

## *Supporting Information*

### **Evolution of nitroaromatic compounds in urban aerosols during China's Blue Sky Protection Campaign: molecular characteristics, sources, and formation pathways**

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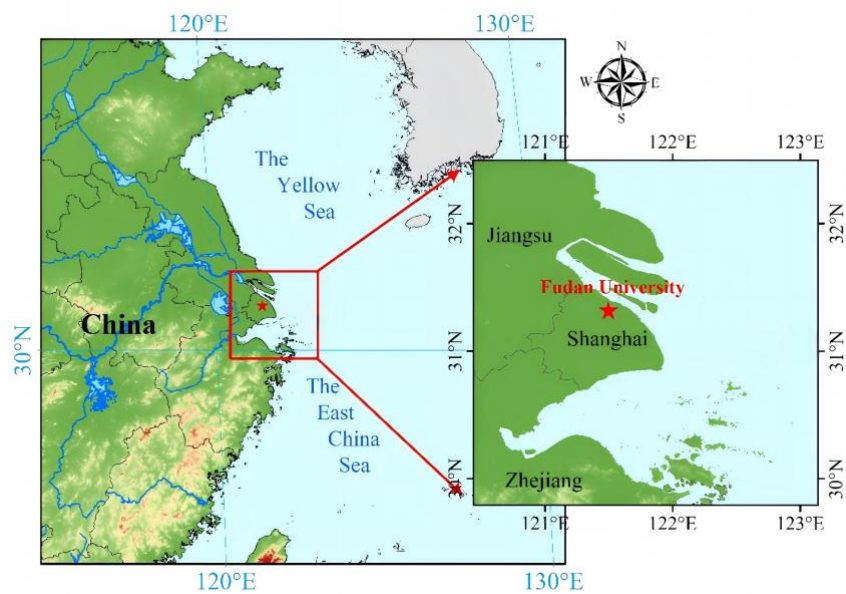


**Figure S1.** The location of the sampling site (red pentagram) in the Yangpu district of Shanghai.

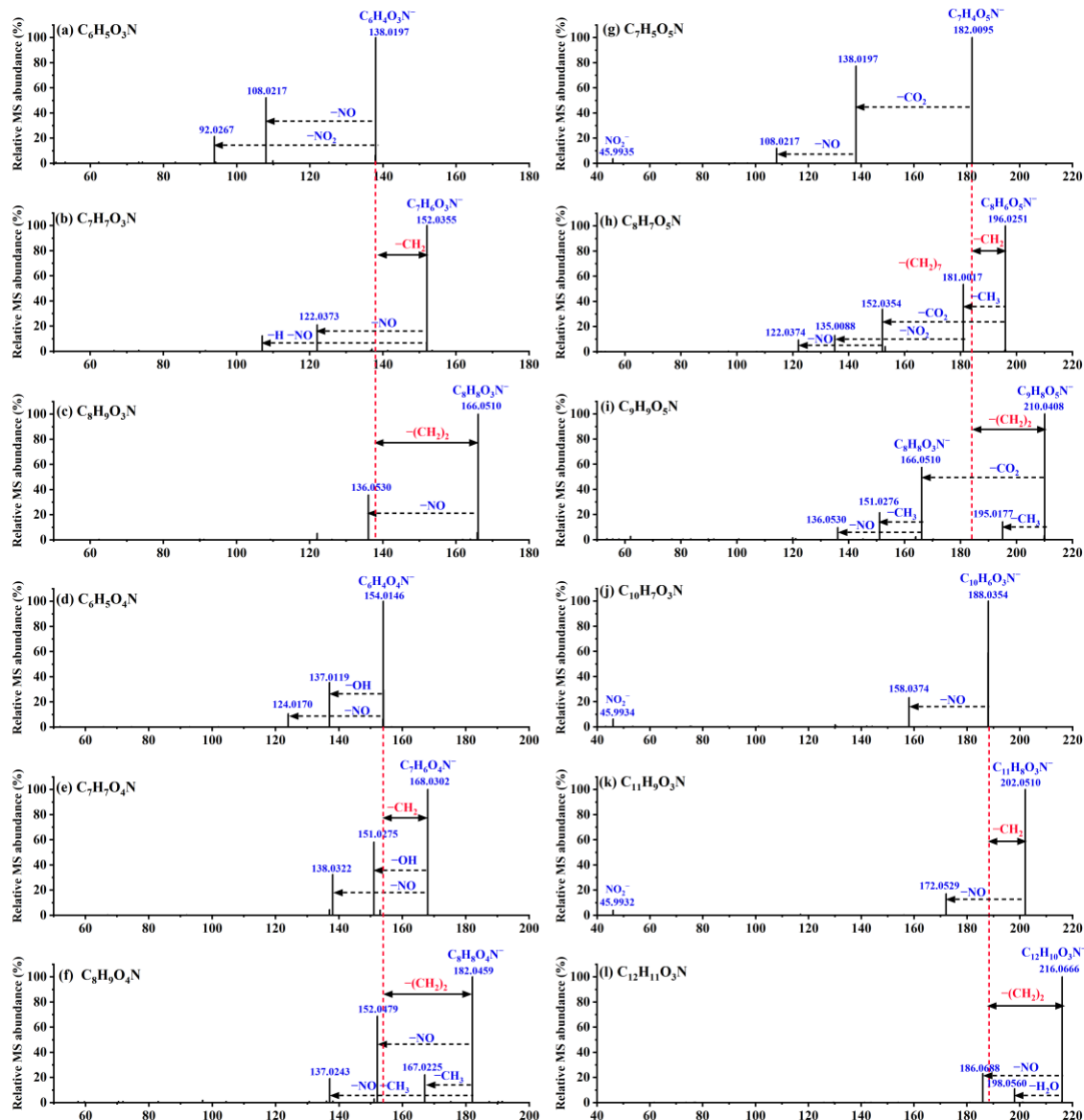


**Figure S2.** MS/MS fragmentation spectra of twelve representative nitroaromatic compounds. The parent ions correspond to four homologous series: (a–c)  $C_6H_5NO_3(CH_2)_n$ , (d–f)  $C_6H_5NO_4(CH_2)_n$ , (g–i)  $C_7H_5NO_5(CH_2)_n$ , and (j–l)  $C_{10}H_7NO_3(CH_2)_n$ , where  $n = 0, 1$ , and  $2$ , respectively.

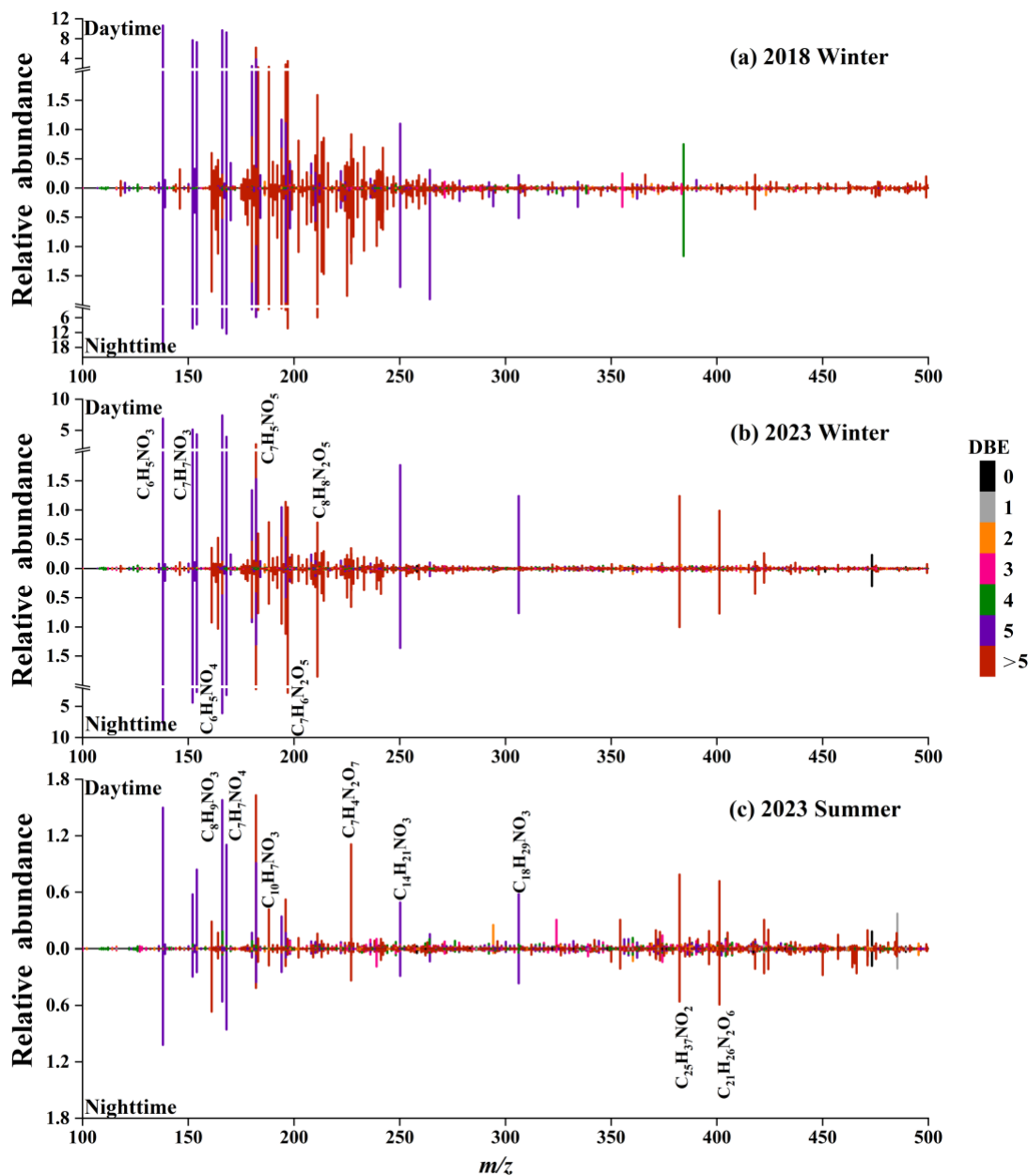


**Figure S3.** The relative abundance distribution of detected CHON compounds in 2018 (a, winter) and 2023 (b, winter; c, summer). The color bar denotes the DBE of CHON compounds. Note that the abundances of IS in each sample were set arbitrarily to 100%. The molecular formulas of the major high-abundance compounds highlighted in panels (a-c) are  $C_6H_5NO_3$ ,  $C_7H_7NO_4$ ,  $C_8H_9NO_3$ , and  $C_7H_6N_2O_5$ .

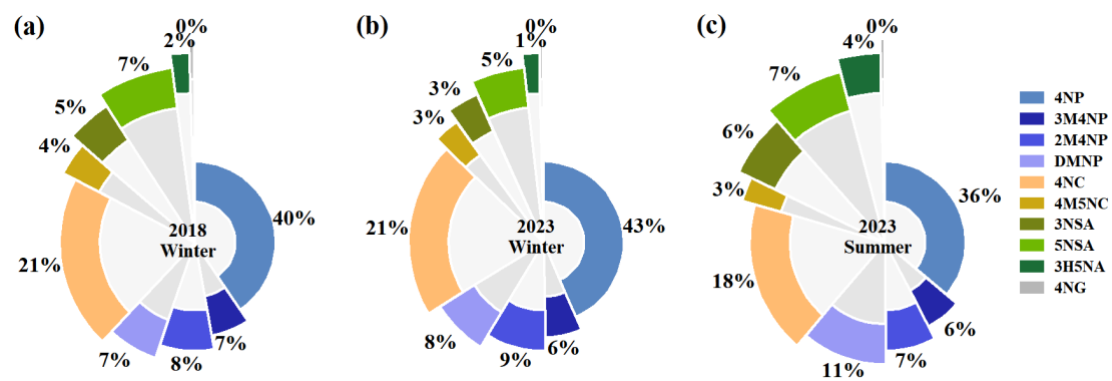


**Figure S4.** Composition of individual nitroaromatic compounds (NACs) in (a) winter 2018, (c) winter 2023, and (e) summer 2023.

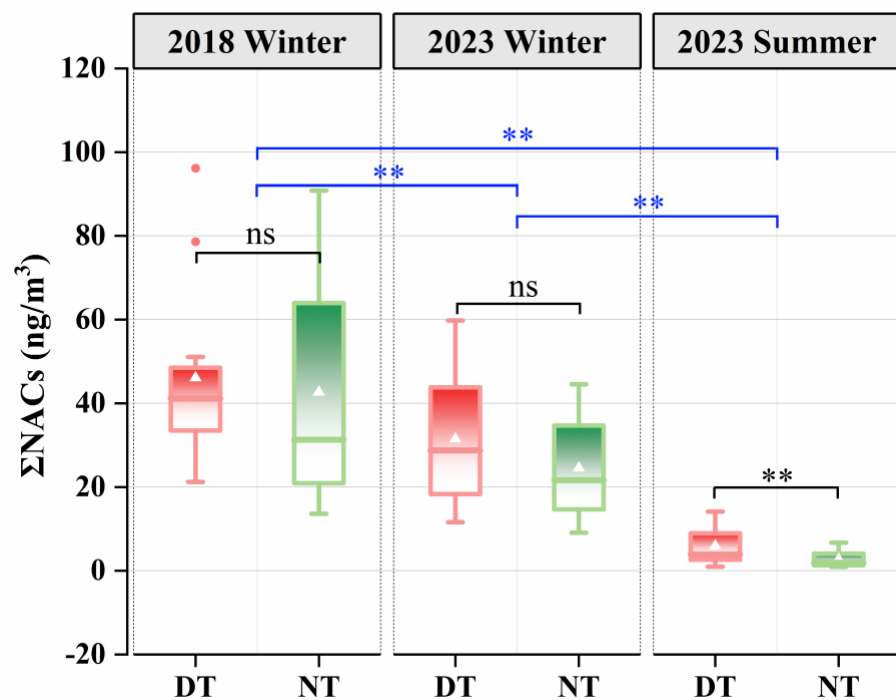


**Figure S5.** Interannual, seasonal, and day-night variability of total NACs ( $\Sigma$ NACs). The whisker plot boundaries are between the 10th and 90th percentiles, the box displays the 25th, 50th (median), and 75th percentiles, and the triangles display the mean values. Asterisks denote statistical significance (t-test,  $*p < 0.05$ ,  $**p < 0.01$ ), while “ns” indicates no significance.

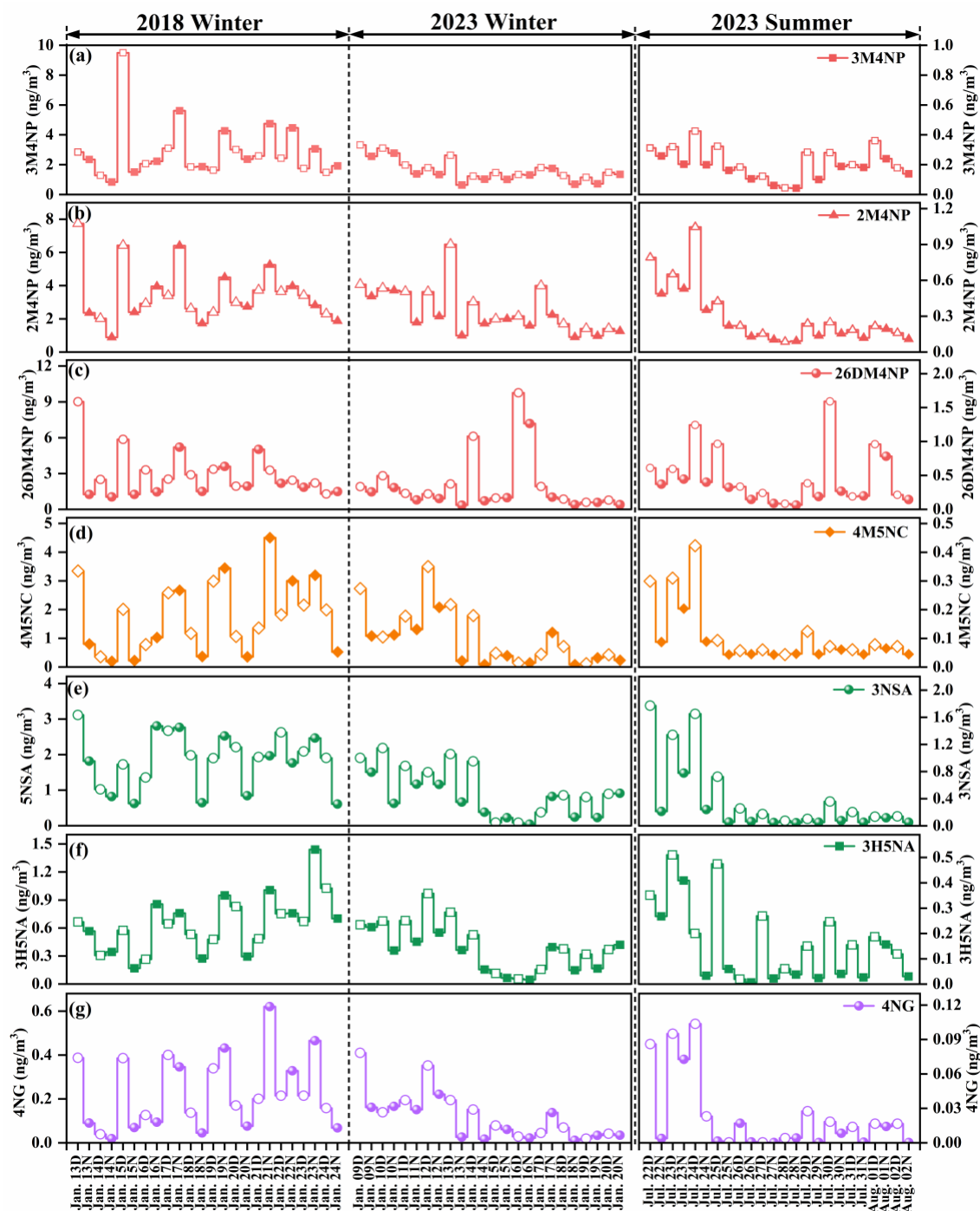


**Figure S6.** Temporal variations in (a) 3M4NP, (b) 2M4NP, (c) 26DM4NP, (d) 4M5NC, (e) 3NSA, (f) 3H5NA, and (g) 4NG during winter 2018, winter 2023, and summer 2023. Daytime and nighttime samples are denoted by empty and solid marks, respectively.

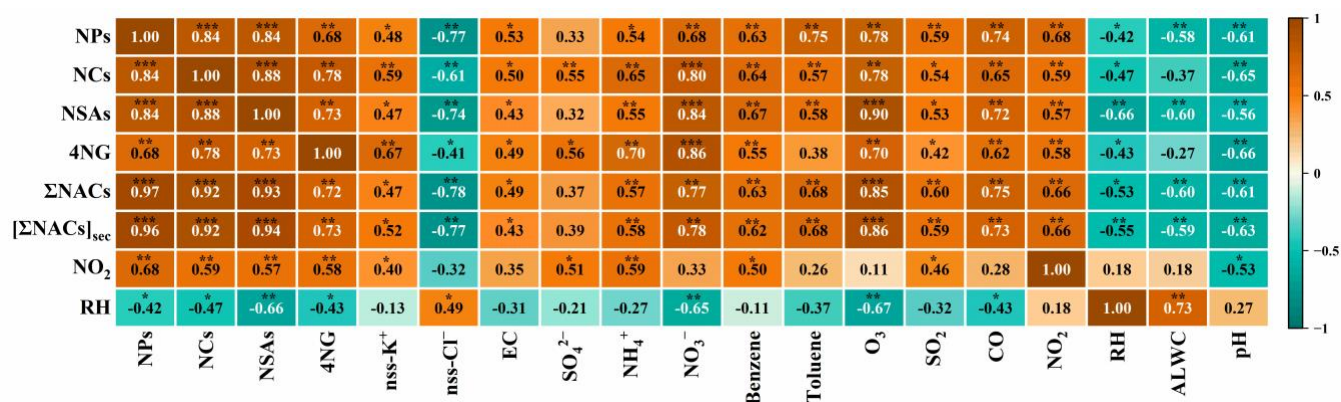


**Figure S7.** Pearson correlation matrix between four NAC groups as well as their correlations with VOC precursors, aerosol components, oxidants (NO<sub>2</sub> and O<sub>3</sub>), ALWC and pH in summer 2023 (n=24).

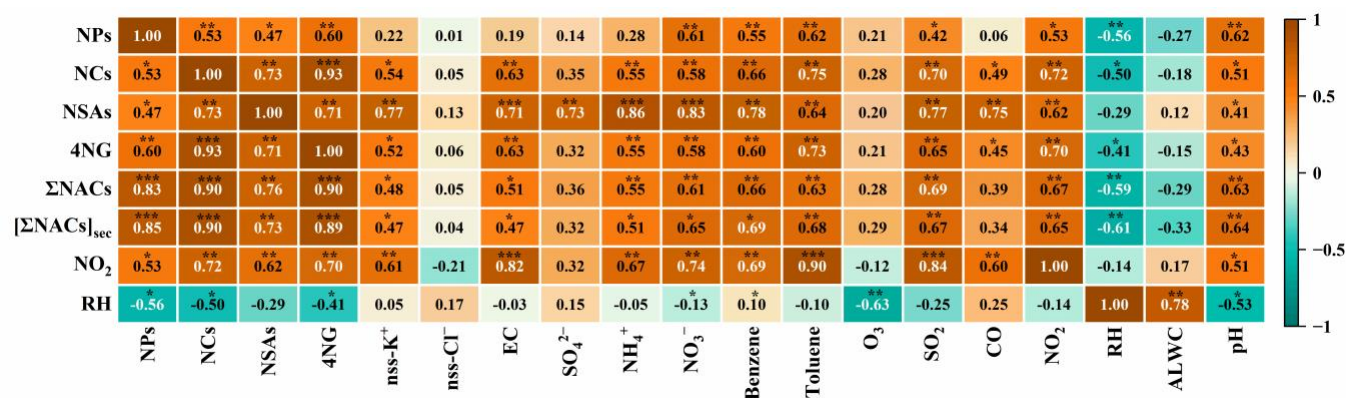


**Figure S8.** Pearson correlation matrix between four NAC groups as well as their correlations with VOC precursors, aerosol components, oxidants (NO<sub>2</sub> and O<sub>3</sub>), ALWC and pH in winter 2023 (n=24).

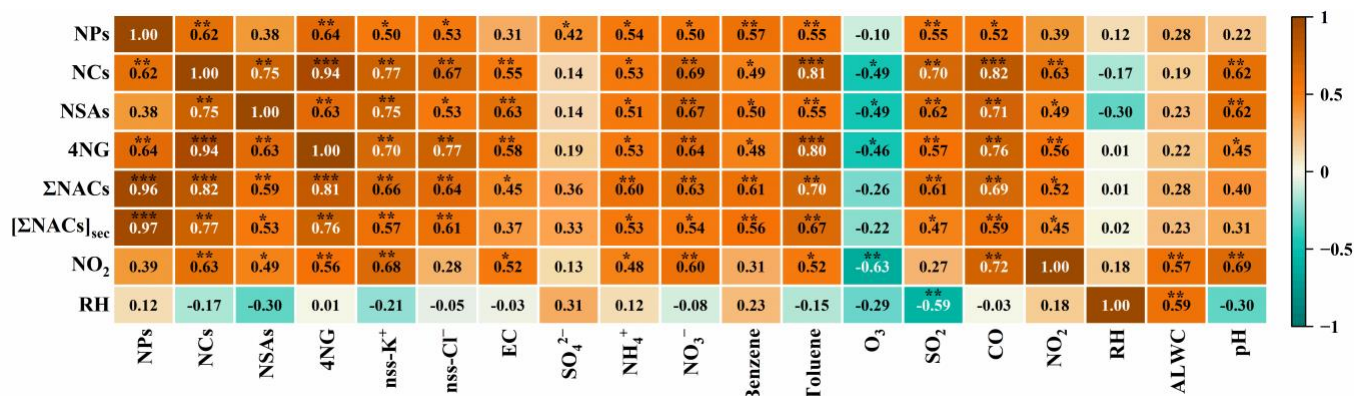


**Figure S9.** Pearson correlation matrix between four NAC groups as well as their correlations with VOC precursors, aerosol components, oxidants (NO<sub>2</sub> and O<sub>3</sub>), ALWC and pH in winter 2018 (n=24).

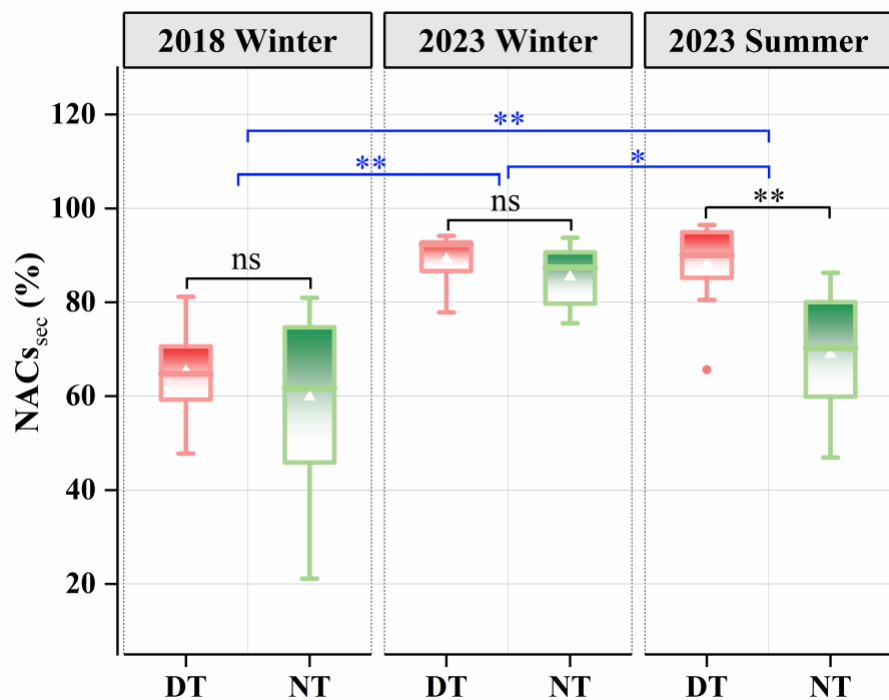


**Figure S10.** Interannual, seasonal, and day-night variability for average contributions of secondarily formed NACs to the total measured NAC mass. The whisker plot boundaries are between the 10th and 90th percentiles, the box displays the 25th, 50th (median), and 75th percentiles, and the triangles display the mean values. Asterisks denote statistical significance (t-test,  $*p < 0.05$ ,  $**p < 0.01$ ), while “ns” indicates no significance.

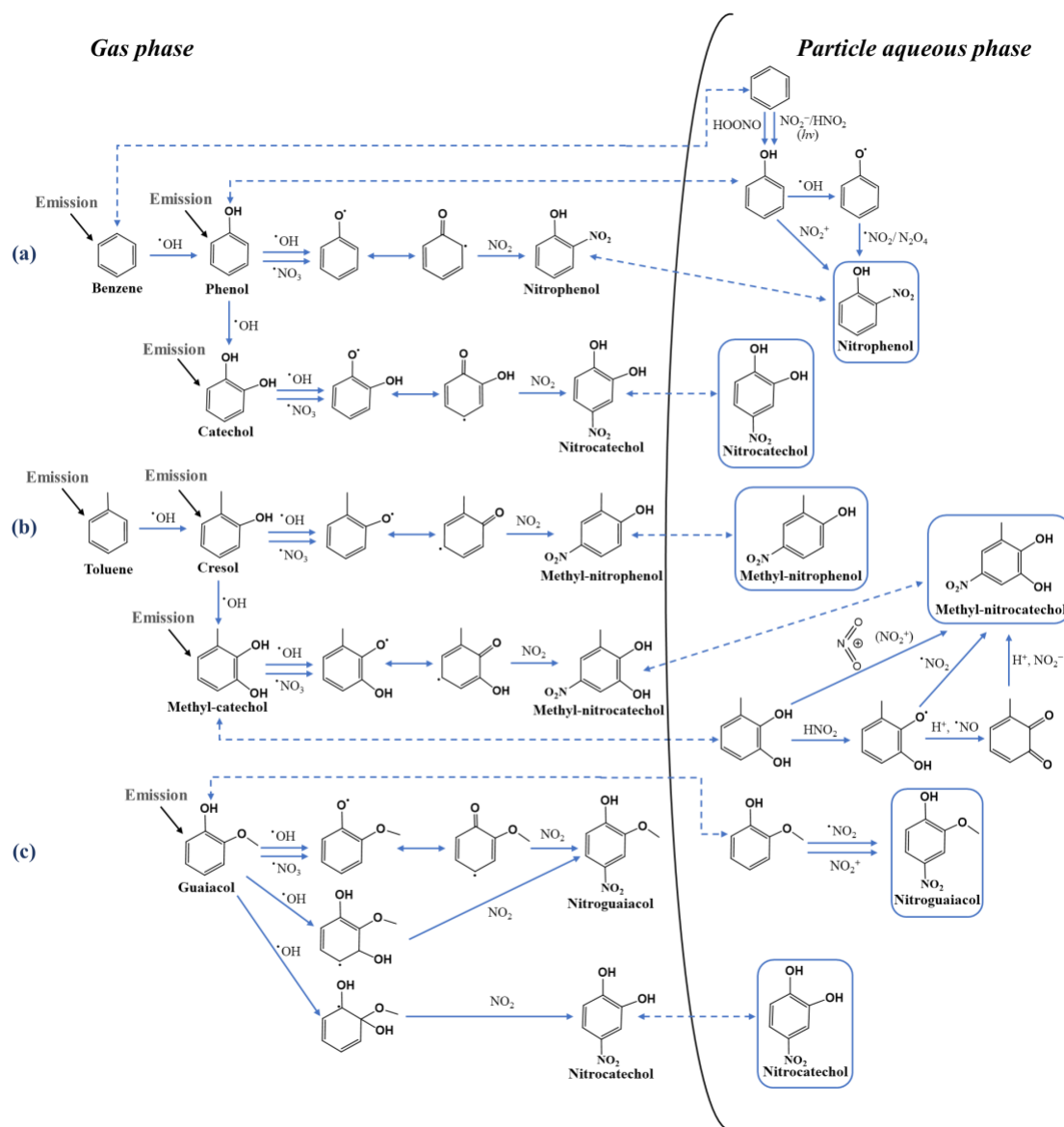


**Figure S11.** Schematic presentation for the formation pathways of secondary NACs from the oxidation of benzene, toluene, phenol, methylcatechol, and guaiacol (2-methoxyphenol) in the atmosphere (Vione et al., 2004; Finewax et al., 2018; Frka et al., 2016; Vidovic et al., 2018; Berndt and Böge, 2003; Bolzacchini et al., 2001; Inuma et al., 2010; Lauraguais et al., 2014; Kroflic et al., 2015; Yang et al., 2016). For simplicity, only single isomers and not all reaction pathways are shown.

**Table S1.** Summary of PM<sub>2.5</sub>, gaseous pollutants, major PM<sub>2.5</sub> components, aromatic VOCs, and meteorological parameters during the winter 2018 and summer and winter 2023 sampling campaigns in Shanghai. Abbreviations include aerosol liquid water content (ALWC), temperature (T), relative humidity (RH), and wind speed (WS). Values are presented as mean  $\pm$  standard deviation.

| Species                                                                                                                          | 2018            |                 |                 | 2023            |                 |                 | 2023            |                 |                 |
|----------------------------------------------------------------------------------------------------------------------------------|-----------------|-----------------|-----------------|-----------------|-----------------|-----------------|-----------------|-----------------|-----------------|
|                                                                                                                                  | Winter          | Daytime         | Nighttime       | Winter          | Daytime         | Nighttime       | Summer          | Daytime         | Nighttime       |
| I. PM <sub>2.5</sub> , gaseous pollutants, major PM <sub>2.5</sub> components ( $\mu\text{g m}^{-3}$ ), and aromatic VOCs (ppbv) |                 |                 |                 |                 |                 |                 |                 |                 |                 |
| PM <sub>2.5</sub>                                                                                                                | 76.6 $\pm$ 30.4 | 79.2 $\pm$ 25.3 | 74.0 $\pm$ 34.6 | 50.8 $\pm$ 34.0 | 50.1 $\pm$ 31.4 | 51.6 $\pm$ 36.4 | 14.4 $\pm$ 7.68 | 16.3 $\pm$ 7.43 | 12.5 $\pm$ 7.43 |
| SO <sub>2</sub>                                                                                                                  | 11.1 $\pm$ 3.40 | 12.0 $\pm$ 3.32 | 10.2 $\pm$ 3.24 | 8.08 $\pm$ 2.18 | 8.34 $\pm$ 2.29 | 7.81 $\pm$ 2.03 | 4.56 $\pm$ 1.15 | 5.05 $\pm$ 1.37 | 4.06 $\pm$ 0.90 |
| NO <sub>2</sub>                                                                                                                  | 74.7 $\pm$ 26.4 | 65.1 $\pm$ 14.3 | 84.4 $\pm$ 31.7 | 55.5 $\pm$ 26.5 | 53.8 $\pm$ 21.8 | 59.2 $\pm$ 30.4 | 17.8 $\pm$ 6.12 | 16.8 $\pm$ 4.59 | 19.5 $\pm$ 5.17 |
| CO                                                                                                                               | 1174 $\pm$ 307  | 1194 $\pm$ 202  | 1154 $\pm$ 384  | 996 $\pm$ 274   | 988 $\pm$ 266   | 1004 $\pm$ 282  | 524 $\pm$ 102   | 548 $\pm$ 81.3  | 500 $\pm$ 114   |
| O <sub>3</sub>                                                                                                                   | 43.9 $\pm$ 20.8 | 53.1 $\pm$ 16.9 | 34.6 $\pm$ 20.3 | 42.8 $\pm$ 22.5 | 50.9 $\pm$ 21.4 | 34.6 $\pm$ 23.2 | 61.3 $\pm$ 17.3 | 75.9 $\pm$ 24.3 | 46.7 $\pm$ 11.7 |
| OM                                                                                                                               | 19.4 $\pm$ 7.21 | 20.6 $\pm$ 6.85 | 18.3 $\pm$ 7.37 | 11.6 $\pm$ 9.62 | 11.2 $\pm$ 7.19 | 12.0 $\pm$ 11.5 | 5.56 $\pm$ 4.30 | 6.43 $\pm$ 4.82 | 4.57 $\pm$ 3.01 |
| EC                                                                                                                               | 3.89 $\pm$ 2.69 | 3.93 $\pm$ 1.63 | 3.85 $\pm$ 3.44 | 1.66 $\pm$ 1.06 | 1.64 $\pm$ 0.78 | 1.68 $\pm$ 1.28 | 1.03 $\pm$ 0.40 | 1.23 $\pm$ 1.07 | 0.83 $\pm$ 0.62 |
| SO <sub>4</sub> <sup>2-</sup>                                                                                                    | 10.5 $\pm$ 5.33 | 11.9 $\pm$ 6.72 | 9.51 $\pm$ 5.50 | 3.99 $\pm$ 2.05 | 4.12 $\pm$ 2.11 | 3.86 $\pm$ 1.98 | 2.16 $\pm$ 1.37 | 2.19 $\pm$ 1.13 | 2.13 $\pm$ 1.52 |
| NO <sub>3</sub> <sup>-</sup>                                                                                                     | 26.9 $\pm$ 13.9 | 28.9 $\pm$ 11.6 | 24.8 $\pm$ 15.7 | 18.6 $\pm$ 15.9 | 18.0 $\pm$ 14.4 | 19.2 $\pm$ 17.3 | 1.30 $\pm$ 0.77 | 1.17 $\pm$ 0.38 | 0.91 $\pm$ 0.73 |
| NH <sub>4</sub> <sup>+</sup>                                                                                                     | 10.2 $\pm$ 4.94 | 11.0 $\pm$ 4.69 | 9.46 $\pm$ 5.07 | 5.45 $\pm$ 3.97 | 5.36 $\pm$ 3.71 | 5.53 $\pm$ 4.20 | 0.84 $\pm$ 0.60 | 0.87 $\pm$ 0.57 | 0.82 $\pm$ 0.62 |
| nss-Cl <sup>-</sup>                                                                                                              | 1.60 $\pm$ 0.65 | 1.67 $\pm$ 0.64 | 1.53 $\pm$ 0.65 | 1.34 $\pm$ 0.62 | 1.42 $\pm$ 0.64 | 1.27 $\pm$ 0.59 | 0.56 $\pm$ 0.39 | 0.49 $\pm$ 0.36 | 0.62 $\pm$ 0.41 |
| ALWC                                                                                                                             | 55.6 $\pm$ 57.2 | 30.9 $\pm$ 24.1 | 80.3 $\pm$ 68.8 | 60.2 $\pm$ 44.6 | 40.4 $\pm$ 35.6 | 80.1 $\pm$ 43.9 | 5.65 $\pm$ 3.50 | 4.41 $\pm$ 2.55 | 6.88 $\pm$ 3.87 |
| pH                                                                                                                               | 3.57 $\pm$ 0.39 | 3.59 $\pm$ 0.38 | 3.56 $\pm$ 0.40 | 3.18 $\pm$ 0.21 | 3.22 $\pm$ 0.23 | 3.14 $\pm$ 0.19 | 2.84 $\pm$ 1.57 | 2.77 $\pm$ 1.56 | 2.91 $\pm$ 1.58 |
| Benzene                                                                                                                          | 0.95 $\pm$ 0.32 | 0.98 $\pm$ 0.29 | 0.92 $\pm$ 0.36 | 0.75 $\pm$ 0.34 | 0.76 $\pm$ 0.33 | 0.75 $\pm$ 0.35 | 0.17 $\pm$ 0.11 | 0.14 $\pm$ 0.11 | 0.21 $\pm$ 0.09 |
| Toluene                                                                                                                          | 2.38 $\pm$ 1.48 | 2.40 $\pm$ 1.17 | 2.36 $\pm$ 1.74 | 1.52 $\pm$ 1.22 | 1.52 $\pm$ 1.03 | 1.53 $\pm$ 1.38 | 0.37 $\pm$ 0.24 | 0.38 $\pm$ 0.26 | 0.37 $\pm$ 0.22 |
| II. Meteorological parameters                                                                                                    |                 |                 |                 |                 |                 |                 |                 |                 |                 |
| T (°C)                                                                                                                           | 6.18 $\pm$ 2.95 | 8.29 $\pm$ 2.07 | 4.08 $\pm$ 2.06 | 7.64 $\pm$ 3.28 | 9.43 $\pm$ 3.44 | 5.84 $\pm$ 3.13 | 30.8 $\pm$ 1.54 | 32.4 $\pm$ 1.38 | 29.1 $\pm$ 0.59 |
| RH (%)                                                                                                                           | 65.7 $\pm$ 16.3 | 54.2 $\pm$ 14.5 | 77.2 $\pm$ 7.65 | 75.7 $\pm$ 13.9 | 67.6 $\pm$ 14.3 | 83.7 $\pm$ 7.26 | 80.8 $\pm$ 7.60 | 76.0 $\pm$ 7.94 | 85.5 $\pm$ 2.78 |
| WS (m s <sup>-1</sup> )                                                                                                          | 3.32 $\pm$ 1.52 | 4.03 $\pm$ 1.23 | 2.61 $\pm$ 1.44 | 3.39 $\pm$ 1.60 | 3.90 $\pm$ 1.16 | 2.88 $\pm$ 1.81 | 4.70 $\pm$ 1.13 | 5.07 $\pm$ 1.21 | 4.33 $\pm$ 0.91 |

**Table S2.** HPLC-Orbitrap MS method parameters used for the analysis of NOCs.

| Instrument                  | Thermo Scientific HPLC-Orbitrap MS (Exploris 120) |                                                     |       |
|-----------------------------|---------------------------------------------------|-----------------------------------------------------|-------|
| HPLC settings               | Column                                            | Waters Acquity HSS T3 column (1.8 μm, 100 × 2.1 mm) |       |
|                             | Injection Volume                                  | 5 μL                                                |       |
|                             | Mobile Phases                                     | A: Milli-Q water + 0.1% formic acid                 |       |
|                             |                                                   | B: acetonitrile + 0.1% formic acid                  |       |
|                             | Solvent flow                                      | 0.3 mL/min                                          |       |
|                             | Gradient                                          | Time (min)                                          | B (%) |
|                             |                                                   | 2                                                   | 10    |
|                             |                                                   | 2–25                                                | 10–30 |
|                             |                                                   | 25–50                                               | 30–90 |
|                             |                                                   | 50–60                                               | 90    |
| 60–61                       |                                                   | 90–10                                               |       |
| 61–70                       |                                                   | 10                                                  |       |
| MS settings                 | Scan parameters                                   |                                                     |       |
|                             | Scan type                                         | Full MS                                             |       |
|                             | Scan range                                        | 60 to 900 <i>m/z</i>                                |       |
|                             | Resolution                                        | 120000 at <i>m/z</i> 200                            |       |
|                             | Polarity                                          | Negative                                            |       |
|                             | Microscans                                        | 1                                                   |       |
|                             | AGC target                                        | 3e <sup>6</sup>                                     |       |
|                             | Maximum inject time                               | 100 ms                                              |       |
|                             | RF Lens (%)                                       | 70                                                  |       |
|                             | HESI source                                       |                                                     |       |
|                             | sheath gas flow rate                              | 35                                                  |       |
|                             | Aux gas flow rate                                 | 10                                                  |       |
|                             | Sweep gas flow rate                               | 0                                                   |       |
|                             | Spray voltage (kV)                                | –3.0 (ESI–)                                         |       |
|                             | Capillary temperature (°C)                        | 320                                                 |       |
|                             | S-lens RF level                                   | 50                                                  |       |
|                             | Aux gas heater temp. (°C)                         | 350                                                 |       |
| dd-MS <sup>2</sup> settings | Resolution                                        | 30000                                               |       |
|                             | AGC target                                        | 1e <sup>5</sup>                                     |       |
|                             | Maximum inject time                               | 50 ms                                               |       |
|                             | Loop count                                        | 3                                                   |       |
|                             | MSX count                                         | 1                                                   |       |
|                             | Isolation window                                  | 1.0 <i>m/z</i>                                      |       |
|                             | (N)CE / stepped (N)CE                             | ce:10, 20, 40                                       |       |

**Table S3.** Linearity, coefficient of determination ( $R^2$ ), recovery, method limits of detection (LOD) and quantification (LOQ), and precision of nitroaromatic compounds.

| Compound (abbreviation)               | $m/z$    | Linear range<br>(ng mL <sup>-1</sup> ) | $R^2$ | Recovery<br>(%) | LOD <sup>a</sup><br>(ng mL <sup>-1</sup> ) | LOQ <sup>b</sup><br>(ng mL <sup>-1</sup> ) | RSD <sup>c</sup><br>(%) |
|---------------------------------------|----------|----------------------------------------|-------|-----------------|--------------------------------------------|--------------------------------------------|-------------------------|
| 4-Nitrophenol (4NP)                   | 138.0197 | 5.00–500                               | 0.998 | 93.5%           | 0.26                                       | 0.73                                       | 1.9%                    |
| 3-Methyl-4-nitrophenol (3M4NP)        | 152.0353 | 5.00–500                               | 0.999 | 103.7%          | 0.34                                       | 0.67                                       | 2.1%                    |
| 2-Methyl-4-nitrophenol (2M4NP)        | 152.0353 | 5.00–500                               | 0.996 | 93.9%           | 0.19                                       | 0.85                                       | 3.1%                    |
| 2,6-Dimethyl-4-nitrophenol (2,6DM4NP) | 166.0510 | 5.00–500                               | 0.996 | 97.4%           | 0.49                                       | 0.98                                       | 1.5%                    |
| 4-Nitrocatechol (4NC)                 | 154.0146 | 5.00–500                               | 0.995 | 98.7%           | 0.23                                       | 0.71                                       | 2.7%                    |
| 4-Methyl-5-nitrocatechol (4M5NC)      | 168.0302 | 5.00–500                               | 0.997 | 103.4%          | 0.34                                       | 0.84                                       | 3.4%                    |
| 4-Nitroguaiacol (4NG)                 | 168.0302 | 5.00–500                               | 0.998 | 107.6%          | 0.15                                       | 0.56                                       | 2.1%                    |
| 3-nitrosalicylic acid (3NSA)          | 182.0095 | 5.00–500                               | 0.997 | 97.5%           | 0.14                                       | 0.31                                       | 2.8%                    |
| 5-nitrosalicylic acid (5NSA)          | 182.0095 | 5.00–500                               | 0.999 | 92.7%           | 0.51                                       | 1.17                                       | 1.7%                    |
| 3-hydroxy-5-nitrobenzoic acid (3H5NA) | 182.0095 | 5.00–500                               | 0.996 | 96.8%           | 0.28                                       | 0.64                                       | 2.5%                    |

<sup>a</sup> Method detection limit defined as 3-fold standard deviation of 1 ng/mL standard solution signals;

<sup>b</sup> Limit of quantification defined as 10-fold standard deviation of 1 ng/mL standard solution signals;

<sup>c</sup> repeatabilities of six replicate standard injections.

**Table S4.** Summary of molecular characteristics of CHON compounds detected in urban PM<sub>2.5</sub> samples from Shanghai.

| Sample ID              | Number of<br>formulas | % of formulas<br>number with<br>O/N $\geq$ 3 | % of formulas<br>intensity with<br>O/N $\geq$ 3 | MW <sub>w</sub> | H/C <sub>w</sub> | O/C <sub>w</sub> | DBE <sub>w</sub> | Xc <sub>w</sub> |
|------------------------|-----------------------|----------------------------------------------|-------------------------------------------------|-----------------|------------------|------------------|------------------|-----------------|
| <b>2018<br/>Winter</b> | 581 – 1410<br>(2530)  | 67 ± 3                                       | 72 ± 5                                          | 197 ± 10.1      | 0.92 ± 0.04      | 0.37 ± 0.03      | 6.72 ± 0.23      | 2.51 ± 0.05     |
| <b>2023<br/>Winter</b> | 391 – 1426<br>(2685)  | 75 ± 4                                       | 83 ± 5                                          | 199 ± 9.63      | 1.01 ± 0.04      | 0.42 ± 0.04      | 6.02 ± 0.13      | 2.21 ± 0.03     |
| <b>2023<br/>Summer</b> | 678 – 1242<br>(2150)  | 64 ± 5                                       | 67 ± 7                                          | 298 ± 34.6      | 1.23 ± 0.06      | 0.50 ± 0.09      | 5.14 ± 0.21      | 1.85 ± 0.07     |

**Table S5.** Comparison of H/C<sub>w</sub> and O/C<sub>w</sub> ratios of CHON compounds in this study and other studies shown in Figure 3d.

| Sample/type               | Site/type                  | Season          | Extraction solution | Color/shape | H/C <sub>w</sub> | O/C <sub>w</sub> | Instrument  | References                |
|---------------------------|----------------------------|-----------------|---------------------|-------------|------------------|------------------|-------------|---------------------------|
| PM <sub>2.5</sub>         | Shanghai                   | 2018 Winter     | Methanol            | ●           | 0.92±0.04        | 0.37±0.03        | Orbitrap MS | This study                |
|                           |                            | 2023 Winter     |                     | ●           | 1.01±0.04        | 0.42±0.04        |             |                           |
|                           |                            | 2023 Summer     |                     | ●           | 1.23±0.06        | 0.50±0.09        |             |                           |
| BBOA1 <sup>a,b</sup>      | Musa                       |                 | Methanol            | ■           | 1.34             | 0.31             | FT-ICR MS   | (Tang et al., 2020)       |
|                           | Hevea                      |                 | /Water              |             | 1.22             | 0.37             |             |                           |
| CCOA1 <sup>a,c</sup>      | Anthracite                 |                 | Methanol            | ▲           | 0.87             | 0.32             |             | (Tang et al., 2020)       |
|                           | Bituminous coal            |                 | /Water              |             | 0.97             | 0.21             |             |                           |
| Tunnel OA <sup>a</sup>    |                            |                 | Methanol /Water     | ▼           | 1.65             | 0.39             |             | (Tang et al., 2020)       |
| Vehicle <sup>a</sup>      |                            |                 | Methanol /Water     | ◆           | 1.53             | 0.33             |             | (Tang et al., 2020)       |
| BBOA2                     | Hunan                      |                 | Water               | ■           | 1.2              | 0.28             |             | (Cai et al., 2020)        |
|                           | Hubei                      |                 |                     |             | 1.21             | 0.28             |             |                           |
| off-road engine emissions | Excavator (idling)         |                 | Water               | ◆           | 1.14             | 0.32             |             | (Cui et al., 2019)        |
|                           | Excavator (working)        |                 |                     |             | 1.06             | 0.38             |             |                           |
|                           | Excavator (moving)         |                 |                     |             | 1.14             | 0.31             |             |                           |
|                           | Excavator                  |                 |                     |             | 1.08             | 0.37             |             |                           |
|                           | Vessel (heavy oil)         |                 |                     |             | 0.94             | 0.4              |             |                           |
|                           | Vessel (diesel)            |                 |                     |             | 1.14             | 0.33             |             |                           |
|                           | Vessel                     |                 |                     |             | 1.04             | 0.34             |             |                           |
| BBOA3                     | Corn                       |                 | water               | ■           | 1.13             | 0.43             | FT-ICR MS   | (Song et al., 2018)       |
|                           | Rice                       |                 |                     |             | 1.1              | 0.45             |             |                           |
|                           | Pine                       |                 |                     |             | 1.07             | 0.42             |             |                           |
| CCOA2                     |                            |                 | Water               | ▲           | 1.01             | 0.27             |             | (Song et al., 2018)       |
| CCOA3                     | Yulin                      |                 | Water               | ▲           | 1                | 0.3              |             | (Song et al., 2019)       |
|                           | Shuangyashan               |                 |                     |             | 1                | 0.31             |             |                           |
|                           | Yongcheng                  |                 |                     |             | 1                | 0.24             |             |                           |
|                           |                            |                 |                     |             | 0.94             | 0.21             |             |                           |
| CCOA4                     | Yulin                      |                 | Methanol            | ▲           | 0.94             | 0.19             |             | (Song et al., 2019)       |
|                           | Shuangyashan               |                 |                     |             | 0.94             | 0.21             |             |                           |
|                           | Yongcheng                  |                 |                     |             | 0.88             | 0.11             |             |                           |
|                           |                            |                 |                     |             | 0.84             | 0.11             |             |                           |
| Cloud                     | France                     |                 | Water               | ★           | 1.59             | 0.39             |             | (Bianco et al., 2018)     |
|                           | Colorado                   |                 |                     |             | 1.56             | 0.72             |             | (Zhao et al., 2013)       |
| Rainwater                 | Northeastern United States |                 |                     |             | 1.9              | 1.6              |             | (Altieri et al., 2009)    |
| PM <sub>2.5</sub>         | Cambridge                  | Winter          | Acetonitrile /water | ●           | 1.67             | 0.74             | Orbitrap MS | (Rincón et al., 2012)     |
|                           |                            | Summer          |                     |             | 1.65             | 0.8              |             |                           |
| PM <sub>2.5</sub>         | Pearl River Delta          |                 | Water               | ●           | 1.15±0.31        | 0.41±0.19        | Orbitrap MS | (Lin et al., 2012)        |
| TSP                       | Virginia                   |                 | Water               | ●           | 1.41±0.20        | 0.47±0.23        | FT-ICR MS   | (Willoughby et al., 2014) |
|                           |                            |                 | Pyridine            |             | 1.53±0.42        | 0.54±0.32        |             |                           |
|                           |                            |                 | Acetonitrile        |             | 1.66±0.42        | 0.48±0.35        |             |                           |
| PM <sub>0.18-1.8</sub>    | California                 | after midnight  |                     | ●           | 1.5±0.0          | 0.78±0.08        | Orbitrap MS | (O'brien et al., 2014)    |
|                           |                            | morning         |                     |             | 1.5±0.0          | 0.73±0.03        |             |                           |
|                           |                            | afternoon       |                     |             | 1.5±0.0          | 0.62±0.01        |             |                           |
|                           |                            | before midnight |                     |             | 1.5±0.05         | 0.64±0.01        |             |                           |
| PM <sub>2.5</sub>         | Beijing                    | Hazy            | DCM                 | ●           | 1.23±0.40        | 0.48±0.25        | FT-ICR MS   | (Jiang et al., 2016)      |
|                           |                            | Clear           |                     |             | 1.26±0.42        | 0.44±0.23        |             |                           |
|                           |                            | Hazy            | Water               |             | 1.13±0.38        | 0.45±0.22        |             |                           |

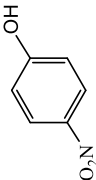
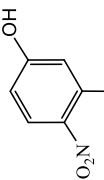
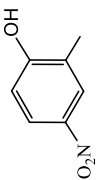
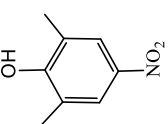
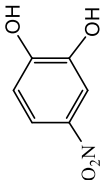
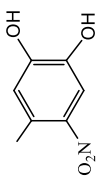
|                   |           |                |                     |   |           |           |             |                      |
|-------------------|-----------|----------------|---------------------|---|-----------|-----------|-------------|----------------------|
|                   |           | Clear          |                     |   | 1.24±0.39 | 0.46±0.19 |             |                      |
| PM <sub>2.5</sub> | Shanghai  | Whole year     | Acetonitrile        | ● | 1         | 0.5       | Orbitrap MS | (Wang et al., 2017)  |
| PM <sub>2.5</sub> | Mainz     | low-pollution  | Acetonitrile /water | ● | 1.06      | 0.56      | Orbitrap MS | (Wang et al., 2018a) |
|                   | Beijing   | low-pollution  |                     | ● | 0.96      | 0.5       |             |                      |
|                   |           | high-pollution |                     |   | 0.92      | 0.5       |             |                      |
| PM <sub>2.5</sub> | Changchun |                | Acetonitrile /water | ● | 0.94±0.01 | 0.51±0.00 | Orbitrap MS | (Wang et al., 2021)  |
|                   | Shanghai  |                |                     | ● | 0.86±0.01 | 0.56±0.01 |             |                      |
|                   | Guangzhou |                |                     | ● | 0.89±0.01 | 0.58±0.01 |             |                      |
| PM <sub>2.5</sub> | Guangzhou | Whole year     | Methanol            | ● | 1.35±0.06 | 0.51±0.04 | FT-ICR MS   | (Jiang et al., 2022) |
| PM <sub>2.5</sub> | Beijing   | Winter         | Methanol            | ● | 1.00      | 0.61      | Orbitrap MS | (Wang et al., 2024)  |
|                   |           | Summer         |                     | ● | 1.09      | 0.39      |             |                      |
|                   | Harbin    | Winter         |                     | ● | 0.96      | 0.54      |             |                      |
|                   |           | Summer         |                     | ● | 1.10      | 0.36      |             |                      |
|                   | Xi'an     | Winter         |                     | ● | 0.95      | 0.48      |             |                      |
|                   |           | Summer         |                     | ● | 0.97      | 0.33      |             |                      |
|                   | Chengdu   | Winter         |                     | ● | 0.99      | 0.50      |             |                      |
|                   |           | Summer         |                     | ● | 1.08      | 0.34      |             |                      |
|                   | Guangzhou | Winter         |                     | ● | 0.97      | 0.53      |             |                      |
|                   |           | Summer         |                     | ● | 0.99      | 0.29      |             |                      |
|                   | Wuhan     | Winter         |                     | ● | 1.00      | 0.55      |             |                      |
|                   |           | Summer         |                     | ● | 0.96      | 0.96      |             |                      |

a, the data obtained from the ref. have been recalculated using different s/n threshold to the original authors.

b, BBOA denotes biomass burning organic aerosols

c, CCOA denotes coal combustion organic aerosols

**Table S6.** The nitroaromatic compounds quantified by UHPLC-Orbitrap MS.

| Compounds          | [M-H] <sup>-</sup>                                         |          | Error<br>(ppm) | Standard | Structure                                                                             | Concentrations (ng/m <sup>3</sup> ) |           |                    |           |                    |           |
|--------------------|------------------------------------------------------------|----------|----------------|----------|---------------------------------------------------------------------------------------|-------------------------------------|-----------|--------------------|-----------|--------------------|-----------|
|                    | Formula                                                    | Mass     |                |          |                                                                                       | 2018 Winter(n=24)                   |           | 2023 Winter (n=24) |           | 2023 Summer (n=24) |           |
|                    |                                                            |          |                |          |                                                                                       | Range                               | Average   | Range              | Average   | Range              | Average   |
| 4NP                | C <sub>6</sub> H <sub>4</sub> NO <sub>3</sub> <sup>-</sup> | 138.0197 | 0.7            | 4NP      |    | 6.03–54.5                           | 18.4±12.7 | 4.55–24.1          | 11.3±4.55 | 0.31–5.74          | 1.54±1.42 |
| 3M4NP              | C <sub>7</sub> H <sub>6</sub> NO <sub>3</sub> <sup>-</sup> | 152.0353 | 0.8            | 3M4NP    |    | 0.84–9.49                           | 2.86±1.80 | 0.64–3.32          | 1.63±0.73 | 0.04–0.42          | 0.21±0.10 |
| 2M4NP              | C <sub>7</sub> H <sub>6</sub> NO <sub>3</sub> <sup>-</sup> | 152.0353 | 0.6            | 2M4NP    |    | 0.89–7.74                           | 3.44±1.61 | 0.90–6.48          | 2.50±1.32 | 0.09–1.05          | 0.29±0.24 |
| 2,6DM4NP           | C <sub>8</sub> H <sub>8</sub> NO <sub>3</sub> <sup>-</sup> | 166.0510 | 0.8            | 2,6DM4NP |    | 1.04–9.01                           | 2.85±1.81 | 0.35–9.76          | 1.96±2.31 | 0.06–1.59          | 0.45±0.39 |
| Σ <sub>4</sub> NPs |                                                            |          |                |          |                                                                                       | 8.79–76.3                           | 27.5±16.6 | 6.69–32.8          | 17.5±6.69 | 0.53–8.45          | 2.49±1.96 |
| 4NC                | C <sub>6</sub> H <sub>4</sub> NO <sub>4</sub> <sup>-</sup> | 154.0146 | 0.5            | 4NC      |  | 1.70–18.5                           | 9.63±5.55 | 0.71–29.2          | 7.18±6.89 | 0.08–3.47          | 0.81±0.76 |
| 4M5NC              | C <sub>7</sub> H <sub>6</sub> NO <sub>4</sub> <sup>-</sup> | 168.0302 | 0.5            | 4M5NC    |  | 0.19–4.51                           | 1.75±1.21 | 0.08–3.50          | 0.98±0.92 | 0.04–0.42          | 0.10±0.10 |

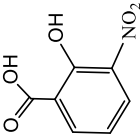
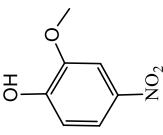
| $\Sigma_2\text{NCs}$     |                                     | 1.89–23.1 | 11.4±6.74 | 0.79–32.7 | 8.16±7.78                                                                            | 0.13–3.77   | 0.91±0.85 |
|--------------------------|-------------------------------------|-----------|-----------|-----------|--------------------------------------------------------------------------------------|-------------|-----------|
| 3NSA                     | $\text{C}_7\text{H}_4\text{NO}_5^-$ | 182.0095  | 0.6       | 3NSA      |   | 0.05–1.77   | 0.36±0.50 |
| 5NSA                     | $\text{C}_7\text{H}_4\text{NO}_5^-$ | 182.0095  | 0.7       | 5NSA      |                                                                                      | 0.04–1.46   | 0.38±0.44 |
| 3H5NA                    | $\text{C}_7\text{H}_4\text{NO}_5^-$ | 182.0095  | 0.6       | 3H5NA     |                                                                                      | 0.01–0.51   | 0.16±0.15 |
| $\Sigma_3\text{NSAs}$    |                                     | 1.77–8.05 | 5.25±1.81 | 0.18–6.07 | 5.42±3.97                                                                            | 0.11–3.59   | 0.91±1.05 |
| 4NG                      | $\text{C}_7\text{H}_6\text{NO}_4^-$ | 168.0302  | 0.4       | 4NG       |  | 0.0003–0.10 | 0.02±0.03 |
| $\Sigma_{10}\text{NACs}$ |                                     | 13.6–96.1 | 44.4±22.6 | 8.67–59.8 | 28.4±14.0                                                                            | 0.93–14.2   | 4.33±3.75 |

**Table S7.** The concentrations of nitroaromatic compounds in this and previous studies.

| Location            | Date                | Concentration (ng m <sup>-3</sup> ) |       |       |                 |                   |       |       |       |      |      |       | References |                           |
|---------------------|---------------------|-------------------------------------|-------|-------|-----------------|-------------------|-------|-------|-------|------|------|-------|------------|---------------------------|
|                     |                     | 4NP(NP)                             | 3M4NP | 2M4NP | 2,6DM4NP (DMNP) | 4NC (NC)          | 4M5NC | 3M6NC | 3M5NC | 3NSA | 5NSA | 4NG   | Total      |                           |
| Ljubljana, Slovenia | Summer, 2010        | 0.15                                | <0.03 | 0.05  |                 | 0.24              | 0.06  | <0.03 | 0.01  | 0.09 | 0.18 | <0.11 | 0.84       | (Kitanovski et al., 2012) |
|                     | Winter, 2010        | 1.8                                 | 0.61  | 0.75  |                 | 75                | 29    | 6.2   | 34    | 1.3  | 1.4  | 0.40  | 150.1      |                           |
| Los Angeles         | Summer, 2010        |                                     |       | 1.24  | 0.92            | 1.67              |       | 0.36  |       | 0.92 |      |       | 5.11       | (Zhang et al., 2013)      |
| Detling, UK         | Winter, 2012        | 0.02                                |       | 5.0   |                 | 2.5               |       | 8.2   |       |      |      |       | 15.72      | (Mohr et al., 2013)       |
| TROPOS, Germany     | Winter, 2014        | 7.09                                | 2.60  | 3.64  | 0.65            |                   |       |       |       | 1.36 | 0.94 |       | 16.28      | (Teich et al., 2017)      |
| Melpitz, Germany    | Winter, 2014        | 4.09                                | 2.44  | 3.64  | 0.91            |                   |       |       |       | 0.66 | 0.32 |       | 12.06      |                           |
|                     | Summer, 2014        | 0.06                                | 0.03  | 0.04  | n.d.            |                   |       |       |       | 0.17 | 0.09 |       | 0.39       |                           |
| Waldstein, Germany  | Summer, 2014        |                                     |       |       |                 |                   |       |       |       | 0.17 | 0.23 |       | 0.4        |                           |
| Mainz, Germany      | 2006–2007           | 2.48(5.98)                          |       |       |                 | 6.40              |       |       |       |      |      |       | 8.88       | (Zhang et al., 2010)      |
| Nagoya, Japan       | Summer, 2013        | 1.1                                 | 0.17  | 0.49  |                 | 0.74              | 0.081 |       |       | 0.33 | 0.75 | 0.04  | 3.70       | (Ikemori et al., 2019)    |
|                     | Fall, 2013          | 7.0                                 | 1.1   | 3.2   |                 | 6.8               | 1.6   |       |       | 0.27 | 0.67 | 0.55  | 21.19      |                           |
| Ostrava, Czechia    | Summer, 2016        | 5.35                                | 3.58  | 4.97  |                 | 37.2              | 21.2  | 3.68  | 13.98 | 0.33 | 0.62 | 0.38  | 91.29      | (Kitanovski et al., 2020) |
|                     | Autumn, 2016        | 0.85                                | 0.15  | 0.26  |                 | 3.88              | 1.44  | 0.23  | 0.70  | 0.31 | 0.73 | <0.03 | 8.55       |                           |
| Hong Kong           | Winter, 2009-2012   | 1.71                                | 0.15  | 1.06  | 0.04            | 3.98              | 1.06  | 0.23  | 1.85  |      |      |       | 10.08      | (Chow et al., 2016)       |
|                     | Spring, 2009-2012   | 0.64                                | 0.06  | 0.37  | 0.01            | 0.83              | 0.15  | 0.10  | 0.22  |      |      |       | 2.38       |                           |
|                     | Summer, 2009-2012   | 0.31                                | 0.02  | 0.19  | 0.01            | 0.77              | 0.16  | 0.07  | 0.34  |      |      |       | 1.87       |                           |
|                     | Autumn, 2009-2012   | 1.07                                | 0.06  | 0.58  | 0.02            | 2.66              | 0.59  | 0.15  | 1.16  |      |      |       | 6.29       |                           |
| Xianghe, China      | July-Aug, 2013      | 0.98                                | 0.09  | 0.32  | 0.06            |                   |       |       |       | 1.21 | 0.88 |       | 3.54       | (Teich et al., 2017)      |
| Wangdu, China       | Summer, 2014        | 2.63                                | 0.21  | 0.68  | 3.45            |                   |       |       |       | 3.14 | 1.63 |       | 11.74      |                           |
|                     |                     | 5.9 <sup>a</sup>                    |       |       |                 |                   |       |       |       |      |      |       |            |                           |
| Yucheng, China      | Summer, 2014        |                                     |       |       |                 | 5.7 <sup>a</sup>  |       |       |       |      |      |       | 5.7        | (Wang et al., 2018b)      |
| Mt. Tai, China      | Summer, 2014        |                                     |       |       |                 | 2.5 <sup>a</sup>  |       |       |       |      |      |       | 2.5        |                           |
| Jinan, China        | Nov, 2013-Jan, 2014 |                                     |       |       |                 | 48.4 <sup>a</sup> |       |       |       |      |      |       | 48.4       |                           |

|                  |                     | 9.8 <sup>a</sup>   |       |       |                   |       |       |       |      | 9.8   |       |                      |
|------------------|---------------------|--------------------|-------|-------|-------------------|-------|-------|-------|------|-------|-------|----------------------|
| Autumn, 2014     |                     |                    |       |       |                   |       |       |       |      |       |       |                      |
| Winter, 2016     |                     | 105.4 <sup>b</sup> |       |       |                   |       |       |       |      | 105.4 |       |                      |
| Spring, 2016     |                     | 34.0 <sup>b</sup>  |       |       |                   |       |       |       |      | 34.0  |       |                      |
| Summer, 2016     |                     | 13.5 <sup>b</sup>  |       |       |                   |       |       |       |      | 13.5  |       |                      |
| Qingdao, China   | Winter, 2019        | 37.94              | 17.21 | 10.43 | 7.86              | 16.67 | 3.78  | 24.78 | 1.70 | 1.46  | 121.8 | (Liang et al., 2020) |
|                  | Dec, 2012-Mar, 2013 | 35.1               |       | 27.2  |                   | 27.1  | 7.38  | 18.6  |      | 8.5   | 123.9 | (Ma et al., 2018)    |
|                  | Apr-Nov, 2012       | 2.86               |       | 1.91  |                   | 2.91  | 0.51  | 2.25  |      | 6.38  | 16.82 |                      |
| Beijing, China   | Summer, 2016        | 2.15               | 0.27  | 0.76  | 0.55              | 1.89  | 0.56  | 0.07  | 0.44 |       | 6.62  | (Wang et al., 2019)  |
|                  | Winter, 2016        | 43.0               | 16.7  | 17.2  | 13.1              | 39.4  | 9.3   |       | 3.6  | 2.4   | 17.6  | (Li et al., 2020b)   |
|                  | Autumn, 2017        | 6.76               | 0.61  |       |                   | 4.54  | 0.85  |       | 2.04 | 4.03  | 0.94  | 20.27                |
|                  | Winter, 2017        | 24.07              | 6.77  |       |                   | 19.49 | 6.50  |       | 5.41 | 5.75  | 5.54  | 77.23                |
| Mt. Wuyi, China  | Spring              | 0.18               | 0.03  |       | 0.06 <sup>c</sup> | 0.34  | 0.20  |       | 0.07 | 0.12  | 0.07  | 1.3                  |
|                  | Summer              | 0.05               | 0.05  |       | 0.06 <sup>c</sup> | 0.14  | 0.19  |       | 0.04 | 0.07  | 0.07  | 0.97                 |
|                  | Autumn              | 0.32               | 0.04  |       | 0.08 <sup>c</sup> | 0.64  | 0.34  |       | 0.09 | 0.23  | 0.07  | 2.1                  |
|                  | Winter              | 0.74               | 0.06  |       | 0.09 <sup>c</sup> | 1.6   | 0.39  |       | 0.19 | 0.55  | 0.05  | 3.9                  |
| Harbin, China    | Winter, 2019        | 6.02               | 9.37  | 16.83 | 0.52              | 4.38  | 14.97 |       | 2.79 | 4.38  | 7.77  | 158.79               |
|                  | Summer, 2021        | 1.09               | 0.42  | 1.39  | 0.04              | 1.07  | 0.13  |       | 0.47 | 1.80  | 0.59  | 7.18                 |
| Beijing, China   | Winter, 2019        | 0.75               | 0.73  | 1.43  | 0.06              | 4.22  | 0.52  |       | 0.80 | 1.50  | 0.37  | 10.80                |
|                  | Summer, 2021        | 1.10               | 0.92  | 1.34  | 0.04              | 0.46  | 0.04  |       | 0.51 | 1.54  | 1.49  | 8.02                 |
| Xi'an, China     | Winter, 2019        | 3.15               | 1.95  | 4.30  | 0.06              | 23.19 | 4.98  |       | 1.30 | 2.42  | 0.80  | 42.97                |
|                  | Summer, 2021        | 0.18               | 0.03  | 0.15  | 0.003             | 0.72  | 0.10  |       | 0.31 | 1.45  | 0.02  | 3.00                 |
| Wuhan, China     | Winter, 2019        | 1.56               | 0.79  | 2.55  | 0.05              | 17.77 | 3.09  |       | 1.26 | 3.04  | 0.22  | 31.04                |
|                  | Summer, 2021        | 0.09               | 0.01  | 0.09  | 0.002             | 0.67  | 0.08  |       | 0.26 | 0.79  | 0.01  | 2.02                 |
| Chengdu, China   | Winter, 2019        | 0.22               | 0.13  | 0.35  | 0.01              | 5.86  | 1.26  |       | 0.35 | 0.81  | 0.01  | 9.15                 |
|                  | Summer, 2021        | 0.89               | 0.55  | 1.52  | 0.06              | 1.15  | 0.11  |       | 0.86 | 3.12  | 0.78  | 9.39                 |
| Guangzhou, China | Winter, 2019        | 0.73               | 0.37  | 1.56  | 0.02              | 9.29  | 1.63  |       | 1.05 | 3.87  | 0.19  | 19.35                |
|                  | Summer, 2021        | 0.32               | 0.04  | 0.24  | 0.01              | 1.60  | 0.17  |       | 0.47 | 0.91  | 0.16  | 3.97                 |
| Lanzhou, China   | Winter, 2017        | 6.53               | 1.69  | 1.96  | 1.76 <sup>c</sup> | 5.79  | 0.23  |       | 0.75 | 0.61  | 1.23  | 20.56                |
|                  |                     | 12.77              | 2.89  | 3.39  | 1.87 <sup>c</sup> | 15.5  | 0.19  |       | 1.15 | 1.08  | 3.29  | 42.13                |
| Xi'an, China     |                     |                    |       |       |                   |       |       |       |      |       |       | (Xu et al., 2026)    |

(Huang et al., 2024)

(Xu et al., 2026)

|                    |       |      |      |                   |       |      |      |      |       |       |
|--------------------|-------|------|------|-------------------|-------|------|------|------|-------|-------|
| Beijing, China     | 6.31  | 2.00 | 2.08 | 1.06 <sup>c</sup> | 4.83  | 0.27 | 0.42 | 0.47 | 1.25  | 18.69 |
| Harbin, China      | 10.57 | 2.96 | 3.74 | 0.38 <sup>c</sup> | 20.71 | 0.42 | 0.55 | 0.41 | 5.12  | 44.87 |
| Taiyuan, China     | 11.68 | 4.75 | 4.47 | 2.68 <sup>c</sup> | 9.84  | 0.49 | 0.67 | 0.73 | 2.98  | 38.3  |
| Guangzhou, China   | 0.87  | 0.01 | 0.02 | 0.25 <sup>c</sup> | 3.28  | 0.16 | 0.17 | 0.38 | 0.41  | 5.55  |
| Chengdu, China     | 5.23  | 0.65 | 1.00 | 0.46 <sup>c</sup> | 8.43  | 0.16 | 0.46 | 0.78 | 1.58  | 18.75 |
| Guiyang, China     | 1.62  | 0.21 | 0.28 | 0.19 <sup>c</sup> | 5.41  | 0.22 | 0.19 | 0.19 | 0.75  | 9.04  |
| Kunming, China     | 1.32  | 0.20 | 0.24 | 0.30 <sup>c</sup> | 3.66  | 0.26 | 0.04 | 0.18 | 0.67  | 6.88  |
| Wuhan, China       | 2.60  | 0.29 | 0.42 | 0.52 <sup>c</sup> | 3.40  | 0.18 | 0.42 | 0.55 | 0.52  | 8.89  |
| Hangzhou, China    | 2.53  | 0.26 | 0.43 | 0.67 <sup>c</sup> | 2.16  | 0.16 | 0.31 | 0.43 | 0.39  | 7.33  |
| Summer, 2017       | 2.80  | 0.16 | 0.67 | 0.59              | 1.52  | 0.30 | 0.32 | 0.92 | 0.014 | 7.83  |
| Winter, 2017       | 20.9  | 3.37 | 4.55 | 2.01              | 11.3  | 1.32 | 1.51 | 3.61 | 0.89  | 52.3  |
| Winter, 2018       | 18.4  | 2.86 | 3.44 | 2.85              | 9.63  | 1.75 | 1.84 | 2.77 | 0.23  | 44.4  |
| Winter, 2023       | 11.4  | 1.63 | 2.50 | 1.96              | 7.18  | 0.98 | 0.92 | 1.38 | 0.12  | 28.4  |
| Summer, 2023       | 1.54  | 0.21 | 0.29 | 0.45              | 0.81  | 0.10 | 0.36 | 0.38 | 0.021 | 4.33  |
| (Cai et al., 2022) |       |      |      |                   |       |      |      |      |       |       |
| This study         |       |      |      |                   |       |      |      |      |       |       |

<sup>a</sup> The identified NACs excluded 2,6-dimethyl-4-nitrophenol (2,6DM4NP).

<sup>b</sup> The identified NACs also included 2,4-dinitrophenol (2,4DNP), and 4-methyl-2,6-dinitrophenol (4M2,6DNP).

<sup>c</sup> The only identified NAC was 2,4-dinitrophenol (2,4DNP)

**Table S8.** Average daytime and nighttime concentrations and relative contributions of primary and secondary NACs estimated using the CO-tracer method during the three campaigns.

|                        |                  | [NACs] <sub>sec</sub>                 |                   | [NACs] <sub>pri</sub>                 |                   |
|------------------------|------------------|---------------------------------------|-------------------|---------------------------------------|-------------------|
|                        |                  | Concentration<br>(ng/m <sup>3</sup> ) | Proportion<br>(%) | Concentration<br>(ng/m <sup>3</sup> ) | Proportion<br>(%) |
| <b>2018<br/>Winter</b> | <b>Daytime</b>   | 31.7 ± 18.5                           | 65 ± 9            | 14.4 ± 2.43                           | 35 ± 9            |
|                        | <b>Nighttime</b> | 28.8 ± 21.7                           | 60 ± 17           | 13.9 ± 4.66                           | 40 ± 17           |
|                        | <b>Average</b>   | 30.2 ± 20.2                           | 63 ± 14           | 14.2 ± 3.73                           | 37 ± 14           |
| <b>2023<br/>Winter</b> | <b>Daytime</b>   | 30.5 ± 14.8                           | 89 ± 6            | 2.97 ± 0.80                           | 11 ± 6            |
|                        | <b>Nighttime</b> | 20.5 ± 10.2                           | 85 ± 6            | 2.93 ± 0.90                           | 15 ± 6            |
|                        | <b>Average</b>   | 25.5 ± 13.7                           | 87 ± 6            | 2.95 ± 0.85                           | 13 ± 6            |
| <b>2023<br/>Summer</b> | <b>Daytime</b>   | 5.52 ± 4.35                           | 88 ± 8            | 0.43 ± 0.05                           | 12 ± 8            |
|                        | <b>Nighttime</b> | 2.07 ± 1.76                           | 69 ± 13           | 0.64 ± 0.17                           | 31 ± 13           |
|                        | <b>Average</b>   | 3.79 ± 3.74                           | 78 ± 15           | 0.54 ± 0.17                           | 21 ± 14           |

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