The paper by Wang et al. tackles a key issue of the identification of organosulfates in three megacities at the middle and lower reaches of the Yangtze river. At the early stage I reviewed the paper by these authors submitted to ACPD. At that time I was highly critical about the way of presentation and the lacking of a number of analytical data. Now, I can say the authors made the major revision of the ACPD manuscript addressing the most controversial points. Taking into account my review report (please see in the separate document), I would suggest the manuscript to be accepted for publication for ACP, however considering a minor revision.

p. 3, line 42: "Organosulfates (OSs)-i.e., sulfate esters and..."

In my opinion the analyzed and discussed throughout the text are compounds that are sulfate monoesters and/or nitroxy derivatives thereof. Thus, I would suggest replacing sulfate esters for sulfate monoesters.

p. 3 , line 54-64

The authors are presenting the pathways how OSs could be formed. However, the state of the knowledge on this subject is incomplete. There is no single word about other mechanisms published recently to rationalize the formation of organosulfates through the aqueous phase chemistry. In my opinion the authors are discussing the fragmentary data ignoring the route of the addition of sulfate radical anions to the C=C bonds. Was it made intentionally? If no, I would suggest completing the state of the knowledge by stressing the aqueous-phase processing of organic precursors with sulfate radical anions. For example refer to the papers by Szmigielski (*Atmos Environ*, 2015), Schoene et al. (*Phys Chem Chem Phys*, 2014); Szmigielski (Chemistry of Organic Sulfates and Nitrates in the Urban Atmosphere; NATO Science for Peace and Security Series C-Environmental Security, p 211-226); Noziere et al., (*Geoph Res Lett*, 2010); Rudzinski et al. (*Atmosph Chem Phys*, 2009).

p. 7, line 176-182

Were MS parameters, such as spray voltage, gas flows etc., optimized? If yes, please add this info to the text.

p. 7, line 180: "The Q-Exactive mass spectrometer was externally mass calibrated daily..."

Does it mean the mass spectrometer was calibrated before each analysis?

p.8, line 192

Why mass tolerance was set at plus/minus 2? Was there a difference if this parameter was lowered to 1 unit, a sit is a case of Orbitrap MS analysis?

Line 201-212: "In this study, the abundance of an OS refers to the area of its chromatographic peak, and the number of isomers for an OS is based on the number of chromatographic peaks observed for given m/z values."

This approach might lead to a systematic error since no response from internal standard was considered. I realize this approach is popular in the literature. However, the replication of the risky method is not a good manner in the analytically-oriented paper. My doubts are also about the assumption that the number of isomers for an OS is based on the number of chromatographic peaks

observed for given m/z values. What happens when peak overlapping occurs? The latter is a common case of analysis of SOA extracts!

p. 9, line 233: "Not taking into account the two double bonds involved in each sulfate group..."

Why DBE does not involve each sulfate group? Does it apply when nitroxy-organofulfates are considered?

p. 10, line 266-269

Again, in this paragraph the authors are missing the recently obtained data regarding the WM 182 OS from aqueous-phase-generated secondary organic aerosols and/or ambient SOA from Diabla Gora rural site (Szmigielski Atmos Environ 2015; DOI:10.1016/j.atmosenv.2015.10.072). In my opinion it should be taken into account while discussing the number of isomers in Riva's samples. Did the authors record the ESI spectrum (first order or MS/MS) for the isomers of the MW 182 OSs? If so, it would be benefitable to put these data in the supplementary text to let the reader see the isomeric differences

p. 16, line 438 onward

The author should also refer to other documented pathways of the OSs formation from isoprene through aqueous-phase processing (please see my comments above).