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_3 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/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 $PM_{2.5}$ 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 Bejing, 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 Bejing, 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_2, NO_2, and O_3. Was NO measured? Usually, NO and NO_2 are measured together by gas analyzers.

Response: Thanks for the comment. In this work, NO and NO_2 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 (SO_2, *NO*, NO_2, and 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 ($P_{2NP} = 1.5 \times 10^{-4}$ atm and $H_{2NP} = 81.1$ M atm^{-1}) compared to 4NP ($P_{4NP} = 6.6 \times 10^{-7}$ atm and $H_{4NP} = 3.0 \times 10^{5}$ 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 (±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 ±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 ±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 ±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, NO_2, and O_3). The optimal number of factors was determined to be four based on the ratio of Q_{true} to Q_{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, NO_2, and 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 $PM_{2.5}$ 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 ± 32.7 ng m^{-3} across four seasons at eleven sampling sites."

Revised Table 1:

Sampling site	Season	ΣNACs	SO_2 (ppbv)	NO_2 (ppbv)	O_3 (ppbv)	CO (ppbv)	T (°C)	RH (%)
Jinan	Spring	**34.0 ± 24.0**	13.7 ± 7.7	43.7 ± 23.2	79.3 ± 19.9	920.2 ± 307.1	20.1 ± 2.5	37.2 ± 13.5
	Summer	**10.4 ± 4.5**	14.7 ± 14.7	26.7 ± 13.7	42.6 ± 26.3	1049.2 ± 573.5	24.0 ± 4.3	66.9 ± 16.4
	Autumn	**26.3 ± 27.9**	4.4 ± 1.8	35.2 ± 15.8	21.7 ± 14.5	812.1 ± 354.5	11.7 ± 3.0	44.8 ± 12.2
	Winter	**60.7 ± 31.9**	21.4 ± 9.6	26.3 ± 12.1	30.2 ± 17.6	1053.0± 403.3	8.6 ± 3.8	36.3 ± 11.7
Guangzhou	Summer	**19.8 ± 10.5**	3.0 ± 0.5	20.0 ± 3.8	13.0 ± 13.2	566.2 ± 82.0	27.1 ± 3.0	79.3 ± 11.2
Nanjing	Autumn	**8.2 ± 3.3**	3.4 ± 0.8	30.2 ± 11.0	23.5 ± 15.8	529.4 ± 126.2	14.1 ± 3.3	69.6 ± 15.2
Beijing	Winter	**42.1 ± 27.1**	3.7 ± 3.0	21.1 ± 13.2	21.2 ± 9.9	691.0 ± 489.6	-3.3 ± 4.4	36.4 ± 13.5
Yucheng	Summer	**5.8 ± 2.7**	3.2 ± 3.0	20.9 ± 12.5	45.9 ± 18.9	665.8 ± 146.8	24.5 ± 3.5	69.3 ± 15.3
Wangdu	Summer	**5.9 ± 3.7**	7.0 ± 5.6	14.2 ± 7.6	56.9 ± 23.3	521.2 ± 203.9	27.0 ± 4.4	55.4 ± 18.1
Dongying	Summer	**20.9 ± 12.5**	3.6 ± 1.5	5.1 ± 2.2	77.0 ± 28.5	478.2 ± 173.0	27.7 ± 3.6	60.2 ± 11.9
	Winter	**41.7 ± 27.6**	4.6 ± 3.6	11.6 ±5.2	21.8 ± 7.1	1494.8 ± 553.9	-2.6 ± 1.9	76.9 ± 13.7
Qingdao	Winter	**53.6 ± 53.2**	3.7 ± 2.1	16.8 ± 9.3	22.2 ± 11.1	757.2 ± 382.5	4.0 ± 5.5	64.3 ± 18.0
Mount Tai	Spring	**10.8 ± 4.9**	2.1 ± 1.4	2.1 ± 1.3	72.7 ± 8.9	445.1 ± 121.3	8.5 ± 4.0	67.4 ± 18.5
	Summer	**2.5 ± 1.6**	2.6 ± 2.0	2.7 ± 0.8	70.3 ± 18.6	331.6 ± 148.9	19.7 ± 2.6	86.9 ± 8.9
	Winter	**30.3 ± 13.6**	2.0 ± 1.3	4.2 ± 2.7	40.9 ± 7.6	308.2 ± 168.3	-3.8 ± 3.3	51.8 ±20.5
Mount Lao	Spring	**12.3 ± 8.3**	1.0 ± 0.8	7.7 ± 3.7	50.3 ± 12.2	273.0 ± 99.2	16.7 ± 3.6	56.0 ± 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
Jinan	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
		2016.02.19-2016.03.07			1, 2, 3, 5, 6, 7, 8, 9, 10
Guangzhou	urban	2017.06.28-2017.07.08	summer	20	1, 2, 3, 4, 5, 6, 7, 8, 9, 10, 11, 12
Nanjing	urban	2017.10.22-2017.10.31	autumn	16	1, 2, 3, 4, 5, 6, 7, 8, 9, 10, 11, 12
Beijing	urban	2018.01.15-2018.01.31	winter	14	1, 2, 3, 4, 5, 6, 7, 8, 9, 10, 11, 12

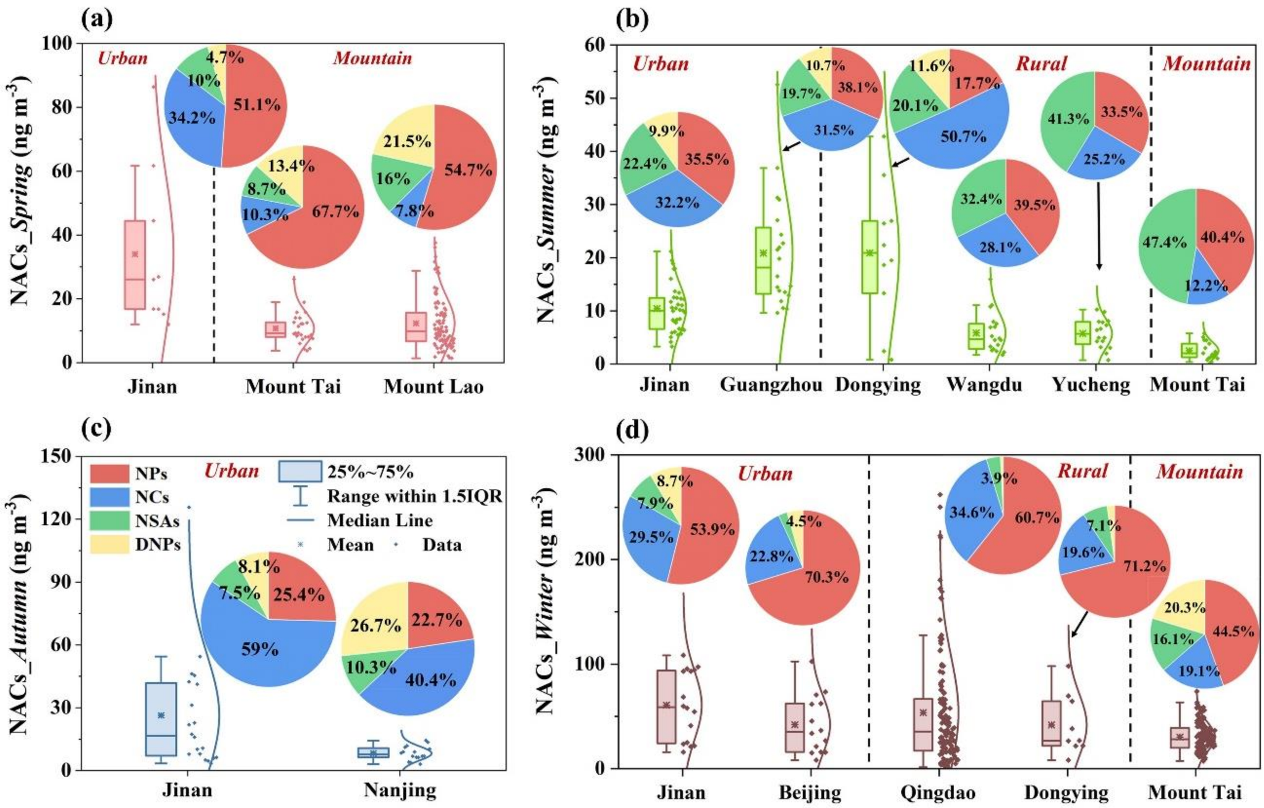
Yucheng	rural	2014.06.09-2014.06.20	summer	16	1, 2, 3, 5, 6, 7, 8, 9, 10
Wangdu	rural	2014.06.19-2014.06.29	summer	18	1, 2, 3, 5, 6, 7, 8, 9, 10
Dongying	rural	2017.06.04-2017.06.15	summer	10	1, 2, 3, 4, 5, 6, 7, 8, 9, 10, 11, 12
		2017.01.15-2017.01.23	winter	9	1, 2, 3, 4, 5, 6, 7, 8, 9, 10, 11, 12
Qingdao	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
Mount Tai	mountain	2018.03.22-2018.04.05	spring	25	1, 2, 3, 4, 5, 6, 7, 8, 9, 10, 11, 12
		2014.07.27-2014.08.06	summer	17	1, 2, 3, 5, 6, 7, 8, 9, 10
		2017.11.28-2017.12.09	winter	157	1, 2, 3, 4, 5, 6, 7, 8, 9, 10, 11, 12
		2019.12.01-2019.12.31			1, 2, 3, 4, 5, 6, 7, 8, 9, 10, 11, 12
Mount Lao	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

Rubio, M. A., Lissi, E., Herrera, N., Perez, V., and Fuentes, N.: Phenol and nitrophenols in the air and dew waters of Santiago de Chile, Chemosphere, 86, 1035-1039, https://doi.org/10.1016/j.chemosphere.2011.11.046, 2012.

Wang, H., Gao, Y., Wang, S., Wu, X., Liu, Y., Li, X., Huang, D., Lou, S., Wu, Z., Guo, S., Jing, S., Li, Y. J., Huang, C., Tyndall, G., Orlando, J. J., and Zhang, X.: Atmospheric processing of nitrophenols and nitrocresols from biomass burning emissions, Journal of Geophysical Research: Atmospheres, 125, e2020JD033401, https://doi.org/10.1029/2020JD033401, 2020.

Wang, Z., Zhang, J., Zhang, L., Liang, Y., and Shi, Q.: Characterization of nitroaromatic compounds in atmospheric particulate matter from Beijing, Atmos. Environ., 246, 118046, https://doi.org/10.1016/j.atmosenv.2020.118046, 2021.

Response (in blue) to Reviewer's comments #2

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 #2:

The manuscript investigated the sources and drivers of particulate nitro-aromatic compounds (NACs) in eastern China using a combination of machine learning and receptor modelling. The study's main focus is how primary emissions, secondary formation, and meteorological factors contribute to ambient NAC levels across different locations and seasons. The authors proposed an ensemble machine learning (EML) model coupled with SHAP (SHapley Additive exPlanation) values and a PMF (Positive Matrix Factorization) source apportionment to interpret NAC variations. Eleven sampling sites (urban, rural, mountain) over multiple seasons provide a robust dataset of NAC concentrations and related variables. The EML model achieves high predictive performance (as can be expected from statistical modelling). The authors conclude that strengthened control of combustion emissions is necessary to mitigate particulate NAC pollution, as their modelling highlights the outsized role of human sources even in a region with complex meteorological and secondary processes.

Overall, this work is important. It extends existing literature on NAC sources (which previously relied on linear models or standalone PMF) by providing a more interpretable quantification of each factor's contribution. The study is well grounded in current literature and clearly exhibits its novelty by bridging source apportionment with explainable AI. A few methodological clarifications and edits (detailed below) could further strengthen the work before this paper could be submitted.

We sincerely thank the review for the valuable feedback that we have used to improve the quality of our manuscript. According to the comments, we have made extensive modifications to this manuscript to make results convincing. In the revised version, the reviewer comments are laid out below in **bold black** font. Below, we provide a point-to-point response to each comment. Our response is given in blue and changes to our manuscript are all highlighted by using *blue italic* text. We have tried our best to improve the manuscript and we hope the revision would satisfactorily address the comments and concerns of the reviewer.

Furthermore, we would like to show the details as below:

1. I agree that the ensemble machine learning approach is appropriate for capturing complex nonlinear relationships, but some details would benefit from clarification to enhance confidence in the results. The authors note an 80/20 random split with cross-validation, but given data from multiple sites and seasons, it would be helpful to discuss whether any site-specific bias could affect the model. If, for instance, all data from a particular location or season mostly fell into the training set, the reported high R2 might not fully reflect generalizable performance. An ideal approach (if data allow) would be to test the model's predictive skill in a leave-one-site-out or leave-one-season-out manner to ensure it generalizes across different scenarios.

Response: We appreciate the reviewer's insightful comment regarding the potential influence of site-specific or seasonal bias in our model evaluation. To address this concern and rigorously assess the spatial generalizability of our ensemble machine learning (EML) model, we performed a leave-one-site-out cross-validation analysis.

Under this cross-validation scheme, data from each site were systematically withheld from model training in turn, and predictions were made exclusively on the excluded site. The procedure ensured a strict separation between training and testing data, thereby providing an unbiased estimate of model transferability across different scenario.

As shown in the revised manuscript (Figure S7), the leave-one-site-out results demonstrated good predictive performance, with an overall R^2 of 0.84 and a regression slope of 0.92 between observed and predicted NACs concentrations, which further indicates that the model is generalizable rather than a location-specific model. The relevant description has been incorporated into the revised manuscript accordingly.

Revised sentence in manuscript (Line 212–216):

"*To further evaluate the generalizability of the EML model, a leave-one-site-out cross-validation approach was implemented. The data from each site were iteratively excluded from model training and used exclusively for testing, ensuring complete independence between training and testing sets. The results show that this model exhibits robustness and transferability rather than limited to specific scenarios (see Fig. S7).*"

Added Figure S7 in Supporting Information:

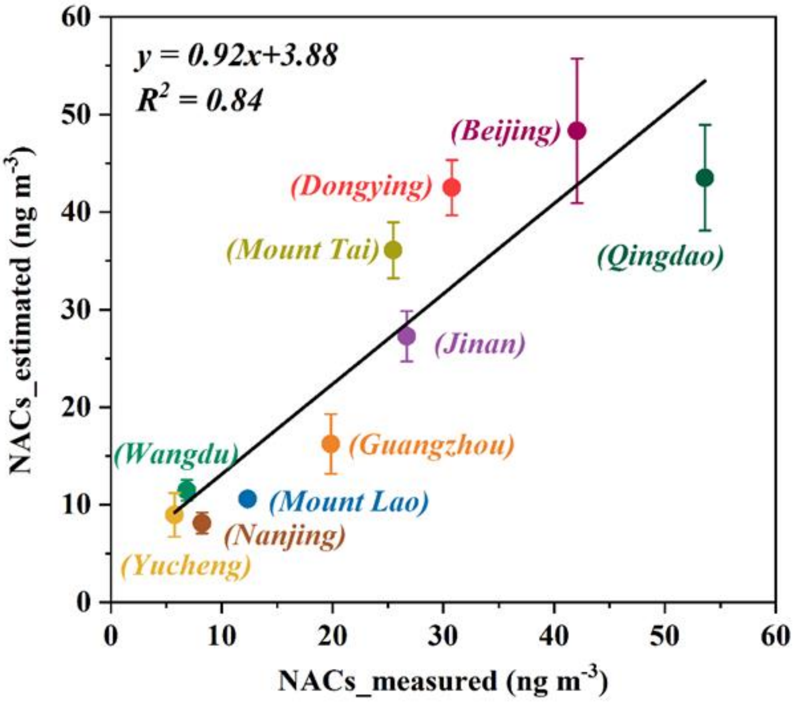


Figure S7. Comparison of observed and simulated NACs at different sites with a leave-one-site-out cross-validation approach.

While the integration of PMF source contributions as input features is innovative, this could introduce circular reasoning if not carefully handled – since NAC concentrations themselves (via their speciation) inform the PMF factors. The authors should reassure that using PMF outputs (four source factor contributions) as predictors does not inadvertently "double count" NAC information. One way to address this would be to emphasize that the ML model's target was the total NAC (or NAC subgroups) concentration and that PMF factors, being based on species patterns, serve as

independent explanatory variables capturing source-type influences. Clarifying these points will help readers understand the modelling strategy and trust that the conclusions (e.g., anthropogenic share of ~49%) are data-driven and not an artifact of the model design.

Response: Thanks for the reviewer's valuable comment regarding the potential risk of circular reasoning due to the integration of PMF source contributions as input features in the ML model. In this study, the target variable of the ML model is the total concentration of NACs, which includes not only the NACs species used in the PMF model (*i.e.*, 4NP, 3M4NP, 2M4NP, 4NC, 4M5NC, 3M6NC, 5NSA, 3NSA), but also other NACs that were not included in the PMF input matrix (as shown in Table S1). Therefore, there is no direct overlap between the target variable and the PMF input.

Additionally, the PMF outputs are composite source-type signatures derived from the covariation of these eight NACs and tracer gases, rather than reconstructions of individual NAC concentrations.

Importantly, in the ML modeling process, we used only the PMF-derived source contributions as input features, and no individual NAC concentrations were directly included. As a result, the ML model avoids any potential data leakage or double-counting of NACs, which further supports the robustness of the conclusion regarding anthropogenic influence.

In the revised manuscript, we have added relevant sentence and provided a comprehensive description of the data-driven modeling framework on PMF and ML.

Revised sentence in manuscript (Line 192–198):

"*The* dataset (613 rows) *used* for the four ML algorithms *consisted of eleven parameters as inputs, including PMF-derived source contributions,* meteorological conditions (T, BLH, RH, SSR, WS_H, and WS_V), and heterogeneous reaction represented by the aerosol surface area *(Sa), all of which influence the sources and sinks of NACs. To avoid circular reasoning, the ML model was constructed to predict the total concentration of NACs as target variable. The four PMF-derived source contribution factors, which serve as independent explanatory variables capturing source-type influences, were used as input features instead of individual NAC species. This approach ensures a clear separation between PMF inputs and the ML target, effectively preventing data leakage or double counting*."

Added Table S1 in Supporting Information:

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

Sampling site	Site type	Sampling period	Season	Number of samples	Detected species
Jinan	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
		2016.02.19-2016.03.07			1, 2, 3, 5, 6, 7, 8, 9, 10
Guangzhou	urban	2017.06.28-2017.07.08	summer	20	1, 2, 3, 4, 5, 6, 7, 8, 9, 10, 11, 12
Nanjing	urban	2017.10.22-2017.10.31	autumn	16	1, 2, 3, 4, 5, 6, 7, 8, 9, 10, 11, 12
Beijing	urban	2018.01.15-2018.01.31	winter	14	1, 2, 3, 4, 5, 6, 7, 8, 9, 10, 11, 12
Yucheng	rural	2014.06.09-2014.06.20	summer	16	1, 2, 3, 5, 6, 7, 8, 9, 10
Wangdu	rural	2014.06.19-2014.06.29	summer	18	1, 2, 3, 5, 6, 7, 8, 9, 10

Dongying	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
Qingdao	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
Mount Tai	mountain	2018.03.22-2018.04.05	spring	25	1, 2, 3, 4, 5, 6, 7, 8, 9, 10, 11, 12
		2014.07.27-2014.08.06	summer	17	1, 2, 3, 5, 6, 7, 8, 9, 10
		2017.11.28-2017.12.09	winter	157	1, 2, 3, 4, 5, 6, 7, 8, 9, 10, 11, 12
		2019.12.01-2019.12.31			1, 2, 3, 4, 5, 6, 7, 8, 9, 10, 11, 12
Mount Lao	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).

2. The claim of "enhanced anthropogenic emissions" driving NAC pollution needs to be positioned against existing studies to ensure the manuscript's novelty is clear. Prior works have already pointed to combustion sources (coal, biomass burning, vehicle emissions) as major NAC contributors. My understanding is, the manuscript's novelty is primarily methodological, and this study's added value lies in quantifying the contributions with a new method and revealing nuanced patterns (like seasonal driver shifts and differences between urban/rural/mountain sites). The authors should ensure readers recognize that the significance lies in using an explainable ML approach to confirm and detail known drivers, rather than in discovering an entirely new source of NACs. This steer will prevent any impression that the study is merely repeating known information, instead of providing new insights into the magnitude and context of anthropogenic influence.

Response: Thanks for the constructive comment. As suggested, we have clarified that the key contribution of this study lies not in identifying new NAC sources, but in employing an explainable ensemble machine learning framework to provide high-resolution, quantitative assessment of the relative importance of known sources under complex atmospheric conditions. By applying this approach across urban, rural, and mountain sites and throughout different seasons, we revealed nuanced shifts in drivers, which have not been captured in prior NAC source apportionment studies.

To ensure the manuscript's novelty is clear, we have changed the title and add some sentences in the revised manuscript:

a. Title has been changed into "Explainable ensemble machine learning revealing *spatiotemporal heterogeneity in driving factors of* particulate nitro-aromatic compounds in eastern China"

b. One supplemental clarification is in the Introduction, to position this study with the context of prior source apportionment research and emphasize our methodological innovation.

c. Another supplemental clarification is in Section 3.2, to highlight the added value of the new approach in refining the understanding of known drivers under different environmental conditions.

Revised sentence in manuscript (Line 84–86 and Line 299–303):

"*This study makes a methodological contribution by employing a novel approach to quantify the seasonal shifts in drivers and spatial variations across urban, rural, and mountain regions in a nuanced manner.*"

"*This enhancement in anthropogenic emissions is consistent with the findings reported in previous NAC* *studies (Wang et al., 2018; Yuan et al., 2021). However, the integration of the explainable EML framework constitutes a methodological advancement by enabling quantitative evaluation of source contributions, thereby providing a more nuanced and context-specific understanding of the driving factors across diverse atmospheric conditions.*"

3. The use of SHAP values is a strong point of the study, but some aspects of the SHAP-based findings could be explained more clearly to avoid confusion. One issue is the meaning of negative SHAP contributions for certain factors. For example, the authors mention that at the mountain site, primary emissions had a mean SHAP contribution of –5.7 ng m^{-3}, which initially sounds like primary sources were somehow reducing NAC levels. The intended meaning is presumably that local primary emissions are minimal at the mountain (so their absence corresponds to lower baseline NAC, hence a negative SHAP relative to other sites). Also for discussions regarding "temperature" and "BLH", providing one or two sentences of intuition (e.g., "a negative SHAP value for a factor means that higher values of that factor are associated with lower NAC concentrations") when introducing the SHAP results would make the explanation more accessible, especially for readers new to SHAP analysis.

Response: Thanks for the review's valuable comment. We have revised the manuscript to clarify the interpretation of SHAP values. Specifically, the negative SHAP contribution of primary emissions at the mountain site reflects minimal local emissions, which results in lower NAC concentrations, rather than indicating an actual reduction in NACs due to these sources. Additionally, we also provided concise explanations for the SHAP interpretations of the input variables. These revisions aim to improve the clarity and accessibility of the SHAP-based analysis, especially for readers who are less familiar with the method.

Revised sentence in manuscript (Line 315–317 and Line 423–424):

"*A positive SHAP value indicates that the variable increases the predicted NAC concentration relative to the baseline, whereas a negative SHAP value suggests that higher values of the variable are associated with a decrease in NAC concentrations.*"

"*This negative value reflects the minimal contribution of local anthropogenic emissions in this region, resulting in lower concentrations of NACs compared to other sites.*"

SHAP can sometimes capture pairwise interactions, the authors could discuss on interactions or co-variability among factors if any were observed. For example, did the authors notice if certain meteorological conditions amplify the effect of emissions (high humidity aiding secondary formation of NACs, etc.)? Ensuring the SHAP results are clearly linked back to physical processes (mixing, photochemistry, emissions timing) will make the conclusions more convincing and useful for policy implications.

Response: Thanks for the insightful comment. In response, we conducted a detailed analysis of pairwise SHAP interaction values among key variables. Notably, a significant interaction between temperature (T) and aerosol surface area (Sa) was identified, as shown in the newly added Figure S8. The interaction pattern indicates that high Sa facilitates NACs formation under low-temperature conditions (T < 10°C), suggesting the enhanced gas-particle partitioning and heterogenous reactions. In contrast, high Sa appears to inhibit NACs formation at high temperature (T > 10°C), potentially due to intensified photochemical reactions shifting towards gas-phase products, high temperature promoting to particle-to-gas partitioning, or dilution effects arising from elevated mixing heights in hot seasons. The temperature-dependent behavior highlights the complex role of heterogenous reaction in atmospheric aerosol formation.

However, no other variable pairs exhibited comparable interaction effects across the dataset. Future research incorporating more comprehensive datasets with machine learning or deep learning model is required to better elucidate the synergistic effects on ambient NAC concentrations.

Revised sentence in manuscript (Line 332–337):

"*Notably, at low temperature (approximately < 10°C), the contribution on NACs exhibited an explosive enhancement, accompanied by a pronounced synergistic effect with higher Sa (Fig. S8), indicating enhanced gas-to-particle partitioning and heterogeneous formation. Conversely, at higher temperature, high Sa appears to suppress NAC formation, possibly as a result of intensified photochemical reactions facilitating gas-phase products, high temperature promoting to the partitioning to particle phase, or dilution effects caused by increased mixing heights in hot seasons.*"

Added Figure S7 in Supporting Information:

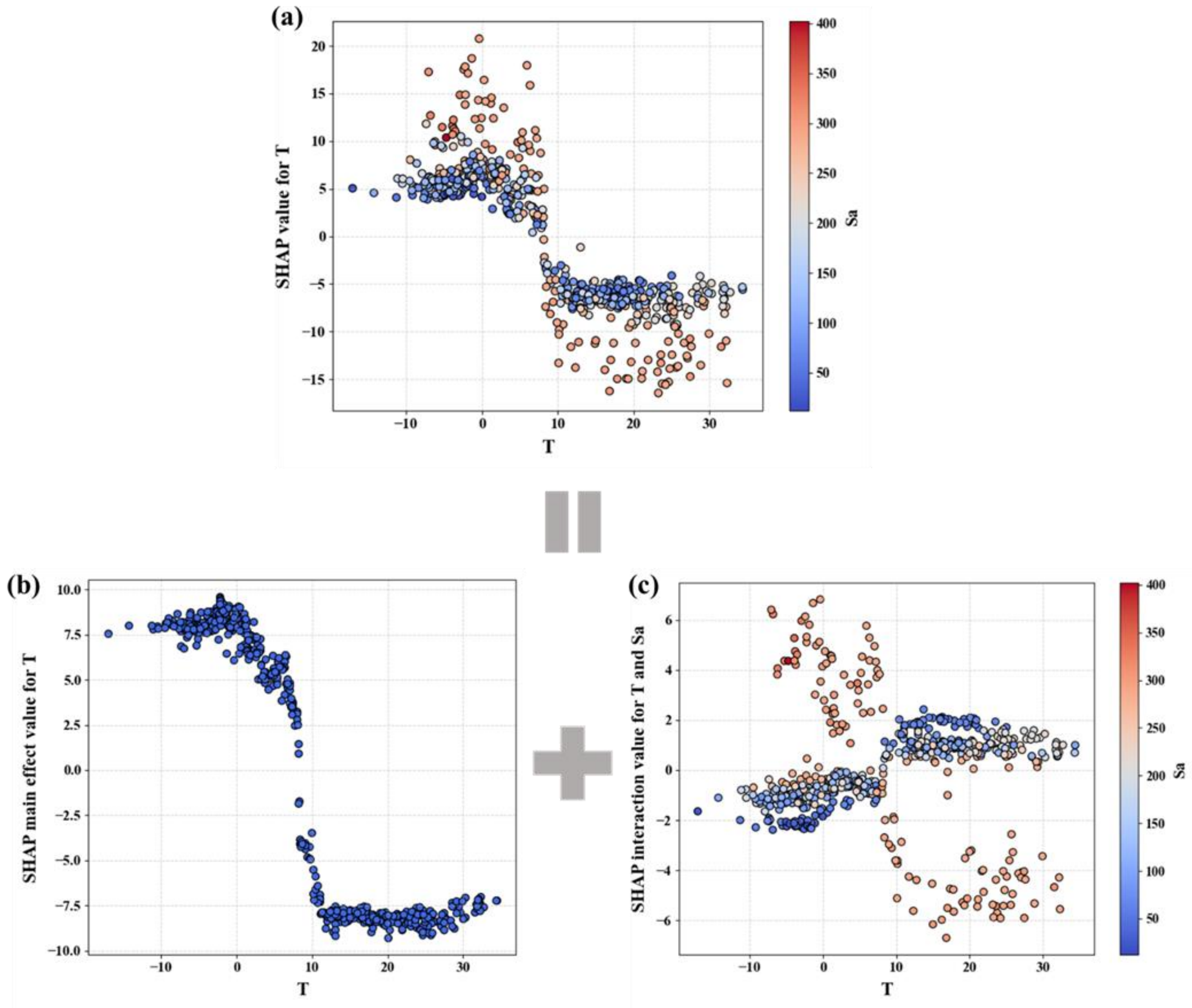


Figure S8. (a) The interaction effect of temperature (T) and aerosol surface area (Sa), (b) the main effects of T on NACS, and (c) the interaction SHAP value between T and Sa shows how the effect of T on NACs varies with Sa.

4. Line 186, the multi-target modelling approach, where NPs, NCs, and NSAs were predicted simultaneously (mentioned in the Methods), is an interesting aspect but is not very prominently discussed in the results. The conclusion hints that different functional groups had different key

drivers (e.g., gas-phase oxidation dominating NSAs). It would strengthen the paper to emphasize these findings a bit more in the Results section 3.3 or 3.4 – for instance, explicitly stating which sources were most important for each NAC subclass. This adds depth to the analysis (showing the model's strength in capturing subtle differences).

Response: Thanks for the comment. As suggested, in the revised manuscript, we have incorporated a summary paragraph at Section 3.3 to explicitly highlight the distinct sources for different NAC subclasses. Specifically, coal combustion was identified as the primary contributor to NPs, biomass burning emerged as the dominant source for NCs, and NSAs were predominantly associated with gas-phase formation. These results demonstrate the model's capacity to resolve nuanced differences in source attribution across functional groups and underscore the importance of implementing targeted control strategies.

Revised sentence in manuscript (Line 395–399):

"*Overall, the results demonstrate that the multi-target EML model effectively captured the distinct source contributions and formation pathways associated with different NAC subclasses. Coal combustion was identified as the most important driver for NPs, biomass burning dominated the formation of NCs, and NSAs were primarily linked to gas-phase formation. These findings highlight the strength of this integrated EML approach in differentiating functional group-specific drivers and emphasize the importance of targeted mitigation strategies for various NAC species.*"

Also, given that the data span 2014–2021, there could be a question that if trends over that period were considered – for example, have emission controls in China over the years impacted NAC levels? This may be outside the scope of the current paper's focus on spatial drivers, but a short note in the discussion could acknowledge that temporal trends were not the focus here (assuming no strong trend was observed after accounting for other factors).

Response: We appreciate the reviewer's thoughtful comment. We appreciate the suggestion to explore potential temporal trends in NAC concentrations, particularly considering the emission control measures implemented in China over the years. We have conducted a thorough analysis of the data spanning from 2014 to 2021, and our results indicate that, under consistent seasonal and site-type conditions, NAC concentrations did not exhibit any significant temporal trends, suggesting that the factors influencing NAC levels during this period were primarily spatial rather than temporal. We have added a brief note in the Discussion section to acknowledge that temporal trends were not the focus of this study, and clarify that no strong significant trends were observed after accounting for other variables.

Revised sentence in manuscript (Line 248–249):

"*Moreover, data from 2014 to 2021 revealed no significant trends in NAC concentrations across the same seasonal and site-type conditions, therefore temporal variation was not considered as a primary focus of this study.*"

Minor issues:

The paper is generally well-written, but a few sentences should be edited for clarity or correctness. Here below are examples but the authors need to read through the manuscript for such minor language issues:

Line 73, "Given the complex nonlinear links… it is necessary to establish an effective and reliable evaluation method to comprehensively understand and assess the importance and contribution of each factor…". This could be broken into two sentences to avoid confusion.

Response: Thanks for the helpful suggestion. In response, we have revised the sentence by dividing it into two parts to enhance clarity.

Revised sentence in manuscript (Line 76–80):

"Given the complex nonlinear links between primary emissions, secondary formation, and meteorological conditions and the ambient particulate NACs, *a clear understanding of the separate role of each factor is challenging. Therefore,* it is necessary to establish an effective and reliable evaluation method to comprehensively understand and assess the importance and contribution of each factor on the abundances of NACs under complicated atmospheric conditions."

Line 258, the phrasing "is in coincided with" is grammatically incorrect.

Response: Thanks for the comment. It has been corrected in the revised manuscript.

Revised sentence in manuscript (Line 272–273):

"The dominance of NPs and NCs in this study *coincides* with *the findings from previous studies* in other locations (Cai et al., 2022; Li et al., 2020c; Wang et al., 2019)."

Line 182, a typo "leaner" should be "learner"

Response: Thanks for the comment. It has been corrected.

Line 389, "Jinan ang Beijng" should be "and Beijing"

Response: Thanks for the comment. It has been corrected.

Line 363, "confirmed by a previous observational study"

Response: Thanks for the comment. It has been corrected.

1. As noted, the manuscript uses many abbreviations (NACs, PMF, EML, SHAP, NP, NC, NSA, BLH, SSR, WS_V, WS_H, etc.). It would be very helpful to provide a list of abbreviations early on to improve readability.

Response: Thanks for the valuable comment. As suggested by the reviewer, a comprehensive list of abbreviations was added early in the manuscript to enhance clarity and ensure the accessibility of the main text.

Abbreviation			
NACs	Nitro-aromatic compounds	**BrC**	Brown carbon
NPs	Nitrophenol and its derivatives	**VOCs**	Volatile organic compounds
NCs	Nitrocatechol and its derivatives	**T**	Temperature
NSAs	Nitrosalicylic acids	**RH**	Relative humidity
DNPs	Dinitrophenol and its derivatives	**SSR**	Surface net solar radiation
4NP	4-nitrophenol	**PMF**	Positive matrix factorization
3M4NP	3-methyl-4-nitrophenol	**PCA**	Principal component analysi
2M4NP	2-methyl-4-nitrophenol	**ML**	Machine learning
2,6DM4NP	2.6-dimethyl-4-nitrophenol	**SHAP**	SHapley Additive exPlanation
4NC	4-nitrocatechol	**EML**	Ensemble machine learning
4M5NC	4-methyl-5-nitrocatechol	**BLH**	Boundary layer height
3M6NC	3-methyl-6-nitrocatechol	**WS_H**	Horizontal wind speed

3M5NC	3-methyl-5-nitrocatechol	**WS_V**	Vertical wind speed
5NSA	5-nitrosalicylic acid	**Sa**	Aerosol surface area
3NSA	3-nitrosalicylic acid	**EDTA**	ethylenediaminetetraacetic acid
2,4DNP	2,4-dinitrophenol	**BB**	Biomass burning
4M2,6DNP	4-methyl-2,6-dinitrophenol	**RF**	Random forest
CC	Coal combustion	**XGBoost**	Extreme gradient boosting
TE	Traffic emission	**LightGBM**	Light gradient boosting machine
GR	Gas-phase reaction	**MLP**	Multilayer perceptron
PE	Primary emission	**R^2**	Coefficient of determination
SF	Secondary formation	**MAE**	Mean absolute error
		RMSE	Root mean squared error

2. Line 95, there is a minor point about terminology. Calling Mount Lao a "mountain" site when it's only 166 m altitude is a bit confusing as Mount Tai is at 1534 m a.s.l. It might be worth clarifying that Mount Lao site is at a lower elevation (perhaps a foothill or a coastal mountain location) to avoid readers questioning if it truly represents a clean mountain background.

Response: Thanks for the comment. In response, we have revised the manuscript to characterize Mount Lao more precisely as a lower-elevation site situated in a coastal mountainous region. This clarification aims to provide a more accurate depiction of the site's geographic and environmental context, and to avoid potential ambiguity regarding its representativeness as a clean mountain background location.

Revised sentence in manuscript (Line 100–102):

"…and two mountain sites: Mount Tai (36.27° N, 117.10° E, 1,534 m a.s.l.), *a typical high-elevation background site;* and Mount Lao (36.15° N, 120.68° E, 166 m a.s.l.), *a lower-elevation site situated in a coastal mountainous region*."

3. Line 184, when talking about the performance of a model, it cannot be validated or verified as natural systems are never closed, it can only be evaluated.

Response: Thanks for the careful comment. Accordingly, we have revised the manuscript to replace "validation" to "evaluation" to ensure accuracy.

4. Linked to point 2, it's needed to ensure Figure (e.g., Figures 4-7) legends and captions fully describe what the plots represent. The caption may list the variables by name (or refer to a legend) so readers don't have to infer abbreviations (e.g., PE, SF, etc).

Response: Thanks for the comment. In accordance with your suggestion, we have revised the captions of Figure 4-7 to explicitly list the variables by name and, where applicable, refer to the corresponding legends to avoid the need for reader to infer abbreviations.

Revised caption of Figure 4 in manuscript (Line 305–313):

"Figure 4: (a) The ranking of the importance for all input variables *("CC": coal combustion; "TE": traffic emission; "T": temperature; "BB": biomass burning; "Sa": aerosol surface area; "GR": gas-phase reaction; "BLH": boundary layer height; "SSR": surface net solar radiation; "RH": relative humidity; "WS_H": horizontal wind speed; "WS_V": vertical wind speed)* calculated via SHAP algorithm (average absolute contribution), (b) the impacts of driving factors on variations of NACs from SHAP analysis during the whole sampling periods *("PE" and "SF" represent primary emissions and* *secondary formation, respectively)*, (c) SHAP summary plots for all samples with the shift in colour of the scatter plot from blue to red indicating an increase in driving factor values, and the relationships between the SHAP values and parameter values for (d) temperature *(T)*, (e) aerosol surface area *(Sa)*, (f) boundary layer height *(BLH)*, and (g) surface net solar radiation *(SSR)* with the right y axis corresponding to the frequency distribution of the measured variables."

Revised caption of Figure 5 in manuscript (Line 368–370):

"Figure 5: The absolute contributions of (a) meteorological conditions, (b) all factors, and (c) primary emissions on the variations of NACs in four seasons from SHAP analysis and box plots with the order of SHAP values for each driving factor in (d) spring, (e) summer, (f) autumn, and (g) winter. *"PE" and "SF" refer to primary emissions and secondary formation, respectively*."

Revised caption of Figure 6 in manuscript (Line 401–406):

"Figure 6: The impacts of primary emissions (PE), meteorological conditions, and secondary formation (SF) on the variations of (a) NPs, (b) NCs, and (c) NSAs from SHAP analysis and relative importance of (d) primary emissions *(including coal combustion (CC), traffic emission (TE), and biomass burning (BB))*, (e) meteorological conditions *(including temperature (T), boundary layer height (BLH), surface net solar radiation (SSR), relative humidity (RH), horizontal wind speed (WS_H), and vertical wind speed (WS_V)*), and (f) secondary formation (*including gas-phase reaction (GR) and heterogeneous reaction represented by aerosol surface area (Sa)*)."

Revised caption of Figure 7 in manuscript (Line 444–448):

"Figure 7: The combined contributions of sources (*i.e.*, coal combustion *(CC)*, traffic emission *(TE)*, and biomass burning *(BB)*), meteorological conditions (*i.e.*, *temperature (*T*)*, *boundary layer height (*BLH*)*, *surface net solar radiation (*SSR*)*, *vertical wind speed (*WS_V*)*, *horizontal wind speed (*WS_H*)*, and *relative humidity (*RH*)*), and secondary formation (*i.e.*, gas-phase reaction *(GR)* and heterogeneous reaction represented by the aerosol surface area *(Sa)*) to the variations of NACs in (a) urban, (b) rural, and (c) mountain areas."