

Interactive comment on “Long-term observational constraints of organic aerosol dependence on inorganic species in the southeast US” by Yiqi Zheng et al.

Anonymous Referee #2

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Summary: The author compared the long-term trend of organic aerosol and sulfate mass loading from field measurement with GEOS-Chem simulations in this manuscript, with the model showing a steeper decrease in the OA mass loading and larger month-to-month variability than the field data. The long-term trend of sulfate, on the other hand, was well captured by the model when comparing with the field data. By applying coating effect, constant aerosol acidity, and a different NH₃ emission product, the modeling results match the field data better, suggesting further study is needed to address the weak dependence of OA formation and sulfate. The manuscript is overall clearly written, but may need to address the following aspects before publishing.

C1

First, the author did a good job explaining the modeling results in Section 3, but it was not very clear at certain sections which modeling results agree with the field measurement and which do not. The author may need to improve clarity about the modeling-field data comparisons when describing the modeling results, especially in line 251 when the author discusses that the contribution of IEPOX-SOA to total OA mass decreases from 61% to 28% from the early 2000s to 2013. Does this modeling result agree with previous field measurements (such as the results from Xu. et al. 2015 and Budisulistiorini, et al. 2016)? Maybe the author can include a sentence or two to compare the modeling data with the field data.

The manuscript also concluded that coating can improve the modeling result because thinner coating may enhance the formation of IEPOX-SOA. There are a few papers that also measured/modelled the effects of SOA coating on the formation of IEPOX-SOA. For instance, Zhang et al. 2018, Jo et al., 2019, and the subsequent study by Schmedding et al. 2019 discussed the effects of pre-existing coating on the formation of IEPOX-SOA. Does the result in this manuscript using ethylene glycol agree with previous studies using authentic SOA?

In addition, Jo et al. 2019 shows that the uptake of IEPOX would increase with increasing coating for most of the situations using GEOS-Chem due to increasing surface tension, contrary to a decreasing uptake with coating effect in this manuscript. Could the author explain why the trends are different in these two studies?

The manuscript used a fixed acidity to reduce the modelled month-to-month variation of IEPOX-SOA so that the results match better with the field data. It seems this large month-to-month variation in modeling only appeared before 2008, while the month-to-month variation decreased significantly after 2008 even with the default modeling scenario. Can the author explain why there is such a large difference in the month-to-month variation before and after 2008 in the default modeling scenario? Was it due to difference in NH₃ emission inventory before and after 2008 or other reasons? In addition, after updating the NH₃ inventory with the new emission inventory from CrIS

C2

satellite observation, the author stated in line 384-385 that this scenario (CT_newNH3) performed a bit worse than the fixed acidity scenario (CT_H01). I wonder whether that was due to coating effects not correctly represented by using ethylene glycol rather than the results from authentic coatings. Besides the paper mentioned above, Li et al. 2020 and Zhang et al. also used simplified equations that can estimate the phase state of a few IEPOX-SOA species that might be helpful in performing future modeling.

Minor comments: Why would there be a large increase of the default IEPOX-SOA during 2005-2008? The manuscript mentioned about higher correlation of IEPOX-SOA with acidity during this period of time. Was this abrupt increase of IEPOX-SOA caused by lower NH3 emissions between 2005-2008?

Line 171: There are multiple papers discussing about different Henry's law constants for IEPOX. The author did discuss in line 439 but probably should include other relevant papers, such as Woo et al., 2015, Budisulistiorini et al., 2016, Pye et al., 2017, and Zhang et al., 2018.

Line 477-478: One other potential mechanism I can think of is the non-linear feedback between sulfate and IEPOX-SOA production discussed in recent studies. For instance, Riva et al. 2019 and Zhang et al. both show that IEPOX-SOA fraction and sulfate are nonlinear due to chemical reactions, acidity, and the coating effects of IEPOX-SOA are intertwined and nonlinear due to the formation of organosulfates.

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C3

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C4

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