Atmos. Chem. Phys. Discuss., 11, C7544–C7546, 2011
www.atmos-chem-phys-discuss.net/11/C7544/2011/

Atmospheric Chemistry and Physics Discussions

Interactive comment on "Organic carbon and non-refractory aerosol over the remote Southeast Pacific: oceanic and combustion sources" *by* L. M. Shank et al.

Anonymous Referee #3

Received and published: 9 August 2011

The paper is very well written with a logical progression through the data and a concise presentation of the results. The result is very intriguing (no evidence of a marine source of submicrometer organics) given the many recently published papers based on field measurements and laboratory studies indicating enrichment of organics in ocean-derived aerosol. The data analysis appears to be rigorous. The paper should be publishable in ACP after the issues listed below have been addressed.

General concerns:

It needs to be made clear that the AMS data provide only mass concentrations of organics, not number concentrations (the relevant parameter for CCN and aerosol-cloud

interactions) . A low submicrometer mass concentration does not necessarily imply a low number concentration. Could it be that the number concentration of organic-containing particles in the 30 to 300 nm size range was larger than the number concentration of sulfate-containing particles? Several studies have shown the existence of 50 nm organic inclusions coated by sulfates (e.g., Leck and Bigg, GRL, vol. 32, 2005).

The use of the org/so4 ratio needs to be discussed in more detail given the different production mechanisms. Oxidation of DMS to so4 has been shown to occur in the upper troposphere. The resulting so4 may be transported long distances from the point of emission. Organic aerosols may be produced locally through wind-driven mechanisms or may result from gas to particle conversion. How do the spatial and temporal scales of these different production mechanisms impact the utility of the org/so4 ratio in looking for evidence of marine organics?

Specific comments:

p. 16898, lines 11 – 17: In O'Dowd et al., GRL, 35, 2008, the correlation between water insoluble organics in aerosol and chlorophyll is weak ($r^2 \sim 0.25$). Hence, the statement that "...chlorophyll-a concentrations have been correlated with OC concentrations in clean marine aerosol collected there (Mace Head)" should indicate that the correlation was weak.

p. 16902, lines 26 – 27: How were these values determined to be representative of "background conditions"? Also, based on what is said on p. 16898, background conditions at Mace Head were determined to be 138 ppbv CO and 40 nm/m3 of BC. Why are VOCALS conditions of 61 ppbv CO and 5 ng/m3 BC the "best direct comparison between clean cases in the SEP and the North Atlantic"?

ACE Asia: How were the "org" values determined since only impactor measurements of C measured?

p. 16905, line 14: Charlson et al. (1987) doesn't provide evidence for an ocean source

of SO4 but rather summarizes results of others. The original papers should be cited.

p. 16906, line 25: For clarity, change to “In contrast, during periods of high biological activity, O’Dowd et al. reported a tenfold increase. . ..”

Figure 7: Include a legend or list in the caption label for the different colored lines in 7c.

p. 16909, lines 22 – 27: How do the size distributions of org and OPC non-volatile mass compare? Do the org size distributions indicate a larger mode?

p. 16909, line 23: aerodynamic diameter or vacuum aerodynamic diameter?

p. 16910,lines 11 – 12: Is it possible that the m/z ions chosen to represent the “org” component of the aerosol did not include organic ions that would result from wind-driven production of sea spray organic aerosol?

p. 16914, lines 11 – 16: The O’Dowd et al. (2008) relationship involved chlorophyll vs. water insoluble organics measured in ambient aerosol. It is likely that the ambient aerosol had been modified relative to nascent, ocean-derived aerosol or, as argued in the paper, is not of marine origin. If the organic concentrations were higher for the modified/non-marine aerosol than nascent aerosol, then the relationship is biased high. Assuming an intercept through zero won’t get around this problem. Including the O’Dowd figure and relationship in the paper and suggesting a way to improve it is not justified. If the point is to show that the relationship doesn’t apply to the study regions considered here, then its inclusion in the paper is justified.

Interactive comment on Atmos. Chem. Phys. Discuss., 11, 16895, 2011.