



Supplement of

Light absorption properties and composition of water-soluble brown carbon in North China Plain: implication for an enhancing role of nitrogenous organic compounds

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Sect. S1 Positive Matrix Factorization (PMF) source apportionment

PMF, as a receptor model, decomposes the sample matrix into two matrices (factor contributions and factor profiles), and has been widely used in source apportionment of atmospheric pollutants (Wu et al., 2024; Li et al., 2023). Details regarding the PMF model are available on the EPA website (<https://www.epa.gov/air-research/epa-positive-matrix-factorization-50-fundamentals-and-user-guide>, last access: 16 May 2025). Herein, the EPA PMF 5.0 was employed to conduct source apportionment of organic matter (OM), with input parameters including the mass concentrations of major species (OM, EC, WSOC, NO₃⁻, NH₄⁺, Mg²⁺, Ca²⁺, WSOC, NACs), and organic markers (benzo(b)fluoranthene (BbF), benzo(e)pyrene (BeP), indeno(1,2,3-c,d)pyrene (IP), levoglucosan). The samples from all the sampling sites were added together as one data matrix, and the model was run numerous times with 3–7 factors to obtain the optimal solution. Base on the Q value (Q true:14982 and Q robust:14478) and interpretability, three factors were obtained as the optimal solution, which are indicative of a strong correlation between input species and simulated ones (R²>0.83).

Sect. S2 Estimation of secondary WSON

To obtain the relative contributions of the primary emission and secondary formation to the particulate WSON, a tracer method analogous to the EC-based approach commonly used for secondary organic carbon was employed here. The equation is as follow:

$$\text{WSON}_{\text{sec}} = \text{WSON} - \left[\frac{\text{WSON}}{\text{EC}} \right]_{\text{pri}} \times \text{EC} \quad \text{Eq. S1}$$

Here, WSON, WSON_{sec} and EC denote the mass concentrations of the particle-

phase WSON, secondary WSON and EC, respectively. Herein, EC serves as the tracer of the combustion source. $[\frac{WSON}{EC}]_{pri}$ is estimated by fitting the lowest 15% of $[\frac{WSON}{EC}]$ data points. To assess the robustness of the inferred secondary WSON, we further quantified the relative differences (RD) in WSON/EC ratios between the 15th percentile and other alternative percentiles, and all RD values were less than 20% (Table S3). This result indicated that the estimation of secondary WSON was robust. Additionally, the primary WSON (WSON-WSONsec) robustly and positively correlated with CO that mainly originates from biomass burning and fossil fuel combustion. Thus, the EC-based method can capture the variation of secondary WSON, despite potential overestimation caused by non-combustion primary WSON emissions. Even so, the derived secondary fractions should be interpreted as semi-quantitative estimates rather than precise source apportionments, due to the inherent uncertainty of the method

$$RD = \frac{|C_{x\%} - C_{15\%}|}{(C_{x\%} + C_{15\%})/2} \times 100\% \quad \text{Eq. S2}$$

Where, $C_{x\%}$ and $C_{15\%}$ denote the lowest X% and 15% of $[\frac{WSON}{EC}]$, respectively. Above method was further extended to estimate the secondary NACs in the present study.

Sect. S3 Generalized Additive Model (GAM)

As a nonparametric modeling framework, GAM model is inherently data-driven rather than bound by statistical distribution constraints. Thus, it excels at capturing complex nonlinear relationships between explanatory variables and the target response, with flexible functional response patterns adaptable to various research scenarios. Herein, the WSONsec is response variable, while the explanatory variables include NH_4^+ , NO_2 , O_3 , ALWC, pH and meteorological factors, consistent with the setup of the RF model. GAM analyses were conducted using R package version 4.2.1 and its mgcv package (Wood, 2011). Model parameterization and analytical methods can be found in our pervious study (Lv et al., 2023), and the p-value, adjusted R^2 , and explained deviance given by GAM were adopted to evaluate factor significance for WSONsec and model fitting accuracy. As summarized in Table S4, the explained deviance was 88.0% with

adjusted $R^2=0.87$. A strong linear correlation was observed between the observed and simulated values, and all explanatory variables reached statistical significance. Collectively, these statistical metrics verify that the GAM can effectively reproduce the variability of WSONsec. As illustrated in the Figure S9, the GAM outputs agree well with the RF results, which also indicated the significant impacts of NH_4^+ and ALWC on the variation of WSONsec.

To quantify multicollinearity among explanatory variables, the variance inflation factor (VIF) was applied here, where a higher VIF value denotes stronger collinearity of one variable with the others. According to the empirical criterion (Lin et al., 2018), VIF exceeding 4.0 implies severe collinearity and unstable regression fitting. From Figure S9, we noted that the VIF values of all explanatory variables were <4 , implying a weak multicollinearity among these factors. Finally, additional sensitivity tests were carried out, in which all other explanatory variables were held constant while only NH_4^+ and NACs were proportionally reduced, to further evaluate their individual impacts on WSONsec. As demonstrated in Figure S9d, the variation of the predicted WSONsec is far more sensitive to change, further highlighting the key importance of NH_4^+ on WSONsec. Notably, these model-based results may not fully capture actual atmospheric responses and should therefore be further corroborated by chemical evidences.

Sect. S4 Theoretical estimation of chemical forms of SNA

To reveal the major SNA forms during the sampling period, the theoretical ammonium concentration was calculated according to thermodynamic equilibrium with the atmospheric sulfate and nitrate levels. The theoretical ammonium levels were calculated as follows:

$$\text{NH}_4^+_{\text{calculate}} = \left(\frac{[\text{SO}_4^{2-}]}{48} + \frac{[\text{NO}_3^-]}{62} \right) \times 18 \quad \text{Eq. S3}$$

$$\text{NH}_4^+_{\text{calculate}} = \left(\frac{[\text{SO}_4^{2-}]}{96} + \frac{[\text{NO}_3^-]}{62} \right) \times 18 \quad \text{Eq. S4}$$

where $[\text{SO}_4^{2-}]$ and $[\text{NO}_3^-]$ refer to atmospheric concentrations measured in $\mu\text{g}/\text{m}^3$. The NH_4^+ calculate is achievable through Equation (3) if $(\text{NH}_4)_2\text{SO}_4$ and NH_4NO_3 serve as the dominant species. Conversely, equation (4) demonstrates that NH_4HSO_4 and NH_4NO_3 are abundantly present in the analyzed aerosols.

Sect. S5 Aerosol liquid water content (ALWC), in-situ pH and gas-to-particle-phase partitioning coefficient of ammonia

A robust thermodynamic model (ISORROPIA-II) was employed to theoretically estimate the aerosol liquid water content (ALWC) and in-situ pH in $\text{PM}_{2.5}$, which has been widely used in previous studies (Song et al., 2018; Pye et al., 2020). By combining the actual temperature (T) and relative humidity (RH) in the atmosphere and the water-soluble ions composition, the ALWC and H^+ loads can be simulated by running the model under the metastable mode. And the in-situ pH can be calculated by Eq. S5.

$$\text{pH} = \frac{[\text{H}^+] \times 1000}{\text{ALWC}} \quad \text{Eq. S5}$$

Where the units for H^+ and ALWC are the $\mu\text{g m}^{-3}$. Gaseous ammonia was not detected in this study, thus, which may lead to overestimation of aerosol acidity. Furthermore, the gas-to-particle-phase partitioning coefficient of ammonia ($\epsilon_{\text{NH}_4^+}$) was theoretically calculated following the method reported by Nah et al. (2018) and Guo et al. (2017):

$$\epsilon(\text{NH}_4^+) \cong \frac{\frac{\gamma_{\text{H}^+} 10^{-\text{pH}}}{\gamma_{\text{NH}_4^+}} \text{H}_{\text{NH}_3}^* \text{ALWC} \times \text{R} \times \text{T} \times 0.987 \times 10^{-14}}{1 + \frac{\gamma_{\text{H}^+} 10^{-\text{pH}}}{\gamma_{\text{NH}_4^+}} \text{H}_{\text{NH}_3}^* \text{ALWC} \times \text{R} \times \text{T} \times 0.987 \times 10^{-14}} \quad \text{Eq. S6}$$

$$\ln(\text{H}_{\text{NH}_3}^*) = 25.393 - 10373.6 \left(\frac{1}{\text{T}_r} - \frac{1}{\text{T}} \right) + 4.131 \left(\frac{\text{T}_r}{\text{T}} - \left(1 + \ln \left(\frac{\text{T}_r}{\text{T}} \right) \right) \right) \quad \text{Eq. S7}$$

Here, γ_{H^+} and $\gamma_{\text{NH}_4^+}$ are the activity coefficients of H^+ and NH_4^+ , respectively; and both were assumed to be 1 in this study. $\text{H}_{\text{NH}_3}^*$ is equilibrium constant calculated by Eq S7; R is the gas constant. For the the $\epsilon_{\text{NH}_4^+}$ calculation, the pH value was adjusted slightly, while parameters adopted the average values obtained during the campaign.

Table S1 Information of nitroaromatic compounds detected in this study.

Compounds	Molecular	Formula	CAS number	Abbreviation
4-Nitrophenol	139.11	C ₆ H ₅ NO ₃	100-02-7	4NP
3-Methoxy-4-nitrophenol	153.14	C ₇ H ₇ NO ₃	2581-34-2	3M4NP
2, 4-Dinitrophenol	184.11	C ₆ H ₄ N ₂ O ₅	51-28-5	2,4DNP
4-Nitroguaiacol	169.14	C ₇ H ₇ NO ₄	3251-56-7	4NGA
5-Nitroguaiacol	169.14	C ₇ H ₇ NO ₄	636-93-1	5NGA
4-Nitrocatechol	155.11	C ₆ H ₅ NO ₄	59030-13-6	4NC
4-Methyl-5-nitrocatechol	169.13	C ₇ H ₇ NO ₄	68906-21-8	4M5NC
3-Nitrosalicylic acid	183.12	C ₇ H ₅ NO ₅	85038-1	3NSA
5-Nitrosalicylic acid	183.12	C ₇ H ₅ NO ₅	96-97-9	5NSA

Table S2 Statistical metrics for evaluation of RF model performance

Statistical metrics	
R ²	0.94
MSE	0.024
RMSE	0.16
MAE	0.1

Note: MSE: mean square error; RMSE: root-mean-square error; MAE: mean absolute error.

Table S3 The abs_{365} and MAE_{365} of water-soluble BrC in $\text{PM}_{2.5}$ among different cities in the world.

Location	Year	abs_{365} (Mm^{-1})	MAE ($\text{m}^2 \text{g}^{-1}$)	Reference
Beijing	2011	10.2 ± 6.9	1.22 ± 0.11	Cheng et al. (2016)
	2013	14 ± 5.2	1.54 ± 0.16	Yan et al. (2015)
	2018	12.5	1.2	Li et al. (2020b)
	2021	3.5 ± 3.4	1.1 ± 0.4	You et al. (2024)
	2023	2.3 ± 1.9	0.7 ± 0.5	This study
Tianjin	2018	7.9 ± 5	1.04 ± 0.3	Chen et al. (2024)
	2020	7.5 ± 5	1.27 ± 0.3	
	2023	5.7 ± 3.8	1.12 ± 0.05	This study
Handan	2018	16.8 ± 2.7	1.22 ± 0.3	Chen et al. (2024)
	2020	11.1 ± 2.3	1.1 ± 0.27	
	2023	8.2 ± 5.0	1.05 ± 0.02	This study
Jinnan	2014	10.3 ± 6.5	0.88 ± 0.4	Shi et al. (2023)
	2018	5.42	0.65	Zhang et al. (2022)
	2023	6.9 ± 3	1.4 ± 0.02	This study
Nanjing	2018	9.47 ± 5.83	1.27 ± 0.34	Bao et al. (2022)
	2020	3.12 ± 2.41	0.58 ± 0.26	Feng et al. (2023)
	2021	4.73 ± 1.97	1.23 ± 0.26	Cui et al. (2025)
	2023	3.73 ± 2.46	0.62 ± 0.18	Zhou et al. (2025)
Xi'an	2012	22.6 ± 12.3	1.31 ± 0.4	Lei et al. (2023)
	2016	28 ± 16	1.2 ± 0.06	Wu et al. (2020)
	2018	17.8 ± 9.6	2.0 ± 0.6	Li et al. (2020a)
	2021	6.7 ± 1.4	0.83 ± 0.22	Wang et al. (2025)
Guangzhou	2012	3.57 ± 1.34	0.81 ± 0.16	Liu et al. (2018)
	2016	5.8	0.98	He et al. (2023)
	2018	2.5 ± 2	1 ± 0.21	Zou et al. (2023)
	2019	4.3	0.34	Wang et al. (2022)
Seoul, Korea	2012	7.31	1.02	Kim et al. (2016)
Leipzig, Germany	2014	3.91 ± 2.5		Teich et al. (2017)
Milan, Italy	2016	3.0 ± 1.3		Gilardoni et al. (2020)
Pasadena, USA*	2010	0.88 ± 0.71	0.77	Zhang et al. (2011)
Atlanta, USA*	2010	0.61 ± 0.38	1.15	
Southeastern US*	2013	0.42 ± 0.43	0.84 ± 0.37	Xie et al. (2019)

Note: * indicates the samples collected in summer

Table S4 Statistical metrics for evaluating the EC-based method of secondary WSON and NACs

Sites	WSON			NACs		
	RD _{15% vs. 10%}	RD _{15% vs. 20%}	R ² WSON _{pri&CO}	RD _{15% vs. 10%}	RD _{15% vs. 20%}	R ² NAC _{Spri&CO}
BJ	8±13	19±13	0.52**	20±15	9±9	0.68**
TJ	12±16	18±13	0.78**	16±17	17±20	0.73**
LC	12±23	18±24	0.74**	14±17	12±8	0.70**
HD	17±11	19±14	0.30*	12±4	18±20	0.56**
JN	15±30	8±7	0.37**	27±12	19±13	0.58**

Note: RD_{15% vs. 10%} indicates the relative difference between WSON/EC results from the 15% and 10% lowest percentiles; RD_{15% vs. 20%} indicates the relative difference between WSON/EC results from the 15% and 20% lowest percentiles; R² WSON_{pri&CO} denotes the correlation coefficient between primary WSON and CO; R² NAC_{Spri&CO} denotes the correlation coefficient between primary NACs and CO (**: p<0.01; *:p<0.05).

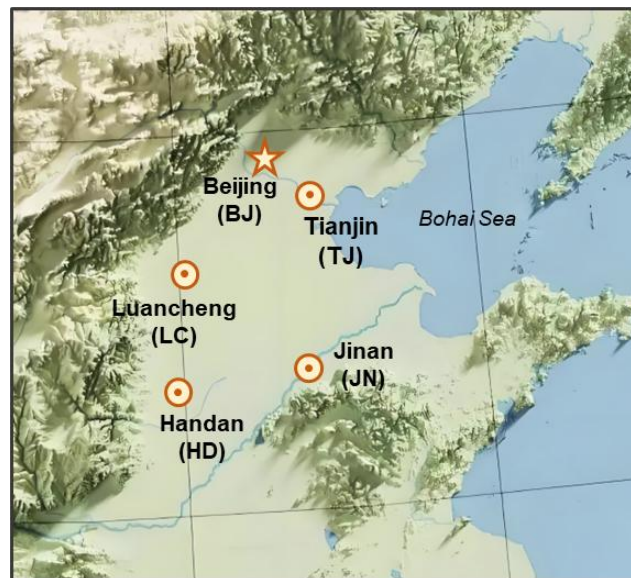


Figure S1 Topographic distribution the sampling sites in the NCP. (The maps are the reproductions from ©Baidu (<https://image.baidu.com/>, last access: 16 July 2025))

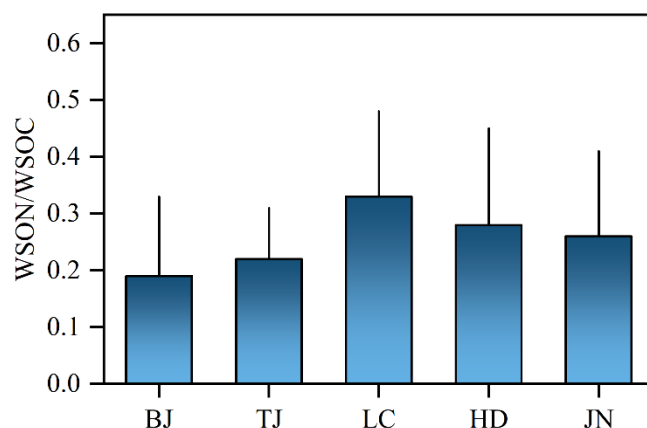


Figure S2 The WSON/WSOC ratio in different sampling sites.

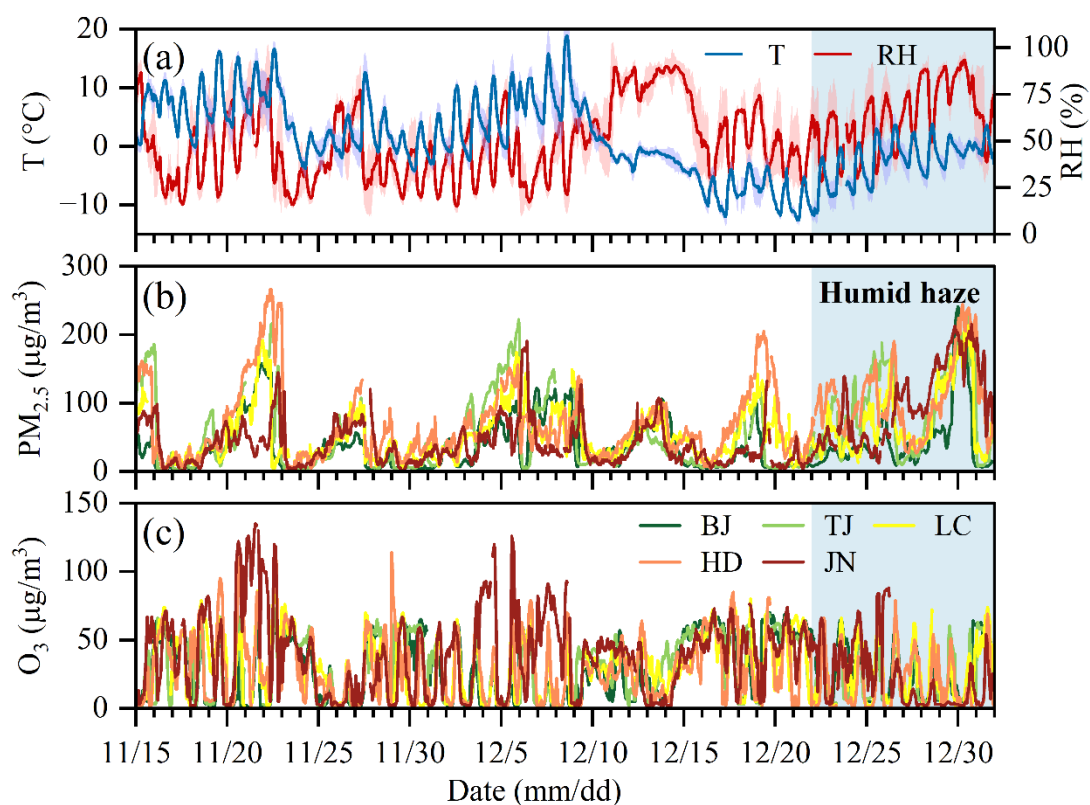


Figure S3 Temporal variations of meteorological factors, PM_{2.5} and O₃ at five sampling sites. (The shade indicates the humid haze event.)

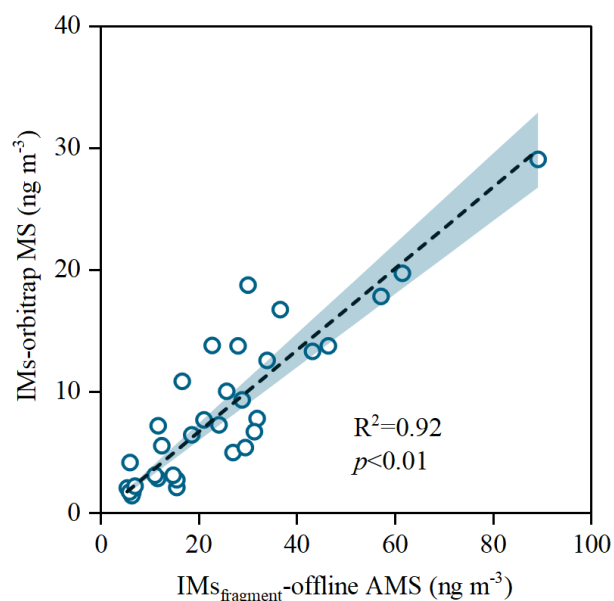


Figure S4 Scattering plot of IMs-related fragments derived from offline AMS (unpublished data) and IMs concentration determined by orbitrap mass spectrometry (Xiao et al. (2025)).

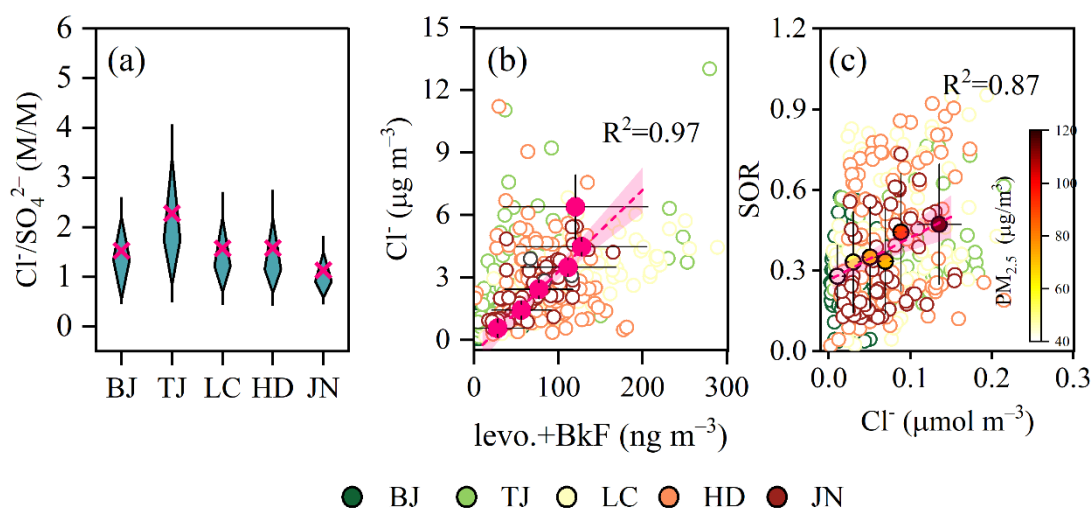


Figure S5 The source of the abundant chloride across the NCP and its effect on sulfate formation. **(a)** Molar ratio of Cl^- and SO_4^{2-} ; Linear fit regression for Cl^- with BkF+ levoglucosan **(b)** and SOR **(c)**. (Circles in **(b)** and **(c)** show grouped mean statistics over all sampling site)

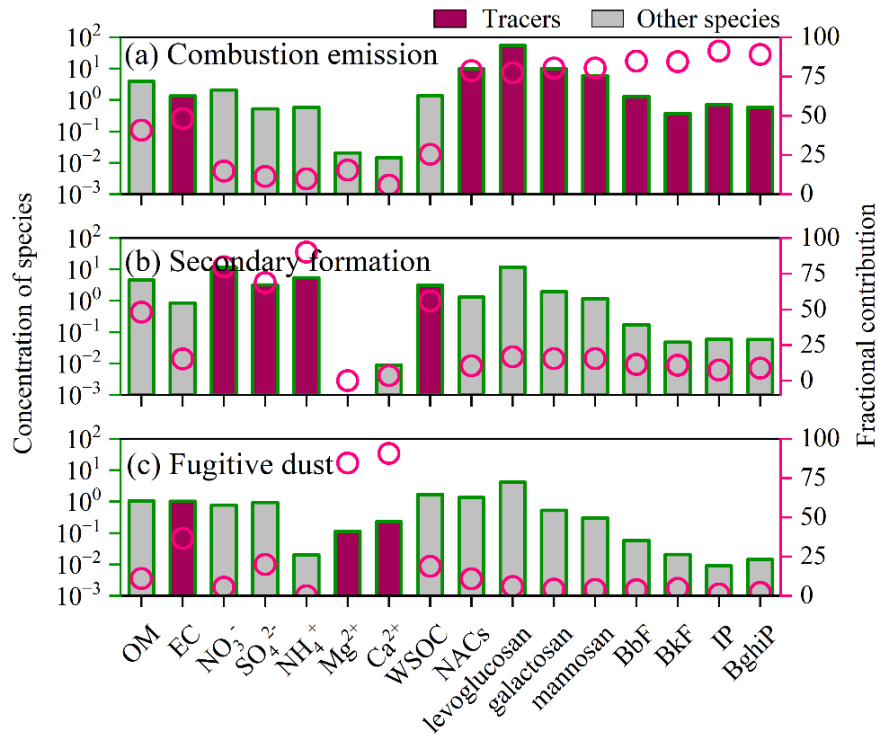


Figure S6 Source apportionment for OM at five sampling sites during the campaign.

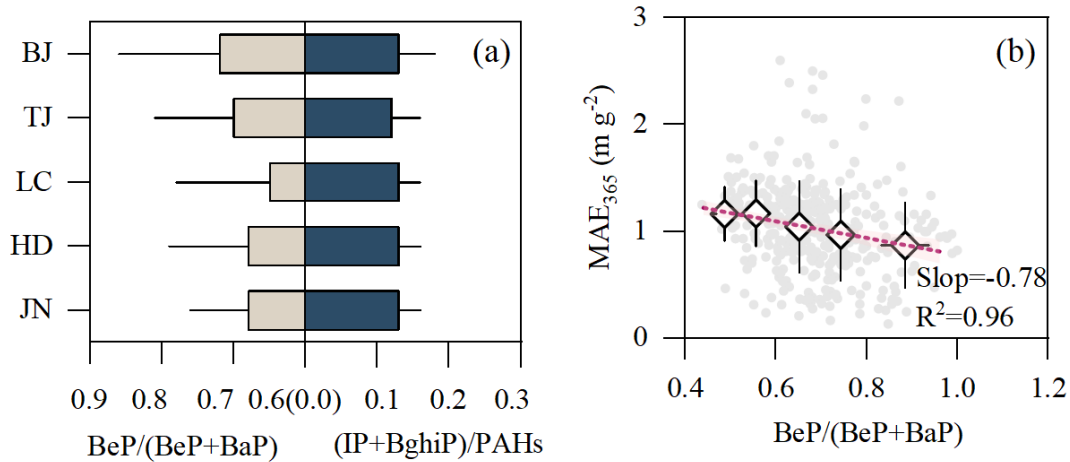


Figure S7 BeP/(Bep+Bap) and (IP+BghiP)/PAHs ratios at all the sampling sites (a); Dependence of MAE₃₆₅ on BeP/(Bep+Bap) across the NCP (Diamonds denote the mean values of MAE₃₆₅ and Bep/(Bep+Bap) across all sites for each binned Bep/(Bep+Bap) interval, with cross bars representing standard deviations) (b).

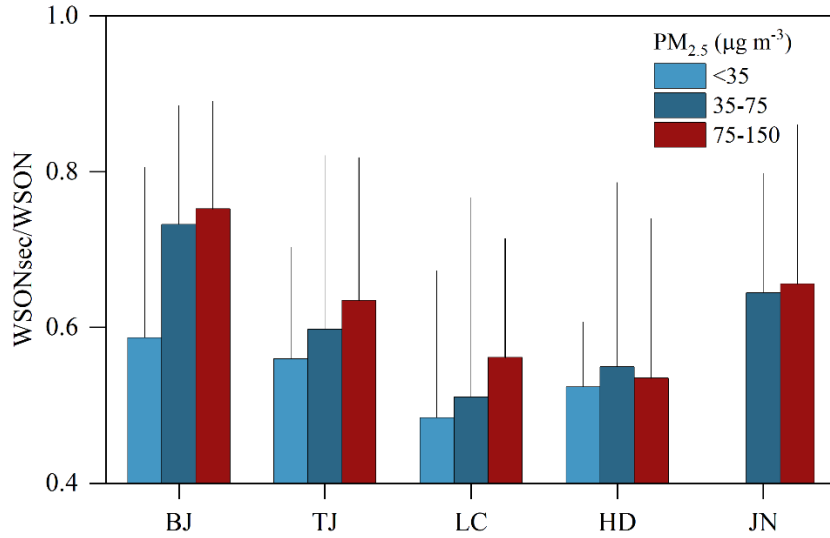


Figure 8 Ratios of secondary to total water-soluble organic nitrogen (WSNsec/WSN) across five cities at different PM_{2.5} levels.

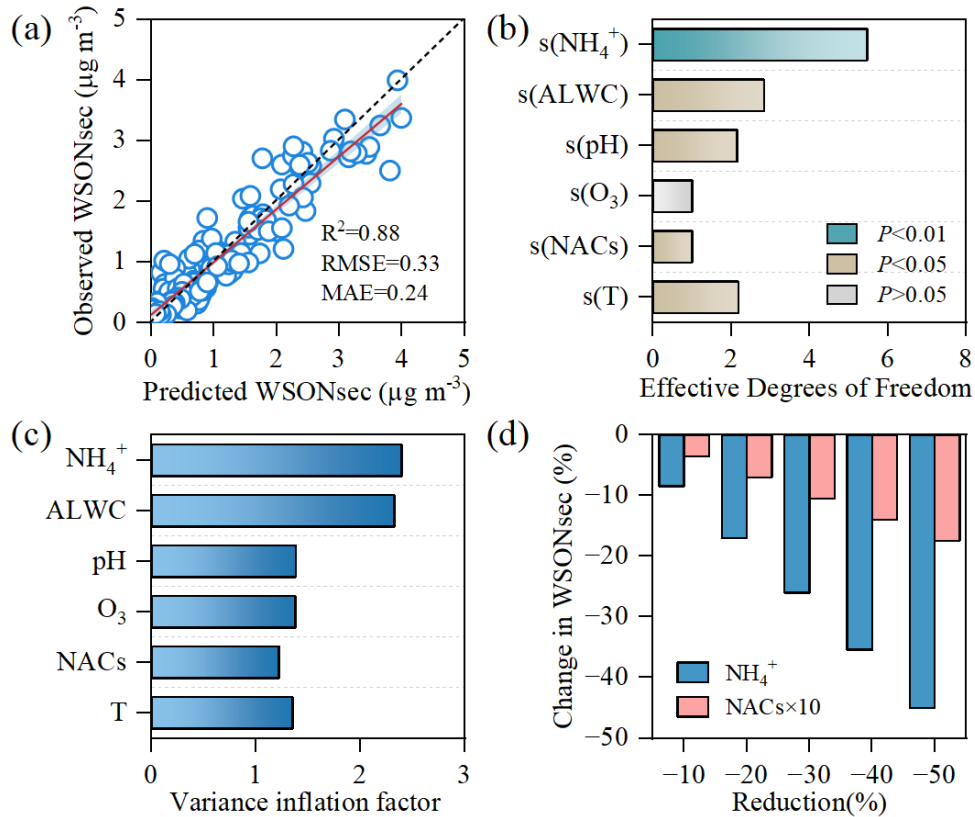


Figure 9 GAM model analysis for recognizing the key factors controlling WSNsec formation in the NCP. (a) Evaluation of GAM predictions against observed WSNsec concentrations; (b) Effective degrees of freedom of GAM smooth terms; (c) Variance

inflation factor of the predictors; (d) Sensitivity tests on the response of WSONsec to the reductions of NH_4^+ and NACs.

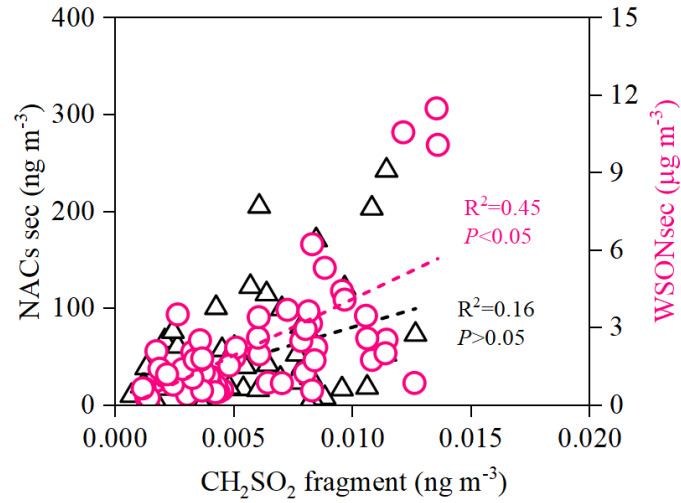


Figure S10 The relationship between CH₂SO₂ fragment, secondary NACs and WSON during humid haze episode.

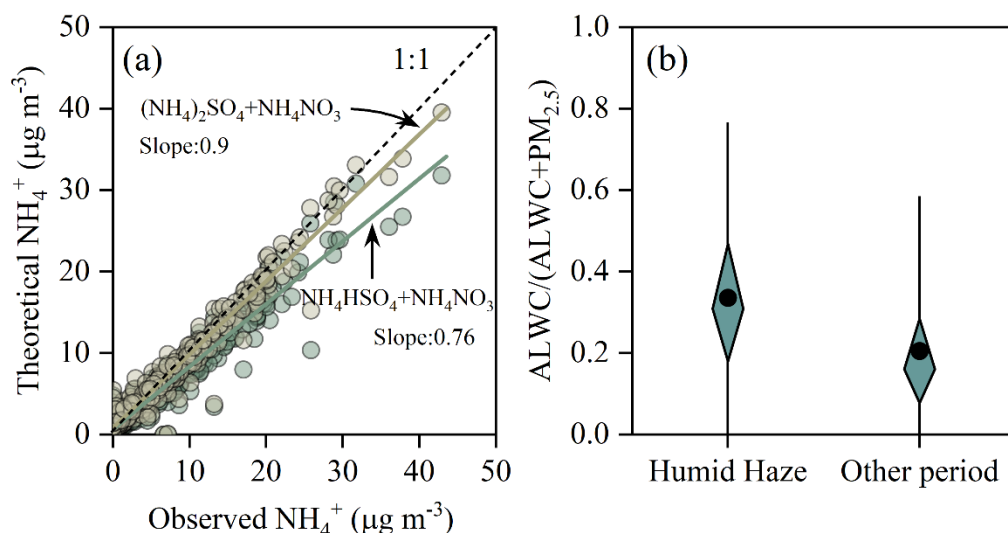


Figure S11 Comparison of the calculated and observed NH_4^+ concentrations across the NCP (a); The $\text{ALWC}/(\text{ALWC} + \text{PM}_{2.5})$ ratio at different periods (b).

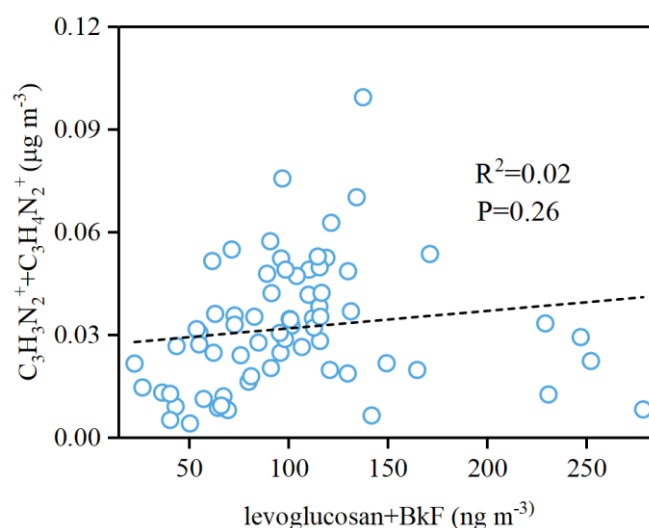


Figure S12 Scattering plot of Ims-related fragments versus levoglucosan+BkF

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