

Interactive comment on “Online gas and particle phase measurements of organosulfates, organosulfonates and nitrooxyorganosulfates in Beijing utilizing a FIGAERO ToF-CIMS” by Michael Le Breton et al.

Anonymous Referee #3

Received and published: 12 December 2017

This paper presents the characterization of organic aerosol sampled in Beijing using a FIGAERO ToF-CIMS with a focus on the organosulfates (CHOS and CHONS). In this manuscript, the authors have attempted to quantify these species and look at their distribution between gas and particle phases. While the method/idea proposed in this work is interesting and could lead to a new way to characterize such compounds, this paper cannot be accepted as it is. Indeed, the identification of the OSs is based on peak fittings that are highly questionable. In addition, the authors concluded on the validity of the method/results without any strong support/ evidence. As it is the paper

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is speculative and major improvements are needed to support this work. The authors seem to have intentionally left crucial information out of this manuscript to write (an)other paper(s). If this current manuscript cannot stand by itself (and it is the case) additional information have to be added.

The first line of the abstract: “Fast” Inlet? The correct name of the FIGAERO is Filter inlet.

SCO identification/quantification: There is clearly a lack of information in these sections and more efforts are needed to better describe validate the method/identification:

- The mass calibration is crucial in the identification of the compounds, have the authors checked if the parameters didn't change over time? The authors should propose the time series of the parameters in the SI.
- The authors need to provide the peak fittings for all of the peaks and not only two compounds. The peaks reported in Figure 1 are very small and poorly resolved, which make the “identification” questionable. If they claim they identified 17 SCOs, they should report 17 HR fittings. In addition, please add the masses and the formula of the compounds observed on the HR fittings.
- It is hard to believe that the mass resolution of the instrument was 4000 based on the peak shape/HR fittings proposed in Figure 1. The authors should bring more evidence and make sure the resolution was the same throughout the campaign.
- The authors mentioned that they have compared LC-MS and FIGAERO data. Where are these data? They need to be reported in this work. The authors said: “This analysis is not within the scope of this work and provides the basis of the correct identification to which a future paper will probe the caveats observed between the measurement techniques.” It is actually the scope of the paper, demonstrating that the FIGAERO is able to quantify OSs. Therefore comparison with well-known techniques is more than crucial. LC-MS vs FIGAERO should be added ([C],...).
- Page 8. Lines 9-12: The authors have to provide this information, that's an important parameter. In addition, we do not know if the ramp chooses during the field campaign leads to high fragmentation or not.
- Page 8. Lines 13-15. The authors mentioned that the OS concentrations are relatively

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low but in another section that they contribute to a significant fraction page 7 line 14. Make sure your statements are consistent. - Lines 14-19: The authors discussed the fragmentation issue. It is recognized that the FIGAERO or in general thermo desorption leads to the fragmentation of organic species (Thornton's group, Stark et al., 2017; . . .). While they acknowledged this problem, the authors didn't discuss this point when they determined/discussed the volatility of the OSs (Section 3.2). In the existing literature, previous FIGAERO studies have discussed this potential artifact and mention that the decomposition of oligomers could lead to lower T_{max} than expected. Would it be the case for the OSs? The authors compare the volatility of acid compounds between the Knudsen and the FIGAERO. It is interesting but not relevant to the current study as they didn't quantify/look at the carboxylic acids. Have they done the comparison with the OSs used in this study (e.g. LAS & NP OS)? - The authors should provide the T_{max} & thermograms for all the SCO and look at the evolution of individual T_{max} throughout the campaign.

Page 9. The authors should provide simple analysis before going into too many details to validate the FIGAERO data (e.g. sum of organics vs OA; sum of organics vs SO₄; sum of SCO vs OA, SO₄, . . .).

Page 10. Lines 30-34. How is it possible that IEPOX-OS could be more volatile than less oxidized compounds, such as GAS? If it was the case previous measurements realized by Stone's group in the SE-US would have revealed such phenomena. Overall this discussion is lacking comparisons with previous studies (Lopez-Hilfiker et al., 2016; Hettiyadura et al., 2017) and evidence/better constraints to validate such "results".

Page 11. Lines 23-26: That is not true. Liu et al. 2017, ACP reported the formation of such OS from the photooxidation of cyclohexene.

Interactive comment on Atmos. Chem. Phys. Discuss., <https://doi.org/10.5194/acp-2017-814>, 2017.