

## Response (in blue) to Reviewer's comments #1

**Manuscript number:** egusphere-2025-165

**Title:** Explainable ensemble machine learning revealing enhanced anthropogenic emissions of particulate nitro-aromatic compounds in eastern China

### Response to reviewer #1:

Li et al. investigates the influencing factors of particulate nitro-aromatic compounds (NACs) in eastern China, including meteorological factors, primary and secondary sources of NACs. The machine learning combined with PMF model is a key feature. Also, machine learning has not been applied to study NACs before. So, this paper studies an important component of atmospheric aerosols (i.e., NACs) with an innovative approach. Results are presented in a logical and organized manner with thorough discussion. Conclusions are clear and reasonable. Nevertheless, there are still some places where clarifications are needed, which are not major or critical issues. Also, the language could be further improved. Overall, I would recommend a minor revision before this paper could be accepted.

We appreciate the opportunity to revise our manuscript and are grateful for the insightful comments. Those comments are all valuable and very helpful for revising and improving our paper, as well as the important guiding significance to our researches. In the following, we have provided detailed responses to reviewer's comments. The reviewer's comments are in **bold black**, while our responses and changes in the manuscript are in blue and in *blue italic*, respectively. In addition, we also made some changes to improve the language of the manuscript. And here we did not list the changes but marked in *blue italic* in the revised paper. We have tried our best to make all the revisions clear, and hope that the revised manuscript meets the requirements for publication.

### Minor comments:

**Line 24-25:** The authors state that “temperature had the largest impact in winter”. It is still not clear whether higher temperatures impose a positive or negative impact on NAC abundances in winter. Please briefly elaborate here.

**Response:** Thanks for the valuable comments. In the revision, we have carefully clarified that lower temperatures generally have a positive impact on NAC abundance in winter.

### Revised sentence in manuscript (Line 23–24):

“Seasonal variations analysis showed that direct emissions presented positive responses to NACs concentrations in spring, summer, and autumn, while *lower* temperature had the largest *positive* impact in winter.”

**Line 48-49:** (1) The “in-situ” used here sounds not necessary. (2) Specifically, only aromatic VOCs could be oxidized to produce NACs. (3) More recent references should also be cited here. A recommended reference is shown as follows.

Men Xia et al., 2023, JGR: A, Observations and Modeling of Gaseous Nitrated Phenols in Urban Beijing: Insights From Seasonal Comparison and Budget Analysis.

**Response:** Thanks for your valuable suggestions. We have addressed these comments as follows:

- (1) We agree that the use of “in-situ” in this context is not essential and may cause confusion. Accordingly, we have removed the term to improve clarity.
- (2) We appreciate the clarification. We have revised the sentence to specify that aromatic volatile organic compounds (VOCs) are the key precursors that can undergo atmospheric oxidation to form NACs, which improves the precision of the description.
- (3) As suggested by the reviewer, we have checked the literature carefully and incorporated the suggested references into the revised manuscript and cited them appropriately in the relevant context to strengthen the literature support.

**Revised sentence in manuscript (Line 47–49):**

“They can be also produced through nitration of anthropogenic *aromatic* volatile organic compounds (VOCs) initiated by OH and NO<sub>3</sub> radicals in either the gas or aqueous phases (Harrison et al., 2005; Atkinson et al., 1989; Atkinson et al., 1992; *Xie et al., 2017; Xia et al., 2023*).”

**Added reference:**

*Xie, M., Chen, X., Hays, M. D., Lewandowski, M., Offenberg, J., Kleindienst, T. E., and Holder, A. L.: Light absorption of secondary organic aerosol: composition and contribution of nitroaromatic compounds, Environ. Sci. Technol., 51, 11607-11616, <https://doi.org/10.1021/acs.est.7b03263>, 2017.*

*Xia, M., Chen, X., Ma, W., Guo, Y., Yin, R., Zhan, J., Zhang, Y., Wang, Z., Zheng, F., Xie, J., Wang, Y., Hua, C., Liu, Y., Yan, C., and Kulmala, M.: Observations and Modeling of Gaseous Nitrated Phenols in Urban Beijing: Insights From Seasonal Comparison and Budget Analysis, J. Geophys. Res.-Atmos., 128, e2023JD039551, <https://doi.org/10.1029/2023JD039551>, 2023.*

**Line 55: How could solar radiation inhibit NAC photolysis? In my understanding, solar radiation should enhance NAC photolysis. Also, “NACs photolysis production and loss” lacks clarity. Please double check the expressions here.**

**Response:** Thanks for the reviewer’s critical comments. We agree that the original sentence was misleading and lacked clarity. Our intended meaning was that surface net solar radiation (SSR) has a dual role in the atmospheric behavior of NAC: it can promote their photochemical formation via oxidation of aromatic precursors, and also enhance their photolytic degradation. In the revised manuscript, we have modified the sentence in the manuscript to better reflect this twofold effect and avoid confusion.

**Revised sentence in manuscript (Line 54–55):**

“...while surface net solar radiation (SSR) *exerted a dual effect by enhancing both the photochemical production and photolytic degradation of NACs* (Peng et al., 2023b).”

**Line 56: What does “their” refer to, the abundance of NACs or the influencing factors of NACs? Please clarify. Also, please check the potential abuse or overuse of “it” and “they” in other places.**

**Response:** Thanks for your helpful comment. We agree that the reference of the pronoun “it” and “their” was ambiguous in the original sentence. In the revised version, we have clarified that “it” refers to the challenge of quantification, and “their” refers to the impacts of influencing factors. Additionally, we have reviewed the manuscript to avoid potential abuse or overuse of pronouns such as “it” and “they”, and all unclear pronoun references have been corrected accordingly.

**Revised sentence in manuscript (Line 55–57):**

“The complex and synergetic effects of primary emissions, secondary formation, and meteorological conditions on the abundances of NACs make *the quantification of the individual contribution of each factor* a challenge.”

**Line 67: So far, it is inappropriate to judge that machine learning is a more advanced method than PMF or PCA analysis. As an emerging method that is only recently applied in atmospheric chemistry, some scholars also hold a conservative attitude toward the usage of machine learning.**

**Response:** Thanks for the insightful comments. Machine learning is a relatively new tool in atmospheric chemistry, and it may not be appropriate to claim that it is categorically more advanced than traditional receptor models such as PMF and PCA. In the revised manuscript, we have carefully rephrased the relevant sentence and clarified that machine learning is used as a complementary tool to explore complex and nonlinear relationships that may not be captured well by conventional linear models.

**Revised sentence in manuscript (Line 64–68):**

“However, these methods are typically *based on* linear algorithms that *may overlook* the multivariate nature and nonlinear relationships between NACs and the potential sources as well as the complex influences *from* meteorological conditions, *potentially resulting in biased interpretations of NACs under complex atmospheric conditions*. *Therefore, a complementary* data analysis *approach* is *warranted* to *more efficiently* uncover the hidden complicated relationships.”

**Line 72-73: It is not clear whether Qin et al. and Peng et al. investigated NACs or other compounds.**

**Response:** Thanks for pointing this out. We acknowledge that the original sentence lacked clarity regarding the compounds investigated in the cited studies. The reference of Qin et al. and Peng et al. employed machine learning model in combination with SHAP to analyze the drivers of gaseous elemental mercury and atmospheric visibility, respectively. These examples were cited to demonstrate the growing use of machine learning in atmospheric chemistry, which supports its applicability in this study of particulate NACs. To improve clarity, we have revised the manuscript as follows:

**Revised sentence in manuscript (Line 72–76):**

“The interpretable ML methods in combination with *interpretable* SHAP *algorithm* have been recently applied to investigate the formation mechanism and influencing factors *of atmospheric pollutants. For example*, Qin et al. (2022) *quantified the drivers of gaseous elemental mercury by using an ML model in combination with SHAP*. Peng et al. (2023a) *utilized an ML model coupled with SHAP to assess the effects of PM<sub>2.5</sub> sources and RH on atmospheric visibility*.”

**Line 79: The authors mention “source apportionment”. Does that mean the authors also use methods like PMF or PCA? Please do clarify this key point.**

**Response:** Thanks for the comment. In this study, we applied positive Matrix Factorization (PMF) in this study to identify the major sources of particulate NACs from primary emissions and secondary formation. In the revised manuscript, we have clarified the methodology accordingly.

**Revised sentence in manuscript (Line 82–83):**

“*By integrating observational* datasets of NACs, meteorological data, particle loading (*i.e.*, aerosol surface area data), and source apportionment results *derived from PMF model, .....*”

**Line 84-85: The combination of PMF and machine learning is a highlight in this paper, which should be emphasized more clearly and thoroughly here, and maybe emphasized again in other places, e.g., the last paragraph in the conclusion part.**

**Response:** Thanks for your insightful suggestion. In response, we have revised the manuscript to emphasize this highlight more clearly and thoroughly in the last paragraph of conclusion part.

**Revised sentence in manuscript (Line 471–475):**

“Particularly, the integration of *PMF-based source apportionment with* a data-driven ensemble machine learning model and SHAP analysis method proved as a potent tool for rapidly diagnosing the driving factors for organic aerosols, which is helpful for the control strategies targeting aerosol pollution. *This hybrid approach not only enhances the interpretability of ML results but also allows for a more robust and quantitative assessment of the contributions of individual sources and environmental drivers.*”

**Line 97-98: The authors honestly acknowledge that some data has been reported in previous studies, which is of course good manners. Nevertheless, it is more important to emphasize what data is newly reported here, if any.**

**Response:** Thanks for your kind comment. In the revised manuscript, we have clarified which data are newly reported in this study. Specifically, the NACs data from Mount Tai and Mount Lao in spring, Guangzhou and Dongying in summer, Jinan and Nanjing in autumn, and Beijing, Dongying and Mount Tai (2017) in winter are newly measured and reported in this study for the first time. These new data not only enrich the seasonal coverage, but also allow for a more comprehensive comparison across urban, rural, and mountain sites.

**Revised sentence in manuscript (Line 106–111):**

“*In contrast, the NAC data collected during the campaigns in spring at Mount Tai and Mount Lao, the campaigns in summer in Guangzhou and Dongying, the campaigns in autumn in Jinan and Nanjing, and the campaigns in winter in Beijing, Dongying and Mount Tai (2017) are newly reported in this study. More importantly, the novelty of this work lies in the integration of multi-season, multi-site dataset with an ensemble machine learning algorithm to comprehensively assess the key driving factors of particulate NACs across different sampling locations and seasons.*”

**Line 112: Check for typo of “filed campaigns”.**

**Response:** Thanks for pointing out the typo. We have corrected “filed campaigns” to “field campaigns” in the revised manuscript.

**Line 115: The authors mention SO<sub>2</sub>, NO<sub>2</sub>, and O<sub>3</sub>. Was NO measured? Usually, NO and NO<sub>2</sub> are measured together by gas analyzers.**

**Response:** Thanks for the comment. In this work, NO and NO<sub>2</sub> are measured at most sites using online instruments. However, for the Nanjing and Guangzhou sites, due to the lack of on-site trace gases measurements, we obtained the corresponding data from the nearest national air quality monitoring stations, which did not provide NO concentrations. Therefore, NO was excluded from our data analysis for consistency.

**Revised sentence in manuscript (Line 125–128):**

“Several major tracer gases ( $\text{SO}_2$ ,  $\text{NO}$ ,  $\text{NO}_2$ , and  $\text{O}_3$ ) were simultaneously monitored by online analyzers *at most sampling sites. For* the Nanjing and Guangzhou sites, *where on-site gas measurements were not available, the corresponding data were* downloaded from the China National Environmental Monitoring Centre (available at: <http://106.37.208.233:20035/>). *NO concentrations were unavailable at these two sites due to data limitations and were therefore excluded from this study.”*

**Line 135: Was 2-nitrophenol detected? Why or why not?**

**Response:** Thank you for your thoughtful comment. In this study, 2-nitrophenol (2NP) was not detected in the fine particulate matters. This is due to its relatively high vapor pressure and lower Henry's law constant ( $P_{2\text{NP}} = 1.5 \times 10^{-4}$  atm and  $H_{2\text{NP}} = 81.1 \text{ M atm}^{-1}$ ) compared to 4NP ( $P_{4\text{NP}} = 6.6 \times 10^{-7}$  atm and  $H_{4\text{NP}} = 3.0 \times 10^5 \text{ M atm}^{-1}$ ), resulting in a stronger tendency for 2NP to partition into the gas phase rather than the particle phase (Wang et al., 2020). Additionally, as reported by Rubio et al. (2012) and Wang et al. (2021), 2NP is characterized by a faster removal rate than 4NP in the atmosphere, which could contribute to its absence in the particle-phase samples.

**Line 142-146: Please elaborate what is the overall/total uncertainty of measuring NACs?**

**Response:** Thank you for your valuable comment. The total uncertainty in the measurement of NACs was estimated by considering the main error sources, including extraction recovery rates, instrumental precision ( $\pm 3.8\%$ , obtained from five times replicate measurements of samples with same concentrations using UHPLC-MS), and blank correction. Based on this analysis, the total measurement uncertainty was estimated to be approximately  $\pm 19.1\%$ . This information has been added to the revised manuscript.

**Revised sentence in manuscript (Line 158–162):**

*“Moreover, the instrumental precision was determined by repeated analysis of standard solutions ( $n = 5$ ) under the same operating conditions, yielding relative standard deviations of  $\pm 3.8\%$  for the target NACs, which indicates high analytical reproducibility. Taking into account errors from extraction recovery rates, instrumental precision, and blank subtraction, the total measurement uncertainty for NACs was estimated to be approximately  $\pm 19.1\%$ .”*

**Line 159-161: Although more details could be found in SI, it is still necessary to state other data or parameters input into the PMF. Also, the key message of PMF methods stated in SI should also be briefly summarized and mentioned in the main text.**

**Response:** Thanks for your suggestions. In this study, the dataset used for PMF analysis consisted of 613 aerosol samples with 10 kinds of species (including 4NP, 3M4NP, 2M4NP, 4NC, 4M5NC, 3M6NC, 5NSA, 3NSA,  $\text{NO}_2$ , and  $\text{O}_3$ ). The optimal number of factors was determined to be four based on the ratio of  $Q_{\text{true}}$  to  $Q_{\text{robust}}$ , as detailed in Text S4 in SI. As suggested, a brief summary of the PMF methodology has been added to the main text to enhance clarity for readers.

**Revised sentence in manuscript (Line 175–182):**

*“In this study, the PMF input matrix consisted of 613 daily aerosol samples and ten components (including 4NP, 3M4NP, 2M4NP, 4NC, 4M5NC, 3M6NC, 5NSA, 3NSA,  $\text{NO}_2$ , and  $\text{O}_3$ ). For the input data, the treatment and calculation of the concentrations and associated uncertainties for each species followed the methodology described in our previous study (Li et al., 2020a). Here, by comparing the  $Q$  value results with two to six factor numbers, the optimal number of source factors was determined to be four. Specific details of the PMF model configuration and evaluation can be found in Text S4 and Fig. S3. Based on the outputs from the PMF model, four major sources of NACs, including coal combustion*

(CC), traffic emission (TE), secondary formation associated with gas-phase reaction (GR), and biomass burning (BB), were identified from samples collected at the 11 sampling sites (Text S4) and the corresponding source profiles are presented in Fig. S4.”

**Line 164:** The expression “were considered firstly in this study” sounds misleading. The mentioned machine learning algorithms have already been applied in previous studies.

**Response:** Thanks for the comment. Our intention was to express that these four machine learning algorithms have been widely applied in prior environmental studies, and were therefore selected as the initial candidates in our model framework. In the revised version, we have revised the sentence accordingly to improve clarity and academic accuracy.

**Revised sentence in manuscript (Line 184–185):**

“Four widely employed ML algorithms, including random forest (RF), extreme gradient boosting (XGBoost), light gradient boosting machine (LightGBM), and multilayer perceptron (MLP), were *selected* in this study *for model development*.”

**Line 182: Check for typo of “leaners”.**

**Response:** Thanks for the comment. The typo has been corrected.

**Line 195: Check the grammar for “for quantify”. Please also carefully check the grammar issues in other places.**

**Response:** Thank you for your careful review. The grammar error “for quantify” has been corrected to “for quantifying” in the revised manuscript. We have also thoroughly checked the manuscript for other grammar issues and made necessary corrections accordingly.

**About section 2.5 Aerosol surface area density (Sa) prediction. This section needs to be moved to SI. The prediction of Sa by machine learning is not a major scientific goal of this study.**

**Response:** Thanks for the constructive suggestion. In accordance with the review’s comment, the prediction of aerosol surface area density (Sa) by machine learning has been moved to the SI, as it is not a central scientific objective of this study. Relevant descriptions in the main text have been revised accordingly.

**Revised sentence in manuscript (Line 135–138):**

“Additionally, Sa data for the remaining sites were estimated by using *predictive capability* machine learning algorithms based on the *input variables* of PM<sub>2.5</sub> and meteorological parameters. *Detailed descriptions on the estimation method of Sa can refers to Text S2 and Fig. S1, and the predicted Sa results were shown in Fig. S2.*”

**In Table 1, at least the total NACs concentrations, which is key to this study, should be mentioned. Since the season has been mentioned, the detailed sampling period is less interesting and may be recorded in SI.**

**Response:** Thanks for the comment. In the revised manuscript, the total and average concentration of NACs has been added in Table 1 and main text. Meanwhile, the detailed sampling periods have been removed from Table 1 and provided in the SI.

**Revised sentence in manuscript (Line 230–231):**

*“The particulate NACs measured in this study exhibited relatively high levels, with an average total concentration of  $28.5 \pm 32.7 \text{ ng m}^{-3}$  across four seasons at eleven sampling sites.”*

**Revised Table 1:**

Sampling site	Season	$\Sigma\text{NACs}$	$\text{SO}_2$ (ppbv)	$\text{NO}_2$ (ppbv)	$\text{O}_3$ (ppbv)	$\text{CO}$ (ppbv)	T (°C)	RH (%)
<i>Jinan</i>	Spring	<b><math>34.0 \pm 24.0</math></b>	$13.7 \pm 7.7$	$43.7 \pm 23.2$	$79.3 \pm 19.9$	<b><math>920.2 \pm 307.1</math></b>	$20.1 \pm 2.5$	$37.2 \pm 13.5$
	Summer	<b><math>10.4 \pm 4.5</math></b>	$14.7 \pm 14.7$	$26.7 \pm 13.7$	$42.6 \pm 26.3$	<b><math>1049.2 \pm 573.5</math></b>	$24.0 \pm 4.3$	$66.9 \pm 16.4$
	Autumn	<b><math>26.3 \pm 27.9</math></b>	$4.4 \pm 1.8$	$35.2 \pm 15.8$	$21.7 \pm 14.5$	<b><math>812.1 \pm 354.5</math></b>	$11.7 \pm 3.0$	$44.8 \pm 12.2$
	Winter	<b><math>60.7 \pm 31.9</math></b>	$21.4 \pm 9.6$	$26.3 \pm 12.1$	$30.2 \pm 17.6$	<b><math>1053.0 \pm 403.3</math></b>	$8.6 \pm 3.8$	$36.3 \pm 11.7$
<i>Guangzhou</i>	Summer	<b><math>19.8 \pm 10.5</math></b>	$3.0 \pm 0.5$	$20.0 \pm 3.8$	$13.0 \pm 13.2$	<b><math>566.2 \pm 82.0</math></b>	$27.1 \pm 3.0$	$79.3 \pm 11.2$
<i>Nanjing</i>	Autumn	<b><math>8.2 \pm 3.3</math></b>	$3.4 \pm 0.8$	$30.2 \pm 11.0$	$23.5 \pm 15.8$	<b><math>529.4 \pm 126.2</math></b>	$14.1 \pm 3.3$	$69.6 \pm 15.2$
<i>Beijing</i>	Winter	<b><math>42.1 \pm 27.1</math></b>	$3.7 \pm 3.0$	$21.1 \pm 13.2$	$21.2 \pm 9.9$	<b><math>691.0 \pm 489.6</math></b>	$-3.3 \pm 4.4$	$36.4 \pm 13.5$
<i>Yucheng</i>	Summer	<b><math>5.8 \pm 2.7</math></b>	$3.2 \pm 3.0$	$20.9 \pm 12.5$	$45.9 \pm 18.9$	<b><math>665.8 \pm 146.8</math></b>	$24.5 \pm 3.5$	$69.3 \pm 15.3$
<i>Wangdu</i>	Summer	<b><math>5.9 \pm 3.7</math></b>	$7.0 \pm 5.6$	$14.2 \pm 7.6$	$56.9 \pm 23.3$	<b><math>521.2 \pm 203.9</math></b>	$27.0 \pm 4.4$	$55.4 \pm 18.1$
<i>Dongying</i>	Summer	<b><math>20.9 \pm 12.5</math></b>	$3.6 \pm 1.5$	$5.1 \pm 2.2$	$77.0 \pm 28.5$	<b><math>478.2 \pm 173.0</math></b>	$27.7 \pm 3.6$	$60.2 \pm 11.9$
	Winter	<b><math>41.7 \pm 27.6</math></b>	$4.6 \pm 3.6$	$11.6 \pm 5.2$	$21.8 \pm 7.1$	<b><math>1494.8 \pm 553.9</math></b>	$-2.6 \pm 1.9$	$76.9 \pm 13.7$
<i>Qingdao</i>	Winter	<b><math>53.6 \pm 53.2</math></b>	$3.7 \pm 2.1$	$16.8 \pm 9.3$	$22.2 \pm 11.1$	<b><math>757.2 \pm 382.5</math></b>	$4.0 \pm 5.5$	$64.3 \pm 18.0$
<i>Mount Tai</i>	Spring	<b><math>10.8 \pm 4.9</math></b>	$2.1 \pm 1.4$	$2.1 \pm 1.3$	$72.7 \pm 8.9$	<b><math>445.1 \pm 121.3</math></b>	$8.5 \pm 4.0$	$67.4 \pm 18.5$
	Summer	<b><math>2.5 \pm 1.6</math></b>	$2.6 \pm 2.0$	$2.7 \pm 0.8$	$70.3 \pm 18.6$	<b><math>331.6 \pm 148.9</math></b>	$19.7 \pm 2.6$	$86.9 \pm 8.9$
	Winter	<b><math>30.3 \pm 13.6</math></b>	$2.0 \pm 1.3$	$4.2 \pm 2.7$	$40.9 \pm 7.6$	<b><math>308.2 \pm 168.3</math></b>	$-3.8 \pm 3.3$	$51.8 \pm 20.5$
<i>Mount Lao</i>	Spring	<b><math>12.3 \pm 8.3</math></b>	$1.0 \pm 0.8$	$7.7 \pm 3.7$	$50.3 \pm 12.2$	<b><math>273.0 \pm 99.2</math></b>	$16.7 \pm 3.6$	$56.0 \pm 22.6$

**Revised sentence in Supporting Information (Line 24–26):**

“Detailed *information on* sampling sites and online measurements *is* available below, *with the specific sampling periods for each field campaign illustrated in Table S1. As indicated, three field campaigns were conducted in spring, six in summer, two in autumn, and five in winter.*”

**Added Table S1:**

*Table S1. Sampling sites and sampling periods involved in this study.*

Sampling site	Site type	Sampling period	Season	Number of samples	Detected species
<i>Jinan</i>	urban	2016.04.12-2016.04.27	spring	9	1, 2, 3, 4, 5, 6, 7, 8, 9, 10, 11, 12
		2014.09.04-2014.09.21	summer	37	1, 2, 3, 5, 6, 7, 8, 9, 10
		2016.06.27-2016.07.11			1, 2, 3, 4, 5, 6, 7, 8, 9, 10, 11, 12
		2017.10.22-2017.11.01	autumn	20	1, 2, 3, 4, 5, 6, 7, 8, 9, 10, 11, 12
		2013.11.26-2014.01.05	winter	16	1, 2, 3, 4, 5, 6, 7, 8, 9, 10, 11, 12
<i>Guangzhou</i>	urban	2016.02.19-2016.03.07			1, 2, 3, 5, 6, 7, 8, 9, 10
		2017.06.28-2017.07.08	summer	20	1, 2, 3, 4, 5, 6, 7, 8, 9, 10, 11, 12
<i>Nanjing</i>	urban	2017.10.22-2017.10.31	autumn	16	1, 2, 3, 4, 5, 6, 7, 8, 9, 10, 11, 12
<i>Beijing</i>	urban	2018.01.15-2018.01.31	winter	14	1, 2, 3, 4, 5, 6, 7, 8, 9, 10, 11, 12

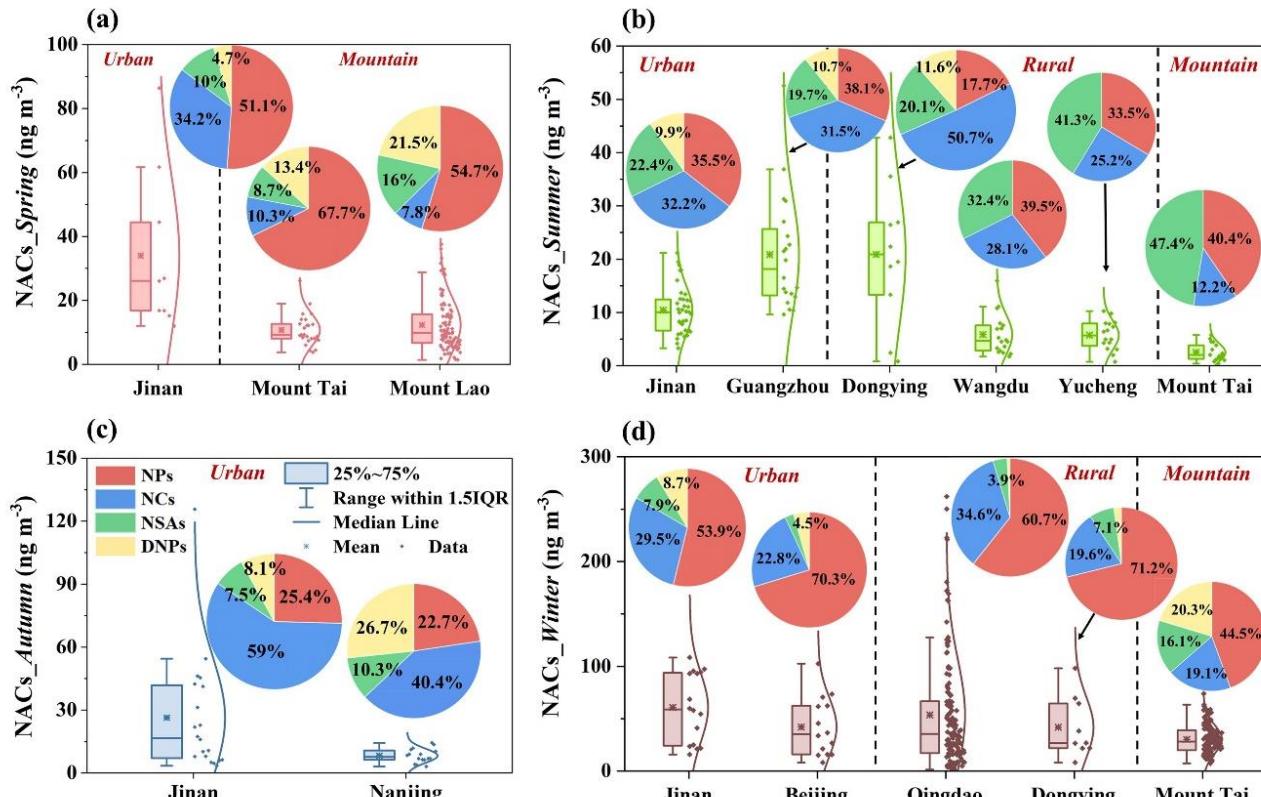
<i>Yucheng</i>	rural	2014.06.09-2014.06.20	summer	16	1, 2, 3, 5, 6, 7, 8, 9, 10
<i>Wangdu</i>	rural	2014.06.19-2014.06.29	summer	18	1, 2, 3, 5, 6, 7, 8, 9, 10
<i>Dongying</i>	rural	2017.06.04-2017.06.15	summer	10	1, 2, 3, 4, 5, 6, 7, 8, 9, 10, 11, 12
		2017.01.15-2017.01.23	winter	9	1, 2, 3, 4, 5, 6, 7, 8, 9, 10, 11, 12
<i>Qingdao</i>	rural	2019.01.10-2019.02.23	winter	132	1, 2, 3, 4, 5, 6, 7, 8, 9, 10, 11, 12
		2019.11.11-2019.12.25			1, 2, 3, 4, 5, 6, 7, 9, 10, 11, 12
		2018.03.22-2018.04.05	spring	25	1, 2, 3, 4, 5, 6, 7, 8, 9, 10, 11, 12
<i>Mount Tai</i>	mountain	2014.07.27-2014.08.06	summer	17	1, 2, 3, 5, 6, 7, 8, 9, 10
		2017.11.28-2017.12.09			1, 2, 3, 4, 5, 6, 7, 8, 9, 10, 11, 12
		2019.12.01-2019.12.31	winter	157	1, 2, 3, 4, 5, 6, 7, 8, 9, 10, 11, 12
<i>Mount Lao</i>	mountain	2021.04.16-2021.05.19	spring	97	1, 2, 3, 4, 5, 6, 7, 8, 9, 10, 11, 12

NOTE: 1 4-nitrophenol (4NP). 2 3-methyl-4-nitrophenol (3M4NP). 3 2-methyl-4-nitrophenol (2M4NP). 4 2,6-dimethyl-4-nitrophenol (2,6DM4NP). 5 4-nitrocatechol (4NC). 6 4-methyl-5-nitrocatechol (4M5NC). 7 3-methyl-6-nitrocatechol (3M6NC). 8 3-methyl-5-nitrocatechol (3M5NC). 9 5-nitrosalicylic acid (5NSA). 10 3-nitrosalicylic acid (5NSA). 11 2,4-dinitrophenol (2,4DNP). 12 4-methyl-2,6-dinitrophenol (4M2,6DNP).

In Figure 2, it is not clear how to understand these box plots. Please clearly state what does the boxes and data dots mean in this figure. For example, in the box plot, which mark represents the mean and median value, which marks show the interquartile range.

**Response:** Thanks for the comment. In the revised manuscript, we have added a clear explanation in figure caption of Figure 2. Specially, the box represents the interquartile range (IQR, i.e., the 25th to 75th percentiles), the line inside the box indicates the median value, and the asterisk marker denotes the mean. The whiskers extend to 1.5 times the IQR, and diamond-shaped markers represent individual data points in the figure.

## Revised Figure 2:



**Line 329: Check typo for “expect winter”. Check for grammar for “which with a little high contribution”.**

**Response:** Thanks for pointing this out. We have corrected the typo and revised the sentence in the revised manuscript.

**Revised sentence in manuscript (Line 351–352):**

“Additionally, the impacts of secondary formation *on* ambient NACs *exhibit* minimal fluctuation across different seasons, *except in* winter, *when* a *slightly higher* contribution (27.8%) *was observed.*”

**Line 345: What do PE and SF mean? To help readers understand the figure clearly, please elaborate here even if they are defined elsewhere.**

**Response:** Thanks for the comment. In the revised manuscript, we have added explicit definitions of the abbreviations “PE” (Primary Emissions”) and “SF” (Secondary Formation) at line 566 to enhance clarity and improve the readability of the figure.

**Revised sentence in manuscript (Line 370):**

“*PE*” and “*SF*” refer to primary emissions and secondary formation, respectively.”

## Reference

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