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Supplement of

Six-year source apportionment of submicron organic aerosols from near-continuous highly time-resolved measurements at SIRTA (Paris area, France)

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Table S1. Trend analysis using the Mann-Kendall test and the seasonal Mann-Kendall test, respectively. The seasonality was exanimated using the Kruskal-Wallis test. Bold text values were selected for the quantification of final trends according to Kruskal-Wallis test at the 5% significance level. Note that the HOA factor without (w/o) December data was also tested.

Composition	Kruskal-Wallis test	Mann-Kendall test		Seasonal Mann-Kendall test	
	(seasonality)	*p*-value	Sen's slope	*p*-value	Sen's slope
PM_1	0.1353	**0.0123**	**-0.6440**	0.0056	-0.7552
SO_4	0.1210	**0.5117**	**-0.0237**	0.4562	-0.0241
NO_3	0.0000	0.0844	-0.0961	**0.0106**	**-0.1447**
OA	0.5594	**0.0028**	**-0.3823**	0.0078	-0.4227
HOA	0.1001	**0.0776**	**-0.0512**	0.0554	-0.0579
HOA (w/o Dec.)	0.1622	**0.0300**	**-0.0594**	0.02302	-0.0601
BBOA	0.0000	0.0548	-0.0674	**0.0106**	**-0.0647**
MO-OOA	0.0234	0.0184	-0.1813	**0.0431**	**-0.1754**
LO-OOA	0.0001	0.7446	-0.0159	**0.2871**	**-0.0383**
eBC_{ff}	0.0101	0.0282	-0.0198	**0.0040**	**-0.0198**
eBC_{wb}	0.0000	0.7482	-0.0014	**0.8073**	**-0.0006**

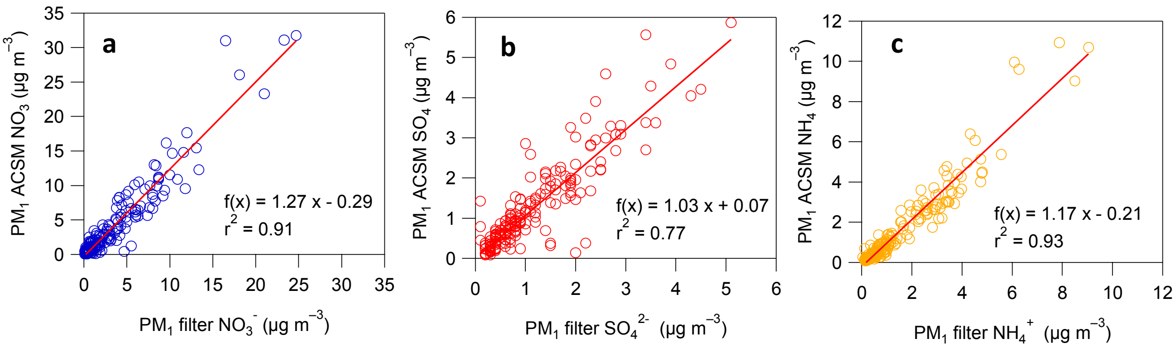

Figure S1. Correlations between the ACSM and 4-h PM$_1$ filter-based measurements for main inorganic species in Spring 2016 (from March 3 to April 22). Satisfactory correlation coefficients (r^2 = 0.77 - 0.93) are obtained for each species. For nitrate, a slope of 1.27 is observed, which can be partially explained by the fact that ammonium nitrate is semi-volatile, possibly leading to negative sampling artefacts in off-line measurements (Sciare et al., 2007; Ianniello et al., 2011). Consistently, ammonium also presents a slope higher than 1 (i.e., 1.17), but lower than the slope obtained for nitrate. This is in good agreement with ammonium being mainly combined with nitrate and sulfate, and the non-volatility of ammonium sulfate in ambient conditions. Moreover, the slope obtained for sulfate is much closer to 1 (i.e., 1.03). Overall, those results confirm the validity of the calibration parameters (including response factor and relative ions efficiency values) determined during the 2016 intercomparison exercise (Freney et al., 2019).

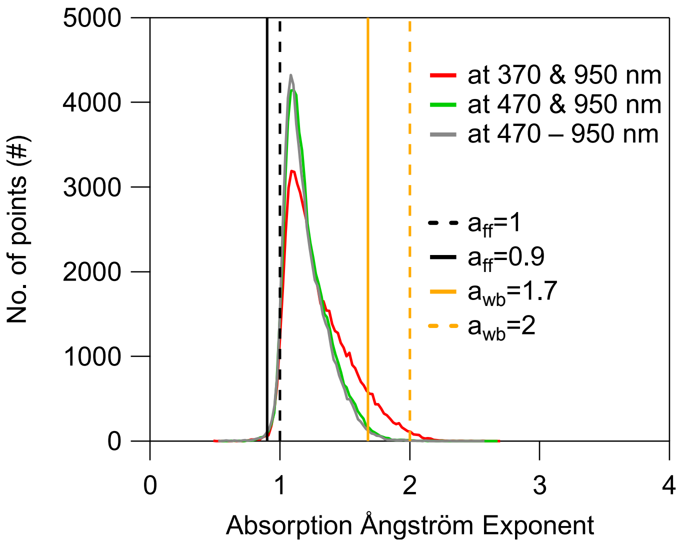

Figure S2. Frequency distributions of ambient absorption Angström exponent (AAE) values calculated at different paired-wavelengths (i.e., 370 and 950 nm, as well as 470 and 950 nm) and for the 6 wavelengths comprised between 470 and 950 nm.

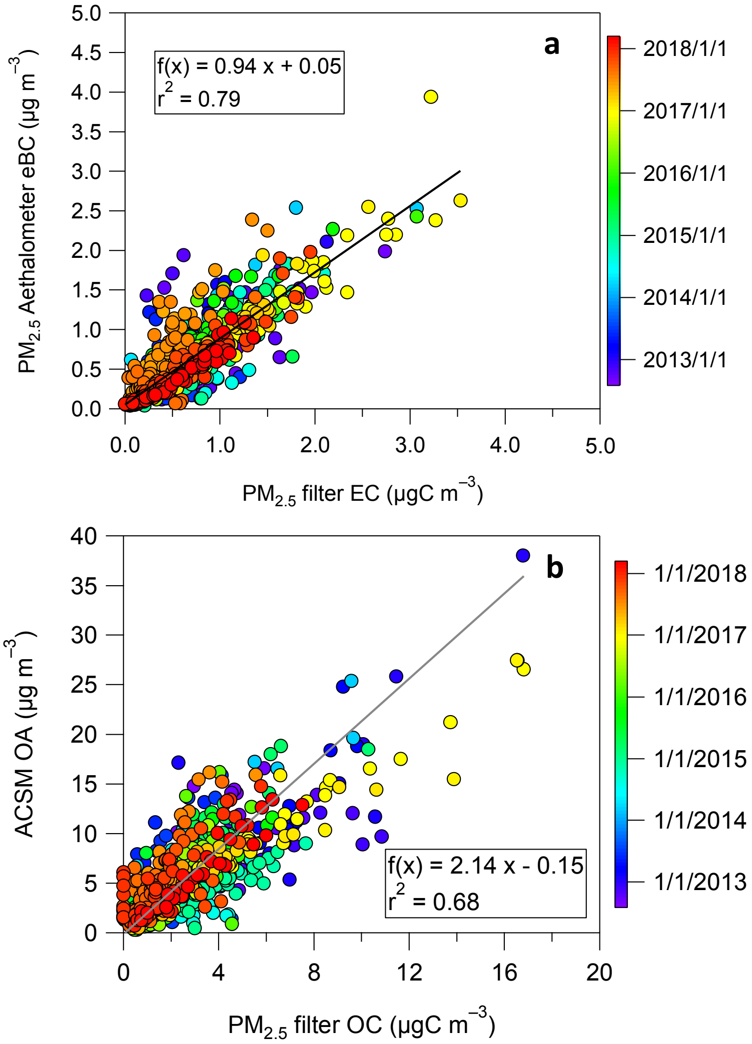

Figure S3. Comparison between online (AE33 and ACSM) and offline ($PM_{2.5}$ daily filters) carbonaceous aerosol measurements: (a) eBC (AE33) vs. EC, and (b) OA (ACSM) vs. OC.

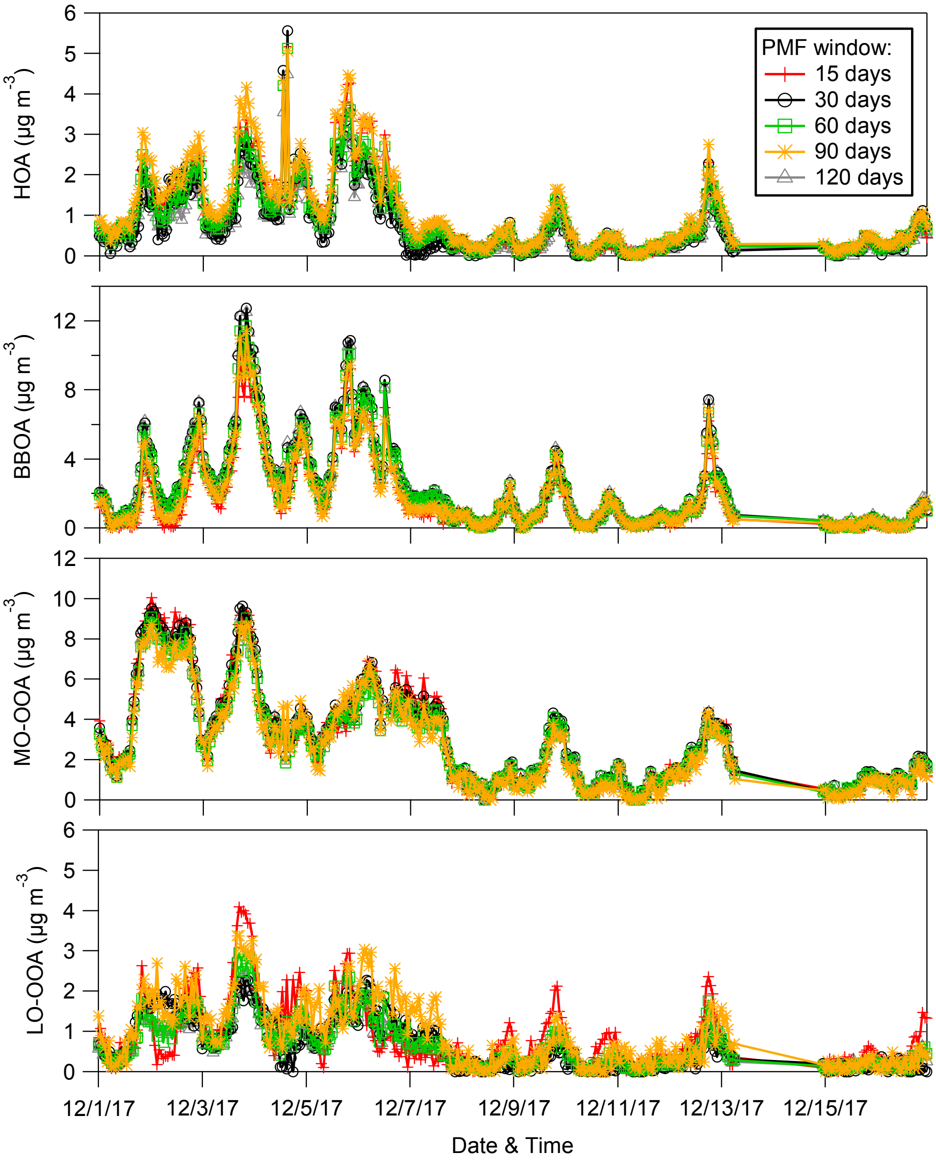

Figure S4. Comparisons of mass concentrations of four OA factors resolved from different PMF windows runs with setting of 15, 30, 60, 90 and 120 days, respectively.

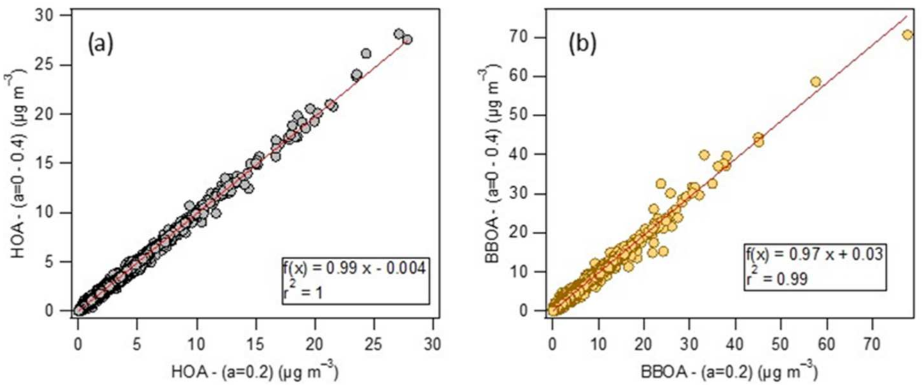

Figure S5. Comparisons of ME-2 runs between two different a-value settings, including the a-values varying range (*a*=0 - 0.4) and a specific *a*-value (a=0.2).

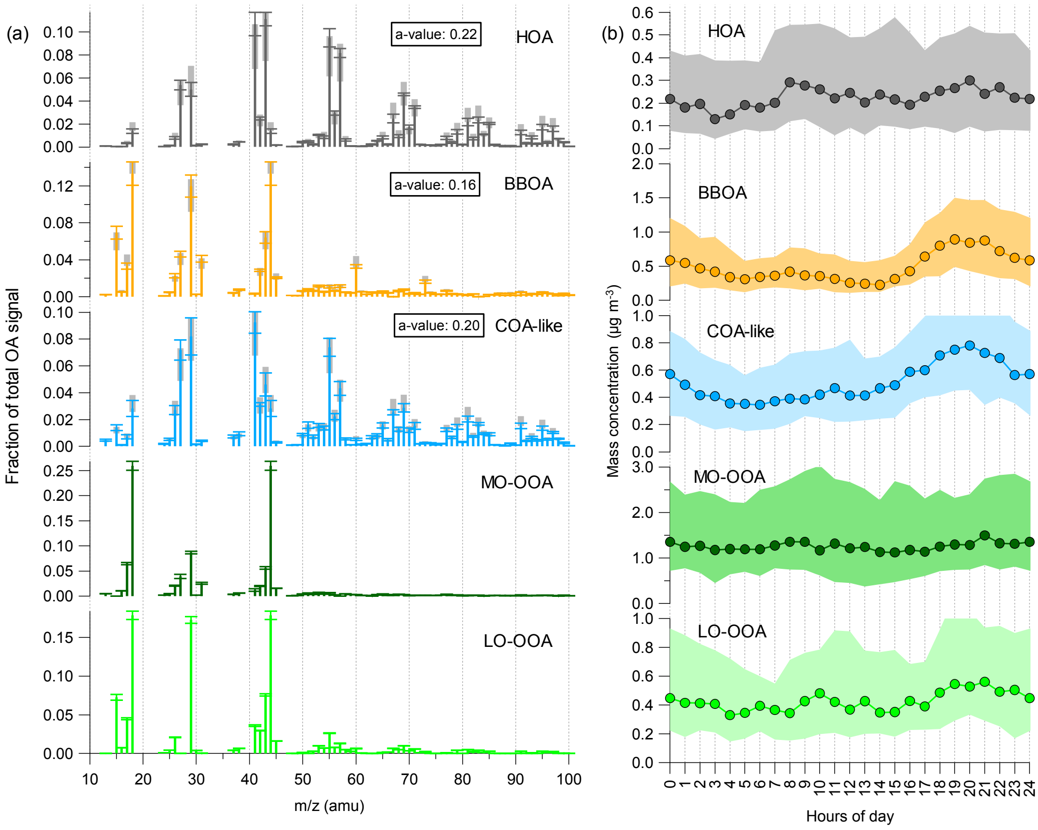

Figure S6. Mass spectra (a) and diel variations (b) of OA factors obtained from the 5-factor solution ME-2 runs for the winter 2017-2018 period. In (a), gray bars describe the range of *a*-value changes for constrained factors (i.e., HOA BBOA and COA-like) during ME-2 runs with the random mode. Errors in each plot present 1 standard deviation. Stick lines indicate average values over all selected ME-2 runs. Averages a-value for constrained factors during the ME-2 runs is also shown**.** In (b), shadow areas present the 25th and 75th percentiles respectively. Solid circle lines are median values.

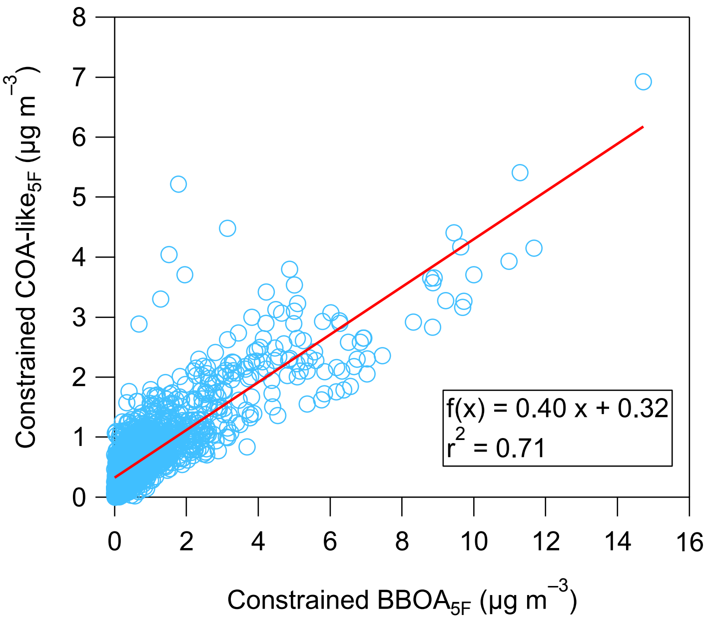

Figure S7. Relationships of constrained COA-like versus BBOA from the 5-factor solution.

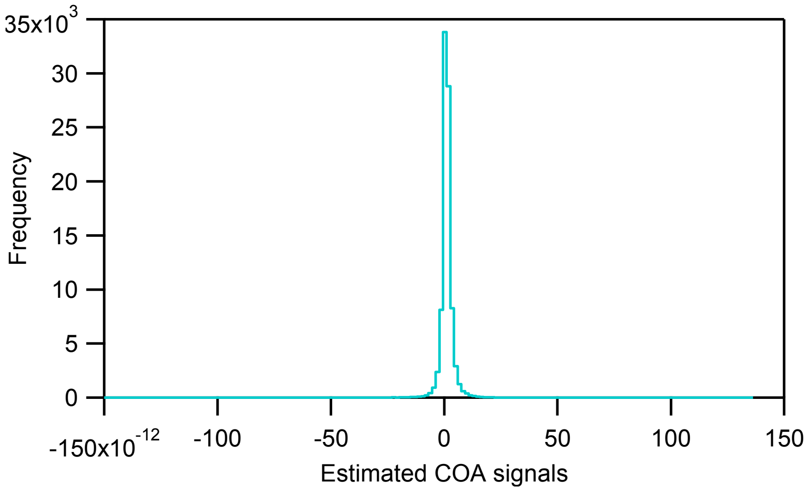

Figure S8. Distribution of the estimated COA signals by using a m/z-tracer method (Mohr et al., 2012) during the entire period. Briefly, the COA signals can be calculated based on the time series of the signals of *m/z* 55, *m/z* 57, and *m/z* 44 measured by the ACSM (using the equation: $COA_{est} = \left[\frac{\frac{1}{a}\cdot m/z55 - m/z57 + \left(c - \frac{b}{a}\right)\cdot m/z44}{\frac{1}{a} - \frac{1}{d}}\right]$). The parameters in this equation, a, b, c and d, are determined by the corresponding signals intensity in mass spectra of OA factors, which have been clearly described by Mohr et al. (2012).

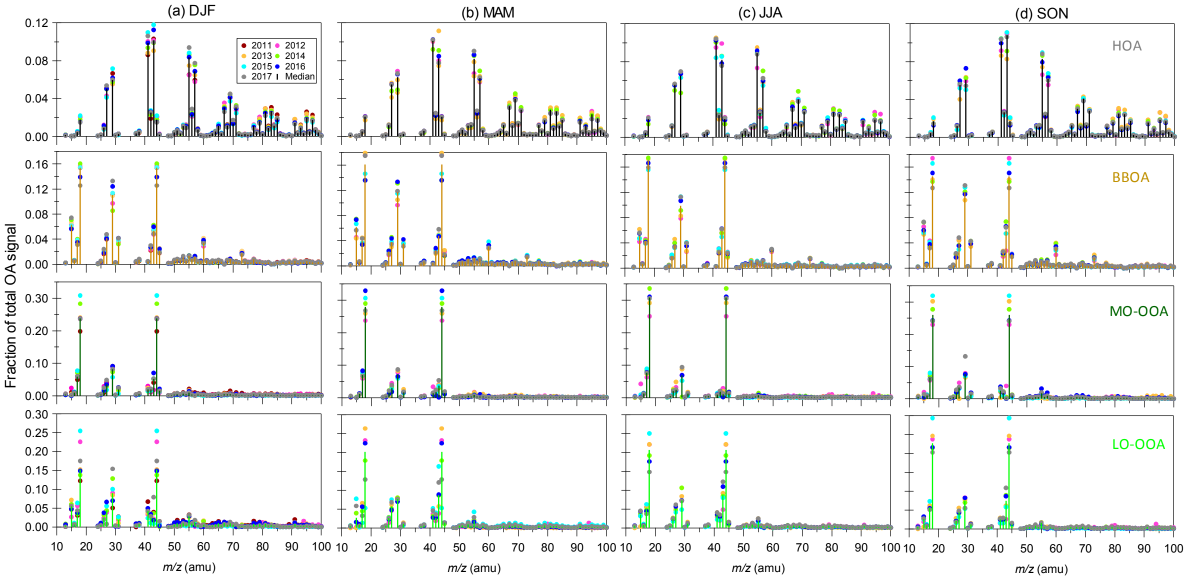

Figure S9. Seasonal mass spectral profiles of the four PMF OA factors determined from winter 2011-2012 to winter 2017-2018.

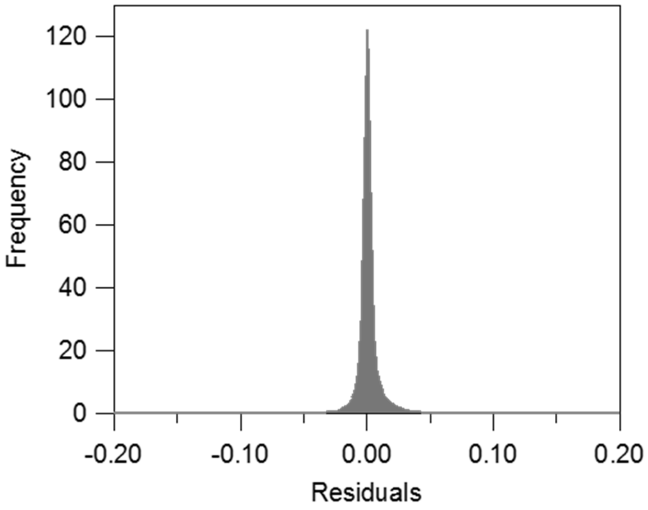

Figure S10. Residual distributions of PMF OA analysis using ME-2 approach (random mode with *a*-value 0 – 0.4) for the entire investigated period.

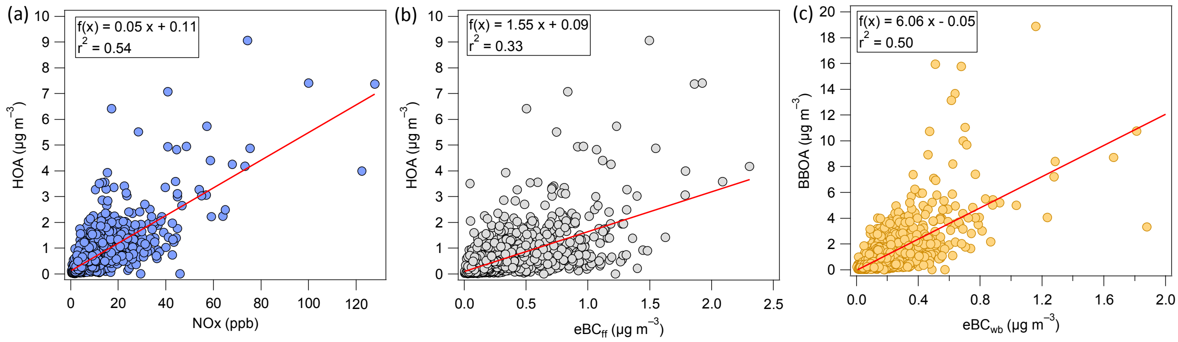

Figure S11. Correlations of POA factors with tracers. (a) HOA versus NOx, (b) HOA versus eBC$_{ff}$, and (c) BBOA versus eBC$_{wb}$. The data points refer to daily mean values during the entire period.

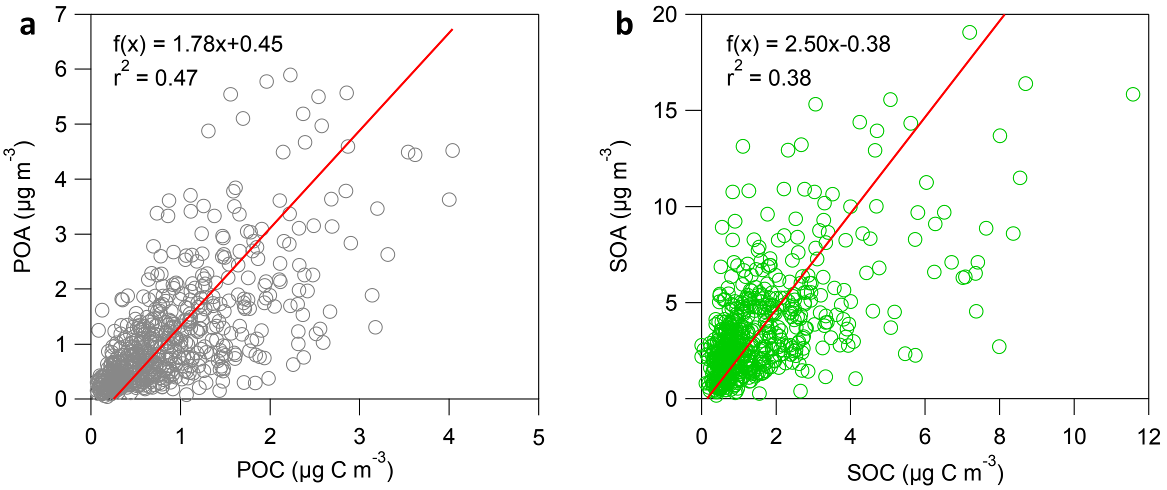

Figure S12. Intercomparison between POA (resp. SOA) and POC (resp. SOC), where POC and SOC were calculated using the OC/EC mass ratio method. The seasonal minimum OC/EC mass ratios – DJF (2.45), MAM (1.32), JJA (1.52), and SON (1.46) – were assumed being from primary combustion source emissions. Such OC/EC ratio method to has been widely applied to isolate POC and SOC from total OC by numerous previous studies (e.g., Srivastava et al., 2018, and references therein).

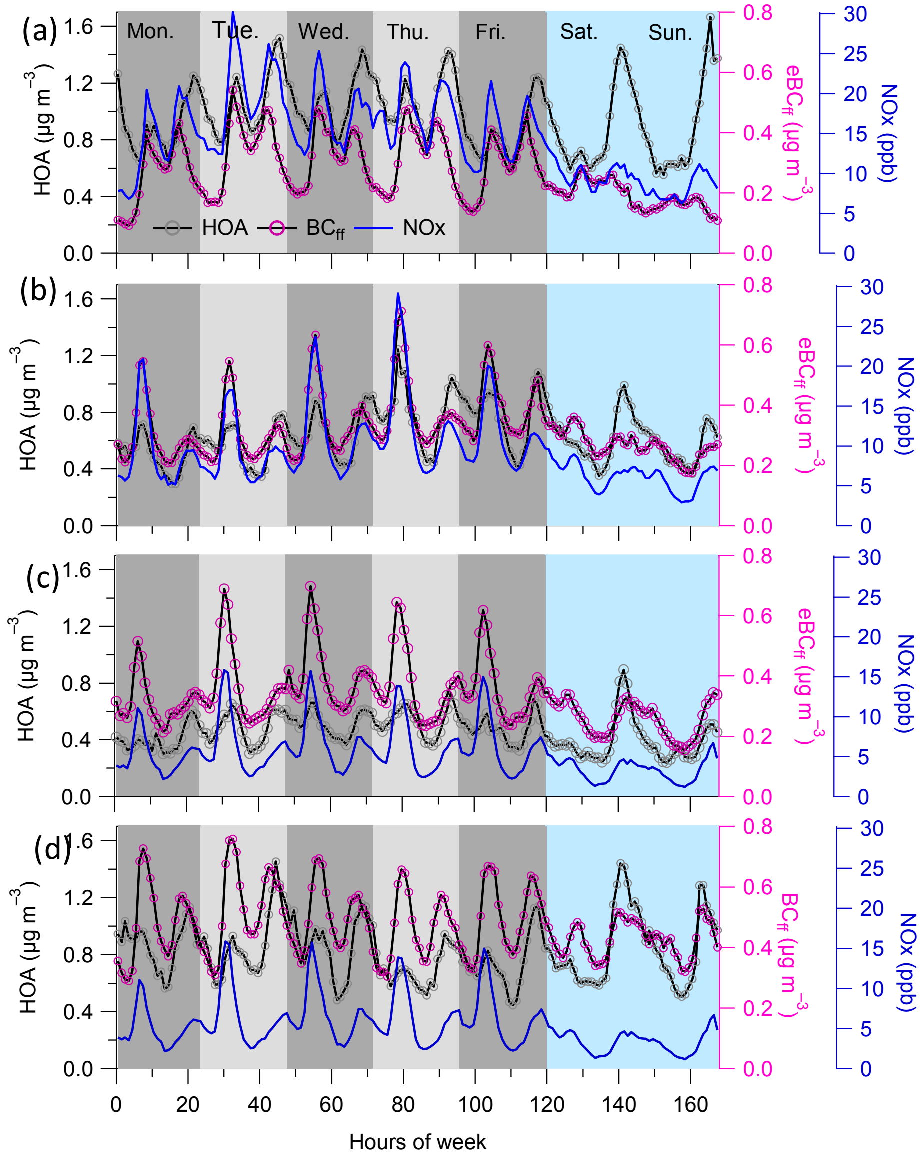

Figure S13. Seasonal weekly diel cycles obtained for HOA along with external tracers (eBC$_{ff}$ and NO$_x$) in (a) winter, (b) spring, (c) summer, and (d) fall.

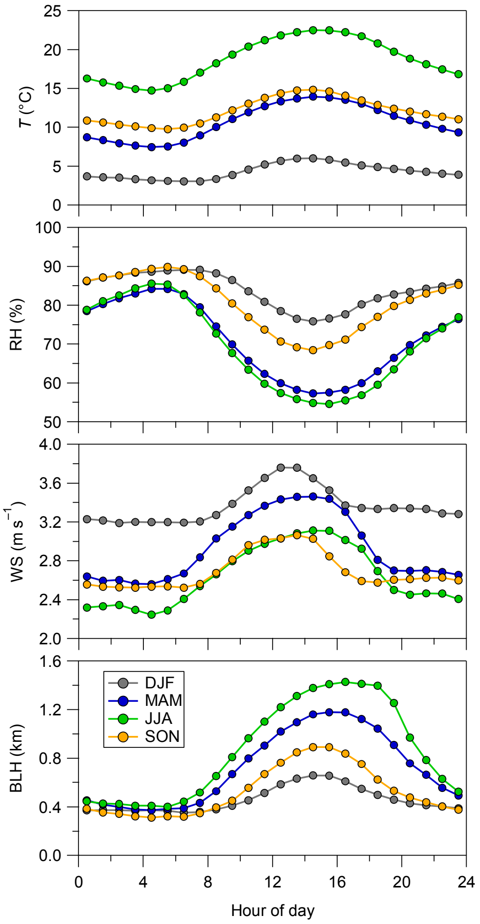

Figure S14. Diel cycles of meteorological parameters (including T, RH, WS, and BLH) in each season, i.e., winter (DJF), spring (MAM), summer (JJA), and fall (SON).

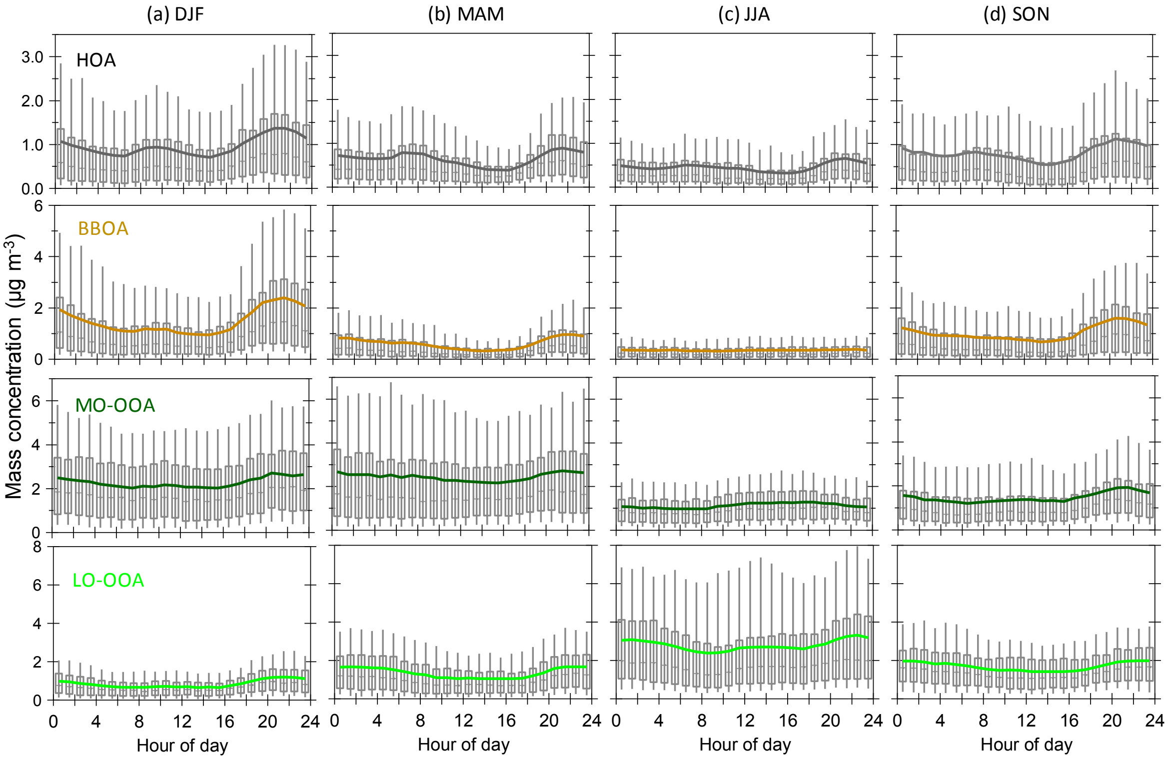

Figure S15. Average diel variations of the four ME-2 OA factors in each season.

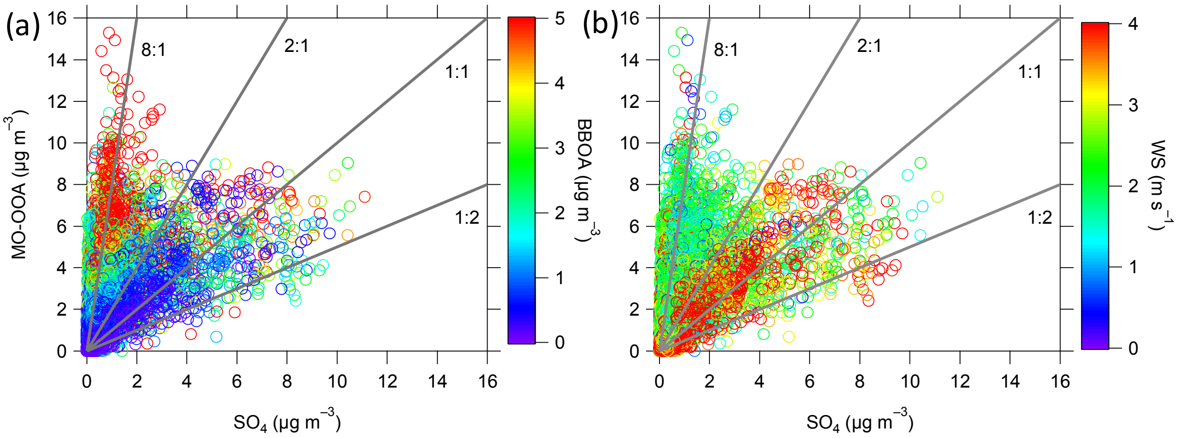

Figure S16. Relationship between MO-OOA and sulfate (SO$_4$) during wintertime, where the data points are colored by (a) mass concentration of BBOA; and (b) wind speed (WS).

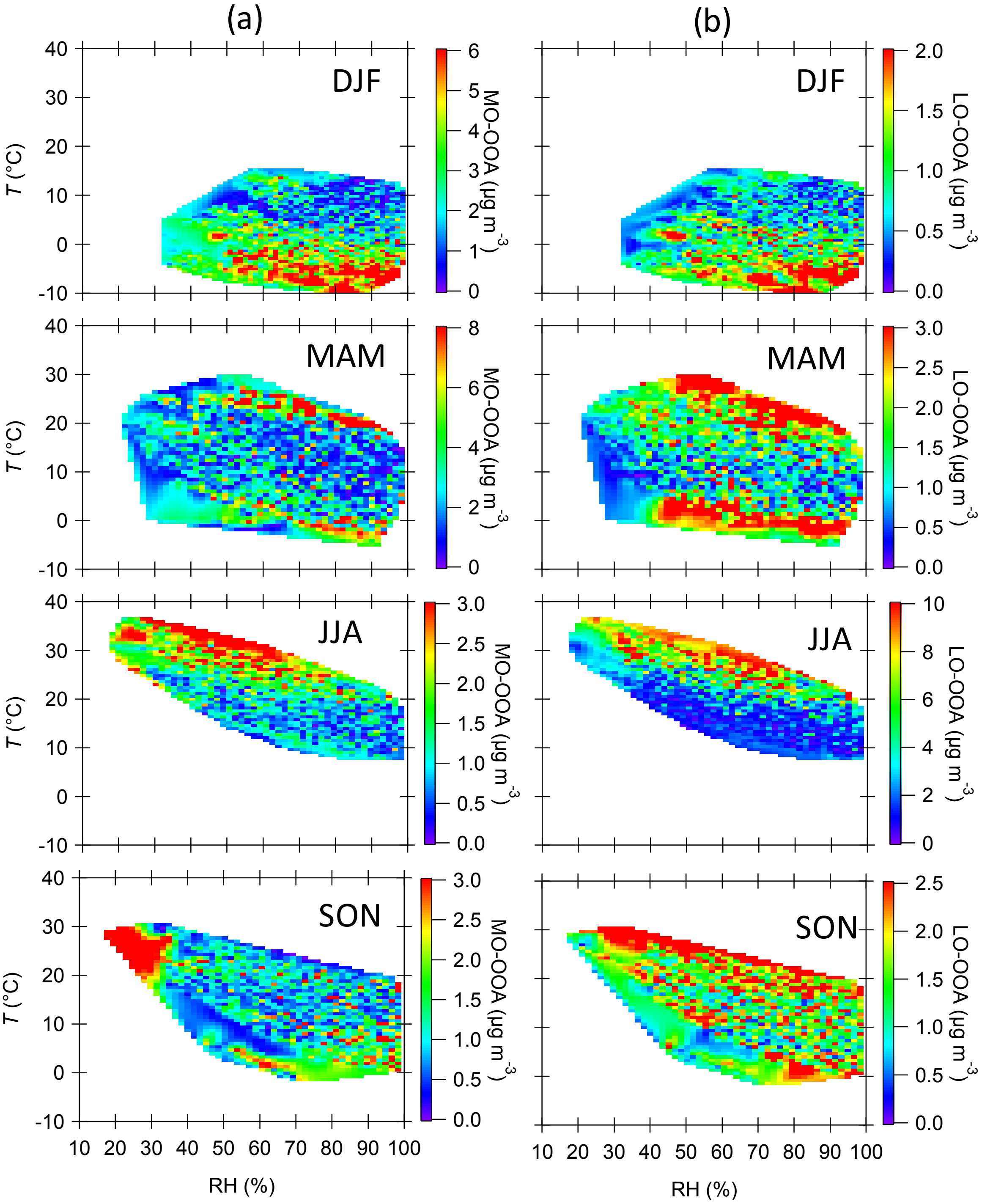

Figure S17. Temperature (*T*) and relative humidity (RH) dependence variations of the mass loadings of two OOA fractions.

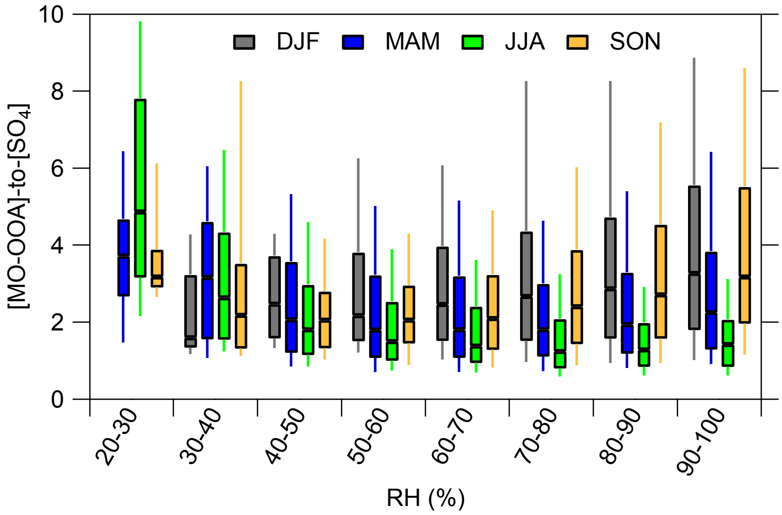

Figure S18. [MO-OOA]-to-[SO_4] ratio as a function of RH during each season.

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