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Atmospheric
Chemistry
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Discussions

Interactive comment on “The formation, properties and impact of secondary organic aerosol: current and emerging issues” *by* M. Hallquist et al.

M. Hallquist et al.

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We would like to thank the reviewer for the comments and helpful suggestions. We have made changes to the manuscript in order to address these issues and also performed editing and updates to the document. Most of these changes are minor and the overall content and conclusions of the work remain the same. Nevertheless, the comments of the reviewer have helped us to improve the quality of the manuscript and are very much appreciated. Our responses to the specific comments are given below.

Anonymous Referee #3

1. In general, I am somewhat surprised that relatively little focus was placed on potential SOA formation processes in the marine boundary layer (i.e., ocean organics leading to particles ala the CLAW hypothesis (Meshkidze et al., 2006); nanoparticle

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formation/growth in the MBL (Russell et al., 2007; O'Dowd et al., 2002); chlorine as an oxidant (Karlsson et al., 2000; Cai et al., 2008)).

Response:

The following text has been inserted into the second paragraph to address the potential for oceanic sources of organics to produce SOA:

"The sources of biogenic VOCs (BVOCs) are mainly derived from terrestrial ecosystems. However, there are also important emissions of BVOCs from the oceans, in particular of dimethylsulfide, which is oxidized to methanesulfonic acid aerosol (Kettle and Andreae, 2000). Other identified marine SOA components are dicarboxylic acids (Kawamura and Sakaguchi, 1999) and dimethyl- and diethylammonium salts (Facchini et al., 2008). Meskhidze and Nenes (2007) suggested that marine emissions from isoprene could also be a source of SOA. The latter is still an open question, although it has been estimated that the global production of SOA from marine isoprene is insignificant in comparison to terrestrial sources (Arnold et al., 2009)."

The following text has been inserted to address the potential role of chlorine as an oxidant:

(i) Section on Atmospheric Degradation of SOA precursors (section 2.2 in ACPD version)

"Under certain conditions in the marine atmosphere, chlorine atoms (Cl) may also initiate the oxidation of VOCs."

(ii) Section on Laboratory Studies (section 2.5.1 in ACPD version):

"Obviously, the NO3 radical initiated oxidation of VOCs can be a source of SOA during nighttime conditions, as outlined in Section 3.1. The reaction of VOCs with Cl atoms is also a potential source of SOA in the marine boundary layer (e.g., Cai et al., 2008)."

2. I also believe that a greater amount of focus is needed (at least citation!) on what I

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consider to be pioneering work: that of Sullivan et al. that characterized WSOC.

Response:

The work of Sullivan et al. (2004) was in fact cited on p. 3605 of the original ACPD article, with the reference appearing on p. 3728-3729. We have expanded the section on the use of PILS-WSOC analysis to cover the work of Sullivan et al. (2004) and added three new references, including further papers by the Sullivan/Weber group. This part of the text now reads as follows:

"Particle into liquid samplers (PILS) collect particles into water for subsequent analysis (e.g., IC) (Weber et al., 2001; Orsini et al., 2003; Sullivan et al, 2004; Sorooshian et al., 2006a). A continuous measurement of WSOC (as well as inorganic and organic ions by IC) with a time resolution of minutes has been coupled to a PILS instrument and deployed in several aircraft campaigns (e.g., Sorooshian et al., 2006a, b; Peltier et al., 2007a; Sorooshian et al., 2007a, b; Weber et al., 2007). The main sources of WSOC are SOA and biomass burning OA (Sullivan et al., 2006). Miyazaki et al. (2006) showed that WSOC in Tokyo correlated well with SOC estimated with the EC-tracer method, while water-insoluble organic carbon (WIOC) correlated well with EC and CO in that study. Kondo et al. (2007) and Docherty et al. (2008) have shown that the WSOC measurement is very similar to the total AMS OOA in Tokyo and Riverside, respectively. Based on measurements made in Cairo, Favez et al. (2008) suggested that anthropogenically-dominated SOA may have a higher insoluble fraction. Recently, a measurement of total carbon following PILS collection has also been demonstrated in the field, yielding a slope similar to 1:1 within the uncertainties of both measurements. However, a non-zero intercept was also determined, which is possibly related to the inability of the TOC analyzer to digest larger insoluble particles (Peltier et al., 2007b). Another very promising recent development is the simultaneous measurement of particle and gas-phase WSOC by Hennigan et al. (2008, 2009). During the summer in Atlanta at elevated RH levels ($>$70%), a significant increase in WSOC partitioning to the particle phase was observed and followed the predicted water uptake by fine particles. These results suggest that SOA formation involving partitioning to liquid water may be a significant pathway that is often not considered. Sorooshian et al. (2007a,b) have pioneered the analysis of organic acids by IC analysis following PILS collection. These authors report that organic acids averaged 3.4% of the water-soluble PM mass during an airborne study in the Houston area. Organic acids were most abundant above clouds, presumably as a result of aqueous phase chemistry in cloud droplets, followed by subsequent droplet evaporation above cloud tops with the main product of this chemistry being oxalic acid. Suppressed organic acid formation was observed in clouds with relatively acidic droplets and lower liquid water content."

3. Clarification of Footnote 1 on page 3561 would be appreciated.

Response:

The text in this footnote has been clarified and now reads:

"A question arises on how to estimate the range of a summed quantity from the ranges of the summed components. For example if one wants to calculate the range of total SOA from the published estimated ranges for BSOA (9-50 TgC y-1) and ASOA (1.4-8.6 TgC y-1), one could add the extremes to come up with a total SOA estimate of (10.4-58.6 Tg y-1). However this procedure artificially inflates the uncertainty range. Thus here, and in later estimates, the ranges have been determined by first calculating the mean of the mid-range value (i.e. (9+50)/2=29.5 for BSOA and (1.4+8.6)/2=5.0 for ASOA)) and a confidence interval of the difference between the range values (i.e., 50-9=41 for BSOA and 8.6-1.4=7.2 for ASOA). Uncertainty propagation rules are then applied to those values to estimate the mid-range value for the sum (i.e. 29.5+5.0=34.5 for total SOA) and the size of the new confidence interval (i.e. sqrt((41*41)+(7.2*7.2))= 41.6 for total SOA). Finally the range for the summed quantity is calculated as the new mid-range value plus and minus the new confidence interval (i.e. 34.5-41.6/2=13.7 and 34.5+41.6/2=55.3 for total SOA) (D. Fahey, NOAA CSD, Boulder, personal communication, 2008). Although the various range estimates which are being added in this way have been derived in different ways and may have different statistical properties, this method is preferable to other approaches. The fluxes presented here are also rounded to reflect the large uncertainties in these calculations."

4. Correction for page 3666, line 22. The model of Griffin et al. allows organics to partition to the aqueous phase but does not account adequately for the interaction of organics and inorganics within this phase.

Response:

We thank the reviewer for pointing this out. The text has been changed accordingly.

5. Correction for page 3745. I believe that "isoprene" should be replaced with "a-/b-pinene" in the caption to Table 5. Should this also include limonene based on the 3rd entry down from the top?

Response:

This has been changed accordingly

Interactive comment on Atmos. Chem. Phys. Discuss., 9, 3555, 2009.

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