The article by Yang et al. investigates the kinetics and products of liquid-phase OH radical oxidation of three aromatic organosulfates (OSs), namely phenyl sulfate, p-tolyl sulfate, and 4-ethylphenyl sulfate. The authors employed a batch reactor containing an aqueous solution of aromatic OSs, H_2O_2, and dissolved O_2 under xenon lamp irradiation to study OH radical oxidation processes. Concentrations of the OSs were quantified using LC–MS analysis, while UV–visible absorption and fluorescence spectra were also measured. The results and conclusions provide new insights into this field, including faster OH oxidation rates under more alkaline conditions, the formation of inorganic sulfate, and the production of brown carbon–like species. Incorporation of these findings into atmospheric models could improve understanding of the atmospheric impacts of aromatic OS oxidation. However, the manuscript would benefit from additional work and refinement prior to publication in an EGU journal, particularly through the inclusion of missing references, more detailed discussion of the results, and a clearer overall structure.

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 changed the title of the manuscript.
(2) We have revised the discussion about the possible explanations for the difference in rate constant at different pH values.
(3) We have reconstructed the discussion about atmospheric lifetime of aromatic OSs under different environmental conditions.
(4) We have provided more details about the quantitation method of three aromatic OSs and benzoic acid.
(5) We have cited the related references, 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) *Change "multiphase" to "liquid-phase" or delete "multiphase" from the title, the whole manuscript and supplement. In atmospheric science, the "multiphase reaction" generally means heterogeneous reactions happening between different phases (e.g., gas and liquid). To study multiphase reactions, people generally react gas-phase VOCs or oxidants with liquid-phase aerosol inside a chamber. All papers with titles including "multiphase" the authors cited used a chamber to study the aerosol-gas reaction. This study used a water solution containing reactants in batch reactor and studied liquid-phase reactions.*

Response: We have changed the title of manuscript to "Liquid-phase reactions of aromatic organosulfates with OH radicals: Kinetics, mechanisms, and environmental effects".

(Q2) *In Section 2.3, explain in more detail the quantification of OSs compounds and do a mass closure calculation. The authors said that the concentration was detected by LCMS analysis. However, the specific quantification process was unclear in the paper.*

Two essential questions the reader may ask: What standards the authors used to quantify OSs and their oxidation products? What percentage of total compounds quantified using both IC and LC/MS (a mass closure calculation)?

Response: Thanks for your comment. In this study, for kinetic studies, the concentrations of aromatic OS and benzoic acid were simultaneously quantified by their corresponding commercial standards using UPLC. For mechanism studies, due to the lack of authentic standards, qualitative rather than quantitative measurements of new OSs formed during the reaction of aromatic with OH radicals were carried out using LC-MS. Moreover, it is noted that inorganic sulfate species can also be in the form of SO_4^{2-} (*m/z* 48) in addition to HSO_4^- (*m/z* 97) in the mass spectra. Peak at *m/z* 48 is below the mass range of the instrument (50–500), and the distribution of different forms of inorganic sulfate species is significantly affected by the solution pH. Therefore, it seems that accurately measuring inorganic sulfate concentration via LC-MS using the external standard method is not feasible. In the revised manuscript, we have provided further details regarding the quantitation method for the three aromatic OSs (i.e., phenyl sulfate, p-tolyl sulfate, and 4-ethylphenyl sulfate) and for benzoic acid.

(Q3) *The authors observed that aromatic OSs reacting with OH radical more rapidly in alkaline solution (pH 8) compared to acidic solution (pH 3). However, I am not entirely convinced that the acidity is the reason for this change in OH oxidation kinetics. As the solution at pH 3 contains HCl and the solution at pH 8 contains phosphate buffer (Na_2HPO_4 and NaH_2PO_4). These additional compounds in solutions may affect the OH radical oxidation kinetics. The authors need to prove that the change in oxidation kinetics is indeed caused by acidity change. The authors also need to explain why they chose phosphate buffer to adjust the pH, as phosphate is not a ubiquitous species in atmosphere.*

Response: The acidity of the solution increased slightly after the reactions for kinetic studies. This increase is negligible at an initial pH of 3 but becomes significant at an initial pH of 8. Therefore, although phosphate is not a ubiquitous species in the atmosphere, a phosphate buffer was still used to maintain the pH at 8 throughout the reactions. Phosphate buffers have also been employed in similar kinetic studies in previous work (Witkowski et al., 2019).

Previous studies have shown that even though methoxyphenols, benzene-diols, and highly substituted phenols exist predominantly in protonated form within the pH range of 2 to 6, their second-order reaction rate constants with OH radicals at pH 2 are generally lower than those at pH 5 or 6. (Smith et al., 2015; Arciva et al., 2022). Moreover, we observed variations in product distributions at different pH values, suggesting the involvement of distinct reaction mechanisms, thereby affecting the kinetics of OH oxidations. Other possible reasons for the difference in rate constants at different pH values may include uncertainty in the rate constant of the reaction of reference with OH radicals, as well as differences in the reaction matrix (HCl *vs* phosphate buffer) as noted by the reviewer (Arciva et al., 2022). In the revised manuscript, we have revised the discussion about the possible explanations for the difference in the rate constant at different pH values.

(Q4) *Add a section specifically explaining the calculation of the lifetime of OSs against OH radical oxidation. The authors could include more information on the ambient OH concentrations. Ambient OH concentrations [•OH] could fluctuate greatly from remote to urban environments, the authors could discuss a little more about the ambient OH concentrations across different regions, especially on the urban sites. Moreover, in Lines 298-299, the authors mentioned that "The lifetime of aromatic OSs is significantly shorter than those of aliphatic OSs (Gweme and Styler, 2024; Lai et al., 2025)." However, the authors didn't further explain why. Also, the cited studies reported oxidation kinetics in liquid-phase. There are other laboratory studies (listed below) reported the heterogeneous OH oxidation kinetics of aliphatic OSs. The authors may also want to include these studies in the discussion of aerosol lifetimes.*

Response: Thanks for your suggestion. Using the determined *k* value along with modeled OH radical concentrations (Herrmann et al., 2005, 2010), the corresponding lifetime ($\tau=1/k_{OS}\times[\bullet OH]$) of aromatic OSs was calculated. In contrast to urban areas ($[OH]_{\text{urban aerosol}} = 4.4 \times 10^{-13}\,M^{-1}$; $[OH]_{\text{urban cloud}} = 3.5 \times 10^{-15}\,M^{-1}$), remote areas exhibit higher liquid-phase OH concentrations ($[OH]_{\text{remote aerosol}} = 3 \times 10^{-12}\,M^{-1}$; $[OH]_{\text{remote cloud}} = 2.2 \times 10^{-14}\,M^{-1}$). The concentration of OH radicals in aerosol is typically higher than that in cloud. Consequently, the lifetime of aromatic OSs is about 1 minute in remote aerosols and can be up to 16 hours in urban cloud water, highlighting the significant influence of environmental conditions on their persistence. Previous study reported that the lifetime of aliphatic OSs in such varied environments ranges from 31 minutes to 32 days (Gweme and Styler, 2024). Aromatic OSs seem to have higher reactivity towards OH radicals compared to aliphatic OSs, suggesting that they play a more important role in the sulfur cycle given that some of aromatic OSs can return to inorganic sulfate as well. In addition to lifetimes in aqueous environments, previous studies have also calculated the atmospheric lifetimes of several aliphatic OSs via heterogeneous OH radical oxidation based on measurements of their uptake coefficient (Kwong et al., 2018; Lam et al., 2019; Xu et al., 2022). The results indicate that the atmospheric lifetimes of these OSs differ between liquid-phase and heterogeneous OH radical oxidation pathways. However, heterogeneous OH radical oxidation experiments for aromatic OSs were not conducted in this study. Thus, we cannot directly compare the lifetime of aromatic and aliphatic OSs through the OH radical heterogeneously oxidation, and this warrants further investigations. In the revised manuscript, we have added the discussion on the atmospheric lifetime of aromatic OSs under different environmental conditions and improved the comparative analysis of the lifetimes of various OSs, including heterogeneous OH radical oxidation.

(Q5) *The first row of the Table S3 lists the concentration of OH radicals while other rows list the lifetimes (days). It would be better to put the concentration of OH radicals as a note at the end of the table or on the title.*

Response: We have put the concentration of OH radical as a note at the end of the table.

(Q6) *Lines 21-25: Cite study Hu, W. W., P. Campuzano-Jost, B. B. Palm, et al. "Characterization of a Real-Time Tracer for Isoprene Epoxydiols-Derived Secondary*

Organic Aerosol (IEPOX-SOA) from Aerosol Mass Spectrometer Measurements." Atmospheric Chemistry and Physics 15, no. 20 (2015): 11807–33. https://doi.org/10.5194/acp-15-11807-2015.
Response: We have cited this reference in the revised manuscript.

(Q7) *Lines 34-35: The focus of the study is on the aromatic OSs, I would suggest the authors focus more on the introduction of aromatic OSs in urban sites. The authors could lists more studies which detected aromatic OSs in the urban atmosphere (Huang et al.,2020; Li et al., 2023). Then mention the Ma et al., 2025 study to show their contributions to ambient aerosol. a. Huang, Liubin, Tongshan Liu, and Vicki H. Grassian. Radical-Initiated Formation of Aromatic Organosulfates and Sulfonates in the Aqueous Phase. Environmental Science & Technology 54, no. 19 (2020): 11857–64. https://doi.org/10.1021/acs.est.0c05644. b. Li, Ailin, Xiaodi Shi, Xinghua Qiu, et al. "Organosulfur Compounds in Ambient Fine Particulate Matter in an Urban Region: Findings of a Nontargeted Approach." Science of The Total Environment 887 (August 2023): 164114. https://doi.org/10.1016/j.scitotenv.2023.164114.*
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)."

(Q8) *Lines 41-42: Add references to "previous studies revealed that the addition of sulfate radicals on the C=C bond can result in the formation of OSs in aqueous aerosols".*
Response: We have cited related references at the end of sentence in the revised manuscript.

(Q9) *Lines 54-55: I don't see the point of stating the effects of carbon chain length on the OH oxidation kinetics in the introduction part, as the authors later explains the chain length effects in Section 3.1. So I would recommend the authors to just summarize the oxidation kinetics reported from previous studies for aliphatic OSs in introduction part.*
Response: In the revised manuscript, we have shortened the discussion about chain length effects in Section 1 (Introduction) and added more details about this effect in Section 3.1.

(Q10) *Line 67. The author mentioned that "a very recent study…". However there is no reference to this study.*
Response: We have cited the related reference in the revised manuscript

(Q11) *Lines 72-74: The sentence "In this study, we investigated the multiphase reaction of atmospherically relevant aromatic OSs (i.e., phenyl sulfate, p-tolyl sulfate, and 4-ethylphenyl sulfate) with OH radicals (He et al., 2022)" is somewhat confusing, as the reference (Hu et al., 2022) appears to support the present study rather than the atmospheric relevance of the compounds. I suggest separating this into two sentences. For example, the authors could write: "Phenyl sulfate, p-tolyl sulfate, and 4-ethylphenyl sulfate have been observed as atmospherically relevant aromatic OSs in Chengdu, an urban environment during winter (He et al., 2022). In this study, we investigated their liquid-phase reactions with OH radicals."*
Response: Thanks for your suggestion. We have rephrased this sentence in the revised manuscript.

(Q12) *Lines 85-86: add reference Pye et al., 2020 review paper on aerosol acidity, focus more on the aerosol in the urban sites.*
Response: We have cited this reference in the revised manuscript.

(Q13) *Line 88: the authors could explain more why choose the initial concentration of aromatic OSs to be 0.05 mM? How is this value compared to that in ambient aerosol?*
Response: The typical concentrations of aromatic OSs in aerosols range from a few hundredths to several ng m^{-3} (Kundu et al. 2013; He et al., 2022). Assuming an aerosol liquid water content (ALWC) of 1–100 μg m^{-3} (Guo et al., 2015; Su et al., 2022), the corresponding concentration in aerosols is estimated to be on the order of several μM to several mM. In this study, the concentration of aromatic OSs (50 μM) used falls within this atmospherically relevant range.

(Q14) *Lines 89-90: "additional experiments with elevated concentrations of OSs (0.5 or 1 mM) were carried out in order to observe obvious product signals and optical change characteristics." Does the high concentration of OSs change the oxidation kinetics determined using lower concentration (0.05mM)? The authors could report both the oxidation kinetics (k) for low and high concentration experiments if possible.*
Response: Unlike studies that directly determined the rate constants of reactants with oxidants based on a pseudo first-order assumption, our study employed a competition kinetics methods using BA as the reference to measure the second-order rate constants for the oxidation of aromatic OSs by OH radicals. Therefore, we did not investigate the effect of concentration on the rate constants. Although experiments were also performed at elevated OS concentrations to observe obvious changes in optical properties, BA was not added to the solution for these experiments to avoid interference with product analysis. Consequently, despite having decay curves of aromatic OSs under high-concentration conditions, we could not calculate their reaction rate constants due to the absence of BA.

(Q15) *Lines 154-155: I recommend the authors also cite following laboratory studies (listed below) on the heterogeneous OH radical oxidation of isoprene epoxy diol-derived secondary organic aerosol (IEPOX-SOA). These two studies also suggest that*

reaction rate is faster for aliphatic OSs with longer carbon chain length: a. Yan, J.; Zhang,Y.; Chen,Y.; Armstrong, N.C.; Buchenau, N. A.; Lei, Z.; Xiao,Y.; Zhang, Z.; Lambe, A. T.; Chan,M.N. Kinetics and Products of Heterogeneous Hydroxyl Radical Oxidation of Isoprene Epoxydiol-Derived Secondary Organic Aerosol. ACS Earth Space Chem. 2023, 7, 1916. b. Armstrong, N. C.; Chen, Y.; Cui, T.; Zhang,Y.; Christensen, C.; Zhang, Z.; Turpin, B. J.; Chan, M. N.; Gold, A.; Ault, A. P. Isoprene Epoxydiol-Derived Sulfated and Nonsulfated Oligomers Suppress Particulate Mass Loss during Oxidative Aging of Secondary Organic Aerosol. Environ. Sci.Technol. 2022, 56, 16611

Response: We have cited these two references in the revised manuscript.

(Q16) *Lines 207-208: add references to the "previous studies".*

Response: We have cited the related reference in the revised manuscript.

(Q17) *Line 212: change "sulfate anions ($\bullet SO_4^-$)" to "sulfate radical anions ($\bullet SO_4^-$)" assuming the "•" means "Radicals"*

Response: We have corrected this typo in the revised manuscript.

(Q18) *Lines 222-224: add references on "benzoic acid can undergo decarboxylation reactions", and "the elimination of the sulfate group from phenyl sulfate".*

Response: We have cited the related reference in the revised manuscript.

(Q19) *Lines 230-231: the author wrote "Figure S7 reveals that $C_6H_5O_5S^-$ exists as three isomeric forms: ortho, meta, and para hydroxyphenyl sulfate, exhibiting distinct distribution patterns at different pH values." However, Figure S7 only shows the abundance peak of $C_6H_5O_5S^-$ including all structures proposed in Figure 2 mechanism. Figure S7 can only tell us that the OH oxidation happened much faster in alkaline environment (pH 8) compared to more acidic environment (pH 3). I suggest the author don't mention the three isomeric forms, and reword the sentence.*

Response: Thanks for your suggestion. In the revised manuscript, we have deleted the discussion of the effects of pH on the products from the OH radical oxidation of phenyl sulfate.

(Q20) *Line 304: In ambient air, a compound with "shorter lifetime" generally considered not as important as the "long lifetime" species regarding their climate impacts. You could reword this to be "the high abundance of aromatic OSs and their fast reactivity with OH radicals in urban environments further underscores their potentially significant role…".*

Response: Thanks for your suggestion. We have changed this expression in the revised manuscript.

(Q21) *In section 4, the author didn't mention the formation of sulfate anions, which is directly related to the sulfur cycle.*

Response: We have added the related discussion about the implication of inorganic

sulfate formation during the oxidation of aromatic OSs in Section 4 in the revised manuscript.

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