



Supplement of

Time-dependent source apportionment of submicron organic aerosol for a rural site in an alpine valley using a rolling positive matrix factorisation (PMF) window

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Table of Contents

S1	Determination of Collection Efficiency (CE).....	3
S2	Black carbon measurement and source apportionment	5
S3	Rolling PMF analysis.....	7
S3.1	Factor analysis of the organic mass spectra.....	7
S3.2	Preparations and settings for Rolling PMF with ME-2	9
S3.2.1	Seasonal PMF <i>pre-tests</i>	9
S3.2.2	Bootstrap seasonal PMF analysis.....	11
S3.2.3	PMF Window settings.....	12
S3.2.4	Criteria settings	13
S3.3	Definition of “good” PMF runs using the <i>t</i> -test	15
S4	Optimised time window size.....	20
	References	23

S1 Determination of Collection Efficiency (CE)

The CE value typically depends on the particulate water content (Matthew et al., 2008), ammonium nitrate mass fraction (ANMF) and acidity (Middlebrook et al., 2012). We installed a Nafion membrane dryer (Perma Pure MD) in front of the sampling inlet to minimise humidity effects on CE. In addition, more than 93.5% of data have an ANMF smaller than 0.4; only 6.5% of data would be influenced by a time dependent CE correction. Therefore, the ANMF did not significantly affect CE for this dataset. Nevertheless, **Fig. S1a** showed good agreements between the ACSM data corrected with a constant CE of 0.45 and the SO_4^{2-} , NO_3^- , and Cl^- anions from $\text{PM}_{2.5}$ filter samples (with R^2 of 0.83, 0.82, and 0.50, respectively). It also showed relatively good consistencies with the anions measured using chromatography from Mini-denuder (MD) (Dämmgen et al., 2010) samples (**Fig. S1b**). Besides, when adding the equivalent black carbon (eBC) concentration to the corrected ACSM data (CE=0.45), this reconstructed mass agreed well with external TEOM measurement of both $\text{PM}_{2.5}$ and PM_{10} daily mass concentrations with R^2 of 0.81 and 0.67, respectively (**Fig. S1c**). In addition, it (CE=0.45) had a slightly better correlation and a slope closer to 1 to these external measurements than the time-dependent CE corrected data as suggested by Middlebrook et al. (2012). Therefore, we used a constant CE value of 0.45 to quantify ACSM data.

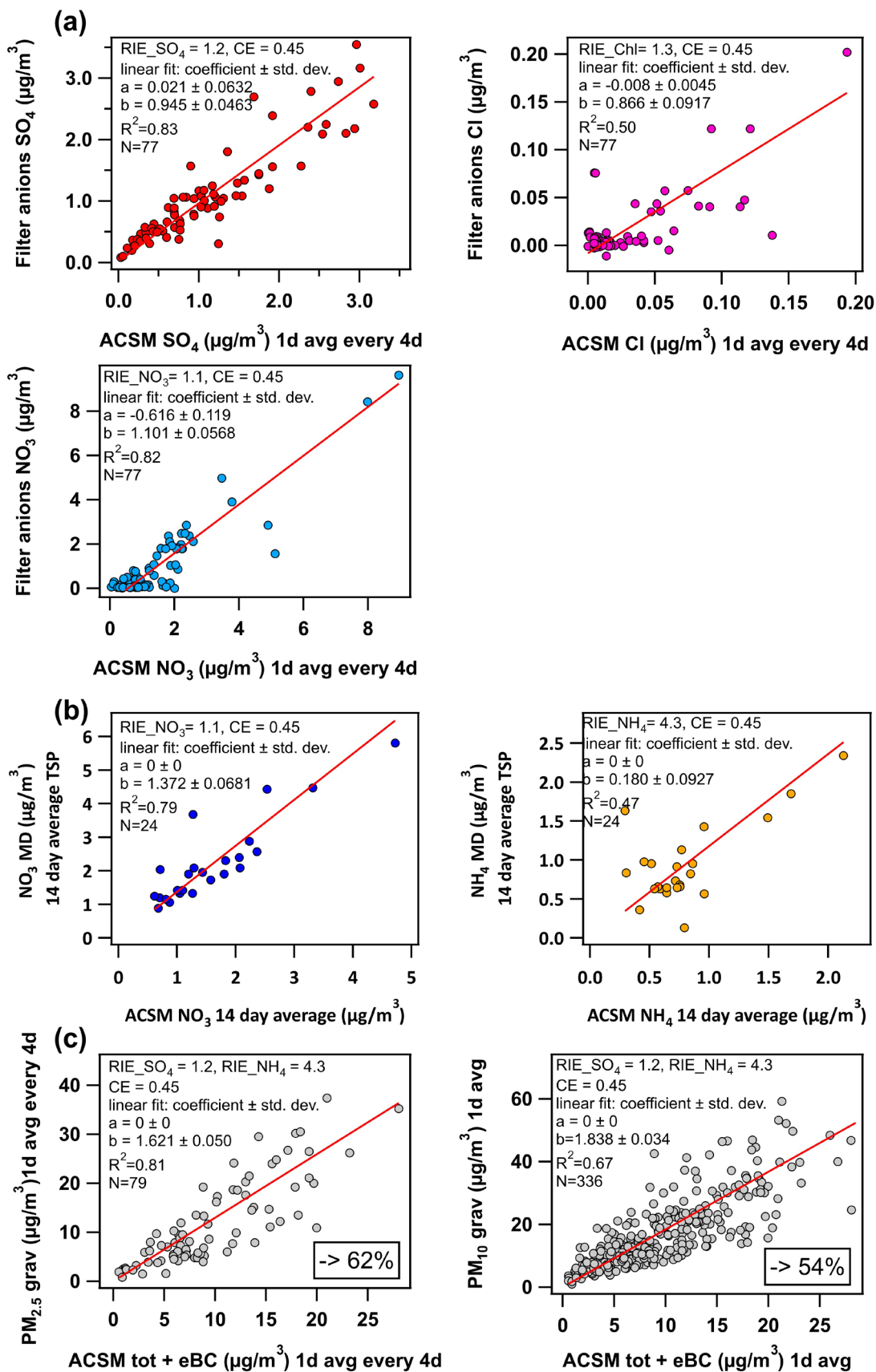


Fig. S1 Mass closure analysis of the dataset. (a) Linear correlations between the filter anions SO_4^{2-} , NO_3^- and Cl^- and the corresponding ASCM inorganic species. (b) Correlations of the NO_3^- and NH_4^+ concentrations measured with mini-denuders and by the ACSM. (c) Correlations of $\text{PM}_{2.5}$ and PM_{10} with NR- PM_1 defined as the sum of the total ACSM mass plus the equivalent black carbon concentration.

S2 Black carbon measurement and source apportionment

The aethalometer (AE 31 model by Magee Scientific Inc.) measures equivalent black carbon (eBC) concentrations via light transmission through a sample spot at multiple wavelengths ($\lambda = 370, 470, 520, 590, 660, 880, \text{ and } 950 \text{ nm}$). In this study, we installed a $\text{PM}_{2.5}$ cyclone and a Nafion dryer (Perma Pure MD) in front of the sampling inlet that the AE31 and the ACSM shared. The light absorption coefficients b_{abs} were calculated by correcting the measured attenuation coefficients for the filter loading effect (Weingartner et al., 2003). To convert the optical absorption to the eBC mass concentration, eBC_{tot} into $\mu\text{g m}^{-3}$ (Petzold et al., 2013), the absorption coefficient at a given wavelength λ , $b_{\text{abs}}(\lambda)$ was divided by the corresponding aerosol mass absorption cross-section $\sigma_{\text{abs}}(\lambda)$ in $\text{m}^2 \text{ g}^{-1}$ (Weingartner et al., 2003):

$$\text{eBC}_{\text{tot}} = b_{\text{abs}}(\lambda) / \sigma_{\text{abs}}(\lambda) \quad (1)$$

with $\sigma_{\text{abs}}(470) = 22.9 \text{ m}^2 \text{ g}^{-1}$ and $\sigma_{\text{abs}}(950) = 8.8 \text{ m}^2 \text{ g}^{-1}$, as previously reported for Magadino (Herich et al., 2011).

The light absorption coefficients measured at wavelengths $\lambda_1 = 470 \text{ nm}$ and $\lambda_2 = 950 \text{ nm}$ were used to retrieve the relative contributions of traffic (eBC_{tr}) and wood burning (eBC_{wb}) to the total

equivalent black carbon mass concentration eBC_{tot} (Herich et al., 2011; Sandradewi et al., 2008; Zotter et al., 2017). The two-component model implies that at a given wavelength λ the absorption coefficient b_{abs} is approximated by the sum of the absorption coefficients of eBC emitted from traffic exhaust $b_{abs,tr}$ and from wood burning $b_{abs,wb}$ (Eq. (2)), which in turn depend on λ through Eq. (3) and Eq.(4):

$$b_{abs}(\lambda) = b_{abs,tr}(\lambda) + b_{abs,wb}(\lambda) \quad (2)$$

$$\frac{b_{abs,tr}(\lambda_1)}{b_{abs,tr}(\lambda_2)} = \left(\frac{\lambda_1}{\lambda_2}\right)^{-\alpha_{tr}} \quad (3)$$

$$\frac{b_{abs,wb}(\lambda_1)}{b_{abs,wb}(\lambda_2)} = \left(\frac{\lambda_1}{\lambda_2}\right)^{-\alpha_{wb}} \quad (4)$$

The Ångstrom exponents for eBC from traffic $\alpha_{tr} = 0.9$ and wood burning $\alpha_{wb} = 1.68$ sources were chosen in accordance with Zotter et al. (2017) suggested for the same sampling site, Magadino.

Note that despite of utilising the aethalometer corrections proposed in Weingartner et al. (2003), the eBC data were not fully free of filter loading artefacts, as evidenced by a discontinuity in $b_{abs}(\lambda)$ measurements on filter tape advancement. Since the filter loading effect is more pronounced at shorter wavelengths due to higher attenuation (Drinovec et al., 2015; Weingartner et al., 2003), b_{abs} measured at 470nm will have more intense signals. As a result, when high eBC loadings triggered more frequent filter advances for winter days, artificial peaks appeared in the time series of apportioned eBC_{wb} . However, when averaging data points for the eBC diurnal cycles that we

used to validate positive matrix factorisation (PMF) solutions, transient peaks due to the filter loading artefacts had negligible effects.

S3 Rolling PMF analysis

S3.1 Factor analysis of the organic mass spectra

PMF has been demonstrated to be a useful tool to retrieve the sources of measured organic aerosol mass spectra with a bilinear factor model (Paatero and Tapper, 1994; Ulbrich et al., 2009):

$$x_{ij} = \sum_{k=1}^p g_{ik} \times f_{kj} + e_{ij} \quad (5)$$

where x_{ij} is the mass concentration of the j^{th} mass spectral variable in the time point i^{th} ; g_{ik} is the contribution of the k^{th} factor in the i^{th} time point; f_{kj} is the concentration of the j^{th} mass spectral variable in the k^{th} factor; and e_{ij} is the residual of j^{th} variable of the mass spectra in i^{th} time point. The superscript, p represents the number of factors, which the user determines. The cost function of PMF uses least-squares algorithm by iteratively minimising the following quantity Q :

$$Q = \sum_{i=1}^n \sum_{j=1}^m \left(\frac{e_{ij}}{\sigma_{ij}} \right)^2 \quad (6)$$

where σ_{ij} is an element in the $n \times m$ matrix of the measurement uncertainties, which corresponds point-by-point to x_{ij} . In addition, we normalised the quantity $\frac{Q}{Q_{exp}}$ as a mathematical metric during PMF analysis, where the Q_{exp} is:

$$Q_{exp} = (n \times m) - p \times (n + m) \quad (7)$$

The $\frac{Q}{Q_{exp}}$ supports the user to determine the number of factors required for the model by investigating the effects on this quantity of adding/removing a factor. However, PMF itself suffers from rotational ambiguity because the object function, Q does not provide unique solutions, that is, when $\mathbf{G} \cdot \mathbf{F} = \mathbf{G} \cdot \mathbf{T} \cdot \mathbf{T}^{-1} \cdot \mathbf{F}$, PMF provides a similar value of Q but very different solutions (rotated matrix $\bar{\mathbf{G}} = \mathbf{G} \cdot \mathbf{T}$ (rotated factor time series) and $\bar{\mathbf{F}} = \mathbf{T}^{-1} \cdot \mathbf{F}$ (rotated factor profiles)). Only one or even none of these rotated solutions may be atmospherically relevant. The ME-2 solver (Paatero, 1999) enables theoretically complete rotational control over the factor solutions, which is implanted here by imposing constraints via the a -value approach on one or more elements of \mathbf{F} and/or \mathbf{G} (Paatero and Hopke, 2009). The a -value (ranging from 0 to 1) determines how much the resulting factor ($f_{j,solution}$) or time series ($g_{j,solution}$) can vary from the input reference factor ($f_{j,reference}$) or time series ($g_{j,reference}$) as shown in Eq.(8) and Eq.(9):

$$f_{j,solution} = f_{j,reference} \pm a \cdot f_{j,reference} \quad (8)$$

$$g_{j,solution} = g_{j,reference} \pm a \cdot g_{j,reference} \quad (9)$$

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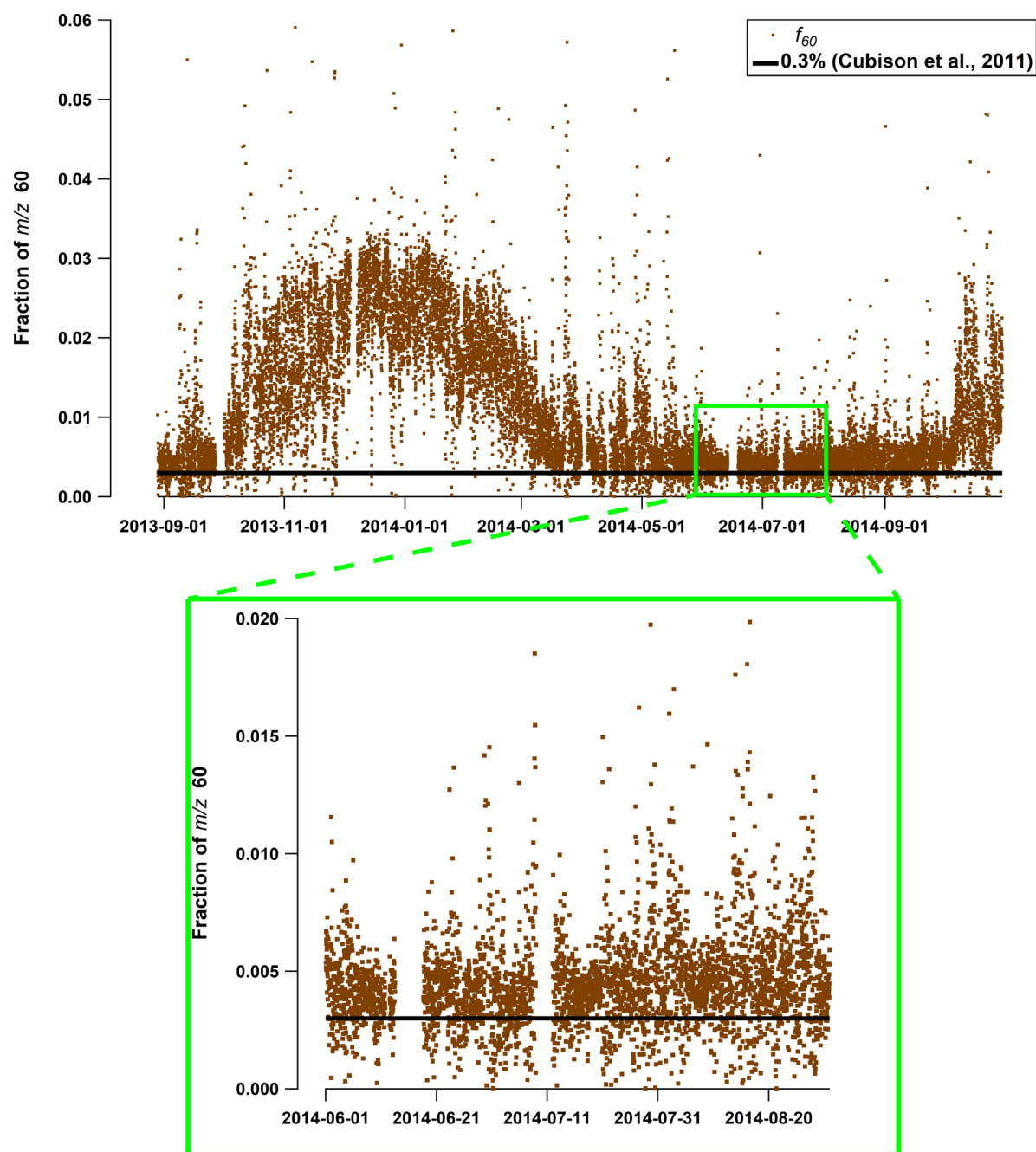
123 Previous work using α -values has shown to retrieve environmentally reasonable PMF solutions
124 efficiently. The presence of legitimate *a priori* constraints decrease the degree of rotational
125 ambiguity (Canonaco et al., 2013, 2021; Crippa et al., 2014; Lanz et al., 2008). Here we configured
126 the ME-2 solver and analysed PMF results using SoFi (Source Finder, Datalystica Ltd., Villigen,
127 Switzerland) Pro 6.D interface (Canonaco et al., 2013, 2021), developed within the IGOR Pro
128 software (WaveMetrics Inc., Lake Oswego, OR, USA).

129 **S3.2 Preparations and settings for Rolling PMF with ME-2**

130 **S3.2.1 Seasonal PMF *pre-tests***

131 To understand the potential sources over different seasons in Magadino, PMF *pre-tests* were
132 conducted based on different seasons. It provides information about the potential number of factors
133 in different seasons, which is necessary before the rolling PMF analysis. In addition, the PMF
134 solutions from rolling PMF analysis tend to be more robust if the reference profiles used to
135 constrain are retrieved from seasonal PMF analysis. Thus, site-dependent reference profiles are
136 necessary (at least for BBOA) to get more accurate estimations of OA sources (i.e., better
137 correlation with external tracers in this study compared to the PMF solution using literature
138 reference profiles). In this study, the whole dataset was divided into five parts based on months
139 (i.e., DJF represents the winter season during December, January, and February; MAM represents
140 the spring season during March, April, and May, etc.). A preliminary “good” PMF solution (so-
141 called base case) can be obtained for each season by following the guidelines provided by Crippa
142 et al. (2014).

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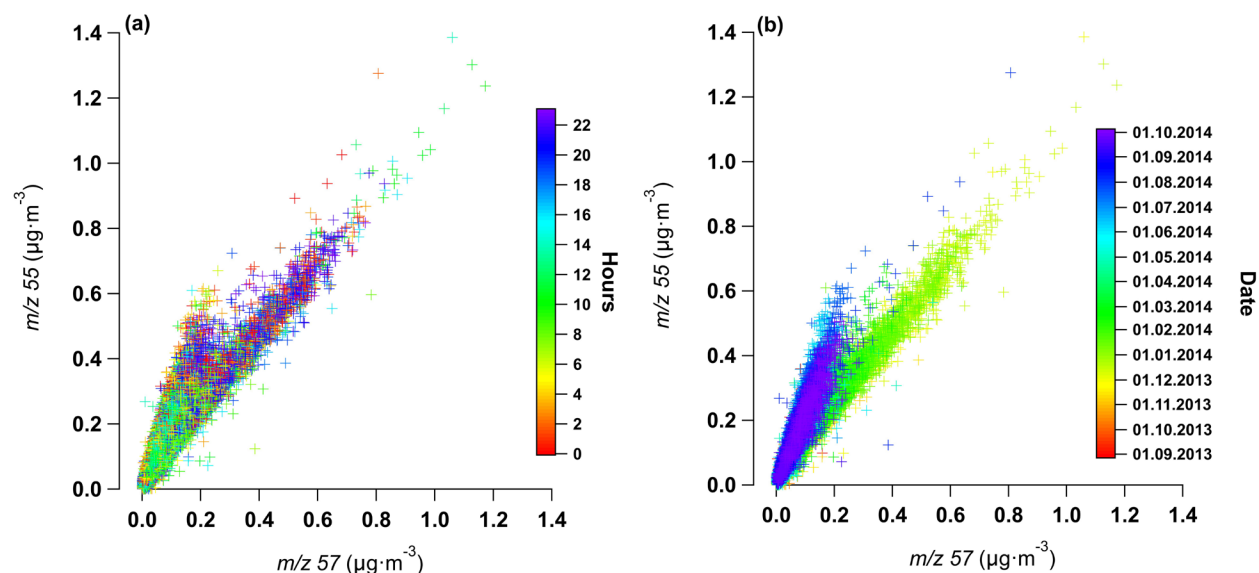


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Fig. S2 Time series of the measured fraction of mass-to-charge ratio (m/z) of 60 .

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149 **Fig. S3** Measured absolute mass concentrations of *mass-to-charge ratio* (m/z)=55 and m/z =57
 150 with colour coded by hours of the day (a) and date and time (b).

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152 S3.2.2 Bootstrap seasonal PMF analysis

153 In order to get stable reference profiles, the bootstrap resampling technique was applied in this
 154 study to test the stability of the base cases from seasonal PMF *pre-tests*. The bootstrap resampling
 155 randomly chooses a subset of the original matrix and replicates some rows/columns to create a
 156 new matrix with the same size (Efron, 1979). Given sufficient bootstrapped runs (>100) can
 157 provide the statistical uncertainty of the PMF solutions.

158 First, the primary factor profiles (hydrocarbon-like OA factor (HOA), BBOA) were retrieved from
 159 preliminary tests during seasonal PMF runs, while an additional mass-to-charge ratio (m/z) 58
 160 related OA (58-OA) factor was obtained in summer, then 1000 PMF runs were conducted for each
 161 season by constraining the POA factor profiles using random *a*-values with a step of 0.1 and
 162 ranging from 0 to 0.5. We used the same criterion list as the base case (as shown in **Table S1**) and

a novel technique, t -test (Section 3.3) to define “good” PMF runs. Then, from the averaged bootstrapped PMF solutions (**Fig. S4**), the reference profiles can be obtained for rolling PMF analysis.

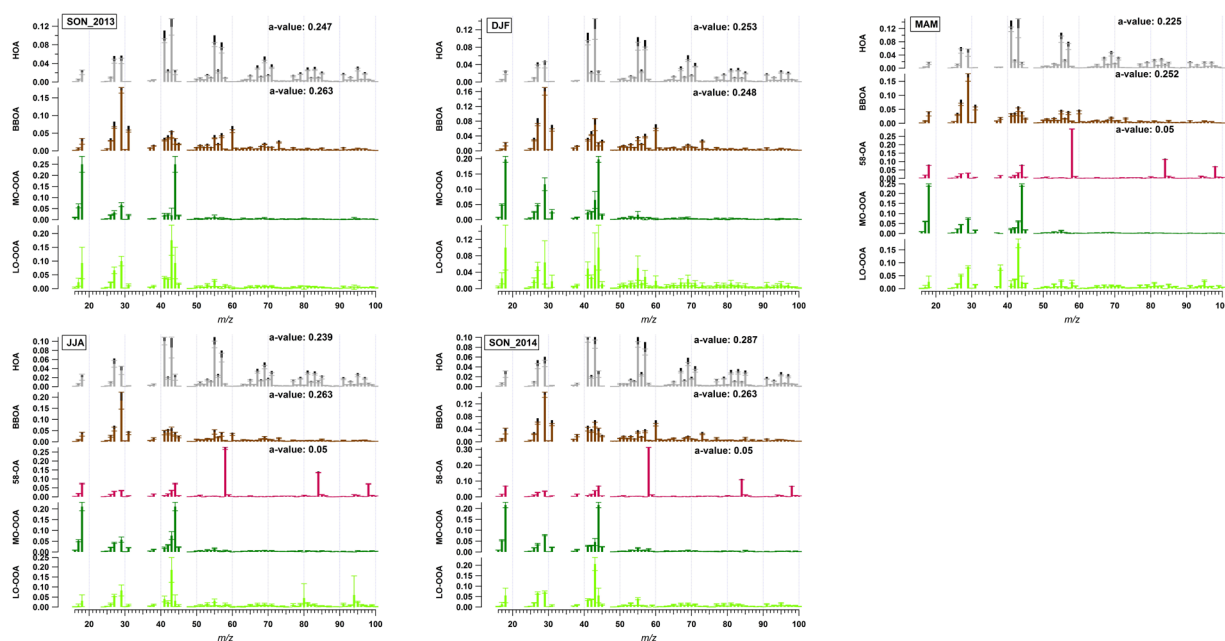


Fig. S4 Averaged factor profiles from seasonal bootstrap solutions for five different periods. The error bars of each factor represent the standard deviation of the averaged bootstrapped solution, the thick dark sticks are the variabilities that each variable allowed to vary with the corresponding averaged a -value. SON = September, October and November, DJF = December, January and February, MAM = March, April, and May, JJA = June, July, and August.

S3.2.3 PMF Window settings

In order to retrieve appropriate constraints, we performed PMF *pre-tests* and bootstrap analysis for different seasons. Here, we constrained primary OA factor profiles (hydrocarbon-like OA (HOA) and biomass burning OA (BBOA)) as well as the factor profile of the 58-OA using the a -value technique in the rolling PMF analysis. The reference profiles of HOA and BBOA were from the winter bootstrapped PMF solution (December, January, and February), as shown in **Fig. S4**. With

a higher contribution of the biomass burning trace ion m/z 60 in the winter, we expect a more representative and robust BBOA profile from the winter solution than from the other seasons. The 58-OA profile was retrieved from the summer bootstrapped PMF solution (June, July, and August) (Fig. S4). To allow the factor profile to adapt itself over time, we applied an a -value randomly from a set of a -values, including 0, 0.1, 0.2, 0.3, and 0.4 (so-called random a -value approach). Canonaco et al. (2021) suggested that an upper a -value of 0.4 is sufficient to cover the temporal variation of OA source profiles. Moreover, due to the uniqueness of the 58-OA chemical profile, it was tightly constrained with a constant a -value of 0.05.

In total, we constrained the HOA and BBOA factors with a random a -value (0–0.4, with a step of 0.1) and an exact a -value (0.05) for the 58-OA factor in the rolling PMF analysis. There are 25 (5×5) possible a -value combinations within an individual rolling window. Therefore, 50 PMF iterations for each time window are sufficient to cover all possibilities of the a -value combinations. With the rolling window of 50 repeats, each data point (except the data within the first and last time window) will actually have many PMF iterations (i.e., length of the window×50), where bootstrap resampling and random combinations of constraints is performed. It allows to estimate the statistical and rotational uncertainties of the PMF factors (Canonaco et al., 2021). To find the optimum length of the time windows, we tested four different lengths of the time windows (1, 7, 14, and 28 days) using the same approaches as in Canonaco et al. (2021). We determined the optimum length of the time window based on the number of missing data points (un-modelled data due to the selection based on the criteria) while applying the same thresholds for the same criteria.

S3.2.4 Criteria settings

Performing a rolling analysis for one-year data with 50 repeats per window requires tens of thousands of PMF runs. Manual inspection of all PMF runs is impractical and therefore was

replaced by monitoring user-defined criterion scores (Canonaco et al., 2021). In this study, R^2 values of the time series of modelled HOA vs NO_x and eBC_{tr} were used for HOA. The BBOA factor was inspected using the variation of $m/z=60$ explained by BBOA (**Table S1**). For these time series based criteria (criterion 1 to criterion 3 in **Table S1**), we deployed a student t -test to minimise subjective judgments while determining the thresholds (more discussions in Section 3.3 of this document).

Typically, OOA factors are dominated by the signals of f_{43} ($\text{C}_2\text{H}_3\text{O}^+$ at $m/z = 43$) and f_{44} (CO_2^+ at $m/z = 44$) that correspond to the less and more oxygenated ion fragments (Crippa et al., 2014; Ng et al., 2010), where f is the fraction of a variable, *i.e.* the intensity $I_{m/z}$ normalised by the sum of the intensities of all organic m/z variables. In this study, we were able to retrieve two OOA factors (*i.e.*, more oxidised OOA (MO-OOA) and less oxidised OOA (LO-OOA)) for the whole year. Since we left two factors unconstrained (4th and 5th factor), MO-OOA can be either at the 4th or the 5th position in these 20750 runs. Thus, we used the f_{44} for the 4th factor to sort the unconstrained OOA factors to ensure MO-OOA and LO-OOA sitting on the 4th and the 5th position, respectively. The details of the sorting scheme can be found in Canonaco et al. (2021). At the same time, we also monitored f_{43} in LO-OOA and f_{44} in MO-OOA to make sure they are not zero. With this set of criteria, we were able to only select “good” (atmospherically relevant) PMF runs before averaging.

S3.2.4.1 Explained variation (EV) of $m/z = 60$ by BBOA

The uncertainties of the aethalometer model for eBC source apportionment are very high when the mass concentration of eBC_{wb} is small (Harrison et al., 2013), which was the case in summer 2014. Thus, the summer BBOA factor was poorly correlated with eBC_{wb} . In this work, we used the

variation of $m/z = 60$ explained by BBOA to justify the summer solution, which is calculated using Eq. (10) (Paatero, 2010):

$$EV_{j,k} = \frac{\sum_{i=1}^n (|g_{ik} \cdot f_{kj}| / \sigma_{ij})}{\sum_{i=1}^n ((\sum_{h=1}^p |g_{ih} \cdot f_{hj}| + e_{ij}) / \sigma_{ij})} \quad (10)$$

Paatero (2010) suggested that if there is a dominant ion in a specific factor, it should explain more than 30-35% of variation of this measured variable. Canonaco et al. (2021) used an EV of 0.25 at $m/z=60$ for BBOA as a threshold to select “good” runs for BBOA. In this study, we only selected PMF runs with EV of $m/z=60$ for BBOA that were significantly larger than those of other factors by t -test with a p -value ≤ 0.05 . In the end, the $EV_{60,BBOA}$ values for selected PMF runs for both seasonal and rolling results are all larger than 0.4.

Table S1 Criterion List for both seasonal and rolling PMF.

	Criterion	Type	Threshold
1	HOA vs NO _x	$R_{pearson}^2$, normal time series	p -value ≤ 0.05
2	HOA vs eBC _{tr}	$R_{pearson}^2$, normal time series	p -value ≤ 0.05
3	$EV_{60,BBOA}$	Average, normal time series	p -value ≤ 0.05
4	factor_4[44]	Profiles, fraction, sorting criterion	>0
5	factor_5[43]	Profiles, fraction	>0

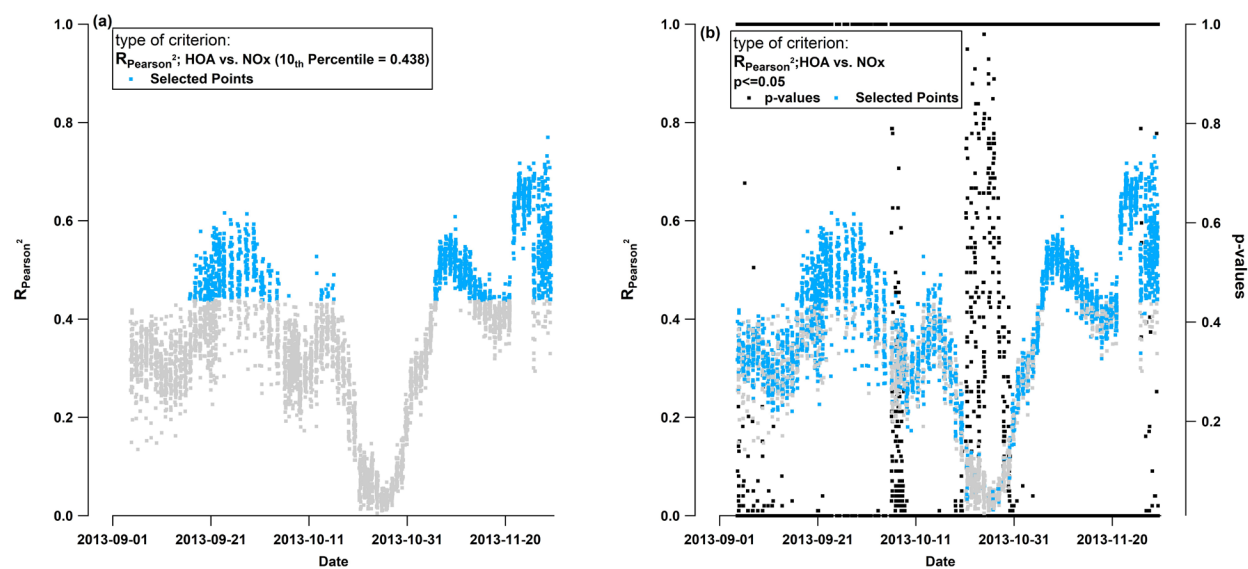
S3.3 Definition of “good” PMF runs using the t -test

The conventional PMF analysis remains subjective on how to define “good” (environmentally reasonable) PMF runs. In this study, we tried to use the criteria-based approach to have a quantitative analysis (e.g., correlations between the time series of a PMF factor and corresponding external tracers, intensities of key ions of the PMF factor profile) on all PMF runs as suggested by Canonaco et al. (2021). However, it is still subjective to decide the lower limit for “good” PMF runs, as Canonaco et al. (2021) suggested.

Canonaco et al. (2021) proposed to define thresholds of criteria for the rolling PMF runs based on the seasonal PMF analysis. For instance, for the criterion of the R^2 -Pearson between NO_x vs HOA, SoFi Pro can resample the time series of both the BBOA factor (from averaged seasonal bootstrapped solutions) and NO_x by bootstrap. It then uses the resampled time series to conduct correlation analysis, which provides systematic statistic metrics, including mean, median, minimum, maximum, and 10th/90th percentile, probability distribution function, etc. Canonaco et al. (2021) proposed to use the 10th percentile as the lower limit of the criteria in the rolling PMF analysis. However, it remains subjective when the user defines the thresholds for the “good” seasonal solutions. In addition, there could also be a dilemma when the thresholds are too strict to allow sufficient data coverage in the end. As shown in **Fig. S5a**, the 10th percentile ($R^2=0.438$) caused a high rejection rate for the majority of data points in fall 2013. This is potentially due to the fact that the resampling size for the seasonal solution during bootstrap of criteria is not small enough, therefore, the resampled correlations appeared to be not representative for the rolling solution.

In this study, we proposed a new technique to minimise subjective judgements from the user. We use the student t -test with the null hypothesis of un-correlation between the two variables (e.g., R^2 of the time series of modelled HOA vs NO_x). For other typical criteria that are based on temporal information (e.g., explained variation of $m/z = 60$ for BBOA), we used the null hypothesis of $EV_{60,BBOA}$ being not larger than EV_{60} of all other factors. With these t -tests, we utilised the p -value to filter out “bad” PMF runs. The statistical significance level of a p -value ≤ 0.05 was applied for criterion 1, 2, and 3 (**Table S1**) to define “good” solutions with minimum subjective judgements. In addition, compared with the 10th percentile technique Canonaco et al. (2021) proposed, the t -test approach typically would accept more data points as illustrated in **Fig. S5b**.

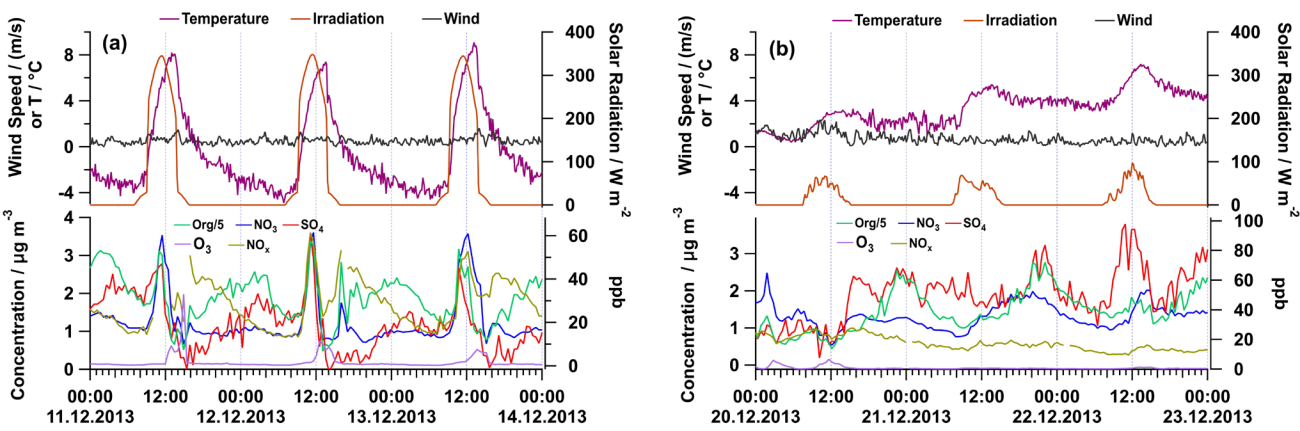
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267 **Fig. S5** Score plot the criterion for the R^2 of HOA vs NO_x in rolling PMF for fall, 2013.

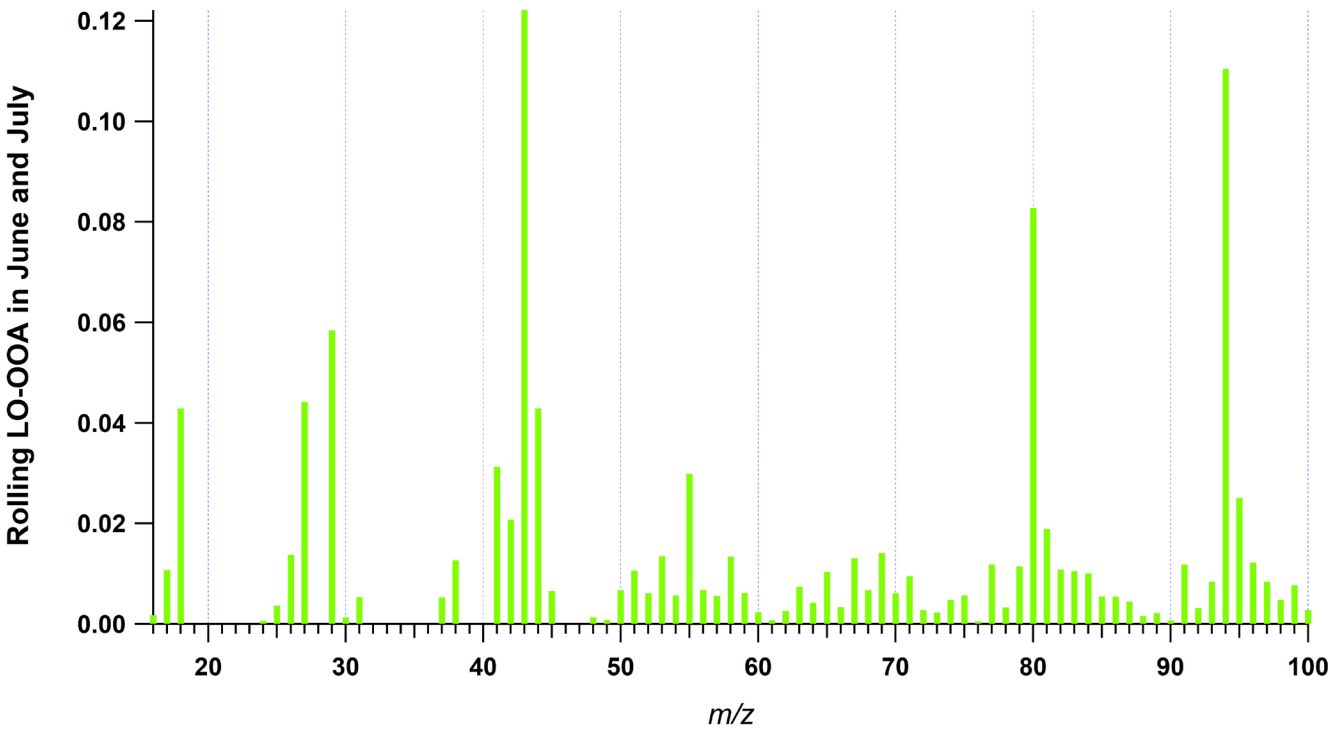
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271 **Fig. S6** Diurnal cycles of the organic (Org), NO₃, SO₄, O₃, NO_x, and corresponding meteorological
272 data on sunny (a) and cloudy (b) days in winter. On sunny days (a), a transport phenomenon was
273 observed in the noontime which caused a sharp enhancement of pollutants, followed by a
274 breakthrough of the boundary layer resulting in simultaneous dilution for all pollutants. Also, the
275 delay of the irradiation peak is because the monitoring station lies in the shadow of surrounding
276 mountains during the winter season. (b) No such situation was observed during cloudy days which
277 indicates that indeed irradiation and temperature gradient might play a role in this phenomenon.

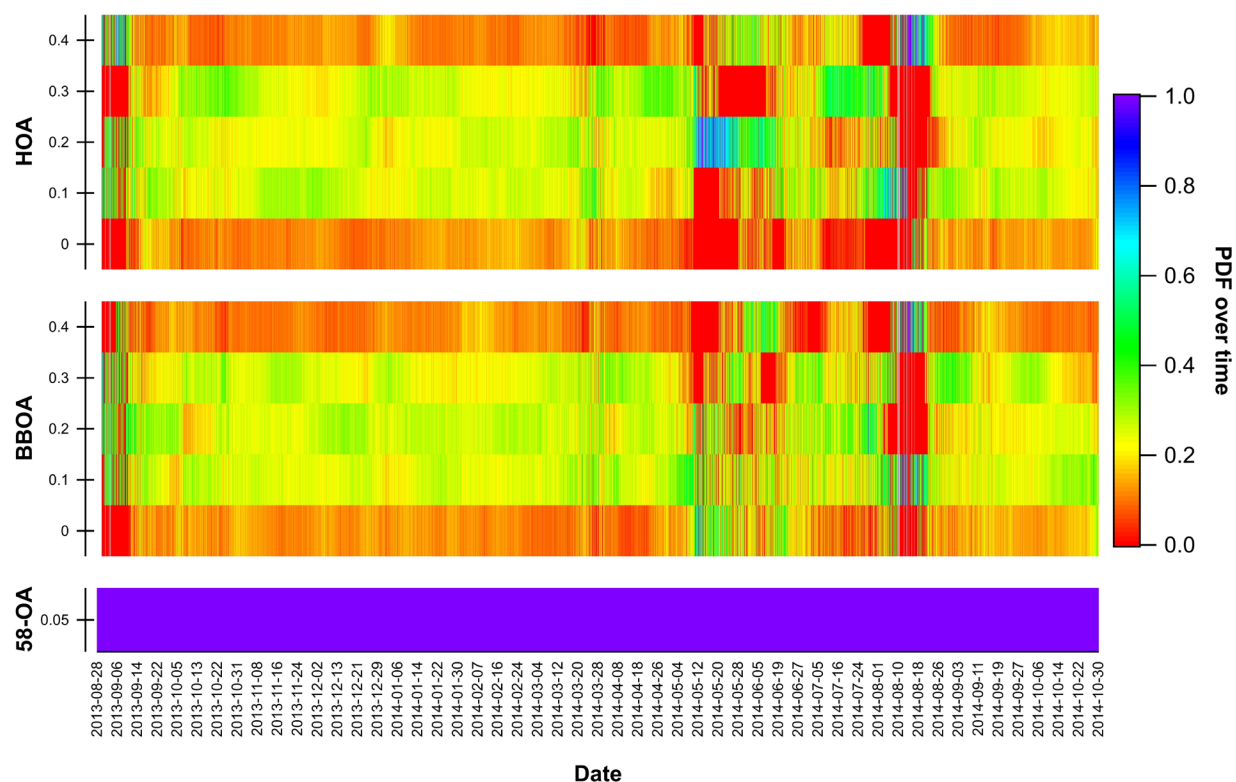
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280 **Fig. S7** Mass spectra for LO-OOA in June and July from the rolling results.

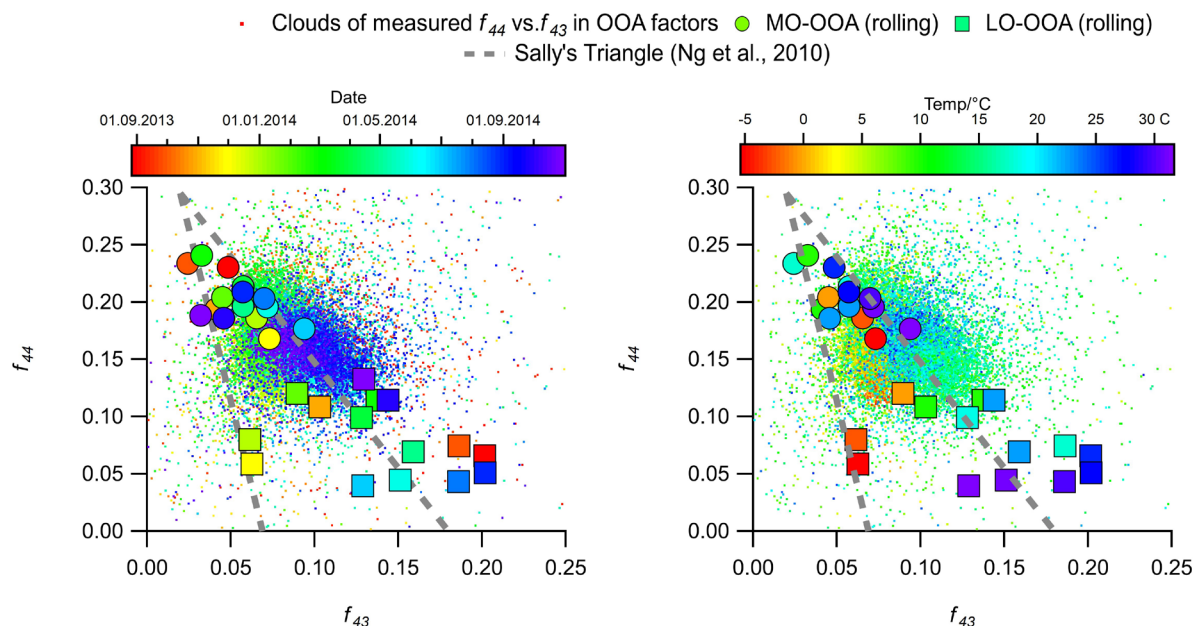
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283 **Fig. S8** The probability distribution function (PDF) of employed α -values of selected PMF runs
 284 for constrained factors as a function of time.

285



286
287 **Fig. S 9** f_{44} vs. f_{43} for OOA factors (after subtraction of signals contributed by the primary HOA,
288 BBOA and 58-OA factors as shown in Eq. (S11) and (S12)) in monthly resolution, colour coded
289 by month (left) and temperature (right).

290

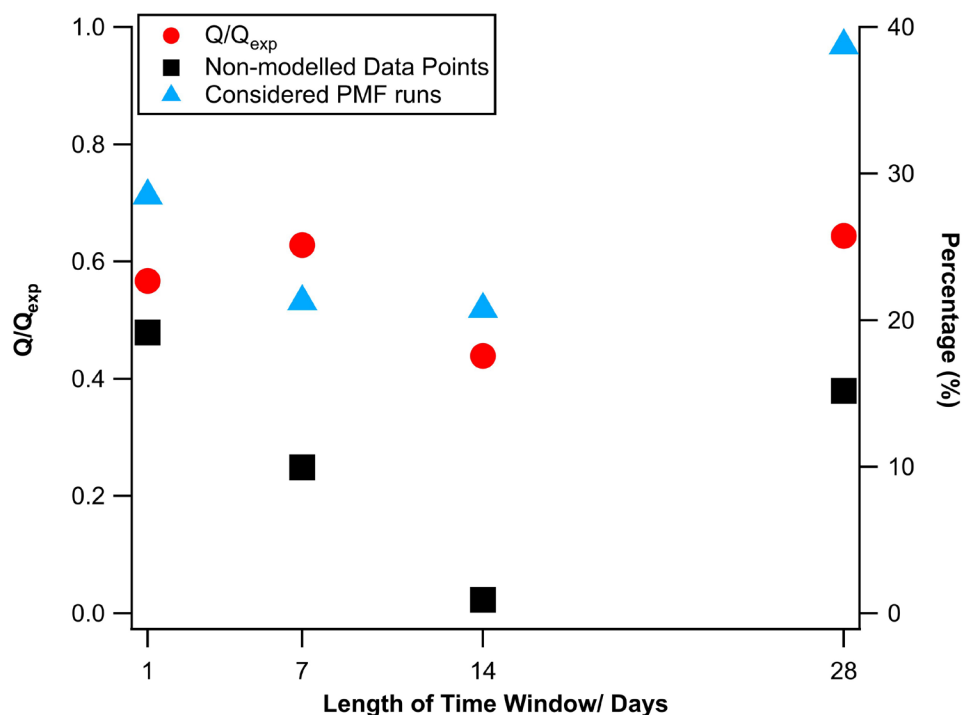
$$\text{subtracted } f_{44} = \frac{\text{mass conc. of OOA @}[m/z 44]}{\text{mass conc. of OOA + residual of total OA}} \quad (11)$$

$$\text{subtracted } f_{43} = \frac{\text{mass conc. of OOA @}[m/z 43]}{\text{mass conc. of OOA + residual of total OA}} \quad (12)$$

291 S4 Optimised time window size

292 We tested different time window size (1, 7, 14, and 28 days) and compared the solutions by
293 applying the same thresholds for the same criteria. We found the optimum window sizes for this
294 dataset to be 14 days, with only 29 (0.15%) non-modelled points (due to the criteria-based
295 selection) as shown in **Fig. S10**. The averaged Q/Q_{exp} for different time window sizes are similar,
296 but the 14-day window solution still has the smallest Q/Q_{exp} (0.448). However, the Q/Q_{exp} for all
297 window sizes are smaller than one, which is likely due to the high uncertainty from the

298 measurement of the ACSM (27/67 variables have signal to noise ratio (S/N) <2), and SoFi
 299 simplifies the equation of Q_{exp} to $n \times m$ because $n \times m \gg p \times (n+m)$ when measured points are
 300 sufficiently large. Nevertheless, we selected and presented the 14-day window solution in this
 301 manuscript with its significantly smaller number of missing (non-modelled) points in the model.



302
 303 **Fig. S10** Non-modelled time points (due to criteria-based selection) and Q/Q_{exp} vs rolling
 304 window size.

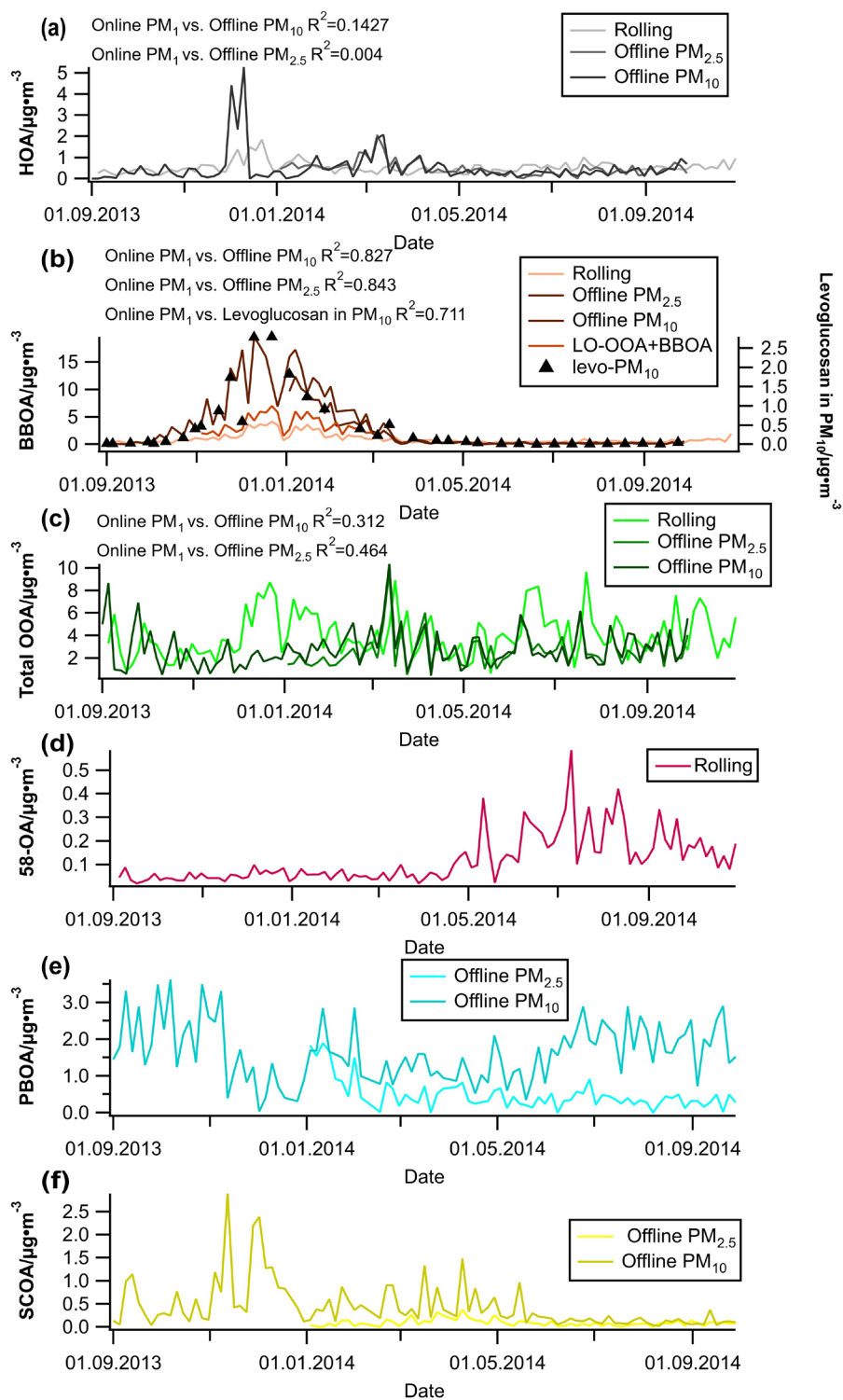


Fig. S11 The comparison between source apportionment results from offline AMS PM₁₀/PM_{2.5} samples and online ACSM.

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