Yang et al. investigated the multiphase reaction of aromatic organosulfates with OH radicals, providing pH dependent rate constants, organosulfate products, and produced inorganic sulfate. The authors additionally show evidence for increased absorbance in OS reaction products. The main kinetically related goals of this manuscript are closely related to that of Gweme and Styler (2024) (referenced by the authors). And indeed, these authors find good agreement between their kinetic results and those from Gweme and Styler (2024). But at least as worded in the introduction, the authors suggest that the novelty and necessity of their new study here is to investigate the possibility of inorganic sulfate as a product (something Gweme and Styler did not provide evidence for, at least for aromatic organosulfates). I have concerns about the experimental design and quality control related to their observation of inorganic sulfate as described below. Recent work from their own institution on inorganic sulfate products is also not referenced (https://acp.copernicus.org/articles/25/8575/2025/). I have listed my concerns below, which largely pertain to novelty and need for increased experimental evidence in some cases. Overall, I believe that my concerns should be addressed before I can recommend publication.

We gratefully thank for your comments and suggestions to improve the manuscript. We have revised the manuscript by carefully considering the comments. The major revisions are specified as follows:

(1) We have provided additional evidences to support the finding of inorganic sulfate formation in the reactions of the studied aromatic OSs with OH radicals, and the related discussion has been revised accordingly.
(2) We have reconstructed the discussion about the reliability of the method employed for kinetic studies.
(3) We have added the discussion about the mechanism for the changes in optical properties at the molecular-level.
(4) We have rewritten the vague sentences, and corrected the typos and grammatical errors of the manuscript.

More details about the revisions can be found in the revised manuscript and in our response below.

(Q1) *Recent work from their own institution on inorganic sulfate products is also not referenced (https://acp.copernicus.org/articles/25/8575/2025/).*
Response: We have briefly introduced and cited this work in Section 1 (Introduction) in the revised manuscript.

(Q2) *Line 35-36: This statement is unsupported and too speculative for an introduction.*
Response: We have reconstructed the introduction about aromatic OSs in the atmosphere as "In urban areas, in addition to isoprene and monoterpenes derived OSs, other OSs containing an aromatic ring were also observed in collected aerosols (Kundu et al., 2013; Huang et al., 2018; He et al., 2022; Wang et al., 2021). He et al. (2022) identified four kinds of aromatic OSs (i.e., phenyl sulfate, methylphenyl sulfate, benzyl

sulfate and phenethyl sulfate) with concentrations ranging from 0.04±0.08 to 2.37±3.59 ng m^{-3} in $PM_{2.5}$ collected in Chengdu, China. Previous study observed that aromatic OSs can account for up to 63.5% of the total identified OSs in a megacity in China (Ma et al., 2014)." This sentence is now deleted in the revised manuscript.

(Q3) *Line 51: No need to say 'potential' – Hydrolysis is indeed known to removed OSs.*
Response: We have deleted it in the revised manuscript.

(Q4) *Line 54: …radicals that reacted…*
Response: We have corrected this typo in the revised manuscript.

(Q5) *Line 57: "border kind" is at least an unfamiliar term to me, consider using something else.*
Response: We have changed it to "more kinds of" in the revised manuscript.

(Q6) *Line 63: "… the partial of OSs…" is confusing as written.*
Response: We have rewritten this sentence as "During the oxidation of some OSs (e.g., methyl sulfate, ethyl sulfate, and α-pinene derived OSs) by OH radicals, it is found that OSs can also return back to inorganic sulfate except for new OS formation (Kwong et al., 2018; Xu et al., 2020; Xu et al., 2024)."

(Q7) *Lines 65 – 70: No reference is given for this. Please correct, as the authors seem to use this evidence as the paramount motivation for their study here. Presumably this is Gweme and Styler (2024), but it isn't confirmed.*
Response: Yes, this reference is Gweme and Styler (2024). We have added this citation at the end of sentence.

(Q8) *Line 88: Is this a summed concentration? Or for each individual component?*
Response: The value of 0.05 mM is the initial concentration of each aromatic OS.

(Q9) *Line 96 – 97: This statement needs experimental or referenced support. Even though I appreciate that hydrolysis of aromatic organosulfates is slow, it is critical that the authors make it defensible beyond doubt that hydrolysis isn't a kinetically meaningful contributor to their aqueous phase analyses.*
Response: The assumption that aromatic OS and the reference compound are just consumed due to the OH radical oxidation is one of important principles in measuring reaction rate constant using the competition kinetics method. We agree well with reviewer that it is essential to preclude interference from other reactions, such as hydrolysis, during kinetic measurements. In this study, control experiments were conducted for the reactions of aromatic OS, BA, and H_2O_2 in the absence of illumination. The results show that hydrolysis of aromatic OS has a negligible effect on the rate constant measurement. Additionally, we performed experiments on the direct photolysis of each aromatic OS in the presence of H_2O_2 to confirm that photodegradation of OS cannot interfere with kinetic measurements either. In the

revised manuscript, we have added reconstructed the discussion about the reliability of method employed.

(Q10) *Line 122: Change 'spectrometry' to 'spectrometer'*
Response: "spectrometry" is now changed to "spectrometer" in the revised manuscript.

(Q11) *Line 150: Missing superscript formatting*
Response: We have corrected this typo in the revised manuscript.

(Q12) *Line 196: Typo for 'radical'*
Response: We have corrected this typo in the revised manuscript.

(Q13) *Line 202-203: This is more interesting to me than the motivation that was framed in the introduction.*
Response: Thanks for your suggestion. In the revised manuscript, we have moved this conclusion statement to the end of Section 3.2 after we discussed the products of the OH radical oxidation of p-tolyl sulfate and 4-ethylphenyl sulfate.

(Q14) *Line 213: Did the authors conduct any full scan mass spectrometry acquisitions of individual organosulfate standards? The detection of m/z 97 does not exclusively indicate inorganic sulfate, as it may also be the result of in-source fragmentation of their organosulfate standards. While electrospray ionization is soft, is still doesn't eliminate the possibility of such byproduct fragments formed from organic precursors. This needs to be clarified in detail. Line 228: I'm not sure this is true as written. Again, I need to see more quality control experiments of the standards themselves to be sure of what the exact origin is of the author's inorganic sulfate peak.*
Response: The mass spectra of samples from reactions without illumination under the same experimental conditions were also collected as the comparison. Although a small SO_4^{2-} peak was observed in IC prior to illumination, originating from impurities in the water blank and the aromatic OS commercial standards, the gradual increase of SO_4^{2-} peak with reaction time after illumination indicates that SO_4^{2-} was formed through the oxidation of phenyl sulfate by OH radicals. This is further supported by mass spectrometry results. A peak at *m/z* 97, which can be assigned to HSO_4^-, was observed in the mass spectra of phenyl sulfate reacting with OH radicals. The possibility that this signal resulted from fragmentation of phenyl sulfate during electrospray ionization can be ruled out, as its intensity was substantially higher than that of the *m/z* 97 signal in the extracted ion chromatogram (EIC) of the organosulfate standard, as well as in the same reactions performed under dark conditions. The pronounced signal of m/z 97 (HSO_4^-) observed in the mass spectra provides robust evidence for the formation of inorganic sulfate during the reactions. Based on the IC and mass spectrometry results, it can be concluded that phenyl sulfate can revert to inorganic sulfate during the liquid-phase reaction with OH radicals. In the revised manuscript, we have reconstructed the discussion about the observation of inorganic sulfate formed during the reactions and provided more evidences including IC results, mass spectra of reactions with/without

illumination and extracted ion chromatogram (EIC) comparison results to support this conclusion.

(Q15) *Line 225 – 227: If the authors cannot adequately defend their observations (especially for a chemical product that is so important to their studies motivations and novelty), then it should not be included in the manuscript.*
Response: We have reconstructed the discussion about the formation of inorganic sulfate during the reactions. This sentence is now deleted in the revised manuscript.

(Q16) *Line 243 – 244: This is vague and unspecific. Statements such as this are best introduced in the introduction with referenced support.*
Response: We have moved this statement to the Introduction to introduce the aim of our work as "Moreover, given that the oxidation of aromatic organic compounds often induces significant alterations in the optical properties of the reaction system (Li et al., 2021; Arciva et al., 2024), the changes of optical properties during these processes were also examined.".

(Q17) *Line 250: This is just my opinion, but descriptors like 'remarkable' don't offer much for a scientific observation. What exactly makes it 'remarkable?*
Response: We have changed it to "increased" in the revised manuscript.

(Q18) *Section 3.3: The authors talk about the formation of brown carbon species. They do not provide molecular-level data to better defend this observation despite having the capability to do so (i.e., a high-resolution mass spectrometry system). There are indeed interesting and exciting observations reported in this paragraph. But while they do have the absorbance data, some molecular-level evaluation of these formed compounds would be much more convincing and comprehensive to defend/prove these observations. This type of data, if acquired, should then also be featured in a key manuscript figure.*
Response: Thanks for your comment. In addition to the absorption spectra obtained by UV–vis spectrophotometry, we have identified the molecular composition of the main chromophores generated in the liquid-phase reactions of aromatic OSs with OH radicals and characterized their optical properties using UPLC-PDA-HRMS. The characteristic absorption peaks of the three aromatics were located at 262 nm for phenyl sulfate, 266 nm for p-tolyl sulfate, and 266 nm for 4-ethylphenyl sulfate. After OH radical oxidation, the newly formed chromophores contributing most to light absorption were $C_6H_5O_5S^-$ (characteristic absorption peak at 274 nm), $C_7H_5O_5S^-$ (characteristic absorption peak at 258 nm), and $C_8H_7O_5S^-$ (characteristic absorption peak at 254 nm), respectively. The formation of these new OSs indeed significantly alters the optical properties of the solution. Additionally, the light absorption properties of other products were characterized. Hydroxylated products exhibited a red shift in their characteristic absorption peaks due to the electron-donating effect of the hydroxyl group, whereas the formation of carbonyl functional groups results in a blue shift. Multi-hydroxylated products are likely responsible for the enhanced light absorption and the red shift of the fluorescence band observed in the reaction system. In the revised manuscript, we have

reconstructed the discussion on the mechanism underlying the changes in optical properties at the molecular-level.

(Q19) *Figure 4: Please be more descriptive (i.e., avoid acronyms) whenever possible in figure captions and axes. While I hope that most don't just look at figures, many inevitably do, so it is best to avoid acronyms as much as possible.*
Response: We have replaced the abbreviations with their corresponding full name in figure captions and axes.