Author Responses (AR) to Referee Comments (RC) and resulting revisions of ms egusphere-2025-5334: "Isotopic apportionment of sulfate aerosols between natural and anthropogenic sources in the outflow of South Asia" by Sean Clarke, Henry Holmstrand, Krishnakant Budhavant, Manoj Remani, Sophie Haslett, Katerina Rodiouchkina, Ellen Kooijman, and Örjan Gustafsson

Reference: https://doi.org/10.5194/egusphere-2025-5334

All reviewer comments are included below in *black italic font* each followed by our detailed author responses, formatted as indented blue text.

> Corrected Table 1 to Table S1 in revised SI (revised SI line 103):
>
> *Table S1: Samples used to derive the anthropogenic IGP end-member*

Anonymous Referee #1: *Major comments:*

Reviewer 1:

Major comments:

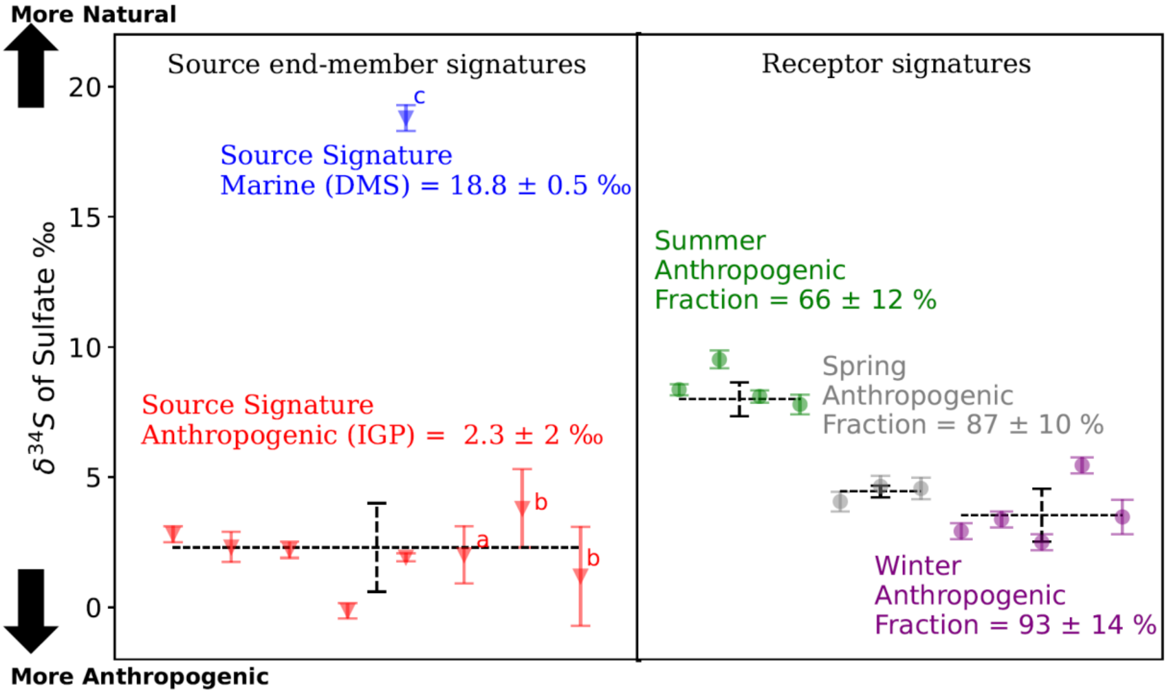
- *I assume that methanesulfonic acid (MSA) is not separated through the anion-retaining mesh method described in 2.4.1. Please describe how much you expect MSA contamination to alter measured d34S(SO4).*

> Author Reply:
>
> We thank the reviewer for highlighting this important aspect, however, we expect MSA contamination to have a negligible effect on measured $\delta^{34}S(SO_4^{2-})$. The isotopic composition of MSA has been reported as 17.4 ± 0.7‰ (Sanusi et al., 2006). In the Indian Ocean region, MSA concentrations are typically ~30–60 ng m^{-3} (Aswini et al., 2020), compared with sulfate concentrations on the order of ~1 - 16 µg m^{-3} during this study, implying an average MSA:SO_4 mass fraction of $\lesssim$1–6%. A simple two-endmember mass balance ($\Delta\delta \approx f_MSA(\delta_MSA - \delta_SO4)$ indicates that for typical f_MSA $\lesssim$1%, any shift would be ≤~0.1–0.2‰ even under conservative isotopic contrasts for the period.
>
> Lines added to revised SI ms (lines 43-49):
>
> Methanesulfonic acid (MSA) is not separated through the anion-retaining mesh and may in principle therefore influence measured $\delta^{34}S(SO_4^{2-})$. Methanesulfonic acid (MSA) has been reported to have $\delta^{34}S$ = 17.4 ± 0.7‰ (Sanusi et al., 2006). In the Indian Ocean region, MSA concentrations are typically ~30–60 ng m^{-3} (Aswini et al., 2020), which are small relative to sulfate (~1–16 µg m^{-3} reported in this study). Using

a simple mass-balance estimate, $\Delta\delta^{34}S(SO_4^{2-}) \approx f\_MSA(\delta\_MSA - \delta\_SO4)$, the expected influence of any MSA carryover is ≤~0.1–0.2‰ for typical f_MSA ≲1%, and therefore negligible relative to analytical uncertainty.

- *Figure 2 is confusing. I recommend splitting the source signatures and the d34S measurements into two subplots in figure 2.*

Author reply



We thank the reviewer for helping us make the graphic clearer and have now drafted the edited version above. We have also changed the values due to the erroneous DMS end member and rounding errors (see next reply).

- *Jongebloed et al. (2023) also compiled DMS-derived sulfurous compounds, including the measurements from Amrani et al. (2013) and other studies, to estimate $d34S(SO4)_{DMS}$ = 18.8 permille, which is slightly lower than the 19.7 used here. Justify why you use 19.7 – are you sure 18.8 is not a more accurate value?*

Author Reply:

We thank the reviewer and agree that we should use 18.8‰. In the original version, we effectively used the $\delta^{34}S$ value of DMSP(aq) (~19.7‰) reported by Amrani et al., 2013, rather than the more appropriate DMS-derived sulfate endmember. This choice did not fully account for the isotopic fractionations associated with conversion from DMSP to DMS (−~0.5‰) and subsequent sea–air transfer (DMS(aq) → DMS(g),

−~0.5‰). Accounting for these steps lowers the expected DMS-related endmember and is consistent with the $\delta^{34}S(SO_4^{2-})$(DMS) reported by Jongebloed et al., 2023. We therefore update the mixing model using 18.8‰.

This revision changes the inferred anthropogenic contribution and its associated error propagation. A rounding error was found in the anthropogenic contributions, and a sample was double-counted in the separated anthropogenic concentration

The values have now been updated in the revised manuscript (lines 23-24 and 268):

    - Summer: 67% ± 11 % → 66 ± 14 %
    - Spring: 88% ± 9 % → 87% ± 10 %
    - Winter: 94% ± 13 % → 93% ± 12 %
- These changes alter the r^2 correlation between anthropogenic SO_4 (δ34S) and fossil fuel BC (Δ14C) from 0.79 to 0.75 (lines 25 and 303 revised manuscript).
- The BC to anthropogenic SO_4 ratio changes to 0.083 (from 0.082) (line 325 revised manuscript).
- Fig. 2 and Fig. 3 were also corrected.

The anthropogenic sulfate concentration has now been corrected in the revised manuscript (lines 269-270):
These fractions translate into anthropogenic sulfate concentrations of 0.6 ± 0.2 μg m^{-3} (summer), 4.~~6~~8 ± 0.8 μg m^{-3} (spring), and 8.9 ~~9.1~~± 5 μg m^{-3} (samples with δ34S; winter).

- *(Harris et al., 2013and others have shown that d34S(SO4) is different from d34S(SO2) due to fractionation during oxidation of SO2. Why is it justifiable to create a mixing model here that ignores this fractionation? Is it because all your source signatures are determined based on sulfate that has already been oxidized? I think this is briefly mentioned in Text S1, but should be mentioned in 1-2 sentences in the main text.*

Author reply

We thank the reviewer and agree that additional lines should be added to the ms to clairfy the effect of $\delta^{34}S(SO_2) \rightarrow \delta^{34}S(SO_4^{2-})$. These effects are ideally captured by measuring at source aerosols but remain an uncertainty in estimations due to the effect of oxidative pathway.

The advantage of analysing receptor-integrated source region aerosols is that they already incorporate the isotope fractionation associated with SO_2 oxidation to sulfate for the anthropogenic endmembers. We agree that this point should be highlighted in the manuscript.

Added lines to revised manuscript (lines 223-224):

Analysing source aerosols captures isotope fractionation during SO_2 oxidation to sulfate, which can be substantial (≈3‰ enrichment in urban environments; Lee et al., 2023); see SI Text S1.

- *The way that the anthropogenic source signature was determined is not clearly explained. This is an important and uncertain aspect of your analysis, so it should be addressed more explicitly in the main text. I assume it is determined by just taking the d34S(SO4) measurements from IGP and BCOB based on this sentence: "Samples from BCOB, which were used to represent the anthropogenic end-member of sulfate, were assumed to be predominantly anthropogenic in origin (~100 %)"*

Author reply:

Yes, the reviewer is correct that this sentence should be clarified. The integrated continental anthropogenic signature was calculated from $\delta^{34}S(SO_4^{2-})$ measured on aerosol filters collected in the IGP (Delhi and BCOB), which we interpret as representative of anthropogenic sulfate in the region. We note, however, that if sulfate formation pathways and oxidation conditions differ between locations or periods, the associated isotope fractionation during SO_2-to-sulfate conversion may vary; consequently, the measured $\delta^{34}S(SO_4^{2-})$ could deviate from the instantaneous source $\delta^{34}S(SO_2)$ and from a simple isotopic equilibrium assumption. This also partially motivated sampling at BCOB, located at the outflow of the IGP (downwind of Delhi), where aerosol composition is expected to reflect a more integrated IGP-region signal. We did not observe clear systematic changes in $\delta^{34}S$ with more aged air masses within our dataset.

Hence, we have revised the sentence to clarify and is now found in the revised manuscript (lines 218-221)

The integrated continental anthropogenic signature was calculated directly from $\delta^{34}S(SO_4^{2-})$ measured on aerosol filters in the IGP (Delhi and BCOB), with samples measured in this study and from the literature, resulting in an end-member value of 2.3 ± 1.7 ‰ (see Fig. 2, Table S1; Dasari & Widory, 2024; Sawlani et al., 2019, n = 50).

- *Even in a highly polluted region, is it realistic that 100% of sulfate is anthropogenic? I wonder if this assumption results in an anthropogenic signature that is too high or too low. It would be good to test this assumption by seeing how your results would change if you assumed that the sulfate in BCOB is only 90% anthropogenic, for example.*

Author reply

We thank Reviewer #1 for highlighting this point. Although sulfate is unlikely to be entirely anthropogenic, it is expected to be close to 100% in this highly polluted IGP system. Our sensitivity test shows that relaxing this assumption does not change the results beyond the propagated uncertainty (shown below). Dasari & Widory, 2024 estimated contributions from biomass burning (2 ± 2%) and road dust (4 ± 3%) for the IGP/Delhi region. Biomass burning in this region is largely anthropogenically driven, but we acknowledge it represents a distinct source. The Dasari & Widory, 2024 estimates are based on PM_{10}, which have a higher dust fraction than our $PM_{2.5}$ measurements; therefore, the reported ~4% road-dust contribution can be considered an upper-bound estimate for our study. Dasari & Widory, 2024 also adopt a road-dust $\delta^{34}S$ value of 2 ± 1‰ from Sawlani et al. (2019). This value is more consistent with resuspended, pollution-derived sulfate than with purely mineral sulfate, which is typically higher (e.g., ~+7.4‰ for gypsum-derived sulfate; Olson et al., 2021). We will incorporate these clarifications in the Supporting Information and include a sensitivity test evaluating the effect of a 5–10% non-anthropogenic sulfate fraction within the BCOB anthropogenic end-member.

Added lines to the rev ms (225-228)

The IGP end-member in this study was assumed to be predominantly anthropogenic. Potential crustal and biogenic contributions are expected to be minor (Dasari & Widory, 2024). Our sensitivity analysis shows that including a 5–10% non-anthropogenic sulfate fraction does not change the source apportionment beyond the propagated uncertainty (see SI Text S3).

Addition to Supplementary information (SI Text S3 and SI Table 2)

Text S3: Sensitivity analysis of non-anthropogenic inputs to the continental end-member

The source-derived "anthropogenic" end-member assumes that sulfate collected at the receptor site represents predominantly anthropogenic sulfate. In practice, this end-member may also include some contributions of SO_4^{2-} from source categories not explicitly treated in the main mixing model (e.g., biomass burning or metallurgical emissions). For the IGP/Delhi region, biomass burning has been estimated at 2 ± 2% (Dasari & Widory, 2024). This partly motivated our use of the term 'anthropogenic end-member' rather than a 'fossil-fuel end-member'."

For non-anthropogenic sources Dasari & Widory, (2024) estimated a road-dust contribution of 4 ± 3% for the IGP/Delhi region (based on PM_{10}), which is likely an upper bound relative to our $PM_{2.5}$ dataset. The road-dust $\delta^{34}S$ value used in that study (2 ± 1‰; Sawlani et al., 2019) is consistent with resuspended, pollution-derived sulfate rather than purely mineral sulfate, which is typically higher (~+7.4‰ for gypsum-derived sulfate; Olson et al., 2021).

To test the sensitivity of our conclusions to potential non-anthropogenic sulfate in the anthropogenic end-member, we performed a dilution/sensitivity analysis in which 5% and 10% of the anthropogenic end-member are replaced by "other" sulfate spanning a wide range of $\delta^{34}S$ values (DMS-derived, volcanic, terrestrial biogenic, and mineral dust sulfate; Table S2). The most extreme case assigns the full 10% "other" fraction to DMS-derived sulfate ($\delta^{34}S = +18.8‰$), which is unlikely for aerosols originating from Delhi but provides an extreme upper bound on the potential effect. Across scenarios, the maximum change in inferred anthropogenic contribution is ~9 percentage points, comparable to our overall model uncertainty; therefore, our main conclusions are robust.

Table S2: Change in anthropogenic contribution when anthropogenic end-member consider non 100% anthropogenic

Non anthropogenic contribution	**100% Anthropogenic ;0% Other**		**95% Anthropogenic ;5% Other**		**90% Anthropogenic ;10% Other**	
	Season	**Percentage anthropogenic**	**Season**	**Percentage anthropogenic**	**Season**	**Percentage anthropogenic**
DMS (+18.8 ‰)[a]	Winter	94	Winter	89	Winter	85
	Spring	87	Spring	83	Spring	79
	Summer	65	Summer	61.7	Summer	59
Volcanic (+4.1 ‰)[b]	Winter	94	Winter	93	Winter	93
	Spring	87	Spring	87	Spring	86
	Summer	65	Summer	65	Summer	65
Terrestrial biogenic (~−5 ‰)[c]	Winter	94	Winter	96	Winter	99
	Spring	87	Spring	89	Spring	92
	Summer	65	Summer	67	Summer	69
Soil dust (+7.4 ‰)[d]	Winter	94	Winter	93	Winter	91
	Spring	87	Spring	86	Spring	84
	Summer	65	Summer	64	Summer	63

a = Amrani et al., 2013 ; b = Jongebloed, Schauer, Cole-Dai, et al., 2023; c = Kamyshny et al., 2014; d= Olson et al., 2021 . Note: Kamyshny et al. (2014) reports a broad range of possible values. For the purposes of this sensitivity test, −5 ‰ was selected as a rough approximation of the mean. The full range of isotopic compositions is large and lies outside the scope of this paper.

- *Another way to examine this would be to create a Keeling Plot (plot 1/nssSO4 vs. d34S(nssSO4)) of the data in Table S1. If anthropogenic sulfate is the more variable sulfate source, the y-intercept will represent the anthropogenic source signature. A Keeling Plot might not be appropriate if the other sources are too variable (see Pataki et al., 2003). I leave the decision of whether to evaluate a Keeling Plot to the authors of this study.*

Author reply

We thank reviewer #1 for this helpful suggestion. We evaluated a Keeling plot approach ($\delta^{34}S(nssSO_4^{2-})$ vs. $1/[nssSO_4^{2-}]$; Pataki et al., 2003). Using all samples yields an intercept of 2.50 ± 0.36‰ (1σ), which is consistent with our derived anthropogenic end-member. However, the result is sensitive to season: excluding the summer samples produces a more depleted intercept of 1.2 ± 0.64‰. This sensitivity suggests that variability in non-anthropogenic sources and mixed source contributions may violate the assumptions of the Keeling approach for these aerosol samples. We therefore do not adopt the Keeling-plot-derived end-member, but we report the analysis in the SI for completeness.

Lines added to the revised manuscript (lines 228-232):

We also investigated the use of a Keeling plot to determine the anthropogenic end-member, but did not apply this approach because mixed and variable sources could bias the inferred end-member toward depleted values (see SI Text 3 and SI Fig. 3). ~~Anthropogenic sulfate~~

~~also includes minor contributions from crustal and biomass burning sources, which have not been separated in this study (Dasari and Widory, 2024).~~

Additional section added to the revised SI mansuscript (lines 50-55):

Text S2: Keeling-plot approach for determining the anthropogenic end-member:

A Keeling plot analysis (1/nssSO_4^{2-} vs. $\delta^{34}S$(nssSO_4^{2-}), Fig. S1) gives an intercept of 2.50 ± 0.36‰ (1σ), similar to our anthropogenic endmember (Fig. 1, Table. S2). However, because the intercept is sensitive to sample selection (e.g., excluding summer samples yields 1.2 ± 0.64‰), we do not use the Keeling approach here due to concerns that mixed and variable sources violate its assumptions for aerosol sulfate (Pataki et al., 2003, Jongebloed et al., 2023).

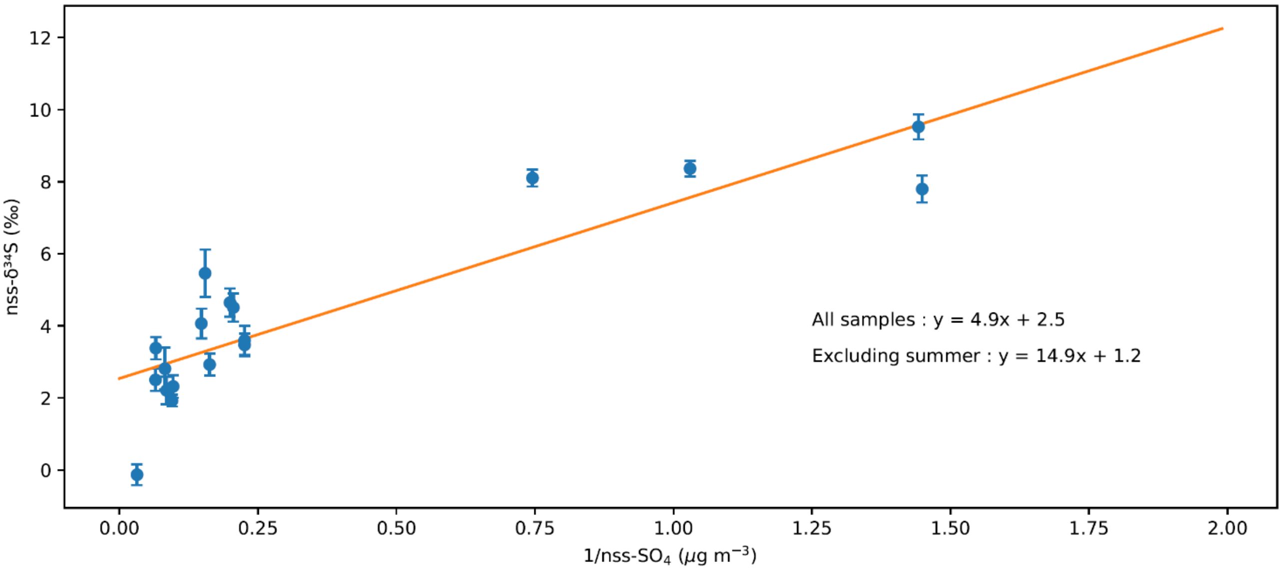


Fig. S1: Keeling plot which shows the (1/nssSO_4^{2-} vs. $\delta^{34}S$(nssSO_4^{2-}). The y-intercept represents the expected anthropogenic end-member. Blue points denote the nss- $\delta^{34}S$ from this study, and the orange line denotes the linear best fit.

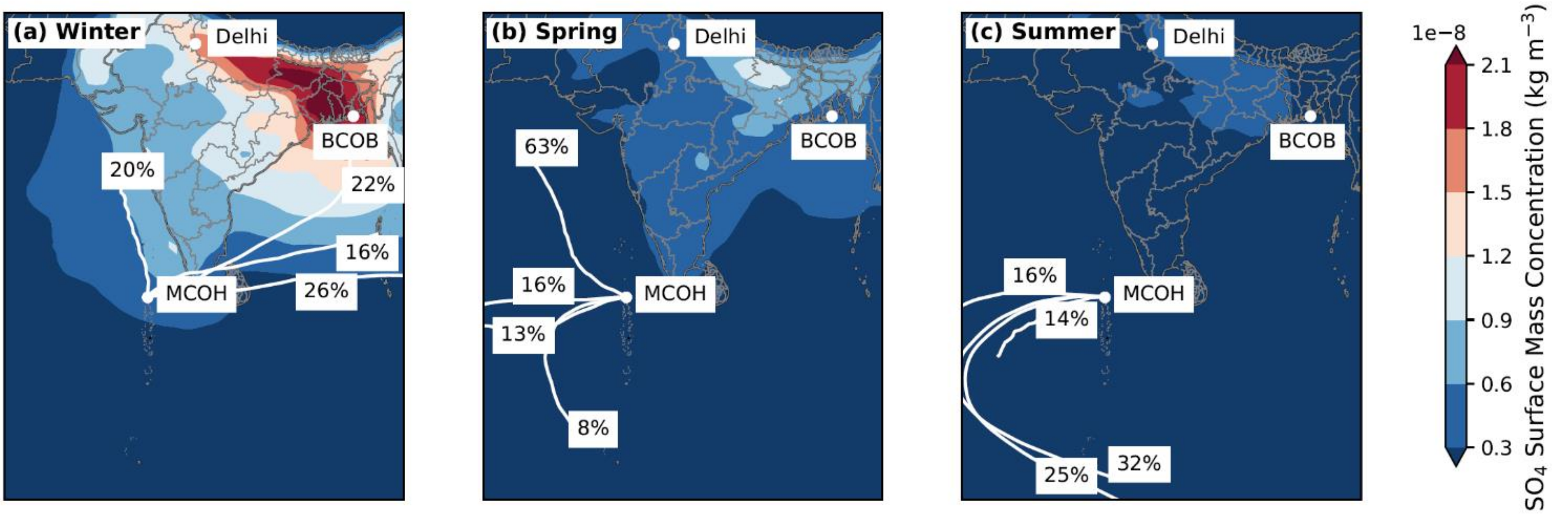
Additionally, with the addition of the new Fig. S1, the original Figs. S1–S3 have been renumbered as Figs. S2–S4.

Minor comments:

- *Figure 1: the labels of the seasons (winter, autumn, summer) don't align with the caption (winter, spring, summer), unless if I'm missing something.*

Author reply

Thank you for catching this error. It should read spring and is now fixed.



- *This sentence needs a citation at the end: The ship end-member had a δ34S of 3 ± 3 ‰ taken from studies that measured δ34-SO4 and δ34-SO2 in the North Atlantic, which is thought to be representative of remote air masses with strong ship emissions from heavy fuel oil. Perhaps (Seguin et al., 2010, 2011; Wadleigh, 2004)?*

Author reply

Agreed the sentence will be updated to have the references as stated in the first section of results.

References (Seguin et al., 2010, 2011; Wadleigh, 2004) added to revised manuscript (line 177)

Anonymous Referee #2: *Major comments:*

Lines 170–176 and Section 3.1: The discussion regarding the choice of δ^{34}S end-member values for both biogenic and anthropogenic sources should be expanded, as these choices directly affect the subsequent source apportionment.

- *Biogenic sources: Several previous studies have reported δ^{34}S values for biogenic sulfur sources ranging from approximately 17.4 to 19.7 ‰ (e.g., (Jongebloed et al., 2023; Seguin et al., 2011, and references therein). The use of lower δ^{34}S values could potentially reduce the inferred contribution of anthropogenic sources in the present analysis. Please provide clearer justification for the selected biogenic end-member values and discuss the potential sensitivity of the results to this choice.*

  Author reply

  We agree with Reviewer #2 that this discussion should be expanded and clarified. We have reassessed the use of 19.7‰ for the marine-biogenic end-member. The value used in the submitted manuscript corresponds to DMSP in aqueous solution

(≈19.7‰), whereas our end-member would more accurately represent DMS in air. During conversion and transfer there is fractionation, including from DMSP → DMS (≈ −0.5‰) and during sea–air transfer (DMS(aq) → DMS(g); ≈ −0.5‰). Therefore, the value reported by Jongebloed et al. (2023) is more appropriate, as it reflects atmospheric DMS. We will revise the manuscript to use 18.8‰ as the marine-biogenic (DMS) end-member

The changes made are shown in Anonymous Referee #1 major comment as they rightly brought up the same issue.

- *Anthropogenic sources: A ship emission end-member of 3 ± 3 ‰ is introduced; however, in the final source apportionment it appears that a value of 2.3 ± 1.7 ‰ was used as the anthropogenic end-member without explicitly accounting for the ship contribution. Please clarify how this value was derived and explain how ship emissions were considered (or excluded) in determining the anthropogenic end-member.*

  Author reply

  We thank Reviewer #2 for this comment and agree that our explanation was not sufficiently clear. The continental anthropogenic end-member and the ship-emission end-member are treated as distinct end-members and are not mixed in our analysis. Because these end-members are relatively similar, the source-apportionment results are only weakly sensitive to this choice. Switching between them changes the estimated anthropogenic contribution by at most ~3%.

  We separated these two end-members as we stated that the appropriate anthropogenic signature can vary spatially and seasonally. During the monsoon, air masses reaching MCOH frequently originate over the open ocean, for which a ship-emissions end-member may be more appropriate than a continental anthropogenic end-member derived from the IGP. We acknowledge that ship emissions may also influence MCOH during winter and spring; however, both top-down (MERRA-2) and bottom-up (CEDS) constraints indicate that ship emissions are minor during these periods compared to the dominant continental pollution outflow (Buchard et al., 2017; Randles et al., 2017; Hoesly et al., 2018).

  Lines added to the revised manuscript (lines 213-217):

  The oceanic anthropogenic and continental anthropogenic end-members were treated as distinct to reflect their different origins. Ship emissions may contribute to the continental end-member, but available top-down (MERRA-2) and bottom-up (CEDS) constraints indicate that these contributions are much smaller than land-based continental emissions (Buchard et al., 2017; Hoesly et al., 2018; Randles et al., 2017). In any case, the choice of end-member changes the inferred anthropogenic contribution by at most ~3%.

- *Lines 296–311: The discrepancy between the observed BC/SO*$_4$ *ratios (0.075 ± 0.03) and the inventory-based BC/SO*$_2$ *ratio (0.097) warrants further discussion, particularly because a stated goal of this study is to provide guidance for future mitigation strategies in South Asia.*

- *How might atmospheric processes following emission (such as SO_2 oxidation to sulfate, differential deposition of BC and sulfate, or other removal mechanisms) affect the BC/SO_4 ratio? After accounting for these processes, is the difference between observed and inventory-based ratios still significant? In addition, could emissions of H_2S from mangrove ecosystems influence the observed ratios? If possible, the authors are encouraged to provide suggestions on how emission inventories for this region could be improved based on these findings.*
- *Finally, as noted above, uncertainties associated with the choice of $\delta^{34}S$ end-member values may further propagate into the estimated BC/SO_4 ratios and should be acknowledged.*

Author reply

We agree with reviewer #2 that these issues are important and would be relevant for mitigation guidance. However, a quantitative treatment of that topic is beyond the scope of this paper and would require a chemical transport model. This includes post-emission atmospheric processing (oxidation of SO_2 to sulfate and differential deposition/removal of BC and sulfate) as well as potential reduced-sulfur inputs (e.g., mangrove H_2S).

This comment however prompted us to compare BC to anthropogenic sulfate rather than to total nssSO_4^{2-}, since BC is predominantly anthropogenic. This yields BC/SO_4^{2-}_anth = 0.08~~2~~3 ± 0.03 (spring and winter), which is closer to the inventory-based BC/SO_2 ratio (0.097). We note that the quoted ±0.03 reflects measurement variability only and does not include additional uncertainty propagated from the $\delta^{34}S$ end-member constraints.

Evaluating why these ratios differ quantitatively remains challenging. As the reviewer notes, differential transport and removal of BC and sulfate, as well as additional terrestrial sulfur inputs, could play a role. Black carbon at MCOH has been reported to have a longer atmospheric lifetime than more hygroscopic species (e.g.,Budhavant et al., 2020). Preferential removal of sulfate during transport would therefore be expected to increase BC/SO_4^{2-} in situ relative to the emission-inventory ratio. In contrast, unaccounted biogenic sulfur inputs from mangroves (e.g., reduced-sulfur emissions that oxidize to sulfate) would add sulfate without co-emitted BC and thus decrease BC/SO_4^{2-}.

Lines added to the revised manuscript (324-329):

~~, with MERRA-2 being higher.~~ When we compare to the in situ anthropogenic sulfate fraction (BC/anthro-SO_4^{2-}), the agreement improves (0.083 ± 0.03; spring and winter). The quoted uncertainty reflects measurement variability only and does not include additional uncertainty propagated from the $\delta^{34}S$ end-member constraints. Notably, the inventory-based ratio is closer to our in-situ constraint than the MERRA-2 estimate. The emission-inventory ratio does not include biogenic sulfur inputs, which could lower BC/SO_4^{2-}, nor does it account for preferential removal of hygroscopic sulfate during transport, which could act in the opposite direction by increasing BC/SO_4^{2-} (Budhavant et al., 2020).

Specific comments:

- Line 113: Please provide information on the IC column used for ion quantification.'

  Author Reply

  The analytical details on analytical columns have been added to the revised manuscript (113-114):

  Ionic concentrations (Cl^-, Br^-, NO_2^-, NO_3^-, PO_4^{3-}, SO_4^{2-}, Na^+, NH_4^+, K^+, Mg^{2+}, Ca^{2+}) were measured using a Dionex Aquion ion chromatography (IC) system (Thermo Finnigan LLC, Dionex IonPAc CS12A) anion (Dionex IonPac AS22 fast).

Line 118: Please explicitly state whether the sulfate-to-sodium ratio refers to a mass ratio or a molar ratio.

Author Reply:

We thank reviewer #2 and agree that this should be stated. The ratio is the mass ratio. This is now explicitly stated in the revised manuscript (line 119):

The nss-SO42- is the non-sea salt sulfate, with the mass ratio of sulfate to sodium from sea water being 0.253 taken from (Keene et al., 1986).

- Lines 130–132: Please specify the chemical forms of the solutions used for adding Si and Na. For example, were these added in acidic (e.g., H_3SiO_4) and alkaline (e.g., NaOH) forms, respectively? Although readers can consult Rodiouchkina, (2018) for methodological details such as solution concentrations, the simplified procedure presented here is difficult to follow from an analytical chemistry perspective without this clarification.

  Author Reply

  We agree that these details should be included and the ms will be revised accordingly.

  Lines added to the revised manuscript (lines 131-134):

  Silicon (Si) was added to all solutions as ammonium hexafluorosilicat $(NH_4)_2SiF_6$), at a concentration ratio of 1:1 (S:Si, µg mL-1:µg mL-1) for internal standardization. Additionally, sodium (Na) was added as sodium carbonate (Na2CO3) at a molar ratio of 2 (Na/S) to all measurement solutions.

Line 159: Replacing "%nssSO_4" with "F_nssSO_4" would improve readability and maintain consistency with other equations in the manuscript (e.g., F_biomass in Eq. (3) and F_DMS-SO_4 in Eq. (5)).

Author Reply :

Reviewer #2 is correct.

Equation 4 in revised manuscript (line 160) now reads:

$$\delta^{34}S_{nss} = (\delta^{34}S - \left(\frac{100 - F\_nssSO_4}{100}\right) \times \delta^{34}S_{sea\ water}) \div \frac{\%nssSO_4}{100}$$

References

Amrani, A., Said-Ahmad, W., Shaked, Y., & Kiene, R. P. (2013). Sulfur isotope homogeneity of oceanic DMSP and DMS. *Proceedings of the National Academy of Sciences of the United States of America*, *110*(46). https://doi.org/10.1073/pnas.1312956110

Aswini, A. R., Hegde, P., Aryasree, S., Girach, I. A., & Nair, P. R. (2020). Continental outflow of anthropogenic aerosols over Arabian Sea and Indian Ocean during wintertime: ICARB-2018 campaign. *Science of the Total Environment*, *712*. https://doi.org/10.1016/j.scitotenv.2019.135214

Buchard, V., Randles, C. A., da Silva, A. M., Darmenov, A., Colarco, P. R., Govindaraju, R., Ferrare, R., Hair, J., Beyersdorf, A. J., Ziemba, L. D., & Yu, H. (2017). The MERRA-2 aerosol reanalysis, 1980 onward. Part II: Evaluation and case studies. *Journal of Climate*, *30*(17). https://doi.org/10.1175/JCLI-D-16-0613.1

Budhavant, K., Andersson, A., Holmstrand, H., Bikkina, P., Bikkina, S., Satheesh, S. K., & Gustafsson, Ö. (2020). Enhanced Light-Absorption of Black Carbon in Rainwater Compared With Aerosols Over the Northern Indian Ocean. *Journal of Geophysical Research: Atmospheres*, *125*(2). https://doi.org/10.1029/2019JD031246

Dasari, S., & Widory, D. (2024). Retrospective Isotopic Analysis of Summertime Urban Atmospheric Sulfate in South Asia Using Improved Source Constraints. *ACS ES&T Air*, *1*(5), 357–364. https://doi.org/10.1021/acsestair.3c00060

Hoesly, R. M., Smith, S. J., Feng, L., Klimont, Z., Janssens-Maenhout, G., Pitkanen, T., Seibert, J. J., Vu, L., Andres, R. J., Bolt, R. M., Bond, T. C., Dawidowski, L., Kholod, N., Kurokawa, J. I., Li, M., Liu, L., Lu, Z., Moura, M. C. P., O'Rourke, P. R., & Zhang, Q. (2018). Historical (1750-2014) anthropogenic emissions of reactive gases and aerosols from the Community Emissions Data System (CEDS). *Geoscientific Model Development*, *11*(1), 369–408. https://doi.org/10.5194/gmd-11-369-2018

Jongebloed, U. A., Schauer, A. J., Cole-Dai, J., Larrick, C. G., Wood, R., Fischer, T. P., Carn, S. A., Salimi, S., Edouard, S. R., Zhai, S., Geng, L., & Alexander, B. (2023). Underestimated Passive Volcanic Sulfur Degassing Implies Overestimated Anthropogenic Aerosol Forcing. *Geophysical Research Letters*, *50*(1). https://doi.org/10.1029/2022GL102061

Jongebloed, U. A., Schauer, A. J., Hattori, S., Cole-Dai, J., Larrick, C. G., Salimi, S., Edouard, S. R., Geng, L., & Alexander, B. (2023). Sulfur isotopes quantify the impact of anthropogenic activities on industrial-era Arctic sulfate in a Greenland ice core. *Environmental Research Letters*, *18*(7). https://doi.org/10.1088/1748-9326/acdc3d

Kamyshny, A., Druschel, G., Mansaray, Z. F., & Farquhar, J. (2014). Multiple sulfur isotopes fractionations associated with abiotic sulfur transformations in Yellowstone National Park geothermal springs. *Geochemical Transactions*, *15*(1). https://doi.org/10.1186/1467-4866-15-7

Keene, W. C., Pszenny, A. A. P., Galloway, J. N., & Hawley, M. E. (1986). Sea-salt corrections and interpretation of constituent ratios in marine precipitation. *Journal of Geophysical Research: Atmospheres*, *91*(D6), 6647–6658. https://doi.org/10.1029/jd091id06p06647

Lee, G., Ahn, J., Park, S. M., Moon, J., Park, R., Sim, M. S., Choi, H., Park, J., & Ahn, J. Y. (2023). Sulfur isotope-based source apportionment and control mechanisms of PM2.5 sulfate in Seoul, South Korea during winter and early spring (2017–2020). *Science of the Total Environment*, *905*. https://doi.org/10.1016/j.scitotenv.2023.167112

Olson, E., Michalski, G., Welp, L., Larrea Valdivia, A. E., Reyes Larico, J., Salcedo Peña, J., Fang, H., Magara Gomez, K., & Li, J. (2021). Mineral dust and fossil fuel combustion dominate sources of aerosol sulfate in urban Peru identified by sulfur stable isotopes and water-soluble ions. *Atmospheric Environment*, *260*. https://doi.org/10.1016/j.atmosenv.2021.118482

Pataki, D. E., Ehleringer, J. R., Flanagan, L. B., Yakir, D., Bowling, D. R., Still, C. J., Buchmann, N., Kaplan, J. O., & Berry, J. A. (2003). The application and interpretation of Keeling plots in terrestrial carbon cycle research. *Global Biogeochemical Cycles*, *17*(1). https://doi.org/10.1029/2001GB001850

Randles, C. A., da Silva, A. M., Buchard, V., Colarco, P. R., Darmenov, A., Govindaraju, R., Smirnov, A., Holben, B., Ferrare, R., Hair, J., Shinozuka, Y., & Flynn, C. J. (2017). The MERRA-2 aerosol reanalysis, 1980 onward. Part I: System description and data assimilation evaluation. *Journal of Climate*, *30*(17). https://doi.org/10.1175/JCLI-D-16-0609.1

Rodiouchkina, K. (2018). *Development of a multi-collector inductively coupled plasma massspectrometry method for measurement of stable sulphur isotope ratios in aerosol sulphate (Dissertation)*.

Sanusi, A. A., Norman, A. L., Burridge, C., Wadleigh, M., & Tang, W. W. (2006). Determination of the S isotope composition of methanesulfonic acid. *Analytical Chemistry*, *78*(14). https://doi.org/10.1021/ac0600048

Sawlani, R., Agnihotri, R., Sharma, C., Patra, P. K., Dimri, A. P., Ram, K., & Verma, R. L. (2019). The severe Delhi SMOG of 2016: A case of delayed crop residue burning, coincident firecracker emissions, and atypical meteorology. *Atmospheric Pollution Research*, *10*(3). https://doi.org/10.1016/j.apr.2018.12.015

Seguin, A. M., Norman, A. L., Eaton, S., & Wadleigh, M. (2011). Seasonality in size segregated biogenic, anthropogenic and sea salt sulfate aerosols over the North Atlantic. *Atmospheric Environment*, *45*(38). https://doi.org/10.1016/j.atmosenv.2011.09.033