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 $m/z 60$ 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.

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