Response to Reviewer 1

Overall, I endorse this revised version. The manuscript is well-written with thorough data analysis. I have only one minor comment: In lines 369-370 regarding the description of GAS, I suggest supplementing the text with recent literature indicating that the gas-phase pathway also makes an important contribution to GAS formation (Sun et al., 2025).

We thank the reviewer for this endorsement. We have updated the manuscript as suggested.

Changes to the manuscript

Lines 399–402: Glycolic acid sulfate (GAS) was observed during this campaign. This compound has traditionally been considered a product of aqueous photochemistry after glyoxal has been taken up by sulfate-containing aerosol (Galloway et al., 2009), although recent research suggests that a gas-phase pathway, involving the reaction between the SO_3 radical and glycolic acid, can also contribute to GAS formation (Sun et al., 2025).

Reference

Sun, H., Liu, Y., Nie, W., Li, Y., Ge, D., Xu, T., Yin, J., Liu, C., Fu, Z., Qi, X., Liu, T., Zha, Q., Yan, C., Wang, Z., Chi, X., and Ding, A.: Unexpected Gas-Phase Formation of Glycolic Acid Sulfate in the Atmosphere, Environ. Sci. Technol., 59, 16556–16566, https://doi.org/10.1021/acs.est.5c07888, 2025.

Response to Reviewer 2

The authors have carefully addressed my comments and added some analysis, including PMF analysis of the AMS data, CWT analysis, and a more detailed discussion of the organic compounds. These changes improve the manuscript. However, some main conclusions are still stated more strongly than can be supported by the current measurements. Therefore, I suggest that it could be considered for publication before the following comments was addressed.

We thank the reviewer for these comments, and have now made the requested changes, as outlined below.

Major comments:

1. For the AMS PMF analysis, the authors should provide more information on the PMF evaluation, including the Q/Qexp, residuals, solution stability across different seeds and rotations, and the similarity of the identified factor spectra to reference HOA, BBOA, SV-OOA, and LV-OOA spectra. The reason for combining the two LV-OOA-like factors should also be demonstrated more clearly. Without these checks, it is difficult to assess whether the three reported factors represent stable atmospheric components or a particular mathematical solution.

Some of the interpretations of these factors also appear speculative. For example, the slight nighttime increase in SV-OOA is used to suggest local formation, and the higher contribution in northerly air masses is linked to VOC emissions from nearby islands or the ocean surface. These possibilities may be reasonable, but the current analysis does not distinguish them from changes in transport, boundary-layer dynamics, or partitioning. These interpretations should either be supported by additional evidence or presented more cautiously.

We agree with the reviewer that more details of the PMF analysis should have been included, and we have now added: (1) a more thorough description of the PMF runs to the method section in the main text, including mention of runs using different seeds and rotations, (2) a figure showing the main PMF result in the results section, and (3) figures and tables showing further details of the PMF results in the supplement. We have also now compared our observed spectra with findings presented by Ng et al. (2010), who used PMF factor solutions from a large number of different sites, in order to identify specific characteristics.

Furthermore, we agree with the reviewer's comment that the interpretation of the SV-OOA factor was largely speculative. In fact, based on the comparison with representative spectra that we have now carried out, this factor much more closely resembles a further LV-OOA factor than it does SV-OOA, making SV-OOA a likely mis-attribution. A correlation analysis between the three more oxidised factors showed that they were extremely chemically similar, with r-values of 0.975 (factor 1, factor 2); 0.997 (factor 1, factor 3) and 0.972 (factor 2, factor 3).

In an effort to avoid over-interpretion, we have now removed the SV-OOA label, and merged the three LV-OOA factors, given that there was very little to distinguish between them chemically. We have adjusted the text accordingly. Nevertheless, we recognise that this merging of factors was a judgement call, and have therefore included the full four-factor solution, and individual concentration-weighted back trajectory images for both merged and separate factors, in the supplement.

The solution presented now is not identical to our original, as, when revisiting this, we identified some peaks that were included as organic compounds, but in fact, on closer inspection, were highly influenced by the tail of large sulfate peaks and therefore did not reflect genuine organic signal. These should not have been included in the original peak list and have now been removed. The influence that this has had on the retrieved factors is minimal and does not alter our interpretation.

Changes to the manuscript

- We have shifted the description of PMF to the method section, and expanded it to include a more thorough description of retrieved parameters, and more consideration of reference spectra.

Lines 198–223: Positive matrix factorisation (PMF; Ulbrich et al., 2009) was applied to the high-resolution AMS organic mass spectra to investigate the sources and degree of processing of organic aerosol observed at MCOH. This approach is commonly employed with AMS data to break down the observed mass spectrum into non-negative factor contributions representing different sources or chemical signatures, and has been described in detail elsewhere (Crippa et al., 2013). This analysis was carried out using the PMF Evaluation Tool (PET) v3.08E (Aerodyne). The error matrix was generated by the AMS analysis software, with standard signal-to-noise (SNR)-based down-weighting of weak ions (SNR < 2) and removal of bad ions (SNR < 0.2). Peaks whose signals were strongly dominated by the tails of adjacent sulfate peaks removed from the matrix.

The value of Q/Q_{exp} was found to decrease gradually from 0.429 for one factor to 0.372 for five, with no clear inflection point (Fig. S13a). Values below 1 indicate that uncertainties are conservative. A four-factor solution was required, here, before a clear hydrocarbon-like organic aerosol (HOA) signal emerged, while increasing the number of factors beyond this further divided the more oxidised factors. For this reason, the four-factor solution was chosen. The residuals for this solution were normally distributed around zero (Fig. S13d,e). Thirty seed runs were carried out for the four-factor solution, all of which converged to broadly the same solution, demonstrated by Q/Q_{exp} ranging only from 0.3810030 to 0.3810044 (Fig. S13b). Sensitivity to rotation was assessed by carrying runs with

fPeak values varying between -1 and 1. The PET software was unable to produce solutions for some values (successful runs were fPeaks = -0.3, -0.2, 0, 0.4, 0.6, 0.8, 1). The value of Q/Q_{exp} was lowest at fPeak = 0 and rose by less than 2% across the range of successful fPeak runs (Fig. S13c).

The chosen four-factor solution consisted of one HOA/BBOA factor – characterised by the ion series $C_nH^+_{2n1}$ and $C_nH^+_{2n-1}$ (Ng et al., 2010), as well as a peak at *m/z* 60, which is indicative of biomass burning (Alfarra et al., 2007; Schneider et al., 2006) – and three factors that were highly oxidised, dominated by *m/z* 28 and 44. In all three of these cases, the fractional contribution of *m/z* 44 to the total organic spectrum (f_{44}) was comfortably within typical parameters to be interpreted as low-volatility oxidised organic aerosol (LV-OOA) (Ng et al., 2010), with f_{44} values ranging from 0.19 to 0.29. The mass spectra for these three more oxidised factors were very similar (r = 0.97 to 0.997, or 0.85 to 0.94 when *m/z* 28 and 44 were excluded from the correlation analysis). There was no distinct signature in the mass spectra to separate these three factors, and there was no distinct relationships with air mass back trajectory. For this reason, we have presented these, here, as a single LV-OOA factor. However, we recognise that this is a subjective decision, and have therefore included the unmerged solution in Fig. S12.

- We have removed the speculative discussion about SV-OOA from the results, and included a broader discussion about what these results demonstrate.

Lines 487–514: The chosen PMF solution for the campaign is displayed in Fig. 7, with the oxygenated factors 1–3 merged. The HOA/BBOA factor accounted for approximately 7% of the total organic aerosol mass. It was characterised by large peaks at *m/z* 55 (C_4H_7) and *m/z* 57 (C_4H_9), both of which are typically associated with traffic exhaust (Canagaratna et al., 2004), and a small signal at *m/z* 60 ($C_2H_4O_2$), which is typically identified as a fragment of levoglucosan or other anhydrous sugars, and considered a tracer for fresh biomass burning (Alfarra et al., 2007; Schneider et al., 2006). It shows a slight decrease in concentration during the day, which can be attributed to a combination of variability in sources – with biomass burning for cooking, for example, being more likely to occur in the evenings – and changes in boundary layer height allowing more dilution during daylight hours. Concentration-weighted back trajectory analysis indicated that there was no clear preference in source region for this factor (Fig. S14).

The merged LV-OOA factor accounted for approximately 93% of organic aerosol mass. Its spectrum was dominated by *m/z* 28 and 44 (with f_{44} = 0.25 and f_{43} = 0.039), placing it solidly in the LV-OOA space (Ng et al., 2010). In this merged factor, CHO compounds made up 72% of the organic aerosol mass. There was variation in its concentration over the period of the campaign, but it did not show a diel pattern, which is consistent with the expected behaviour of long-range transported aerosol. A concentration-weighted back trajectory analysis showed no strong preferred source direction for LV-OOA, although there are slight differences between the three merged factors.

Organic markers that have previously been identified with marine organic aerosol, such as CH_3SO_2 (*m/z* 79) (Crippa et al., 2013), were not observed in high concentrations in any of the identified factors, which suggests that the influence of secondary aerosol formation from marine sources such as methanesulfonic acid (MSA) is not a dominant source of aerosol in this region. This finding is consistent with the low concentrations of dimethyl sulfide (DMS) observed at MCOH.

Together, these results indicate that the vast majority of organic aerosol at MCOH during the winter represents a single, highly-oxidised component. A small HOA/BBOA factor is also present, which may reflect local emissions, but it is extremely small in comparison with the mass of aged aerosol. This supports that conclusion that organic aerosol reaching MCOH is highly processed during long-range

transport. While this finding is consistent with previous observations at the site, previous analyses were not able to analyse the organic composition at high time-resolution, and thus it has not previously been possible to establish whether the aerosol mass has distinct chemical characteristics associated with specific air mass origins. Our results indicate that, over the course of this campaign, the chemical composition of organic aerosol was not strongly dependent on transport pathways. Organic composition at MCOH cannot, therefore, be used to identify individual source regions. Rather, the clearest chemical signature that can be used to distinguish between air masses is the sulfate-to-organic ratio.

- We have included the full details of the PMF solution, including seeds and rotations, and the full four-factor solution before the merging of the LV-OOA factors, in the supplement (Figs. S12 and S13).

2. The manuscript now shows that sulfate concentration is higher in air masses associated with the IGP, whereas organic aerosol has a less distinct geographical pattern. However, the CWT analysis shown in Fig. 3 are based on sulfate concentration, while the text subsequently suggests that the sulfate fraction may be used as an indicator of IGP influence. An analysis based on sulfate fraction or the sulfate-to-organic ratio would be more appropriate.

The trajectory methods also need clarification. Section 2.2 states that ten-day trajectories were calculated at 00:00, 06:00, 12:00, and 18:00 UTC, whereas the Results section and Fig. S1 refer to hourly generated trajectories. The manuscript should state consistently how many trajectories were used for the cluster and concentration-weighted analyses.

We have now updated Fig. 3 in the main text to show (a) absolute sulfate concentration, and (b) sulfate/organic aerosol ratio, to support this point. The figure showing absolute organic aerosol concentrations has been moved to the supplement (Fig. S5).

We thank the reviewer for noting the inconsistency with the back trajectory timing. The illustrative trajectories shown in Fig. 1c were in fact generated hourly. However, for the concentration-weighted analysis, trajectories were 3-hourly. The text in the methods section has now been updated.

Changes to the manuscript

Lines 115–6: Ten-day backward trajectories were computed hourly for the sampling location, allowing for an assessment of both regional and long-range air-mass origins.

And, in the section about the concentration-weighted trajectories:

Lines 125–6: For this analysis, three-hourly back trajectories were used over the course of the full campaign.

3. The author acknowledges that photochemical ageing, cloud processing, and heterogeneous sulfate chemistry are inferred rather than directly observed. However, several strong statements remain in the manuscript. For example, Section 3.5 states that "the importance of aqueous processing is also evident," Section 3.4 states that the results "unambiguously highlight" sulfate chemistry, and the Conclusions state that photochemical and aqueous processing during transport is "the most important factor governing" aerosol composition.

The abundance of oxalic acid and other dicarboxylic acids is consistent with aqueous processing, but it does not uniquely demonstrate that this process occurred during the sampled transport period. The particle size distribution reported in an earlier study at the same site is also not a direct constraint on cloud processing during this campaign. Unless the authors can provide a more direct

link to cloud exposure, rainfall history, aerosol liquid water, or relevant meteorological conditions, these statements should be revised throughout the Abstract, Results, and Conclusions.

We agree with the reviewer that, while our results here are consistent with this interpretation, they do not uniquely demonstrate it. On the reviewer's advice, we have revised the abstract, results and conclusions to soften this language.

Changes to the manuscript

- In the abstract:

Lines 31–3: Our results show the strong influence of long-range transport on aerosol composition over the Indian Ocean, and are consistent with extensive atmospheric processing, including possible heterogeneous sulfate chemistry.

- In section 3.4:

Lines 429–33: Taken together, these observations are consistent with the influence of both acid-catalysed epoxide reactions and aqueous phase chemistry in driving the formation of SCOs in this region. It is likely that species identified in the CIMS dataset represent a combination of genuine parent molecules present in ambient air, and of thermal decomposition products of larger parent species or products of further chemical reactions during desorption. However, these results suggest that sulfate chemistry, and interactions between biogenic/anthropogenic VOCs and sulfate aerosol, may play a role in the aerosol plume.

- In the conclusion:

Lines 602–3: Results presented here are consistent with extensive photochemical processing, and possibly aqueous processing, during transport as an important control on the chemical composition of aerosol over the Indian Ocean during the winter.

We have also re-worded several parts of the conclusion to ensure that we are not over-stating the interpretation of findings reported here.

4. Table 3 gives the percentage contribution of individual molecular formulae to the detected CHO and CHON signal. This is useful descriptive information, but it should not be us as a mass fraction without compound-specific sensitivity calibrations. The manuscript currently refers to "22% of the signal by mass," while the response describes these values as contributions to the total organic aerosol signal. Table 3 instead defines them as fractions of the CHO+CHON signal. These descriptions should be made consistent.

We thank the reviewer for noting this. The table represents percentage of the total CHO + CHON signal. The text has now been updated to reflect this.

Changes to the manuscript

Line 447: Of the total peak-fitted CHO + CHON compounds, 22% of the signal was made up of dicarboxylic acids

5. The authors now acknowledge that C4H9O4S may be generated by thermal decomposition or by reactions occurring during FIGAERO desorption. However, the same signal is then described as a "genuine tracer" for sulfate chemistry in an anthropogenic plume from the IGP. I suggested that the author could analyze the thermogram of C4H9O4S to investigate whether it was decomposition product.

We agree that the term “genuine tracer” was a poor choice of wording. The intention was to note that this product reliably appears in air masses that originate from the IGP. This, combined with the thermogram shape that matches that of sulfuric acid perfectly, suggest the importance of sulfur chemistry in its formation.

We have now removed the use of “genuine tracer” from the text, and included, in the section analysing thermograms, that this compound’s thermogram shape perfectly matches that of sulfuric acid and a few other SCO compounds. This supports the idea of this compound being a thermal decomposition product, or formation product of reactions taking place on the filter.

Changes to the manuscript

Lines 411–2: The inclusion of $C_4H_9O_4S^-$ in this group supports the interpretation of this compound as a possible product of thermal decomposition of, or reactions involving, sulfate and organic material during desorption.

Specific comments:

1. Section “Source apportionment of organic aerosol using Positive Matrix Factorisation” and “Transport of continental emissions and comparison with CIMS measurements from Delhi” have a same section number.

2. In Section 2.4, the calibration curves are referred to as Fig. S1, but they are shown in Fig. S2.

3. Line 312: The average CIMS mass spectrum is referred to as Fig. 3. It appears to be Fig. 4

4. Line 324: It should be “Fig.5”.

These mis-labellings have now been corrected, and a more thorough read-through has been carried out before this submission to check that all figure and table references are correct throughout the manuscript and supplement.

Response to Reviewer 3

Thank the authors for their revised manuscript and for the substantial additional analyses provided in response to the previous reviews. The manuscript has improved, particularly through the inclusion of AMS-PMF analysis, additional discussion of sulfur-containing organic compounds (SCOs), thermogram information, and CIMS calibration for MSA and ammonium sulfate. The dataset is valuable because molecular-level aerosol observations over the northern Indian Ocean remain limited.

However, I still have several important concerns. In my view, the manuscript is not yet ready for final acceptance, mainly because the central scientific novelty and several quantitative/methodological aspects remain insufficiently resolved.

We thank the reviewer for this assessment and comments, which have helped to improve our manuscript. We have responded to these individually, below.

1 The main scientific novelty remains unclear.

The manuscript emphasizes that this is the first application of high-time-resolution, high-mass-resolution soft-ionization mass spectrometry at MCOH and that several SCOs are reported for the first time at this site. However, it is still not clear what new atmospheric process or mechanism has

been discovered. Many conclusions, including sulfate dominance, anthropogenic influence from the Indo-Gangetic Plain, aged organic aerosol, and extensive transport processing, are broadly consistent with previous studies. The authors should more explicitly state what new scientific understanding is obtained beyond the application of advanced instrumentation and molecular formula identification.

We thank the reviewer for this comment. We note that the intention with this paper is not to outline the discovery of a new atmospheric process or mechanism. However, we would like to highlight the following, specific contributions:

(1) This is the first time that online, high-time-resolution instrumentation has been used to characterise aerosol at this site.
(2) We found that the composition of organic aerosol is largely consistent across incoming air masses, regardless of back trajectory history. While previous studies have noted that organic aerosol at this site is highly aged, methodological limitations made it impossible to compare molecular composition between specific air masses at high time resolution in the way that we have been able to do here. Our results indicate that the organic aerosol composition cannot easily be used for source attribution at this site, while the ratio of SO_4/Org is more useful for distinguishing air masses that are more heavily influenced by the IGP. This is a genuine answer to the question of whether molecular organic information can be a useful tool to distinguish air mass history at this location.
(3) Our comparison with the molecular composition of aerosol in Delhi shows that certain distinctive features, including high concentrations of levoglucosan and CHON compounds, were largely absent after long-range transport to MCOH. While Delhi is only a single site, its source profile – including notable contributions from biomass burning and fossil fuel combustion – are broadly representative of cities in this region.
(4) We report the presence of previously unreported SCO species. We also highlight a potential artefact observed during FIGAERO measurements, which will be valuable for future studies that may encounter the same thing.

In response to the reviewer's comment, we have made some changes to the manuscript to attempt to highlight these findings more clearly.

Changes to the manuscript

- In section 3.6:

Lines 506–514: Together, these results indicate that the vast majority of organic aerosol at MCOH during the winter represents a single, highly-oxidised component. A small HOA/BBOA factor is also present, which may reflect local emissions, but it is extremely small in comparison with the mass of aged aerosol. This supports that conclusion that organic aerosol reaching MCOH is highly processed during long-range transport. While this finding is consistent with previous observations at the site, previous analyses were not able to analyse the organic composition at high time-resolution, and thus it has not previously been possible to establish whether the aerosol mass has distinct chemical characteristics associated with specific air mass origins. Our results indicate that, over the course of this campaign, the chemical composition of organic aerosol was not strongly dependent on transport pathways. Organic composition at MCOH cannot, therefore, be used to identify individual source regions. Rather, the clearest chemical signature that can be used to distinguish between air masses is the sulfate-to-organic ratio.

- In the conclusions:

Lines 573–80: This study presents the first online, high-time-resolution molecular characterisation of aerosol at MCOH. Results presented here show changes in the sulfate-to-organic ratio, but extremely low variability in organic aerosol chemical composition over the course of this campaign, despite air masses originating from distinct source regions. Concentration-weighted back trajectory analysis indicates that sulfate aerosol was more abundant in air masses originating from the IGP. However, the dominance of sulfate in all air masses, together with the extremely similar organic aerosol signature regardless of transport history, suggests that the regional atmosphere is dominated by well-mixed aerosol that has been aged in a manner consistent with both photochemic ageing and cloud processing. This indicates that the source region information is largely lost by the time the aerosol reaches the site.

2 The FIGAERO-CIMS background subtraction is not adequately described.

The manuscript presents an average background-subtracted CIMS mass spectrum, but the procedure used to determine and subtract the FIGAERO-CIMS background is unclear. The authors should specify whether the background was based on empty filters, or other methods. They should also describe how frequently backgrounds were measured and how background drift was handled.

We have now added the following description of the blank subtractions to the method section:

Lines 155–9: Every fourth collection was carried out through a HEPA filter to provide a background measurement, giving 24 background measurements over the course of the campaign. Although blank signals showed some variability, the relative standard deviation in the blanks was below 30% for the majority of identified compounds. The mean integrated blank signal for each compound was subtracted from each sample signal.

3 CIMS quantification remains incomplete.

The added calibrations for MSA and ammonium sulfate are useful, but most CIMS-detected organic and sulfur-containing organic species remain unquantified. The manuscript sometimes discusses relative signal contributions in a way that may be interpreted as quantitative mass contribution. The authors should clearly distinguish between signal fraction, semi-quantitative abundance, and true mass concentration. If quantitative conclusions are drawn from CIMS organic/SCO signals, compound-specific or class-level sensitivity assumptions and associated uncertainties should be provided.

We thank the reviewer for noting this inconsistency in language use. We have now worked through the manuscript and updated the language to make clear where we are referring to the mass, specifically, and where we are only referring to CIMS signal. We have updated the captions for tables and figures displaying CIMS data to make it clear that this is a comparison of signal, not mass.

Minor revision

4 The Delhi comparison is useful, but it should be framed as an illustrative contrast rather than direct source-to-receptor proof.

We agree with the reviewer that the framing here is important. For this reason, the section about the Delhi comparison already begins with the following:

"Here, we compare the characteristics of organic aerosol observed at MCOH with that of organic aerosol observed using the CIMS during a campaign in Delhi in 2019 (Haslett et al., 2023; Mishra et al., 2023), which is used as an illustrative example of aerosol composition in a highly-polluted continental environment within South Asia. The purpose of this comparison is not to demonstrate a direct

influence of Delhi emissions on aerosol observed at MCOH, but rather to provide insight into the compositional differences between a continental source region and the aged aerosol observed after regional transport over the northern Indian Ocean."

Changes to the manuscript

- We have now removed one instance of Delhi being referred to as a "source region" in section 3.7.
- In the abstract, we have changed the wording in the following sentence:

Lines 591–2: In comparison with mass spectra observed in the city of Delhi, which is an illustrative example of one of the key source regions for observed air masses…

5 The PMF section would benefit from clearer reporting of solution diagnostics, residuals, and uncertainty/rotational ambiguity.

We agree with the reviewer, and have now added substantial additional information about the PMF solution. Our more detailed response can be seen in our response to Reviewer 2, above.

6 Please ensure that terms such as "quantitative", "mass fraction", "concentration", and "signal fraction" are used consistently.

We have worked through the manuscript and changed instances where these words were used inconsistently.