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Interactive comment on “Identification of particulate organosulfates in three megacities at the middle and lower reaches of the Yangtze River” by X. K. Wang et al.

JDS Surratt

surratt@unc.edu

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Although I am not a reviewer for this paper, when my Postdoctoral Scholar (Matthieu Riva) and I saw this published online in ACPD we read this with very high interest. I think the intentions of the authors are very good and this kind of data from China are highly needed in the literature. I strongly agree with the reviewer comments on the technical aspects they raised in their reviews. Specifically, I do want to point out that I agree that comparing seasonal and site trends really requires more filters, so these comparisons probably need to be modified with this in mind. However, the degree of characterization provided in this manuscript is very interesting and potentially useful, espe-

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cially the supplemental tables listing the detailed list of organosulfates (OSs) (including nitrated derivatives) identified from these samples. From carefully reviewing these detailed tables, it is clear that a lot of biogenic VOCs contribute to these OSs. However, the authors should be aware that a new study by Riva et al. (2015, ES&T) recently published from my group (in collaboration with Professor Eric Villenave's group and Professor Betsy Stone's group) revealed that organosulfur compounds (both organosulfates and sulfonates) form from the photooxidation of PAHs in the presence of sulfate aerosol. Several of the laboratory-generated PAH organosulfur compounds were identified in ambient samples collected from Pasadena, CA, USA and Lahore, Pakistan. Notably, many of the ions you report in your supplemental tables were recently characterized as sulfonates (i.e., m/z 201 [C₇H₅O₅S⁻], 215 [C₈H₇O₅S⁻], 227 [C₉H₇O₅S⁻], 229 [C₉H₉O₅S⁻] and organosulfates (i.e., m/z 231 [C₉H₁₁O₅S⁻], 257 [C₁₀H₉O₆S⁻], 274 [C₁₀H₉O₇S⁻]) from PAH (naphthalene and 2-methylnaphthalene) oxidations in presence of sulfate aerosol (Riva et al., 2015, ES&T). In addition, we noted that many of the OSs you identified were recently observed in Riva et al. (2015, Atmos. Environ.). In that study, OSs distinct to isoprene ozonolysis were identified. Many of these OSs previously observed from isoprene ozonolysis were also identified in your supplemental tables, including m/z 181 [C₅H₉O₅S⁻], 197 [C₅H₉O₆S⁻], 199 [C₅H₁₁O₆S⁻], 213 [C₅H₉O₇S⁻], 227 [C₆H₁₁O₇S⁻], 249 [C₉H₁₃O₆S⁻], and 267 [C₉H₁₅O₇S⁻]. One lesson we learned from the isoprene ozonolysis study is that some of the OSs can have similar elemental formulas as monoterpene OSs, and thus, reporting retention times or showing extracted ion chromatograms (as suggested by one of the reviewers) will be very helpful. I also wonder if the authors agree if adding another column to their supplemental tables listing the potential VOC precursor(s) and relevant study(studies) that supports this is useful? The authors may want to add a cautionary note that even if an OS has a C₅ or C₁₀ backbone (based on accurate mass measurements), these might not necessarily come from isoprene or monoterpenes as our recent work has shown that PAHs might have similar compositions or nominal masses as these products.

Thank you for considering our short comment. Sincerely, Jason Surratt and Matthieu

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