Atmos. Chem. Phys. Discuss., 7, S5875–S5878, 2007
www.atmos-chem-phys-discuss.net/7/S5875/2007/

Atmospheric Chemistry and Physics Discussions

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

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Received and published: 10 October 2007

The manuscript presents simultaneous in situ measurements of aerosol and cloud properties at two different sites in Puerto Rico. Results of the two sites are compared in a "quasi Lagrangian" framework with respect to the interaction of aerosols with clouds. Special emphasize is placed on measurements of the aerosol (total, interstitial) and cloud droplet chemical composition by means of an aerosol mass spectrometer, aerosol hygroscopic growth factors, CCN activity and particle number size distributions. Equivalent black carbon values were measured using an Aethalometer and an absorption photometer to account for anthropogenically influenced air masses. The data was classified into three periods of different air mass origin - two "polluted" and a "clean" period. Both, aerosol composition and cloud properties showed clear differences between the polluted and the clean case; the polluted one was moderately influenced by populated islands upwind and generally smaller, externally mixed, carbonaceous particles were measured. Along with this a decrease in hygroscopicity and

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fractional cloud activation potential of the measured aerosol was observed compared to the clean case. A comparison to previous studies in Puerto Rico also revealed a difference in the observed anthropogenic organic fraction of the aerosol measured - the organic matter measured during PRACS (this campaign) was about a fifth of the mass measured previously. The authors stress the difference in the used measurement and analysis methods and mention the possibility that the main source of disagreement between the past and the current study "may be due to technical issues associated with differences in methodologies". However, the authors also conclude that it is likely that the organic fraction observed at this location is not ubiquitous as a "discrepancy between the analytical techniques themselves is not deemed likely".

Overall remarks:

The topic is certainly appropriate for publication in ACP. The combination of measurement techniques and suitable locations with respect to the observation of aerosol-cloud interactions lead to a comprehensive dataset necessary to make use of. The data is well presented and the links between aerosols and clouds sufficiently discussed within the scope of this paper and using the underlying dataset. However, a follow-up publication dealing with a more quantitative closure between the aerosol composition, hygroscopicity and size-resolved CCN measurements is highly desirable as already mentioned by the authors.

The manuscript compares quantitative results of a large number of instruments but mainly focuses on the chemical composition, CN number concentration, CCN activity, and hygroscopicity. 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

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more detail (see also special remark with respect to the organic fraction measured by the AMS). 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 a separate section of this manuscript.

I just found instrument uncertainties for the FSSP and the PVM instruments.

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?

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 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

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the conclusions given in the paper.

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.

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.

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 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.

Allan et al., Submicron aerosol composition at Trinidad Head, California, during ITCT 2K2: It's relationship with gas phase volatile organic carbon and assessment of instrument performance, JGR, 109, 2004

Stohl et al., A replacement of simple back trajectory calculations in the interpretation of atmospheric trace substance measurements, Atmos. Environ., 36, 2002

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