

## ***Interactive comment on “Organics in the Northeastern Pacific and their impacts on aerosol hygroscopicity in the subsaturated and supersaturated regimes” by K. K. Crahan et al.***

### **Anonymous Referee #1**

Received and published: 16 July 2006

Review of the paper “Organics in the Northeastern Pacific and their impacts on aerosol hygroscopicity in the subsaturated and supersaturated regimes” by K. K. Crahan, D. A. Hegg, D. S. Covert, J. L. Santarpia, H. Jonsson, G. Buzorius, D. Collins

The authors present some nice aerosol related data from flights off the Californian coast. The data seems to be of high quality and span over physical properties related to particle size and water up-take as well as chemical analysis of some inorganics and organics. The paper fits well within the focus of Atmospheric Chemistry and Physics and is well written. It also deals with a subject - organics in aerosols and water uptake of particles - that needs more elucidation. I therefore recommend publishing after some

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changes and the authors' seriously considering some comments.

Major efforts are made in the paper to find closure between chemical and physical properties. These aims are, however, not stated. I would suggest that the authors help the readers by clearly stating the aims of the paper.

My main comments are regarding two of the conclusions and I strongly encourage the authors to be very careful in how these conclusions are stated. I would especially recommend the authors to look carefully on these conclusions in the abstract.:

-1: The authors state on page 4225 that “..treating water activity as ideal is not a good assumption at subsaturations ..” and “Organics likely play a large role in the poor agreement between the models and observations,..”. This is, as I understand it, based on the fact that the observed hygroscopic growth is smaller than expected from the chemical composition, assuming ideal behavior. A similar discrepancy was observed by Svenningsson et al., for the mixture referred to in table 4 in the Crahan et al. paper. In fact, it was observed already in 1987 by Cohen et al. for ammonium sulfate-sodium chloride mixtures. The reason for the discrepancy is unclear and non-ideality is only one possible explanation.

-2: The existence of surface layers is an explanation often put forward when no other explanations can be found. One has to be very careful when doing so - strictly speaking ALL other possibilities should be explored, which is an impossible mission! In this case I think that there could be explanations found for example in variability in the chemical composition over particle size- what about if the smallest particles consist of almost only organics as could be the case if they are produced from film droplets?

I do very much like the calculations of surface area coverage of insoluble organics (figure 11) that show that there is enough material to cover the surface at 85% RH. But the surface area increases rapidly at higher RH and the material will then not be sufficient. The hindering of water vapor condensation must thus take place at RH below 85-90% where the response time to RH changes for 100 nm particles is very short and

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thus the relative increase in growth rate must be very high. What is the authors' view in this?

The hygroscopic growth observed for the marine aerosol in this study is compared with laboratory results on an artificially produced and strongly simplified imitation of a continentally influenced marine aerosol (Svenningsson et al., 2006). In my opinion, these results can not be used as a model for all marine aerosols, but rather as a step bringing laboratory generated, relatively well known aerosols closer to real atmospheric aerosols. There is however much more to learn about marine aerosols, and studies like the one presented here by Crahan et al. will be very useful.

As I understand it, the laboratory results are used as they are, without correcting for variations in the relative amounts of organics and inorganics from the chemical analysis of samples taken during flights. Is that correct?

The experimental section does not describe the hygroscopic growth and critical supersaturation measurements. In the case of hygroscopic growth, a description is found in section 3.2, but I could not find any description of the CCN spectrometer used. Mentioning in the Experimental section that the hygroscopic growth measurements are described together with the results would help the reader. A short description of the CCN measurements is also suggested.

On page 4219, the correlation matrix in table 3 is discussed. I do very much appreciate this analysis and find it very interesting. However, the authors state that there is a correlation (0.624) between SPOM and “biomass burning” but no strong linkage between dicarboxylic acids and any other chemical components. But what about dicarboxylic-sea salt (0.575) and dicarboxylic acids-ammonium nitrate (0.501)?

At the bottom of page 4219, the authors say that the IPOM found is not of anthropogenic nature since the air has spent more than the atmospheric life time of these compounds over sea. But, are there no anthropogenic sources over sea?

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I do find the negative correlation between hygroscopicity and IPOM very interesting. This will be an interesting task for the scientific community!

Thanks for really nice reading!

Cohen, M. D., Flagan, R. C., and Seinfeld, J. H.: Studies of concentrated electrolyte solutions using the electrodynamic balance. 2. Water activities for mixed-electrolyte solutions, *J. Phys. Chem.*, 91, 4575-4582, 1987.

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Interactive comment on *Atmos. Chem. Phys. Discuss.*, 6, 4213, 2006.

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