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 #1

General comments

The article entitled "Organosulfates in atmospheric aerosols in Shanghai, China: seasonal and interannual variability, origin, and formation mechanisms" by Wang et al., presents the OS that are quantified across the 4 seasons in 2015/2016 and 2018/2019, before and after reduction of anthropogenic pollution in the Eastern China, giving valuable insights to their formation mechanisms. They have shown that Ox level (O3+NO2) plays a key role in OS formation particularly in summer, while sulfate, aerosol acidity, ALWC are also important factors. Overall, this study is well executed, and the article is well written. The authors have provided sufficient evidence to support their results and

conclusions. I recommend acceptance of this manuscript after minor revision listed below.

Specific comments

Line 21: Indicate the reason for the observed decrease in OM in 2018/2019 with compared to 2015/2016 upfront

Line 22: Indicate percent contribution of OS to OA in 2015/2016 and 2018/2019 next to their average concentrations

Line 24, 29-30, 32-34: Is this the average contribution (%) in 2015/2016 and 2018/2019 of each species? If yes, indicate it clearly here as well as in the main text

Line 55-56: Replace POM with FPOM to denote "fine particulate organic mass"

Section 2.1: Give the coordinates of the sampling site

Line 137: Indicate the temperature at which the sonication took place

Line 140: Indicate time and speed of centrifugation. Also include the model of the centrifuge used

Line 145-147: Revise water as (A) and methanol as (B) to match with the gradient elution procedure, as in reversed phase separation starts with high aqueous and then elution progressed with increasing organic mobile phase

Line 155: Indicate why the purities of LAS and GAS are very low.

Line 153-158: Indicate the chemical formula of all the standards in parentheses next to their names

Section 2.3: Add table S3 to section "2.3 Auxiliary measurements" and change the order of table S2 and S3 in the SI and throughout the main text

Line 186: Indicate the spike concentration/s

Line 187: Indicate that in addition to these standards d-carene sulfate, b-caryophyllene sulfate, camphor sulfate were also used in QC.

Line 191-193: Indicate briefly how the matrix effect is evaluated for those that were already present in the samples, whether the response is neglected if it is very small compared to the spike concentration or subtracted from the total (sample+standard) before calculating the ratio

Line 148: Indicate the ESI source was operated in negative ion mode

Line 230: Is it high MW CHOS (>400 Da?) or high MW S-containing compounds (>400 Da? CHOS+CHONS) and what is the size range?

Line 231: Indicate what anthropogenic sources

Line 246: What about $C_{27}H_{53}O_{11}S^-$?

Line 255-256: Revise the sentence "The quantified OS and NOS accounted for 14-18% and 47-67% by intensity of identified CHOS and CHONS, respectively, in polluted winter days and 15-37% and 58-87%, respectively, in polluted summer days (Fig. 3c)." to read "The quantified OS and NOS accounted for 14-18% and 47-67% by intensity of identified CHOS and CHONS in polluted winter days and 15-37% and 58-87%, in polluted summer days (Fig. 3c), respectively."

Line 274: Replace the values of quantified OS in Centreville, AL using the values given in Hettiyadura et al., 2019 and revise the sentence accordingly

Line 274-275: OC%, instead of OM% is given in Hettiyadura et al. indicate it as follows e.g Atlanta, GA (2366.4 ng m-3, 16.5% of OC) (Hettiyadura et al., 2019)

Line 314-316: Also add Chen et al., 2020, which gives evidence for formation of m/z 211 and 213 from 2-MT-OS

Section 3.4: How well the OS correlates with individual Ox species (O3 and NO2 separately)?

Line 416-421: I suggest also discussing this result in the context of "Multiphase buffer theory explains contrasts in atmospheric aerosol acidity" by Zheng et al, Science, 2020 (https://science.sciencemag.org/content/369/6509/1374)

Tables and Figures

Figure 1: Indicate the coordinates of the sampling site

Figure 1: Increase the symbol size of sampling site or use a star to show the sampling site

Figure 2 and 4: Indicate the 4 seasons in the time series either in the figure or in the caption

Figure 2: Indicate the whole Y axis scale of [H+] concentration in scientific notation (10-x..)

Figure 5: Indicate what is Ox in the figure caption

Figure S1: Indicate collision energy of each

Figure S1: Revise the caption to indicate that the S-containing fragments are labeled or label all the fragment ions

Table 1: Add retention times

Table S1: Indicate the spike concentration/s

Table S1: Add spike recoveries of LAS and GAS

Table S2: Clearly indicate the spike concentration of individual OS and/or add the information presented in the table caption to the table

Table S2: Indicate the dates of the samples used

Technical corrections

Table 1: Subscript the numbers of the formulas in the first 2 entries

Table 1: Adjust the column width to indicate anthropogenic in one line

Line 86, 138, 142, 144: Define all acronyms where it first appears (OM, PTFE, UPLC, BEH...)

Line 116: PRD should be corrected as YRD

Line 144: Add 'Waters' in parentheses

Line 169: Organic matter is already defined at line 86, use acronym

Line 288: Correct "(C7H9O7S)" as "(C7H9O7S-)"

Line 297: Use 2-MA-OS, as the acronym is introduced before

Use acronyms consistently e.g. OSi instead of OSI (line 394) and Ox or Ox (subscripted) consistently

Add GA after Atlanta and AL after Centreville (e.g. Atlanta, GA and Centreville, AL)