

Interactive comment on “Organosulfates in atmospheric aerosols in Shanghai, China: seasonal and interannual variability, origin, and formation mechanisms” by Yao Wang et al.

Anonymous Referee #3

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This manuscript presents a comprehensive characterization of the seasonality and interannual variability of the abundance, sources, and formation processes of organosulfates (OS) in ambient aerosols in Shanghai, China. The authors found that the contribution of OS to organic aerosols (OA) has increased obviously in recent years and that biogenic emissions such as isoprene and monoterpenes contributed significantly to the production of OS and SOA in this polluted region. The authors also found that the production of OS was largely controlled by atmospheric oxidizing capacity (i.e., Ox level), and inferred that the mitigation of Ox pollution can effectively reduce the production of OS and SOA in eastern China. In general, the measurements are well done, the manuscript is nicely written, and the results are properly discussed. This work adds

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new understandings of the characteristics and mechanisms of OS formation in polluted atmosphere. I recommend its publication in ACP after considering the following concerns.

Specific comments: L230-231. The authors stated that the high-molecular-weight CHOS species are likely to arise from anthropogenic sources. What are the molecular identities of these species? Are they accretion reaction products of smaller CHOS species? Sect. 3.3 and Table 1. Some of quantified OS species may have one or more structural isomers. Have all the isomers been considered in quantification? If so, information regarding the number of isomers and their retention times for each OS species should be provided. L360-361. The authors claimed that the abundance of anthropogenic OS decreased in 2018/2019 compared to that in 2015/2016. However, only a tiny portion of anthropogenic OS have been quantified in this study. Therefore, care must be taken when making conclusions regarding the inter-annual variation of the formation of anthropogenic OS. L368-369. Further discussions regarding the origin of phenyl sulfate and benzyl sulfate may need to be added. For example, a recent study by Huang et al. (ES&T, 2020) found that phenyl sulfate and benzyl sulfate can also be produced by sulfate radical-initiated aqueous-phase oxidation of aromatic compounds. Fig. 5. The authors may plot the averages of OS concentrations within regular Ox or ALWC intervals in the figure to better demonstrate the trends of OS vs. Ox and OS vs. ALWC.

Ref: Huang, L.B., Liu T.S., and Grassian V.H.: Radical-initiated formation of aromatic organosulfates and sulfonates in the aqueous phase, *Environ. Sci. Technol.*, 54, 11857–11864, 2020.

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