Response to Reviewer #3

We are grateful to the reviewer for their thoughtful comments on the manuscript. Our point-to-point responses to each comment are as follows (reviewer comments are in black, our responses are in blue, and revised texts that appear in the manuscript are in quotes and underlined).

Comments:

General Comments:

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 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?

Response: We have listed the molecular formulas of high-molecular-weight CHOS species in Table S4. We found that although several species had two sulfur atoms, which may be accretion reaction products of smaller CHOS species, most of them only had one sulfur atom.

Table S4. The molecular formulas of high-molecular-weight CHOS species observed during the pollution periods of winter and summer in 2019.

Winter in 2019		Summer in 2019	
Formula	m/z	Formula	m/z
$C_{18}H_{33}O_8S^-$	409.1896	*$C_{18}H_{13}O_8S_2^-$	421.0052
$C_{20}H_{37}O_7S^-$	421.226	$C_{22}H_{21}O_7S^-$	429.1008
$C_{19}H_{37}O_8S^-$	425.2209	$C_{14}H_{13}O_{14}S^-$	437.0026
$C_{22}H_{43}O_6S^-$	435.278	$C_{15}H_{15}O_{14}S^-$	451.0183
*$C_{16}H_{23}O_{10}S_2^-$	439.0733	$C_{15}H_{17}O_{14}S^-$	453.0339
$C_{18}H_{31}O_{10}S^-$	439.1638	*$C_{15}H_{19}O_{12}S_2^-$	455.0318
$C_{24}H_{37}O_8S^-$	485.2209	$C_{15}H_{15}O_{15}S^-$	467.0132
$C_{30}H_{27}O_{12}S^-$	611.1223	$C_{24}H_{37}O_8S^-$	485.2209
$C_{37}H_{49}O_7S^-$	637.3199	$C_{34}H_{49}O_5S^-$	569.3301
$C_{31}H_{57}O_{14}S^-$	685.3469	$C_{35}H_{53}O_6S^-$	601.3563
		$C_{36}H_{47}O_6S^-$	607.3093
		$C_{37}H_{49}O_7S^-$	637.3199
		$C_{37}H_{59}O_7S^-$	647.3981
		$C_{43}H_{63}O_5S^-$	691.4396

*The high-molecular-weight CHOS species with two sulfur atoms may be accretion 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.

Response: All the isomers were considered in quantification and we have added the retention times for each isomer to Table 1.

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.

Response: Thanks for the suggestion. We have rephrased the statement in L360-363.

"Although the concentration of quantified OS_a decreased in 2018/2019, we are not sure whether the total concentration of OS_a decreased because only a small fraction of OS_a were quantified. The inter-annual variations of OS_a warrants further studies."

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.

Response: We have added further discussions regarding the origin of phenyl and benzyl sulfate to the manuscript.

"Phenyl sulfate and benzyl sulfate may be produced by photooxidation of naphthalene and 2-methylnaphthalene (Riva et al., 2015) and/or sulfate radical reaction with aromatic compounds such as benzoic acid and toluene in the aqueous phase (Huang et al., 2020)"

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.

Response: We have added the averages of OS concentrations within regular Ox or ALWC intervals (every ten unit-length) in the figure.