from the DMT CCN. This is stated in the revised version.

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



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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.

Response: True, but we also know that while sulphate is found at the upwind site, it is not found in the interstitial aerosol. The text has been revised as follows: "The measured sulphate mass concentrations in the cloud residuals were higher by a factor of about 20. While it is possible that some of the mass could have been added in the cloud through aqueous processes, the fact that very little remained in the interstitial aerosol indicates that almost all sulphate-containing particles were activated as CCN."

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?

Response: These have been added to the revised version.

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.

Response: The discussion actually refers the data in table 2 (some of this is later repeated in table 4). This is stated in the revised text.

The supersaturation for the CCN/CN ratios needs to be included in the Table

Response: This has been added to the revised version.

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



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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?

Response: These are addressed in the response to the general point about cloud dynamics above. We feel that we have done as good a job as possible in constraining these effects with the data available.

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.

Response: Agreed that the logic of the current wording is in error. The statement has been reworded as follows: "There is no evidence to suggest that the changes in cloud microphysical behaviour noted during this study are attributable to anything other than the influence of anthropogenic emissions...";

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.

Response: The frequency plots have been made more distinct to indicate that each grouping by case falls in the same interval of values. This is also clarified in the text as follows: "The frequency distributions shown in Fig. 6 highlight the differences in some of the aerosol properties related to air mass origins. The frequency of occurrence for each of the three cases (red, blue and black) is given as a percentage of all events during the respective time periods. The same number of intervals is used for the three



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cases and for each aerosol parameter." and also: "The frequency diagrams shown in Fig. 10 illustrate the changes in bulk cloud properties as a function of air mass origin. The frequency of events is presented in the same manner as discussed for Fig. 6"

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.

Response: This is not exactly what we are trying to argue; the low hygroscopicity particles do contribute to the CCN concentrations, but their contribution is not proportional to the addition they make to the CN concentrations. This is stated more clearly in the text as follows: "The addition of a primary aerosol is also manifested in an increase in CCN concentrations, but this increase is not proportional to the (much larger) increase in CN, resulting in lower CCN to CN ratios for particles of a given size. The overall ratio of CCN to CN is likewise suppressed, although this may have more to do with the larger number of small particles under these conditions..." and also, "The measured changes in the cloud microphysics during the polluted cases would seem to suggest that even at the modest mass concentrations observed and in spite of the low hygroscopicity of the particles, the pollution from the upwind islands contributes to the CCN concentrations and has a measurable effect on the properties of warm clouds..."

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



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quantitative comparison suggested earlier is implemented.

Response: Agreed that this would be desirable, however as stated before, this would not be a trivial exercise, especially when trying to quantitatively separate the low-hygroscopicity particles from the high-hygroscopicity particles in the HTDMA and CCN spectra. This work will be presented in a future publication.

Estimate the cloud supersaturation.

Response: Unfortunately, we do not think we have sufficient data to constrain this with any confidence, either based on meteorological measurements or by comparing the upwind aerosol properties with the cloud microphysics.

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

Response: While the conclusion is mainly qualitative, we do not feel that the CCN data and the cloud microphysical data are at odds with each other. Both show an increase in CCN and therefore cloud droplet number concentrations that is not proportional with the corresponding increase in CN observed. This is consistent with the idea that a subset of the pollution particles are able to act as viable CCN. While this subset as a fraction is small, it is still able to measurably affect the cloud microphysics. We also show that both composition and size are important in dictating whether these particles will activate or not.

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