



Supplement of

Sources and processes of water-soluble and water-insoluble organic aerosol in cold season in Beijing, China

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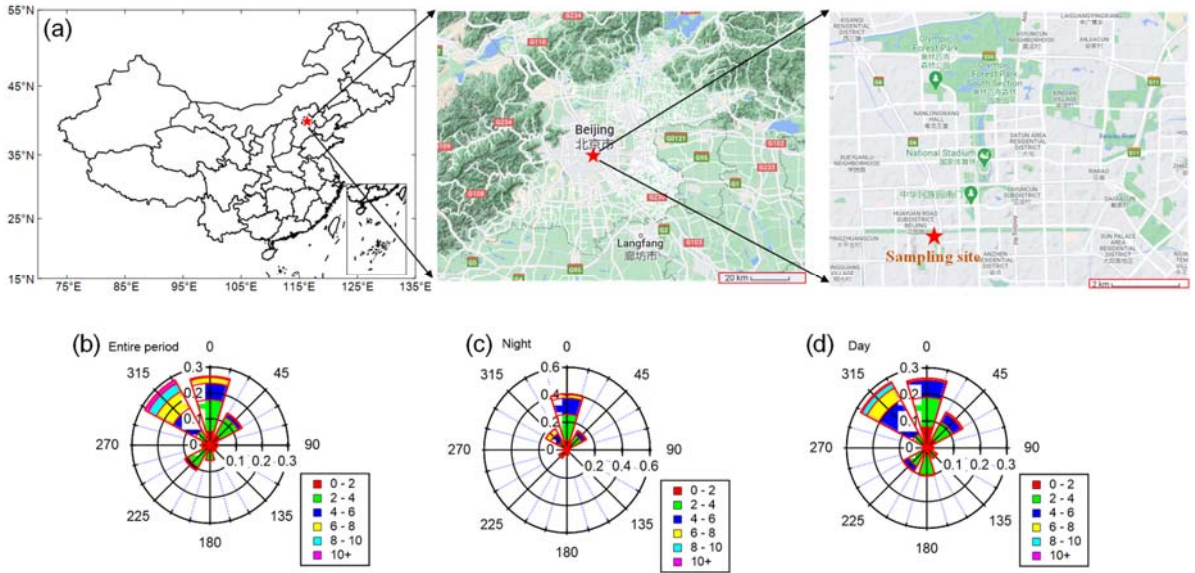


Figure S1. (a) Sampling site position (© Google Maps), and the wind rose plots of (b) the entire period, (c) nighttime (from 6 November to 5 December 2020) and (d) daytime, respectively.

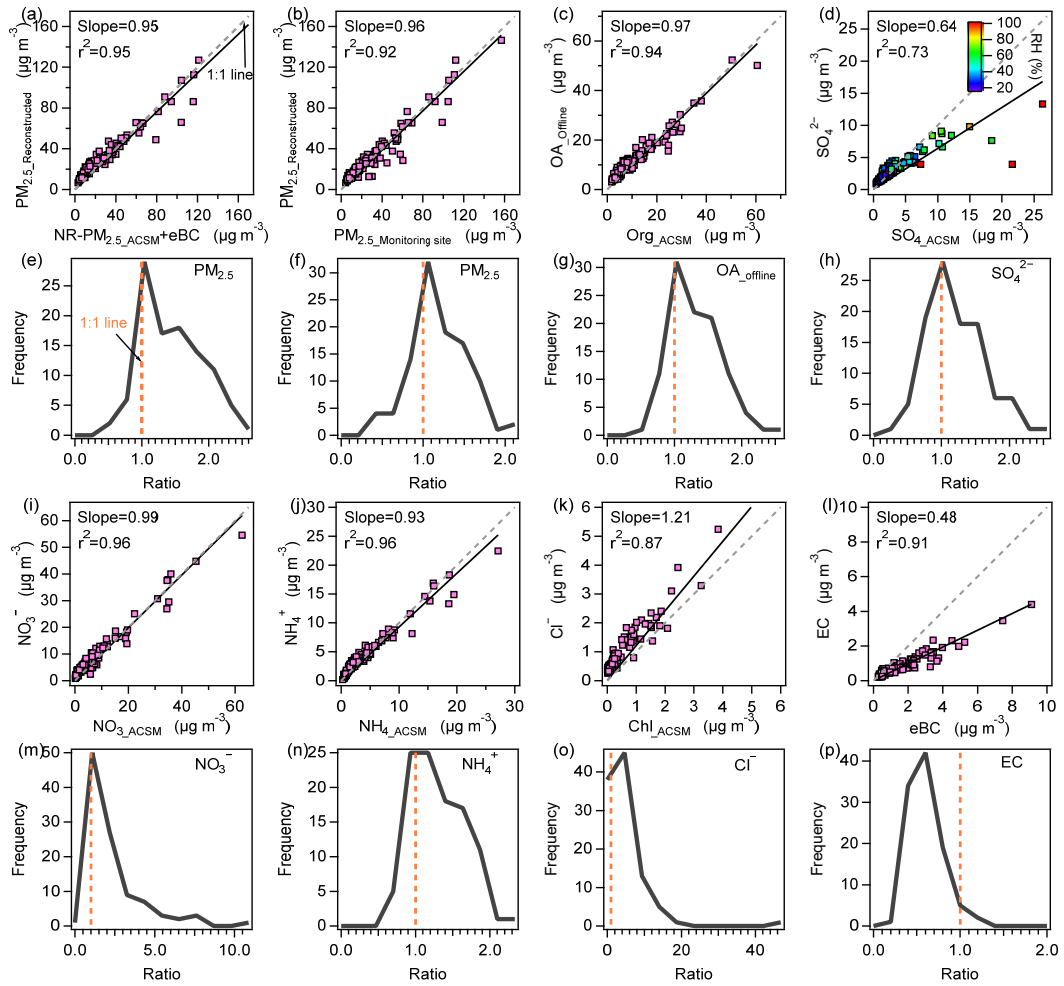


Figure S2. Mass concentration comparisons of online and offline measurements, and the offline-to-online ratio frequency (103 samples in total) distribution.

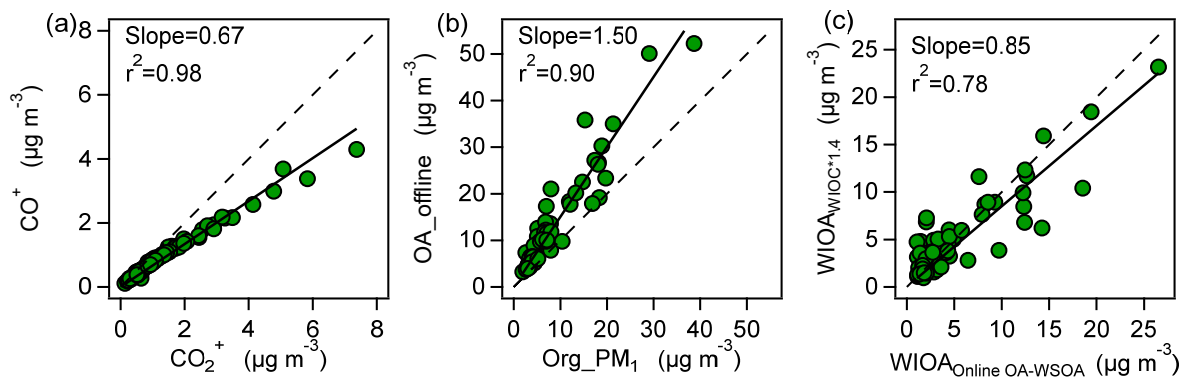


Figure S3. Scatter plots of (a) CO^+ vs. CO_2^+ in WSOA in this study, (b) offline-reconstructed OA (WSOA + WIOA) vs. Org in PM_1 measured with AMS, and (c) reconstructed WIOA ($\text{WIOC} \times 1.4$) vs. WIOA calculated from the difference between OA and WSOA. The dashed lines indicate the 1:1 lines.

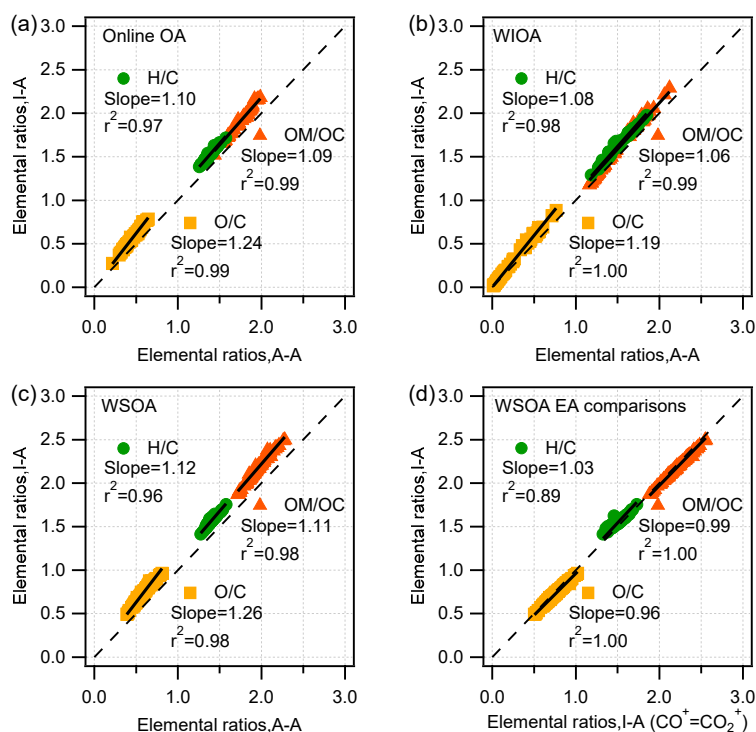


Figure S4. Elemental ratio comparisons of I-A method and A-A method of (a) online OA, (b) WIOA, (c) WSOA (CO^+ use original values). Panel (d) shows the WSOA elemental ratio comparisons when CO^+ use original values and when CO^+ values are equal to CO_2^+ values.

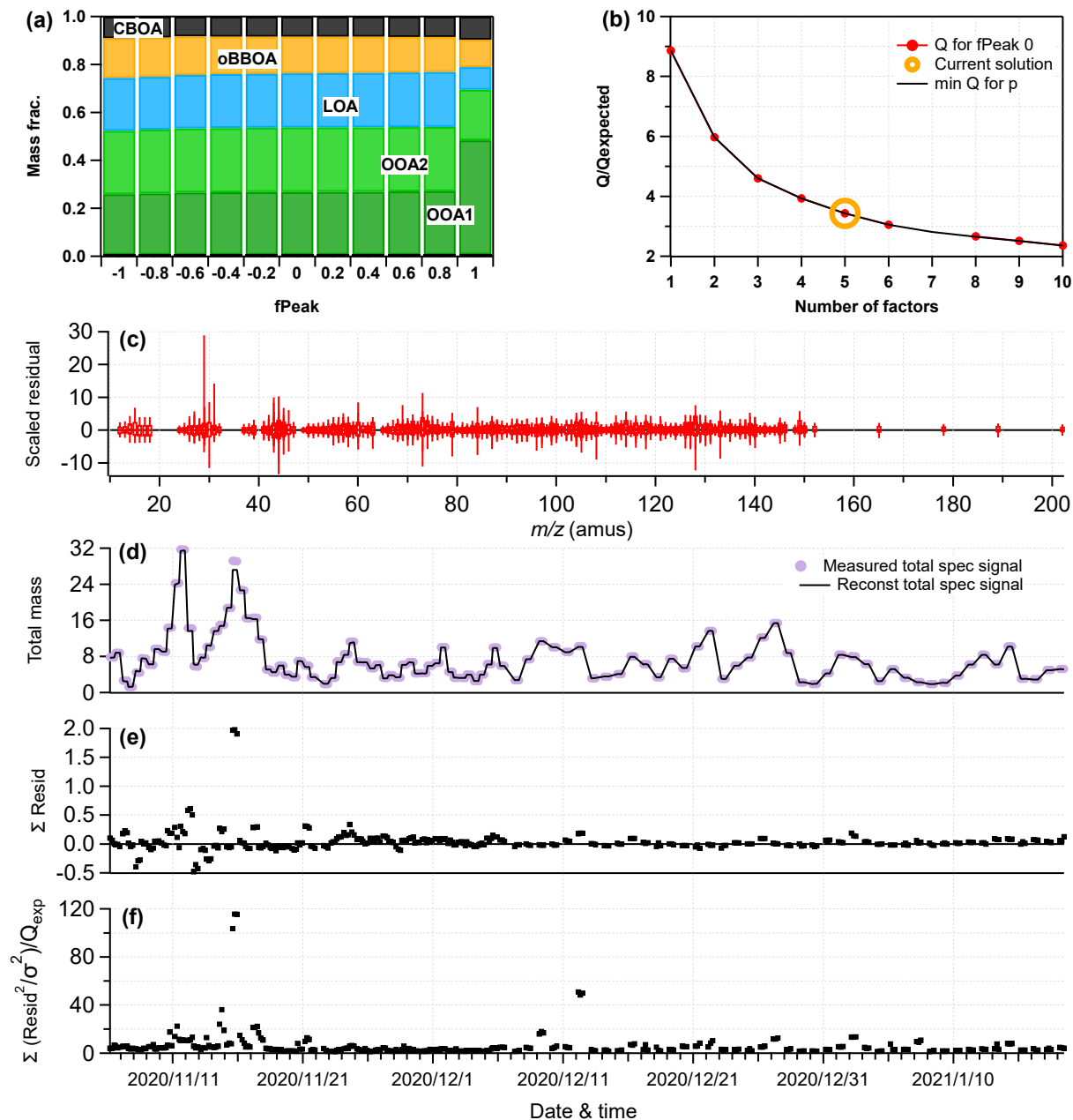


Figure S5. Diagnostic plots of PMF results for 5-factor solution: (a) fractions of OA factors vs. f_{peak} ; (b) Q/Q_{exp} as a function of number of factors selected for PMF modeling; (c) box and whiskers ($\pm 25\%$ of box) presenting the distribution of scaled residuals for each m/z ; (d) time series of the measured organic mass and reconstructed (reconstructed WSOA = OOA1 + OOA2 + LOA + oBBOA + CBOA) organic mass; (e) time series of the residuals (residuals = measured mass – reconstructed mass); (f) time series of Q/Q_{exp} .

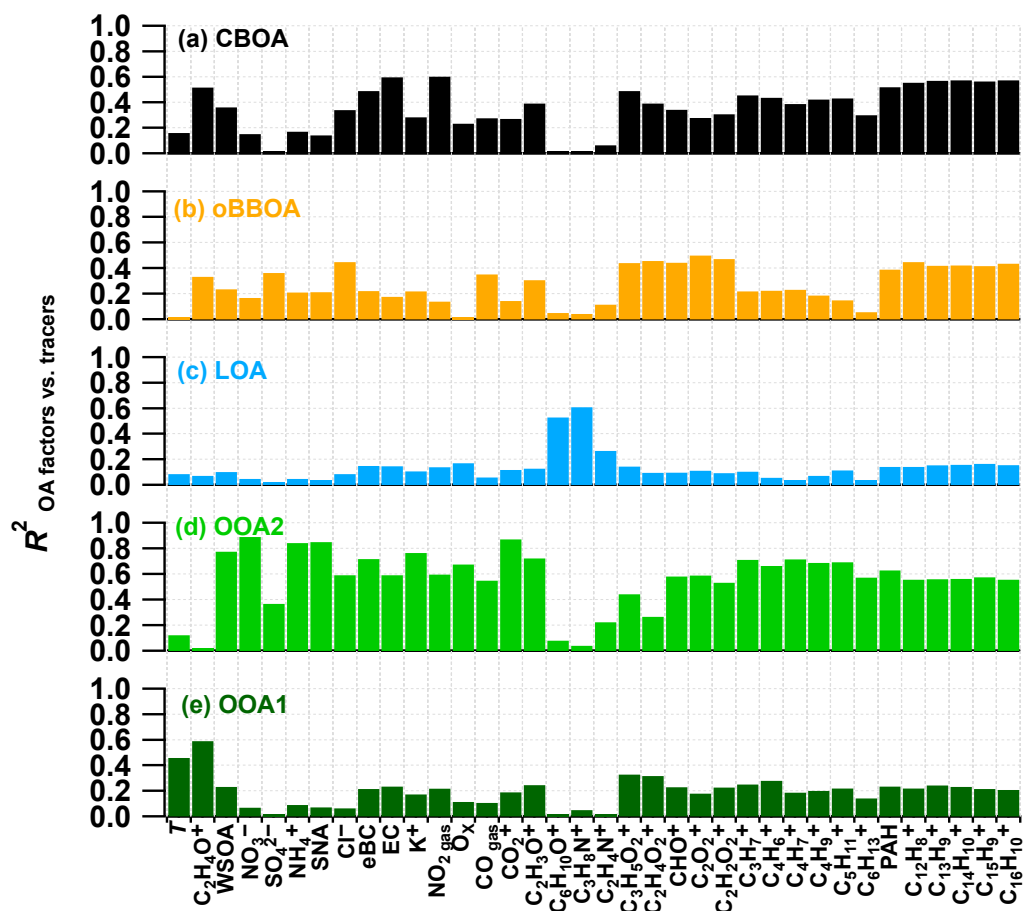


Figure S6. Correlations (R^2) between WSOA factors and common tracers.

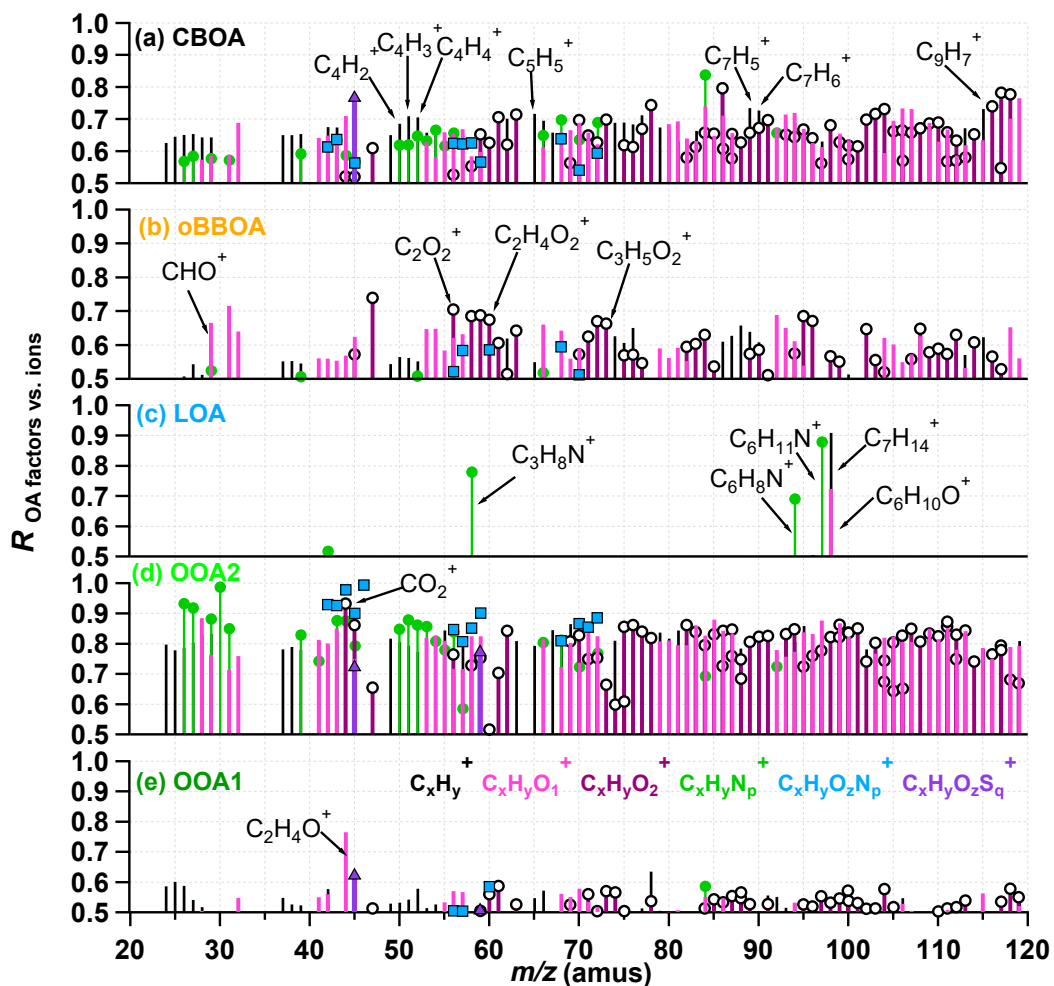


Figure S7. Correlation ($R > 0.5$) of WSOA factors and individual fragment up to m/z 120.

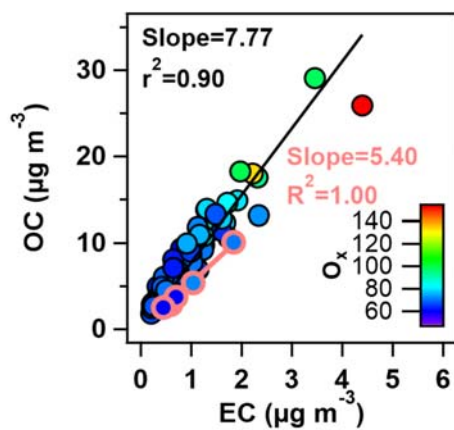


Figure S8. EC-tracer method (OC vs. EC), the points with pink edges have the least OC/EC ratios.

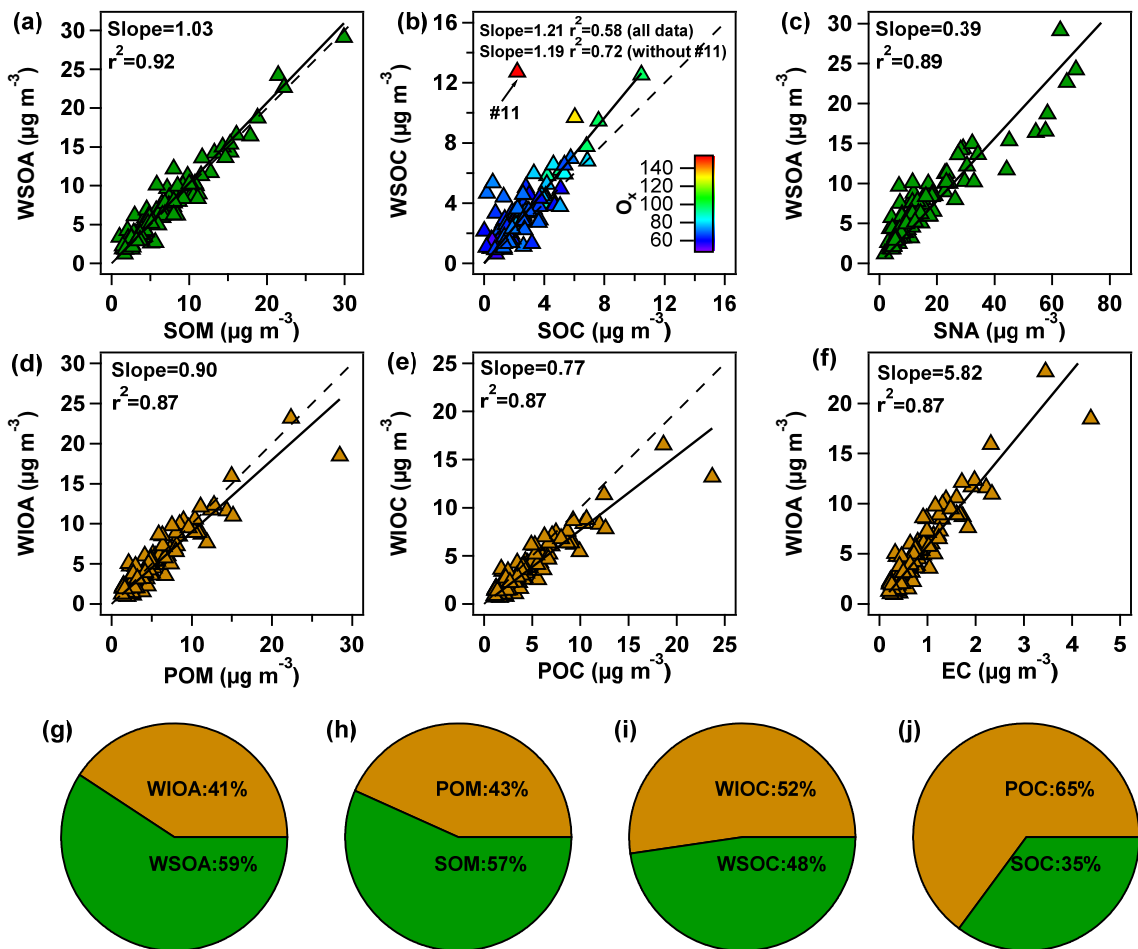


Figure S9. Scatter plots that compare (a) WSOA vs. SOM, (b) WSOC vs. SOC, (c) WSOA vs. SNA, (d) WIOA vs. POM, (e) WIOC vs. POC and (f) WIOA vs. EC, as well as composition pie charts of (g) WIOA & WSOA, (h) POM & SOM, (i) WIOC & WSOC and (j) POC & SOC. The black dashed lines indicate the 1:1 lines.

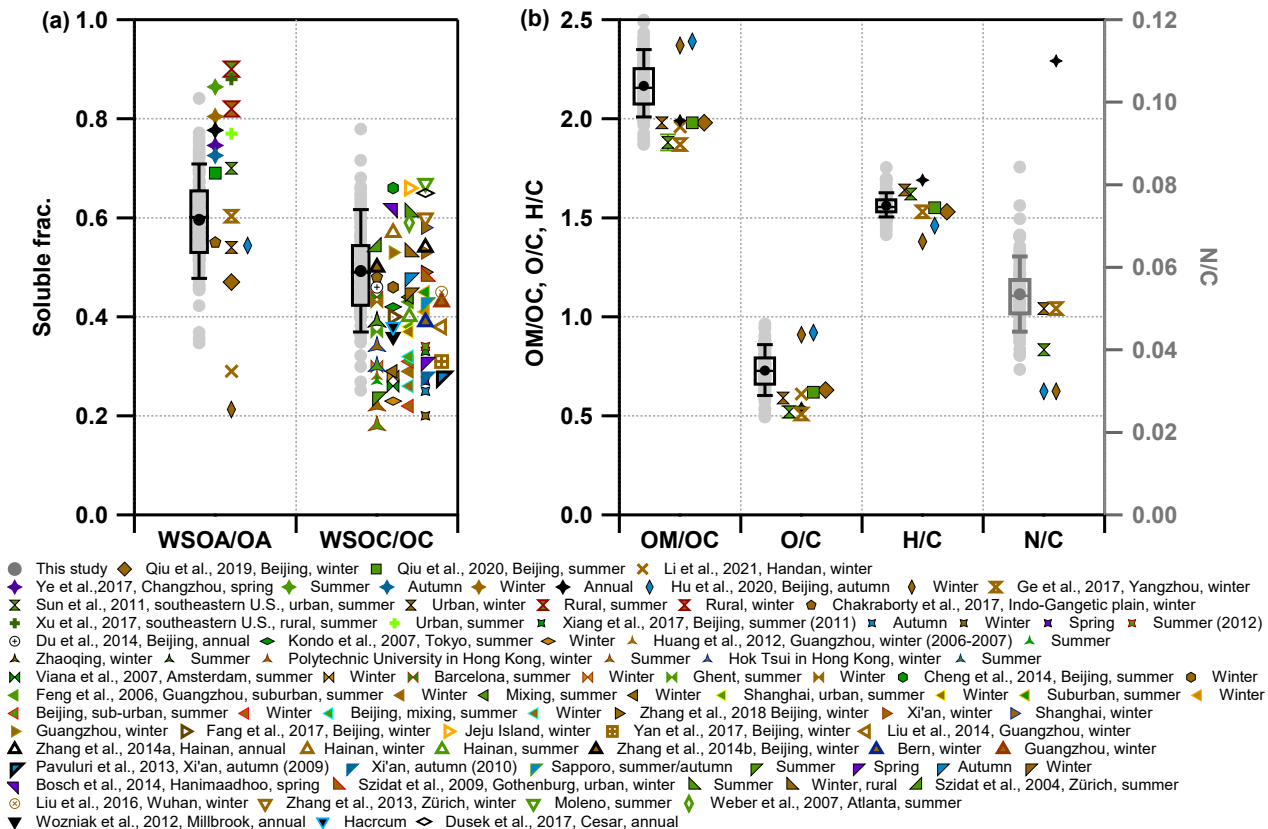


Figure S10. The same picture as Fig. 2, and the detailed references are listed below (Qiu et al., 2019; Qiu et al., 2020; Li et al., 2021; Ye et al., 2017; Hu et al., 2020; Ge et al., 2017; Sun et al., 2011; Chakraborty et al., 2017; Xu et al., 2017; Xiang et al., 2017; Du et al., 2014; Kondo et al., 2007; Huang et al., 2012; Viana et al., 2007; Cheng et al., 2014; Feng et al., 2006; Zhang et al., 2018; Fang et al., 2017; Yan et al., 2017; Liu et al., 2014; Zhang et al., 2014a; Zhang et al., 2014b; Pavuluri et al., 2013; Bosch et al., 2014; Szidat et al., 2009; Szidat et al., 2004; Liu et al., 2016; Zhang et al., 2013; Weber et al., 2007; Wozniak et al., 2012; Dusek et al., 2017).

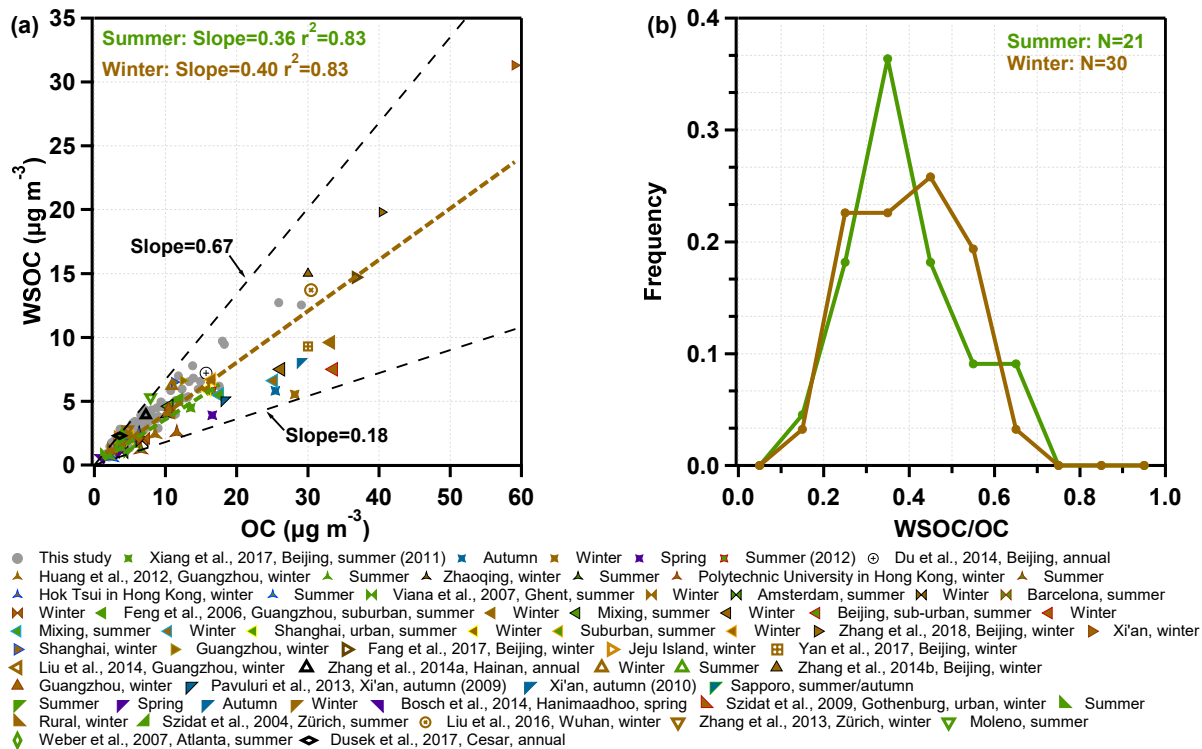


Figure S11. WSOC and OC values summarized from the previous studies (Xiang et al., 2017; Du et al., 2014; Huang et al., 2012; Viana et al., 2007; Feng et al., 2006; Zhang et al., 2018; Fang et al., 2017; Yan et al., 2017; Liu et al., 2014; Zhang et al., 2014a; Zhang et al., 2014b; Pavuluri et al., 2013; Bosch et al., 2014; Szidat et al., 2009; Szidat et al., 2004; Liu et al., 2016; Zhang et al., 2013; Weber et al., 2007; Dusek et al., 2017), the multiple datasets in summer and winter are fitted for comparison in panel (a), and the frequency distribution of WSOC/OC in summer and winter are shown in panel (b). Colored markers represent data from previous studies, which are colored in purple in spring, green in summer, blue in autumn and brown in winter, the data across the year are in black. The results of different seasons and different sites in the same reference are shown with different edge colors in panel (a).

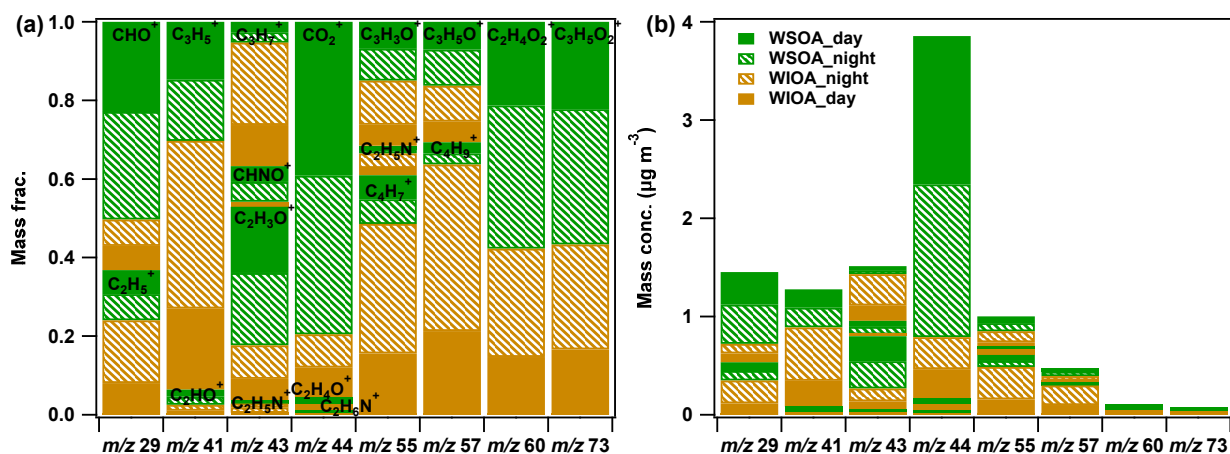


Figure S12. (a) Mass fractions (WSOA_day + WSOA_night + WIOA_night + WIOA_day) and (b) concentrations in m/z 29, m/z 41, m/z 43, m/z 44, m/z 55, m/z 57, m/z 60 and m/z 73.

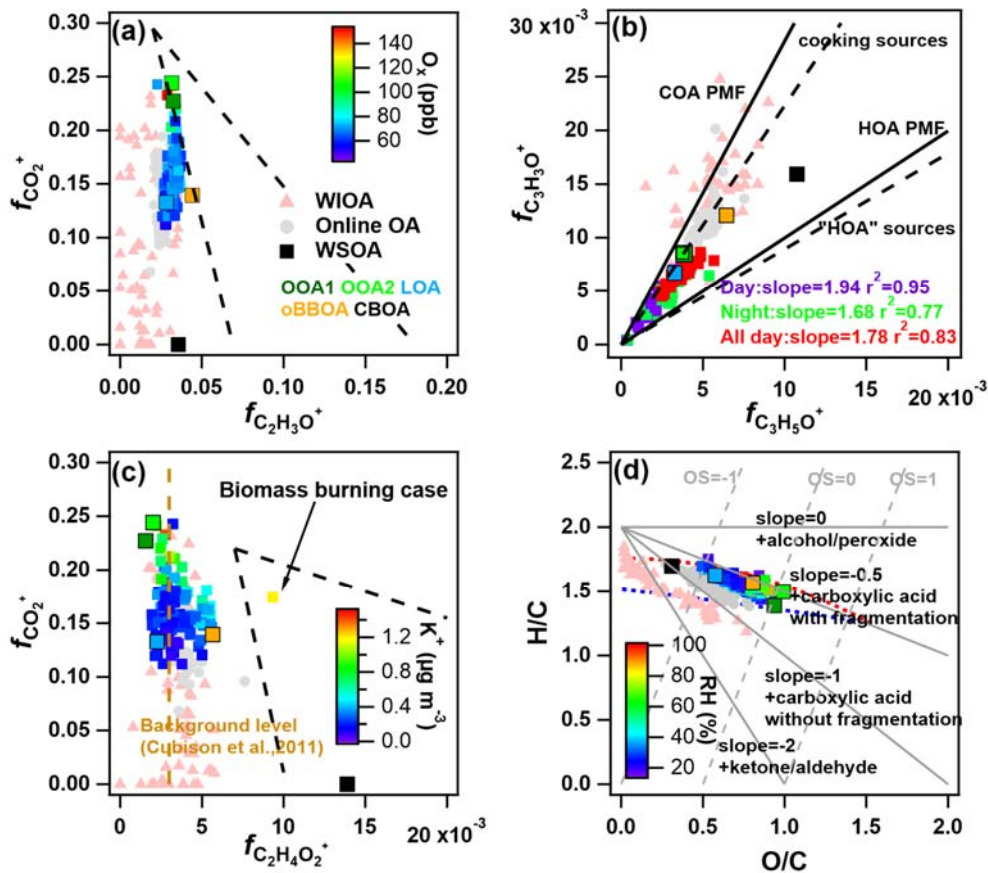


Figure S13. Triangle plots of main fragments corresponding to the unit m/z s in Fig. 7, including (a) $f_{CO_2^+}$ vs. $f_{C_2H_3O^+}$, (b) $f_{C_3H_3O^+}$ vs. $f_{C_3H_5O^+}$, (c) $f_{CO_2^+}$ vs. $f_{C_2H_4O_2^+}$. Panel (d) shows the Van Krevelen diagram, in which the WSOA data are colored by RH. The reference lines are the same as the lines in Fig. 7.

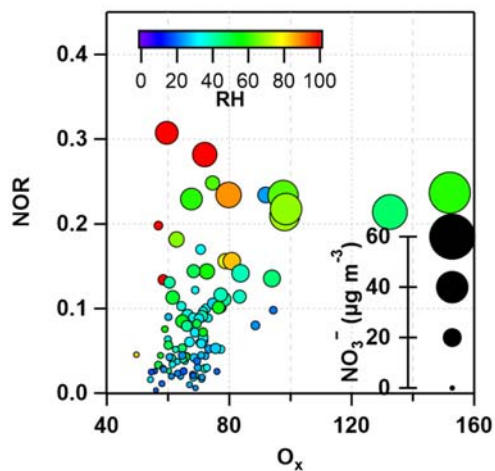


Figure S14. The nitrogen oxidation ratio (NOR) variation as a function of O_x , the data are colored by RH, and the size of markers indicates the mass concentration of NO_3^- .

Table S1. Night-day differences of average values of offline-measured species, elemental ratios (from 6 November 2020 to 5 December 2020, I-A method, CO^+ use original values) and online-measured meteorological parameters and gases, and the average values of the entire time and each divided episode, values of one standard deviation are also shown in the table.

Species	Night	Day	Night-day absolute change	Night-day relative change	Entire period	P1	P2	P3	P4
PM_{2.5} and components ($\mu\text{g m}^{-3}$)									
PM _{2.5}	37.2 ± 33.3	34.6 ± 28.7	2.6	7.5%	32.6 ± 26.0	73.1 ± 37.7	126.9	21.9 ± 9.0	28.4 ± 16.6
OA _{offline}	15.2 ± 12.2	12.2 ± 8.8	3.0	24.7%	12.6 ± 9.0	26.2 ± 12.4	52.3	8.2 ± 3.8	11.3 ± 6.0
WSOA	8.8 ± 7.0	8.0 ± 5.9	0.8	9.8%	7.5 ± 5.5	16.3 ± 7.4	29.1	5.2 ± 2.1	6.3 ± 3.4
WIOA	6.4 ± 5.4	4.2 ± 3.0	2.2	53.3%	5.1 ± 3.8	10.0 ± 5.5	23.2	3.0 ± 1.9	5.0 ± 2.8
SO ₄ ²⁻	3.4 ± 2.0	3.7 ± 2.4	-0.4	-9.4%	3.5 ± 2.1	5.5 ± 3.1	8.4	3.1 ± 0.8	3.4 ± 2.0
NO ₃ ⁻	10.0 ± 12.4	10.6 ± 11.9	-0.6	-5.6%	8.9 ± 9.9	24.5 ± 15.7	37.5	5.4 ± 2.9	7.1 ± 5.3
NH ₄ ⁺	4.9 ± 5.2	4.9 ± 4.9	0.02	0.4%	4.3 ± 4.2	10.9 ± 6.3	16.9	2.8 ± 1.4	3.6 ± 2.6
Cl ⁻	1.2 ± 1.2	0.9 ± 0.7	0.3	28.5%	1.1 ± 0.9	2.0 ± 1.4	5.2	0.7 ± 0.3	1.1 ± 0.8
EC	1.1 ± 1.0	0.8 ± 0.5	0.3	44.3%	0.8 ± 0.7	1.7 ± 1.0	3.5	0.6 ± 0.3	0.7 ± 0.5
Other ions	1.4 ± 0.8	1.5 ± 0.7	-0.1	-5.7%	1.3 ± 0.5	2.2 ± 0.7	3.2	1.1 ± 0.3	1.2 ± 0.4
WSOA factors ($\mu\text{g m}^{-3}$)									
OOA1	2.7 ± 2.4	2.5 ± 2.2	0.2	6.8%	2.0 ± 1.9	4.6 ± 2.5	10.0	1.2 ± 0.9	1.2 ± 0.7
OOA2	2.5 ± 5.7	2.4 ± 5.1	0.1	4.3%	2.0 ± 4.3	8.2 ± 8.0	7.5	0.3 ± 0.5	1.4 ± 2.0
LOA	1.8 ± 1.1	2.2 ± 1.4	-0.4	-17.7%	1.7 ± 1.0	0.9 ± 1.0	0.9	2.9 ± 0.6	1.3 ± 0.3
oBBOA	0.9 ± 1.4	0.4 ± 0.6	0.5	110.0%	1.1 ± 1.3	1.3 ± 1.7	7.0	0.4 ± 0.5	1.8 ± 1.2
CBOA	0.8 ± 0.8	0.5 ± 0.5	0.4	85.3%	0.6 ± 0.6	1.2 ± 0.7	1.8	0.3 ± 0.4	0.6 ± 0.6
Other ions ($\mu\text{g m}^{-3}$)									
Ca ²⁺	0.17 ± 0.14	0.17 ± 0.14	-0.003	-1.7%	0.22 ± 0.16	0.23 ± 0.12	0.22	0.10 ± 0.05	0.29 ± 0.16
F ⁻	0.15 ± 0.06	0.17 ± 0.04	-0.02	-10.8%	0.14 ± 0.05	0.22 ± 0.07	0.35	0.15 ± 0.02	0.11 ± 0.02
Na ⁺	0.13 ± 0.12	0.10 ± 0.09	0.03	28.8%	0.15 ± 0.12	0.24 ± 0.14	0.57	0.07 ± 0.04	0.19 ± 0.11
NO ₂ ⁻	0.07 ± 0.07	0.12 ± 0.09	-0.05	-39.6%	0.08 ± 0.07	0.11 ± 0.09	0	0.08 ± 0.08	0.07 ± 0.04
K ⁺	0.42 ± 0.32	0.35 ± 0.26	0.07	18.7%	0.35 ± 0.24	0.74 ± 0.34	1.19	0.24 ± 0.08	0.30 ± 0.14
Mg ²⁺	0.45 ± 0.17	0.56 ± 0.12	-0.11	-19.6%	0.40 ± 0.17	0.62 ± 0.13	0.85	0.46 ± 0.16	0.27 ± 0.03
Elemental ratios									
OM/OC	2.15 ± 0.14	2.16 ± 0.17	-0.01	-0.7%	2.16 ± 0.13	2.34 ± 0.11	2.32	2.07 ± 0.09	2.18 ± 0.10
O/C	0.72 ± 0.10	0.73 ± 0.13	-0.01	-1.5%	0.73 ± 0.10	0.86 ± 0.07	0.85	0.65 ± 0.07	0.74 ± 0.07
H/C	1.56 ± 0.06	1.56 ± 0.07	-0.002	-0.1%	1.56 ± 0.05	1.51 ± 0.04	1.55	1.59 ± 0.05	1.56 ± 0.03
N/C	0.055 ± 0.008	0.055 ± 0.009	-0.000	-0.1%	0.053 ± 0.008	0.059 ± 0.013	0.053	0.055 ± 0.003	0.052 ± 0.006
Meteorological parameters									
RH (%)	48.6 ± 20.2	43.1 ± 23.9	5.4	12.5%	40.5 ± 19.7	61.3 ± 25.3	69.9	43.9 ± 18.0	33.5 ± 12.9
T (°C)	5.0 ± 5.0	7.0 ± 5.4	-2.0	-29.0%	2.6 ± 6.2	11.1 ± 2.1	10.1	2.2 ± 2.6	-1.8 ± 4.0
WS (m s ⁻¹)	3.5 ± 1.3	3.7 ± 1.5	-0.11	-3.0%	3.9 ± 1.6	2.7 ± 0.9	2.2	3.9 ± 1.3	4.3 ± 1.7
Gases (ppb)									
NO ₂	53.5 ± 28.8	38.4 ± 27.7	15.1	39.4%	42.6 ± 25.5	78.0 ± 29.5	96.6	31.2 ± 16.1	38.5 ± 19.2
CO	63.5 ± 43.4	56.6 ± 41.2	6.9	12.2%	58.9 ± 37.9	110.0 ± 50.0	181.4	41.1 ± 14.9	57.0 ± 31.5
O ₃	17.1 ± 15.8	35.0 ± 17.4	-17.9	-51.1%	28.0 ± 17.5	11.1 ± 14.2	1.5	32.6 ± 16.7	30.2 ± 15.8
SO ₂	3.7 ± 1.9	3.8 ± 2.3	-0.1	-2.6%	4.6 ± 2.5	4.5 ± 2.3	3.4	3.6 ± 2.1	5.8 ± 2.6
O _x	70.6 ± 19.0	73.4 ± 16.2	-2.8	-3.8%	70.6 ± 14.1	89.1 ± 24.0	98.1	63.9 ± 6.0	68.7 ± 6.6

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