

Results on the aging of an organic plume in the marine atmosphere and impacts on aerosol CCN activity and hygroscopic growth are presented. The paper provides a unique perspective on aging processes and time and should be published once the concerns listed below have been addressed. I found the paper structure to be distracting from the main points. As written, the CCN and GF measurements are described first, then aerosol chemical composition is presented, and then the CCN and GF measurements are discussed in the context of the composition measurements. It would be much simpler (and easier to read) if composition were described first to lay a foundation for the CCN and GF results. In addition, this is a very complete data set with multiple measures of several parameters. It would be nice to see comparisons that would reveal the consistency between different measurements. For example, comparisons between different measures of size distributions (AMS vs. SEMS-OPS) and organic matter concentrations (AMS vs. FTIR). See specifics in the comments below.

Response: We have re-arranged the paper according to the reviewer's suggestions. Chemical composition is now discussed in Section 3.3, the contents of Section 4 (hygroscopic properties in the context of chemical composition measurements) have been moved into corresponding sub-sections of Section 3, and Section 4 has been eliminated.

p. 11921, line 7: change to "increased at LONGER plume ages"

Response: The sentence has been removed as a result of other edits.

p. 11927, lines 18 -27: How well did the AMS and combined SEMS-OPS-APS size distributions agree? Did the agreement indicate that a collection efficiency for the AMS of 0.5 was reasonable?

Response: The correlation analysis between these size-distributions indicates that a collection efficiency of 0.5 is reasonable for the background measurements. For the plume measurements, it is very hard to estimate a collection efficiency –see Figure 1: Due to the different time resolutions of the sizing instruments (and the AMS itself) in a situation in which rapid changes in particle concentrations were common and total measurement time was not long enough to build up good statistics.

For the plot, the merged total particle size distributions was corrected to represent the size distribution of particles measured by the AMS (i.e. the transmission efficiency of the AMS inlet was taken into account) following the assumptions by Quinn et al. (2006): 100% transmission at 500 nm vacuum aerodynamic diameter, 0% transmission at 1 um vacuum aerodynamic diameter, and linear decrease in transmission between these points. For conversions from vacuum aerodynamic to mobility diameter and number to mass concentration spherical particles with 2 bounding densities were considered, 1 and 2 g/cm³, corresponding to the blue and red markers in the plots.

Reference:

Quinn, P. K., Bates, T. S., Coffman, D., et al (2006). Impacts of sources and aging on submicrometer aerosol properties in the marine boundary layer across the Gulf of Maine, *J Geophys Res*, 111, D23S36, doi:10.1029/2006JD007582

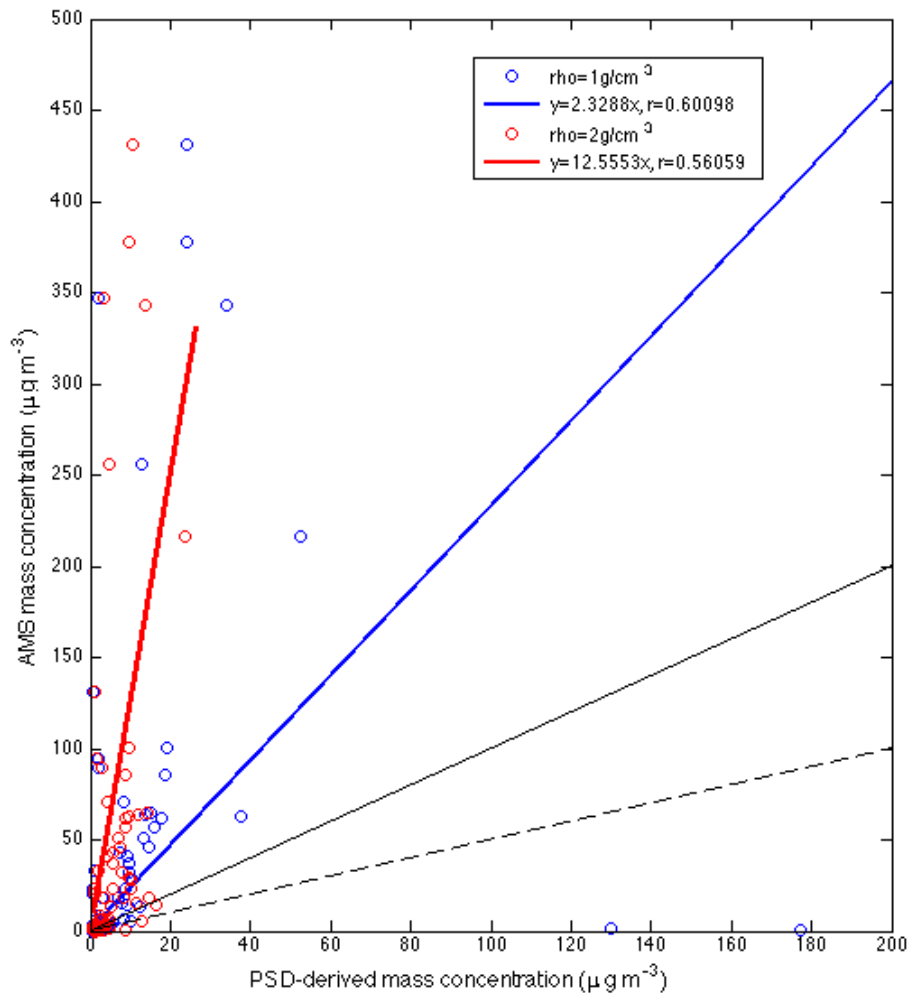


Figure 1: Comparison of particle size distribution (PSD) – derived and AMS mass concentrations for the plume measurements.

p. 11928, lines 1 – 9: How well did the AMS and FTIR organic matter agree? Again, this would lend insight into the collection efficiency of the AMS and also uncertainty associated with calculated WIOM concentrations.

Response: While differences in the sampling time for the plume measurements caused variability, the AMS and FTIR organic matter measurements were generally consistent, within the uncertainty of each instrument (+/-20%), consistent with past intercomparisons (Liu et al., 2012; Russell et al., 2009a/b; Corrigan et al., 2013 ACPD; Hawkins et al., 2010).

References:

Corrigan, A. L., Russell, L. M., Takahama, S., Äijälä, M., Ehn, M., Junninen, H., Rinne, J., Petäjä, T., Kulmala, M., Vogel, A. L., Hoffmann, T., Ebben, C. J., Geiger, F. M., Chhabra, P., Seinfeld, J. H., Worsnop, D. R., Song, W., Auld, J., and Williams J.: Biogenic and biomass burning organic

aerosol in a boreal forest at Hyytiälä, Finland, during HUMPPA-COPEC 2010, *Atmos Chem Phys Discuss*, 13, 16151-16210, 2013.

Hawkins, L. N., Russell, L. M., Covert, D. S., Quinn, P. K., and Bates, T. S.: Carboxylic acids, sulfates and organosulfates in processed continental organic aerosol over the Southern Pacific Ocean during VOCALS-Rex 2008, *J. Geophys. Res.*, 115, D13201, doi: 10.1029/2009JD013276, 2010.

Liu, S., Shilling, J. E., Song, C., Hiranuma, N., Zaveri, R. A., and Russell, L.M.: Hydrolysis of organonitrate functional groups in aerosol particles, *Aerosol Sci Tech*, 46, 12, doi: 10.1080/02786826.2012.716175, 2012.

Russell, L.M., Bahadur, R., Hawkins, L. N., Allan, J., Baumgardner, D., Quinn, P. K., and Bates, T. S.: Organic aerosol characterization by complementary measurements of chemical bonds and molecular fragments, *Atmos Environ*, 43, 38, 6100-6105, <http://dx.doi.org/10.1016/j.atmosenv.2009.09.036>, 2009a.

Russell, L. M., Takahama, S. Liu, S., Hawkins, L. N., Covert, D. S., Quinn, P. K., and Bates, T. S.: Oxygenated fraction and mass of organic aerosol from direct emission and atmospheric processing measured on the R/V Ronald Brown during TEXAQS/GoMACCS 2006, *J. Geophys. Res.*, 114, doi: 10.1029/2008jd011275, 2009b.

p.11930, lines 23 – 24: How was the organic mass fraction calculated – based on total mass measured by the AMS?

Response: Organic mass fraction was calculated as AMS organic mass:total AMS mass. We have added this clarification to the sentence: “During the plume tracking events (green shading in Figure 3), the organic mass fraction (AMS organic mass:total AMS mass) in the fresh plume (plume age A1) was $\geq 97\%$,...”

p. 11931, line 9: Figure 11 is mentioned out of order.

Response: This was done on purpose. The figure cannot be discussed to its full extent in Section 2.6 (as “Figure 5”), as it mostly shows the volume concentrations during plume tracking in the context of an analysis discussed in detail only in Section 3.5. A part of the information contained in this figure, namely the comparison of large mode aerosol volume during the two plume events, is needed in one supportive sentence in section 2.6, but does not warrant a separate figure (raising the total number of figures to 12) which would contain the same information as Figure 12.

Figure 6b: Are 30 nm growth factors shown? It is difficult to see the difference between the blue and black symbols used for the two different particle sizes.

Response: We have changed the color of the 30 nm growth factors.

p. 11935, lines 20 – 26: What is the physical reason for why primary plume particles would activate at a 0.25% supersaturation but not at 0.87%? Were the measurements at difference supersaturations made sequentially so that 0.25% was sampled before 0.87%?

Response: What appears as increase of CCN concentrations at moderate S before high S is a result of sequential measurements, rather than a true physical effect. We have deleted the confusing discussion from the paragraph.

p. 11936, Lines 5 – 23: This discussion would be easier to follow if values of d_{act} and d_K were put into a table or figure. Also, details on how the Kelvin diameter was calculated should be given.

Response: The Kelvin diameter was calculated using the Kelvin equation (e.g. Seinfeld & Pandis, “Atmospheric Chemistry and Physics”, 2nd edition, p. 463, eq. 10.86). We refer to the Kelvin diameter as the diameter of the smallest droplet of pure water that would grow through condensation at the given supersaturation. The activation diameter of a given population of (internally mixed and wettable) particles is expected to be smaller or equal to the Kelvin diameter. Since this is standard textbook content, we do not specifically cover it in the manuscript.

We agree that the paragraph as it stands is not an easy read. However, the activation diameter changed over the course of the plume sampling, and is thus not easily presentable in a table. An additional figure would lengthen the article considerably, and would inevitably present more information than needed: the specific values of d_K and their change in time were not such much of interest in this analysis than the simple “yes or no”- answer to the question: is d_{act} larger than d_K or not? Therefore, we decided to stick with the verbal description of these results.

p. 11940, lines 15 – 16: Is this supposed to say that there is a higher peak in hydroxyl in the fresh smoke than in the oil that was used to generate the smoke?

Response: The paragraph had gotten disarranged. We changed it to: “An increase in hydroxyl functional groups ($3700 - 3100 \text{ cm}^{-1}$) can be observed in the spectrum of the ship stack emissions (black line, upper panel, Fig. 7) compared to the laboratory spectrum of the ship diesel (black line, lower panel, Fig. 7). Similarly, the spectrum of the smoke oil plume (orange, yellow, and blue lines, upper panel, Fig. 7) shows a higher peak in hydroxyl than the spectrum of the generator oil (green line, lower panel, Fig. 7).”

p. 11941, lines 20 – 24: This sentence should be re-written to say that that newly-formed particles after aging contributed to CCN concentrations. As currently written it appears to say that CCN inactive particles contributed to CCN concentrations.

Response: The sentence has been removed as a result of re-arranging and editing.