

Interactive comment on “Aerosol hygroscopicity in the marine atmosphere: a closure study using high-resolution, size-resolved AMS and multiple-RH DASH-SP data” by S. P. Hersey et al.

S. P. Hersey et al.

Received and published: 11 November 2008

General author comments:

Based on the reviews from both anonymous referees, it is clear that the ACPD version of this paper contained a number of phrases that led to methodological misunderstandings. These phrases pertained particularly to the AMS instrument used and the way in which AMS composition data were interpreted.

First, while the phrase ‘high resolution’ was intended to imply ‘high time-resolution’ this was not explicitly stated, and led to confusion about which AMS instrument was used in this study. All references to ‘high resolution’ have either been removed or modified to explicitly state ‘high time-resolution.’ Additionally, text has been added to be clear that

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

Discussion Paper



the AMS used was a Compact Time of Flight Aerodyne Aerosol Mass Spectrometer (C-ToF-AMS).

In analyzing AMS-specific syntax used in the paper, it also became clear that the ACPD version of this paper included imprecise references to oxygenated and hydrocarbon-like organics as OOA and HOA, respectively. All references to OOA and HOA have been removed to avoid any confusion, and terminology is limited to 'oxidized,' with evidence of 'oxidized' organics coming solely from O:C ratios investigated in the paper.

Responses to specific reviewer comments:

Anonymous Reviewer 1:

This paper assumes that inorganic salts are in a solid state and do not uptake water. However, many laboratory experiments have found the existence of supersaturated droplets in many inorganic systems. Santarpia et al. (J. Geophys. Res., 110, D03206, doi:10.1029/2004JD005279, 2005) have observed supersaturated aerosols in coastal environments. It would be interesting to know how the closure analysis would be affected by relaxing the assumption of dry solid at RH below RHD of the salts.

This assumption warrants further discussion in the paper, and the following text has been added to the discussion section:

While particles entering the DASH-SP are dried to well below relative humidity of efflorescence (RHE) for the inorganic salts encountered in the marine atmosphere, it is possible that some particles remain in a supersaturated state after drying. Such a supersaturated particle would exhibit gradual water uptake with increased relative humidity, even at RH lower than the RHD for the inorganic salt. Assuming deliquescent behavior of the inorganic salt (i.e. the inorganic fraction is crystalline at $RH < RHE$), this enhanced water uptake at low RH would be attributed to solely the organic fraction, thereby leading to a potential overestimation in the organic GF.

Anonymous Reviewer 2:

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

Discussion Paper



P16794 L12: Why was 1.65 assumed as the density? Please cite a source or explain how the density was determined.

A density of 1.65 was assumed based on a preliminary analysis of the density required for vacuum aerodynamic diameters (D_{va}) measured by the AMS to match electrical mobility diameters (D_m) measured by the DMA. In response to the reviewer's comments, a more thorough analysis of the density required to align the AMS and DMA distributions was performed. The methods section has been updated to explain in detail how this analysis was performed and density numbers are presented in the first part of the results section. Since the density numbers are slightly different than those originally assumed, all related data have been updated ($V F_{organic}$ numbers, organic GF calculations, R^2 value for the volume-weighted prediction in figure 3, and coefficients for the simple parameterization presented in section 3.4), though with the exception of $V F_{organic}$ magnitudes, these results typically changed on the order of 1

P16798 L4: For the observed trends, can the authors rule out potential hysteresis effects related to pressure to temperature changes?

The DASH-SP monitors temperature and pressure at a number of points in the instrument, including the DMA entrance/exit, each of the 4 humidified sample flows, each of the 4 humidified sheath flows, and at the exit of each OPC (Sorooshian et al., 2008a). Neither temperature nor pressure showed significant variability during any flight. This is now noted in the methods section.

P16801 L3: What was the pure organic acid used?

The 'organic acid' used in the calculation was a parameterization from Peng et al. (2001), which characterizes the hygroscopic growth of 'hydrophilic organics' that they studied (dicarboxylic and multifunctional acids). This has been noted in the text.

P16802 L22: Is the reason these relatively high GFs indicative of this particular case (since most of the flight were on back-to-back days in the same particular location) or

[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)[Discussion Paper](#)

is there some compelling reason to believe the results from these 7 flights would apply more generally to other air masses/locations?

While these flights were all undertaken in similar atmospheric conditions in relatively close proximity to one another, these results may be applicable to the marine atmosphere in general, given a similarly internally-mixed, uniform, highly-oxidized organic component with $V F_{organic}$ in the range presented in the current study. As discussed at the end of section 3.4, we discourage expanding these results to situations where the RH, $V F_{organic}$, or aerosol composition is significantly different than those encountered during MASE-II. These results are valid only for: 1) the range of RHs studied, 2) the range of $V F_{organic}$ observed, 3) and the characteristic O:C ratios observed in the study area.

This comment also relates to the analysis of air mass origin, presented in section 3.1. It appears (from HYSPLIT back-trajectories) that there is a strong correlation between continental air mass influence and growth factor suppression. We observed dramatic GF differences with just slight shifts in air mass origin toward a continental influence. It is conceivable that similar shifts toward urban/anthropogenically-influenced air masses could cause dramatic shifts in O:C ratios, $V F_{organic}$, and aerosol loading, and may lead to breakdowns in our parameterization. We have added brief text at the end of section 3.1, highlighting the importance of accurately predicting air mass origin in global models, in order to predict aerosol composition, and thus hygroscopic characteristics.

Brief clarifying statements about the internally-mixed nature of the aerosol, the uniformity of the organic fraction, and the highly-oxidized organic state have been added in several places in the results and discussion, in order to qualify the results presented here, and to more explicitly limit the validity of these results to the specific aerosol characteristics encountered during these MASE-II flights. While O:C ratios were stated to have been relatively constant throughout the mission, explicit O:C values for each air-mass type and flight leg type have been added, in order to support this claim of organic uniformity.

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

Discussion Paper

Interactive comment on Atmos. Chem. Phys. Discuss., 8, 16789, 2008.

ACPD

8, S8942–S8946, 2008

Interactive
Comment

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

Discussion Paper

S8946

