Point-to-point responses

Reviewer #1

This manuscript presents a comprehensive source apportionment study of non-methane hydrocarbons (NMHCs) in Taiwan's Kaoping region using high-resolution year-round measurements from three Photochemical Assessment Monitoring Stations (PAMS). Positive Matrix Factorization (PMF) was applied to resolve eight distinct NMHC source factors, complemented by Conditional Probability Function (CPF) analysis and a novel "triggered backtrajectory" approach based on episodic peaks in PMF time series. The study finds that petroleum and mixed (vehicular/solvent) sources dominate NMHC contributions and ozone formation potential (OFP), with significant influence even during moderate ozone days.

The manuscript is timely, methodologically innovative, and policy-relevant. It provides strong observational evidence and demonstrates a refined framework for linking receptor modeling with spatial source attribution. However, there are some improvements necessary regarding methodological uncertainty and interpretive depth, which should be addressed before publication. This amounts to a minor revision.

Author's response: On behalf of the co-authors, we appreciate your constructive comments on improving our manuscript. All the comments have been read through, and responses have been made accordingly. The detailed responses are as follows. Revisions made to the manuscript in response to these comments are indicated in Red.

1. Dependence on PMF and CPF assumptions

o The analysis relies on PMF, which is sensitive to input selection, number of factors, and uncertainty estimates. While stability tests are mentioned, the details of these test are not given and unclear what test are done (e.g., factor rotations, influence of excluded low-S/N species). This would strengthen confidence in the robustness of source identification.

Author's response: In this study, uncertainty estimation followed the U.S. EPA PMF 5.0 protocol, incorporating species-specific analytical error fractions and MDL-based rules (below MDL: ½ MDL concentration, ⅚ MDL uncertainty). Low S/N species (with ratio S/N < 0.1) were excluded, while weak species (0.1 ≤ S/N < 2) were down-weighted to prevent instability from low-quality inputs. The stability of PMF solutions was examined through multiple diagnostics, including 100-run random seed tests for each factor number (from 3 to 8), evaluation of Q(robust)/Q(true) ratios, and interpretability checks. Additional Bootstrap (BS) tests were conducted to assess factor stability. The selected eight-factor solution showed consistent profiles reproduced in over 95% of bootstrap runs with Q(robust)/Q(true) ratios that is close to 1, confirming the robustness of the source identification. The related text in the method section has been revised to enhance the readability. Please see lines **196-215.**

Revised text: "In this study, we applied the U.S. EPA's PMF 5.0 software to perform source apportionment of NMHCs measured at the three PAMS sites in the Kaoping region. The input to the model consisted of concentration and uncertainty matrices constructed from hourly NMHC measurements. Uncertainty (Uij) was calculated based on species concentrations (Xij) and minimum detection limits (MDL) as follows:

$$U_{ij} = \sqrt{\left(0.5 \times MDL_j\right)^2 + \left(error\ fraction \times X_{ij}\right)^2} \quad (1)$$

For concentrations below MDL, the value was substituted with half MDL with uncertainty set at $\frac{5}{6}$ MDL, and an error fraction (10%). Missing values were excluded from the input dataset to maintain model reliability. To ensure a consistent and robust dataset across the three sites and four seasons, species selection was based on signal-to-noise (S/N) ratios and detection frequency following EPA PMF guidance. Specifically, species with S/N ratios < 0.1 or missing data with occurrences more than 4 instances were classified as "bad" and excluded, while those with $0.1 \leq S/N < 2$ were down-weighted as weak species. This screening process resulted in a final set of 22 out of the 54 species that were consistently detectable and quantitatively reliable across all sites and seasons (Table S1). Model stability was then evaluated through multiple diagnostic procedures. First, factor numbers from 3 to 8 were tested, each with 100 independent runs using random seed initialization. The optimal number of factors was selected based on $Q_{(robust)}/Q_{(true)}$ values that were close to 1.0, reproducibility of factor profiles across runs assessed via Bootstrap (BS) analyses (>95% matching), and the interpretability and physical plausibility of the resulting source profiles. Together, these procedures confirm that the PMF solution provides a robust and well-constrained representation of NMHC sources in the study region, while the consistent selection of 22 species ensures comparability across all seasonal and site-specific analyses."

o The model stability of the CPF could be visualized.

Author's response: Thank you for your constructive comment. While the 0.75 quantile is commonly used and generally robust for CPF analysis at urban or industrial sites (Pekney et al., 2006; Chen et al., 2019; Wu et al., 2024; Huang and Hsieh, 2019), our sensitivity tests indicated that it was not the optimal threshold in our case. Using one full year of hourly NMHC observations, we compared CPF results across several percentile thresholds, from 0.60 to 0.85. Acetylene was used as a reference factor because its potential source direction—originating from the northern area—is well established, providing a reliable basis for evaluating CPF stability. The CPF patterns remained relatively consistent between the 0.60 and 0.70 quantiles but began to degrade at higher thresholds. Specifically, CPF values dropped sharply and directional features became less meaningful above the 0.75 quantile, as the number of high-percentile events per wind sector decreased substantially, reducing statistical reliability. We therefore selected the 0.70 quantile as an optimal solution. This threshold effectively filters out moderate events, focuses on high-impact episodes, and retains sufficient samples per sector for robust and statistically stable CPF estimation. Although higher thresholds (0.75–0.80) emphasize more episodic or localized plumes, in our dataset they provided too few valid samples for reliable directional inference. The 0.70 quantile thus balances reliability and selectivity while preserving the major CPF directional features relevant for interpretation. The CPF method section was accordingly revised to clarify this selection (see lines **223-231**), and the sensitivity test results are provided in Table S2 of the Supplementary Materials.

Revised text: "CPF was computed as the ratio of the number of times the factor contributions exceeded the threshold within a given wind sector ($n_{\Delta\theta}$) to the total number of valid observations in that sector ($m_{\Delta\theta}$). Wind direction was divided into 16 equal intervals (22.5° per sector) to ensure robust analysis. For each PMF-resolved factor, the 70th percentile was the threshold to isolate the plume events, as it effectively filtered out moderate events while retaining sufficient data for statistically stable and interpretable CPF results (Table S2). Higher CPF values in specific wind sectors indicated stronger contributions from sources in that direction. CPF plots were generated for each PMF factor to visualize dominant source directions and assess consistency with known emission source locations, meteorological patterns, and local topography."

"Table S2. Sensitivity of CPF directional patterns to percentile thresholds. Acetylene, whose source direction is well established as primarily originating from the northern industrial area, was used as a reference factor for this evaluation. CPF patterns were consistent between the 0.60 and 0.70 quantiles but became unstable above 0.75, as higher thresholds yielded fewer valid samples per sector and less coherent directional features. Therefore, the 0.70 quantile was determined to be the most appropriate solution, effectively filtering out moderate events while retaining sufficient data for statistically stable and interpretable CPF results."

Wind_sector	CPF-0.6	CPF-0.65	CPF-0.7	CPF-0.75	CPF-0.8	CPF-0.85
1	1	1	1	1	1	0.895
2	0.803	0.799	0.799	0.747	0.631	0.486
3	0.379	0.379	0.379	0.268	0.172	0.111

4	0.202	0.191	0.169	0.135	0.079	0.034
5	0.140	0.116	0.093	0.093	0.093	0.047
6	0.233	0.178	0.068	0.041	0.027	0.014
7	0.287	0.207	0.069	0.023	0.011	0.011
8	0.319	0.181	0.026	0	0	0
9	0.595	0.214	0.048	0	0	0
10	0.308	0.192	0.077	0.077	0.038	0.038
11	0.367	0.306	0.245	0.143	0.122	0.082
12	0.181	0.111	0.056	0.028	0.014	0.014
13	0.170	0.112	0.076	0.054	0.036	0.022
14	0.545	0.545	0.545	0.500	0.455	0.364
15	0.900	0.900	0.900	0.900	0.800	0.700

o The inability to fully separate vehicular and solvent sources is acknowledged. However, this limitation has important implications for regulatory application and should be discussed more explicitly in the context of control strategies.

Author's response: We have expanded the discussion in the OFP section to address more about the implications of incomplete separation between vehicle and solvent sources. Please see lines **704-714**.

Revised text: "Under moderate-ozone conditions, the mixed-source factor contributes the largest share of OFP. These episodes are frequently associated with lower wind speeds and reduced mixing heights (Aacog, 2015), favoring the accumulation of locally emitted species from traffic, solvents, and smaller-scale industrial activities. The coexistence of these emissions provides a balanced supply of reactive aromatics and olefins, sustaining ozone production even in the absence of strong photochemical aging. This pattern reflects the VOC-limited photochemical environment prevalent in southern Taiwan, where ozone formation is more sensitive to reactive VOCs than to NOx levels (Chang et al., 2022), emphasizing that moderate-ozone episodes are primarily governed by local accumulation and NMHC composition. However, the mixed-source factor reflects overlapping characteristics of vehicular and solvent-related species. This incomplete separation introduces some uncertainty in source-specific attribution of OFP. Nevertheless, this overlap mirrors the reality of urban environments."

2. Treatment of Uncertainty

Uncertainty quantification appears limited. For example, details of the OFP estimates are not shown or explained and the specific chemistry assumptions are not mentioned. Needs to be added.

Author's response: We thank the reviewer for pointing this out. In the revised manuscript, we have added a description of the OFP estimation method, including the chemical mechanism, and a description of the uncertainty. Please see lines: **243-258**.

Revised text: "2.5 Ozone formation potential and uncertainty consideration

The OFP of each NMHC species was estimated using the Maximum Incremental Reactivity (MIR) coefficients developed by (Carter, 2010). The OFP for compound *i* was calculated as:

$$OFP_i = C_i \times MIR_i \quad (3)$$

Where C_i (ppb) is the measured mixing ratio of the species, and MIR_i (g O_3 g^{-1} VOC) is its reactivity coefficient (Table S1). The MIR scale represents ozone yield under low VOC/NO_x (i.e., high-NO_x or VOC-limited) conditions, where ozone formation is primarily sensitive to changes in VOC abundance. This assumption aligns with previous photochemical studies indicating that ozone formation in southern Taiwan is predominantly VOC-limited (Chang et al., 2022).

The uncertainty associated with OFP estimation was inherently accounted for during the PMF analysis. These uncertainties were used to construct the PMF input uncertainty matrix, which determines the weighting of each data point in the model fitting. As the OFP was calculated from the PMF-resolved factor contribution time series, the measurement uncertainties are inherently reflected in the factor contributions. While this approach propagates measurement uncertainty into OFP estimates, it is important to note that OFP represents a photochemical potential, not realized ozone formation, and does not capture the nonlinear interactions that govern actual ozone production."

3. Interpretation of Moderate Ozone Days

The finding that mixed sources dominate OFP under moderate ozone conditions is highly relevant, but the mechanistic explanation is underdeveloped. Are these results consistent with VOC-limited regimes? How do meteorological conditions (e.g., mixing height) shape these patterns? Expanding the discussion would enhance both scientific and policy relevance.

Author's response: We thank the reviewer for this valuable comment. We have enhanced the discussion of OFP patterns under moderate-ozone conditions (Section 3.6, lines **704-714**). The revised text now provides a clearer mechanistic explanation linking the dominance of mixed sources to local photochemical regimes and meteorological influences. Specifically, we added the following points:

a-Mechanistic explanation of photochemical regimes under moderate ozone days:

Southern Taiwan, particularly Kaohsiung, is characterized by a VOC-limited ozone formation regime (Chang et al., 2022). Under such conditions, ozone production is more sensitive to changes in VOC reactivity than in NOx. Therefore, the dominance of mixed sources—which combine traffic, solvent, and industrial VOCs with moderate-to-high MIR values—during moderate ozone episodes is consistent with this chemistry. Small increases in reactive VOCs from these sources can effectively enhance ozone production.

b-Meteorological conditions:

Moderate ozone days typically occur under weak horizontal transport and low-to-moderate mixing height conditions (Aacog, 2015), leading to the accumulation of local VOCs. As a result, the lower boundary layer and limited ventilation during moderate ozone days enhance the influence of locally mixed sources with highly reactive species on OFP.

c-Implications for control strategy:

These findings suggest that under typical VOC-limited conditions, reductions in mixed-source emissions—particularly aromatics and light alkenes—could yield more effective ozone mitigation during local accumulation events, whereas regional transport events would require coordinated upwind emission control.

Revised text: "Under moderate-ozone conditions, the mixed-source factor contributes the largest share of OFP. These episodes are frequently associated with lower wind speeds and reduced mixing heights (Aacog, 2015), favoring the accumulation of locally emitted species from traffic, solvents, and smaller-scale industrial activities. The coexistence of these emissions provides a balanced supply of reactive aromatics and olefins, sustaining ozone production even in the absence of strong photochemical aging. This pattern reflects the VOC-limited photochemical environment prevalent in southern Taiwan, where ozone formation is more sensitive to reactive VOCs than to NOx levels (Chang et al., 2022), emphasizing that moderate-ozone episodes are primarily governed by local accumulation and NMHC composition. However, the mixed-source factor reflects overlapping characteristics of vehicular and solvent-related species. This incomplete separation introduces some uncertainty in source-specific attribution of OFP. Nevertheless, this overlap mirrors the reality of urban environments."

Author's response: As the discussion on OFP is further enhanced, the related text in the conclusion has been revised accordingly. Please see lines: **763-786.**

Revised text: "In addition, this study explored the dynamics of OFP. They were calculated specifically for daytime periods—when photochemical activity is most pronounced. Seasonally averaged OFP was highest at Xiaogang (113.20 ± 23.60 μg/m³), followed by Linyuan (102.73 ± 40.93 μg/m³), and lowest at the downwind rural site Chaozhou (65.38 ± 9.00 μg/m³). Although petroleum-related sources contributed the largest fraction of NMHC concentrations, the mixed source factor—enriched in highly reactive species such as aromatics and olefins—often dominates the OFP, particularly at Xiaogang. Under moderate-ozone conditions (MDA8 40–60 ppb), the factor became the principal driver of ozone formation, consistent with local accumulation under stagnant meteorological conditions and limited vertical mixing. The coexistence of both reactive aromatics and olefins within this factor sustained ozone production despite weak photochemical aging, reflecting a VOC-limited regime typical of southern Taiwan. Across ozone pollution levels, petroleum and mixed sources remained dominant, but their relative influence varied with site characteristics. Mixed sources exerted stronger effects during moderate-ozone episodes at urban–industrial locations, whereas petroleum-related sources dominated in Linyuan under similar conditions. These results suggest that frequent, moderate-ozone episodes are primarily driven by locally accumulated reactive NMHCs. This pattern may also reflect the early impacts of emission control measures, which are more effective under high-pollution conditions but less so during moderate episodes. Given their higher occurrence, moderate pollution episodes still offer valuable insights into the interplay between local accumulation, VOC reactivity, and emission composition that governs ozone formation in the region.

Overall, this study demonstrates the strength of a refined source apportionment approach using multi-site, year-round, high-frequency NMHC measurements, each reflecting distinct source–receptor dynamics. The findings offer a more comprehensive spatiotemporal understanding of ozone formation mechanisms and provide a scientific basis for coordinated control of mobile, solvent-related, and petroleum-associated emissions across southern Taiwan."

Technical Comments

1. Abstract and Summary

o The abstract is long and technical. Consider reducing methodological detail in favor of emphasizing findings and policy implications.

Author's response: We thank for your suggestion. The abstract has been revised significantly as follows.

Revised text: "Ozone pollution remains a persistent challenge in Taiwan's Kaoping region due to dense industrial and urban emissions. Using high-resolution, hourly observations from three Photochemical Assessment Monitoring Stations (PAMS) combined with Positive Matrix Factorization (PMF), this study resolved eight distinct sources of non-methane hydrocarbons (NMHCs), key precursors of ozone formation. The time-resolved PMF output captured source-specific temporal patterns—as shown by the acetylene factor at Linyuan ($R^2 = 0.99$ with observations)—providing an intrinsic check on model performance and facilitating spatial interpretation of sources. Petroleum-related emissions dominate NMHC mass at all sites but are more prominent at Xiaogang, while aged air masses substantially enhance ozone pollution at downwind locations such as Linyuan and Chaozhou, underscoring the role of regional transport and atmospheric aging. Although mixed sources from vehicular and solvent emissions contributed less mass, they dominated ozone formation potential (OFP) due to higher chemical reactivity. Notably, their influence persisted even under moderate ozone conditions, indicative of a VOC-limited regime. Overall, these results emphasize that effective ozone mitigation in southern Taiwan requires coordinated control of petroleum, mobile, and solvent emissions, and demonstrate the value of multi-site, year-round, high-time-resolution NMHC measurements for constraining ozone precursor."

2. Figures

o Some figures (e.g., Fig. 3, Fig. S3) are very information-heavy. Simplifying or providing summary schematics could aid readability.

Author's response: We acknowledge the reviewer's concern. However, these figures present essential information that cannot be further simplified without losing key analytical details. To enhance readability and self-explanation, we have refined the figure captions to help readers better interpret the results.

Revised text: "**Figure 3:** Summary of common source profiles of NMHC at the three sites in 2024. This figure presents the percentage contribution of six common source factors (Petro I, Petro II, Refinery, Industrial fugitive emissions, Mixed, and Aged air mass) to the total NMHC burden at three monitoring sites: Xiaogang (red), Linyuan (orange), and Chaozhou (green) across four seasons (Winter, Spring, Summer, and Fall). Each panel represents a specific source factor and season combination. The stacked bars within each panel show the relative contribution of NMHC species to that source factor. Consistent source profiles of fingerprint species were observed across seasons and at all three monitoring sites, underscoring the robustness of the PMF results."

"**Figure S4** Seasonal PMF-resolved factor profiles at the study sites (a) winter, (b) spring, (c) summer, and (d) fall. Panels correspond to PAMS sites. Percentage contributions reflect relative patterns within each site; therefore, similar percentages do not necessarily correspond to similar absolute acetylene levels or emission characteristics. Species contribution in factor profile is denoted by the red dot."

"**Figure S5** Seasonal variation of daily mean contributions of NMHC concentration from PMF-resolved factors. This figure illustrates the daily variability of identified source factors (Petro I, Petro II, Refinery, Industrial fugitive emissions, Mixed, Aged air mass and Acetylene) over the course of a year at three monitoring sites: Xiaogang (red), Linyuan (orange), and Chaozhou (green). To interpret this figure, follow the trends of the lines for each site within each season to observe how a specific source factor changes over time and reveal episodic emission events that indicate the changes in source activity. Shaded columns represent weekends. "

o S7 legend not readable, S5a vertical axis units are missing.

Author's response: There were errors in these figures, and corrections have been made. The legend in Figure S7 has been updated for clarity which is now Figure S10, and a vertical axis label has been added to Figure S5a which is now Figure S7a

3. Literature Context

o While many relevant studies are cited, the discussion could further situate the work in the context of ozone regime sensitivity studies (VOC- vs. NOx-limited conditions), which are critical for interpreting the results.

Author's response: We have added text to further situate the work in the context of ozone regime sensitivity studies (VOC- vs. NOx-limited conditions), which are aligns with the interpreting of our OFP results. Please see lines: **39-50.**

Revised text: "The impact of VOCs on ozone formation critically depends on the local chemical regime—whether ozone production is VOC-limited or NO_x-limited (Kleinman et al., 2002; Sillman, 1999). In VOC-limited environments, common in densely industrialized and urbanized areas, ozone levels are more responsive to changes in reactive VOC concentrations, whereas in NO_x-limited conditions, ozone formation is constrained by nitrogen oxide availability. Several studies in East and Southeast Asia have emphasized this spatial heterogeneity of ozone sensitivity (Li et al., 2019; Wang et al., 2021; Ren and Xie, 2022). In Taiwan, both modeling and observational evidence indicate that southern and western regions typically exhibit VOC-limited or transition regimes, while rural and downwind areas are more NO_x-limited (Chang et al., 2022; Chen et al., 2021). This regime dependence underscores the need for region-specific precursor management and highlights the importance of identifying the dominant reactive VOC sources that most effectively drive ozone formation. Understanding these sensitivities provides an essential framework for interpreting ozone formation potential (OFP) derived from NMHC source contributions."

Recommendation: Minor revision

This is a strong and innovative paper that makes a meaningful contribution to atmospheric chemistry and air quality management. However, addressing the above issues—particularly uncertainty analysis, interpretation regarding ozone formation under moderate conditions, and clearer presentation of policy-relevant results—would further strengthen the manuscript.

Author's response: Thank you for your positive assessment of our work. We appreciate your insightful comments and have addressed all of your concerns with revisions to the manuscript. We believe these changes have significantly strengthened the manuscript.

Reviewer #2

The study by Nguyen et al. investigates NMHCs using long-term, multi-site measurements in southern Taiwan. By applying PMF-based seasonal source apportionment, the authors attempt to resolve the complex mixture of emissions using a 22-species input matrix that produces eight factors. They complement the PMF analysis with directional information from the Conditional Probability Function and a "triggered back-trajectory" approach based on episodic factor spikes They also estimate the ozone formation potential (OFP) of the resolved factors, analyse seasonal and spatial variability, and conclude that petroleum-related activities dominate the NMHC burden. Such long-term, multi-site datasets are valuable for the literature as they enable robust receptor modelling, provide better-defined PMF profiles, and capture seasonality and episodic events. In this occasion they also support policy-relevant assessments of emission sources affecting local air quality and ozone formation. Overall, this is an interesting study, and I could recommend acceptance in ACP only if the following issues are fully addressed.

Author's response: On behalf of the co-authors, we appreciate your constructive comments on improving our manuscript. All the comments have been read through, and responses have been made accordingly. The detailed responses are as follows. Revisions made to the manuscript in response to these comments are indicated in Red.

General comments

1. The attribution of the so-called gasoline evaporation factor is likely incorrect for two reasons. First, the reported pentane ratios do not match established literature patterns. In the submitted factor profile, the ratio appears reversed relative to typical gasoline evaporation signatures and is more consistent with oil and gas operations. Second, Supplement Fig. S2 shows a markedly different profile at Chaozhou, especially in winter, which further challenges the assignment. The authors should reconsider the interpretation of this factor and revise the respective subsection. Examining correlations with temperature and diel cycles could help clarify the source origin.

Author's response: We appreciate the reviewer's observation. We initially assigned this factor as 'gasoline evaporation' because it shows overall elevation of pentanes, a steady fugitive evaporative emissions consistent with Kaoping source characteristic as it hosts of extensive petrochemical operations, gasoline storage and transfer infrastructure, fuel storage terminals, etc. However, after re-examining the ratio, we agree that the original interpretation as "gasoline evaporation" requires revision. Specifically, the PMF-resolved profile yielding an i-pentane/n-pentane ratio < 1. This ratio is inconsistent with established gasoline evaporation signatures, which typically range from 1.5 to 3.0 (Baker et al., 2008; Gentner et al., 2009). Conversely, the observed ratio aligns well with oil and gas fugitive emissions and petrochemical processing sources, where ratios of 0.8–1.0 are commonly reported (Gilman et al., 2013; Bourtsoukidis et al., 2019). However, oil and gas operations are upstream activities, while Kaoping host downstream operations such as refining crude oil and producing petrochemical feedstocks. Because of that, the initial gasoline evaporation factor was revised into industrial fugitive emission, likely originating from petrochemical processing, storage tanks, and refinery-associated leakage. We have revised related Section 3.3.1.c accordingly and now interpret this factor as representing industrial fugitive emissions. Please see lines **384-415.**

Author's response: After revising the original gasoline evaporation factor to industrial fugitive emissions factor, the markedly different profile observed at Chaozhou—particularly in winter—can be reasonably explained by the distinct seasonal atmospheric processes and spatial characteristics of the Kaoping region. The seasonal variability reflects the interplay between local fugitive releases (most influential in winter) and transported fugitive emissions from the upstream industrial corridor (dominant in spring, summer, and fall). Chaozhou generally exhibits lower PMF-resolved contributions (Fig. S5) and more diluted profiles, consistent with its downwind location relative to major industrial sources. However, in winter, the factor rises to levels comparable with Xiaogang, which is attributable to local accumulation under stable boundary-layer conditions; even then, the species pattern remains consistent with a processed industrial fugitive signature rather than a distinct source type.

During spring and fall, the temporal profiles are weaker and compositionally diluted, and the CPF results (Fig. 4) show a stronger influence from the western sector (Linyuan/Xiaogang). This agrees with the regional transport of fugitive alkane emissions that become mixed and diluted as they move toward Chaozhou. In summer, the PMF-resolved contributions are minimal, and the resolved profile is diffuse, reflecting efficient atmospheric dispersion and reduced accumulation under well-mixed, warm-season conditions. Overall, the combined seasonal, spatial, and directional evidence consistently supports the assignment of this factor as industrial fugitive emissions, despite the distinct winter profile at Chaozhou. Please see lines **414-424.**

Revised text: "Isopentane and n-pentane are recognized tracers of natural gas operations (Gilman et al., 2013) or gasoline vapor emissions (Gentner et al., 2009). In our PMF results, both species showed elevated concentrations, indicating a strong contribution from fugitive sources (Fig. 3). Because isopentane and n-pentane exhibit similarly OH reaction rates (Atkinson, 1986), their ratio (iC_5/nC_5) is commonly used to identify emission sources (Bourtsoukidis et al., 2019). Accordingly, we examined this ratio across all sites using the PMF-resolved concentration of species from factor profile output to reveal their distinct relationship between isopentane and n-pentane.

Many studies report that an iC_5/nC_5 ratio in the range of approximately 0.8–1.1 is characteristic of oil & natural gas operations or raw-gas emissions (Gilman et al., 2013; Thompson et al., 2014; Swarthout et al., 2013; Gilman et al., 2010). In our analysis, the iC_5/nC_5 ratio was consistently <1 across the sites and seasons, with site-specific seasonally mean values of 0.53 ± 0.02 at Linyuan, 0.76 ± 0.06 at Xiaogang, and 0.87 ± 0.10 at Chaozhou, all within or below the typical oil & natural gas range. The Kaoping region hosts only downstream industrial activities—such as refining, petrochemical processing, and feedstock production—rather than upstream extraction. Therefore, this factor was attributed to industrial fugitive emissions, likely originating from petrochemical processing units, storage tanks, and refinery-related leakage. The lowest ratio at Linyuan reflects a very local, fresh fugitive leak from proximate refinery/petrochemical sources. In contrast, the downwind receptor (Chaozhou) or the mixed urban-industrial area (Xiaogang) can result in higher ratios.

The PMF-resolved time series of industrial fugitive emission at all three sites—Xiaogang, Linyuan, and Chaozhou—exhibit a stable, low-level temporal pattern characteristic of continuous industrial fugitive emission sources across all seasons (Fig. S5). Occasional minor peaks occurred, with slightly higher values at Xiaogang and Linyuan than at Chaozhou at times, but no major episodic events were observed. This pattern underscores the nature of fugitive pollution, which remains a minor and relatively steady contributor to ambient NMHC levels year-round.

CPF analysis further reveals distinct spatial and seasonal patterns. At Linyuan and Xiaogang, elevated CPF values are associated with winds from the N-NW, and SE-SSE sectors, respectively, suggesting that elevated concentrations are most likely to occur under these prevailing wind conditions (Fig. 4). Winter and spring generally show slightly higher CPF values compared with summer and fall, particularly at these industrial sites, reflecting seasonal variations in atmospheric transport or emission dynamics. ” (lines **384-413)**

“In contrast, Chaozhou consistently displays low CPF values and the lowest NMHCs abundance across all seasons and wind directions, reflecting its rural setting, lack of industrial activity, and its downwind position relative to Xiaogang and Linyuan. The combination of weaker PMF-resolved timeseries (Fig. S5) and nearly uniform ratios of pentane isomers (0.87 ± 0.10) indicates that fugitive emissions reaching Chaozhou are largely diluted and regionally transported rather than locally generated. The winter enhancement is also consistent with the compositionally altered profile observed at Chaozhou (Fig.S4a), where atmospheric processing during transport and seasonal stagnation modifies the alkane distribution while retaining the broader characteristics of industrial fugitive emissions. Overall, the temporal and directional indicators confirm that Chaozhou is primarily influenced by diluted, downwind industrial fugitive emissions, with local contributions becoming detectable only under wintertime stagnation." (lines **414-424)**

2. The manuscript relies heavily on PMF-based interpretation, but does not provide simple, supporting relationships with routinely measured trace gases such as CO, NO, NO2 or CH4. These correlations are standard practice for validating factor attribution. Without them, confidence in several of the source assignments remains limited and the scientific analysis weaker than it could be.

Author's response: We thank the reviewer for highlighting the potential utility of trace gases for supporting PMF-based source identification. In our study, we did acknowledge and conduct some test PMF runs incorporating CO to evaluate its usefulness. However, as several key vehicle indicator species were missing from our dataset—due to unexpected data gaps or undetected (e.g., 2,4,4-trimethylbenzene, 1,3-butadiene, nonane, undecane). As a result, such as tracer gas of CO were not sufficient to establish a stable PMF solution and were therefore excluded from the final analysis. Despite these limitations, our PMF results produced highly stable factors that were consistent across sites and seasons, with bootstrap results higher than 95%. While the mixed factor representing both vehicle emissions and solvent usage could not be fully resolved for the 2024 dataset, its presence remains informative. This mixed factor highlights overlapping anthropogenic sources and underscores gaps in the current emission inventories, which we have emphasized in the revised manuscript.

Moreover, our PMF analysis accurately reproduced the temporal pattern of the acetylene-related factor (section 3.4, line 610), demonstrating the robustness and reliability of our approach. The ability to capture these temporal variations reflects the high quality of our PAMS dataset and supports confidence in the factor interpretation.

Overall, while routinely measured trace gases are generally valuable for PMF analyses (Huang and Hsieh, 2019), in our study region their effectiveness is limited by overlapping sources and the absence of several vehicle-specific tracers. We have included this discussion as a limitation in the revised manuscript to clarify the constraints of our source apportionment.

Revised text: "A major limitation in separating these sources is the absence of key tracers, such as 2,2,4-trimethylpentane (Huang and Hsieh, 2019), 2,3-dimethylbutane as recognized tracer of motor vehicle exhaust (Chang et al., 2004), or such as methyl tert-butyl ether (MTBE), as an indicator for vehicle exhaust or evaporation (Rubin et al., 2006; Chang et al., 2003; Lin et al., 2005), which makes it challenging to fully separate and resolve vehicle emissions. Another limitation is that CO, a commonly used tracer of combustion sources (Huang and Hsieh, 2019), was considered in the test PMF runs; however, its diagnostic value was restricted in the Kaoping region because multiple overlapping CO sources (e.g., traffic, industrial, residential burning, etc.) all contributed to elevated CO levels. As a result, the PMF model grouped vehicles and solvent-related emissions into a single mixed factor, reflecting the complex emission environment and overlapping NMHC signatures." (lines **432-442)**

3. The PMF analysis uses 22 NMHC species out of the full suite measured, but the manuscript does not explain the criteria for exclusion, nor does it provide any sensitivity analysis demonstrating that the eight-factor solution is robust to species selection. The selection of these species should be better justified.

Author's response: We have revised the manuscript to clearly describe how to select the 22 NMHC species. Because the study relied on PAMS data from three sites and conducted PMF analyses separately by season to improve policy-relevant insights, a consistent species list across all PMF runs was required. Following EPA PMF guidance, we performed a signal-to-noise (S/N) evaluation on all 54 species using 12 seasonal records (3 sites × 4 seasons). Species with more than 4 occurrences of missing data or S/N < 0.1 were classified as "bad" and excluded. This process resulted in 22 species that were consistently detectable and quantitatively reliable across all stations and seasons. This approach ensured that the PMF solution was based on a robust and internally consistent dataset, avoiding introducing artificial season- or site-dependent biases caused by inconsistent species availability. The revised text incorporates these updates in response to Reviewer #1, further clarify the selection of the 22 species, as read below.

Revised text: "For concentrations below MDL, the value was substituted with half MDL with uncertainty set at $\frac{5}{6}$ MDL, and an error fraction (10%). Missing values were excluded from the input dataset to maintain model reliability. To ensure a consistent and robust dataset across the three sites and four seasons, species selection was based on signal-to-noise (S/N) ratios and detection frequency following EPA PMF guidance. Specifically, species with S/N ratios < 0.1 or missing data with occurrences more than 4 instances were classified as "bad" and excluded, while those with 0.1 ≤ S/N < 2 were down-weighted as weak species. This screening process resulted in a final set of 22 out of the 54 species that were consistently detectable and quantitatively reliable across all sites and seasons (Table S1). Model stability was then evaluated through multiple diagnostic procedures. First, factor numbers from 3 to 8 were tested, each with 100 independent runs using random seed initialization. The optimal number of factors was selected based on $Q_{(robust)}/Q_{(true)}$ values that were close to 1.0, reproducibility of factor profiles across runs assessed via Bootstrap (BS) analyses (>95% matching), and the interpretability and physical plausibility of the resulting source profiles. Together, these procedures confirm that the PMF solution provides a robust and well-constrained representation of NMHC sources in the study region, while the consistent selection of 22 species ensures comparability across all seasonal and site-specific analyses." (lines **201-215)**

Specific comments

4. Consider a less technical title. The current one includes 3 acronyms and is difficult to follow.

Author's response: The title has been improved accordingly

Revised text: "Multi-Site Non-Methane Hydrocarbon Source Apportionment and Ozone Insights in Southern Taiwan Using Positive Matrix Factorization"

5. L11-15 & 35-41. Key points, short summary and keywords are not required for ACP submissions.

Author's response: All these sections are removed from the manuscript

6. L44. Consider adding a more widely adopted publication (e.g. https://doi.org/10.1080/1073161X.1993.10467187

Author's response: Thank you for the suggestion. We have included this new citation in the suggested sentence. (line 29)

7. L92. Please define PMF at its first appearance.

Author's response: Since the title has been revised to remove the acronyms, the full definition of PMF is in the abstract at its first appearance. (line 14)

8. L145-146. Note that agricultural emissions are classified as anthropogenic by the IPCC.

Author's response: We appreciate the reviewer's comment. The related sentence has been revised to be more clarified.

Revised text: "Chaozhou, by contrast, is more inland and characterized by agricultural and vegetative land use, with minimal influence from industrial and urban anthropogenic sources." (line 149)

9. Ch2.2. The sampling method requires clearer description, especially because potential inlet artifacts could alter the chemical composition and hence, PMF profiles. The flow rates are very low, and there is no information on the use of an ozone scrubber or a Nafion dryer. Long residence times in the inlet tube can enhance wall interactions, and the absence of humidity control means water vapour will influence FID peak responses in a disproportional way for different NMHCs.
Author's response: We have further clarified the description of Taiwan PAMS system.

Revised text: "Ambient air is sampled at a flow rate of 15 mL/min over 40 minutes, yielding approximately 600 mL of air per sample, and then passes through a Nafion dryer (500 sccm counter-flow) to eliminate excess humidity during sample collection. No ozone scrubber is needed, as the system features short, inert-coated sampling tubing, effectively minimizing the likelihood of ozone reactions." (lines 164-167)

10. Ch. 3.2.1.c. As noted in the first general comment, the pentane-based interpretation of the "gasoline evaporation" factor is not consistent with established literature (eg https://acp.copernicus.org/articles/19/7209/2019/ and references therein). Additional analysis is needed to clarify this factor.
Author's response: We have revised related Section 3.3.1.c accordingly and now interpret this factor as representing industrial fugitive emissions. Related text as read in lines 384-401.

Revised text: "Isopentane and n-pentane are recognized tracers of natural gas operations (Gilman et al., 2013) or gasoline vapor emissions (Gentner et al., 2009). In our PMF results, both species showed elevated concentrations, indicating a strong contribution from fugitive sources (Fig. 3). Because isopentane and n-pentane exhibit similarly OH reaction rates (Atkinson, 1986), their ratio (iC_5/nC_5) is commonly used to identify emission sources (Bourtsoukidis et al., 2019). Accordingly, we examined this ratio across all sites using the PMF-resolved concentration of species from factor profile output to reveal their distinct relationship between isopentane and n-pentane.

Many studies report that an iC_5/nC_5 ratio in the range of approximately 0.8–1.1 is characteristic of oil & natural gas operations or raw-gas emissions (Gilman et al., 2013; Thompson et al., 2014; Swarthout et al., 2013; Gilman et al., 2010). In our analysis, the iC_5/nC_5 ratio was consistently <1 across the sites and seasons, with site-specific seasonally mean values of 0.53 ± 0.02 at Linyuan, 0.76 ± 0.06 at Xiaogang, and 0.87 ± 0.10 at Chaozhou, all within or below the typical oil & natural gas range. The Kaoping region hosts only downstream industrial activities—such as refining, petrochemical processing, and feedstock production—rather than upstream extraction. Therefore, this factor was attributed to industrial fugitive emissions, likely originating from petrochemical processing units, storage tanks, and refinery-related leakage. The lowest ratio at Linyuan reflects a very local, fresh fugitive leak from proximate refinery/petrochemical sources. In contrast, the downwind receptor (Chaozhou) or the mixed urban-industrial area (Xiaogang) can result in higher ratios."

11. Ch. 3.2.1.d. The mixed source profile is heavily dominated by toluene, yet the interpretation does not reflect this. The authors should use established NMHC ratios and also examine relationships with co-measured trace gases (see general comment 2).
Author's response: We have elaborated more for the mixed factor to enhance the factor interpretation. Fig.S4 caption for details factor profile in the supplementary is also enhanced for better readability.

Revised text: "Importantly, toluene is the dominant compound in the mixed factor, especially in Xiaogang, which provides a key constraint on source interpretation. The elevated toluene contribution points primarily to substantial solvent usage, as toluene is widely employed in paints, adhesives, coatings, and various cleaning agents, and chemical manufacturing processes common in industrial zones, along with ethylbenzene, m,p-xylene, n-hexane, and 1,2,4-trimethylbenzene (Wu et al., 2016; Shen et al., 2018; Shao et al., 2016). Together, the combined presence of combustion tracers, evaporative fuel markers, and solvent-related aromatics demonstrates that this factor represents a true mixture of vehicular exhaust and industrial solvent usage, with toluene-dominated solvent emissions (Bari and Kindzierski, 2018), serving as a major driver of its chemical profile. This mixed source factor highlights gaps in current emission inventories and underscores the need for improved, locally speciation-resolved data to support future research in this region. " (lines 457-467)

"**Figure S4** Seasonal PMF-resolved factor profiles at the study sites (a) winter, (b) spring, (c) summer, and (d) fall. Panels correspond to PAMS sites. Percentage contributions reflect relative patterns within each site; therefore, similar percentages do not necessarily correspond to similar absolute acetylene levels or emission characteristics. Species contribution in factor profile is denoted by the red dot."

12. Ch. 3.3.2.a. Consider adding a discussion of anthropogenic isoprene and its potential link to biomass burning (e.g. https://doi.org/10.5194/acp-24-7063-2024 , https://doi.org/10.1016/j.scitotenv.2023.166592)

Author's response: We thank the reviewer for highlighting the potential role of anthropogenic isoprene and biomass-burning emissions. Our earlier discussion referenced anthropogenic isoprene mainly in the context of traffic-related influences observed in predominantly urban environments, based on our previous work; however, the studies suggested by the reviewer have helped us refine and strengthen the interpretation. The revised discussion has been updated accordingly. (lines 536-549)

In brief: The revised text clarifies that the Chaozhou biogenic factor is dominated by isoprene with strong seasonal and diurnal patterns consistent with temperature and radiation driven biogenic emissions. It also acknowledges that the factor includes co-emitted VOCs, especially in summer, and incorporates emerging evidence that anthropogenic processes, including episodic biomass burning, can influence ambient isoprene and related VOCs. Supporting indicators, including acetylene factor with summer spikes from biomass burning at Chaozhou are used to justify the potential non-biogenic contributions within biogenic factor. Later on from trajectories section 3.4, evidences from air-mass trajectories further support the discussions. (lines 637-641)

Revised text: "The PMF results at Chaozhou identify a biogenic factor dominated by isoprene, with contributions present in both summer and fall (Fig.5). Temporal patterns (Fig. S7a,b) show that isoprene levels are substantially higher and more variable in summer, consistent with strong temperature and solar radiation dependence of biogenic isoprene emissions (Zeng et al., 2023; Vettikkat et al., 2023). These conditions are more intense and sustained during the summer months in southern Taiwan, and the surrounding agricultural landscape provides a plausible source. The pronounced late-morning to mid-afternoon peak (10:00–15:00) further supports a photosynthetically driven biogenic origin.

Notably, the biogenic factor profile also contains co-emitted VOCs, forming a mixed-species profile in which the isoprene signature is less distinct in summer but appears cleaner and more purely biogenic in fall. The observation is consistent with emerging evidence that urban isoprene budgets can include other non-biogenic sources (Peron et al., 2024), or traffic emissions (Chang et al., 2014; Hsieh et al., 2017). Residential biomass burning is another plausible contributor that can modify ambient mixtures during episodic events (Desservettaz et al., 2023). Given the agricultural setting at Chaozhou, biomass burning is likely a significant local source." (lines 536-549)

"Further trajectories are also run for Chaozhou, which supports the hypothesis that the high peak in August originates from local sources, indicating an intermittent episode peak from biomass burning in such a highly agricultural landscape (Fig. S1). This interpretation is further strengthened by the trajectory analysis (Fig. S9), which shows locally originating and spatially scattered air mass pathways around the monitoring site." (lines 637-641)

13. Ch 3.5. The OFP calculation is not shown. Please include a supplementary table listing the reaction rate constants used and elaborate further in the methodology.

Author's response: As responded to Reviewer#1**,** we have added a description of the OFP estimation in the methodology section. A supplementary table for species reactivity coefficients has been included in Table S1. (lines 243-251)

Revised text: "2.5 Ozone formation potential and uncertainty consideration

The OFP of each NMHC species was estimated using the Maximum Incremental Reactivity (MIR) coefficients developed by (Carter, 2010). The OFP for compound *i* was calculated as:

$$OFP_i = C_i \times MIR_i \quad (3)$$

Where C_i (ppb) is the measured mixing ratio of the species, and MIR_i (g O_3 g^{-1} VOC) is its reactivity coefficient (Table S1). The MIR scale represents ozone yield under low VOC/NO_x (i.e., high-NO_x or VOC-limited) conditions, where ozone formation is primarily sensitive to changes in VOC abundance. This assumption aligns with previous photochemical studies indicating that ozone formation in southern Taiwan is predominantly VOC-limited (Chang et al., 2022)."

14. Ch3.5. The discussion of ozone formation relies solely on OFP estimates without considering the prevailing chemical regime. Many regions in southern Taiwan are known to be VOC-limited, in which case the OFP ranking of factors does not directly translate into realized ozone production. The manuscript should at least acknowledge this limitation and ideally provide basic regime indicators (e.g. NOx levels, VOC:NOx ratios, or references to regional studies).

Author's response: We thank the reviewer for this insightful comment. We fully agree that OFP provides only an estimate of the potential for ozone formation and that the prevailing chemical regime strongly influences the realized ozone production. To address this concern, we have substantially revised the discussion in Section 3.6 (lines 704–714) and updated the conclusion (lines 763–786). The revised text now explicitly acknowledges the limitations of OFP in VOC-limited environments and integrates relevant regime information from regional studies.

First, we clarify that OFP represents potential ozone formation derived from the PMF-resolved VOC contributions; it is not interpreted as realized ozone production. Measurement uncertainties associated with each species are included in the PMF uncertainty matrix, ensuring that OFP calculations inherently reflect the uncertainty structure of the underlying VOC data. (lines 252-258)

Second, we added a mechanistic explanation of why the dominance of the mixed-source factor during moderate-ozone episodes is consistent with the VOC-limited regime prevalent in southern Taiwan (Chang et al., 2022). Under VOC-limited conditions, ozone production is more sensitive to reactive VOCs than to NOx. Mixed sources, which contain aromatics and light olefins with moderate-to-high MIR values, therefore exert a disproportionate influence even when absolute concentrations are moderate. This provides a photochemical justification for the observed OFP patterns. Then, we expanded the discussion under moderate-ozone days. In the region typically occur under weak transport and low-to-moderate mixing heights (AACOG, 2015), conditions that enhance the accumulation of locally emitted VOCs and increase the relative importance of mixed-source emissions. Finally, we added policy-relevant implications in the discussion, noting that the overlap of vehicular and solvent signals within the mixed-source factor introduces some uncertainty in precise source attribution, but also reflects real-world urban emission mixtures. This overlap highlights the need for coordinated VOC reduction strategies targeting both mobile and solvent-related emissions. The enhanced text clearly described how moderate-ozone episodes provide insights into how local and reactive NMHC composition shape ozone formation under VOC-limited conditions (lines 704–714). We believe these revisions considerably strengthen both the scientific interpretation and policy relevance of our OFP analysis.

Revised text: "The uncertainty associated with OFP estimation was inherently accounted for during the PMF analysis. These uncertainties were used to construct the PMF input uncertainty matrix, which determines the weighting of each data point in the model fitting. As the OFP was calculated from the PMF-resolved factor contribution time series, the measurement uncertainties are inherently reflected in the factor contributions. While this approach propagates measurement uncertainty into OFP estimates, it is important to note that OFP represents a photochemical potential, not realized ozone formation, and does not capture the nonlinear interactions that govern actual ozone production." (lines 252-258)

"Under moderate-ozone conditions, the mixed-source factor contributes the largest share of OFP. These episodes are frequently associated with lower wind speeds and reduced mixing heights (Aacog, 2015), favoring the accumulation of locally emitted species from traffic, solvents, and smaller-scale industrial activities. The coexistence of these emissions provides a balanced supply of reactive aromatics and olefins, sustaining ozone production even in the absence of strong photochemical aging. This pattern reflects the VOC-limited photochemical environment prevalent in southern Taiwan, where ozone formation is more sensitive to reactive VOCs than to NOx levels (Chang et al., 2022), emphasizing that moderate-ozone episodes are primarily governed by local accumulation and NMHC composition. However, the mixed-source factor reflects overlapping characteristics of vehicular and solvent-related species. This incomplete separation introduces some uncertainty in source-specific attribution of OFP. Nevertheless, this overlap mirrors the reality of urban environments." (lines 704–714).

Reviewer #3

The authors have leveraged high-resolution PAMS data, PMF technique along with a novel back-trajectory analyses to identify sources and quantify their contribution to NMHCs across three dynamic sites in Taiwan. The study also focuses on ozone formation making it interesting for both the academic community and policymakers. The manuscript is comprehensive and well written. However, some aspects need to be further clarified before this manuscript is accepted for publication.

General comments

1) There is a period (looks like August 2024) where there are high concentrations of acetylene in Chaozhou. What is this driven by? The authors mention that Chaozhou is likely influenced by more diffuse or intermittent combustion-related activities but that would have led to multiple spikes throughout the year, right? Also, how would the PMF results and other findings change if that time period was excluded?

Author's response: We appreciate the reviewer's comment. Chaozhou is an agricultural area where three annual harvest cycles lead to open-field biomass burning beginning in late summer and extending to the rest of the year. Acetylene, a tracer of incomplete combustion, therefore shows its strongest episodic peaks during this period, with smaller enhancements in fall and early winter. This pattern reflects intermittent but seasonally intensified combustion activities rather than continuous, year-round emissions.

Because PMF resolves factors based on distinct chemical–temporal signatures, the strong August peak allows the model to identify an acetylene-rich factor in summer. As demonstrated in Section 3.4, this supports the reliability of our PMF results in capturing source-specific signatures when driven by strong observational patterns. If this major episode were removed, the summer dataset would lack a clear combustion-related signal, and the acetylene factor would no longer be resolved—while other source factors would remain unaffected.

Later on from trajectories section, evidences from air-mass trajectories further support the discussions in Chaozhou.

Revised text: "Chaozhou is an agricultural area that leads to open-field biomass burning beginning in late summer and extending to the rest of the year. Acetylene is an emission from incomplete combustion processes, such as biomass burning (Burling et al., 2010; Wang et al., 2014), and therefore exhibits its strongest summer spikes and weaker, intermittent fall and winter contributions (Fig.2a, Fig.S1). This seasonal pattern reflects intermittent but intensified combustion activities rather than continuous emissions. Consequently, although PMF resolves an acetylene-rich factor at both Linyuan and Chaozhou, its contribution at Chaozhou is much lower (Fig.S5). Unlike Linyuan—where a known nearby acetylene filling plant provides a persistent local source—Chaozhou is influenced mainly by diffuse biomass burning and other small-scale, variable combustion activities. The absence of a strong directional signal in the CPF analysis for Chaozhou further supports the interpretation that acetylene in Chaozhou originates from non-point or variable sources rather than a single dominant emitter." (lines 585–595)

"Further trajectories are also run for Chaozhou, which supports the hypothesis that the high peak in August originates from local sources, indicating an intermittent episode peak from biomass burning in such a highly agricultural landscape (Fig. S1). This interpretation is further strengthened by the trajectory analysis (Fig. S9), which shows locally originating and spatially scattered air mass pathways around the monitoring site." (lines 637–641)

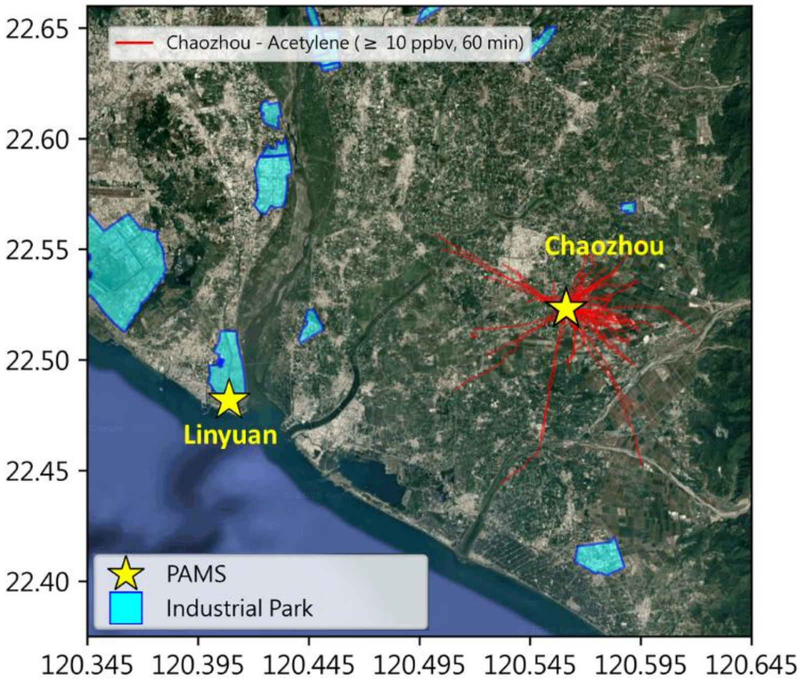


Fig.S9 Triggered back trajectory analysis of spike levels in Chaozhou. Base map from Google Maps (Map data ©2025 Google).

2) Figure captions in both manuscript and SI need to provide more details so that they are self-explanatory. Many figures have either missing legends or the legends are blurred. High resolution images are recommended.

Author's response: To enhance readability and self-explanation for the figures, we have refined the figure captions to help readers better interpret the results. Additionally, there were errors in supplementary figures, and corrections have been made: legend in Fig. S10 has been updated for clarity, and a vertical axis label has been added to Fig. S7a.

Revised text: "**Figure 3:** Summary of common source profiles of NMHC at the three sites in 2024. This figure presents the percentage contribution of six common source factors (Petro I, Petro II, Refinery, Industrial fugitive emissions, Mixed, and Aged air mass) to the total NMHC burden at three monitoring sites: Xiaogang (red), Linyuan (orange), and Chaozhou (green) across four seasons (Winter, Spring, Summer, and Fall). Each panel represents a specific source factor and season combination. The stacked bars within each panel show the relative contribution of NMHC species to that source factor. Consistent source profiles of fingerprint species were observed across seasons and at all three monitoring sites, underscoring the robustness of the PMF results."

"**Figure S4** Seasonal PMF-resolved factor profiles at the study sites (a) winter, (b) spring, (c) summer, and (d) fall. Panels correspond to PAMS sites. Percentage contributions reflect relative patterns within each site; therefore, similar percentages do not necessarily correspond to similar absolute acetylene levels or emission characteristics. Species contribution in factor profile is denoted by the red dot."

"**Figure S5** Seasonal variation of daily mean contributions of NMHC concentration from PMF-resolved factors. This figure illustrates the daily variability of identified source factors (Petro I, Petro II, Refinery, Industrial fugitive emissions, Mixed, Aged air mass and Acetylene) over the course of a year at three monitoring sites: Xiaogang (red), Linyuan (orange), and Chaozhou (green). To interpret this figure, follow the trends of the lines for each site within each season to observe how a specific source factor changes over time and reveal episodic emission events that indicate the changes in source activity. Shaded columns represent weekends."

<u>Specific comments</u>

3) Title: Multiple acronyms in the title make it difficult to follow. The authors could consider revising the title as well as including "Taiwan" in it.

Author's response: The title has been improved accordingly.

Revised text: "Multi-Site Non-Methane Hydrocarbon Source Apportionment and Ozone Insights in Southern Taiwan Using Positive Matrix Factorization"

4) Introduction. Lines 43-52. NOx is also mentioned here and it would benefit the reader if more details on the "complex atmospheric interactions" are added here. I am assuming that ozone production in all three study regions is sensitive to VOCs and not NOx, but this needs to be explicitly mentioned in both the introduction and the discussion.

Author's response: We have added text to further situate the work in the context of ozone regime sensitivity studies (VOC- vs. NOx-limited conditions). Further discussion and implications for control strategy has also been added in section 3.6

Revised text: "The impact of VOCs on ozone formation critically depends on the local chemical regime—whether ozone production is VOC-limited or NO_x-limited (Kleinman et al., 2002; Sillman, 1999). In VOC-limited environments, common in densely industrialized and urbanized areas, ozone levels are more responsive to changes in reactive VOC concentrations, whereas in NO_x-limited conditions, ozone formation is constrained by nitrogen oxide availability. Several studies in East and Southeast Asia have emphasized this spatial heterogeneity of ozone sensitivity (Li et al., 2019; Wang et al., 2021; Ren and Xie, 2022). In Taiwan, both modeling and observational evidence indicate that southern and western regions typically exhibit VOC-limited or transition regimes, while rural and downwind areas are more NO_x-limited (Chang et al., 2022; Chen et al., 2021). This regime dependence underscores the need for region-specific precursor management and highlights the importance of identifying the dominant reactive VOC sources that most effectively drive ozone formation. Understanding these sensitivities provides an essential framework for interpreting ozone formation potential (OFP) derived from NMHC source contributions." (lines 39–50)

5) Line 54. "Such as" part of previous sentence.

Author's response: The error is fixed; sentence is revised as read below.

Revised text: "Traditional VOC source analysis often relies on passive sampling techniques, such as using canisters at strategic locations to capture spatial and temporal variations in ambient concentrations…" (line 51)

6) Introduction. The authors should also include what is already known for Taiwan? And where has the PMF and CPF approach already been applied?

Author's response: The introduction has been improved as suggested.

Revised text: "The impact of VOCs on ozone formation critically depends on the local chemical regime—whether ozone production is VOC-limited or NO_x-limited

(Kleinman et al., 2002; Sillman, 1999). In VOC-limited environments, common in densely industrialized and urbanized areas, ozone levels are more responsive to changes in reactive VOC concentrations, whereas in NO_x-limited conditions, ozone formation is constrained by nitrogen oxide availability. Several studies in East and Southeast Asia have emphasized this spatial heterogeneity of ozone sensitivity (Li et al., 2019; Wang et al., 2021; Ren and Xie, 2022). In Taiwan, both modeling and observational evidence indicate that southern and western regions typically exhibit VOC-limited or transition regimes, while rural and downwind areas are more NO_x-limited (Chang et al., 2022; Chen et al., 2021). This regime dependence underscores the need for region-specific precursor management and highlights the importance of identifying the dominant reactive VOC sources that most effectively drive ozone formation. Understanding these sensitivities provides an essential framework for interpreting ozone formation potential (OFP) derived from NMHC source contributions." (lines 39–50)

"In Taiwan, PMF has been widely applied to support evidence-based air-quality policymaking; however, most studies have concentrated on northern (Kuo et al., 2014; Liao et al., 2017; Liao et al., 2024) and central regions (Su et al., 2019). Applications of CPF in conjunction with PMF have also been largely limited to central Taiwan (Huang and Hsieh, 2019; Chen et al., 2019), leaving the Kaoping region comparatively understudied—particularly in terms of combining PMF, CPF, and trajectory analyses to identify industrial and transported NMHC sources. Thus, this study aims to bridge this gap by applying PMF and CPF to identify and characterize NMHC sources in the Kaoping region and analyze the temporal features of PMF factor contributions to guide trajectory-based source tracking. By identifying high-contribution episodes for source factor and performing back-trajectory analyses during those episodes, we can confidently infer the likely geographic origins of the emissions. In addition, the OFP associated with each source was estimated to evaluate its relative impact on ozone production. Compared to most PMF studies that rely on data from a single receptor site and limited time resolution, our study leverages three PAMS sites with year-round hourly data and distinct source–receptor characteristics, allowing for a more robust source apportionment and regional representation. This multi-PAMS framework offers insights into NMHC dynamic emissions and transport in one of the most industrialized areas in Southern Taiwan." (lines 119–134)

7) Section 2.2 Data collection. It would be good to add more details here on how the data cleaning was carried out and also clarify whether the 8,596 hourly samples refers to the PAMS dataset or the TAQMN or combined dataset?

Author's response: Additional details on the data cleaning procedure have been incorporated into Section 2.2 of the revised manuscript.

Revised text: "Data cleaning was performed prior to analysis to ensure consistency across datasets. TAQMN measurements were first screened to remove invalid entries, including missing values, instrument flags, any non-numeric values associated with routine maintenance, calibration events, power outages, or temporary instrument shutdowns. After this quality-control procedure, the TAQMN 2024 dataset achieves approximately 98% data coverage across the three sites." (lines 181–185)

8) Equation 1 may be missing an "=".
Author's response: The equation was corrected.
Revised text: $$U_{ij} = \sqrt{\left(0.5 \times MDL_j\right)^2 + \left(error\ fraction \times X_{ij}\right)^2} \quad (1)$$

9) Page 6. Line 192. Screening resulted in 22 out of 54 measured species. It is important to quantify how much NMHCs by mass or volume are being investigated. The phrase "lacking sufficient data" is vague. Was there a threshold that was applied?
Author's response: We have added further description for the selection of 22 species. The screened out 22 species accounting for 88.9%, 91.7%, and 93.8% of the full 54 NMHCs concentration at Xiaogang, Linyuan, and Chaozhou, respectively. It was described in line 327.

Revised text: "For concentrations below MDL, the value was substituted with half MDL with uncertainty set at $\frac{5}{6}$ MDL, and an error fraction (10%). Missing values were excluded from the input dataset to maintain model reliability. To ensure a consistent and robust dataset across the three sites and four seasons, species selection was based on signal-to-noise (S/N) ratios and detection frequency following EPA PMF guidance. Specifically, species with S/N ratios < 0.1 or missing data with occurrences more than 4 instances were classified as "bad" and excluded, while those with 0.1 ≤ S/N < 2 were down-weighted as weak species. This screening process resulted in a final set of 22 out of the 54 species that were consistently detectable and quantitatively reliable across all sites and seasons (Table S1). Model stability was then evaluated through multiple diagnostic procedures. First, factor numbers from 3 to 8 were tested, each with 100 independent runs using random seed initialization. The optimal number of factors was selected based on $Q_{(robust)}/Q_{(true)}$ values that were close to 1.0, reproducibility of factor profiles across runs assessed via Bootstrap (BS) analyses (>95% matching), and the interpretability and physical plausibility of the resulting source profiles. Together, these procedures confirm that the PMF solution provides a robust and well-constrained representation of NMHC sources in the study region, while the consistent selection of

22 species ensures comparability across all seasonal and site-specific analyses." (lines 201–215)

10) Section 3.1. PAMS data overview. Looking at the timeseries data in Figure 2, I was wondering if median statistics should be presented instead of mean. Also, Chaozhou has a short period with elevated concentrations that should be discussed, and statistics should be presented with and without this period. Also, I would not say "low standard deviation" if it is similar in value to the mean.

Author's response: The comment has been incorporated with revised text in lines 266-284. We keep both mean and median to have more comprehended discussion.

Revised text: "However, the corresponding median values (10.92, 13.68, and 6.92 ppb, respectively) indicate that these distributions are skewed, particularly at Linyuan, where the mean is substantially higher than the median, reflecting occasional extreme emission events likely driven by dense petrochemical operations. Meanwhile, Xiaogang, with a mean slightly higher than its median, represents a mixed urban-industrial setting, where both vehicular emissions and industrial activities contribute to ambient NMHC levels. In contrast, Chaozhou exhibits lower mean and median concentrations, characteristic of a predominantly agricultural environment. Notably, an anomalous elevation was observed in Chaozhou during August, and this feature will be further examined in later sections. Statistical analysis reveals that, after removing the elevated window, the extreme short-duration did not alter the central tendency of the observations, with minimal impact observed on the statistical metrics (Table S3). A comparative analysis with other urban environments reveals that NMHC concentrations in this study were generally lower than those reported in Guangzhou (42.74 ppb), Wuhan (34.65 ppb), Chengdu (41.8 ppb), and Beijing (29.12 ppb), (Li et al., 2022; Hui et al., 2018; Zou et al., 2015; Song et al., 2018). This overall lower presence of NMHC may suggest effectiveness in emission control, supported by stringent regulations, improved fuel quality, and industrial emission standards (Moe, 2022, 2023). Additionally, meteorological factors, such as higher wind speeds at Xiaogang and Chaozhou, likely contribute to dilution and dispersion, further shaping the observed NMHC distribution. The combined consideration of mean and median reveals both typical ambient levels and the influence of short-term emission peaks in understanding NMHC exposure across the region." (lines 266–284)

11) There is a brief comparison of NMHC concentrations with other urban environments. Why were these locations chosen? Same latitude? Same sources? Are these values for same time of year? References should be added to support emission

controls, stringent regulations and improved fuel quality.

Author's response: These comparison cities (Guangzhou, Wuhan, Chengdu, and Beijing) were selected because they represent major Asian megacities with extensive NMHC literature. They were chosen for their relevance in urban emission source types (traffic and industry dominated NMHC profiles). The cited values were taken from studies that generally reported annual. Additionally, some references from the Taiwan Ministry of Environment (MOE) have been added to support the statement

Revised text: "A comparative analysis indicates that NMHC concentrations in this study were generally lower than those reported in Guangzhou (42.74 ppb), Wuhan (34.65 ppb), Chengdu (41.8 ppb), and Beijing (29.12 ppb), which were selected due to their similar urban environments (Li et al., 2022; Hui et al., 2018; Zou et al., 2015; Song et al., 2018). The difference reflects the effectiveness of emission control measures in Taiwan, supported by stringent regulations, cleaner fuels, and strengthened industrial emission standards (Moe, 2022, 2023). Additionally, meteorological factors, such as higher wind speeds at Xiaogang and Chaozhou, likely contribute to dilution and dispersion, further shaping the observed NMHC distribution." (line 276-283).

12) Figure 2 describes the percent contribution and so we see a high percentage contribution of acetylene in Linyuan. How do the absolute concentrations look? Are the absolute alkane concentrations similar across the three regions? The authors could consider adding one more row in Figure 2 to show the absolute concentrations for key species.

Authors' response: We have added a new Fig.S3 to show the annual mean of species across the seasons. In general, the alkanes highest at Xiaogang and lowest at Chaozhou.

Revised text: "Figure 2: Time series of total NMHC concentrations and mean composition of NMHC groups at Chaozhou, Linyuan, and Xiaogang in 2024. (a) Hourly variations illustrate temporal patterns and notable episodic peaks at each site. (b) Mean NMHC concentrations (ppb) and percentage contributions of individual compounds within major chemical groups. The mean concentrations of individual species are presented in Fig. S3."

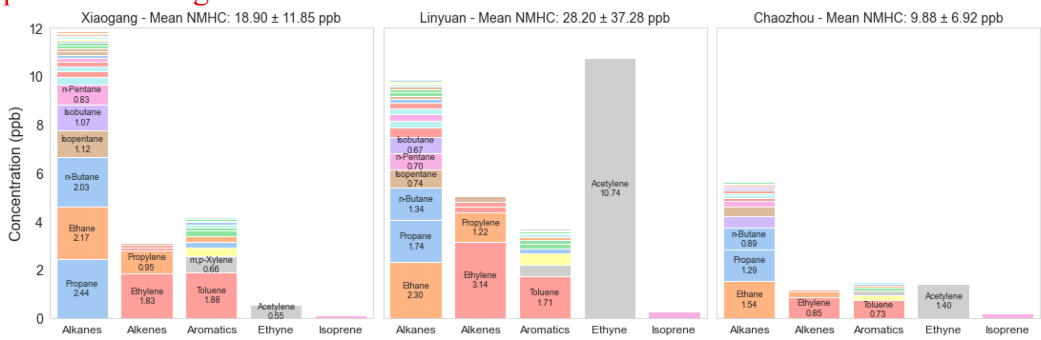


Fig. S3 Annual mean of notable NMHC species

13) Line 260-262. Is acetylene excluded from all three sites for this revised ranking?
Authors' response: Yes, acetylene was excluded consistently across all three stations when generating the revised ranking. Please see the new figure plot without Acetylene. The figure will be added in the supplementary as Fig. S2. And the related sentences will be enhanced for clarity.

Revised text: "When acetylene is excluded, the relative ranking changes, and Linyuan shifts to second place after Xiaogang (Fig. S2), highlighting the disproportionate influence of a single pollutant species on the site's total NMHC burden." (lines 301–303)

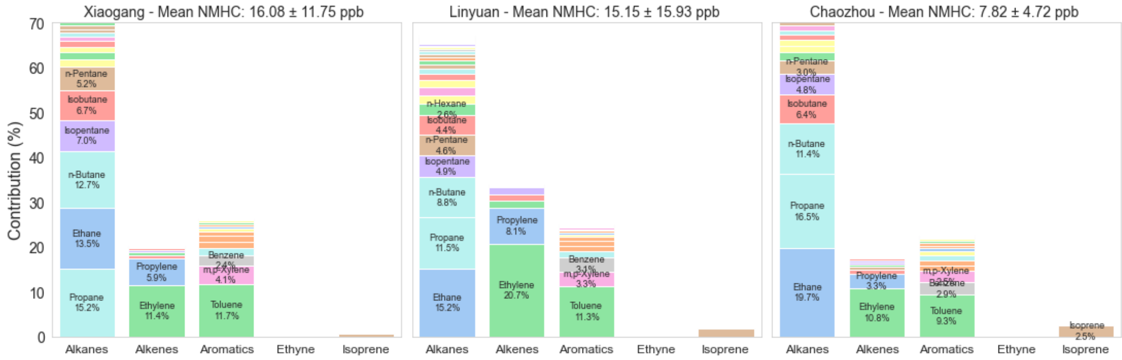


Fig. S2: Mean NMHC concentrations (ppb) and percentage contributions of individual compounds within major chemical groups after removing acetylene

14) The authors should add a brief description of the eight distinct sources. For example, distinguish between Petro I and Petro II. How is it different from petroleum refineries? This could be in the form of a table in the SI.
Authors' response: We have added a supplementary table to have a brief description of the eight distinct sources

Table S4. Summary and brief description of the eight distinct sources

Source	Key Species	Description
Petro I	Ethylene	Ethylene is the most important feedstock in the synthetic organic chemical manufacturing industry
Petro II	Propylene	Propylene is a critical precursor in producing various petrochemical products.
Refinery	C_3–C_5 alkanes	Associated with petroleum refining processes; distinct from petrochemical sources due to C_3–C_5 dominance and stability

Source	Key Species	Description
Industrial fugitive emissions	n-Pentane, Isopentane	Isopentane and n-pentane are recognized tracers of natural gas or gasoline vapor emissions. The ratio of iC_5/nC_5 that smaller than 1, leading to the signs of downstream petrochemical operations, gasoline/fuel storage
Mixed factor	Toluene, Benzene, Ethylbenzene, m,p-Xylene, Cyclopentane, 2-methylpentane, methylcyclopentane, acetylene, n-hexane, and 1,2,4-trimethylbenzene	Combination of vehicle and solvent emissions; difficult to separate due to overlapping NMHCs
Aged air mass	Ethane, Propane, Acetylene, Benzene	Long-lived NMHCs from regional transport; represents secondary and transported sources
Biogenic	Isoprene	Vegetation emissions, sensitive to temperature and sunlight; local to agricultural areas (Chaozhou)
Acetylene	Acetylene	Local industrial source (cylinder filling); distinct single-species source at Linyuan

15) I appreciate that it was difficult to distinguish between vehicular and solvent related emissions in the PMF analysis. Would it help to use a high-resolution emission inventory such as CEDS (now available at 10km resolution) to aid in interpretation? This could also be explored to investigate the non-biogenic sources of isoprene for section 3.3.2. If emission inventories are not capturing this, then it is very important to include that in the implications of this study.

Authors' response: We thank the reviewer for this thoughtful suggestion. We agree that emission inventories are valuable tools for supporting source interpretation. However, CEDS and other global inventories remain too coarse for separating solvent usage and vehicular emissions in Kaohsiung's highly industrialized environment, where emission gradients occur at scales well below 10 km. In addition, CEDS does not resolve facility-specific emissions from petrochemical plants, shipyards, port operations, or fugitive industrial sources that strongly influence NMHC levels at Xiaogang and Linyuan. Therefore, while such inventories are useful for regional trends, they cannot currently discriminate the co-located sources relevant to our study.

For these reasons, we therefore did not include it in the manuscript. Nevertheless, we agree with the reviewer that the limitations of current inventories are an important scientific implication. We have added a brief statement in the discussion noting that improved, higher-resolution, and more speciated emission inventories would greatly benefit future source-apportionment work in industrial regions such as Kaohsiung.

Authors' response: We have further refined the non-biogenic source of isoprene in section 3.3.2

Revised text: "This mixed source factor highlights gaps in current emission inventories and underscores the need for improved, locally speciation resolved data to support future research in this region. " (lines 465–467)

"Notably, the biogenic factor profile also contains co-emitted VOCs, forming a mixed-species profile in which the isoprene signature is less distinct in summer but appears cleaner and more purely biogenic in fall. The observation is consistent with emerging evidence that urban isoprene budgets can include other non-biogenic sources (Peron et al., 2024), or traffic emissions (Chang et al., 2014; Hsieh et al., 2017). Residential biomass burning is another plausible contributor that can modify ambient mixtures during episodic events (Desservettaz et al., 2023). Given the agricultural setting at Chaozhou, biomass burning is likely a significant local source." (lines 543–549)

16) For the acetylene factor, it would be a good idea to state the absolute contributions for all three sites. Are they similar for Chaozhou and Xiaogang? They look like from fig. S1 but the percent contribution from Chaozhou and Linyuan is similar. A brief discussion perhaps before moving to the back trajectory analyses?

Authors' response: Although the temporal patterns appear visually similar between Xiaogang and Chaozhou during periods of low concentration (Fig.S1), their annual mean levels—and therefore the overall magnitude of acetylene emissions—are clearly distinct (Fig.S3). Regarding the reviewer's observation that the percentage contributions of the acetylene factor appear similar across sites, we clarify that the percent contribution is a relative metric within each site's PMF solution and should not be directly compared across sites. Percentage contributions reflect how a factor behaves relative to other sources at the same site, not absolute emission strength. Thus, similarities in percentage contributions do not imply similar absolute acetylene levels or emission characteristics across locations. These clarifications have been incorporated in the caption of Fig.S4 to avoid misinterpretation.

A brief discussion between Chaozhou and Linyuan has been added before moving to the back trajectory analyses.

Revised text: "Chaozhou is an agricultural area that leads to open-field biomass burning beginning in late summer and extending to the rest of the year. Acetylene is an emission from incomplete combustion processes, such as biomass burning (Burling et al., 2010; Wang et al., 2014), and therefore exhibits its strongest summer spikes and weaker, intermittent fall and winter contributions (Fig.2a, Fig.S1). This seasonal pattern reflects intermittent but intensified combustion activities rather than continuous emissions. Consequently, although PMF resolves an acetylene-rich factor at both Linyuan and Chaozhou, its contribution at Chaozhou is much lower (Fig.S5). Unlike Linyuan—where a known nearby acetylene filling plant provides a persistent local source—Chaozhou is influenced mainly by diffuse biomass burning and other small-scale, variable combustion activities. The absence of a strong directional signal in the CPF analysis for Chaozhou further supports the interpretation that acetylene in Chaozhou originates from non-point or variable sources rather than a single dominant emitter." (lines 585–595)

"**Figure S4** Seasonal PMF-resolved factor profiles at the study sites (a) winter, (b) spring, (c) summer, and (d) fall. Panels correspond to PAMS sites. Percentage contributions reflect relative patterns within each site; therefore, similar percentages do not necessarily correspond to similar absolute acetylene levels or emission characteristics. Species contribution in factor profile is denoted by the red dot."

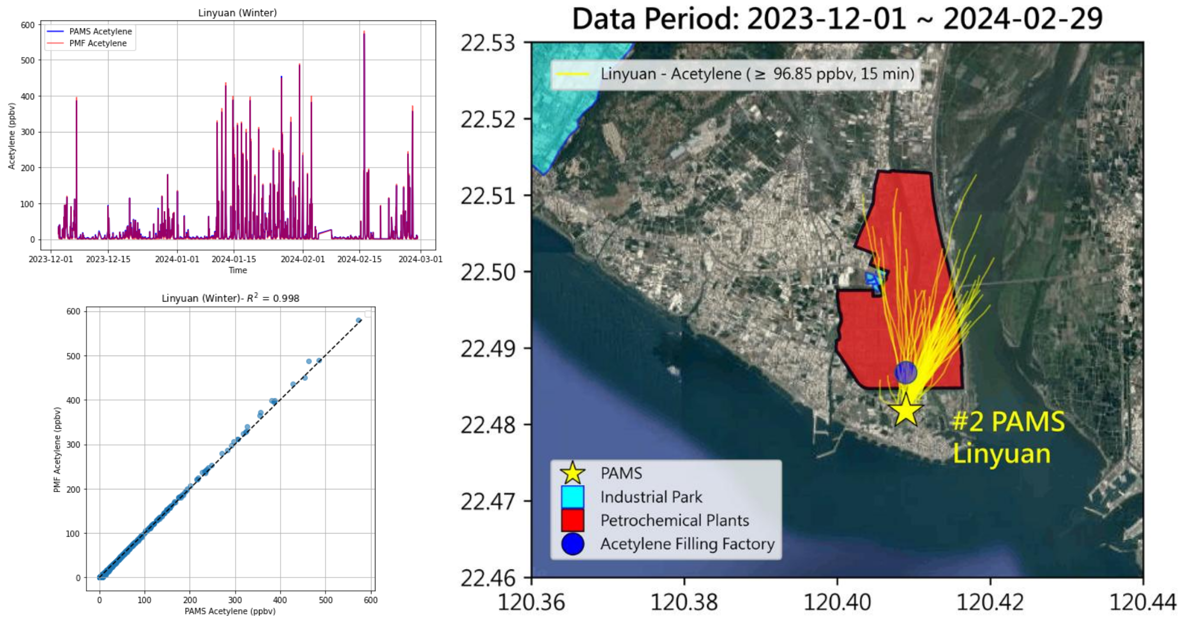
17) Line 530. Correlation coefficient is R.

Authors' response: Thank you for pointing this out. We have corrected the mistake.

Revised text: "Beyond factor profiles and species contributions, the PMF-resolved acetylene factor at Linyuan also resulted in a well-defined temporal profile that closely matched the observed data. Figure 6a demonstrates this consent with a coefficient of determination (R^2) exceeding 0.99." (line 608)

18) Figure 6. The acetylene filling factory is not visible.

Authors' response: The Fig. was revised and enhanced the visualize. (line 642)



a) Agreement of PAMS and PMF-resolved factor

b) Triggered back trajectory

Figure 6: The acetylene factor at Linyuan, (a) time series comparison and (b) Triggered back trajectory analysis of spike levels. Base map from Google Maps (Map data ©2025 Google).

19) Figure 7. Similar to my comment above, it would be helpful to add another row that presents the absolute concentrations as well. Also, a brief discussion on that.

Authors' response: The absolute value for OFP estimation was actually presented in Figure S11

20) Section 3.5 OFP calculation should be described. What is the peak ozone season in Taiwan? Anything special about OFP at this time of the year? Some of the sources will increase both VOCs as well as nitrogen oxides. Is it possible to comment on the resulting impact on ozone production (and regimes)?

Authors' response: As responded to Reviewer#1**,** we have added a description of the OFP estimation in the methodology section. A supplementary table for species reactivity coefficients has been included. In addition, the revised manuscript expands the discussion of OFP source contributions under different ozone conditions, emphasizing the role of meteorological conditions, photochemical activity, and source composition. The interpretation is framed within a VOC-limited photochemical regime, which is characteristic of southern Taiwan, allowing the impacts of concurrent VOC and NOx emissions on ozone formation to be addressed implicitly through source-specific OFP behavior rather than seasonal classification.

Revised text: "2.5 Ozone formation potential and uncertainty consideration

The OFP of each NMHC species was estimated using the Maximum Incremental Reactivity (MIR) coefficients developed by (Carter, 2010). The OFP for compound i was calculated as:

$$OFP_i = C_i \times MIR_i \quad (3)$$

Where C_i (ppb) is the measured mixing ratio of the species, and MIR_i (g O_3 g^{-1} VOC) is its reactivity coefficient. The MIR scale represents ozone yield under low VOC/NOx (i.e., high-NOx or VOC-limited) conditions, where ozone formation is primarily sensitive to changes in VOC abundance. This assumption aligns with previous photochemical studies indicating that ozone formation in southern Taiwan is predominantly VOC-limited (Chang et al., 2022)." (lines 243–251)

"Under moderate-ozone conditions, the mixed-source factor contributes the largest share of OFP. These episodes are frequently associated with lower wind speeds and reduced mixing heights (Aacog, 2015), favoring the accumulation of locally emitted species from traffic, solvents, and smaller-scale industrial activities. The coexistence of these emissions provides a balanced supply of reactive aromatics and olefins, sustaining ozone production even in the absence of strong photochemical aging. This pattern reflects the VOC-limited photochemical environment prevalent in southern Taiwan, where ozone formation is more sensitive to reactive VOCs than to NOx levels (Chang et al., 2022), emphasizing that moderate-ozone episodes are primarily governed by local accumulation and NMHC composition. However, the mixed-source factor reflects overlapping characteristics of vehicular and solvent-related species. This incomplete separation introduces some uncertainty in source-specific attribution of OFP. Nevertheless, this overlap mirrors the reality of urban environments." (lines 704–714)

21) Figure S2 is a multi-page multi-panel figure and should be split into multiple figures.

Authors' response: We appreciate the reviewer's suggestion regarding Figure S2. This figure serves as a unified collection of the seasonal PMF factor profiles, and presenting all four seasons within one figure preserves direct comparability of the profiles across seasons. To improve clarity, Fig. S2 is now Fig.S4 with enhanced title caption. We hope that this format maintains readability while preserving the integrity of the seasonal comparison.

Revised text: “**Figure S4** Seasonal PMF-resolved factor profiles at the study sites (a) winter, (b) spring, (c) summer, and (d) fall. Panels correspond to PAMS sites. Percentage contributions reflect relative patterns within each site; therefore, similar percentages do not necessarily correspond to similar absolute acetylene levels or emission characteristics. Species contribution in factor profile is denoted by the red dot.”

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