

Secondary processes driven by multi-factor interactions dominate the aerosol nitroaromatic compound pollution during winter in China

Yu Xu^{1,2}, Yu-Cheng You^{1,2}, Ting Yang^{1,2}, Lin Gui^{1,2}, Jun-Liang Tao^{1,2}, Tian-Shu Cheng^{1,2}, Hao Xiao^{1,2*}, Mei-Ju Yin^{1,2}, Hong-Wei Xiao^{1,2}, Hua-Yun Xiao^{1,2}

¹School of Agriculture and Biology, Shanghai Jiao Tong University, Shanghai 200240, China

²Shanghai Yangtze River Delta Eco-Environmental Change and Management Observation and Research Station (Shanghai Urban Ecosystem Research Station), Ministry of Science and Technology, National Forestry and Grassland Administration, 800 Dongchuan Rd., Shanghai 200240, China

*Corresponding authors

Hao Xiao

E-mail: xiaohao@sjtu.edu.cn

Phone: +86-152-7081-8726

20 **Contents of the file**

21 Tables S1–S5

22 Figures S1–S7

23

24

Table S1. Mean values ($\pm$ SD) of various parameters observed in different periods and locations.

Parameter	Northern cities in China								
	HEB			TY			XA		
	Average	Clean period	Poll. period	Average	Clean period	Poll. period	Average	Clean period	Poll. period
T ($^{\circ}$ C)	-17.27 ± 5.42	-17.04 ± 7.2	-17.58 ± 1.62	-2.56 ± 2.14	-3 ± 2.69	-1.95 ± 0.95	1.69 ± 2.24	1.91 ± 2.36	1.39 ± 2.28
RH (%)	66.16 ± 6.15	63.84 ± 4.52	69.4 ± 7.12	42.74 ± 8.78	36.18 ± 3.62	51.93 ± 3.41	49.27 ± 17.47	39.3 ± 7.78	63.23 ± 18.2
ALW	31.53 ± 24.65	17.35 ± 13.66	51.39 ± 23.38	21.18 ± 21.21	7.5 ± 5.98	40.33 ± 19.94	51.63 ± 81.74	7.72 ± 5.69	113.11 ± 101.13
pH	4.35 ± 1.08	3.96 ± 1.28	4.89 ± 0.35	5.99 ± 0.92	6.52 ± 0.85	5.25 ± 0.28	5.13 ± 0.90	5.36 ± 1.07	4.82 ± 0.56
SR (W m^{-2})	71.11 ± 6.34	70.27 ± 6.66	72.3 ± 6.41	119.95 ± 17.58	124.06 ± 16.06	114.2 ± 19.81	141.74 ± 31.62	159.05 ± 18.81	117.51 ± 30.98
PBLH (m)	339.03 ± 191.45	457.34 ± 162.41	173.40 ± 49.43	410.47 ± 361.30	600.59 ± 368.43	144.30 ± 59.63	166.74 ± 67.59	176.70 ± 84.45	152.80 ± 38.10
$\cdot\text{OH}$ ($\times 10^5$ molecule cm^{-3})	3.05 ± 1.25	3.54 ± 1.23	2.35 ± 1.00	3.00 ± 2.14	4.27 ± 1.96	1.22 ± 0.31	1.70 ± 1.14	2.42 ± 0.86	0.68 ± 0.51
SO ₂ ($\mu\text{g m}^{-3}$)	56.24 ± 18.84	44.45 ± 10.23	72.75 ± 15.34	60.36 ± 40.28	31.62 ± 23.46	100.6 ± 12.94	31.73 ± 12.13	24.15 ± 5.08	42.34 ± 11.16
PM _{2.5} ($\mu\text{g m}^{-3}$)	90.62 ± 65.15	45.31 ± 18.11	154.04 ± 50.60	81.02 ± 65.20	33.58 ± 20.46	147.45 ± 40.08	115.33 ± 88.85	52.45 ± 16.8	203.37 ± 68.42
O ₃ ($\mu\text{g m}^{-3}$)	37.84 ± 10.47	43.88 ± 8.21	29.38 ± 6.86	30.07 ± 22.84	43.57 ± 21.10	11.18 ± 1.04	24.67 ± 13.35	31.41 ± 11.8	15.23 ± 9.53
NO ₃ ⁻ ($\mu\text{g m}^{-3}$)	9.16 ± 6.64	7.66 ± 7.74	11.25 ± 4.68	14.95 ± 14.27	6.50 ± 2.83	26.78 ± 15.75	40.56 ± 31.79	20.89 ± 10.2	68.11 ± 31.58
SO ₄ ²⁻ ($\mu\text{g m}^{-3}$)	11.45 ± 7.34	7.26 ± 3.73	17.31 ± 7.33	16.51 ± 13.23	9.17 ± 1.35	29.36 ± 15.31	21.58 ± 17.3	11.14 ± 3.05	36.19 ± 18.75
Ca ²⁺ ($\mu\text{g m}^{-3}$)	0.93 ± 0.39	0.72 ± 0.30	1.23 ± 0.31	6.09 ± 1.72	5.21 ± 1.67	7.31 ± 0.85	6.69 ± 5.72	4.85 ± 1.92	9.26 ± 8.38
Mg ²⁺ ($\mu\text{g m}^{-3}$)	0.10 ± 0.06	0.09 ± 0.08	0.10 ± 0.03	0.34 ± 0.13	0.25 ± 0.07	0.47 ± 0.09	0.36 ± 0.36	0.21 ± 0.09	0.56 ± 0.51
NH ₄ ⁺ ($\mu\text{g m}^{-3}$)	8.23 ± 5.71	5.26 ± 3.63	12.39 ± 5.72	12.11 ± 11.66	5.86 ± 1.64	20.86 ± 14.34	20.58 ± 17.78	8.79 ± 2.98	37.1 ± 16.5
nss-K ⁺ ($\mu\text{g m}^{-3}$)	1.87 ± 1.37	1.10 ± 0.70	2.94 ± 1.39	1.19 ± 0.97	0.63 ± 0.25	1.98 ± 1.09	2.63 ± 1.99	1.63 ± 0.91	4.05 ± 2.32
nss-Cl ⁻ ($\mu\text{g m}^{-3}$)	5.19 ± 4.44	2.81 ± 2.24	8.51 ± 4.79	6.51 ± 5.60	3.49 ± 2.20	10.74 ± 6.37	6.01 ± 3.82	4.38 ± 2.10	8.3 ± 4.71
C ₈ H ₁₇ O ₄ S ⁻ (ng m^{-3})	0.21 ± 0.09	0.21 ± 0.12	0.20 ± 0.05	0.49 ± 0.21	0.43 ± 0.21	0.57 ± 0.20	0.49 ± 0.16	0.51 ± 0.20	0.46 ± 0.09
C ₅ H ₇ O ₆ S ⁻ (ng m^{-3})	17.90 ± 11.61	21.93 ± 13.51	12.27 ± 5.37	12.19 ± 3.18	12.03 ± 3.14	12.42 ± 3.59	12.75 ± 3.61	13.93 ± 4.12	11.08 ± 2.10
4NP (ng m^{-3})	10.57 ± 5.73	9.23 ± 6.74	12.46 ± 3.82	11.68 ± 2.88	11.23 ± 3.47	12.31 ± 1.97	12.77 ± 3.51	12.88 ± 4.25	12.62 ± 2.60
3M4NP (ng m^{-3})	2.96 ± 1.49	2.64 ± 1.70	3.40 ± 1.15	4.75 ± 2.22	5.67 ± 2.18	3.48 ± 1.70	2.89 ± 1.08	3.18 ± 1.29	2.47 ± 0.58

2M4NP (ng m ⁻³)	3.74 ± 1.97	3.31 ± 2.31	4.34 ± 1.38	4.47 ± 2.04	5.24 ± 2.17	3.39 ± 1.35	3.39 ± 1.18	3.78 ± 1.41	2.85 ± 0.48
2,4DNP (ng m ⁻³)	0.38 ± 0.20	0.35 ± 0.19	0.42 ± 0.21	2.68 ± 1.70	1.78 ± 0.86	3.95 ± 1.85	1.87 ± 1.20	1.57 ± 0.41	2.27 ± 1.83
Total NPs (ng m ⁻³)	17.65 ± 9.28	15.53 ± 10.89	20.62 ± 6.34	23.59 ± 6.79	23.92 ± 8.00	23.12 ± 5.49	20.92 ± 5.64	21.42 ± 6.73	20.22 ± 4.31
4M5NC (ng m ⁻³)	0.42 ± 0.31	0.32 ± 0.16	0.56 ± 0.44	0.49 ± 0.32	0.47 ± 0.33	0.51 ± 0.35	0.19 ± 0.12	0.22 ± 0.16	0.16 ± 0
4NC (ng m ⁻³)	20.71 ± 7.55	17.85 ± 7.57	24.71 ± 6.02	9.84 ± 6.24	7.89 ± 3.64	12.58 ± 8.43	15.5 ± 5.50	17.15 ± 5.6	13.19 ± 4.96
Total NCs (ng m ⁻³)	21.13 ± 7.55	18.17 ± 7.48	25.27 ± 6.00	10.33 ± 6.28	8.36 ± 3.82	13.09 ± 8.38	15.69 ± 5.51	17.37 ± 5.61	13.35 ± 4.96
5NSA (ng m ⁻³)	0.41 ± 0.36	0.34 ± 0.43	0.51 ± 0.25	0.73 ± 0.66	0.35 ± 0.26	1.28 ± 0.68	1.08 ± 0.41	0.78 ± 0.20	1.50 ± 0.11
3NSA (ng m ⁻³)	0.55 ± 0.48	0.45 ± 0.55	0.69 ± 0.38	0.67 ± 0.82	0.18 ± 0.19	1.35 ± 0.89	1.15 ± 0.77	0.55 ± 0.19	1.99 ± 0.27
Total NSAs (ng m ⁻³)	0.96 ± 0.84	0.80 ± 0.97	1.20 ± 0.62	1.40 ± 1.47	0.53 ± 0.43	2.62 ± 1.57	2.23 ± 1.16	1.33 ± 0.34	3.49 ± 0.37
4NG (ng m ⁻³)	5.12 ± 1.90	4.18 ± 1.80	6.43 ± 1.18	2.98 ± 1.77	2.94 ± 1.73	3.04 ± 2.03	3.29 ± 1.43	3.75 ± 1.47	2.64 ± 1.24
Total NACs (ng m ⁻³)	44.87 ± 17.83	38.68 ± 19.19	53.52 ± 12.70	38.3 ± 12.2	35.75 ± 12.81	41.87 ± 11.66	42.13 ± 12.13	43.87 ± 13.64	39.7 ± 10.63

Table S2. Mean values ($\pm$ SD) of various parameters observed in different periods and locations.

Parameter	Northern cities in China					
	LZ			BJ		
	Average	Clean period	Poll. period	Average	Clean period	Poll. period
T ($^{\circ}$ C)	-5.72 $\pm$ 2.77	-7.26 $\pm$ 2.30	-3.56 $\pm$ 1.79	-1.58 $\pm$ 2.21	-1.16 $\pm$ 2.20	-2.83 $\pm$ 2.10
RH (%)	53.85 $\pm$ 10.59	58.14 $\pm$ 11.03	47.85 $\pm$ 6.98	48.53 $\pm$ 21.71	39.59 $\pm$ 14.70	75.34 $\pm$ 17.00
ALW	20.44 $\pm$ 13.37	17.03 $\pm$ 15.84	25.22 $\pm$ 8.16	26.48 $\pm$ 42.79	9.90 $\pm$ 15.37	76.24 $\pm$ 64.6
pH	5.18 $\pm$ 0.93	4.86 $\pm$ 0.36	5.64 $\pm$ 1.33	6.31 $\pm$ 1.80	6.96 $\pm$ 1.59	4.37 $\pm$ 0.45
SR (W m ⁻²)	139.16 $\pm$ 13.98	136.92 $\pm$ 18.18	142.28 $\pm$ 4.54	112.91 $\pm$ 9.06	112.31 $\pm$ 10.26	114.70 $\pm$ 4.93
PBLH (m)	273.10 $\pm$ 163.15	347.22 $\pm$ 174.53	169.32 $\pm$ 66.57	407.88 $\pm$ 327.18	466.34 $\pm$ 351.64	232.48 $\pm$ 180.68
$\cdot$ OH ($\times 10^5$ molecule cm ⁻³)	2.22 $\pm$ 1.74	3.19 $\pm$ 1.65	0.85 $\pm$ 0.50	3.67 $\pm$ 1.84	4.41 $\pm$ 1.45	1.45 $\pm$ 0.64
SO ₂ (μ g m ⁻³)	48.00 $\pm$ 20.40	35.77 $\pm$ 15.67	65.13 $\pm$ 12.15	6.95 $\pm$ 3.34	6.59 $\pm$ 3.24	8.04 $\pm$ 4.10
PM _{2.5} (μ g m ⁻³)	86.37 $\pm$ 42.14	54.16 $\pm$ 14.04	131.46 $\pm$ 15.17	52.67 $\pm$ 53.71	25.99 $\pm$ 21.38	132.71 $\pm$ 35.00
O ₃ (μ g m ⁻³)	30.36 $\pm$ 12.83	36.29 $\pm$ 14.15	22.06 $\pm$ 2.24	31.15 $\pm$ 18.29	37.24 $\pm$ 16.73	12.89 $\pm$ 7.39
NO ₃ ⁻ (μ g m ⁻³)	19.91 $\pm$ 14.91	12.00 $\pm$ 6.59	30.99 $\pm$ 16.83	10.77 $\pm$ 15.24	4.38 $\pm$ 5.57	29.96 $\pm$ 20.43
SO ₄ ²⁻ (μ g m ⁻³)	18.15 $\pm$ 11.7	11.61 $\pm$ 4.56	27.31 $\pm$ 12.87	7.40 $\pm$ 8.46	3.89 $\pm$ 3.86	17.91 $\pm$ 10.65
Ca ²⁺ (μ g m ⁻³)	5.83 $\pm$ 3.90	4.03 $\pm$ 1.00	8.35 $\pm$ 5.18	2.33 $\pm$ 0.74	2.52 $\pm$ 0.49	1.75 $\pm$ 1.16
Mg ²⁺ (μ g m ⁻³)	0.31 $\pm$ 0.36	0.17 $\pm$ 0.04	0.51 $\pm$ 0.51	0.15 $\pm$ 0.03	0.14 $\pm$ 0.02	0.17 $\pm$ 0.04
NH ₄ ⁺ (μ g m ⁻³)	10.13 $\pm$ 7.04	6.78 $\pm$ 3.08	14.82 $\pm$ 8.65	6.6 $\pm$ 9.50	2.68 $\pm$ 3.04	18.37 $\pm$ 13.50
nss-K ⁺ (μ g m ⁻³)	0.66 $\pm$ 0.41	0.39 $\pm$ 0.21	1.03 $\pm$ 0.30	0.57 $\pm$ 0.49	0.40 $\pm$ 0.32	1.08 $\pm$ 0.61
nss-Cl ⁻ (μ g m ⁻³)	1.23 $\pm$ 1.14	0.83 $\pm$ 0.72	1.78 $\pm$ 1.46	2.03 $\pm$ 1.90	1.40 $\pm$ 1.17	3.92 $\pm$ 2.68
C ₈ H ₁₇ O ₄ S ⁻ (ng m ⁻³)	0.42 $\pm$ 0.18	0.35 $\pm$ 0.19	0.52 $\pm$ 0.11	0.29 $\pm$ 0.08	0.27 $\pm$ 0.09	0.32 $\pm$ 0.05
C ₅ H ₇ O ₆ S ⁻ (ng m ⁻³)	11.48 $\pm$ 3.79	12.43 $\pm$ 4.72	10.14 $\pm$ 1.53	9.60 $\pm$ 3.64	10.02 $\pm$ 4.14	8.34 $\pm$ 1.11
4NP (ng m ⁻³)	6.53 $\pm$ 2.43	5.35 $\pm$ 1.88	8.17 $\pm$ 2.27	6.31 $\pm$ 2.32	6.35 $\pm$ 2.60	6.20 $\pm$ 1.60
3M4NP (ng m ⁻³)	1.69 $\pm$ 0.47	1.60 $\pm$ 0.53	1.82 $\pm$ 0.39	2.00 $\pm$ 0.83	2.24 $\pm$ 0.80	1.26 $\pm$ 0.37

2M4NP (ng m ⁻³)	1.96 ± 0.60	1.74 ± 0.57	2.27 ± 0.56	2.08 ± 0.73	2.30 ± 0.71	1.42 ± 0.11
2,4DNP (ng m ⁻³)	1.76 ± 2.92	1.01 ± 0.66	2.83 ± 4.51	1.06 ± 0.63	1.18 ± 0.61	0.71 ± 0.66
Total NPs (ng m ⁻³)	11.95 ± 5.13	9.70 ± 3.31	15.09 ± 5.90	11.45 ± 3.50	12.07 ± 3.87	9.59 ± 0.84
4M5NC (ng m ⁻³)	0.23 ± 0.10	0.23 ± 0.09	0.23 ± 0.11	0.27 ± 0.13	0.28 ± 0.13	0.26 ± 0.12
4NC (ng m ⁻³)	5.79 ± 3.70	4.72 ± 3.12	7.30 ± 4.27	4.83 ± 2.91	4.70 ± 3.31	5.22 ± 1.56
Total NCs (ng m ⁻³)	6.02 ± 3.75	4.95 ± 3.19	7.53 ± 4.32	5.10 ± 2.97	4.98 ± 3.39	5.48 ± 1.54
5NSA (ng m ⁻³)	0.61 ± 0.42	0.39 ± 0.25	0.92 ± 0.43	0.47 ± 0.60	0.24 ± 0.36	1.14 ± 0.76
3NSA (ng m ⁻³)	0.75 ± 0.67	0.41 ± 0.35	1.24 ± 0.74	0.42 ± 0.69	0.18 ± 0.43	1.14 ± 0.91
Total NSAs (ng m ⁻³)	1.36 ± 1.09	0.80 ± 0.59	2.16 ± 1.17	0.89 ± 1.28	0.42 ± 0.78	2.28 ± 1.66
4NG (ng m ⁻³)	1.23 ± 0.55	1.13 ± 0.7	1.38 ± 0.24	1.25 ± 0.82	1.32 ± 0.93	1.04 ± 0.35
Total NACs (ng m ⁻³)	20.56 ± 7.95	16.57 ± 7.31	26.16 ± 5.18	18.69 ± 7.14	18.79 ± 8.19	18.39 ± 3.42

Table S3. Mean values ($\pm$ SD) of various parameters observed in different periods and locations.

Parameter	Southern cities								
	CD			KM			WH		
	Average	Clean period	Poll. period	Average	Clean period	Poll. period	Average	Clean period	Poll. period
T ($^{\circ}$ C)	5.82 ± 1.29	6.48 ± 1.19	4.90 ± 0.79	9.63 ± 2.52	9.88 ± 2.98	9.12 ± 1.42	3.89 ± 2.88	2.06 ± 2.48	5.71 ± 2.03
RH (%)	74.75 ± 7.65	71.48 ± 5.65	79.33 ± 8.26	68.25 ± 7.73	68.44 ± 8.83	67.88 ± 6.07	78.84 ± 11.53	81.94 ± 14.27	75.75 ± 8.10
ALW	65.76 ± 96.89	13.76 ± 8.73	138.57 ± 119.76	8.18 ± 6.42	5.64 ± 6.45	13.28 ± 1.42	74.42 ± 89.97	82.53 ± 116.36	66.31 ± 64.1
pH	3.51 ± 0.38	3.43 ± 0.18	3.62 ± 0.57	4.06 ± 1.61	4.33 ± 1.93	3.52 ± 0.48	3.49 ± 0.82	3.57 ± 1.16	3.41 ± 0.35
SR (W m^{-2})	138.45 ± 40.09	117.27 ± 31.56	168.1 ± 32.29	176.3 ± 54.5	176.03 ± 60.38	176.83 ± 48.83	130.02 ± 54.30	127.63 ± 50.32	132.41 ± 62.79
PBLH (m)	365.22 ± 96.81	429.01 ± 71.34	275.90 ± 32.31	451.46 ± 155.51	503.28 ± 155.05	347.82 ± 105.29	269.18 ± 104.97	328.61 ± 93.83	209.74 ± 83.4
$\cdot\text{OH}$ ($\times 10^5$ molecule cm^{-3})	1.96 ± 1.32	2.04 ± 1.68	1.84 ± 0.76	8.14 ± 4.44	9.52 ± 4.58	5.40 ± 2.91	3.10 ± 2.19	3.24 ± 2.59	2.96 ± 1.94
SO ₂ ($\mu\text{g m}^{-3}$)	10.61 ± 2.86	9.24 ± 2.73	12.53 ± 1.84	18.45 ± 5.34	17.39 ± 5.01	20.57 ± 6.08	12.84 ± 6.17	8.60 ± 6.09	17.08 ± 1.87
PM _{2.5} ($\mu\text{g m}^{-3}$)	85.58 ± 58.16	43.99 ± 19.78	143.79 ± 38.10	47.62 ± 30.50	28.28 ± 12.14	86.30 ± 8.63	88.54 ± 35.69	58.43 ± 16.12	118.65 ± 19.13
O ₃ ($\mu\text{g m}^{-3}$)	26.85 ± 9.37	29.22 ± 10.67	23.53 ± 6.85	37.55 ± 16.19	35.69 ± 12.67	41.26 ± 23.65	24.21 ± 10.39	28.83 ± 11.36	19.59 ± 7.55
NO ₃ ⁻ ($\mu\text{g m}^{-3}$)	20.80 ± 19.54	8.80 ± 4.18	37.60 ± 20.47	4.83 ± 3.80	3.11 ± 2.73	8.29 ± 3.42	19.13 ± 12.00	11.25 ± 10.87	27.01 ± 7.04
SO ₄ ²⁻ ($\mu\text{g m}^{-3}$)	10.35 ± 8.82	4.97 ± 1.82	17.88 ± 9.35	8.09 ± 4.53	5.71 ± 3.18	12.84 ± 2.52	9.52 ± 5.17	7.63 ± 4.99	11.42 ± 5.02
Ca ²⁺ ($\mu\text{g m}^{-3}$)	2.08 ± 1.20	1.85 ± 0.96	2.42 ± 1.52	2.91 ± 1.07	2.57 ± 0.83	3.59 ± 1.30	2.34 ± 1.60	1.67 ± 1.63	3.00 ± 1.37
Mg ²⁺ ($\mu\text{g m}^{-3}$)	0.10 ± 0.06	0.08 ± 0.04	0.14 ± 0.06	0.08 ± 0.03	0.07 ± 0.03	0.11 ± 0.02	0.11 ± 0.06	0.07 ± 0.05	0.14 ± 0.05
NH ₄ ⁺ ($\mu\text{g m}^{-3}$)	10.6 ± 10.65	4.45 ± 1.89	19.21 ± 12.14	3.78 ± 2.67	2.47 ± 1.71	6.39 ± 2.36	8.83 ± 4.64	7.14 ± 3.30	10.52 ± 5.44
nss-K ⁺ ($\mu\text{g m}^{-3}$)	1.16 ± 0.77	0.63 ± 0.49	1.90 ± 0.28	0.53 ± 0.36	0.31 ± 0.14	0.95 ± 0.25	1.00 ± 0.61	0.51 ± 0.23	1.50 ± 0.44
nss-Cl ⁻ ($\mu\text{g m}^{-3}$)	2.58 ± 1.92	1.57 ± 1.11	3.99 ± 2.00	0.74 ± 0.38	0.63 ± 0.38	0.96 ± 0.30	1.63 ± 1.16	1.06 ± 0.63	2.21 ± 1.34
C ₈ H ₁₇ O ₄ S ⁻ (ng m^{-3})	0.24 ± 0.06	0.23 ± 0.06	0.26 ± 0.07	0.45 ± 0.36	0.31 ± 0.30	0.73 ± 0.33	0.25 ± 0.17	0.28 ± 0.18	0.21 ± 0.16
C ₅ H ₇ O ₆ S ⁻ (ng m^{-3})	6.52 ± 1.95	5.81 ± 1.81	7.50 ± 1.86	6.03 ± 2.79	5.28 ± 2.97	7.54 ± 1.87	4.92 ± 1.76	4.38 ± 1.31	5.45 ± 2.10
4NP (ng m^{-3})	5.23 ± 2.38	3.89 ± 1.93	7.10 ± 1.59	1.32 ± 0.96	1.18 ± 1.05	1.59 ± 0.84	2.60 ± 1.12	2.19 ± 0.85	3.02 ± 1.28
3M4NP (ng m^{-3})	0.65 ± 0.29	0.54 ± 0.26	0.81 ± 0.27	0.20 ± 0.22	0.26 ± 0.24	0.07 ± 0.08	0.29 ± 0.20	0.25 ± 0.19	0.32 ± 0.22

2M4NP (ng m ⁻³)	1.00 ± 0.40	0.82 ± 0.34	1.25 ± 0.37	0.24 ± 0.25	0.30 ± 0.28	0.12 ± 0.14	0.42 ± 0.29	0.37 ± 0.26	0.46 ± 0.34
2,4DNP (ng m ⁻³)	0.46 ± 0.30	0.52 ± 0.35	0.37 ± 0.23	0.30 ± 0.16	0.25 ± 0.17	0.41 ± 0.04	0.52 ± 0.51	0.52 ± 0.58	0.53 ± 0.49
Total NPs (ng m ⁻³)	7.34 ± 3.15	5.78 ± 2.78	9.52 ± 2.35	2.05 ± 1.38	1.99 ± 1.59	2.18 ± 1.02	3.83 ± 1.88	3.33 ± 1.78	4.34 ± 2.00
4M5NC (ng m ⁻³)	0.16 ± 0	0.16 ± 0	0.16 ± 0	0.26 ± 0.10	0.24 ± 0.05	0.32 ± 0.15	0.18 ± 0.07	0.15 ± 0	0.22 ± 0.09
4NC (ng m ⁻³)	8.43 ± 4.88	5.73 ± 4.45	12.21 ± 2.26	3.66 ± 1.87	3.03 ± 1.96	4.92 ± 0.88	3.40 ± 2.05	2.12 ± 1.30	4.67 ± 1.90
Total NCs (ng m ⁻³)	8.59 ± 4.88	5.89 ± 4.45	12.37 ± 2.26	3.93 ± 1.89	3.27 ± 1.92	5.25 ± 1.00	3.58 ± 2.08	2.26 ± 1.30	4.89 ± 1.92
5NSA (ng m ⁻³)	0.78 ± 0.40	0.53 ± 0.31	1.14 ± 0.15	0.18 ± 0.24	0.07 ± 0.14	0.40 ± 0.28	0.55 ± 0.33	0.35 ± 0.36	0.74 ± 0.14
3NSA (ng m ⁻³)	0.46 ± 0.39	0.19 ± 0.17	0.85 ± 0.23	0.04 ± 0.08	0.02 ± 0.07	0.08 ± 0.10	0.42 ± 0.26	0.32 ± 0.23	0.51 ± 0.28
Total NSAs (ng m ⁻³)	1.24 ± 0.74	0.72 ± 0.43	1.98 ± 0.28	0.22 ± 0.31	0.10 ± 0.20	0.48 ± 0.35	0.97 ± 0.52	0.68 ± 0.51	1.25 ± 0.37
4NG (ng m ⁻³)	1.58 ± 0.90	1.23 ± 0.89	2.08 ± 0.70	0.67 ± 0.31	0.57 ± 0.33	0.86 ± 0.17	0.52 ± 0.30	0.36 ± 0.21	0.67 ± 0.29
Total NACs (ng m ⁻³)	18.75 ± 9.37	13.62 ± 8.44	25.95 ± 4.88	6.88 ± 3.58	5.93 ± 3.88	8.78 ± 2.17	8.89 ± 4.34	6.63 ± 3.66	11.16 ± 3.98

Table S4. Mean values ($\pm$ SD) of various parameters observed in different periods and locations.

Parameter	Southern cities								
	HZ			GZ			GY		
	Average	Clean period	Poll. period	Average	Clean period	Poll. period	Average	Clean period	Poll. period
T (°C)	6.61 $\pm$ 2.49	5.08 $\pm$ 1.62	8.74 $\pm$ 1.85	15.9 $\pm$ 3.25	14.2 $\pm$ 3.00	18.29 $\pm$ 1.85	5.58 $\pm$ 3.29	4.93 $\pm$ 3.22	8.81 $\pm$ 0.46
RH (%)	73.83 $\pm$ 17.61	76.84 $\pm$ 17.38	69.63 $\pm$ 19.03	62.04 $\pm$ 21.39	62.93 $\pm$ 28.03	60.79 $\pm$ 8.73	78.88 $\pm$ 20.50	84.34 $\pm$ 17.71	51.56 $\pm$ 2.92
ALW	64.64 $\pm$ 69.04	57.58 $\pm$ 45.20	74.52 $\pm$ 99.17	19.62 $\pm$ 25.35	21.95 $\pm$ 33.76	16.35 $\pm$ 5.84	37.64 $\pm$ 40.88	42.82 $\pm$ 43.05	11.75 $\pm$ 9.59
pH	3.37 $\pm$ 0.59	3.54 $\pm$ 0.30	3.14 $\pm$ 0.84	2.65 $\pm$ 0.76	2.96 $\pm$ 0.87	2.21 $\pm$ 0.24	3.34 $\pm$ 0.53	3.42 $\pm$ 0.53	2.97 $\pm$ 0.55
SR (W m ⁻²)	128.11 $\pm$ 59.08	139.98 $\pm$ 64.14	111.49 $\pm$ 53.26	140.30 $\pm$ 61.44	149.07 $\pm$ 77.60	128.02 $\pm$ 32.02	124.02 $\pm$ 40.92	120.45 $\pm$ 38.20	141.87 $\pm$ 67.21
PBLH (m)	414.10 $\pm$ 249.60	494.17 $\pm$ 261.18	302.01 $\pm$ 205.12	404.53 $\pm$ 200.27	498.48 $\pm$ 208.57	273.01 $\pm$ 89.32	380.66 $\pm$ 182.81	373.32 $\pm$ 194.26	417.31 $\pm$ 157.42
·OH ($\times 10^5$)	1.82 $\pm$ 1.23	1.98 $\pm$ 1.47	1.60 $\pm$ 0.91	4.06 $\pm$ 3.38	4.83 $\pm$ 4.18	2.99 $\pm$ 1.66	3.98 $\pm$ 2.47	4.01 $\pm$ 2.59	3.87 $\pm$ 2.56
SO ₂ ($\mu\text{g m}^{-3}$)	13.12 $\pm$ 3.59	11.37 $\pm$ 2.60	15.57 $\pm$ 3.53	15.87 $\pm$ 6.29	12.54 $\pm$ 5.960	20.53 $\pm$ 2.98	43.33 $\pm$ 18.64	36.6 $\pm$ 11.06	76.98 $\pm$ 1.38
PM _{2.5} ($\mu\text{g m}^{-3}$)	69.12 $\pm$ 28.14	51.90 $\pm$ 20.62	93.23 $\pm$ 17.15	56.41 $\pm$ 33.06	32.13 $\pm$ 17.95	90.40 $\pm$ 6.85	49.47 $\pm$ 26.17	40.08 $\pm$ 14.71	96.44 $\pm$ 17.12
O ₃ ($\mu\text{g m}^{-3}$)	26.39 $\pm$ 17.90	23.68 $\pm$ 14.35	30.18 $\pm$ 23.26	34.62 $\pm$ 22.76	28.81 $\pm$ 26.40	42.75 $\pm$ 15.40	47.10 $\pm$ 31.46	38.54 $\pm$ 26.20	89.88 $\pm$ 17.80
NO ₃ ⁻ ($\mu\text{g m}^{-3}$)	18.91 $\pm$ 8.03	14.90 $\pm$ 7.15	24.51 $\pm$ 5.78	6.43 $\pm$ 3.53	4.47 $\pm$ 2.29	9.18 $\pm$ 3.20	4.98 $\pm$ 3.76	4.14 $\pm$ 3.14	9.21 $\pm$ 4.90
SO ₄ ²⁻ ($\mu\text{g m}^{-3}$)	9.92 $\pm$ 3.70	8.89 $\pm$ 3.86	11.37 $\pm$ 3.29	8.96 $\pm$ 6.11	4.41 $\pm$ 2.68	15.34 $\pm$ 2.20	9.04 $\pm$ 5.15	7.41 $\pm$ 3.14	17.22 $\pm$ 6.50
Ca ²⁺ ($\mu\text{g m}^{-3}$)	2.19 $\pm$ 1.60	1.82 $\pm$ 1.35	2.69 $\pm$ 1.92	1.01 $\pm$ 0.51	1.05 $\pm$ 0.66	0.94 $\pm$ 0.19	1.71 $\pm$ 1.48	1.53 $\pm$ 1.52	2.60 $\pm$ 1.19
Mg ²⁺ ($\mu\text{g m}^{-3}$)	0.11 $\pm$ 0.06	0.09 $\pm$ 0.04	0.15 $\pm$ 0.07	0.05 $\pm$ 0.02	0.04 $\pm$ 0.01	0.06 $\pm$ 0.02	0.09 $\pm$ 0.08	0.06 $\pm$ 0.05	0.21 $\pm$ 0.05
NH ₄ ⁺ ($\mu\text{g m}^{-3}$)	8.32 $\pm$ 3.04	7.36 $\pm$ 3.35	9.67 $\pm$ 2.16	4.88 $\pm$ 2.41	3.09 $\pm$ 0.90	7.40 $\pm$ 1.10	3.81 $\pm$ 2.53	3.05 $\pm$ 1.57	7.65 $\pm$ 3.61
nss-K ⁺ ($\mu\text{g m}^{-3}$)	0.75 $\pm$ 0.29	0.62 $\pm$ 0.25	0.93 $\pm$ 0.25	0.76 $\pm$ 0.59	0.34 $\pm$ 0.33	1.35 $\pm$ 0.22	0.47 $\pm$ 0.40	0.33 $\pm$ 0.27	1.14 $\pm$ 0.25
nss-Cl ⁻ ($\mu\text{g m}^{-3}$)	1.61 $\pm$ 1.07	1.26 $\pm$ 1.04	2.10 $\pm$ 1.01	0.48 $\pm$ 0.43	0.64 $\pm$ 0.49	0.24 $\pm$ 0.17	0.17 $\pm$ 0.18	0.10 $\pm$ 0.06	0.51 $\pm$ 0.20
C ₈ H ₁₇ O ₄ S ⁻ (ng m ⁻³)	0.26 $\pm$ 0.08	0.26 $\pm$ 0.10	0.27 $\pm$ 0.04	0.07 $\pm$ 0.05	0.09 $\pm$ 0.06	0.05 $\pm$ 0.02	0.08 $\pm$ 0.05	0.07 $\pm$ 0.03	0.13 $\pm$ 0.10
C ₅ H ₇ O ₆ S ⁻ (ng m ⁻³)	4.60 $\pm$ 1.57	4.84 $\pm$ 1.90	4.27 $\pm$ 1.08	3.41 $\pm$ 0.70	3.42 $\pm$ 0.55	3.40 $\pm$ 0.94	3.22 $\pm$ 1.27	3.37 $\pm$ 1.32	2.49 $\pm$ 0.99
4NP (ng m ⁻³)	2.53 $\pm$ 1.49	2.16 $\pm$ 1.73	3.05 $\pm$ 1.00	0.87 $\pm$ 0.43	0.79 $\pm$ 0.52	0.99 $\pm$ 0.25	1.62 $\pm$ 1.22	1.25 $\pm$ 0.91	3.48 $\pm$ 0.67
3M4NP (ng m ⁻³)	0.26 $\pm$ 0.22	0.24 $\pm$ 0.27	0.28 $\pm$ 0.16	0.01 $\pm$ 0.01	0.01 $\pm$ 0.02	0 $\pm$ 0	0.21 $\pm$ 0.21	0.15 $\pm$ 0.16	0.50 $\pm$ 0.14

2M4NP (ng m ⁻³)	0.43 ± 0.39	0.41 ± 0.48	0.47 ± 0.27	0.02 ± 0.05	0.04 ± 0.06	0 ± 0	0.28 ± 0.28	0.2 ± 0.22	0.68 ± 0.21
2,4DNP (ng m ⁻³)	0.67 ± 1.02	0.51 ± 0.66	0.88 ± 1.45	0.25 ± 0.24	0.33 ± 0.28	0.12 ± 0.04	0.19 ± 0.07	0.18 ± 0.05	0.23 ± 0.15
Total NPs (ng m ⁻³)	3.89 ± 2.88	3.33 ± 3.09	4.68 ± 2.66	1.15 ± 0.60	1.18 ± 0.78	1.12 ± 0.29	2.29 ± 1.70	1.77 ± 1.26	4.89 ± 1.16
4M5NC (ng m ⁻³)	0.16 ± 0.03	0.15 ± 0	0.17 ± 0.05	0.16 ± 0.01	0.16 ± 0.02	0.16 ± 0	0.22 ± 0.13	0.20 ± 0.12	0.30 ± 0.19
4NC (ng m ⁻³)	2.16 ± 1.23	2.04 ± 1.53	2.32 ± 0.76	3.28 ± 1.96	2.28 ± 1.79	4.67 ± 1.27	5.41 ± 3.82	4.09 ± 2.49	12.00 ± 0.60
Total NCs (ng m ⁻³)	2.31 ± 1.22	2.19 ± 1.53	2.49 ± 0.72	3.44 ± 1.96	2.45 ± 1.79	4.83 ± 1.27	5.62 ± 3.89	4.29 ± 2.57	12.31 ± 0.41
5NSA (ng m ⁻³)	0.43 ± 0.28	0.35 ± 0.29	0.54 ± 0.26	0.38 ± 0.31	0.22 ± 0.29	0.60 ± 0.16	0.19 ± 0.19	0.16 ± 0.20	0.31 ± 0.06
3NSA (ng m ⁻³)	0.31 ± 0.23	0.26 ± 0.19	0.38 ± 0.28	0.17 ± 0.20	0.03 ± 0.07	0.37 ± 0.13	0.19 ± 0.20	0.16 ± 0.18	0.35 ± 0.33
Total NSAs (ng m ⁻³)	0.74 ± 0.49	0.60 ± 0.46	0.92 ± 0.53	0.55 ± 0.47	0.25 ± 0.34	0.97 ± 0.24	0.37 ± 0.38	0.32 ± 0.37	0.66 ± 0.39
4NG (ng m ⁻³)	0.39 ± 0.29	0.40 ± 0.38	0.36 ± 0.08	0.41 ± 0.28	0.37 ± 0.35	0.46 ± 0.14	0.75 ± 0.61	0.54 ± 0.39	1.83 ± 0.17
Total NACs (ng m ⁻³)	7.33 ± 4.34	6.52 ± 5.23	8.45 ± 2.85	5.55 ± 2.75	4.24 ± 2.75	7.37 ± 1.53	9.04 ± 6.41	6.91 ± 4.44	19.69 ± 1.36

Table S5. Average contribution of secondarily formed NACs to the total measured NACs mass in PM_{2.5} in different periods and locations.

Sites	Clean period	Polluted period	Average
HEB	91.96 ± 4.02 %	89.49 ± 3.32 %	90.93 ± 3.80 %
TY	87.62 ± 7.02 %	70.48 ± 7.35 %	80.48 ± 11.16 %
XA	94.38 ± 1.77 %	88.23 ± 3.46 %	91.82 ± 4.01 %
LZ	85.28 ± 4.24 %	79.25 ± 3.83 %	82.77 ± 4.98 %
BJ	90.99 ± 3.66 %	72.66 ± 7.59 %	86.41 ± 9.43 %
All northern cities	90.00 ± 3.16 %	80.20 ± 7.91 %	87.00 ± 4.59 %
CD	93.53 ± 4.73 %	95.39 ± 1.49 %	94.31 ± 3.73 %
KM	93.17 ± 4.54 %	95.24 ± 1.98 %	93.86 ± 3.90 %
WH	91.52 ± 4.00 %	93.02 ± 3.31 %	92.27 ± 3.59 %
HZ	91.52 ± 4.84 %	94.57 ± 2.17 %	92.79 ± 4.12 %
GZ	95.37 ± 4.80 %	97.99 ± 0.26 %	96.46 ± 3.80 %
GY	86.00 ± 9.23 %	96.81 ± 0.69 %	87.80 ± 9.35 %
All southern cities	92.00 ± 3.15 %	96.00 ± 1.57 %	93.00 ± 2.67 %

Figure S1

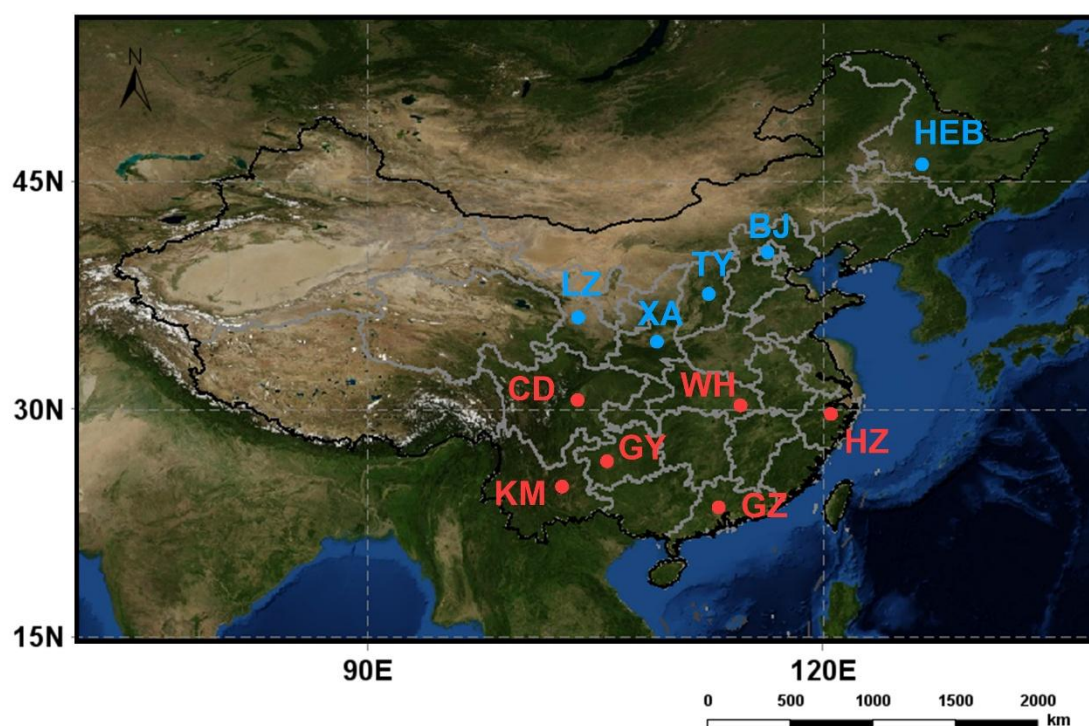


Figure S1. The locations of the sampling sites. The map was derived from ©MeteoInfoMap (version 3.3.0) (Chinese Academy of Meteorological Sciences, China). The latitude and longitude of sampling sites are as follows: BJ (40.043°N, 116.412°E), LZ (36.106°N, 103.731°E), XA (34.249°N, 108.984°E), HEB (45.767°N, 126.635°E), TY (37.803°N, 112.584°E), KM (25.065°N, 102.696°E), GZ (23.180°N, 113.352°E), GY (26.651°N, 106.610°E), HZ (30.295°N, 120.165°E), CD (30.679°N, 104.142°E), and WH (30.546°N, 114.357°E).

Figure S2

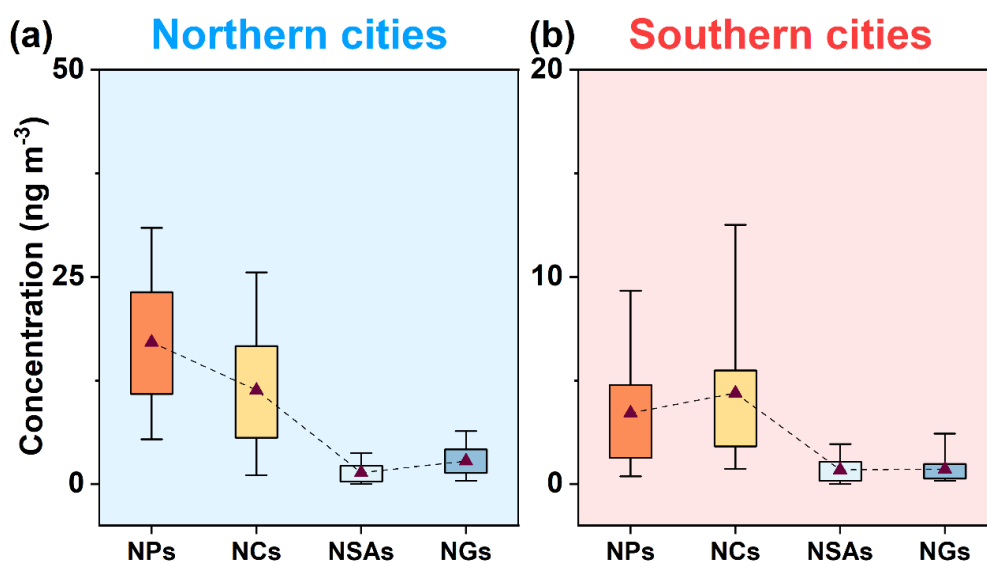


Figure S2. Box-and-whisker plots showing the variations in the concentration of different NAC groups in $\text{PM}_{2.5}$ for the pooled data from (a) northern and (b) southern cities. Each box encompasses the 25th–75th percentiles. The whiskers extend from the 5th to the 95th percentiles. The triangles inside boxes indicate the mean.

Figure S3

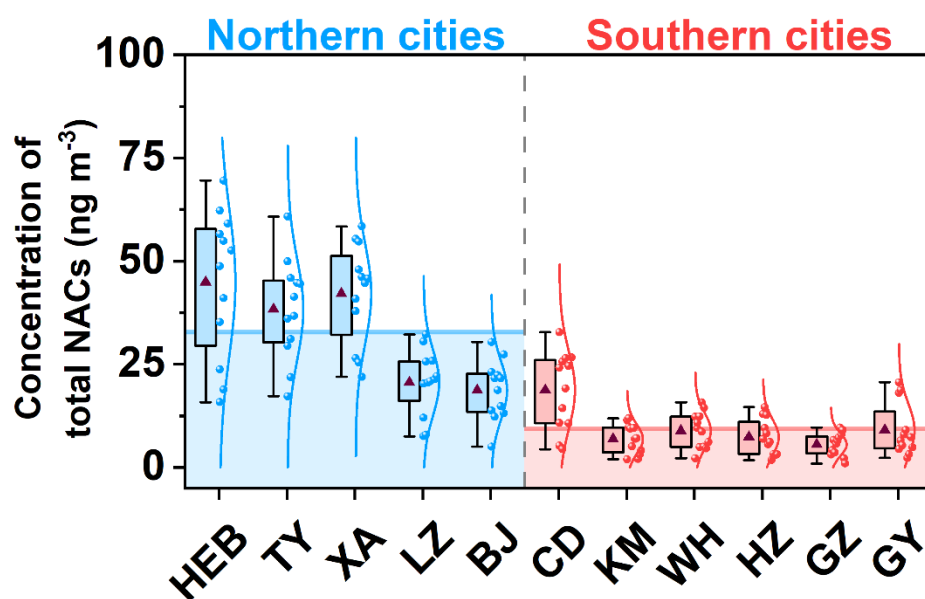


Figure S3. Box and whisker plots showing the variations in the mean concentrations of total NACs in PM_{2.5} collected in 11 Chinese cities during winter. Each box encompasses the 25th–75th percentiles. The whiskers extend from the 5th to the 95th percentiles. The solid triangles inside boxes indicate the mean.

Figure S4

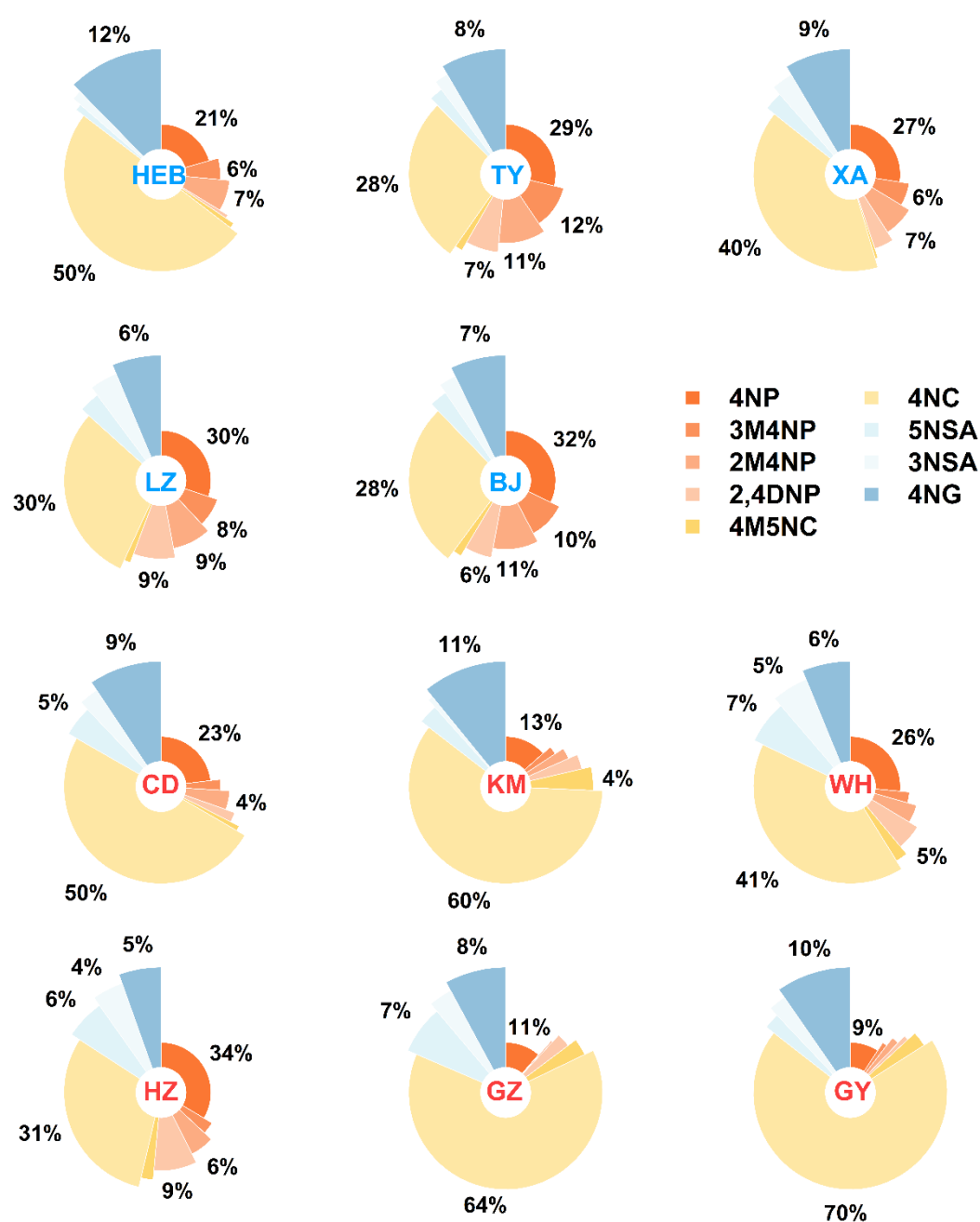


Figure S4. Average percentage distributions of various NAC species in PM_{2.5} collected in 11 Chinese cities during winter.

Figure S5.

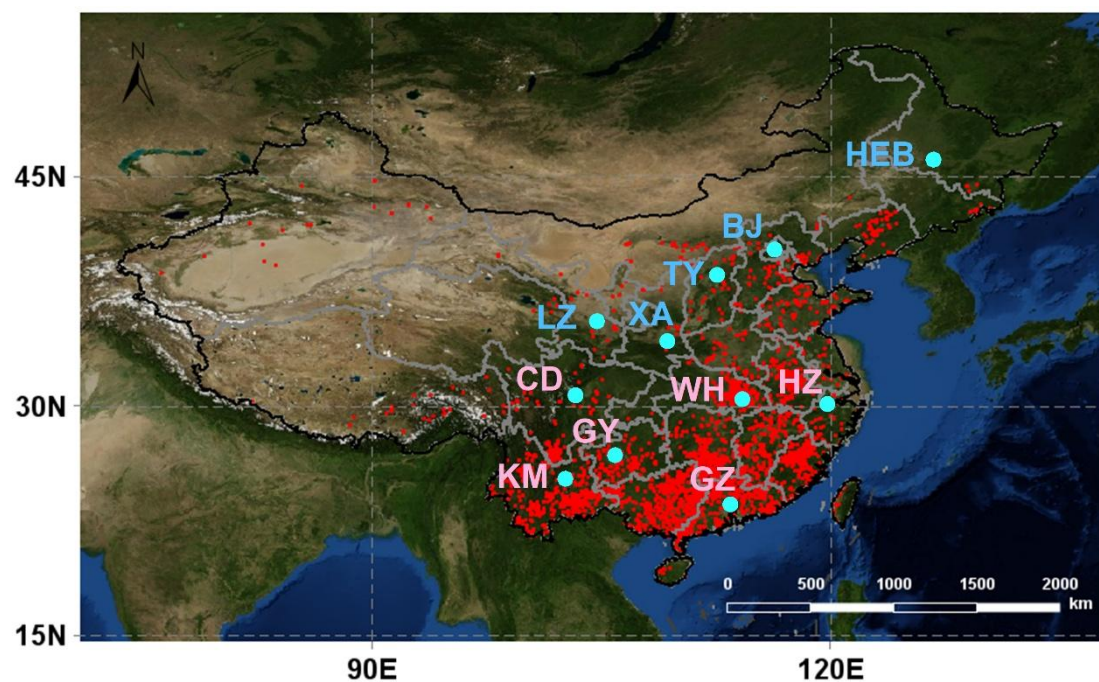


Figure S5. Spatial distribution of open fire spots in China during the sampling period (December 10, 2017 to January 14, 2018). The map was derived from ©MeteoInfoMap (version 3.3.0) (Chinese Academy of Meteorological Sciences, China).

Figure S6

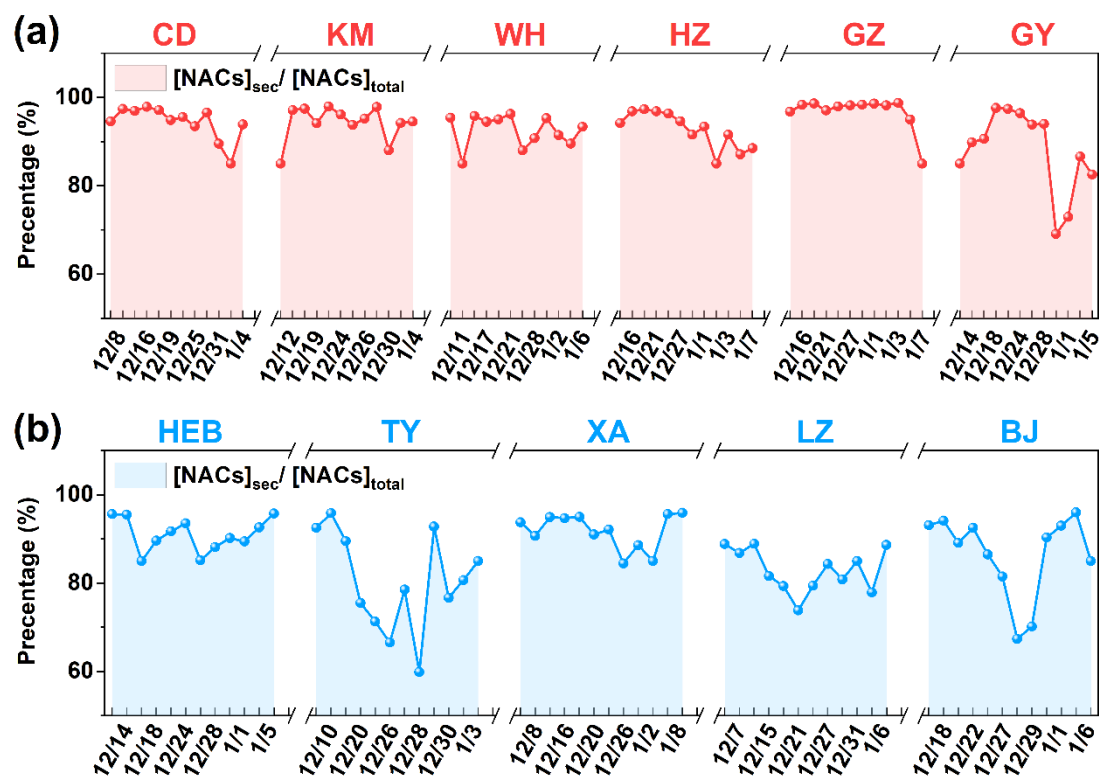


Figure S6. Temporal variations in the contribution of secondarily formed NACs to the total measured NACs mass in $PM_{2.5}$ in (a) northern and (b) southern cities.

Figure S7

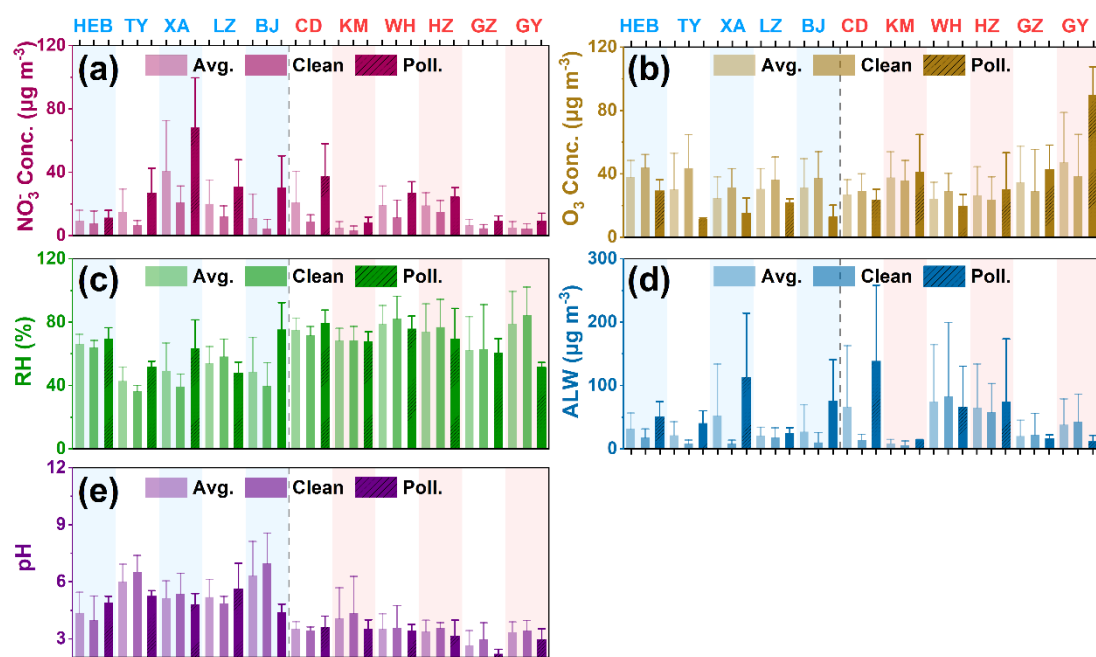


Figure S7. Average levels of (a) NO_3^- , (b) O_3 , (c) RH, (d) ALW, and (e) pH across different periods in 11 Chinese cities.