S1. PMF solutions to determine the number of sources.

The six-factor solution (figure S1.c) shows two split factors (dark blue and green) which correspond to the same source, LVOOA. The five-factor solution (Figure S1.b) shows to be the more acceptable number of sources, with the following sources observed: BBOA, HOA, COA, SVOOA and LVOOA.

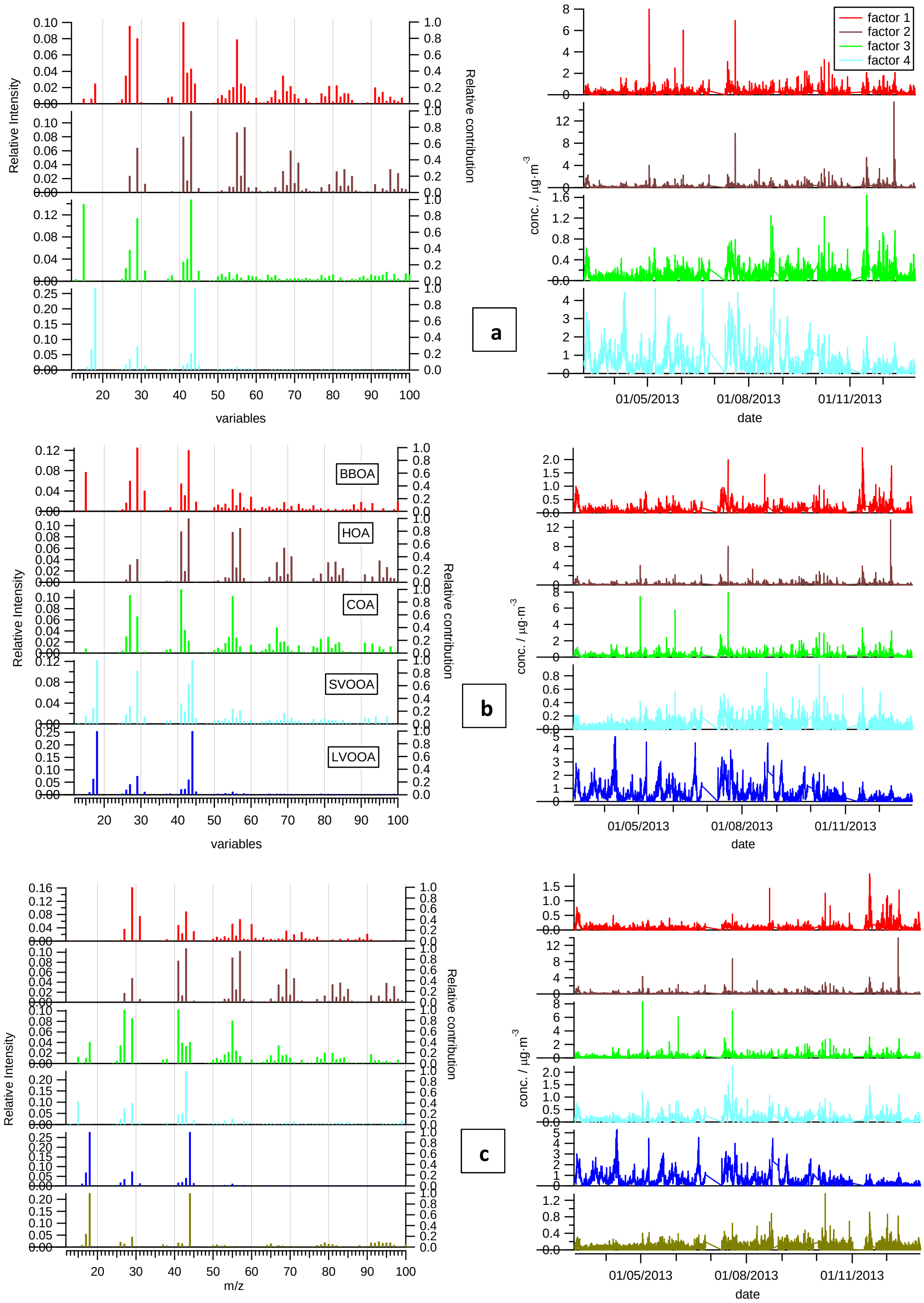


Figure S1: PMF solutions: four-factor solution (a), five-factor solution (b) and six-factor solution.

S2. Seed and mass spectral analysis.

In order to deal with rotational ambiguity, ME-2 runs may be initialised from different random values, also called seeds. Figure S2 shows the analysis carried out to the three different seeds from the best solution chosen for March-December (aB3_H2_C3) to determine stability on the solutions. This stability proves that solutions may be repeatable with the three solutions presenting the same five factors with similar Q/Qexp (S2.a), mass spectrum (S2.b) and time series (S2.c).

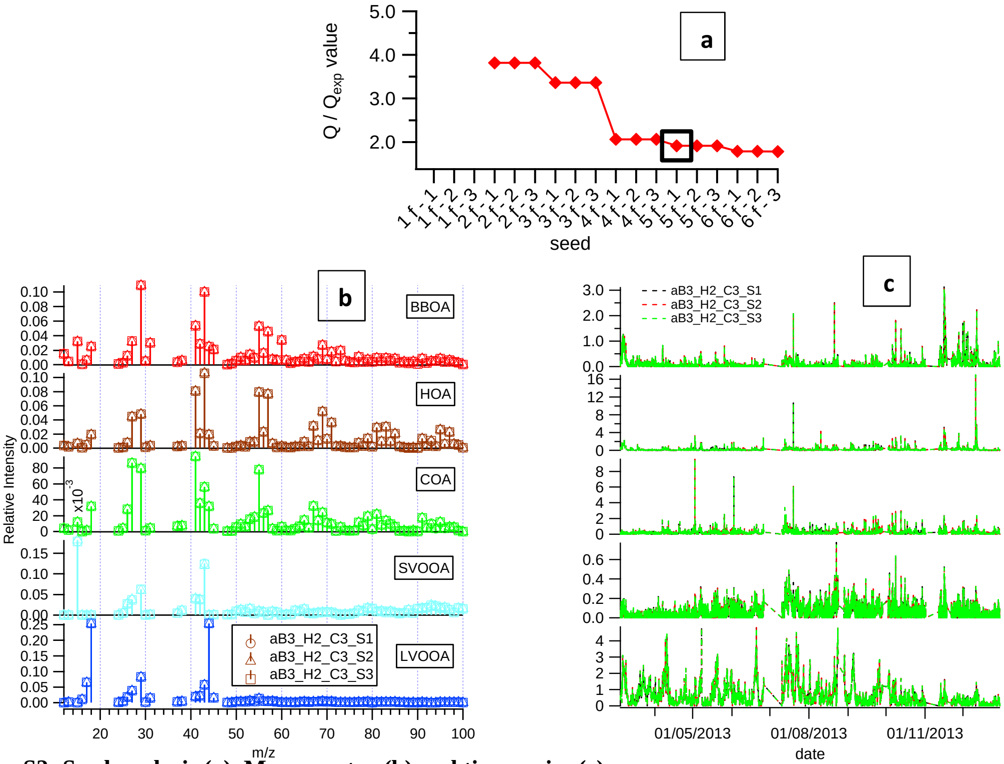


Figure S2: Seed analysis (a). Mass spectra (b) and time series (c).

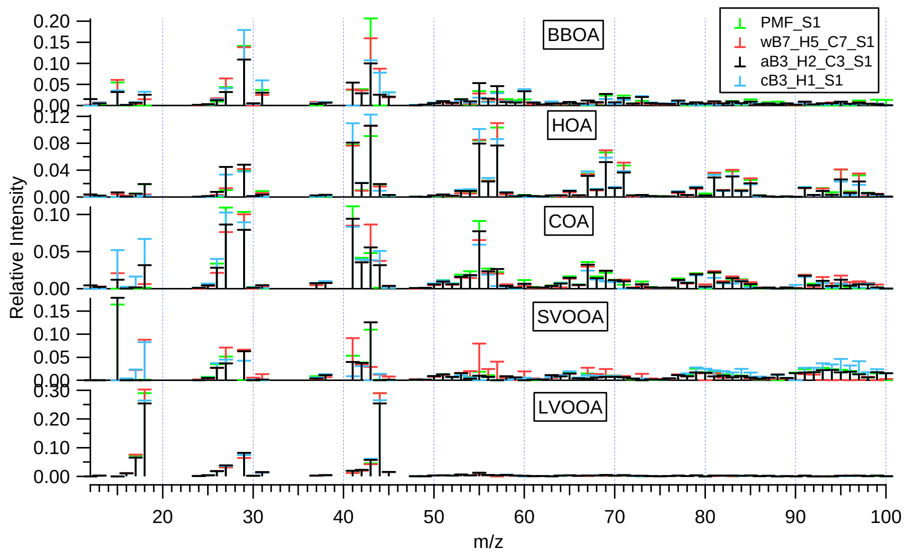
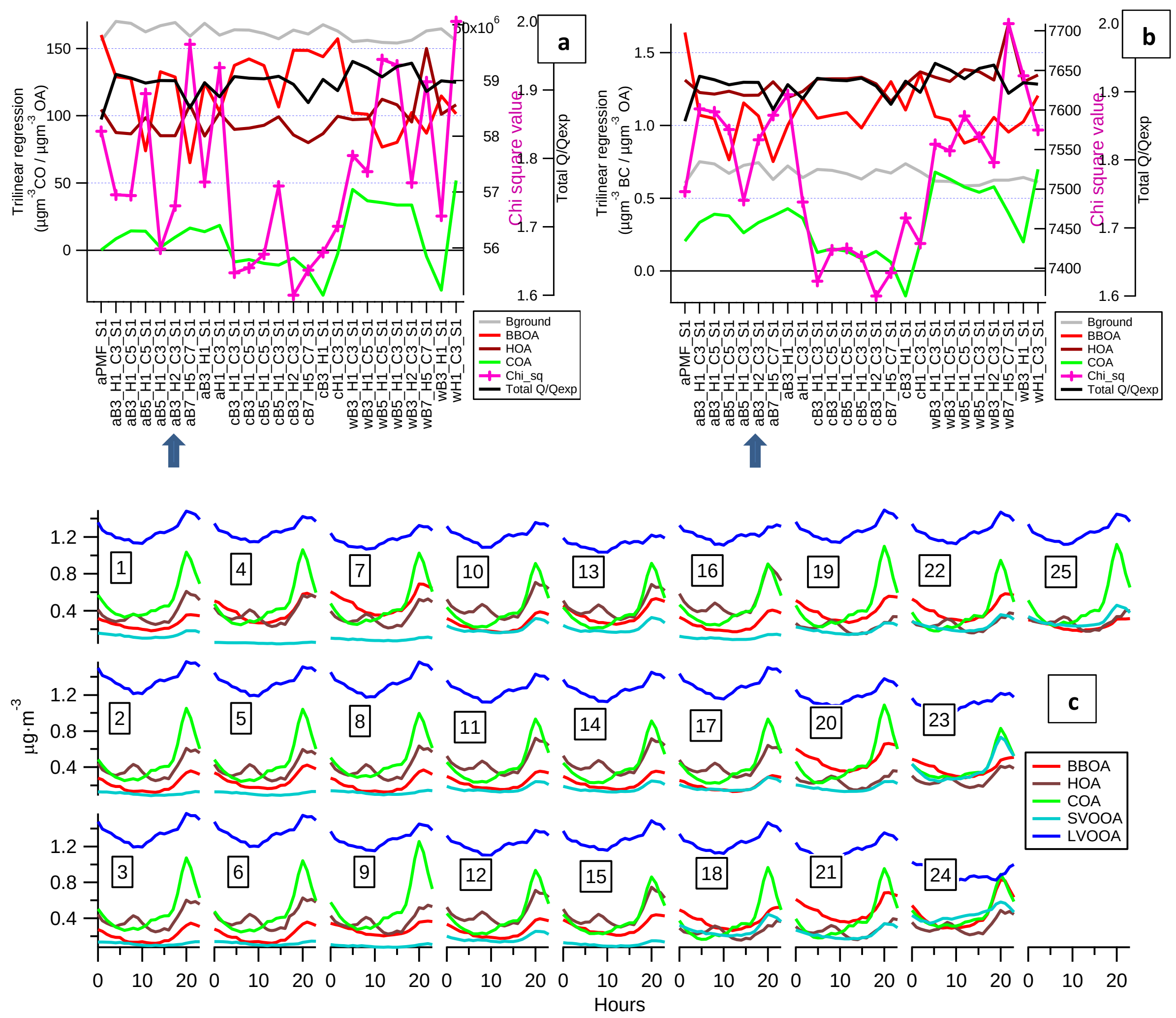


Figure S3: Mass spectra comparison for undesirable solutions for March-December analysis. Example of mass spectra of solutions with mixed factors for unconstrained and contrained solutions.

S3. Analysis to determine the best solution for the different periods of time.

Figure S4 shows the analysis carried out to determine the best solution for the March-December period. As mentioned in the main text of this paper, “c” and “w” target profiles show the less desirable results, “c” target profiles show a high positive residual around 14:00-19:00 hrs. (Figure S4.b) and “w” target profiles show a high chi-square and total $Q/Q_{exp.}$ (Figures C1.a and S4.b). The solution aB3_H2_C3_S1 is chosen to present the best results from this analysis.

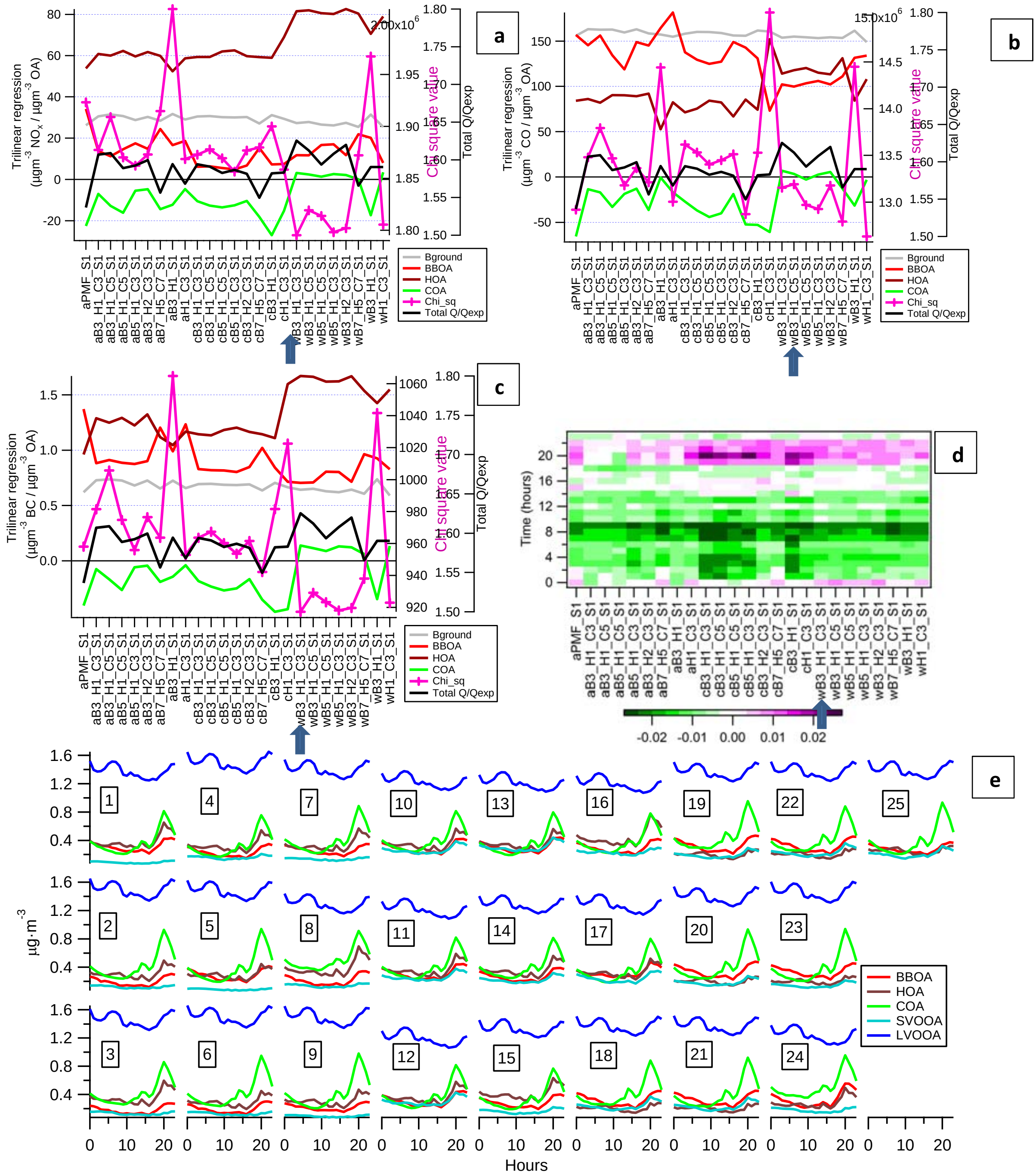


1	aPMF_S1	6	aB3_H2_C3_S1	11	cB3_H1_C5_S1	16	cB3_H1_S1	21	wB5_H1_C3_S1
2	aB3_H1_C3_S1	7	aB7_H5_C7_S1	12	cB5_H1_C5_S1	17	cH1_C3_S1	22	wB3_H2_C3_S1
3	aB3_H1_C5_S1	8	aB3_H1_S1	13	cB5_H1_C3_S1	18	wB3_H1_C3_S1	23	wB7_H5_C7_S1
4	aB5_H1_C5_S1	9	aH1_C3_S1	14	cB3_H2_C3_S1	19	wB3_H1_C5_S1	24	wB3_H1_S1
5	aB5_H1_C3_S1	10	cB3_H1_C3_S1	15	cB7_H5_C7_S1	20	wB5_H1_C5_S1	25	wH1_C3_S1

d

Figure S4: CO and BC trilinear regression (a, b), diurnal concentrations (c) and all solutions for March-Dec analysis (d).

Figure S5 shows the analysis carried out to determine the best solution for spring period. Solutions with "a" and "c" target profiles show the less desirable results with negative concentrations for COA in the trilinear regression analyses (Figures S5.a, S5.b and S5.c) and with high HOA concentrations during the evening (Figure S5.e) apparently mixed with COA. The solution wB3_H1_C3_S1 is chosen to present the best results from this analysis.

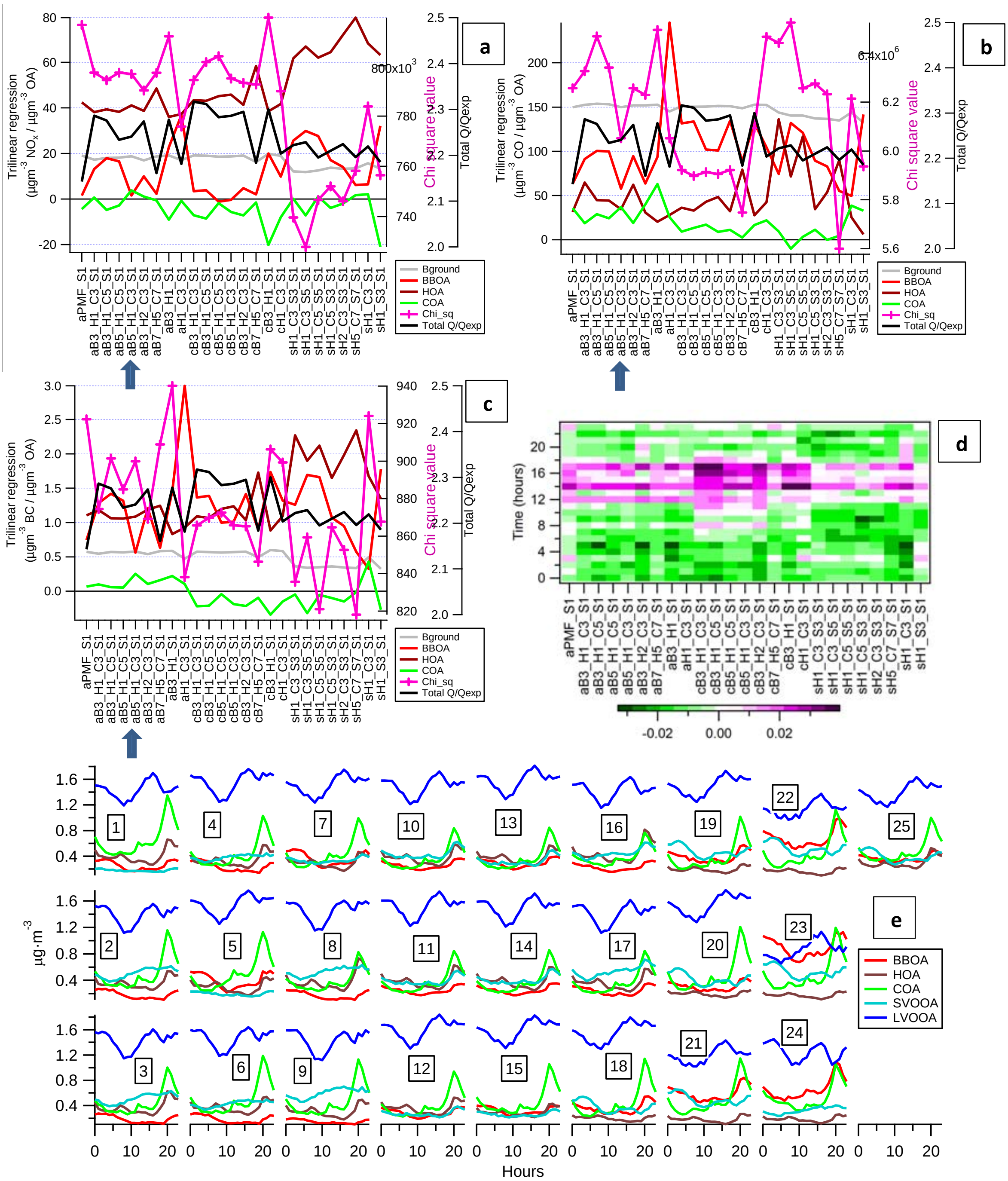


1	aPMF_S1	6	aB3_H2_C3_S1	11	cB3_H1_C5_S1	16	cB3_H1_S1	21	wB5_H1_C3_S1
2	aB3_H1_C3_S1	7	aB7_H5_C7_S1	12	cB5_H1_C5_S1	17	cH1_C3_S1	22	wB3_H2_C3_S1
3	aB3_H1_C5_S1	8	aB3_H1_S1	13	cB5_H1_C3_S1	18	wB3_H1_C3_S1	23	wB7_H5_C7_S1
4	aB5_H1_C5_S1	9	aH1_C3_S1	14	cB3_H2_C3_S1	19	wB3_H1_C5_S1	24	wB3_H1_S1
5	aB5_H1_C3_S1	10	cB3_H1_C3_S1	15	cB7_H5_C7_S1	20	wB5_H1_C5_S1	25	wH1_C3_S1

f

Figure S5: CO and BC trilinear regression (a,b,c), diurnal residual (d),diurnal concentrations (e) all the solutions for spring analysis (f).

Figure S6 shows the analysis carried out to determine the best solution for summer period. Solutions with “c” and “s” target profiles show the less desirable results. “s” target profiles show low chi-square values, however, they present high negative residuals in the morning and at night. “c” target profiles show a high positive residual around 15:00-18:00 hrs. The solution aB5_H1_C3_S1 is chosen to present the best results from this analysis.

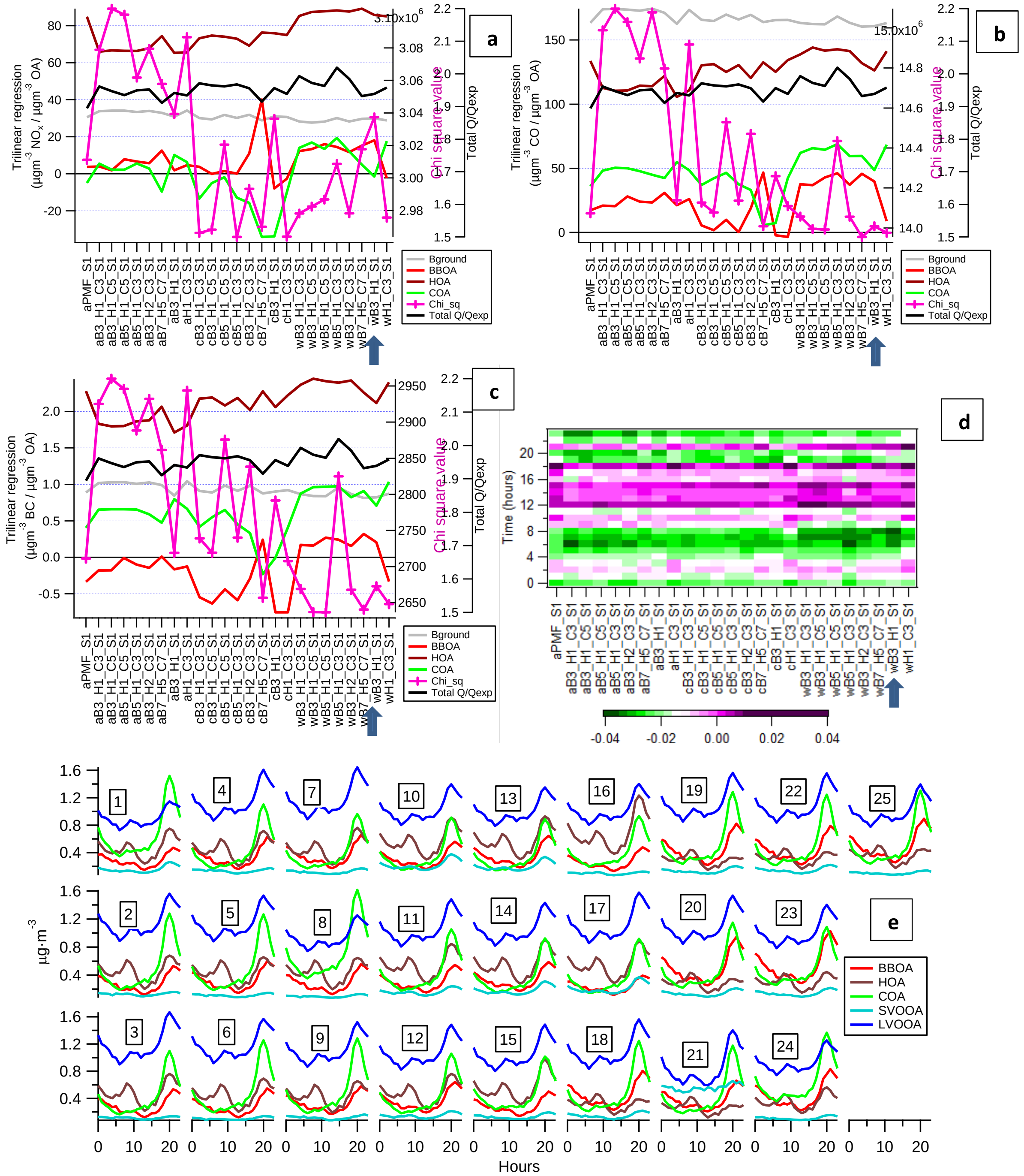


f

1	aPMF_S1	6	aB3_H2_C3_S1	11	cB3_H1_C5_S1	16	cB3_H1_S1	21	sH1_C5_S3_S1
2	aB3_H1_C3_S1	7	aB7_H5_C7_S1	12	cB5_H1_C5_S1	17	cH1_C3_S1	22	sH2_C3_S3_S1
3	aB3_H1_C5_S1	8	aB3_H1_S1	13	cB5_H1_C3_S1	18	sH1_C3_S3_S1	23	sH5_C7_S7_S1
4	aB5_H1_C5_S1	9	aH1_C3_S1	14	cB3_H2_C3_S1	19	sH1_C3_S5_S1	24	sH1_C3_S1
5	aB5_H1_C3_S1	10	cB3_H1_C3_S1	15	cB7_H5_C7_S1	20	sH1_C5_S5_S1	25	sH1_S3_S1

Figure S6: CO and BC trilinear regression (a,b,c), diurnal residual (d),diurnal concentrations (e) all the solutions for summer analysis (f).

Figure S7 shows the analysis carried out to determine the best solution for autumn period. Solutions with “a” target profiles show the less favourable Chi square results and “c” target profiles show negative BBOA slopes residuals in the morning (S7.d), also “a” target profiles show high chi-square values. wB3_H1_S1 solution is chosen to present the best results from this analysis.



1	aPMF_S1	6	aB3_H2_C3_S1	11	cB3_H1_C5_S1	16	cB3_H1_S1	21	wB5_H1_C3_S1
2	aB3_H1_C3_S1	7	aB7_H5_C7_S1	12	cB5_H1_C5_S1	17	cH1_C3_S1	22	wB3_H2_C3_S1
3	aB3_H1_C5_S1	8	aB3_H1_S1	13	cB5_H1_C3_S1	18	wB3_H1_C3_S1	23	wB7_H5_C7_S1
4	aB5_H1_C5_S1	9	aH1_C3_S1	14	cB3_H2_C3_S1	19	wB3_H1_C5_S1	24	wB3_H1_S1
5	aB5_H1_C3_S1	10	cB3_H1_C3_S1	15	cB7_H5_C7_S1	20	wB5_H1_C5_S1	25	wH1_C3_S1

f

Figure S7: CO and BC trilinear regression (a,b,c), diurnal residual (d),diurnal concentrations (e) all the solutions for autumn analysis (f).

S4. OA and meteorology time series to analyse $PM_{2.5}$ concentrations.

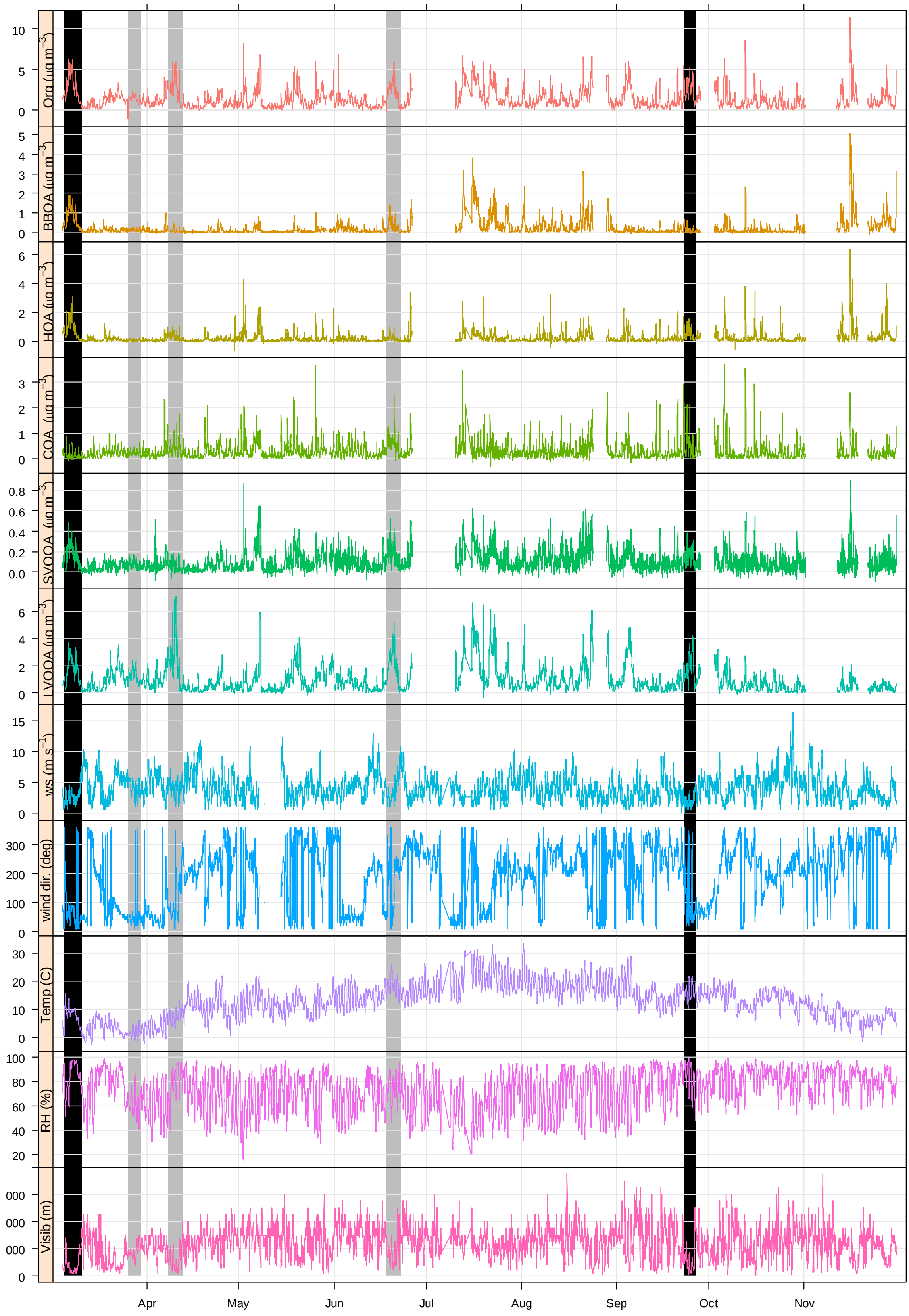


Figure S8: OA and meteorology showing moderate (grey) and high (black) $PM_{2.5}$ concentrations.