Referee Report

Title: Atmospheric Organosulfate Formation Regulated by Continental Outflows and Marine Emissions over East Asian Marginal Seas

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Summary

This manuscript presents cruise observations of atmospheric organosulfates (OSs) over the Yellow and Bohai Seas, quantifying seasonal variability and source influences using authentic standards and offline liquid chromatographic techniques. The study adds valuable insight into the contribution of biogenic *vs.* anthropogenic precursors to OSs formation and discusses the relative roles of OSs and methanesulfonic acid (MSA) in the marine sulfur cycle. The authors structure their work by proposing that biogenic OSs, especially in summer, may be comparable to the role of MSA in some marine boundary layer conditions.

Evaluation of Authors' Response to Major Previous Comments

The authors have responded thoroughly to the previous review comments. I elaborate on each of the key issues previously raised and how the authors have addressed them:

1. OSs vs. MSA – Abundance and Tracer Role

Original Comment: The authors should clarify the respective roles of MSA and OSs as tracers of marine biogenic activity, including a discussion of differences in formation pathways, physicochemical properties, and climate relevance.

Authors' Response: The authors revised both the Introduction and Section 3.2 to more clearly define the chemical and atmospheric distinctions between MSA and OSs. They described their respective precursors (DMS for MSA and isoprene/monoterpenes for OSs) and cited laboratory studies showing differences in hygroscopicity and surface tension. Furthermore, they added a brief statement proposing OSs as supplementary tracers of phytoplankton emissions in high Chl-a and SST regions (Lines 396–398).

2. Analytical Uncertainty of HPLC-MS: LODs, Recovery, and Measurement Precision

Original Comment: Quantitative results should be supported with details on detection limits, extraction efficiency, and replicate variability.

Authors' Response: The authors now report limits of detection (0.07–2.65 μg/L), spike recoveries (94–105%), relative standard deviations (<12.1%), and estimated uncertainty (5.5–13.2%) across OS species in Lines 126–132 and Table S1. These uncertainty ranges are acceptable which have improved confidence in their quantification and establish that observed differences in OS levels across seasons and compounds are meaningful.

3. IEPOX-OS vs. MAE-OS: Seasonal Dominance and NOx Influence

Original Comment: Clarify how the observed OSs composition supports the interpretation of seasonal NOx regimes, particularly regarding IEPOX-OS (low-NO) and MAE-OS (high-NO) products.

Authors' Response: The authors included further analysis (Lines 304–307) comparing IEPOX-OS/MAE-OS mass ratios across seasons: 4.7 in summer, 1.53 in spring, and 0.49 in autumn. This ratio trend supports their claim that the HO_2-dominated (low-NO) pathway prevails in summer, while high-NO pathways become more important in spring and autumn. However, it is noted that no NOx measurement data were available to confirm this. The claim is reasonable but should be stated with some caution due to the absence of back-trajectory NOx data.

4. Principal Component Analysis (PCA)

Original Comment: Clarify how many samples and variables were included in the PCA and whether long filter sampling times limit temporal resolution.

Authors' Response: The authors specify that 26 samples and 18 chemical species were used (Lines 347–349). They also added a discussion of the temporal averaging and limitations of 10–24 h filter sampling (Lines 355–358), acknowledging that while diurnal variability (e.g., impacted by boundary layer height) cannot be resolved, the PCA captures seasonal and regional trends.

5. Use of abbreviation

Original Comment:, and explain abbreviations like IEPOX-OS, MAE-OS, HAS/GAS/LAS clearly in the main text or figure captions.

Authors' Response: The authors indicated they have revised the wordings for consistency and improved language. However, the author should use "OSs" instead of "organosulfates" consistently after first mention.

Minor Comment on Revised Text

- **Lines 61–67**: The statement that OSs are "weakly hygroscopic" and MSA is not should be taken with care. MSA is also known to be less hygroscopic than inorganic sulfate. It would be more accurate to state that both MSA and OSs exhibit moderate-to-low hygroscopicity, but with different physicochemical behaviors that may impact CCN activity.

Recommendation: Minor Revision/ Technical correction

The manuscript presents a valuable contribution to understanding marine secondary organic sulfur species and their interactions with climate-relevant processes. The revised version is well presented with good data quality. I recommend acceptance pending minor revision/technical correction, particularly in:

- Hygroscopicity comparison of MSA and OSs,
- Abbreviation consistency in figures and captions.