

We thank the reviewers for their comments. All comments are addressed below. Reviewers' comments are included in italics, our responses are included in blue, and updated manuscript text is included in red.

### **Reviewer 1**

*The manuscript by Sahil Bhandari et al. presented a new method to conduct source apportionment, which can utilize large datasets collected using long-term monitoring compared to traditional positive matrix factorization approaches that do not resolve the diurnal pattern of factor profiles. In addition, the results showed that the new method resolved a greater diversity of factors compared to the traditional seasonal PMF approach in winter and monsoon seasons. In general, this manuscript is well written, but the following aspects should be fully addressed before it can be considered for publication.*

*1. The authors split the data into six 4-hour time windows, and found the differences of MS and TS of OA factors between new method and traditional positive matrix factorization approaches. My major concern is that are these differences (or the characteristics of MS/TS) affected by time division? For example, what are the differences between the results in 11:00-13:00 LT, 13:00-15:00 LT and 11:00-15:00 LT? The authors need to address such uncertainties in the revised manuscript.*

Response: There could be differences in MS/TS within subgroups of data in the four-hour time windows. To address this issue, we conducted detailed uncertainty analysis to ensure that the identified MS/TS are representative of the four-hour time windows (Sect. 2.5). The different uncertainty quantification using the approaches of bootstrapping (BS), displacement (DISP), and bootstrapping enhanced with displacement (BS-DISP) test data subgroups in 100s-10000s of PMF-like model runs to account for the issues of random error and rotational ambiguity within the four-hour time windows (Paatero et al., 2014). Detailed summary statistics from running these uncertainty analyses are presented as mappings onto the PMF solution for the entire time domain (Tables S8-S10). For BS, we set the number of bootstrap resamples at 100 and the default Pearson correlation coefficient at 0.6 for factor assignment. Thus, the BS analysis tests the random error and to a limited extent rotational ambiguity in each four-hour window using 100 additional PMF-like model runs. We show for the chosen combination of factors that BS-mapping, a metric of how factors for subgroups of data in each four-hour window mapped to the factors of the entire time window, is greater than 90% (Table S8). For DISP and BS-DISP, the exact number of PMF-like models conducted with each run depend on the number of species and the number of PMF factors (Paatero et al., 2014). DISP estimates the rotational ambiguity of solutions by displacing MS of each m/z in a factor slightly and attempting to find a solution with changed MS contributions at other m/zs. DISP runs 100s (~150-320) of PMF-like models within each DISP run. BS-DISP

simultaneously tests for random error and rotational ambiguity by first conducting a BS resample and then exploring the rotationally accessible space around each BS resample (Sect. 2.5). Thus, 10000s of (~15000-32000) PMF-like models are run within each BS-DISP run. Instead of mappings, DISP and BS-DISP track factor swaps; a small number of factor swaps suggests low rotational ambiguity and robustness of the PMF solution. We obtained zero swaps in DISP in all cases, and a low count of factor swaps in BS-DISP (Table S9). Thus, by conducting such detailed uncertainty analysis, we have addressed the uncertainties of choosing a given four-hour time window and found PMF factors representative of that four-hour time window, including for subgroups of data. While it is possible that additional information could be obtained using finer time windows, the 4-hour windows seem to have captured diurnal variations of the mass spectra well (e.g., cooking influence midday in contrast to nighttime) while keeping the computational burden under control. Future work could investigate the optimal length of the time window to sufficiently represent the finer time variations (less than 4 hours) in mass spectral profiles while managing computational burden.

To address the reviewer's comments, we have added the following text in Sect. 2.5 Uncertainty estimation:

“This detailed uncertainty analysis ensures that the identified MS/TS are representative of the four-hour time windows by fitting 100s-10000s of PMF-like model runs to data subgroups within the four-hour time windows (Paatero et al., 2014). Detailed summary statistics from running these uncertainty analyses are presented as mappings onto the PMF solution for the entire time domain (Tables S8-S10). The algorithms and computational workload of these techniques are described in detail elsewhere (Paatero et al., 2014).”

*2. More information needs to be listed to support source apportionment results in HOA and COA, as the current mass spectra appear to be confusing. What about their correlations with tracer species? In fact, the authors showed the correlations in Fig. S22-23 and S7-S8, but more discussion should be included in the main text. In addition, how about the results of 4/5/6-factor solutions?*

Response: We address the second half of the reviewer comment first. The detailed decision-making process regarding the number of factors is described in Sect. S1 (Tables S3–S10, Figs. S2–S13, S16–S30). The process is mostly described in the supplement because most of the steps have been documented in established literature (Ulbrich et al., 2009). We discuss the PMF uncertainty analysis in the main manuscript as it has been used relatively sparsely. The EPA PMF tool provides detailed uncertainty analyses tools to validate how representative the chosen PMF solutions are for the respective time windows. Here, we use the uncertainty analysis to select PMF solutions;

we only finalize solutions that pass the EPA PMF tests of random error and rotational ambiguity (Sect. 2.5). All other solutions are rejected. The application of these detailed uncertainty analyses to select a PMF solution for each time window, including the consideration of 4–6-factor solutions, is documented in Table S6, with supporting information in Tables S5, S7–S10.

We separate HOA and COA-related PMF factors in multiple periods. In monsoon seasonal PMF analysis, we separate an HOA factor with an MS strongly correlated with the reference COA factor MS. However, for separation of cooking organic aerosol in this study, we used the Robinson et al (2018) ratio of contributions at  $m/z$  55:57 of 1.6 as a preliminary test for relative positioning of the HOA and COA profiles (COA factors with the ratio close to or greater than 1.6 and HOA profiles with the ratio substantially lower than 1.6). The monsoon seasonal POA factor MS had a  $m/z$  55 to  $m/z$  57 ratio of 1.2 (Fig. S5). Therefore, the seasonal monsoon POA factor is presented as an HOA factor. In M172303, we observe clear separation in the MS of HOA and COA factors, with strong correlations ( $R > 0.9$ ) with respective reference MS profiles, and ratio of contributions at  $m/z$  55:57 of 1.7 for the COA factor and 1.1 for the HOA factor (Fig. S20). In M171115, the COA-HOA factor mass spectra might appear confusing since the factor MS is strongly correlated to both HOA and COA reference MS profiles. Indeed, the ratio of contributions at  $m/z$  55:57 of 1.4 for the COA-HOA factor reflects factor mixing. In W171115, the SFC-OA profile obtained correlates strongly with a solid-fuel combustion profile obtained using measurements in Delhi elsewhere (Tobler et al., 2020; correlation at all  $m/z$ s but  $m/z$  44, Pearson  $R > 0.95$ , Fig. S18). Also, the ratio of contributions at  $m/z$  55:57 of 1.1 for the SFC-OA factor reflects the limited influence of cooking on that factor.

We present tracer species and their time series correlations with PMF factors in Figs. S6–S8 (seasonal PMF analysis) and Figs. S21–S24 (time-of-day PMF analysis). We use two tracers for HOA-influence: CO and the fossil-fuel component of black carbon, BCFF, estimated using the model of Sandradewi et al. (2008). For the time-series of BBOA factors, we use three tracers: (i) chloride (under the influence of agricultural and other open waste burning-related contributions (Li et al., 2014a, b; Kumar et al., 2015; Fourtziou et al., 2017), (ii)  $\Delta C$ , defined as the difference between UVPM (370 nm) and BC detected by the aethalometer (Wang et al., 2011; Olson et al., 2015; Tian et al., 2019), and (iii) the biomass-burning component of black carbon, BCBB, estimated using the model of Sandradewi et al. (2008). COA-related factors often exhibit weak correlations with external tracers (Huang et al., 2010, Sun et al., 2011, Liu et al., 2012, Sun et al., 2013, Hu et al., 2016, Stavroulas et al., 2019). However, correlations with chloride in COA-like factors is suggestive of the influence of landfill emissions, trash burning, and solid-fuel sources (Dall'Osto et al., 2015, Lin et al., 2017). In the seasonal monsoon PMF run (M17), we observe only one primary factor, an HOA factor with strong correlations with tracers CO (Spearman  $R$ : 0.73) and BCFF

(Spearman R: 0.91) (Fig. S8). The winter midday SFC-OA profile correlates strongly with chloride (Spearman R: 0.71), nitrate (Spearman R: 0.75), BCFF (Spearman R: 0.79), and  $\Delta C$  (Spearman R: 0.60), pointing to the mixing of HOA, BBOA, and possibly COA influence in the factor (Fig. S21). At winter nighttime, we separate an HOA MS profile that correlates strongly with BCFF (Spearman R: 0.84) and CO (Spearman R: 0.83). We obtain one BBOA factor each at winter midday and winter nighttime. Among the two BBOA obtained, winter midday BBOA correlates strongly with chloride (Spearman R: 0.66) and CO (Spearman R: 0.67), suggesting an industrial source (Fig. S21, Sect. 3.1). At nighttime however, winter BBOA correlates strongest with the wood burning component of BC (BCBB, Spearman R: 0.92) and weakly with chloride (Spearman R: 0.40), suggesting at least two different origins of BBOA (Fig. S22). This is consistent with our previous work, where we have separated BBOA-like factors with different correlations with chloride and BCBB in different seasons (Bhandari et al., 2020; Patel et al., 2021a). In monsoon midday, we observe only one primary factor, a COA-HOA factor, with strong correlations with chloride (Spearman R: 0.75), suggesting the influence of landfill emissions, trash burning, and solid-fuel sources (Fig. S23). Otherwise, COA-HOA has weak correlations with external tracers. In the monsoon nighttime PMF run (M172303), we observe stronger correlations of the HOA factor with CO (Spearman R: 0.79) and BCFF (Spearman R: 0.86) compared to correlations of these tracers with the COA factor (CO: Spearman R: 0.70, BCFF: Spearman R: 0.71) (Fig. S24).

To address the reviewer comments regarding methods to obtain PMF solutions, including checking for 4/5/6-factor solutions, we have updated the text in Sect. 2.4:

“Details of the steps for conducting PMF, R code, and criteria for factor selection are discussed in detail in the Supplement (Sect. S1). Briefly, for selection of PMF solutions, we started by analyzing the different statistics of Q/Q<sub>exp</sub> (a measure of fit), correlogram of residual TS and correlation with external tracers, time series patterns in residuals, and PMF fits at different m/zs (Table S4). We also considered the correlation of factor mass spectral profiles with reference mass spectra since MS of different factors are characterized by different spectral signature peaks (Zhang et al., 2011). For example, hydrocarbon-like organic aerosol (HOA) is a proxy for fresh traffic and combustion emissions and shows prominent peaks at m/z values 55 and 57 and a higher fractional organic signal at m/z 43 than m/z 44. For separation of cooking organic aerosol (COA) and distinguishing it from HOA in this study, we used the Robinson et al (2018) ratio of contributions at m/z 55:57 of 1.6 as a preliminary test for relative positioning of the HOA and COA profiles (COA factors with the ratio close to or greater than 1.6 and HOA profiles with the ratio substantially lower than 1.6). We also validated obtained PMF factors by correlation of factor time series with external tracers. We use two tracers for

HOA-influence: CO and the fossil-fuel component of black carbon,  $BC_{FF}$ , estimated using the model of Sandradewi et al. (2008). For the time-series of BBOA factors, we use three tracers: (i) chloride (under the influence of agricultural and other open waste burning-related contributions (Li et al., 2014a, b; Kumar et al., 2015; Fourtziou et al., 2017), (ii)  $\Delta C$ , defined as the difference between UVPM (370 nm) and BC detected by the aethalometer (Wang et al., 2011; Olson et al., 2015; Tian et al., 2019), and (iii) the biomass-burning component of black carbon,  $BC_{BB}$ , estimated using the model of Sandradewi et al. (2008). COA-related factors often exhibit weak correlations with external tracers (Huang et al., 2010, Sun et al., 2011, Liu et al., 2012, Sun et al., 2013, Hu et al., 2016, Stavroulas et al., 2019). Additionally, the EPA PMF tool provides detailed uncertainty analyses tools to validate how representative the chosen PMF solutions are for the respective time windows. Here, we use the uncertainty analysis to select PMF solutions; we only finalize solutions that pass the EPA PMF tests of random error and rotational ambiguity, as described below in Sect. 2.5. The application of these detailed uncertainty analyses to select a PMF solutions for each time window, including the consideration of 3–8-factor solutions, is documented in Table S6, with supporting information in Tables S5, S7–S10.”

To address the reviewer’s comments regarding time series correlations with tracer species, we have updated the text in Sect. 3.1:

“In monsoon, the seasonal PMF HOA MS is also strongly correlated with the reference COA factor MS (Ng et al., 2011a; Pearson  $R \sim 0.90$ ; Fig. S5). However, the monsoon seasonal POA factor MS had a  $m/z$  55 to  $m/z$  57 ratio of 1.2 (Fig. S5). Therefore, the seasonal monsoon POA factor is presented as an HOA factor. This HOA factor has stronger correlations with tracers CO (Spearman  $R$ : 0.73) and  $BC_{FF}$  (Spearman  $R$ : 0.91) than the OOA factors (Fig. S6).”

To address the reviewer’s comments regarding time series correlations with tracer species, we have also updated the text in Sect. 3.2.1:

“The winter midday SFC-OA profile correlates strongly with chloride (Spearman  $R$ : 0.71), nitrate (Spearman  $R$ : 0.75),  $BC_{FF}$  (Spearman  $R$ : 0.79), and  $\Delta C$  (Spearman  $R$ : 0.60), pointing to the mixing of HOA, BBOA, and possibly COA influence in the factor (Fig. S21). At winter nighttime, we separate an HOA MS profile that correlates strongly with  $BC_{FF}$  (Spearman  $R$ : 0.84) and CO (Spearman  $R$ : 0.83). We obtain one BBOA factor each at winter midday and winter nighttime. Among the two BBOA obtained, winter midday BBOA correlates strongly with chloride (Spearman  $R$ : 0.66) and CO (Spearman  $R$ : 0.67), suggesting an industrial source (Fig. S21, Sect. 3.1). At nighttime however, winter BBOA correlates strongest with the wood burning component of BC

(BC<sub>BB</sub>, Spearman R: 0.92) and weakly with chloride (Spearman R: 0.40), suggesting at least two different origins of BBOA (Fig. S22). This is consistent with our previous work, where we have separated BBOA-like factors with different correlations with chloride and BC<sub>BB</sub> in different seasons (Bhandari et al., 2020; Patel et al., 2021a). In monsoon midday, we observe only one primary factor, a COA-HOA factor, with strong correlations with chloride (Spearman R: 0.75), suggesting the influence of landfill emissions, trash burning, and solid-fuel sources (Fig. S23). Otherwise, COA-HOA has weak correlations with external tracers. In the monsoon nighttime PMF run (M172303), we observe stronger correlations of the HOA factor with CO (Spearman R: 0.79) and BC<sub>FF</sub> (Spearman R: 0.86) compared to correlations of these tracers with the COA factor (CO: Spearman R: 0.70, BC<sub>FF</sub>: Spearman R: 0.71) (Fig. S24).”

3. What is the justification for distinguishing between local OOA and regional OOA? Figs. S27-S30 did not support your conclusion in lines 412-414 in my sense.

Response: In Figs. S27–S30 we present the normalized level diurnal variations of the local OOA and the regional OOA factors. Typically, regional OOA is more oxidized (shows weaker correlations with reference SVOOA MS) and has less diurnal variation, in line with its expected average lower volatility and contributions from long-range transport (Drosatou et al., 2019). To quantify the flatness of the diurnal variations in each time window, the table below presents the lowest and the highest levels (relative to the normalization to 1) and the range (difference of the highest to the lowest levels) of the two factors. In all periods, the local OOA exhibits a similar or larger range than the regional OOA factor. However, we see an overlap of the 95% confidence intervals of the normalized levels (Figs. S27–S30).

Period	Lowest level		Highest level		Range (Highest – Lowest)	
	Local OOA	Regional OOA	Local OOA	Regional OOA	Local OOA	Regional OOA
W171115	0.86	0.82	1.21	1.08	0.35	0.26
W172303	0.87	0.71	1.26	1.11	0.39	0.40
M171115	0.81	0.89	1.18	1.15	0.37	0.26
M172303	0.79	0.97	1.12	1.02	0.33	0.05

We have updated the text (Sect. 3.2.2):

“Time-of-day PMF and seasonal PMF generate two OOA factors, local OOA and regional OOA, in each run (Figs. S25 and S26). Typically, regional OOA is more oxidized (shows weaker correlations with reference SVOOA MS) and has less diurnal variation, in line with its expected average lower volatility and contributions from long-

range transport (Drosatou et al., 2019). The time-of-day PMF OOA factors show MS and TS behavior similar to the seasonal PMF OOA factors, as shown in Sect. 3.3. Mass spectra of both local OOA and regional OOA correlate strongly with the reference OOA factor (Pearson  $R > \sim 0.80$ ) (Figs. S25 and Figs. 26). Also, we consistently observe that the more oxidized regional OOA factors have flatter diurnal time series patterns (smaller range) than the less oxidized local OOA factors (larger range) (Figs. S27–S30; Table S11). However, we see an overlap of the 95% confidence intervals of the normalized levels (Figs. S27–S30) and an overlap of external tracers suggesting mixing of the two OOA components (see Sect. S4). This is not surprising considering similarity of the MS of the two OOA factors and a continuum of the level of oxidation in the atmosphere. Since we observe factor mixing of the two secondary components, detailed analysis of the factor MS and TS (correlations with external tracers, features of the mass spectra) are only presented in the Supplement (see Sect. S4).”

We have also added the table to the Supplement (Table S11).

*4. What are the correlations of same type of OA factors between the daytime and nighttime? It would be nice to have some comparison of MS of the same type of OA factors between daytime and nighttime. In my sense, the differences in MS between day and night in OA factors are the highlights of this paper. However, the potential differences between day and night and the reasons have not been discussed in depth.*

Response: We agree with the reviewer that the differences in MS between day (midday) and night (nighttime) in OA factors are key findings of this paper. Currently, we discuss these differences in Sect. 3.3, by combining all POA factors into one POA factor and all OOA factors into one OOA factor in each period. We report the mass spectral correlations for the OOA factors in Figs. S25-S26 and MS comparisons in Figs. S31-S32 and S37-S38. Time-of-day PMF analysis is able to capture differences between day and night MS because it conducts PMF analysis for each period independent of the influence of variability in the other periods. This has been discussed in detail in Sect. 3.3, where the time-of-day PMF MS and TS are compared to seasonal PMF MS and TS.

In the revised manuscript, we have created a separate subsection to discuss the daytime and nighttime MS comparisons, and also added a brief description of the reasons for the differences. We have also added and discuss Figs. S39-S40, showing the MS correlations of the midday and nighttime POA factors across the two techniques, along with their correlations to reference MS profiles. We have updated the text (Sect. 3.4):

“However, this contrast is sharper in time-of-day PMF analysis, in line with the ability of the approach to capture variable MS (Figs. S31b and S32b; Fig. S39, Winter Time-of-day POA Spearman R: 0.93, Winter Seasonal POA Spearman R: 0.97; Fig. S40, Monsoon Time-of-day POA Spearman R: 0.81, Monsoon Seasonal POA Spearman R: 1.0). The seasonal PMF midday–night-time comparison also fails to capture the influence of cooking midday based on the low and similar ratio of contributions at  $m/z$  55 to  $m/z$  57 as night-time, esp. in monsoon ( $\sim 1$ , Figs. S31a and S32a). This contrast between midday and night-time POA MS is higher in time-of-day PMF in winter (midday ratio: 1.2, night-time ratio: 1.0, Fig. S31b) and in monsoon time-of-day PMF analysis (midday ratio: 1.4, night-time ratio: 1.2, Fig. S32b). While seasonal PMF analysis for monsoon suggests no change in MS between midday and night-time, time-of-day PMF analysis suggests large shifts in contributions at key  $m/z$ s such as 41, 43, 44, 55, and 57, in line with the changing importance of cooking from midday to night. These differences demonstrate the ability of time-of-day PMF to capture variable MS corresponding to the source influence of those time-of-day periods (Sect. 3.3.2).

We can also compare OOA MS and TS as well as conduct midday and night-time comparisons for time-of-day PMF and seasonal PMF analysis (Sect. S4). Time-of-day PMF OOA MS and TS are similar to seasonal PMF OOA (Table 3, TS: Pearson  $R > 0.95$ ; Figs. S25–S26 MS: Pearson  $R \geq 0.95$ ). However, the mass spectra of the time-of-day PMF OOA have major differences at  $m/z$  44 relative to the seasonal PMF OOA (Figs. S35–S36a–b). Comparisons of midday and night-time time-of-day PMF OOA MS shows interesting patterns not apparent in seasonal PMF analysis (Figs. S37–S38a–b). For example, time-of-day PMF analysis for monsoon 2017 suggests less oxidized OOA at midday than night-time, likely caused by the presence of semi-volatile compounds (Fig. S38b). Similar behaviour has been observed elsewhere as well, and was attributed to biogenic emissions (Canonaco et al., 2015).

Figure S42 shows all PMF factors obtained in this paper on the triangle plot (Ng et al., 2010). We observe that factors obtained in the time-of-day PMF analysis occupy a larger spread compared to those obtained in seasonal PMF analysis. For example, in time-of-day PMF POA factors, we observe a spread of about 5% in contributions at  $m/z$  43. In contrast, the spread of seasonal PMF POA factors is less than 3%. Overall, because time-of-day PMF conducts PMF analyses for each period independent of the influence of the variability in the other periods, it generates more representative MS for each time-of-day period (Sect. 3.3).”

Since we have created a new subsection, we have updated the text elsewhere as well (Sect. 3):

“In Sect. 3.4, we compare the midday and nighttime POA and OOA MS profile results from the seasonal PMF and the time-of-day PMF approach. Our hypothesis is that the time-of-day PMF approach will show larger variability across the two time periods. In



Sect. 3.5, we discuss period-specific  $Q$  (and  $Q/Q_{exp}$ ) values for the time-of-day PMF approach and the seasonal PMF approach. We also compare the  $Q/Q_{exp}$  TS patterns and  $Q/Q_{exp}$  by  $m/z$  to identify periods and  $m/z$ s with particularly significant changes in  $Q/Q_{exp}$ .”

*5. Zoom the legend in axis in Fig. S22-23 and S7-S8, so that the readers can see them clearly.*

Response: For accessing the expanded figures corresponding to these supplemental figures, we have provided corresponding Supplementary Files, which have larger axis-legends.

*5. Repeated descriptions: lines 216-217 and lines 159-162.*

Response: We have moved lines 159-162 to Sect. 2.4 and removed the repeated descriptions in lines 216–217 in the updated manuscript.

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