

Interactive comment on “Aqueous-phase mechanism for secondary organic aerosol formation from isoprene: application to the Southeast United States and co-benefit of SO₂ emission controls” by E. A. Marais et al.

Anonymous Referee #1

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The manuscript by Marais et al. (2015) explores the SOA formation from isoprene in the Southeast using an aqueous phase SOA mechanism coupled to detailed gas chemistry. This mechanism is shown to capture measured IEPOX SOA well. SOA comprises a large fraction of total aerosol in the atmosphere. However, current models cannot fully reproduce its total mass and spatial and temporal evolutions. One potential reason for the model-observation discrepancy is that current models tend to neglect the SOA formation from aqueous phase, although both lab studies and field measurements have shown the importance of aqueous phase SOA. The manuscript is thus very

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useful for SOA community. My major concern is that the paper seems to exclude the SOA formed from monoterpenes, one source that is comparable to the SOA formed from isoprene in Southeast USA (Xu et al., 2015). I am wondering how the missing monoterpenes SOA would affect the modeled OA and the comparison with measured OA. In addition, the authors should clarify some important processes in the model (e.g., emissions of POA and aerosol depositions) that are important for the prediction of OA concentrations.

Specific comments

Page 32009, line 12-14. The references are not appropriate. Both Virtanen et al. (2010) and Song et al. (2015) showed that under dry conditions, SOA particles are semi-solid or solid phase, rather than liquid phase, which suggests that their formation processes deviate from the reversible partitioning of SVOCs, because mixing within the solid particles is very difficult.

Page 32009, line 22-26. To date several models have been developed to estimate the amount of SOA formed in the aqueous phase from the regional scale to the global scale (e.g., Carlton et al., 2008; Myriokefalitakis et al., 2011; Liu et al., 2012; Lin et al., 2012; Pye et al., 2013; Lin et al., 2014). The authors should briefly discuss previous model efforts on aqueous phase SOA.

Page 32011, line 6. Can the authors explain the unit of yield “7 mol%”? Why is there “mol” in the middle?

Page 32011, line 24-27. Why did the authors change the yield of C5-LVOC reported by Krechmer et al. (2015)? Any scientific justification for this? Or just an arbitrary tuning?

Page 32014, line 14-16. Can the authors give a references for setting up the value of gamma to be 0.1? Or it is an arbitrary large number?

Page 32014, line 25-27. The reasoning is not convincing. First, although the aqueous-phase concentrations are low in cloud water, the total amount of SOA produced in

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cloud is not necessary negligible, given that the cloud water content is usually larger than aerosol water content by 3-4 orders. Second, the mechanisms for aqueous phase SOA formation are different between in cloud water and aerosol water, due to the difference in aqueous-phase concentrations between them. Consequently, the major products in cloud water are carboxylic acids (e.g., oxalic acid), while major products formed from the reactions in aerosol water are oligomers (Ervens et al., 2011).

Page 32015, line 1-2. How about the SOA formed in stratiform cloud or fogs?

Page 32016, line 11-19. Not clear to me what aerosols were treated as aqueous particles. Did they assume only sulfate as aqueous particles? Or also included the OA (POA and/or SOA)?

Page 32013, Equation (2). How did the authors predict the concentrations of [nucl] and [HSO₄⁻] in the model?

Sec. 3 GEOS-Chem simulation and isoprene SOA yields. The authors should briefly describe how they treat the POA emissions and aerosol depositions in the model.

Page 32020, line 5. "without bias" is over-stated. The model cannot capture strong IEPOX SOA peaks around June 25. The model also underestimates C5-LVOC SOA during June 18-20.

Sect. 5. Effect of anthropogenic emission reductions. Do the authors know the relative role of sulfate aerosol volume decrease vs. the aerosol acidity increase on the IEPOX SOA reduction? Which one is more important?

Page 32023, line 9-11. This is not accurate and misleading. The traditional gas-particle partitioning model follows the Pankow (1994) absorption model, which assumes reversible partitioning to pre-existing humid OA, instead of dry OA. The problem lies in that this model was usually fit to the SOA yield data obtained from chamber studies performed under dry conditions, with very low relative humidity.

Figure 3. "OA mixing ratios" should be "OA mass concentrations.

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