

1 *Supplementary Information for*

2 **Measurement report: Formation of tropospheric brown carbon in a**
3 **lifting air mass**

4 Can Wu^{1,2}, Xiaodi Liu¹, Ke Zhang¹, Si Zhang^{1*}, Cong Cao^{3,4}, Jianjun Li³, Rui Li^{1,2}, Fan
5 Zhang^{1,2}, Gehui Wang^{1,2*}

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11 ¹Key Lab of Geographic Information Science of the Ministry of Education, School of
12 Geographic Sciences, East China Normal University, Shanghai 210062, China

13 ²Institute of Eco-Chongming, 20 Cuiniao Rd., Chongming, Shanghai 202150, China

14 ³State Key Laboratory of Loess and Quaternary Geology, Institute of Earth Environment,
15 Chinese Academy of Sciences, Xi'an 710061, China

16 ⁴School of Marine and Atmospheric Science, Stony Brook University, Stony Brook, NY 11794,
17 USA

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24 *Corresponding authors: Dr. Si Zhang, E-mail: szhang@geo.ecnu.edu.cn
25 Prof. Gehui Wang, E-mail: ghwang@geo.ecnu.edu.cn

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33 List of supporting materials:

- 34 1. Two text, Text S1- S2.
35 2. Four tables, Table S1-S4.
36 3. Nine figures, Figure S1-S10.

38 **Text S1** UV–vis light absorption measurement

39 About 8 punches (12 cm³) from each filter sample was extracted three time under sonication
40 with 15 ml Milli-Q pure water (18.2 MΩ), and the extract was subsequently filtered through
41 0.45 μm PTFE pore syringe filter to remove the insoluble component in the suspension. A
42 liquid waveguide capillary UV–vis spectrometer equipped with a 1 m long-effective path
43 detection cell was applied to record the light absorption spectra of all extracts. The light
44 absorption spectrum was finally converted into absorption coefficient (abs_λ, M/m) at a
45 particular wavelength (λ) using the following equation.

Eq.S1

$$abs_{\lambda} = (A_{\lambda} - A_{700}) \frac{v_1}{v_a \times l} \times \ln(10)$$

46 Where A_λ and A₇₀₀ represent the light absorption of the extracts at wavelength λ and 700 nm,
47 respectively. V₁ corresponds to the volume of the extract, e.g., 15 ml; V_a refers to the volume of
48 the air through corresponding to filter punches; l (m) is the absorbing path length. While, ln(10)
49 is used for converting common logarithm that provided by the spectrophotometer to natural
50 logarithm.

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52 **Text S2** Positive matrix factorization (PMF) source apportionment

53 To quantitatively determine the fractional contribution of specific sources to BrC, a PMF
54 receptor model (EPA PMF 5.0 version) coupled with a bootstrap technique was applied here,
55 which has been widely used for the source apportionment of atmospheric pollutants in previous
56 studies. For more details of this method, you can found on the EPA
57 website([https://www.epa.gov/air-research/epa-positive-matrix-factorization-50-fundamentals-
58 and-user-guide](https://www.epa.gov/air-research/epa-positive-matrix-factorization-50-fundamentals-and-user-guide)). Briefly, WSOC, WSON, SO₄²⁻, NH₄⁺, Mg²⁺, Ca²⁺, abs_{365 nm} and organic tracers
59 (BbF, Bghip and levo.) were regarded as the input variables in the present work. As indicated in
60 our previous study on Mt. Hua(Wu et al., 2022), insignificant change in the corresponding
61 emission sources was revealed during air mass lifting process. Thus, the daytime samples from
62 both sites were added together as one data matrix. Considering Q values and interpretability,
63 four factors were obtained as the optimal solution after numerous testes with three to seven
64 factors, and the input species matched well with simulated ones with significant correlations
65 (R²>0.88).

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Table S1 RF model performance for testing dataset.

	MF site	MS site
R ²	0.92	0.86
MSE	0.003	0.008
RMSE	0.054	0.091
MAE	0.04	0.07

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

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85 **Table S2.** Information and mass concentration ($\mu\text{g m}^{-3}$) of nitroaromatic compounds detected in
86 this study.

Compounds	Molecular	Formula	CAS number	Abbreviation	MF site	MS site
4-Nitrophenol	139.11	C ₆ H ₅ NO ₃	100-02-7	4NP	2.6±2.6	0.40±0.25
3-Methoxy-4-nitrophenol	153.14	C ₇ H ₇ NO ₃	2581-34-2	3M4NP	0.07±0.05	0.01±0.01
4-Nitrocatechol	155.11	C ₆ H ₅ NO ₄	59030-13-6	4NC	1.6±2.5	0.25±0.67
4-Methyl-5-nitrocatechol	169.13	C ₇ H ₇ NO ₄	68906-21-8	4M5NC	11.4±11.4	1.8±1.3
3-Nitrosalicylic acid	183.12	C ₇ H ₅ NO ₅	85038-1	3NSA	0.01±0.01	0.004±0.01
5-Nitrosalicylic acid	183.12	C ₇ H ₅ NO ₅	96-97-9	5NSA	0.13±0.13	0.012±0.015

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Table S3 NH₃ concentrations in different regions of China

Region	Location	Period	Ammonia ($\mu\text{g m}^{-3}$)	Reference
NCP	Beijing	Summer of 2009	29.4±11.9	Meng et al.(Meng et al., 2017)
	Gucheng	Mar.2016-May.2017	22.2±12.8	Kuang et al. (Kuang et al., 2020)
		May-Sep 2013	27.5±42.8	Meng et al. (Meng et al., 2018)
	Luancheng Cangzhou	Dec.2015-Feb.2016 Dec.2015-Feb.2016	17.2 22.2	Pan et al. (Pan et al., 2018)
FWP	Xi'an	Winter of 2016	29±7.3	Wu et al. (Wu et al., 2020a) Cao et al. (Cao et al., 2009)
		Summer of 2016	38±9.4	
		2006-2007	12.9	
	Weinan	Dec.2015-Feb.2016	12.4	Pan et al. (Pan et al., 2018)
	Mt. Hua-MS Mt. Hua-MF	Summer of 2020 Summer of 2020	3.1±1.9 27.3±51	Wu et al. (Wu et al., 2022)
YRD	Shanghai	Autumn of 2019	9.5	Wu et al. (Wu et al., 2023)
		July-Dec 2013, Mar-June 2014	9.4±6.9	Wang et al. (Wang et al., 2015)
		Dec.2019-Jan.2020	9.3±4.0	Lv et al. (Lv et al., 2022)
	Nanjing Taihu	Dec.2015-Feb.2016 Dec.2015-Feb.2016	10.8 6.3	Pan et al. (Pan et al., 2018)
	Lin'an	Sep 2009-Dec 2010	12.5±8.5	Meng et al. (Meng et al., 2014)
PRD	Guangzhou	Dec.2015-Feb.2016	5.8	Pan et al. (Pan et al., 2018)
		Oct-Nov 2004	7.3±6.2	Hu et al. (Hu et al., 2008)
	Dinghushan Maoming	Dec.2015-Feb.2016 Dec.2015-Feb.2016	2.8 9.8	Pan et al. (Pan et al., 2018)
	Hongkong	Autumn 2000	2.3±2.7	Yao et al. (Yao et al., 2006)
TP	Lhasa	Dec.2015-Feb.2016	4.8	Pan et al. (Pan et al., 2018)
	Ali	Dec.2015-Feb.2016	1.7	Pan et al. (Pan et al., 2018)

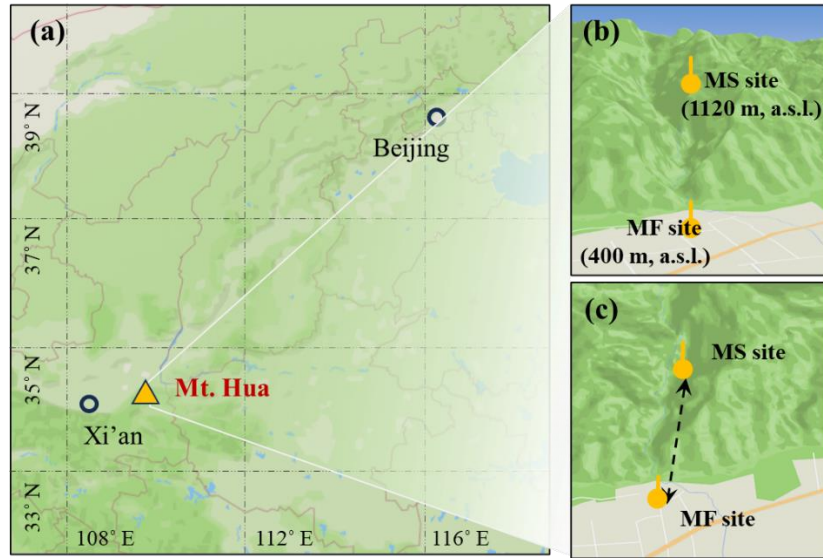
97 Note: In some cities, the NH₃ unit is ppb, which was converted by the formula of standard atmospheric
98 pressure and normal temperature in this study. The abbreviations of NCP, FWP, YRD, PRD and TP indicate
99 North China Plain, Fen-wei Plain, Yangtze River Delta, Pearl River Delta and Tibet Plateau, respectively.

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Table S4 MAE₃₆₅ of water-soluble BrC in PM_{2.5} among different cities in the world.

Region	Location	Year	Season	MAE ₃₆₅	Reference
NCP, China	Beijing	2011	Winter	1.2±0.1	Cheng et al. (Cheng et al.,
		2010-2011	Winter	1.26	Du et al.(Du et al., 2014)
		2010-2011	Summer	0.51	
			Spring	1.4±0.18	
	Xingtai	2018-2019	Summer	0.95±0.18	Li et al. (Li et al., 2023a)
			Autumn	1.5±0.13	
			Winter	1.9±0.16	
	Jinan (TSP)	2016	Spring	1.00±0.23	Wen et al. (Wen et al., 2021)
	Zhangbei	2016	Spring	1.32±0.34	
	Tianjin	2016–2017	Winter	1.54±0.33	Deng et al.(Deng et al., 2022)
Summer			0.84±0.22		
FWP, China	Xi'an	2016–2017	Winter	1.2±0.06	Wu et al. (Wu et al., 2020b)
			Summer	1.1±0.2	
	Licun	2017	Winter	0.78 ± 0.96	Li et al. (Li et al., 2023b)
			Winter	0.94±0.28	Li et al. (Li et al., 2020)
			Summer	1.01±0.18	
			Summer	0.69±0.2	
Mt. Hua-MS	2016	Summer	0.67±0.21	This study	
Mt. Hua-MF					
YRD, China	Changzhou	2018	Winter	0.74	Tao et al.(Tao et al., 2021)
	Yangzhou	2015-2016	Annual	0.75 ± 0.29	Chen et al.(Chen et al., 2018)
			Spring	0.69	
			Summer	0.51	
	Nanjing	2015-2016	Autumn	0.55	Chen et al.(Chen et al., 2018)
			Winter	1.04	
	Shanghai	2018-2019	Winter	1.18±0.42	Zhao et al. (Zhao et al.,
PRD, China	Guangzhou	2012	Winter	0.81	Liu et al.(Liu et al., 2018)
		2018	Winter	1.0 ± 0.21	Zou et al.(Zou et al., 2023)
		2019	Winter	0.34	Wang et al.(Wang et al., 2023)
		2016	Autumn	0.60±0.06	He et al. (He et al., 2023)
	Taipei	2021	Annual	0.86±0.60	Ting et al.(Ting et al., 2022)
TP, China	Lulang	2015-2016	Winter	0.75 ± 0.13	Zhu et al. (Zhu et al., 2018)
	Lhasa	2013-2014	Annual	0.74	Li et al. (Li et al., 2016)
			Summer	0.27 ± 0.10	
	Southeast TP	2013-2014	Winter	0.86 ± 0.17	Wu et al. (Wu et al., 2020c)
			Summer	0.38±0.16	
Nam Co	2015	Summer	0.38±0.16	Zhang et al. (Zhang et al.,	
India	Delhi	2016	Spring	2.5	Dasari et al. (Dasari et al.,
Korea	Seoul	2012-2013	Winter	1.02	Kim et al. (Kim et al., 2016)
			Summer	0.28	
USA	Los Angeles	2018-2019	Winter	0.7±0.2	Soleimanian et al. (Soleimanian et al., 2020)
			Summer	0.5±0.2	
	Yorkville	2010	Winter	0.41	Hecobian et al. (Hecobian et
Switzerland		2013	Summer	0.29	Mie et al. (Xie et al., 2019)
			Winter	0.9	
Greece	Ioannina	2019	Summer	0.28	Moschos et al. (Moschos et al., 2018)
			Summer	0.3 ± 0.1	Paraskevopoulou et al.

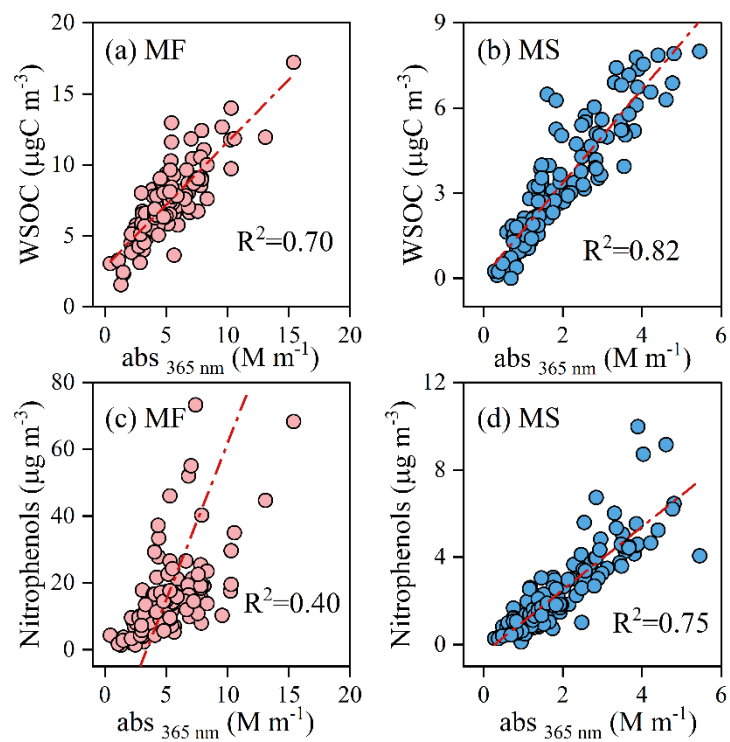
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Figure S1. Locations of the sampling sites in China. **(a)** Topographic view, **(b)** vertical view and bird's-eye view **(c)** of Mt. Hua with the sampling sites marked. The maps are the reproductions from ©Mapbox (<https://account.mapbox.com/>, last access: 16 March 2024)

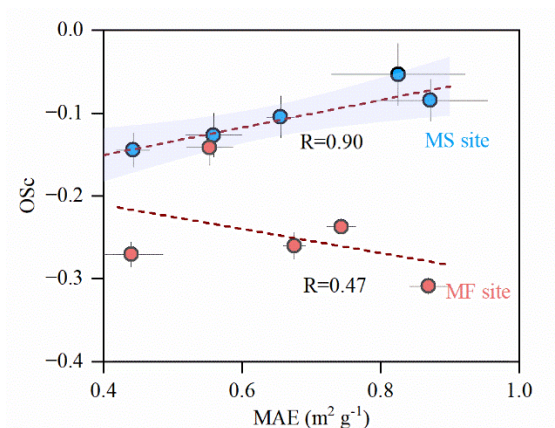
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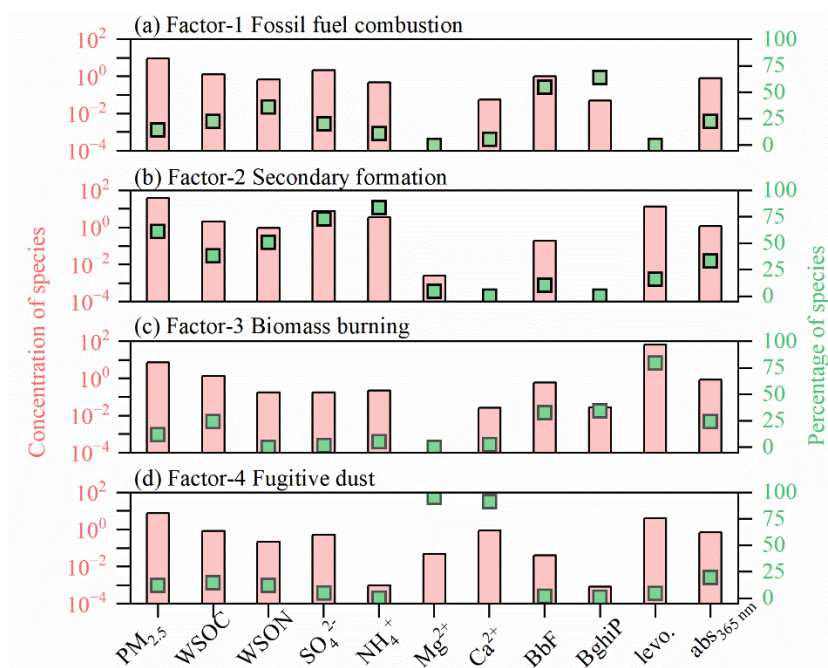
Figure S2. Linear fit regression analysis for Abs_{λ=365 nm} with WSOC and nitrophenols at MF and MS sites

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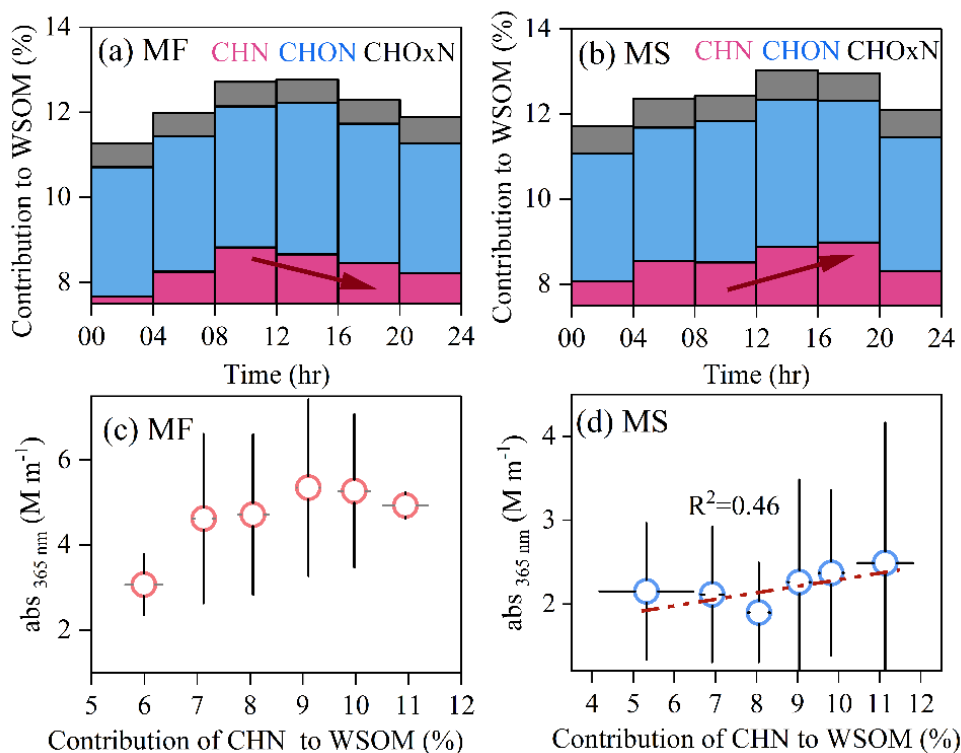
Figure S3. The correlativity between MAE_{365nm} and OSc value at both sampling sites.



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Figure S4. Source apportionment for light absorption of daytime water-soluble BrC (abs_{365 nm}) of all the daytime samples collected during the whole campaign.

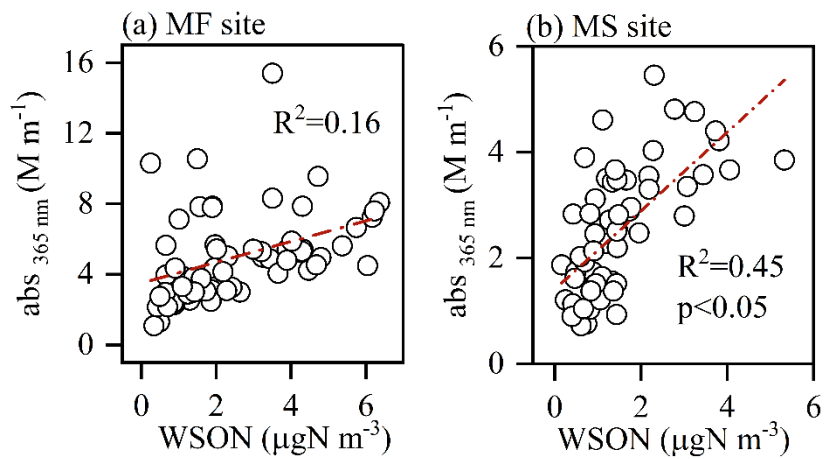
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Figure S5. (a) and (b) Diurnal variations in relative contributions of nitrogen-containing organic fragments to WSOM at MF and MS sites, respectively. (c and d) The dependence of light absorption of WSOC ($\text{Abs}_{\lambda=365\text{ nm}}$) on the relative contributions of CHN fragments to WSOM at the two sampling sites.

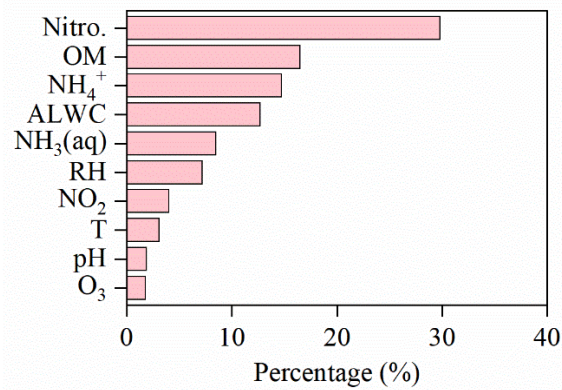
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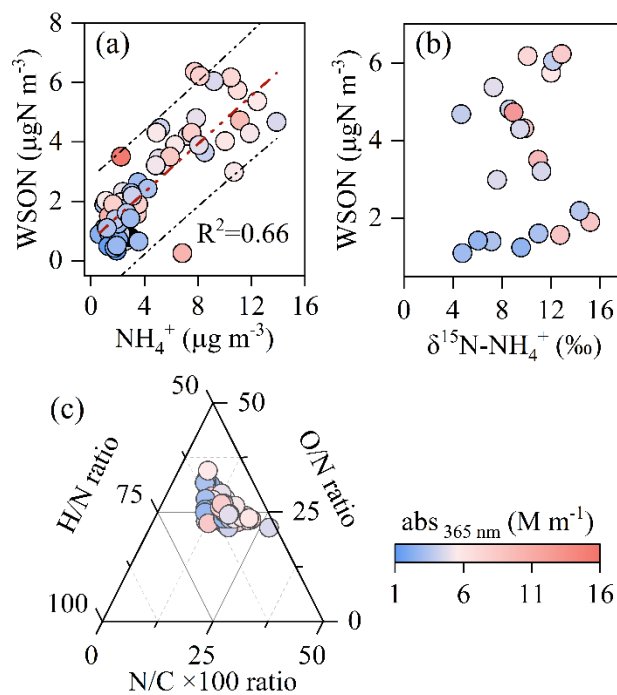
Figure S6 Linear fit regression analysis for $\text{Abs}_{\lambda=365 \text{ nm}}$ and WSON of the daytime samples at two sites

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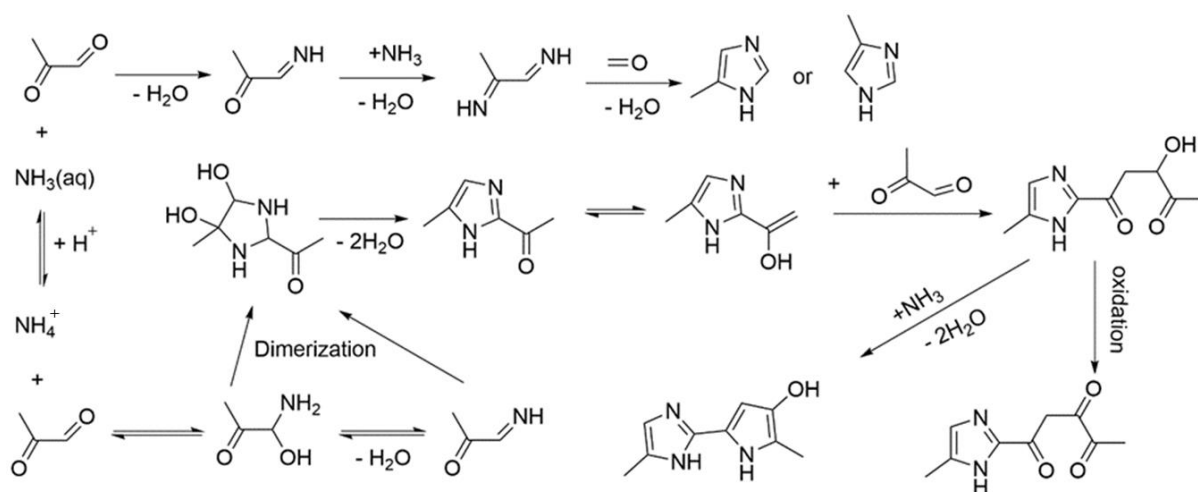
Figure S7 Random forest analysis for daytime WSON of the PM_{2.5} at MF site.



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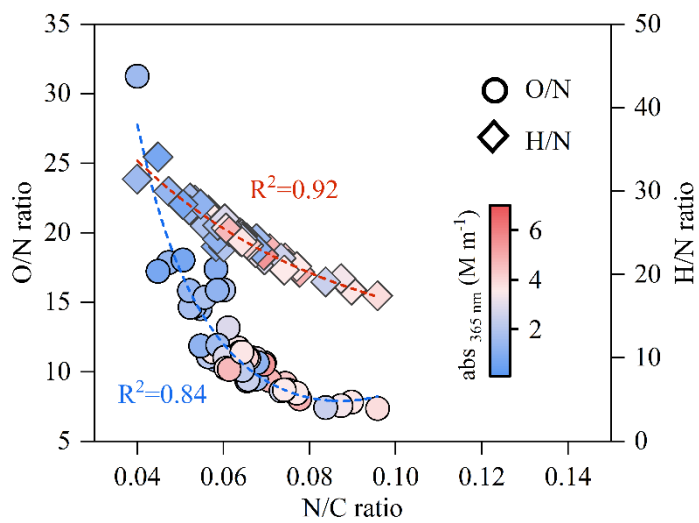
Figure S8 Impacts on WSON formation at MF site. Linear fit regressions for WSON with NH_4^+ + $\text{NH}_3(\text{aq})$ (a) and $\delta^{15}\text{N-NH}_4^+$ (b) at MF site and triangular chart for the elemental ratios of N/C, H/N and O/N of WSOC at MF site (c).

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Figure S9 Simple reaction paths for imidazoles or N-heterocycles (Modified from Aiona et al. (2017) and Jang et al. (2013))



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Figure S10 Elemental composition of daytime WSOC at MS site.

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