We thank the editor and both reviewers for their careful and constructive evaluations. The manuscript has been substantially revised to improve methodological transparency, distinguish evidence from inference, strengthen uncertainty treatment, and moderate policy statements. The principal changes are: (i) a compact PMF robustness assessment in the main text and expanded diagnostics in the Supplement; (ii) an Aethalometer-model sensitivity analysis and explicit qualification of near-zero model-derived eBC_{ff} values; (iii) replacement of volatility-based OOA labels by LO-OOA and MO-OOA; (iv) a more cautious interpretation of OOA precursor sources and regional transport; (v) a dedicated limitations section; and (vi) editing for consistency, self-containment, and scientific style.

Referee 2

The authors report measurements of $PM_{2.5}$ composition using ToF-ACSM and aethalometer measurements combined with source apportionment and wind analysis at a site upwind of Delhi. They find that combustion emissions (BBOA + SFCOA) are the dominant primary source of $PM_{2.5}$ as well as a persistent elevated background of aged OA. These measurements at an upwind site are an important effort to understanding the dynamics of haze pollution in Delhi and throughout the IGP, and this dataset appears valuable. I have a few minor comments

We sincerely thank the reviewer for this positive and encouraging assessment of our work. We are grateful that the reviewer recognizes the value of these upwind measurements and their importance for understanding haze dynamics in the Indo-Gangetic Plain. We address each of the reviewer's comments in detail below.

One of the author's main conclusions is that direct reductions of primary emission will have limitations, as a substantial portion of the carbonaceous aerosol is secondary (e.g. line 381). This is an important finding from a policy standpoint; however, discussion is needed about what the dominant precursor sources of the OOA factors are. Specifically, if the major source of VOCs is from biomass burning, then reductions in primary aerosol will also reduce the secondary aerosol loading. This has very different implications than if the OOA is thought to be from natural sources, such as biogenic VOCs. This topic is brought up near the end of the paper (line 797), but if the authors have hypotheses on this, I would suggest adding it to the PMF discussion, such as in section 3.1.

We thank the reviewer for highlighting this important distinction and its implications for air-quality management. We agree that the originally submitted manuscript did not adequately discuss the likely sources of the gaseous organic precursors contributing to the OOA factors. We have therefore expanded the PMF discussion in Sections. 3.1 (Line 414-431) to evaluate the available evidence regarding the precursor mixture and to clarify the associated mitigation implications.

To address this point, we have expanded the PMF discussion to examine the available evidence regarding the precursor sources of OOA. Co-located aromatic VOC measurements from the same campaign, previously analyzed by Rathore et al., (2025), provide an independent indication of the sources influencing the organic precursor pool. Xylene and ethylbenzene were strongly correlated during the haze periods, with $r^2 = 0.97$, suggesting a common or closely related source influence. The regression slope, expressed as the xylene-to-ethylbenzene enhancement ratio $\Delta X/\Delta E$, was approximately 0.92 ppb ppb^{-1}. This value is lower than the ratio of 2.24 ± 0.33 commonly reported for many non-biomass-burning anthropogenic sources and falls within the range reported for biomass-burning plumes, which is generally below 1.2 (Monod et al., 2001; Nelson and Quigley, 1983; Wang et al., 2014). It is also comparable to the value of approximately 1.12 observed in biomass-combustion plumes in Brazil (Monod et al., 2001).

The low $\Delta X/\Delta E$ value supports an important biomass-burning influence on the aromatic VOC mixture during the haze periods. This interpretation is also consistent with the concurrent enhancement of BBOA, SFCOA, eBC_{bb}, and the oxygenated OA factors, together with the northwesterly transport signatures observed during polluted periods. Collectively, these observations suggest that biomass burning and other solid-fuel combustion sources supplied both directly emitted particles and gaseous organic compounds that subsequently contributed to OOA formation.

We have nevertheless treated this interpretation cautiously. Xylene is removed more rapidly than ethylbenzene during atmospheric oxidation, so photochemical aging can decrease the observed $\Delta X/\Delta E$ ratio. In addition, particle-phase PMF does not directly determine the sources of the VOCs, intermediate-volatility organic compounds, and semi-volatile organic compounds from which OOA forms. The available evidence therefore supports biomass and solid-fuel combustion as important, and potentially major, anthropogenic precursor influences during the observed haze periods. It does not demonstrate that these sources were the exclusive precursors of OOA. Contributions from traffic, industrial activities, residential fuel use, and biogenic VOCs cannot be quantified or excluded using the present dataset.

We have also revised the policy interpretation to distinguish between reducing primary particulate emissions alone and reducing the underlying combustion activity. Source-based reductions in biomass burning and solid-fuel combustion would decrease directly emitted particles together with co-emitted organic precursors, thereby potentially reducing both primary OA and the combustion-influenced contributions to LO-OOA and MO-OOA. By contrast, measures that remove or control only directly emitted particles without reducing gaseous precursor emissions may have a more limited effect on secondary OA. This clarification has been incorporated into the revised PMF discussion and the Summary and Conclusions.

Similar to above, is there any evidence that either of the OOA factors is directly related to combustion processes? This may be difficult to determine with ACSM measurements, however,

there has been other work that has found OOA related to processed biomass burning aerosol appears spectrally similar to OOA (Such as Vasilakopoulou et al., 2023). It may also be important to note that f_{60} is lower when using a capture vaporizer (Zheng et al., 2020)

We thank the reviewer for this important comment and for drawing our attention to these relevant studies. We agree that the ACSM mass spectra alone cannot establish that either OOA factor originated directly or exclusively from biomass-burning or other combustion emissions. OOA factors are resolved principally according to their oxygenated spectral characteristics and may contain secondary material formed from several anthropogenic and biogenic precursor classes. We have revised section 3.1.2 to state this limitation explicitly.

Several independent observations nevertheless support a contribution from processed biomass and solid-fuel combustion emissions to the OOA factors during the haze periods. Both LO-OOA and MO-OOA increased during periods when BBOA, SFCOA, and eBC_{bb} were elevated. The nighttime enhancement of LO-OOA broadly tracked that of eBC_{bb}, suggesting that LO-OOA may contain secondary material formed relatively recently from combustion-related precursors under conditions of weak dispersion. Both oxygenated factors also exhibited northwesterly transport signatures similar to those of the combustion-related factors. In addition, the f_{44}–f_{43} and f_{44}–f_{60} relationships reported for the same campaign by Rathore et al., (2025) are consistent with the presence of aged biomass-burning aerosol. The low $\Delta X/\Delta E$ enhancement ratio provides further supporting evidence for an important biomass-burning influence on the gaseous organic precursor mixture.

We have incorporated the interpretation of Vasilakopoulou et al., (2023), which demonstrates that extensively processed biomass-burning OA can lose its characteristic fresh-combustion markers and become spectrally similar to generic OOA. This finding supports the possibility that aged biomass-burning material contributed to our oxygenated factors, particularly MO-OOA. At the same time, it reinforces the need for caution because such attribution generally requires independent tracers or modelling and cannot be established from the OOA spectrum alone.

We have also added the capture-vaporizer limitation identified by the reviewer. Zheng et al., (2020) showed that biomass-burning marker signals, including f_{60}, are lower for capture-vaporizer measurements than for standard-vaporizer measurements. Atmospheric aging can further reduce these marker signals. The BBOA contribution resolved in our analysis may therefore underestimate the total amount of fresh and processed biomass-burning OA. Some combustion-derived material may instead be represented within LO-OOA or MO-OOA. However, the available evidence does not permit us to reassign a quantitative fraction of either OOA factor to biomass burning.

The revised manuscript therefore interprets LO-OOA and MO-OOA as mixed secondary components with evidence for an important combustion influence during the haze periods. LO-OOA may contain relatively recently formed oxidation products from biomass and solid-fuel

emissions, while MO-OOA may include more extensively aged, regionally transported combustion-derived material. Neither factor is attributed uniquely to combustion, and contributions from fossil-fuel-related and biogenic precursors remain possible. We have added this interpretation to Section 3.1.2 and 3.1.3 (Line 455-522) and included the capture-vaporizer limitation in section 4 Summary and Conclusions (Line1131-1133).

I would suggest that the authors consider changing the name of SV-OOA and LV-OOA as no volatility measurements were conducted. Instead, terminology such as more oxidized OOA (MO-OOA) and less oxidized OOA (LO-OOA) may be more appropriate.

We thank the reviewer for highlighting this point. We have accordingly updated the terminology throughout the main manuscript, supplementary material, figures, and tables.

How was the O/C and H/C for the PMF factors parameterized?

We thank the reviewer for requesting clarification of this methodology. We agree that the originally submitted manuscript did not explain how the elemental ratios assigned to the PMF factors were obtained. We have now added a dedicated description in Section S2 of the Supplementary Information.

The O:C and H:C ratios were not measured directly for individual PMF factors. Instead, they were estimated from the unit-mass-resolution mass spectrum of each factor. For each normalized factor profile, f_{44} and f_{43} were calculated as the fractions of the total organic signal occurring at m/z 44 and m/z 43, respectively. Initial O:C and H:C estimates were then obtained using the Improved-Ambient parameterizations of Canagaratna et al., (2015):

$$\mathrm{O{:}\,C_{IA}} = 0.079 + 4.31 f_{44}$$
$$\mathrm{H{:}\,C_{IA}} = 1.12 + 6.74 f_{43} - 17.77 f_{43}^2$$

As the ToF-ACSM used in this study was equipped with a capture vaporizer, we subsequently applied the capture-vaporizer calibration factors reported by Hu et al., (2018) for ambient OA. The corrected ratios were calculated as:

$$\mathrm{O{:}\,C_{CV}} = 0.73\ \mathrm{O{:}\,C_{IA}} = 0.73(0.079 + 4.31 f_{44})$$
$$\mathrm{H{:}\,C_{CV}} = 0.72\ \mathrm{H{:}\,C_{IA}} = 0.72(1.12 + 6.74 f_{43} - 17.77 f_{43}^2)$$

The same procedure was applied to the representative mass spectrum of each PMF-resolved factor, and the resulting values are reported in Table S1. Canagaratna et al., (2015) noted that the f_{44}-based O:C parameterization is less accurate for primary OA factors with very low f_{44}, while Hu et al., (2018) reported estimated uncertainties of approximately 23% for O:C and 18% for H:C for standard organic compounds measured with a capture vaporizer. We therefore now describe the factor-specific ratios as parameterized estimates and use them mainly to compare

the relative degree of oxygenation among factors rather than as precise measurements of elemental composition.

Can the authors expand on what type of fuel is typically used in this region for solid fuel combustion. Is there evidence that crop burning and SFC have equivalent angstrom exponents?

We thank the reviewer for this important question. We have expanded the manuscript to clarify both types of solid fuels commonly burned in northern India and the limitations of representing these diverse combustion sources using a single absorption Ångström exponent.

The SFCOA factor should be interpreted as a mixed combustion-related source class rather than as the signature of a single fuel. Relevant sources in Delhi-NCR and the surrounding Indo-Gangetic Plain include fuelwood, dung cakes, crop residues, charcoal, and other solid fuels (Tobler et al., 2020) used for household cooking and heating, as well as emissions from open burning of municipal waste. These fuels differ substantially in composition, moisture content, burning conditions, and combustion efficiency and may therefore produce different primary organic aerosol profiles and optical properties.

There is no compelling evidence that open-field crop-residue burning and residential or small-scale industrial solid-fuel combustion have equivalent AAEs. Field measurements of residential heating emissions in India have reported absorption Ångström exponents ranging from approximately 1.34 to 2.57 (Navinya et al., 2025) for emissions associated with firewood, crop residues, and dung cakes, illustrating the considerable variability among solid-fuel combustion conditions. More importantly, measurements in the northwestern Indo-Gangetic Plain have shown that the absorption Ångström exponent is influenced strongly by combustion efficiency and cannot be treated as a unique indicator of fuel type. For example, efficiently flaming biomass combustion may produce an AAE close to 1, whereas poorly optimized fossil-fuel combustion may produce values greater than 1.

The assumed $\alpha_{\mathrm{bb}} = 2.0$ in our Aethalometer analysis therefore does not imply that crop-residue burning, wood combustion, dung burning, and other solid-fuel sources possess identical optical properties. Rather, it represents a conventional reference value used to define an operational biomass or solid-fuel-related optical component. The resulting eBC_{bb} contribution cannot distinguish among crop-residue burning, fuelwood, dung cakes, waste burning, or other brown-carbon-containing combustion sources. Its magnitude also depends on fuel composition, combustion phase, atmospheric aging, particle mixing state, and the assumed AAE values.

We have revised the manuscript accordingly. The SFCOA factor is now described as a mixed source category potentially influenced by residential and small-scale industrial combustion of wood, dung, crop residues, charcoal, and other locally used solid fuels. We have also explicitly clarified that eBC_{bb} is an operational optical source class rather than a source-specific measure of agricultural burning, and that different biomass and solid-fuel sources should not be assumed

to have equivalent AAEs. This clarification has been incorporated into the Methods, the AAE sensitivity discussion, and the limitations paragraph.

Line-by-line Comments

Line 34: “This region…” This sentence is missing a word

Noted. The updated sentence is added to manuscript (Line 36-39): “This densely populated and rapidly developing airshed is influenced by a complex mixture of emission sources, including vehicular traffic, industrial activities, residential biofuel consumption, and seasonal agricultural residue burning (Biswal et al., 2023; Ganguly et al., 2009).“

Line 161: empty parentheses

Removed the empty parentheses.(Line 166)

Line 178: SMPS has not been defined

Acronym Scanning Mobility Particle Sizer (SMPS) has been defined in Line 184 of revised manuscript.

Line 230 “..including air beam corrections..” this was already stated previously

The line had been removed.

Line 322: I’m confused by this sentence - at line 322, it says the five factor solution was obtained using unconstrained PMF, but then that one of the solutions was constrained. Can this be clarified?

We thank the reviewer for pointing out this ambiguity. We agree that the wording in the originally submitted manuscript was imprecise because it did not clearly distinguish between the procedure used to determine the number of factors and the approach used to obtain the final reported solution.

We first explored unconstrained PMF solutions containing three to seven factors. The appropriate factor number was evaluated using changes in Q/Q_{exp}, where Q is the uncertainty-weighted sum of the squared differences between the measured and modelled data and Q_{exp} is the statistically expected value of Q based on the model degrees of freedom. A substantial decrease in Q/Q_{exp} indicates an improvement in model fit, whereas only a small decrease after adding another factor suggests limited additional explanatory value. Factor-number selection was also based on the residual structure, spectral interpretability, temporal behavior,

correlations with independent tracers, and evidence of factor mixing or splitting. Therefore, these diagnostics supported the selection of a five-factor structure.

The unconstrained five-factor solution, however, showed mixing between HOA and SFCOA. We therefore retained the five-factor structure but used the ME-2 framework for the final source-apportionment analysis, constraining only the HOA profile with a literature-based reference spectrum and a randomized a-value between 0 and 0.5. This allowed the HOA profile to vary within a defined range while improving the separation of HOA and SFCOA. Thus, the number of factors was identified through unconstrained exploration, whereas the final five-factor solution reported in the manuscript was obtained using an HOA-constrained ME-2 analysis.

We have revised the corresponding text in Sect. 3.1.1 (Line 358-392) to make this sequence explicit. We have also removed the policy-based justification for applying the constraint and now justify the methodological choice solely on the basis of factor separation, chemical interpretability, and sensitivity testing. Additional details on the factor-number exploration and the stability of the constrained solution are provided in Section S1 of the Supplementary Information.

Line 366. Section numbering is incorrect

Corrected.

Line 571: The statement about the increase in BBOA concentration is repeated, it is also stated at line 560

The repetitive statement at Line 571 of original manuscript has been removed.

Line 578: The authors claim that the elevated SV-OOA during H2 is due to the shorter event duration, however, it is unclear how they came to that reasoning. For example, could it also be due to elevated local emissions, rather than long-range transport. Specifically, if it was the duration of the event that was the primary factor, I would expect the fraction of LVOOA to build up over the course of the events. Instead the opposite trend is observed in H1, with very little SVOOA at the start of the event. This should be reconciled, or additional evidence should be provided.

We thank the reviewer for identifying this inconsistency. We agree that the explanation in the originally submitted manuscript, which attributed the relatively large less-oxidized OOA contribution during H2 primarily to the shorter duration of the episode, was not adequately supported by the observations. As the reviewer correctly notes, the temporal evolution during H1 does not show a simple progressive conversion from a less-oxidized to a more-oxidized factor as the event proceeds. Event duration alone therefore cannot explain the contrasting OOA distributions in H1 and H2.

We have removed this duration-based interpretation from the revised manuscript. The comparable concentrations of less-oxidized oxygenated organic aerosol, LO-OOA, and more-oxidized oxygenated organic aerosol, MO-OOA, during H2 are now interpreted as evidence of a different mixture of emissions and atmospheric processing relative to H1. The temporal coincidence of H2 with the Diwali period, together with elevated SFCOA and the comparatively larger LO-OOA contribution, is consistent with an enhanced influence of recently emitted combustion products and rapid secondary formation or partitioning superimposed on an existing regional aged aerosol background.

We have nevertheless avoided attributing the H2 pattern definitively to local emissions or fireworks. The measurements at a single receptor site and the PMF analysis cannot determine whether the fresher material originated locally or was transported over shorter regional distances. They also cannot uniquely separate Diwali-related emissions from other biomass, solid-fuel, traffic, or industrial sources active during the same period. We therefore describe Diwali-related emissions as a plausible contributing influence rather than the sole explanation for the observed factor distribution.

The revised discussion(Line 730-739) now emphasizes that the relative contributions of LO-OOA and MO-OOA depend on the evolving source mixture, oxidation history, gas-particle partitioning, meteorological conditions, precipitation history, and atmospheric transport. We no longer identify event duration as the primary control. We thank the reviewer for this observation, which has led to a more cautious and physically consistent interpretation of the episode-wise differences.

Line 677-682: HOA and eBC_{ff} was already discussed in the section above.

We thank the reviewer for catching this repetition. We have removed this paragraph (Lines 677–682) from the manuscript, as the discussion of HOA and eBC_{ff} was indeed redundant with the earlier section.

Line 761 and line 749: Cl^- appears to be included in both primary and secondary?

We thank the reviewer for pointing this out. This was indeed a typographical error, and the text has been corrected accordingly - Cl^- is correctly considered a secondary species.(Line 911)

Figure S1b - can BBOA be added to the diurnal profile here? Also, do the authors have any explanations of the diurnal shift between SFCOA and eBC_{bb}?

We thank the reviewer for this helpful suggestion. We have added the diurnal variation of BBOA to the corresponding panel. Following the reorganization of the Supplementary Information, the figure referred to by the reviewer is now Figure S2, and BBOA is included in the upper-right panel of Figure. S2a together with SFCOA and eBC_{bb}.

The revised figure shows that SFCOA, BBOA, and eBC_{bb} share a broadly similar diurnal pattern, with relatively high concentrations during the night and evening and lower concentrations during the daytime. However, their hourly maxima and the timing of their evening increases do not coincide exactly. We agree that this difference warrants clarification, but we do not interpret it as evidence for a uniquely defined aging delay.

SFCOA and eBC_{bb} are derived from different measurements and represent partly overlapping rather than identical source categories. SFCOA is a PMF-resolved organic aerosol factor identified from the ToF-ACSM mass spectra and may represent organic aerosol emitted from several residential, commercial, and other solid-fuel-combustion activities. In contrast, eBC_{bb} is an operational optical component derived from the wavelength dependence of aerosol absorption using assumed values of α_{ff} and α_{bb} (Deng et al., 2020). It may include contributions from crop-residue burning, fuelwood, dung, and other brown-carbon-rich combustion sources and cannot distinguish independently among them.

Differences between the SFCOA and eBC_{bb} diurnal profiles may therefore arise from variations in the timing and relative importance of the contributing combustion sources, differences in OA-to-BC emission ratios among fuels and combustion phases, atmospheric transport and boundary-layer evolution, and the volatility and gas-particle partitioning of combustion-derived organic material. The detailed eBC_{bb} profile may also be affected by the use of fixed AAE values, because aerosol spectral absorption varies with fuel type, combustion conditions, atmospheric processing, and particle mixing state. Previous comparisons of PMF- and Aethalometer-derived biomass-burning components have similarly shown that their diurnal patterns need not coincide exactly because the methods respond to different aerosol properties and source mixtures(Elser et al., 2016).

We have revised the Supplementary Information accordingly (Section S3). We have also removed our earlier speculative explanation that freshly emitted particles may become fully represented by eBC_{bb} only after one to several hours of atmospheric aging. The available measurements do not support such a specific timescale. We now interpret the modest temporal offset as reflecting differences between the source mixtures and atmospheric processes represented by SFCOA and eBC_{bb}, rather than attributing it to a single mechanism. The addition of BBOA provides a further comparison with the biomass-burning-related organic aerosol component.

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