

Response to editor and reviewer comments

We thank the editor and reviewers once again for their valuable comments and suggestions. In the following, we provide our point-by-point responses and the corresponding revisions made to the manuscript.

In response to the editor's request, four minor points for further strengthening the manuscript are listed below, along with our corresponding revisions:

Comments #1:

Pre-holiday K/Ba fluctuations: Please briefly speculate on likely contributors (e.g., biomass burning vs. industrial emissions) based on co-varying PMF factors or other tracers, to sharpen the contrast with the fireworks signature.

In addressing the first point regarding K/Ba fluctuations and potential biomass burning influences, you may wish to consider whether a geographical shift in emissions during holiday periods - such as increased rural residential burning - could contribute to the observed patterns. This mechanism is discussed in a paper you have already cited (Li et al., 2025) for a different region, and its findings may offer a useful perspective for your discussion.

Response:

We thank the reviewer for this constructive suggestion. In response, we have added a short discussion to clarify the potential sources of the pre-holiday K/Ba fluctuations and to better distinguish them from the fireworks signal. As noted in the revised manuscript, before the fireworks episode the fluctuating K/Ba ratios were associated with consistently low Ba concentrations (average $0.01 \mu\text{g}/\text{m}^3$) and moderate contributions from the PMF-resolved biomass burning factor ($0.74 \mu\text{g}/\text{m}^3$, 1.6% of $\text{PM}_{2.5}$), suggesting that the variability was likely influenced by mixed combustion sources (e.g., biomass burning or residential combustion) rather than fireworks emissions. In contrast, chloride concentrations and the PMF-resolved fireworks factor increased sharply during the fireworks episode, highlighting the distinct chemical signature of fireworks emissions. Following the reviewer's suggestion, we also discuss the possibility that such pre-holiday variability may be related to a geographical shift in emissions during the Spring Festival period, such as increased rural residential combustion, as reported by Li et al. (2025). The corresponding discussion has been added in Lines 256-264 of the revised manuscript.

The revised text in the manuscript is as follows:

“Before the fireworks episode, the fluctuating K/Ba ratios were accompanied by consistently low Ba concentrations (average $0.01 \mu\text{g}/\text{m}^3$) and moderate contributions from the biomass burning factor, as identified by the PMF analysis presented later ($0.74 \mu\text{g}/\text{m}^3$, 1.6% of $\text{PM}_{2.5}$), indicating that the variability was likely influenced by mixed combustion sources (e.g., biomass burning or residential combustion) rather than fireworks emissions. Consistently, chloride concentrations and the fireworks factor remained low before the event but increased sharply during the fireworks episode,

reinforcing the distinction between the pre-holiday variability and the characteristic fireworks signature. This pattern may also be related to a geographical shift in emissions under the influence of the Spring Festival holiday-related activities, when increased rural residential combustion can influence urban air quality through regional transport, as discussed by Li et al. (2025).”

Comments #2:

Tracer enrichment vs. mass contribution: Briefly discuss potential mixing-state influences and clarify how tracer enhancement is interpreted in relation to mass attribution within the PMF framework.

Response:

We thank the reviewer for this new suggestion. To clarify the relationship between tracer enrichment and mass contribution within the PMF analysis, we have added a brief explanation in the revised manuscript. Specifically, we note that the enrichment of certain tracer elements does not necessarily correspond directly to the mass contribution of a given source factor. In PMF analysis, tracer species mainly serve as indicators for source identification, whereas the factor-resolved mass contributions represent the overall mixture of chemical species associated with each source profile. We also acknowledge the potential influence of particle mixing state, whereby fireworks-related tracer elements may coexist with secondary species (e.g., sulfate or nitrate). The corresponding discussion has been added at the end of the “Source apportionment results” section (Lines 220-226) in the revised manuscript.

The revised text in the manuscript is as follows:

“Moreover, the enrichment of specific tracer elements does not necessarily correspond directly to the mass contribution of a given source factor. Within the PMF modeling, tracer species primarily serve as indicators for source identification, whereas factor-resolved mass contributions reflect the overall mixture of chemical species associated with each source profile. In addition, atmospheric particles are often internally mixed, and tracer elements from fireworks emissions may coexist with secondary species such as sulfate or nitrate. Consequently, the enhancement of specific tracer elements should be interpreted primarily as evidence of source influence rather than as a direct proxy for the quantitative mass contribution of that source.”

Comments #3: Spatial representativeness of composition: Acknowledge possible spatial variability in chemical source signatures (given that fireworks tracers can show strong local heterogeneity) and discuss the implications for extrapolating single-site PMF results to the city scale.

Response:

The spatial representativeness of the single-site chemical observations has already been discussed in the previous version of the manuscript (Lines 368-383), where we explain why the Wolongqiao station can reasonably capture the city-scale fireworks signal

based on the regional nature of the event and the strong consistency between site-level and city-averaged PM_{2.5} observations. Following the reviewer's suggestion, we have further revised this section to explicitly acknowledge the potential spatial heterogeneity of fireworks-related tracers. In the revised manuscript, we now note that tracers such as K, Ba, and Cl may exhibit local variability due to localized burning activities. We therefore emphasize that, although the single-site measurements effectively capture the city-scale pollution signal during the fireworks episode, future studies with a more extensive chemical monitoring network would be valuable to better resolve potential fine-scale spatial variability. The corresponding revision has been added in Lines 382-385 of the revised manuscript.

The revised text in the manuscript is as follows:

“Here we acknowledge that due to the unavailability of multi-site chemical speciation data in Hangzhou, this study relies solely on speciation data from the Wolongqiao urban station. We contend that the data from this single site is sufficient and representative for validating the city-wide fireworks signal for two primary reasons. First, fireworks emissions during the CSF are not localized point sources but occur nearly simultaneously as an area-wide activity across the city. The resultant pollution plume, characterized by a sharp, synchronous peak in city-level PM_{2.5} (averaged from 14 sites), is a regional atmospheric phenomenon. Second, the Wolongqiao site's PM_{2.5} observations demonstrate a strong positive correlation with the city-wide average (e.g., $R^2 = 0.78$, $N = 1213$). This high consistency confirms that the site's temporal PM_{2.5} variation capture the pollution trends across Hangzhou, especially during the winter haze and Spring Festival season (Fig. S9). Additionally, the chemical fingerprint (e.g., distinct peaks in K, Ba, and the K/EC ratio) measured by the Wolongqiao site aligns perfectly with the timing of this regional PM_{2.5} peak identified by our machine learning model. This strong temporal consistency and physicochemical coherence provide compelling evidence that both methods are detecting the same city-scale pollution event, thereby supporting the representativeness of the single-site chemical data for this specific analysis. Nevertheless, we acknowledge that fireworks-related tracers (e.g., K, Ba, and Cl) may exhibit spatial heterogeneity due to localized burning activities. Future studies with a more extensive chemical monitoring network would be beneficial to further confirm these findings and capture potential finer-scale spatial heterogeneity.”

Comments #4: PMF factor optimization: Provide a brief comparison of Q/Q_{exp} values or bootstrapping results across different factor numbers to confirm that the nine-factor solution is optimal.

Response:

To further justify the selection of the nine-factor solution, we compared PMF solutions with different factor numbers (6-10 factors) using bootstrap diagnostics. The results are summarized in Table S3 in the Supplementary Information. The comparison shows that the nine-factor solution exhibits the highest bootstrap mapping stability (96-100%) with

no unmapped factors, indicating the most stable and interpretable factor structure among the tested solutions.

Accordingly, we have added the following explanation in the PMF factor identification section of the revised manuscript (Lines 190-193):

“An optimal nine-factor solution was obtained, with small, normally distributed residuals, demonstrating good model performance (Table S3). The robustness of this solution was further validated through bootstrap diagnostics across solutions with six to ten factors. Specifically, the nine-factor solution exhibited the highest bootstrap mapping stability (96-100%) and no unmapped factors, confirming it as the most stable and interpretable among all tested solutions.”

Table S3. Comparison of bootstrap diagnostics for PMF solutions with different factor numbers (6-10 factors). The nine-factor solution shows the highest bootstrap mapping stability (96-100%) with no unmapped factors. In contrast, solutions with fewer factors tend to mix multiple sources, whereas the 10-factor solution shows reduced stability and limited improvement in model performance. These results indicate that the nine-factor solution is the most stable and interpretable among the tested solutions.

BS diagnostics	6-factor	7-factor	8-factor	9-factor	10-factor
Lowest %BS mapping	91	94	94	96	94
Highest %BS mapping	100	100	100	100	97
Highest unmapped	1	0	0	0	0