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Atmospheric Chemistry and Physics
Discussions

Interactive comment on “Particle partitioning potential of organic compounds is highest in the Eastern US and driven by anthropogenic water” *by* A. G. Carlton and B. J. Turpin

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Comment: This is a well written and timely study that examines the importance of liquid water and water soluble organic gases on organic aerosol formation in the eastern U.S. The authors use version 4.7 of the Community Multiscale Air Quality (CMAQ) model in order to conduct their study for July 2003. My major concern with this current version of the manuscript is with the model simulations. Specifically, the model simulations lack inclusion of explicit chemistry related to isoprene epoxydiols (IEPOX) (Paulot et al., 2009,Science; Surratt et al., 2010, PNAS; Lin et al., 2012, ES&T) and methacyrlic acid epoxide (MAE) (Lin et al., 2013, PNAS). Considering that most of the measurable

isoprene SOA tracers in ambient PM2.5 come from these two gas-phase water soluble epoxides (Lin et al., 2013, ACPD), it seems the model is missing an important class of water soluble species. Without including these species, the authors may be underestimating the controllable fraction even more than they currently think in this manuscript. I think the authors need to acknowledge this as an issue (or limitation) of the model in this area of the country. Please note that I’m aware of a study that is trying to incorporate these new chemistries into the CMAQ model, but it is currently under review. As a result of the latter, it may not be expected that the authors include this at the present time. I should stress here that isoprene epoxides have been linked with sulfate aerosols as a major source of organic aerosol in this region (Lin et al., 2013, ACPD; Budisulistiorini et al., 2013, ES&T). Specifically, their reactive uptake onto acidified sulfate aerosols (and thus wet sulfate aerosols) and subsequent aerosol-phase chemistry has been demonstrated (Lin et al., 2012, ES&T; Lin et al., 2013, PNAS). In an ideal world, it would be appropriate for the authors to include the isoprene-derived epoxides (MAE and IEPOX) in their simulations, especially since their oxirane ring-opening reactions are critical to SOA formation. For example, by H2O acting as a nucleophile, you get the famous (and quite abundant) 2-methyltetrols and 2-methlyglyceric acid from IEPOX and MAE hydrolysis, respectively. In addition to the lack of inclusion of explicit isoprene epoxides, why are the authors not using the Xie et al. (2012, ACPD) modified mechanism (SAPRC07) for their gas-phase simulations? It seems the modified SAPRC07 model might be more appropriate than the 99 mechanism. Do the authors agree?

It seems strange there is only a results section and no discussion section in this manuscript. In addition, I thought for ACP publications there should also be a conclusions section, right?

Response: Reviewer 2’s major concern is that the gas phase chemical mechanism employed in this CMAQ simulation does not represent formation of gas phase water-soluble epoxides from isoprene oxidation. As a result, the Reviewer is concerned that this work underestimates the controllable fraction of biogenic SOA. We agree that there are improvements in gas phase chemical mechanisms over SAPRC99, including the Xie et al. manuscript in ACPD. However, the main conclusion that water soluble organic gases are spatially widespread and partitioning to liquid water is limited by availability of particle phase liquid water is independent of the SAPRC version used. The main argument of our manuscript is that water soluble organic gases are ubiquitous in the atmosphere over the United States. Partitioning to the liquid/condensed phase is limited not by the availability of water-soluble gases (which include epoxides) but rather by concentrations and spatial patterns in predicted particle phase liquid water. To our knowledge, epoxides do not affect particle phase liquid water concentrations. Thus, inclusion of epoxides would enhance our findings and in no way contradict our conclusions.

In the revised Supplemental Information Section we also calculate gas-to-aerosol water partitioning through a simple Henry's Law approach and reach the same conclusion. We observe gas-to-aerosol water partitioning is predominantly in the Eastern U.S. due to the spatial pattern of H2Optcl. WSOMg is not limiting.

We agree with the Reviewer that formation of water-soluble epoxides is an important new development in the field and should be mentioned. We have changed the text to now explicitly discuss epoxides:

Beginning on page 12748, line 21: "It should be recognized that this summation [of WSOMg] is a lower bound estimate. Other water-soluble organics undoubtedly also form SOAAQ.... Further, gas phase isoprene photooxidation produces water-soluble epoxides (Paulot et al., 2009) recently found in ambient samples in the Southeast U.S. (Budisulistiorini et al., 2013) but not included in this model simulation."

We have added a conclusions section.

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