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 1

The manuscript is a 54-page Egusphere preprint posted on **15 April 2026** and presents high-time-resolution ToF-ACSM and Aethalometer measurements from **Sonipat** during **25 October-15 November 2023**, with PMF, Aethalometer-model BC apportionment, and trajectory/wind analyses. Its central claim is that severe post-monsoon haze reaching Delhi-NCR is already strongly carbonaceous and regionally processed before entering the urban core.

The study is timely, the site is strategically important, and the dataset appears valuable. The paper's strongest contribution is its framing of Sonipat as an upwind receptor that can separate regional transport from Delhi-core emissions, and the reported dominance of OA and biomass-related BC is potentially important for air-quality policy in the Indo-Gangetic Plain. However, several key methodological choices and interpretive steps need to be tightened before publication, especially around PMF constraints, uncertainty quantification, BC source apportionment assumptions, and the degree to which some conclusions go beyond what the evidence directly supports.

We sincerely thank the reviewer for this careful, balanced, and constructive assessment of our manuscript. We appreciate the reviewer's recognition of the value of the high-time-resolution observations, the strategic location of the Sonipat site, and the potential significance of the observed dominance of organic aerosol and biomass-related black carbon for understanding post-monsoon air pollution in the Indo-Gangetic Plain.

We agree that the original manuscript did not provide sufficient quantitative documentation of several important methodological uncertainties and, in some places, expressed the source-attribution and regional-policy implications more strongly than warranted by the available evidence. In response, we have revised the manuscript. In particular, we have expanded the description and evaluation of the PMF solution by including factor-number selection, rotational sensitivity, bootstrap reproducibility, and the influence of the HOA constraint. We have also

added a sensitivity analysis and a more explicit uncertainty discussion for the Aethalometer-model black-carbon apportionment, clarified the assumptions and limitations associated with the trajectory and wind-based source interpretation, and moderated statements that could imply a definitive separation of regional and local contributions.

The revised manuscript now describes Sonipat more precisely as an upwind receptor under the prevailing post-monsoon north-westerly transport conditions, rather than as a site that can independently quantify the complete regional-versus-urban pollution balance. Accordingly, our conclusions emphasize that the measurements provide strong evidence for substantial regional transport, biomass- and solid-fuel-related emissions, and secondary processing at the site. At the same time, we acknowledge the possible influence of local and mixed sources and the limitations inherent in receptor-based attribution.

We are grateful to the reviewer for identifying these issues. We believe that the resulting revisions have substantially improved the methodological transparency, scientific balance, and overall robustness of the manuscript.

Major comments

1. **PMF robustness is not documented strongly enough in the manuscript text.**

The methods say SoFi was used, with FPEAK exploration plus displacement and bootstrap analyses, and the results mention a five-factor solution with a constrained HOA profile using a randomized a-value between 0 and 0.5. But the manuscript text shown here does not give the reader enough quantitative evidence to judge whether the chosen solution is truly robust: for example, how many factor numbers were tested, how Q changed, how often bootstrap runs reproduced the selected factors, whether DISP indicated swaps, or how sensitive source contributions were to the HOA constraint. That is especially important because the paper's interpretation depends heavily on separating **HOA vs SFCOA** and **fresh vs aged oxygenated OA**. I would ask the authors to add a compact but explicit robustness section in the main paper, not only in supplementary material.

Response: We thank the reviewer for this important comment. We agree that the originally submitted manuscript did not provide sufficient quantitative information in the main text for readers to evaluate the robustness of the selected PMF solution, particularly the separation of HOA from SFCOA and the stability of the two oxygenated OA factors. We have therefore added a dedicated subsection 3.1.1 entitled **"**PMF solution selection and robustness"(Line 358-392) in

Section 3.1 of the revised manuscript. This subsection now reports the factor numbers tested, changes in Q/Q_{exp}, FPEAK sensitivity, bootstrap reproducibility, sensitivity to the HOA a-value constraint, and residual diagnostics. The corresponding detailed figures and analyses remain in Section S1 of the Supplementary Information.

We also thank the reviewer for drawing attention to the reference to displacement analysis in the original Methods section. DISP analysis was not performed in this study as we followed the ME-2/SoFi source apportionment framework of Canonaco et al., (2021). We have clarified it in method section 2.3.1(Line 250-252). Rotational sensitivity was evaluated through FPEAK exploration and sensitivity to the HOA a-value constraint, while statistical reproducibility was assessed through bootstrap resampling. We therefore do not make any claim regarding factor swaps based on DISP.

Unconstrained PMF solutions containing three to seven factors were first examined. The decrease in Q/Q_{exp} for the transitions from three to four, four to five, five to six, and six to seven factors were 29.9%, 41.2%, 20.6%, and 17.9%, respectively. The transition from four to five factors produced the largest relative improvement. Although Q/Q_{exp} continued to decrease for the six- and seven-factor solutions, these higher-order solutions produced splitting of existing factors and did not yield additional profiles with sufficiently distinct mass spectra, temporal behaviour, or tracer relationships. The five-factor structure was therefore selected using the combined statistical and chemical diagnostics rather than from the Q/Q_{exp} elbow alone.

The unconstrained five-factor solution showed mixing between HOA and SFCOA. We consequently retained the five-factor structure but used the ME-2 framework for the final reported solution, constraining only the HOA reference profile with a-values between 0 and 0.5. BBOA, SFCOA, LO-OOA, and MO-OOA were left unconstrained. For the five-factor solution, Q/Q_{exp} ranged from 9.17 to 9.69 over FPEAK values from -1.0 to $+1.0$, with the minimum occurring at FPEAK $= -0.2$. The relatively limited variation of the fit statistic across this range indicates that the overall model fit was not strongly sensitive to the applied FPEAK value, although this result alone does not demonstrate the absence of rotational ambiguity.

Bootstrap resampling using 500 iterations reproduced and successfully mapped all five factors in all runs. The result indicated high reproducibility of the factor structure under resampling. In addition, varying the HOA a-value between 0 and 0.5 resulted in campaign-average contributions of 7-9% for HOA, 21-26% for SFCOA, 10-14% for BBOA, 18-20% for LO-OOA, and 36-40% for MO-OOA, without a systematic redistribution between HOA and SFCOA across the tested range. These results indicate solutions are stable to a-value constraint.

The scaled residuals were centered close to zero and did not show a persistent positive or negative bias with time or m/z. However, the fitted residual-distribution width of 3.52 was broader than the ideal unit-width distribution. We have therefore revised the manuscript to state

that the solution showed no strong systematic directional bias while retaining some unresolved variability, rather than describing the residual fit as ideal. Taken together, the factor-number evaluation, FPEAK exploration, bootstrap analysis, HOA-constraint sensitivity, and residual diagnostics support the reproducibility and interpretability of the five-factor solution while also defining its remaining limitations.

2. **The Aethalometer-model source split needs a more cautious treatment and stronger uncertainty discussion.**

The manuscript uses fixed absorption Ångström exponents of **1.0 for fossil fuel** and **2.0 for biomass burning**. That is common practice, but it is also a known source of uncertainty, especially during complex mixed-source periods such as post-monsoon haze and Diwali. The reported result that eBCbb accounts for about **78%** of BC overall and that eBCff falls to **0.03 µg m^{-3}** during H2 is striking. Because such values can be quite sensitive to the assumed exponents, the paper should include a sensitivity analysis showing how the biomass/fossil split changes across a plausible AAE range. Without that, the qualitative conclusion may still be reasonable, but the quantitative confidence appears overstated.

We thank the reviewer for this important and constructive comment. We fully agree that the quantitative separation of equivalent black carbon into fossil-fuel-related and biomass-burning-related components depends strongly on the absorption Ångström exponents assumed in the Aethalometer model. The values of $\alpha_{\mathrm{ff}} = 1.0$and $\alpha_{\mathrm{bb}} = 2.0$ are commonly used in previous studies (Deng et al., 2020; Vaishya et al., 2017), but they cannot be regarded as uniquely constrained values for a complex environment such as Sonipat during post-monsoon haze and Diwali.

To evaluate this uncertainty explicitly, we repeated the source apportionment using combinations of α_{ff} between 0.9 and 1.1 and α_{bb}between 1.7 and 2.2 (Supplementary information Section S3 and Figure S10c). These ranges encompass values commonly reported for fossil-fuel and biomass or solid-fuel combustion aerosols. Across the tested combinations, the arithmetic mean of the time-resolved eBC_{ff}/eBC fraction varied from 20.3% to 61.6%, while the corresponding eBC_{bb}/eBC fraction varied from 38.4% to 79.7%. The sensitivity was substantially greater with respect to α_{bb}than to α_{ff}. For example, at $\alpha_{\mathrm{ff}} = 1.0$, increasing $\alpha_{\mathrm{bb}}$from 1.7 to 2.2 changed the mean eBC_{ff} fraction from 22.4% to 58.2%. In comparison, at $\alpha_{\mathrm{bb}} = 2.0$, varying $\alpha_{\mathrm{ff}}$from 0.9 to 1.1 changed the eBC_{ff} fraction from 43.6% to 49.9%. These results confirm that the numerical biomass and fossil-fuel split is assumption-dependent and should not be interpreted as a uniquely determined source contribution.

We have also clarified an important difference between the averaging metrics used in the original manuscript. The value of approximately 77.8% was calculated from the ratio of the campaign-mean concentrations, $10.9/(10.9+3.1)$, and therefore represents a mass-weighted contribution to the campaign-integrated eBC burden under the reference AAE pair. By contrast, the value of 53.6% obtained for $\alpha_{\mathrm{ff}} = 1.0$ and $\alpha_{\mathrm{bb}} = 2.0$ in the sensitivity analysis represents the arithmetic mean of the time-resolved eBC_{bb}/eBC fractions. The difference arises because the mass-weigted calculation gives greater influence to periods with high eBC concentrations, which were also periods of stronger biomass or solid-fuel influence. We now define these two metrics explicitly and avoid presenting the 77.8% value as an assumption-independent quantity.

We have revised the interpretation of the very small eBC_{ff} value obtained during H2 in the same way. The value of 0.03 μg $m^{-3}$is a model-derived estimate under the reference AAE pair and is particularly sensitive to the selected exponents. It is therefore no longer interpreted as evidence for the absence of fossil-fuel emissions. Instead, we describe it as indicating a strong biomass or solid-fuel optical signature under the reference parameterization. We have also added episode-specific sensitivity results, particularly for H2, to demonstrate how this estimate changes across the tested AAE combinations.

The revised manuscript further clarifies that eBC_{bb} and eBC_{ff} are operational optical source classes rather than uniquely source-specific measurements. The eBC_{bb} component may include contributions from crop-residue burning, wood combustion, dung burning, and other brown-carbon-rich solid-fuel emissions. It should not be interpreted exclusively as agricultural residue burning. We thank the reviewer for prompting this analysis, which has substantially improved the transparency and balance of the BC source-apportionment discussion. We have addressed these comments in main manuscripts section 2.3.2 (Line 270-286), section 3.1.4, section 4(Line 1100-1109) and section S3 in supplementary.

3. **The conclusions about policy relevance are reasonable, but some causal claims should be softened.**

The paper argues that haze over Delhi-NCR “largely forms outside the urban core” and that city-level controls alone cannot effectively mitigate peak PM. The transport evidence is strong enough to support an important regional component, but the phrase “largely forms outside the urban core” is stronger than what a single upwind receptor plus PMF/CWT can fully establish. The results show that Sonipat receives a major regional combustion plume before mixing with Delhi emissions, which is already a valuable conclusion. I would suggest recasting policy claims to

emphasize that **regional controls are necessary in addition to urban controls**, rather than appearing to diminish the relevance of urban sources altogether.

We sincerely thank the reviewer for this important and well-considered comment. We agree that the original manuscript expressed the regional contribution to Delhi-NCR haze more strongly than can be established from observations at a single upwind receptor. The measurements at Sonipat, together with the PMF, trajectory, concentration-weighted trajectory, and wind-based analyses, provide strong evidence that substantial carbonaceous aerosol loading associated with regional biomass and solid-fuel combustion was already present at the site under the prevailing post-monsoon northwesterly transport conditions. However, these analyses cannot quantify the complete regional-versus-urban source balance across Delhi-NCR or determine the additional aerosol mass formed after the sampled air masses enter and mix within the urban environment.

We have therefore removed the statement that severe haze over Delhi-NCR "largely forms outside the urban core" and have revised similar statements throughout the manuscript. The revised text now describes Sonipat as an upwind receptor that intercepts a substantial regional combustion influence before further interaction with Delhi's urban emissions under the observed transport conditions. We also clarify that PMF and trajectory-based analyses identify source characteristics and probable transport pathways but do not provide a complete source budget or absolute source contributions for the wider Delhi-NCR airshed.

The policy interpretation has been revised accordingly. We no longer imply that urban emissions or city-level controls are of secondary importance. Instead, the revised manuscript emphasizes that regional and urban sources contribute at different stages and locations within the airshed and must be addressed together. The presence of substantial carbonaceous aerosol at Sonipat indicates that urban controls alone may not fully address the transported pollution burden during post-monsoon haze episodes. At the same time, regional controls cannot substitute for strong measures targeting traffic, industry, construction, waste burning, and other sources within Delhi-NCR. Effective mitigation therefore requires coordinated airshed-scale reductions in regional combustion emissions and aerosol precursors alongside sustained urban emission controls.

We have incorporated this more balanced interpretation into the Abstract and section 4 "Summary and Conclusions". We thank the reviewer for this suggestion, which has helped us more clearly distinguish what our measurements directly demonstrate from the broader policy implications supported by the results.

4. **The manuscript would benefit from a dedicated limitations paragraph.**

I did not find a real limitation discussion in the main text. This paper especially needs one.

Relevant limitations include: ACSM measures non-refractory PM and misses refractory species; the source apportionment pertains to one site and one season; PMF factor labels are not unique; Aethalometer source apportionment depends on fixed AAEs; and CWT identifies likely source regions but not emissions inventories or source strengths directly. A frank limitations section would strengthen the paper, not weaken it.

We thank the reviewer for this important and constructive suggestion. We agree that the originally submitted manuscript did not include a dedicated discussion of the study limitations. We have addressed this omission by adding a clearly identified limitations paragraph to section 4 "Summary and Conclusions" (Line 1125-1160) of the revised manuscript. We have also revised related statements throughout the manuscript so that the interpretation of the results remains consistent with the scope and constraints of the observations and analytical methods.

The new limitations discussion acknowledges that the measurements cover only one post-monsoon period at a single receptor site. Consequently, the results cannot fully represent interannual variability, spatial heterogeneity, or the complete regional-versus-urban source balance across Delhi-NCR and the wider Indo-Gangetic Plain. We now state that coordinated measurements at upwind, urban, and downwind locations would be needed to quantify changes in aerosol mass and composition as polluted air masses move through the Delhi urban region.

We have also explicitly described the limitations of the ToF-ACSM measurements. The instrument quantifies non-refractory particulate matter but does not measure refractory components such as mineral dust, elemental material, and some trace metals. In addition, the use of a capture vaporizer can reduce the biomass-burning marker signal at $m/z60$ and may cause part of the combustion-related organic aerosol to be apportioned into oxygenated OA factors rather than a distinct BBOA factor (Zheng et al., 2020). We further clarify that the elemental ratios derived using empirical parameterizations (Canagaratna et al., 2015; Hu et al., 2018) should be interpreted as approximate indicators of aerosol oxidation state rather than direct quantitative measurements.

The revised manuscript now includes a more explicit discussion of the uncertainty associated with Aethalometer-based source apportionment. The estimated eBC source fractions depend on prescribed absorption Ångström exponents, which may vary with fuel type, combustion conditions, aerosol aging, brown-carbon contributions, and particle mixing state. We therefore describe eBC_{bb} and eBC_{ff} as operational optical source categories and emphasize that their exact quantitative separation is not uniquely constrained. This discussion is supported by the new AAE sensitivity analysis presented in the Supplementary information section S3.

We have also clarified that PMF factors are statistical representations of covarying mass-spectral signals. Although their interpretation is supported by characteristic spectral features, temporal

behavior, diurnal patterns, and correlations with independent tracers, the factor labels are not unique, and individual factors may include contributions from related sources or atmospheric processes. Similarly, trajectory and concentration-weighted trajectory analyses identify probable transport pathways and potential source regions, but they do not directly quantify emission strengths, provide emission inventories, or replace chemical transport modelling and source-specific measurements such as radiocarbon analysis.

In this way, we have linked these limitations to a more cautious interpretation of the principal findings. The revised manuscript states that the observations provide evidence for an important regional contribution to the carbonaceous aerosol measured at Sonipat under the prevailing northwesterly transport conditions. However, they do not establish the complete source balance across Delhi-NCR or quantify the additional contribution from urban emissions after the sampled air masses enter the city. We believe that the addition of this dedicated limitations discussion has substantially improved the transparency, scientific balance, and overall robustness of the manuscript.

Minor comments

The manuscript briefly shifts into first person in a way that reads unpolished for a research article: "**I have tried to answer some of those questions**." This should be rewritten in standard scientific style.

We thank the reviewer for pointing out this oversight. We entirely agree that the phrasing was unpolished. We have carefully revised this sentence and reviewed the rest of the manuscript to ensure a consistent, formal scientific tone is maintained throughout.

The methods would also read better with a clearer separation between what is reproduced from the prior Rathore et al. study and what is newly done here. Right now, several instrumental details are deferred to Rathore et al. (2025), which is understandable, but the present paper should still be as self-contained as possible for readers and reviewers.

We thank the reviewer for this helpful comment and agree that the distinction between the prior study and the present work should be stated explicitly rather than left implicit. We have added this clarification to the Introduction as below(Line 137-144):

"*Rathore et al., (2025) established this site as an upwind receptor and characterized the haze episodes at Sonipat, identifying two haze and two non-haze episodes through temporal variability, tracer, and fragment analyses combined with VOC emission ratios. Building on this foundation, the present work offers one of the first high-resolution PM2.5 source apportionment*

analyses at this strategically located upwind gateway to Delhi, providing new constraints on regional transport processes and source contributions relevant to air quality management across Delhi-NCR and the broader Indo-Gangetic Plain"

We have also revised the Instrumentation and Measurements section 2.2 (Line 172-227) to include brief standalone descriptions of key instrumental details that were previously only referenced via citation to Rathore et al., (2025), so that the manuscript can be understood independently. We retain citations to Rathore et al., (2025) where readers may wish to consult more extensive calibration, QA/QC, or uncertainty details beyond what is essential for interpreting the present results.

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:

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

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 Fig. S2, and BBOA is included in the upper-right panel of Fig. 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.

References:

Canagaratna, M. R., Jimenez, J. L., Kroll, J. H., Chen, Q., Kessler, S. H., Massoli, P., Hildebrandt Ruiz, L., Fortner, E., Williams, L. R., Wilson, K. R., Surratt, J. D., Donahue, N. M., Jayne, J. T., and Worsnop, D. R.: Elemental ratio measurements of organic compounds using aerosol mass spectrometry: characterization, improved calibration, and implications, Atmos. Chem. Phys., 15, 253–272, https://doi.org/10.5194/acp-15-253-2015, 2015.

Canonaco, F., Tobler, A., Chen, G., Sosedova, Y., Slowik, J. G., Bozzetti, C., Daellenbach, K. R., El Haddad, I., Crippa, M., Huang, R.-J., Furger, M., Baltensperger, U., and Prévôt, A. S. H.: A new method for long-term source apportionment with time-dependent factor profiles and uncertainty assessment using SoFi Pro: application to 1 year of organic aerosol data, Atmos. Meas. Tech., 14, 923–943, https://doi.org/10.5194/amt-14-923-2021, 2021.

Deng, J., Guo, H., Zhang, H., Zhu, J., Wang, X., and Fu, P.: Source apportionment of black carbon aerosols from light absorption observation and source-oriented modeling: an implication in a coastal city in China, Atmos. Chem. Phys., 20, 14419–14435, https://doi.org/10.5194/acp-20-14419-2020, 2020.

Elser, M., Huang, R.-J., Wolf, R., Slowik, J. G., Wang, Q., Canonaco, F., Li, G., Bozzetti, C., Daellenbach, K. R., Huang, Y., Zhang, R., Li, Z., Cao, J., Baltensperger, U., El-Haddad, I., and Prévôt, A. S. H.: New insights into $PM_{2.5}$ chemical composition and sources in two major cities in China during extreme haze events using aerosol mass spectrometry, Atmos. Chem. Phys., 16, 3207–3225, https://doi.org/10.5194/acp-16-3207-2016, 2016.

Hu, W., Day, D. A., Campuzano-Jost, P., Nault, B. A., Park, T., Lee, T., Croteau, P., Canagaratna, M. R., Jayne, J. T., Worsnop, D. R., and Jimenez, J. L.: Evaluation of the New Capture Vaporizer for Aerosol Mass Spectrometers (AMS): Elemental Composition and Source Apportionment of Organic Aerosols (OA), ACS Earth Space Chem., 2, 410–421, https://doi.org/10.1021/acsearthspacechem.8b00002, 2018.

Monod, A., Sive, B. C., Avino, P., Chen, T., Blake, D. R., and Sherwood Rowland, F.: Monoaromatic compounds in ambient air of various cities: a focus on correlations between the xylenes and ethylbenzene, Atmospheric Environment, 35, 135–149, https://doi.org/10.1016/S1352-2310(00)00274-0, 2001.

Navinya, C., Kapoor, T. S., Gupta, A., Haswani, D., Raman, R. S., Kumari, J., Khan, M. S., Habib, G., Venkataraman, C., and Phuleria, H. C.: Carbonaceous Aerosol Emissions From Residential Heating in India: Field Measurements of Emission Factors and Optical Properties, JGR Atmospheres, 130, e2025JD044270, https://doi.org/10.1029/2025JD044270, 2025.

Nelson, P. F. and Quigley, S. M.: The ,. A technique for estimating hydrocarbon age in ambient atmospheres, Atmospheric Environment (1967), 17, 659–662, https://doi.org/10.1016/0004-6981(83)90141-5, 1983.

Rathore, J., Ganguly, D., Singh, V., Gupta, M., Vazhathara, V. J., Biswal, A., Kunchala, R. K., Patra, P. K., Sahu, L. K., Gani, S., and Dey, S.: Characteristics of Haze Pollution Events During Biomass Burning Period at an Upwind Site of Delhi, JGR Atmospheres, 130, e2024JD042347, https://doi.org/10.1029/2024JD042347, 2025.

Tobler, A., Bhattu, D., Canonaco, F., Lalchandani, V., Shukla, A., Thamban, N. M., Mishra, S., Srivastava, A. K., Bisht, D. S., Tiwari, S., Singh, S., Močnik, G., Baltensperger, U., Tripathi, S. N., Slowik, J. G., and Prévôt, A. S. H.: Chemical characterization of PM2.5 and source apportionment of organic aerosol in New Delhi, India, Science of The Total Environment, 745, 140924, https://doi.org/10.1016/j.scitotenv.2020.140924, 2020.

Vaishya, A., Singh, P., Rastogi, S., and Babu, S. S.: Aerosol black carbon quantification in the central Indo-Gangetic Plain: Seasonal heterogeneity and source apportionment, Atmospheric Research, 185, 13–21, https://doi.org/10.1016/j.atmosres.2016.10.001, 2017.

Vasilakopoulou, C. N., Matrali, A., Skyllakou, K., Georgopoulou, M., Aktypis, A., Florou, K., Kaltsonoudis, C., Siouti, E., Kostenidou, E., Błaziak, A., Nenes, A., Papagiannis, S., Eleftheriadis, K., Patoulias, D., Kioutsioukis, I., and Pandis, S. N.: Rapid transformation of wildfire emissions to harmful background aerosol, npj Clim Atmos Sci, 6, 218, https://doi.org/10.1038/s41612-023-00544-7, 2023.

Wang, H., Lou, S., Huang, C., Qiao, L., Tang, X., Chen, C., Zeng, L., Wang, Q., Zhou, M., Lu, S., and Yu, X.: Source Profiles of Volatile Organic Compounds from Biomass Burning in Yangtze River Delta, China, Aerosol Air Qual. Res., 14, 818–828, https://doi.org/10.4209/aaqr.2013.05.0174, 2014.

Zheng, Y., Cheng, X., Liao, K., Li, Y., Li, Y. J., Huang, R.-J., Hu, W., Liu, Y., Zhu, T., Chen, S., Zeng, L., Worsnop, D. R., and Chen, Q.: Characterization of anthropogenic organic aerosols by TOF-ACSM with the new capture vaporizer, Atmos. Meas. Tech., 13, 2457–2472, https://doi.org/10.5194/amt-13-2457-2020, 2020.