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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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Although the links between aerosol and clouds presented in this paper, e.g. the measured particulate sulphate concentrations going along with elevated CCN activity in the polluted case, all make sense, however, a summary of the instrument uncertainties would be highly informative, might support findings by excluding instrumental issues in some cases, and would present a first step towards a more quantitative closure study anyway. At least for the measurements of CCN activity, hygroscopicity and chemical composition these values should be given or discussed in more detail (see also special remark with respect to the organic fraction measured by the AMS).

Response: Instrumental uncertainties, where possible, have been added to the revised manuscript in table 1.

The possibility of the disagreement of results due to technical issues seems to be a big problem for the analysis of the data set and could be worked on more, maybe even in

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a separate section of this manuscript.

Response: This uncertainties associated with the AMS and offline techniques are discussed in the discussion section. In the revised manuscript, this has been given its own subsection and expanded.

Specific remarks: 1) One of the main conclusions of the manuscript is that organic fraction observed at the measurement location is likely not to be ubiquitous ("not totally ubiquitous", 12593, line 14 is a vague word). This is based on the disagreement of results of two different campaigns using different instrumentation and qualitatively discussing the possibility of "technical issues" being responsible for that. For my understanding only the standard deviation of the measurements is given for both campaigns (e.g. 12576, line 4-5) but no actual values of the accuracy/uncertainty of the measurement/instruments are reported. This value also has to be taken into account when discussing this issue. Is there a possibility to quantify accuracy of filter and AMS measurements?

Response: This has been addressed with the addition of the instrument uncertainties and expanding of the discussion as described above. Novakov et al (1997) estimated the uncertainty associated with EGA to be around 10%. This is now stated in the expanded uncertainties section.

2) A follow up point on this would be a more detailed description of the AMS settings used during the Puerto Rico measurements. The quantitative data of the AMS represents a substantial fraction of the results presented in the manuscript and provides the basis of one main conclusion, i.e. the necessity to investigate the organic fraction of the "local" aerosol in more detail. Based on Allan et al. [2004] the quantitative results of the AMS are strongly influenced by the collection efficiency of the sampled particles, the ionization efficiency of the individual components and the application of an empirical correction factor normally used for the quantification of sulfate. The author states that "in the absence of a reliable external constraint, a collection efficiency of 0.5 was

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assumed" (12580, line 8-9). It would be helpful to discuss the uncertainty of the factors mentioned above with regard to the quantification of the mass concentrations - and the organic fraction in particular - in more detail. This might lead to a better valuation of the conclusions given in the paper.

Response: The following discussion has been added to the section on instrument issues, which addresses the points raised by the reviewer:

"The AMS analysis relies on assumed values of the relative ionisation efficiencies and collection efficiency. The former values are based on laboratory characterisation work and with the exception of ammonium, have consistently been found to be robust between instruments and deployments (Allan et al., 2004a;Takegawa et al., 2005). It is possible that the actual collection efficiency could be greater than the 0.5 assumed, but this is only typically observed for pure ammonium nitrate or sulphuric acid particles or those with a very high water content (Crosier et al., 2007;Allan et al., 2004a). While considered unlikely, if this were the case, this would mean the actual organic mass concentrations would be lower than reported by up to a factor of 2."

3) In the abstract and in the introduction the term "some of the latest developments in online instrumentation" (12575, line 5) and "recent developments in the field of aerosol instrumentation" (12578, line 24) are used. It would be helpful to address the actual instruments linked to these expressions.

Response: The AMS, CCNs, and HTDMA are mentioned explicitly in the revised version.

Technical corrections: 1) Page 12606, Fig. 1: An increase of size and sector of the left bottom panel might improve the quality of this figure.

Response: Altered as suggested.

With respect to a possible follow-up campaign: It might be helpful to employ the FLEXPART-Model (Stohl et al., 1998) for the classification of air-masses, too. The

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combination of the dispersion model and cluster analysis and especially the FLEXPART product "footprint residence time" that accounts for the parcel's origin and it's source strength averaged over the lowest 150m above the surface would give a good indication of anthropogenic emissions when having these 'patchy'; distributions of islands upwind of the measurement locations.

Response: Agreed that this would be desirable and this is now mentioned in the revised manuscript.

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