Supplement of

Molecular characteristics, sources, and formation pathways of organosulfur compounds in ambient aerosol in Guangzhou, South China

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Measurements for PM$_{2.5}$ and Organics

A total of 55 PM$_{2.5}$ samples collected on prebaked quartz fiber filters once a week at Guangzhou from July, 2017 to June, 2018 (June–September: summer, October–November: fall; December–February: winter; March–May: spring) over a period of 24 h with a high-volume air sampler at a flow rate of 1 m$^3$·min$^{-1}$. Quartz fiber filters were preheated at 450°C for 6 h before used and weighed. After sampling, each filter was wrapped with prebaked aluminum foil, sealed. Before weighing again, the PM$_{2.5}$ samples were kept at constant temperature and humidity for 24 h. The difference between two weighing is the amount of collected PM$_{2.5}$. A punch of filter (1.5 cm$^2$) was used for carbon concentration measurement. The concentration of organic and elemental carbon were measured using an OC/EC analyzer (Sunset Laboratory, Inc.) following the NIOSH870 thermal optical transmittance (TOT) standard method. We converted OC to organic mass using a typical ratio of OM/OC of 1.8 (Tolocka and Turpin, 2012). Detailed information about the analysis procedures of chemical tracers, and meteorological parameters have been described in previous studies (Jiang et al., 2021b; Jiang et al., 2021a) and are included in the Table S12. The organic tracers’ analysis performed included levoglucosan, polycyclic aromatic hydrocarbons [PAHs], steranes, and hopanes, biogenic SOA tracers (isoprene-derived SOA, MTLs; monoterpane-derived SOA, MSOA), fatty acids, long-chain alkanes. Online data regarding temperature, RH, and NO$_x$ were obtained from a local monitoring station. A gas filter correlation analyzer (Thermo Scientific, Model 48i) was used to observed the CO. SO$_2$ and O$_3$ was measured with the pulsed fluorescence analyzer (Thermo Scientific, Model 43iTLE) and the UV photometric analyzer (Thermo Scientific, Model 49i), respectively. NO and NO$_2$ were determined with a chemiluminescence instrument (Thermo Scientific, Model 42iTL). Meteorological parameters of temperature (T) and relative humidity (RH) were measured with a portable weather station (WXT520, Vaisala, Finland). The concentration of gas-phase OH radical was approximated from a nonlinear Pad$^*$ function, and the NO$_x$ effects were considered.

Results from our previous work (Jiang et al., 2021b): Seven-days backward trajectories were generated using the Hybrid Single Particle Lagrangian Integrated Trajectory (HYSPLIT) model. Trajectories were calculated for air masses starting from the sampling site at 500 m above ground level with 6-h intervals during the 24-h sampling period. All trajectories were classified into four clusters, including marine-origin air masses (summer monsoon period) from the Western Pacific and South East Asia regions, and continental-origin air masses (winter monsoon period) from Mongolia and Central Asia.

From the $^{14}$C-based positive matrix factorization (PMF) analysis, we obtained 5 sources that contributed to the DOM: biomass burning (18%), fossil fuels combustion (32%), secondary inorganic nitrogen chemistry processes (20%), SOA formation associated with photochemical processes and waste combustion (7%), and SOA formation associated with isoprene-derived SOA and organic sulfates (22%). Fossil fuels combustion showed the highest average contribution to DOM but small changes in concentration across the year.
Biomass burning explained 18% of the DOM and showed a marked increasing trend from fall to winter. SOA factors were responsible for 50% of DOM mass, most of which was contributed by the factors that associated with secondary inorganic nitrogen chemistry processes, and isoprene-derived SOA and organic sulfates formations. DOM formed from secondary inorganic nitrogen chemistry processes showed higher concentrations in fall and winter, while DOM formed from secondary processes of isoprene and organic sulfates formations had lower concentrations in winter than in summer.

**Measurements for particulate total sulfur and water-soluble sulfate**

About 1–3 pieces of filters were cut using the steel punchers (1.5 cm²) and then put it into clean tin boats directly. The sample were then crashed into a ball and further analyzed using elemental analyzer (Germany, elementar unicube) coupled with high sensitivity thermal conductivity detector in the CNS mode. The particle sulfur in PM₂.₅ samples were calculated according to the calibration curve which were obtained by analyzing standard samples with different mass. The water-soluble sulfate or SO₄²⁻ was analyzed with ion-chromatography (761 Compact IC, Metrohm, Switzerland). A piece of filter (d=24 mm) was punched for each of collected field filter and dissolved into 12 mL distilled deionized water (≥18.2 Ω). Each sample was sonicated for 30 minutes allowing the solution reaching equilibrium. Then the filtrate was filtered through 0.22 μm PTFE membrane (Jinteng, China) and stored in a prewashed clean bottle at 4 °C until sample analysis. Detailed information about the analysis procedures were described in our previous studies (Jiang et al., 2020; Jiang et al., 2021b). Anions were separated on a Metrohm Metrosep A sup5-250 column with 3.2 mM Na₂CO₃ and 1.0 mM NaHCO₃ as the eluent and 35 mM H₂SO₄ for a suppressor. The injection loop volume for anion was 100 μL. The water-soluble sulfate-sulfur was calculated as 1/3 of the SO₄²⁻ concentration. The organic sulfur (Org-S) is calculated as the amount of sulfate-sulfur (SO₄²⁻-S) subtracted from TS, and the ratio of organic sulfur to TS (Org-S/TS) can be calculated as:

\[
\text{Org-S} = \text{TS} - \text{sulfate-sulfur} \quad (S1)
\]

\[
\text{Org-S/TS} = (\text{TS} - \text{sulfate-sulfur})/\text{TS} \quad (S2)
\]

And the uncertainty of organosulfur fraction of total sulfur (δ OrgS/TS) for filter samples can be calculated using the following equation:

\[
\delta_{\text{OrgS/TS}} = (\text{RSD}_{\text{TS}}^2 + \text{RSD}_{\text{sulfate-sulfur}}^2)^{1/2} \times \text{sulfate-sulfur/TS} \quad (S3)
\]

where RSDₜₜ and RSD_sulfate-sulfur are the relative standard deviations determined for SO₄²⁻ and TS, respectively, both were 0.05 μ g m⁻³ in this study.

**Operating conditions for FT-ICR MS analysis**

The ultrahigh-resolution FT-ICR-MS enables identification of complex atmospheric mixtures by giving accurate m/z value, and each peak was assigned to an ambiguous formula with <1ppm absolute mass
error was achieved (Jiang et al., 2021a). Previous study has indicated that the OSs are readily ionized in the negative ESI mode, and most of them were observed only in the negative mode (Lin et al., 2012b; Kuang et al., 2016). Therefore, the negative ESI FT-ICR-MS analysis could provide a comprehensive understanding about the chemical composition of organosulfur compounds (OSCs) in atmosphere, though the molecular structures such as potential isomers were generally hidden behind a given m/z value.

A total of 55 PM$_{2.5}$ samples were used for negative ESI-FT-ICR MS analysis and each sample were ultrasonic extracted with methanol in cold water bath (Jiang et al., 2021a). Though we did not calculate the extraction efficiency of OSs with methanol in a cold-water bath, many previous studies have suggested that methanol could extracted more than 90% of OC both for filed samples or fresh biomass burning samples(Chen and Bond, 2010; Cheng et al., 2017; Huang et al., 2018). Considering OSs are polar compounds, and most of OSs can be dissolved in methanol(Ye et al., 2020). The potential artifacts resulted from extraction with methanol were not tested in this study. However, in a previous study, methanol was used as eluent to collected the humic-like substance for OSs characterization. Direct using methanol as extraction solvent to extract OSs was reported by Ye et al. (2020). All these studies have successfully characterized the OSs and made comparisons between ambient samples collected at different location. Therefore, we think that there might be small or no potential artifacts resulted from extraction with methanol. The methanol extracts were filtered with PTFE members and concentrated, and direct injected into a 9.4T solariX XR FT-ICR mass spectrometer (Bruker Daltonik GmbH, Bremen, Germany) in negative ESI modes at a flow rate of 180 μL h$^{-1}$ (Jiang et al., 2021a; Jiang et al., 2020). Detailed operating conditions are set as: capillary voltage and capillary column end voltage for the negative ESI-FT-ICR MS analysis were set to 4.5 kV and −500 V , ions were accumulated in a hexapole for 0.65s, and the conditions of Octupole were set as 5 MHz and 350 V of peak to-peak (Vp-p) radio frequency (RF) amplitude. An argon-filled hexapole collision pool was operated at 2 MHz and RF amplitude of 1400 Vp-p, in which ions were accumulated for 0.02 s. The optimized mass for quadrupole (Q1) was 170 Da with the time of flight is 0.65ms. The mass range was set as150–800 Da, and a total of 128 continuous 4M data FT-ICR transients were co-added to enhance the signal-to-noise ratio and dynamic range. Field blank filters were processed and analyzed following the same procedure to detect possible contamination. All mass spectra were calibrated externally with arginine clusters in negative ion mode using a linear calibration. The final spectrum was internally recalibrated with typical O$_2$ class species peaks using quadratic calibration in DataAnalysis 5.0 (Bruker Daltonics). A typical mass-resolving power (m/Δm50 %, in which Δm50% is the magnitude of the mass spectral peak full width at half-maximum peak height) >450 000 at m/z 319 with <0.3 ppm absolute mass error was achieved. In this study, three duplicate representative aerosol samples were analyzed at the beginning, middle, and end of the analysis to test the reproducibility of sample extraction, the peak detection of the method, and the molecular formula assignment procedures. Pearson’s correlation analysis of the relative intensities of all molecules between duplicates confirmed the high level of reproducibility of the selected samples (r = 0.98) (Jiang et al., 2021a).

**FT-ICR MS data processing**
A custom software was used to calculate all mathematically possible formulas for all ions with a signal-to-noise ratio above 4 using a mass tolerance of ±1ppm. The compounds assigned as C\(_s\)H\(_b\)O\(_o\)N\(_n\)S\(_s\) with \(s = 1, 2\) will be collectively referred to as organosulfur compounds (OSs) including CHOS \((n = 0)\) and CHONS \((n = 1, 2)\). The identified formulas containing isotopomers (i.e., \(^{13}\)C, \(^{18}\)O or \(^{34}\)S) was not discussed.

The intensity-weighted elemental ratios such as O/C, H/C, O/S were calculated as described in previous study (Jiang et al., 2021a). The double bond equivalent (DBE) is calculated using the equation:

\[
\text{DBE} = \frac{(2c+2-h+n)}{2} \quad (S4)
\]

Additionally, the modified index of aromaticity equivalent \((X_c)\) which was considered as a better index to describe potential monocyclic and polycyclic aromatic compounds with S atoms, were also calculated using the flowing equation (Ye et al., 2020; Yassine et al., 2014):

\[
X_c = \frac{3[D\text{BE}-(m\times o+n\times s)]^2}{D\text{BE}-(m\times o+n\times s)} \quad (S5)
\]

Where \(m\) and \(n\) correspond to the fraction of oxygen and sulfur involved in the \(\pi\)-bond structure of the compound, respectively. If \(\text{DBE} \leq (m\times o+n\times s)\), then \(X_c=0\) is assumed. For chemical classes including alchohol, ether, sulfide, disulfide, sulfinic and sulfonic acids, \(m=n=0\) should be used. And for chemical classes including carboxylic acid, ester and nitro, \(m=0.5\) was adopted. Assuming the sulfur atom of organosulfur molecule exists in a sulfate group \((R-\text{OSO}_3\text{H})\) or a sulfonate group \((R-\text{SO}_3\text{H})\), the organosulfur molecule can be converted into a virtual organic carbon molecule by replacing \(-\text{OSO}_3\text{H}\) with \(-\text{OH}\) (or \(-\text{SO}_3\text{H}\) with \(-\text{H}\)). Considering negative ESI-FT-ICR MS analysis was performed, and the negative ESI mode is sensitive to compounds containing carboxylate, sulfonate and nitro groups. Thus, the calculation for \(X_c\) of organosulfur compounds can be simplified as (Ye et al., 2020):

\[
X_c = \frac{3[D\text{BE}-0.5\times(o-4)]^2}{D\text{BE}-0.5\times(o-4)} \quad (S6)
\]

We rounded \(0.5\times(o-4)\) down to the next lower integer if \(o\) is an odd number. A value of \(X_c\geq2.5000\) was supposed as the unambiguous minimum criterion for the presence of an aromatic structure. \(X_c\geq2.7143, 2.8000, 2.8333, 2.9231\) were considered as the thresholds for molecules containing cores of naphthalene, anthracene, pyrene and ovalene, respectively.
Figure S1. (a) Formular number percentages of each subgroup which divided based on the DBE value and the length of carbon skeleton in the CHOS formulas; (b) and (c) Intensity percentages and formular number percentages of each subgroup which divided based on the Xc value of formulas.
Figure S2. Molecular distribution of CHONS compounds detected by FT-ICR MS for the sample set collected in Guangzhou. (a) Double bond equivalent (DBE) vs C number for all the CHONS compounds of all samples. The color bar and marker size denote the number of oxidation state and the average sum-normalized relative peak intensities of the compounds; (b) Classification of CHONS species into different subgroups according to the numbers of S and O atoms in their molecules; (c) and (d) Intensity percentages and formular number percentages of each subgroup which divided based on the DBE value and the length
of carbon skeleton in the formulas; (e) and (f) Intensity percentages and formular number percentages of each subgroup which divided based on the $X_c$ value of formulas.

Figure S3. Significant correlations between (a) the sum-normalized intensity of OSs form potential unsaturated fatty acid compounds (UFAC) and RH, and the sum-normalized intensity of OSs classified into the subgroup B2 (with $\text{DBE} \leq 2$, $C > 8$, $3 < O < 7$ for CHOS and $\text{DBE} \leq 2$, $N = 1$, $C > 8$, $6 < O < 10$ for CHONS compounds) and (b) UFAC, (c) RH, the concentrations of (d) sterane and hopanes, (e) $\text{Cl}^-$. 
Figure S4. Significant correlations between the concentration of Org-S and (a) SO$_2$, (b) NO$_2$, (c) NO$_x$, (d) NO$_x$+O$_3$, (e) NO$_3$−/SIA, (f) SO$_4^{2−}$/SIA.
Table S1. Summary of the concentration of organosulfur (Org-S) and fraction in total particulate sulfur (TS), organic carbon (OC), organic matter (OM), and PM$_{2.5}$ mass reported in recent studies (OS denotes organosulfates).

| Sites                      | Org-S ($\mu$g/m$^3$) | Org-S/TS | Org-S /OC | OrgSs /OM | Org-S /PM | Ref.               |
|----------------------------|-----------------------|-----------|-----------|-----------|-----------|--------------------|
| Guangzhou                  | 0.04–1.1 (0.6)        | 0.07–50%  (33%) | 11–89% (42%) | 0–3% (1.4%) |          | This study         |
| Maldives                   | 0.3 (OS)              | 2.1%      | 4.4%      | 0.9%(OS)  |          | (Stone et al., 2012) |
| Gosan                      | 0.1 (OS)              | 1.1%      | 3.5%      | 0.6% (OS) |          |                    |
| Singapore                  | 0.3 (OS)              | 2.5%      |           | 1.4% (OS) |          |                    |
| Lahore                     | 0.9–2 (OS)            | 5.9–7.7%  | 0.4–0.8%  | 0.7–0.9%  (OS) |          |                    |
| Four Asian sites           |                       |           |           |           |           |                    |
| Continental aerosol        |                       |           |           |           |           | 4% (OS) (Hawkins et al., 2010) |
| Whistler, British Columbia |                       |           |           |           |           | < 1%(OS) (Schwartz et al., 2010) |
| Polar region               |                       |           | 6%        | 9–11%(OS) |          | (Frossard et al., 2011) |
| Kpuszta, Hungary           | 0.02–0.09             | 6–12%     | 8–50% (OS) |          |          | (Luk’Acs et al., 2009) |
|                           | 0.33                  | 20%       | 30% (OS)  |          |          | (Surratt et al., 2008) |
| Fairbanks, Alaska          |                       | 1.3%      |           | 0.8%      |          | (Shakya and Peltier, 2013) |
| Eight sites in U.S.        | up to 0.07            | 10–13%    |           | 1–3%      |          | (Shakya and Peltier, 2015) |
| 12 sites in U.S.           | 0.1–1.4               | 1–20% (OS) | 5–10% (OS) |          |          | (Tolocka and Turpin, 2012) |
| Mt Kleiner Feldberg in central Germany |            | 40%       |           |          |          | (Vogel et al., 2016) |
| 21 sites in U.S.           | <0.0376 to 0.3        |           |           |          |          | (Dombek et al., 2020) |
| U.S. (eastern and western, composite) | 0.3±0.2 to 0.5±0.2 | 16±3 to 17±5 |           |          |          | (Chen et al., 2021) |
Table S2. Summary of the calculated molecular characteristics of organosulfur compounds groups detected in the yearlong sample set.

| Group | Subgroup | Number of formulas set | % of total OrgSs \(^a\) | % of total OrgSs with \(o/(4s+3n) \geq 1\) | Number of formulas with \(o/(4s+3n) \geq 1\) | For sample | For OrgSs formulas set \(^b\) |
|-------|----------|------------------------|-------------------------|---------------------------------------|----------------------------------------|------------|-------------------------|
|       |          |                        |                         | MW                                   | H/C                                   | O/C        | O/S        | DBE        | Number of formulas with \(o/(4s+3n) \geq 1\) | % of formulas |                        |                         |                        |
| CHOS  | CHOS\(_1\) | 406-2199               | 57(50-67)               | 70(56-80)                            | 389-2143                              | 349(305-378) | 1.78(1.72-1.84) | 0.52(0.40-0.67) | 6.7(5.8-7.7) | 2.64(2.22-2.90) | 5664 | 5256(93%) |
|       | CHOS\(_2\) | 82-291                 | 64(4-12)                | 2(1-6)                               | 35-149                                | 46(31-63)   | 583(519-649) | 1.50(1.30-1.66) | 0.33(0.21-0.50) | 3.8(3-4.3) | 7.80(5.78-9.38) | 3722 | 2017(54%) |
| Total |          | 498-2383               | 64(58-73)               | 72(59-84)                            | 432-2262                              | 355(315-389) | 1.77(1.72-1.83) | 0.52(0.40-0.68) | 6.7(5.7-7.7) | 2.77(2.39-3.50) | 9386 | 7273(77%) |
| CHON\(_1\)S | 190-1344 | 31(22-35)              | 26(15-37)               | 159-1177                             | 83(75-89)                             | 366(325-399) | 1.72(1.65-1.77) | 0.71(0.63-0.84) | 8.4(7.5-9.5) | 3.46(3.10-4.45) | 4397 | 3253(74%) |
| CHON\(_2\)S | 40-247   | 5(2-10)                | 2(1-6)                  | 25-227                               | 78(48-94)                             | 455(390-553) | 1.69(1.42-1.80) | 0.90(0.61-1.35) | 11.0(9.7-11.9) | 4.85(3.49-8.06) | 2215 | 1357(61%) |
| Total |          | 269-1591               | 36(27-42)               | 28(16-41)                            | 202-1389                              | 373(331-405) | 1.72(1.62-1.76) | 0.72(0.63-0.85) | 8.6(7.7-9.7) | 3.56(3.15-4.89) | 6612 | 4610(70%) |

\(^a\) OrgSs: Organosulfur Compounds

\(^b\) OrgSs formulas set denotes the all organosulfur compounds detected in all samples.
Table S3. Comparison of O/C and H/C ratios of CHOS compounds in this study and other studies.

| Sample/type | Site/type | Extraction solution | O/C     | H/C     | Instrument | Ref.                      |
|-------------|-----------|---------------------|---------|---------|------------|---------------------------|
| PM$_{2.5}$ | CHOS      | Methanol            | 0.52±0.0  | 1.77±0.0  | FT-ICR MS | This study                |
| Rainwater   | Northeaster n United States | Water | 1.3±0.8 | 1.9±0.5 | FT-ICR MS | (Altieri et al., 2009)    |
| PM$_{2.5}$ | Pearl River Delta | Water | 0.55± 0.17 | 1.67±0.3 | Orbitrap MS | (Lin et al., 2012a)       |
| PM$_{2.5}$ | Cambridge | winter/spring Water and acetonitrile | 0.47   | 1.47   | Orbitrap MS | (Rincón et al., 2012)    |
| Cloud | Colorado | Water | 0.43±0.0  | 1.41±0.2  | FT-ICR MS | (Zhao et al., 2013)       |
| PM (0.18-1.8 μm) | California | after midnight/Water/morning | 0.87±0.0 | 1.7±0.05 | Orbitrap MS | (O'brien et al., 2014)   |
| TSP | Virginia | Water | 0.47±0.2  | 1.46±0.3  | FT-ICR MS | (Willoughby et al., 2014) |
| PM$_{2.5}$ | Beijing | Hazy/DCM/Clear/Water | 0.49±0.2 | 1.55±0.4 | FT-ICR MS | (Jiang et al., 2016)      |
| Wuhan | Winter | Methanol | 0.37±0.2 | 1.68±0.4 | Orbitrap MS | (Wang et al., 2016)       |
| Nanjing | Summer | Methanol | 0.43±0.3 | 1.68±0.4 | Orbitrap MS | (Wang et al., 2016)       |
| Shanghai | Winter | Acetonitrile | 0.40±0.2 | 1.68±0.4 | Orbitrap MS | (Wang et al., 2016)       |
| Shanghai | Spring/Fall/Winter | Acetonitrile | 0.2 | 1.1 | Orbitrap MS | (Wang et al., 2017)       |
| PM$_{2.5}$ | Mainz | low-pollution Water | 0.78 | 1.66 | Orbitrap MS | (Wang et al., 2018)       |
| City          | Low-pollution | High-pollution | Water | FT-ICR MS | FT-ICR MS |
|--------------|---------------|----------------|-------|-----------|-----------|
| Beijing      | 0.63          | 0.51           | 0.3   | 1.81      | 1.74      |
| Cloud        |               |                |       |           |           |
| France       |               |                |       |           |           |
| Water        | 0.3           | 1.52           |       |           |           |
| PM$_{2.5}$   |               |                |       |           |           |
| Changchun    | 1.17±0.1      | 1.56±0.1       | 3     | 1         |           |
| Shanghai     | 1.41±0.1      | 1.85±0.0       | 9     | 4         |           |
| Guangzhou    | 1.48±0.0      | 1.85±0.0       | 5     | 2         |           |

(Wang et al., 2021)
| Sample/time | Site/type               | Extraction solution | O/C      | H/C      | Instrument | Ref.                  |
|-------------|-------------------------|---------------------|----------|----------|------------|-----------------------|
| PM$_{2.5}$  | CHONS/Guangzhou         | Methanol            | 0.72±0.0 | 1.72±0.0 | FT-ICR MS  | This study            |
| rainwater   | North eastern United States | Water               | 1.7 ± 0.9 | 1.8 ± 0.6 | FT-ICR MS  | (Altieri et al., 2009) |
| PM$_{2.5}$  | Pearl River Delta       | Water               | 0.81 ± 0.22 | 1.73 ± 0.29 | Orbitrap MS | (Lin et al., 2012a)   |
| PM$_{2.5}$  | Cambridge summer        | Water and acetonitrile | 0.73  | 1.99  | Orbitrap MS | (Rincón et al., 2012) |
| PM$_{2.5}$  | Cambridge winter        | Water               | 0.44±0.0 | 1.17±0.1 | FT-ICR MS  | (Zhao et al., 2013)   |
| Cloud       | Colorado after midnight | Water               | 0.99±0.0 | 1.7±0.0  | Orbitrap MS | (O'Brien et al., 2014) |
| PM (0.18-1.8 μm) | California morning    | Water               | 1.0±0.00 | 1.7±0.0  | Orbitrap MS | (Willoughby et al., 2014) |
| TSP         | Virginia                | Water               | 0.71±0.2 | 1.65±0.2 | FT-ICR MS  | (Willoughby et al., 2014) |
| PM$_{2.5}$  | Beijing Hazy            | DCM                 | 0.69±0.3 | 1.57±0.3 | FT-ICR MS  | (Jiang et al., 2016)  |
| PM$_{2.5}$  | Beijing Clear           | Water               | 0.70±0.3 | 1.51±0.3 | FT-ICR MS  | (Jiang et al., 2016)  |
| PM$_{2.5}$  | Wuhan Winter            | Water               | 0.35±0.1 | 1.58±0.4 | Orbitrap MS | (Wang et al., 2016)   |
| PM$_{2.5}$  | Nanjing Summer          | Methanol            | 0.44±0.2 | 1.69±0.3 | Orbitrap MS | (Wang et al., 2016)   |
| Shanghai    | Winter                  | Methanol            | 0.42±0.2 | 1.64±0.5 | Orbitrap MS | (Wang et al., 2017)   |
| Shanghai    | Summer                  | Acetonitrile        | 0.4     | 1.5     | Orbitrap MS | (Wang et al., 2017)   |
| PM$_{2.5}$  | Spring                  | Acetonitrile        | 0.2     | 1.5     | Orbitrap MS | (Wang et al., 2017)   |
| PM$_{2.5}$  | Fall                    | Acetonitrile        | 0.3     | 1.5     | Orbitrap MS | (Wang et al., 2017)   |
| PM$_{2.5}$  | Winter                  | Acetonitrile        | 0.4     | 1.5     | Orbitrap MS | (Wang et al., 2017)   |
| PM$_{2.5}$  | Mainz low               | Acetonitrile        | 0.91    | 1.54    | Orbitrap MS | (Wang et al., 2017)   |
| PM$_{2.5}$  | Mainz high              | Acetonitrile        | 0.91    | 1.54    | Orbitrap MS | (Wang et al., 2017)   |

Table S4. Comparison of O/C and H/C ratios of CHONS compounds in this study and other studies.
| City          | Pollution Level | Water Type | Method      | Al.                               |
|--------------|----------------|------------|-------------|-----------------------------------|
| Beijing      | Low-pollution   | e-water    | FT-ICR MS   | Bianco et al., 2018               |
|              |                 |            |             |                                   |
|              | High-pollution  | e-water    | FT-ICR MS   | Bianco et al., 2018               |
|              |                 |            |             |                                   |
| Cloud        |                 |            | FT-ICR MS   | Bianco et al., 2018               |
| France       |                 |            |             |                                   |
| PM$_{2.5}$   | Changchun       | Acetonitrile | Orbitrap MS | Wang et al., 2021                |
|              |                 | e-water    |             |                                   |
|              | Shanghai        |            |             |                                   |
|              | Guangzhou       |            |             |                                   |
Table S5. Summary of the calculated molecular characteristics of organosulfur compounds groups detected in source samples, as the FT-ICR MS data are obtained from Cui et al. (2019) and Tang et al. (2020)

| Source Samples                  | Formula number | MW     | H/C   | O/C   | O/S   | DBE   | % of (DBE-N) ≥ 4 | % of Xc ≥ 2.5 | % of ω/(4s+3n) ≥ 1 |
|---------------------------------|----------------|--------|-------|-------|-------|-------|-----------------|--------------|-------------------|
| **Excavator**                   |                |        |       |       |       |       |                 |              |                   |
| BBOA1 (Musa)                    | CHOS           | 444    | 360   | 1.52  | 0.47  | 6.21  | 4.76            | 57           | 43                | 88                |
|                                 | CHONS          | 371    | 379   | 1.55  | 0.50  | 7.21  | 4.98            | 58           | 64                | 64                |
|                                 | Avg/total      | 815    | 367   | 1.53  | 0.48  | 6.59  | 4.85            | 57           | 53                | 77                |
| **BBOA2 (Hevea)**               | CHOS           | 174    | 396   | 1.35  | 0.40  | 5.97  | 7.68            | 69           | 59                | 86                |
|                                 | CHONS          | 65     | 411   | 1.56  | 0.50  | 7.51  | 4.79            | 62           | 69                | 63                |
|                                 | Avg/total      | 239    | 400   | 1.40  | 0.42  | 6.34  | 6.98            | 67           | 62                | 80                |
| **COCOA1 (Anthracite)**         | CHOS           | 549    | 323   | 1.01  | 0.40  | 5.40  | 8.55            | 85           | 82                | 95                |
|                                 | CHONS          | 767    | 340   | 0.98  | 0.52  | 6.49  | 8.99            | 94           | 97                | 47                |
|                                 | Avg/total      | 1316   | 332   | 0.99  | 0.47  | 6.03  | 8.80            | 90           | 91                | 67                |
| **COCOA2 (Bituminous coal)**    | CHOS           | 463    | 340   | 0.99  | 0.31  | 4.64  | 9.90            | 96           | 94                | 85                |
|                                 | CHONS          | 293    | 308   | 0.97  | 0.49  | 5.82  | 8.04            | 92           | 93                | 29                |
|                                 | Avg/total      | 756    | 328   | 0.98  | 0.38  | 5.10  | 9.18            | 94           | 93                | 63                |
| **Vehicle emissions**           | CHOS           | 112    | 441   | 1.31  | 0.25  | 4.47  | 9.54            | 71           | 71                | 75                |
|                                 | CHONS          | 17     | 400   | 1.17  | 0.72  | 8.59  | 6.92            | 59           | 59                | 47                |
|                                 | Avg/total      | 129    | 432   | 1.28  | 0.35  | 5.36  | 8.97            | 69           | 69                | 71                |
| **Tunnel aerosols**             | CHOS           | 635    | 325   | 1.74  | 0.59  | 6.79  | 2.75            | 46           | 23                | 96                |
|                                 | CHONS          | 410    | 340   | 1.81  | 0.90  | 8.73  | 2.78            | 28           | 29                | 91                |
|                                 | Avg/total      | 1045   | 331   | 1.76  | 0.71  | 7.53  | 2.76            | 39           | 25                | 94                |
| **Excavator-idling (diesel)**   | CHOS           | 1004   | 353   | 1.61  | 0.38  | 5.81  | 4.18            | 68           | 58                | 96                |
|                                 | CHONS          | 310    | 325   | 1.47  | 0.41  | 5.59  | 5.18            | 56           | 65                | 42                |
|                                 | Avg/total      | 1314   | 347   | 1.59  | 0.38  | 5.77  | 4.38            | 65           | 60                | 83                |
| **Excavator-moving (diesel)**   | CHOS           | 334    | 326   | 1.51  | 0.46  | 5.20  | 3.58            | 54           | 49                | 98                |
|                                 | CHONS          | 117    | 298   | 1.62  | 0.48  | 5.17  | 5.55            | 59           | 64                | 9                 |
|                                 | Avg/total      | 451    | 314   | 1.35  | 0.42  | 5.19  | 4.38            | 56           | 53                | 75                |
| **Excavator-working (diesel)**  | CHOS           | 631    | 342   | 1.63  | 0.36  | 5.44  | 4.00            | 62           | 55                | 93                |
|                                 | CHONS          | 260    | 323   | 1.47  | 0.40  | 5.41  | 5.26            | 62           | 69                | 27                |
|                                 | Avg/total      | 891    | 337   | 1.58  | 0.37  | 5.19  | 4.35            | 62           | 59                | 74                |
| **Diesel-vessels**              | CHOS           | 334    | 306   | 1.66  | 0.40  | 5.14  | 3.47            | 55           | 50                | 95                |
|                                 | CHONS          | 13     | 461   | 1.50  | 0.36  | 6.74  | 9.38            | 38           | 38                | 46                |
|                                 | Avg/total      | 347    | 310   | 1.66  | 0.40  | 5.17  | 3.60            | 54           | 49                | 93                |
| **Heavy-fuel-oil-vessels**      | CHOS           | 1110   | 311   | 1.48  | 0.36  | 4.77  | 4.85            | 76           | 71                | 83                |
|                                 | CHONS          | 398    | 343   | 1.35  | 0.39  | 5.68  | 6.35            | 80           | 86                | 28                |
|                                 | Avg/total      | 1508   | 314   | 1.47  | 0.36  | 4.86  | 5.00            | 77           | 75                | 68                |
Table S6. Detailed intensity percentages of isoprene-derived OSs detected at Guangzhou. Noted the formulas in the Table S6-S10 were from the summarization of recent studies and the reference in (Bruggemann et al., 2020; Ye et al., 2020; Zhu et al., 2019; Wang et al., 2019).

| Formula [M-H] | MW (Da) | DBE | Average RI (%) |
|---------------|---------|-----|----------------|
| C$_{18}$H$_{10}$O$_5$S | 346.0086 | 3 | 0.039 |
| C$_{18}$H$_{10}$O$_5$S$^-$ | 331.0704 | 1 | 0.028 |
| C$_{18}$H$_{10}$O$_5$S$^-$ | 333.0861 | 0 | 0.070 |
| C$_{18}$H$_{10}$O$_5$S$^-$ | 451.1491 | 0 | 0.035 |
| C$_{18}$H$_{10}$NO$_5$S$^-$ | 244.0133 | 1 | 0.172 |
| C$_{18}$H$_{10}$NO$_5$S$^-$ | 260.0082 | 1 | 0.230 |
| C$_{18}$H$_{10}$NO$_5$S$^-$ | 273.9874 | 2 | 0.099 |
| C$_{18}$H$_{10}$NO$_5$S$^-$ | 304.9933 | 2 | 0.108 |
| C$_{18}$H$_{10}$NO$_5$S$^-$ | 346.0086 | 3 | 0.039 |
Table S7. Detailed intensity percentages of terpene-derived OSs (including limonene) detected at Guangzhou.

| Formula [M-H] | MW (Da)  | DBE | Average RI (%) |
|---------------|----------|-----|----------------|
| C_{10}H_{11}O_{2}S | 179.0384 | 1   | 0.055          |
| C_{11}H_{11}O_{2}S | 199.0282 | 0   | 0.166          |
| C_{11}H_{12}O_{2}S | 200.9711 | 1   | 0.167          |
| C_{11}H_{11}O_{5}S | 211.0282 | 1   | 0.348          |
| C_{12}H_{11}O_{2}S | 215.0231 | 0   | 0.431          |
| C_{12}H_{12}O_{2}S | 219.0697 | 2   | 0.169          |
| C_{12}H_{13}O_{2}S | 221.0853 | 1   | 0.189          |
| C_{13}H_{11}O_{5}S | 223.0282 | 2   | 0.291          |
| C_{13}H_{12}O_{5}S | 223.1010 | 0   | 0.391          |
| C_{13}H_{13}O_{5}S | 225.0438 | 1   | 0.462          |
| C_{13}H_{14}O_{5}S | 226.9867 | 2   | 0.503          |
| C_{13}H_{15}O_{5}S | 229.0024 | 1   | 0.469          |
| C_{14}H_{12}O_{3}S | 229.0176 | 5   | 0.471          |
| C_{14}H_{13}O_{3}S | 229.0540 | 4   | 0.478          |
| C_{14}H_{15}O_{3}S | 231.0697 | 3   | 0.453          |
| C_{15}H_{15}O_{5}S | 235.0646 | 2   | 0.252          |
| C_{15}H_{13}O_{3}S | 237.0438 | 2   | 0.403          |
| C_{14}H_{15}O_{3}S | 237.1166 | 0   | 0.478          |
| C_{14}H_{15}O_{3}S | 241.0176 | 6   | 0.630          |
| C_{15}H_{11}O_{3}S | 241.0751 | 0   | 0.669          |
| C_{15}H_{11}O_{3}S | 243.0180 | 1   | 0.656          |
| C_{16}H_{15}O_{3}S | 245.0125 | 5   | 0.279          |
| C_{15}H_{12}O_{3}S | 247.0646 | 3   | 0.129          |
| C_{15}H_{14}O_{3}S | 249.0438 | 3   | 0.140          |
| C_{15}H_{12}O_{3}S | 249.0802 | 2   | 0.217          |
| C_{16}H_{13}O_{3}S | 250.9867 | 4   | 0.236          |
| C_{16}H_{15}O_{3}S | 251.0231 | 3   | 0.326          |
| C_{16}H_{15}O_{3}S | 251.0595 | 2   | 0.507          |
| C_{16}H_{15}O_{3}S | 251.0959 | 1   | 0.771          |
| C_{16}H_{16}O_{5}S | 253.0024 | 3   | 0.793          |
| C_{16}H_{15}O_{5}S | 253.0387 | 2   | 0.912          |
| C_{17}H_{15}O_{5}S | 253.0751 | 1   | 1.038          |
| C_{16}H_{15}O_{3}S | 253.1115 | 0   | 1.056          |
| C_{17}H_{14}O_{5}S | 258.9918 | 6   | 0.416          |
| C_{16}H_{16}O_{5}S | 259.0282 | 5   | 0.290          |
| C_{16}H_{15}O_{3}S | 261.0438 | 4   | 0.062          |
| C_{17}H_{15}O_{3}S | 263.0231 | 4   | 0.080          |
| C_{17}H_{15}O_{5}S | 263.0595 | 3   | 0.153          |
| C_{18}H_{15}O_{5}S | 265.0024 | 4   | 0.189          |
| C_{18}H_{15}O_{5}S | 265.0387 | 3   | 0.352          |
| C_{18}H_{15}O_{5}S | 265.0751 | 2   | 0.480          |
| C_{18}H_{15}O_{5}S | 267.0180 | 3   | 0.613          |
| C_{19}H_{15}O_{5}S | 267.0544 | 2   | 0.799          |
| C_{19}H_{15}O_{5}S | 267.0908 | 1   | 0.910          |
| C_{20}H_{17}O_{7}S | 269.0700 | 1   | 0.899          |
| C_{20}H_{17}O_{7}S | 271.0129 | 2   | 0.751          |
| C_{20}H_{17}O_{7}S | 273.0074 | 6   | 0.313          |
| C_{21}H_{17}O_{7}S | 273.0650 | 0   | 0.186          |
| C_{20}H_{15}O_{5}S | 279.0544 | 3   | 0.443          |
| C_{21}H_{13}O_{5}S | 281.0337 | 3   | 0.768          |
Table S8. Detailed intensity percentages of other biogenic VOCs-derived OSs (2-Methyl-3-Buten-2-ol; 2-E-pentenal, 2-E-hexenal, 3-Z-hexenal, and cis-3-hexen-1-ol, β-caryophyllene) detected at Guangzhou.

| Formula [-M-H] | MW (Da) | DBE | Average RI (%) |
|----------------|---------|-----|----------------|
| C_{10}H_{15}O_{10}S^- | 281.0700 | 2 | 0.986 |
| C_{12}H_{17}O_{10}S^- | 283.0282 | 7 | 1.001 |
| C_{9}H_{15}O_{6}S^- | 283.0493 | 2 | 1.067 |
| C_{10}H_{16}O_{6}S^- | 283.0857 | 1 | 1.150 |
| C_{9}H_{15}O_{6}S^- | 285.0286 | 2 | 0.826 |
| C_{11}H_{15}O_{6}S^- | 291.0544 | 4 | 0.089 |
| C_{9}H_{15}O_{6}S^- | 295.0129 | 4 | 0.475 |
| C_{10}H_{16}O_{6}S^- | 295.0493 | 3 | 0.595 |
| C_{9}H_{15}O_{6}S^- | 297.0286 | 3 | 0.737 |
| C_{10}H_{17}O_{6}S^- | 297.0650 | 2 | 0.834 |
| C_{9}H_{15}O_{6}S^- | 299.0442 | 2 | 0.580 |
| C_{14}H_{20}O_{6}S^- | 303.1272 | 3 | 0.137 |
| C_{11}H_{17}O_{6}S^- | 309.0650 | 3 | 0.477 |
| C_{10}H_{16}O_{6}S^- | 311.0442 | 3 | 0.642 |
| C_{10}H_{17}O_{6}S^- | 313.0599 | 2 | 0.478 |
| C_{15}H_{20}O_{6}S^- | 317.1428 | 3 | 0.106 |
| C_{14}H_{20}O_{6}S^- | 319.1221 | 3 | 0.152 |
| C_{10}H_{15}NO_{10}S^- | 327.0391 | 3 | 0.358 |
| C_{14}H_{24}O_{6}S^- | 333.1013 | 4 | 0.129 |
| C_{15}H_{20}O_{6}S^- | 333.1377 | 3 | 0.164 |
| C_{10}H_{15}NO_{10}S^- | 341.0184 | 4 | 0.411 |
| C_{15}H_{20}O_{6}S^- | 347.1170 | 4 | 0.136 |
| C_{14}H_{20}O_{6}S^- | 349.0963 | 4 | 0.206 |
| C_{14}H_{20}O_{6}S^- | 351.1119 | 3 | 0.305 |
| C_{15}H_{20}O_{6}S^- | 363.1119 | 4 | 0.188 |
| C_{16}H_{20}O_{6}S^- | 363.1483 | 3 | 0.235 |
| C_{16}H_{20}O_{6}S^- | 379.1432 | 3 | 0.321 |
| C_{20}H_{15}O_{6}S^- | 383.1898 | 5 | 0.240 |
| C_{20}H_{15}O_{6}S^- | 385.2054 | 4 | 0.074 |
| C_{20}H_{15}O_{6}S^- | 481.1571 | 4 | 0.061 |
| C_{10}H_{16}NO_{10}S^- | 294.0653 | 3 | 1.416 |
| C_{9}H_{15}O_{6}S^- | 296.0446 | 3 | 1.483 |
| C_{10}H_{16}NO_{10}S^- | 310.0602 | 3 | 0.130 |
| C_{9}H_{15}O_{6}S^- | 312.0395 | 3 | 0.178 |
| C_{10}H_{16}NO_{10}S^- | 326.0551 | 3 | 0.164 |
| C_{10}H_{16}NO_{10}S^- | 328.0708 | 2 | 0.274 |
| C_{9}H_{15}NO_{10}S^- | 330.0500 | 2 | 0.295 |
| C_{10}H_{16}NO_{10}S^- | 342.0500 | 3 | 0.212 |
| C_{10}H_{15}NO_{10}S^- | 355.0453 | 4 | 0.153 |
| C_{9}H_{13}NO_{10}S^- | 362.1279 | 4 | 0.097 |
| C_{10}H_{15}NO_{10}S^- | 373.0559 | 3 | 0.201 |
| C_{9}H_{13}NO_{10}S^- | 382.1177 | 3 | 0.131 |
| C_{10}H_{17}NO_{12}S^- | 389.0508 | 3 | 0.066 |
Table S9. Detailed intensity percentages of anthropogenic VOCs-derived OSs detected at Guangzhou.

| Formula [M-H] | MW (Da) | DBE | Average RI (%) |
|---------------|---------|-----|----------------|
| C₆H₁₁O₃S     | 211.0282| 1   | 0.607          |
| C₅H₁₀O₄S     | 213.0074| 1   | 0.630          |
| C₄H₉O₄S      | 229.0024| 1   | 0.387          |
| C₆H₁₅O₆S     | 251.0595| 2   | 0.790          |
| C₄H₁₇O₇S     | 269.0700| 1   | 0.910          |
| C₁₄H₂₃O₈S    | 303.1272| 3   | 0.140          |
| C₁₃H₂₃O₈S    | 317.1428| 3   | 0.110          |
| C₁₄H₂₃O₈S    | 319.1221| 3   | 0.199          |
| C₁₄H₂₃O₈S    | 333.1013| 4   | 0.191          |
| C₁₅H₂₅O₉S    | 333.1377| 3   | 0.201          |
| C₁₅H₂₅O₉S    | 347.1170| 4   | 0.190          |
| C₁₄H₂₅O₉S    | 349.0963| 4   | 0.135          |
| C₁₄H₂₅O₉S    | 351.1119| 3   | 0.336          |
| C₁₅H₂₅O₉S    | 363.1119| 4   | 0.237          |
| C₁₆H₂₇O₉S    | 363.1483| 3   | 0.289          |
| C₁₆H₂₇O₉S    | 379.1432| 3   | 0.419          |
| C₁₃H₂₃NO₃S⁻  | 362.1279| 4   | 0.162          |
| C₁₄H₂₃NO₄S⁻  | 382.1177| 3   | 0.151          |

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unsaturated acids, such as Palmitoleic acid, Linoleic acid, Conjugated linoleic acid, 10-

Table S10. Detailed intensity percentages of OSs derived from precursors of multiple sources detected at Guangzhou, including Methyl Vinyl, Methacrolein, glyoxal, methylglyoxal, Oleic acid, and other unsaturated acids, such as Palmitoleic acid, Linoleic acid, Conjugated linoleic acid, 10-Undecenoic acid, as well as some alkanes such as 1-Dodecene.

| Formula [M-H] | MW (Da) | DBE | Average RI (%) |
|---------------|---------|-----|----------------|
| \( \text{C}_3\text{H}_5\text{O}_3\text{S} \) | 155.0020 | 0 | 0.087 |
| \( \text{C}_4\text{H}_7\text{O}_3\text{S} \) | 164.9863 | 2 | 0.076 |
| \( \text{C}_5\text{H}_9\text{O}_3\text{S} \) | 167.0020 | 1 | 0.588 |
| \( \text{C}_6\text{H}_{10}\text{O}_3\text{S} \) | 168.9812 | 1 | 0.127 |
| \( \text{C}_7\text{H}_{11}\text{O}_3\text{S} \) | 179.0020 | 2 | 0.144 |
| \( \text{C}_8\text{H}_{12}\text{O}_3\text{S} \) | 181.0176 | 1 | 0.719 |
| \( \text{C}_9\text{H}_{13}\text{O}_3\text{S} \) | 182.9969 | 1 | 0.683 |
| \( \text{C}_{10}\text{H}_{14}\text{O}_3\text{S} \) | 194.9969 | 2 | 0.907 |
| \( \text{C}_{11}\text{H}_{15}\text{O}_3\text{S} \) | 195.0333 | 1 | 1.546 |
| \( \text{C}_{12}\text{H}_{16}\text{O}_3\text{S} \) | 197.0125 | 1 | 1.113 |
| \( \text{C}_{13}\text{H}_{17}\text{O}_3\text{S} \) | 198.9554 | 2 | 0.004 |
| \( \text{C}_{14}\text{H}_{18}\text{O}_3\text{S} \) | 200.9711 | 1 | 0.015 |
| \( \text{C}_{15}\text{H}_{19}\text{O}_3\text{S} \) | 206.9969 | 3 | 0.312 |
| \( \text{C}_{16}\text{H}_{20}\text{O}_3\text{S} \) | 207.0333 | 2 | 0.487 |
| \( \text{C}_{17}\text{H}_{21}\text{O}_3\text{S} \) | 207.0697 | 1 | 0.392 |
| \( \text{C}_{18}\text{H}_{22}\text{O}_3\text{S} \) | 209.0125 | 2 | 2.961 |
| \( \text{C}_{19}\text{H}_{23}\text{O}_3\text{S} \) | 209.0489 | 1 | 2.110 |
| \( \text{C}_{20}\text{H}_{24}\text{O}_3\text{S} \) | 209.0853 | 0 | 2.239 |
| \( \text{C}_{21}\text{H}_{25}\text{O}_3\text{S} \) | 210.9918 | 2 | 1.181 |
| \( \text{C}_{22}\text{H}_{26}\text{O}_3\text{S} \) | 211.0282 | 1 | 2.907 |
| \( \text{C}_{23}\text{H}_{27}\text{O}_3\text{S} \) | 211.0646 | 0 | 0.858 |
| \( \text{C}_{24}\text{H}_{28}\text{O}_3\text{S} \) | 213.0074 | 1 | 0.565 |
| \( \text{C}_{25}\text{H}_{29}\text{O}_3\text{S} \) | 214.9867 | 1 | 0.002 |
| \( \text{C}_{26}\text{H}_{30}\text{O}_3\text{S} \) | 216.9660 | 1 | 0.017 |
| \( \text{C}_{27}\text{H}_{31}\text{O}_3\text{S} \) | 221.0489 | 2 | 0.742 |
| \( \text{C}_{28}\text{H}_{32}\text{O}_3\text{S} \) | 221.0853 | 1 | 0.344 |
| \( \text{C}_{29}\text{H}_{33}\text{O}_3\text{S} \) | 223.0646 | 1 | 3.136 |
| \( \text{C}_{30}\text{H}_{34}\text{O}_3\text{S} \) | 223.1010 | 0 | 0.657 |
| \( \text{C}_{31}\text{H}_{35}\text{O}_3\text{S} \) | 229.0024 | 1 | 0.084 |
| \( \text{C}_{32}\text{H}_{36}\text{O}_3\text{S} \) | 230.9816 | 1 | 0.007 |
| \( \text{C}_{33}\text{H}_{37}\text{O}_3\text{S} \) | 235.0646 | 2 | 5.496 |
| \( \text{C}_{34}\text{H}_{38}\text{O}_3\text{S} \) | 235.1010 | 1 | 0.431 |
| \( \text{C}_{35}\text{H}_{39}\text{O}_3\text{S} \) | 237.0074 | 3 | 1.350 |
| \( \text{C}_{36}\text{H}_{40}\text{O}_3\text{S} \) | 237.0438 | 2 | 4.505 |
| \( \text{C}_{37}\text{H}_{41}\text{O}_3\text{S} \) | 237.0802 | 1 | 2.513 |
| \( \text{C}_{38}\text{H}_{42}\text{O}_3\text{S} \) | 239.0595 | 1 | 4.788 |
| \( \text{C}_{39}\text{H}_{43}\text{O}_3\text{S} \) | 244.9973 | 1 | 0.006 |
| \( \text{C}_{40}\text{H}_{44}\text{O}_3\text{S} \) | 249.0802 | 2 | 2.914 |
| \( \text{C}_{41}\text{H}_{45}\text{O}_3\text{S} \) | 249.1166 | 1 | 0.448 |
| \( \text{C}_{42}\text{H}_{46}\text{O}_3\text{S} \) | 251.0595 | 2 | 6.871 |
| \( \text{C}_{43}\text{H}_{47}\text{O}_3\text{S} \) | 251.0959 | 1 | 10.186 |
| \( \text{C}_{44}\text{H}_{48}\text{O}_3\text{S} \) | 253.0751 | 1 | 4.825 |
| \( \text{C}_{45}\text{H}_{49}\text{O}_3\text{S} \) | 255.0544 | 1 | 1.826 |
| \( \text{C}_{46}\text{H}_{50}\text{O}_3\text{S} \) | 255.0908 | 0 | 0.549 |
| \( \text{C}_{47}\text{H}_{51}\text{O}_3\text{S} \) | 265.0751 | 2 | 4.866 |
| \( \text{C}_{48}\text{H}_{52}\text{O}_3\text{S} \) | 265.1115 | 1 | 3.640 |
| Compound     | Mass     | Factor | Value  |
|--------------|----------|--------|--------|
| C₄H₁₀O₈S    | 267.0180 | 3      | 2.195  |
| C₃H₈O₄S     | 267.0544 | 2      | 7.408  |
| C₁₀H₁₅O₈S   | 267.0908 | 1      | 4.505  |
| C₄H₁₀O₄S    | 269.0700 | 1      | 2.203  |
| C₄H₁₂O₆S    | 271.0493 | 1      | 0.394  |
| C₃H₇O₁₄S    | 274.9715 | 2      | 0.006  |
| C₁₃H₁₆O₃S   | 277.1479 | 1      | 0.545  |
| C₁₀H₁₄O₄S   | 279.0544 | 3      | 9.100  |
| C₁₁H₁₀O₆S   | 279.0908 | 2      | 3.420  |
| C₁₂H₂₀O₆S   | 279.1272 | 1      | 4.561  |
| C₁₃H₂₁O₆S   | 281.1064 | 1      | 3.002  |
| C₁₀H₁₆O₄S   | 283.0857 | 1      | 2.828  |
| C₄H₁₅O₈S    | 285.0650 | 1      | 0.564  |
| C₁₂H₁₈O₆S   | 291.0908 | 3      | 1.309  |
| C₁₂H₁₂O₆S   | 293.1064 | 2      | 2.970  |
| C₁₃H₂₅O₆S   | 293.1428 | 1      | 5.245  |
| C₁₀H₁₈O₆S   | 295.0493 | 3      | 4.782  |
| C₁₀H₁₉O₆S   | 297.0650 | 2      | 3.585  |
| C₁₁H₂₁O₄S   | 297.1013 | 1      | 1.343  |
| C₁₀H₁₆O₄S   | 299.0806 | 1      | 1.084  |
| C₄H₁₇O₆S    | 301.0599 | 1      | 0.076  |
| C₁₄H₂₅O₆S   | 303.1272 | 3      | 0.671  |
| C₁₄H₂₅O₆S   | 305.1428 | 2      | 1.476  |
| C₁₃H₂₆O₆S   | 305.1792 | 1      | 0.614  |
| C₁₄H₂₅O₆S   | 307.1585 | 1      | 6.946  |
| C₁₅H₂₁O₆S   | 307.1949 | 0      | 1.458  |
| C₁₃H₂₆O₆S   | 309.1377 | 1      | 2.465  |
| C₁₃H₂₅O₆S   | 317.1428 | 3      | 0.720  |
| C₁₄H₂₆O₆S   | 319.1221 | 3      | 1.328  |
| C₁₅H₂₆O₆S   | 319.1585 | 2      | 1.399  |
| C₁₄H₂₆O₆S   | 321.1377 | 2      | 2.457  |
| C₁₃H₂₆O₆S   | 321.1741 | 1      | 7.015  |
| C₁₄H₂₆O₆S   | 323.1534 | 1      | 2.529  |
| C₁₅H₂₁O₆S   | 323.1898 | 0      | 0.906  |
| C₁₃H₂₆O₆S   | 325.1326 | 1      | 1.016  |
| C₁₄H₂₃O₆S   | 333.1013 | 4      | 1.254  |
| C₁₅H₂₅O₆S   | 333.1377 | 3      | 1.362  |
| C₁₆H₂₅O₆S   | 333.1741 | 2      | 1.408  |
| C₁₅H₂₆O₆S   | 335.1534 | 2      | 2.050  |
| C₁₆H₁₃O₆S   | 335.1898 | 1      | 6.059  |
| C₁₄H₂₅O₆S   | 337.1326 | 2      | 2.532  |
| C₁₅H₂₅O₆S   | 337.1690 | 1      | 2.283  |
| C₁₆H₂₃O₆S   | 337.2054 | 0      | 1.863  |
| C₁₅H₂₅O₆S   | 347.1170 | 4      | 1.842  |
| C₁₇H₃₁O₆S   | 347.1898 | 2      | 1.309  |
| C₁₄H₂₆O₆S   | 349.0963 | 4      | 1.610  |
| C₁₅H₂₆O₆S   | 349.1326 | 3      | 2.194  |
| C₁₆H₂₆O₆S   | 349.1690 | 2      | 2.253  |
| C₁₄H₂₅O₆S   | 351.1119 | 3      | 2.031  |
| C₁₅H₂₅O₆S   | 351.1483 | 2      | 2.370  |
| C₁₆H₂₃O₆S   | 351.1847 | 1      | 5.103  |
| C₁₄H₂₃O₆S   | 353.1276 | 2      | 1.433  |
| C₁₅H₂₅O₆S   | 353.1639 | 1      | 1.019  |
| Type                  | CHOS − SO₃ − CHO (1) | CHONS − SO₃ − CHON (2) | Total       |
|----------------------|----------------------|------------------------|-------------|
| Number               | Median               | Range                  | Average±STD | Median   | Range                  | Average±STD | Median   | Range                  | Average±STD | Median   | Range                  | Average±STD | Median   | Range                  | Average±STD |
| Percentage(%)        | 28                   | 11-37                  | 27±7        | 30       | 10-40                  | 28±7        | 30       | 10-40                  | 28±7        | 30       | 10-40                  | 28±7        |
| Intensity percentages (%) | 48                  | 18-62                  | 46±12       | 49                  | 15-61                  | 46±12       |
|                      | 359.1898             | 361.2054               | 363.1119    | 363.1483            | 363.1847               | 363.2211    | 365.1639            | 365.2003               | 367.1432               | 375.1847               | 377.2003               | 377.2367               | 379.1432               | 379.2160               | 381.1589               | 387.0391               | 389.2367               | 391.1796               | 391.2160               | 393.1952               | 395.1745               | 395.2109               | 405.2316               | 405.2680               | 407.1745               | 409.1902               | 417.2680               | 419.2837               | 433.2629               | 433.2993               | 461.2942               | 463.3099               | 477.2891               | 241.9976               | 258.0289               | 326.0551               | 330.0500               | 362.1279               | 382.1177               | 708                   | 480                   | 1158                   | 135-2165               | 1207±578               | 971±6                 | 46±12                  | 46±12                  | 46±12                  |

**Table S11.** Number and percentage occurrences of the plausible reactant– product pairs
Table S12. Selected meteorological parameters and chemical variables that probably have influences on the formation of NOCs. This table has been revised from our previous study and the references therein (Jiang et al., 2021b).

| Abbreviation | Full name | Major Sources/influences |
|--------------|-----------|--------------------------|
| SO₂          | Sulfur dioxide |                          |
| NO           | Nitric oxide   |                          |
| NO₂          | Nitrogen dioxide |                     |
| NO₃⁻         | Nitrogen oxides |                          |
| CO           | Carbon monoxide |                          |
| O₃           | Ozone          |                          |
| NO₃⁺ + O₃    | Oxidants       |                          |
| NH₄⁺         | Ammonium       |                          |
| NO₃⁻         | Nitrates       |                          |
| SO₄²⁻/nss-SO₄²⁻ | Sulfates/ non-sea-salt sulfates | Secondary sulfate formation process |
| Cl⁻          | Chloridion     |                          |
| K⁺/nss-K⁺    | Potassium/non-sea-salt potassium | Biomass burning (also from coal combustion and other sources) |
| Levo         | levoglucosan   |                          |
| MTLs         | sum of 2-methylthreitol and 2-methylerthritol | Isoprene derived SOA |
| MSOA         | monoterpane-derived secondary organic aerosols | α/β-pinene derived SOA |
| FA           | Fatty acids    |                          |
| PAHs         | Polycyclic aromatic hydrocarbons | Combustion sources |
| Alkane       | Long-chain alkanes with C number from 20 to 36 | Combustion sources and high-level plans |
| ΣSH          | steranes and hopanes | Fossil fuels combustion sources |
| LWC          | Liquid water content | Influence the aqueous phase reaction |
| Tem          | Temperature    | Influence the gas-to-particle partitioning |
| RH           | Relative humidity | Influence the aqueous phase reaction |
| OH           | Hydroxyl radical | Influence the oxidation state of precursor/photo-decomposed |
| pH           | Potential of hydrogen | Influence the aqueous phase reaction (range: -0.08-4.90) |
| Δ¹⁴C         | Radiocarbon isotope | Indicator of fossil or non-fossil sources |
Table S13. Number and percentage of compounds classes with significant correlations to the environmental variables.

| Type | Parameters | p-value original | p-value (FDR-adjusted) |
|------|------------|-----------------|------------------------|
|      | CHOS       | CHONS           | CHOS                   | CHONS                   |
|      | Positive   | Negative        | Positive               | Negative               |
| RH   | 591        | 172             | 180                    | 66 (28%)               | 322                      | 20 (74%)                | 65                      | 7 (26%)                 |
|      | (77%)      | (72%)           | (23%)                  | (83%)                  | (17%)                    | (17%)                    | (17%)                    | (26%)                    |
| Tem  | 260        | 697             | 54                     | 514                    | 170                      | 352                      | 22                      | 261                     |
|      | (83%)      | (58%)           | (17%)                  | (42%)                  | (89%)                    | (57%)                    | (11%)                    | (43%)                    |
| MSOA | 478        | 465             | 416                    | 62 (12%)               | 375                      | 260                      | 277                     | 22 (8%)                 |
|      | (53%)      | (88%)           | (47%)                  | (58%)                  | (92%)                    | (42%)                    | (22%)                    | (8%)                     |
| MTLs | 336        | 696             | 124                    | 274                    | 253                      | 451                      | 60                      | 123                     |
|      | (73%)      | (72%)           | (27%)                  | (28%)                  | (81%)                    | (79%)                    | (19%)                    | (21%)                    |
| Δ¹⁴C | 199        | 440             | 87                     | 200                    | 37                       | 225                      | 15                      | 92 (29%)                |
|      | (70%)      | (69%)           | (30%)                  | (31%)                  | (71%)                    | (71%)                    | (29%)                    | (92%)                    |
| NH₄⁺ | 230        | 244             | 306                    | 42 (15%)               | 21 (26%)                 | 56 (89%)                 | 59 (74%)                 | 7 (11%)                 |
|      | (43%)      | (85%)           | (57%)                  | (57%)                  | (26%)                    | (89%)                    | (74%)                    | (11%)                    |
| NO₃⁻ | 283        | 159             | 359                    | 42 (21%)               | 46                       | 40 (75%)                 | 83 (64%)                 | 13 (25%)                |
|      | (44%)      | (79%)           | (56%)                  | (56%)                  | (36%)                    | (75%)                    | (64%)                    | (25%)                    |
| LWC  | 330        | 22 (72%)        | 392                    | 8 (28%)                | 17 (100%)                | 0                        | 43 (100%)                | 0                       |
|      | (46%)      | (72%)           | (54%)                  | (38%)                  | (100%)                   | (100%)                   | (100%)                   | (0%)                     |
| pH   | 65         | 11 (48%)        | 51 (44%)               | 12 (52%)               | 0                        | 0                        | 0                       | 0                       |
|      | (56%)      | (48%)           | (44%)                  | (52%)                  | (52%)                    | (52%)                    | (52%)                    | (52%)                    |
| H⁺   | 11         | 65 (56%)        | 12 (52%)               | 51 (44%)               | 0                        | 0                        | 0                       | 0                       |
|      | (48%)      | (56%)           | (52%)                  | (44%)                  | (52%)                    | (52%)                    | (52%)                    | (52%)                    |
| SO₂⁻ | 247        | 131             | 95 (28%)               | 76 (37%)               | 0                        | 0                        | 0                       | 0                       |
|      | (72%)      | (63%)           | (28%)                  | (37%)                  | (37%)                    | (37%)                    | (37%)                    | (37%)                    |
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