Supplementary Material to:

Variations in the chemical composition of the submicron aerosol and sources of its organic fraction at a regional background site of the Po Valley (Italy)

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S1. Ammonium nitrate and sulfate calibrations

Ammonium nitrate calibrations were performed at least once every season following the procedure described by Aerodyne (2010); results are presented in Figure S1. Although noticeable discrepancies are observed when comparing RF_{NO_3} values season by season - e.g. 3.8E-11±0.2E-11 and 2.65E-11±0.04E-11 amp/(µg/m^3) during April and June, respectively, for ACSM#1 - , Figure S1 suggests that these differences are not attributable to a seasonal behaviour of the instrument but instead represents the uncertainties associated with the calibration procedure (related to e.g. the aerosol generator, the DMPS flow or the relative humidity of the generated particles). Thus, for several-months campaigns with an ACSM, we suggest using an average nitrate response factor determined by the slope of all plotted calibration points, independently of the calibration (Figure S1). We also suggest using an average ammonium RIE, which can be calculated as the average of every seasonal calibration (i.e. average of the slopes) since y-intercepts differ from one calibration to the other, likely due to the lower sensitivity of the instrument to ammonium-related fragments (e.g. *m/z* 17 and 16 from NH_3^+ and NH_2^+, respectively) for which interferences with water and air fragments (HO^+ and O^+, respectively) are usually observed (Ng et al., 2011a). The resulting RIE_{NH4} are 5.8±0.1 and 6.5±1.0 for ACSM#1 and #2, respectively, which is in the 3.2-14.7 range reported in (Crenn et al., 2015) from an inter-ACSM comparison. Sulfate RIEs (RIE_{SO4}) are calculated similarly to RIE_{NH4} when ammonium sulfate (AS) calibrations are performed. A literature-based RIE_{SO4} value of 1.15 (Canagaratna et al., 2007) was applied to ACSM#1 since the custom scan file later developed by Aerodyne for AS calibration was not available yet; two attempts to experimentally determine RIE_{SO4} on ACSM#1 were however made by injecting one AS solution of a given concentration using the AN tool similarly to what is described by Aerodyne (2010), which led to an approximate average RIE_{SO4} of 1.25±0.23 i.e. to experimental values that are comparable to the aforementioned literature one. An experimentally determined RIE_{SO4} value of 0.6±0.1 was applied to ACSM#2 based on two AS calibrations performed following Aerodyne (2013) using 5-10 solutions ranging from 0 to 15 µg/m^3.

S2. Source apportionment configurations

Constrained factor profiles set up

Unconstrained PMF allowed for the identification of an HOA-like factor (with high *m/z* 55 and 57), a BBOA-like factor (with high *m/z* 60 and 73, except for summer where BBOA was not found) and an OOA-like factor (with high *m/z* 44 and 43). However, the BBOA factor profile was not fully satisfactory since specific fragments (e.g. *m/z* 43 and 44, Figure S3) were missing, contrary to what is reported in the literature for primary BBOA emissions (Heringa et al., 2011). Constraining the HOA factor only, with low *a*-values (0.05; Crippa et al., 2014) leads to solutions with high seed variability indicating unstable results. Constraining the BBOA factor only with relatively high *a*-values (0.3-0.5; Crippa et al., 2014) does not allow an HOA factor to be identified (factor with high m/z 44 and 60), and leads to unsatisfactory BBOA time series (which are similar to HOA time series when HOA is constrained). Constraining both HOA and BBOA factors leads to satisfactory solutions with relevant factor profiles, time series and daily cycles that will be discussed later on. Note that an attempt to constrain a COA factor was also made, since it has been reported to be an important source of OA (10-30%) in urban areas widely (e.g. Allan et al., 2010; Huang et al., 2010; Mohr et al., 2012; Sun et al., 2011). No clear evidence indicates the presence of COA from our dataset as it is suggested by i) the absence of lunch and dinner peaks for constrained COA factor contributions, ii) the position of our data in the f_{55} to f_{57} scatter plot suggested by Mohr et al. (2012) (slope closer to HOA-like aerosols) and iii) the dubiously high COA and low HOA relative contributions to OA (13 and 7% on average, respectively) when constraining COA. The absence of a COA factor in our study might be related to i) high similarities existing between HOA and COA unit mass resolution factor profiles, ii) lower sensitivity, time- and mass-to-charge-resolution of the ACSM compared with the (high-resolution time-of-flight-) AMS used in the aforementioned studies, iii) the type of site studied here (rural), which is likely less influenced by local cooking emissions than urban sites, but which could instead be impacted by aged, processed COA particles emitted in urban areas that would then undergo oxidative processes during their transport leading to OOA-like aerosols.

Identification of the optimal number of factors

Solutions applying various numbers of factors and constraining different factor profiles for each season are summarized in Table S1. During summer, the optimal solution is found with two factors, which allows for the identification of HOA and OOA (OOA-2F). Setting three factors splits OOA-2F into SV-OOA and LV-OOA like factors as suggested by their factor profiles and daily cycles. However, the absence of *m/z* 57 in SV-OOA contrary to what is reported in the literature (Ng et al., 2011b), the high correlation observed between SV-OOA and HOA (r^2=0.67) and the poor correlation observed with nitrate (r^2=0.13) highlights the limitations of this SV-OOA factor. During summer, the HOA and OOA 2-factor solution was thus preferred, considering that OOA is here likely influenced by both SV- and LV-OOA. During spring, autumn and winter, the optimal solution is found with three factors, which allows for the identification of HOA, BBOA and OOA. Setting two factors leads to an HOA factor, and a mixing of BBOA and OOA in a single factor, as it is attested by the presence of high f_{43}, f_{44}, f_{55}, f_{57}, f_{60} and f_{73} in the latter. Choosing four factors splits the initial OOA (OOA-3F) into two OOA factors (OOA1-4F and OOA2-4F), for which mass spectra and time series vary depending on the season studied. In general, OOA1-4F is similar to OOA-3F in terms of factor profile and time series, whereas OOA2-4F presents biomass burning characteristics (high f_{60} and f_{73}, correlations with BBOA, etc.) in addition to typical OOA features (high f_{44}). However, this second OOA2-4F factor cannot be clearly interpreted as an OOA-BBOA factor as it has previously been reported (e.g. Crippa et al., 2013), due to the absence of key mass fragments during Spring and Autumn (e.g. *m/z* 29, Alfarra et al., 2007), or higher correlations with primary factors than secondary compounds during winter (see Table S1). During spring, autumn and winter, we therefore chose the HOA, BBOA and OOA 3-factors solution, keeping in mind that OOA is likely influenced by secondary BBOA here.

Identification of the appropriate a-value

The influence of *a*-values on the apportionment of organics has been investigated. Different scenarios were tested applying very low to very high constraints to HOA and BBOA factors (see Table S2 for more details). Solutions were defined as relevant or not depending on the seed variability, factor daily cycles, the independence of factor contributions, etc. They were then compared by investigating i) variations of the seasonal average contribution of each factor and ii) factor time series correlations with independent measurements. Results are summarized in Table S2 and Figure S4. In general, very low and very high constraints lead to irrelevant solutions (e.g high seed variability, traffic peaks for BBOA, etc.) for which the average contributions of each factor vary substantially from other scenarios (e.g. see very high constraint during winter). Low to high constraints results in comparable solutions in terms of both relative contributions and correlations with external measurements. We decided to apply low constraints (i.e. *a*-values of 0.1 and 0.5 for HOA and BBOA, respectively) to give as much freedom as possible to our factor profiles while remaining in the range of plausible solutions. Correlations with independent measurements are further discussed in Sect. **Error! Reference source not found.** of the manuscript.

Set up of the period duration

Choosing the appropriate period duration for SA analysis demands a compromise between i) including as many data as possible to reach robust and statistically significant solutions and ii) limiting this amount of data to respect the assumption that factor profiles are constant during the studied period. In our study, SA was performed on three period durations: i) the entire year, ii) two 6-months periods and iii) four distinct seasons. Comparable solutions are found for the three different configurations with the identification of reasonable (factor profile, diurnal cycle, comparison with external data) HOA, BBOA and OOA factors. However, when performing SA on the entire year and on 6-months periods i) the HOA factor does not exhibit a clear morning (i.e. traffic) peak and ii) the ME-2 assumption that factor profiles are constant during a studied period becomes questionable. Note that in the 1-year and 6-months configurations, BBOA represents 3 and 4% of OA during summer, respectively, which can be viewed as an upper estimate of agricultural waste burning contributions. In addition, performing SA on a seasonal basis allows the seasonal variability of factor profiles to be captured (e.g. higher contribution of f_{60} in OOA during winter). We thus preferred interpreting SA results based on seasonal experiments, although comparable conclusions would be reached with the 6-months and one-year configurations.

Table S1. Tests to identify the optimal number of factors.

Season	Number of factors	Factors fixed	Factors identified	Comments
Spring	2	HOA	HOA, OOA mixed	OOA mixed with primary BBOA
	3	**HOA, BBOA**	**HOA, BBOA, OOA**	**Optimal solution**
	4	HOA, BBOA	HOA, BBOA, OOA1, OOA2	OOA1 factor not fully satisfactory: high time series correlation with BBOA factor (r^2=0.74) suggesting an OOA-BBOA factor but discrepancies with already-found OOA-BBOA factor profile in the literature (e.g. *m/z* 17, 29, 41)
	5	HOA, BBOA	Factors split	High correlations between factor profiles and time series
Summer	**2**	**HOA**	**HOA, OOA**	**Optimal solution**
	3	HOA	HOA, LV-OOA like, SV-OOA like	High seed variability. SV-OOA factor not fully satisfactory: no *m/z* 57, high time series correlations with HOA (r^2=0.67), poor correlation with nitrate (r^2=0.13)
	3	HOA, BBOA	HOA, BBOA, OOA	High contribution of BBOA compared to HOA (22 and 7% of OA, respectively) which is very unlikely in summer
	4	HOA	Factors split	High correlations between factor profiles and time series
Autumn	2	HOA	HOA, OOA mixed	OOA mixed with primary BBOA
	3	**HOA, BBOA**	**HOA, BBOA, OOA**	**Optimal solution**
	4	HOA, BBOA	HOA, BBOA, OOA1, OOA2	OOA1 factor not fully satisfactory: high profile correlations with OOA2 factor (r^2=0.97), high time series correlation with BBOA (r^2=0.69) suggesting an OOA-BBOA factor but discrepancies with already-found OOA-BBOA factor profile in the literature (e.g. *m/z* 17, 29)
	5	HOA, BBOA	Factors split	High correlations between factor profiles and time series
Winter	2	HOA	HOA, OOA	OOA mixed with primary BBOA
	3	**HOA, BBOA**	**HOA, BBOA, OOA**	**Optimal solution**
	4	HOA, BBOA	HOA, BBOA, OOA1, OOA2	OOA1 factor not fully satisfactory: time series correlation with primary factors such as HOA (r^2=0.72) and BBOA (r^2=0.70), but poor correlations with secondary compounds such as NO_3 (r^2=0.22) or SO_4 (r^2=0.12)
	5	HOA, BBOA	Factors split	High correlations between factor profiles and time series

Table S2. Influences of *a*-values on the relative contribution of each factor and the agreement with specific *m/z* or independent measurements. Constraints are defined as: very low: *a*-values calculated for every *m/z* from the relative standard deviation of reference factor profiles; low: a_{HOA}=0.10, a_{BBOA}=0.50; intermediate: a_{HOA}=0.075, a_{BBOA}=0.40; high: a_{HOA}=0.05, a_{BBOA}=0.30; very high: a_{HOA}=0, a_{BBOA}=0. Note that *a*-values used from low to high constraints are chosen following the empiric recommendations of Crippa et al. (2014).

			HOA						BBOA						OOA					
			Contribution	r^2					Contribution	r^2					Contribution	r^2				
	Constraint	Relevance of solution	%	Org_67	Org_81	NOx	CO	BC	%	Org_60	Org_73	NOx	CO	BC	%	Org_43	Org_44	NH4	SO4	NO3
SPRING	very low	Not relevant	13	0.27	0.23	0.00	0.00	0.14	32	0.98	0.95	0.31	0.78	0.66	55	0.88	0.97	0.79	0.43	0.81
	low	Relevant	9	0.60	0.55	0.03	0.08	0.28	32	0.99	0.97	0.32	0.81	0.70	59	0.78	0.67	0.76	0.43	0.77
	intermediate	Relevant	8	0.52	0.46	0.02	0.05	0.24	31	0.99	0.96	0.31	0.80	0.69	61	0.89	0.95	0.75	0.43	0.77
	high	Relevant	8	0.42	0.36	0.01	0.02	0.18	29	0.98	0.96	0.31	0.79	0.67	64	0.92	0.96	0.75	0.40	0.77
	very high	Not relevant	7	0.83	0.80	0.19	0.43	0.56	25	0.90	0.92	0.24	0.69	0.64	64	0.91	0.98	0.79	0.40	0.81
SUMMER	very low	Acceptable	22	0.93	0.95	0.07	0.42	0.56							78	0.94	0.94	0.56	0.63	0.18
	low	Relevant	14	0.90	0.91	0.07	0.40	0.52							86	0.97	0.94	0.54	0.60	0.19
	intermediate	Relevant	13	0.89	0.91	0.07	0.40	0.51			-				87	0.97	0.92	0.54	0.60	0.19
	high	Relevant	13	0.89	0.91	0.07	0.40	0.51							87	0.97	0.93	0.54	0.60	0.19
	very high	Relevant	12	0.88	0.90	0.07	0.39	0.50							88	0.97	0.91	0.54	0.59	0.19
AUTUMN	very low	Acceptable	15	0.43	0.39	0.02	0.01	0.13	23	0.97	0.97	0.07	0.67	0.48	63	0.77	0.94	0.53	0.57	0.42
	low	Relevant	11	0.63	0.61	0.07	0.10	0.24	21	0.99	0.97	0.06	0.68	0.47	68	0.82	0.92	0.47	0.53	0.38
	intermediate	Relevant	11	0.58	0.55	0.06	0.07	0.20	21	0.99	0.97	0.06	0.68	0.47	69	0.81	0.92	0.47	0.54	0.38
	high	Relevant	10	0.56	0.53	0.06	0.06	0.20	21	0.98	0.97	0.06	0.67	0.47	68	0.80	0.91	0.47	0.55	0.37
	very high	Not relevant	11	0.84	0.85	0.11	0.35	0.45	22	0.86	0.91	0.06	0.62	0.50	67	0.82	0.95	0.51	0.52	0.43
WINTER	very low	Relevant	11	0.76	0.75	0.41	0.51	0.55	37	0.97	0.97	0.20	0.66	0.63	52	0.79	0.98	0.51	0.42	0.66
	low	Relevant	11	0.58	0.57	0.34	0.33	0.39	36	0.98	0.97	0.20	0.66	0.63	53	0.80	0.99	0.50	0.39	0.66
	intermediate	Relevant	11	0.51	0.49	0.32	0.29	0.35	35	0.97	0.97	0.20	0.65	0.62	54	0.80	0.99	0.50	0.39	0.66
	high	Relevant	11	0.59	0.58	0.37	0.36	0.42	37	0.97	0.97	0.20	0.65	0.62	53	0.80	0.99	0.50	0.39	0.66
	very high	Not relevant	15	0.91	0.91	0.42	0.69	0.72	24	0.91	0.92	0.17	0.59	0.57	61	0.93	0.95	0.42	0.38	0.56

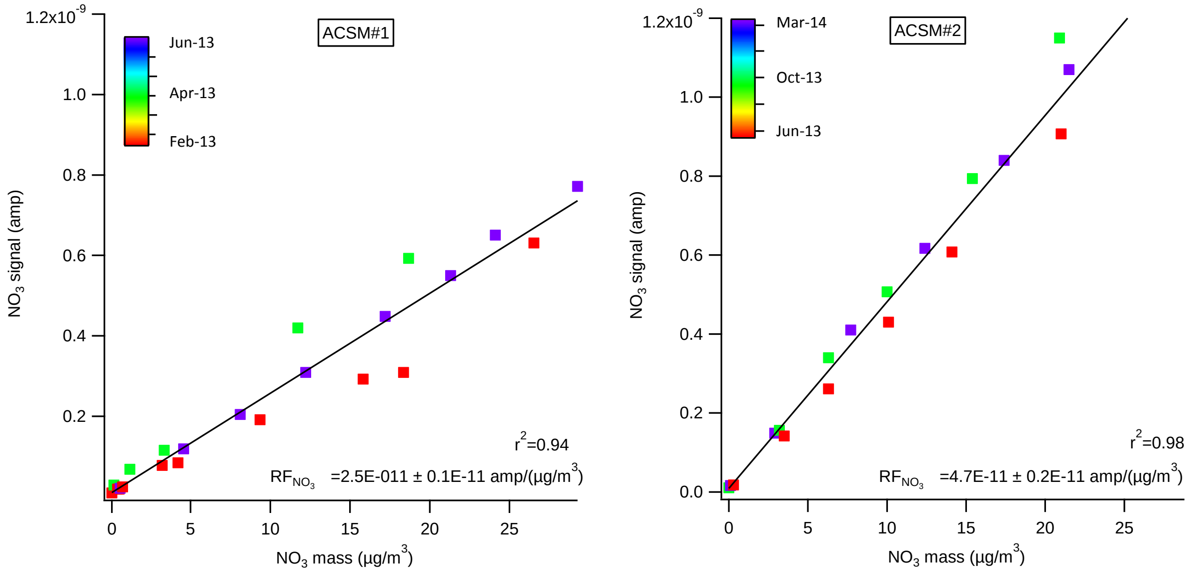

Figure S1. Ammonium nitrate calibrations performed on both ACSMs and colour coded by season. Nitrate response factors (RF_{NO_3}) are indicated ± 1 standard error from linear regression. Note that comparable air beam signals were monitored for every calibration: ACSM#1: (N_2)signal=1.06E-7 ± 0.04E-7 amp (average ± standard deviation) and ACSM#2: (N_2)signal=0.95E-7 ± 0.07E-7 amp; recommended (N_2)signal=1.00E-7 amp.

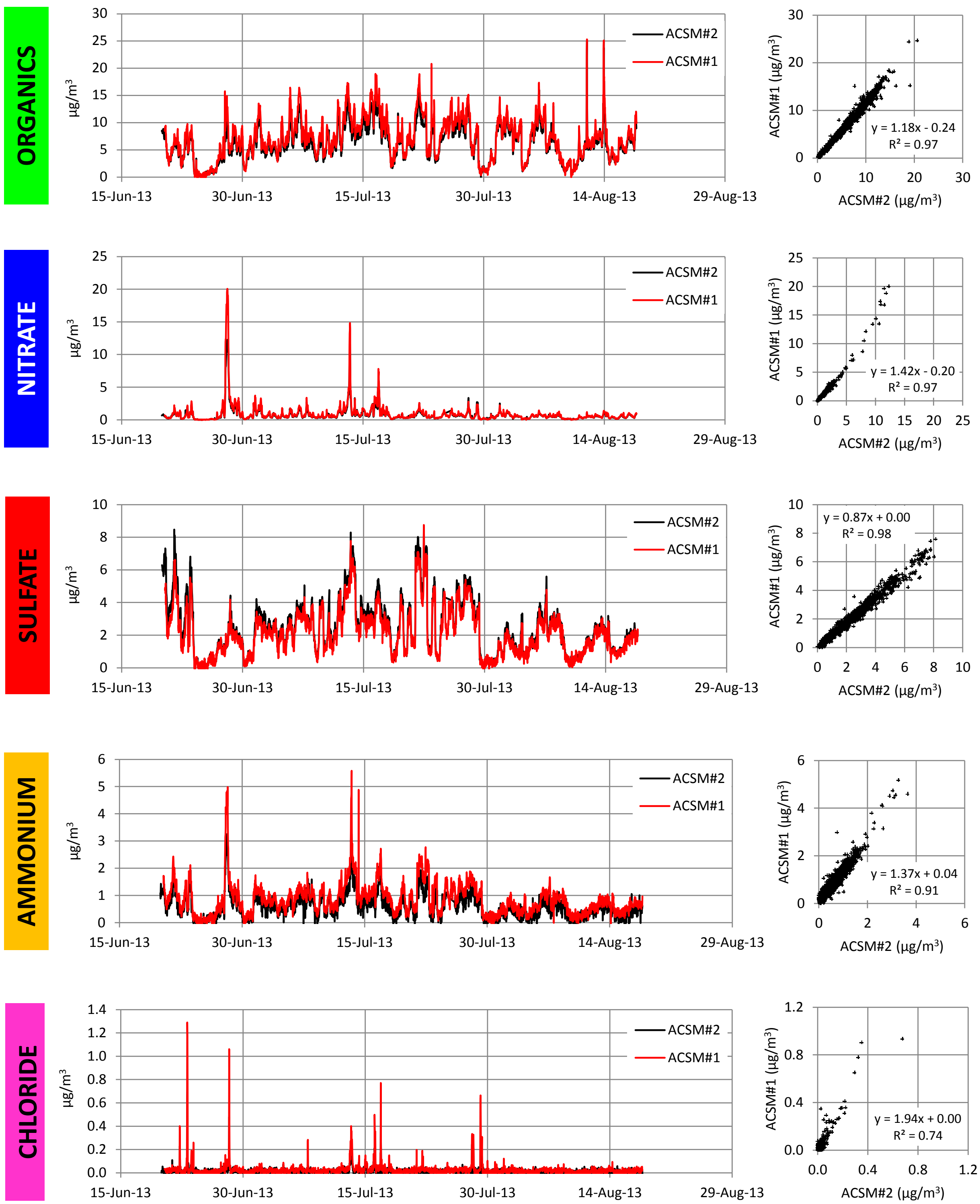

Figure S2. Comparison between two ACSMs for a 2-month summer period. Left panel: time series with 30 min resolution, right panel: scatter plot with 1h time resolution (n=1376, slopes and intercepts are calculated from orthogonal regressions).

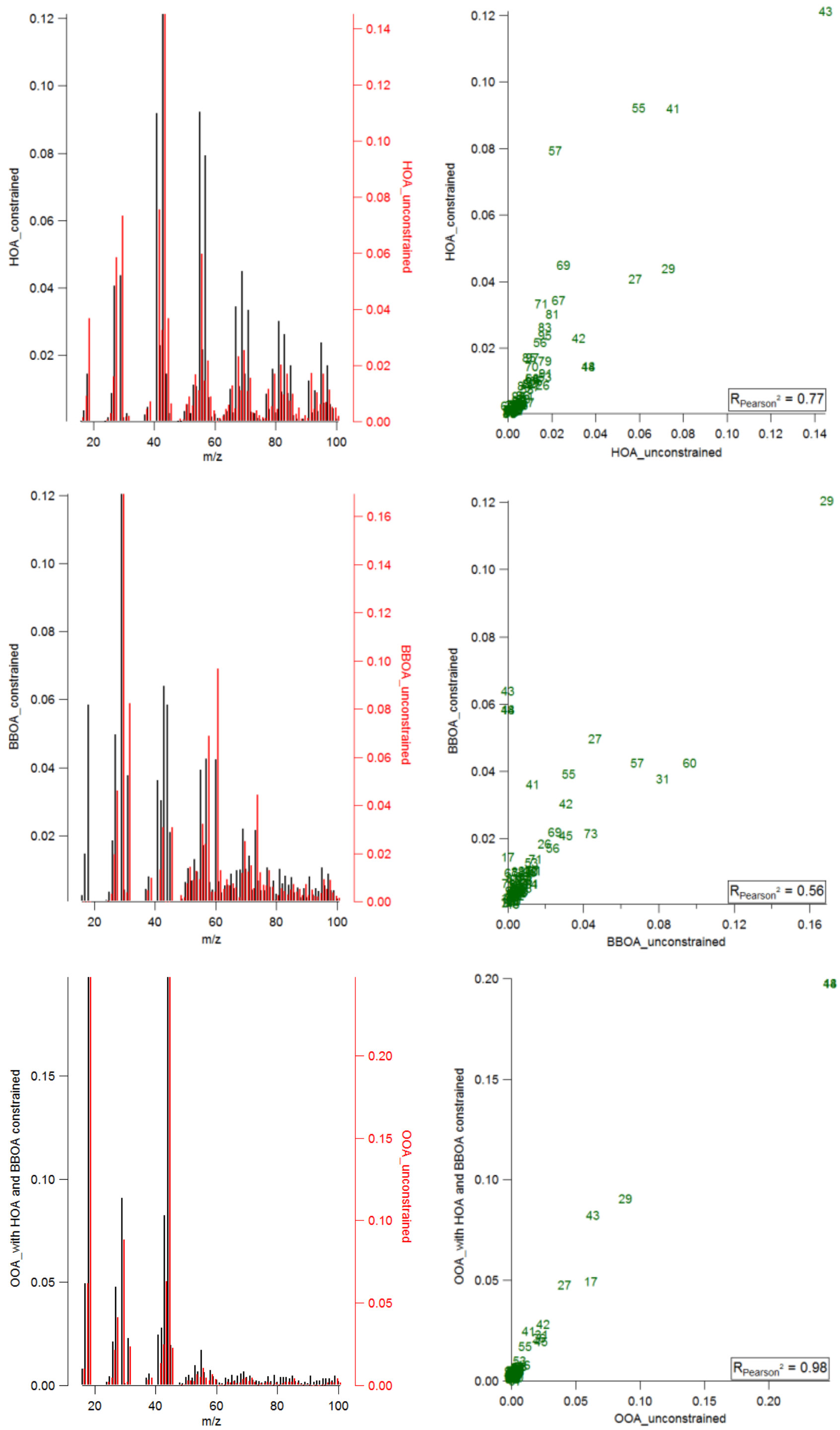

Figure S3. Comparison between constrained and unconstrained factor profiles (spring, three factor solution). Left: relative intensity of every *m/z* in a given factor; right: scatter plots of factor profiles with *m/z* indicated in green.

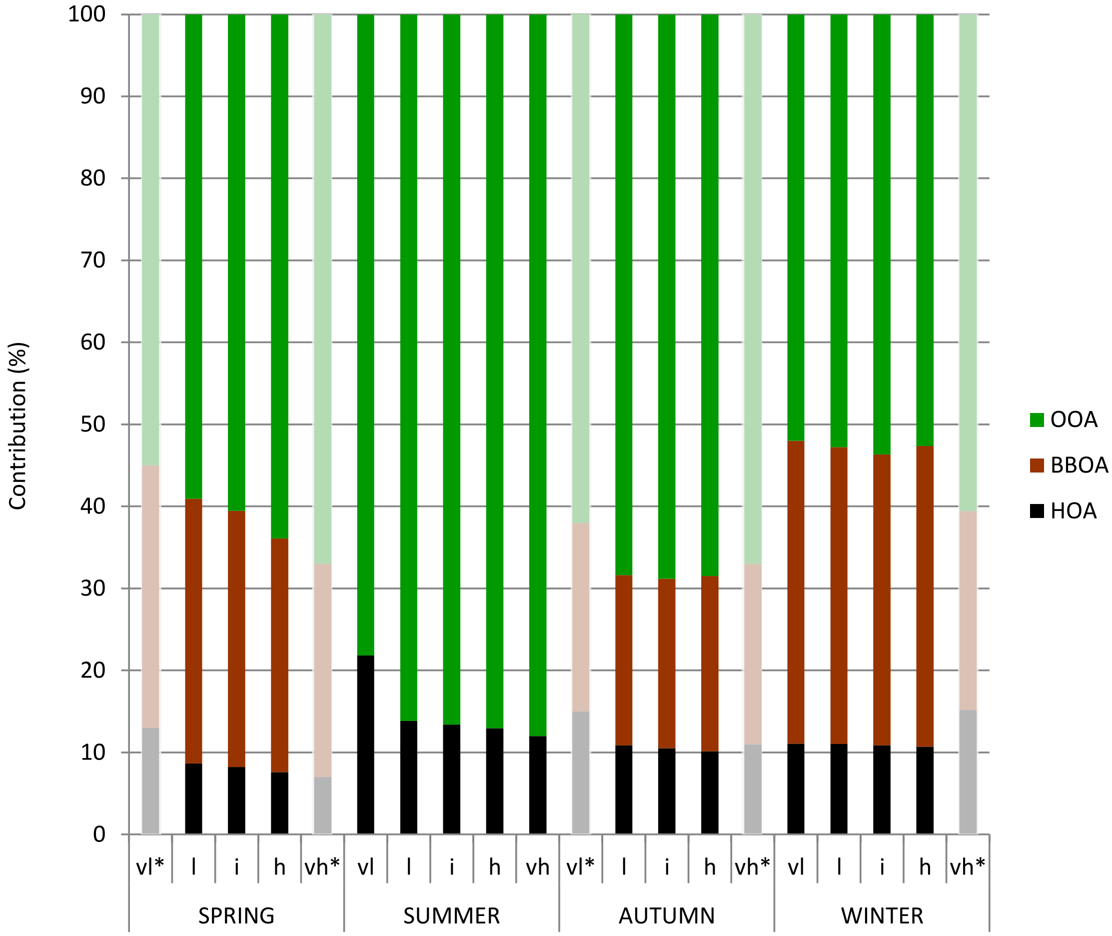

Figure S4. Influence of *α*-values on the average relative contribution of each factor presented by season. vl: very low, l: low, i: intermediate, h: high and vh: very high constraints (see legend Table S2 for more details). Note that scenarios with an asterisk (*) and shaded in grey are regarded as irrelevant due to high seed variability, unexpected daily cycles, etc.

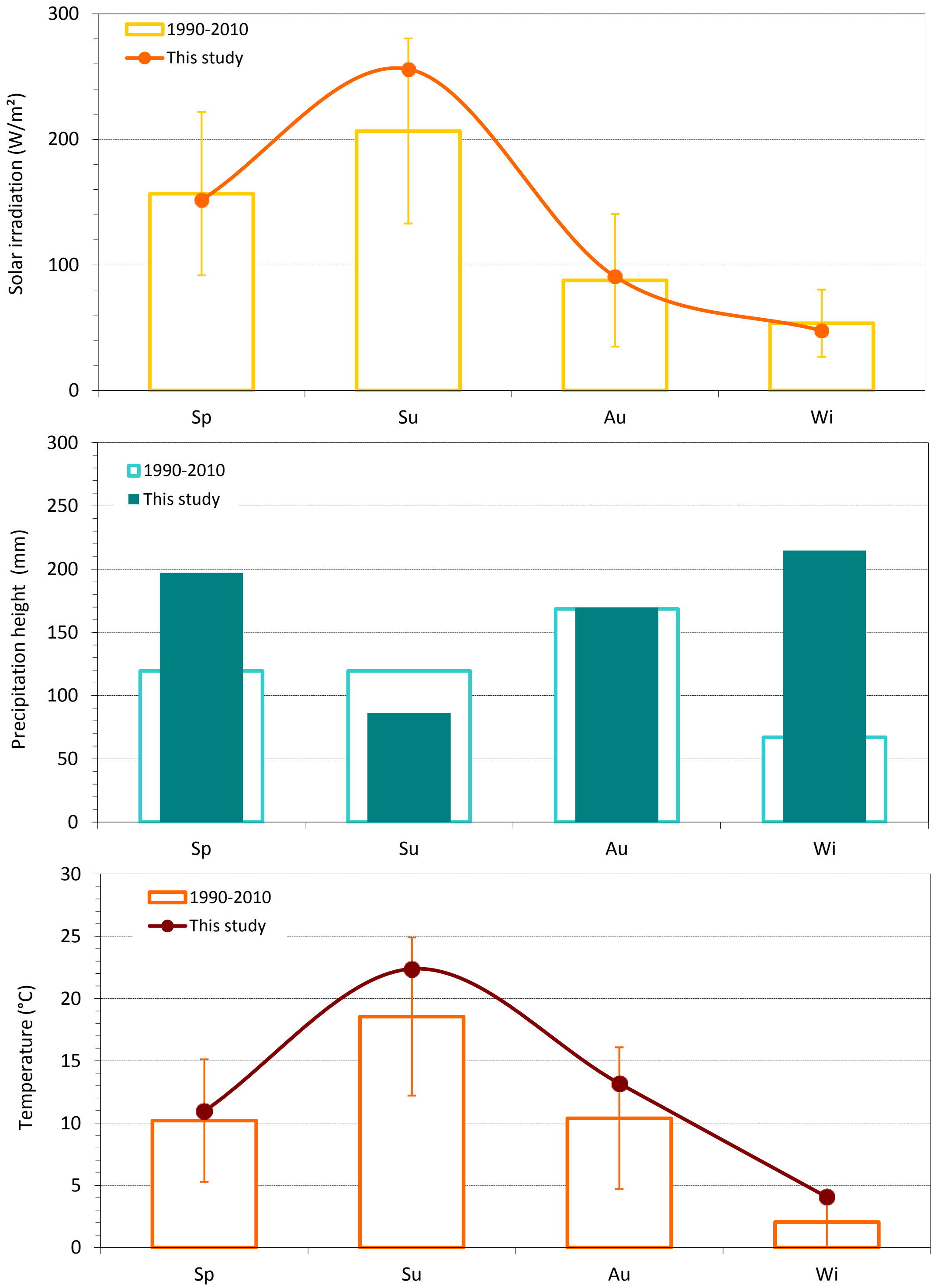

Figure S5. Solar irradiation (top), precipitation height (middle) and temperature (bottom) seasonal averages monitored during this 1-year study, in comparison with the 1990-2010 period averages ± standard deviations measured at the JRC-Ispra site (see Putaud et al., 2014 for more details).

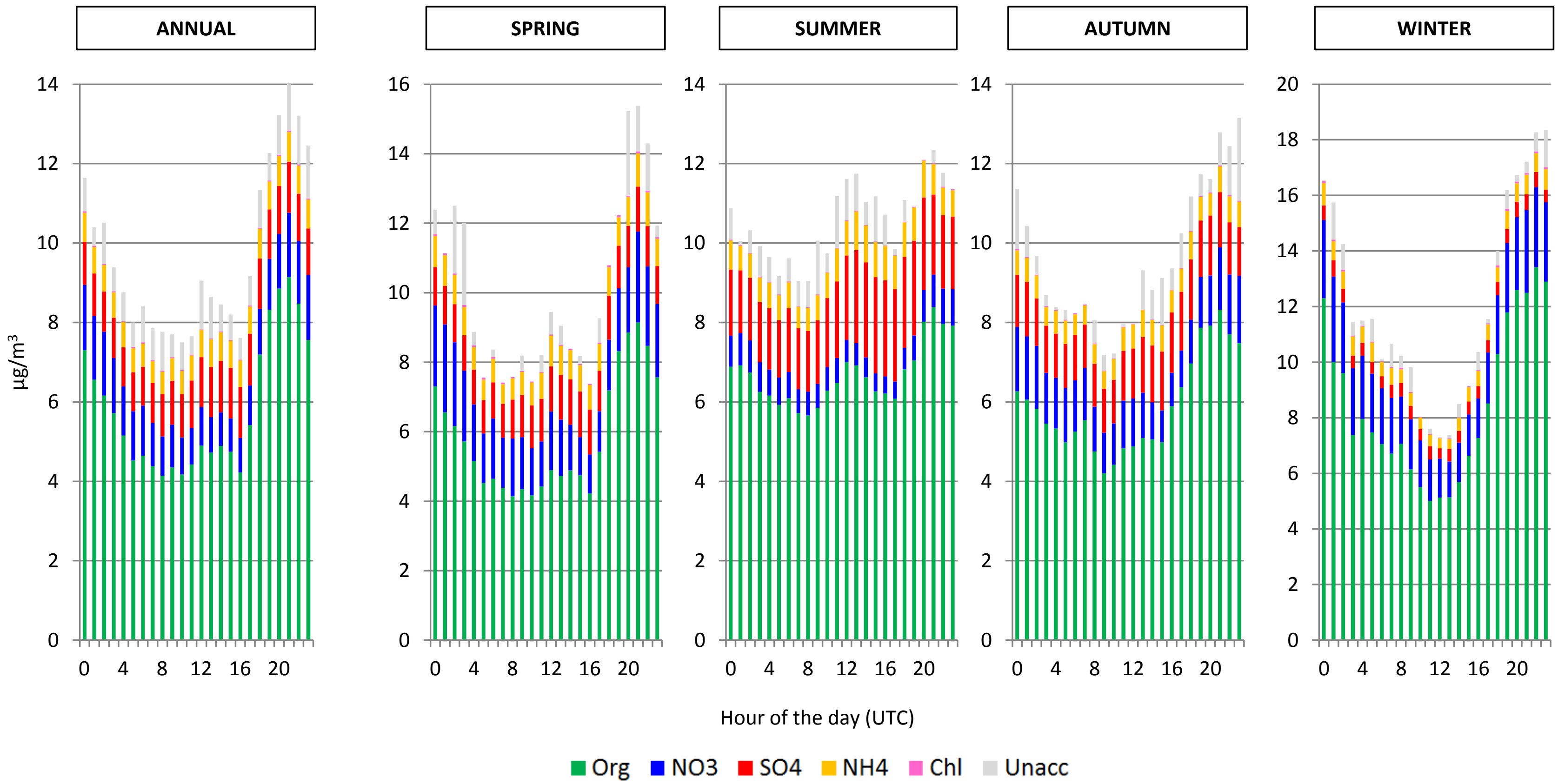

Figure S6. Daily cycles of the median NR-PM$_1$ chemical composition (µg/m^3) presented on the annual scale and by season.

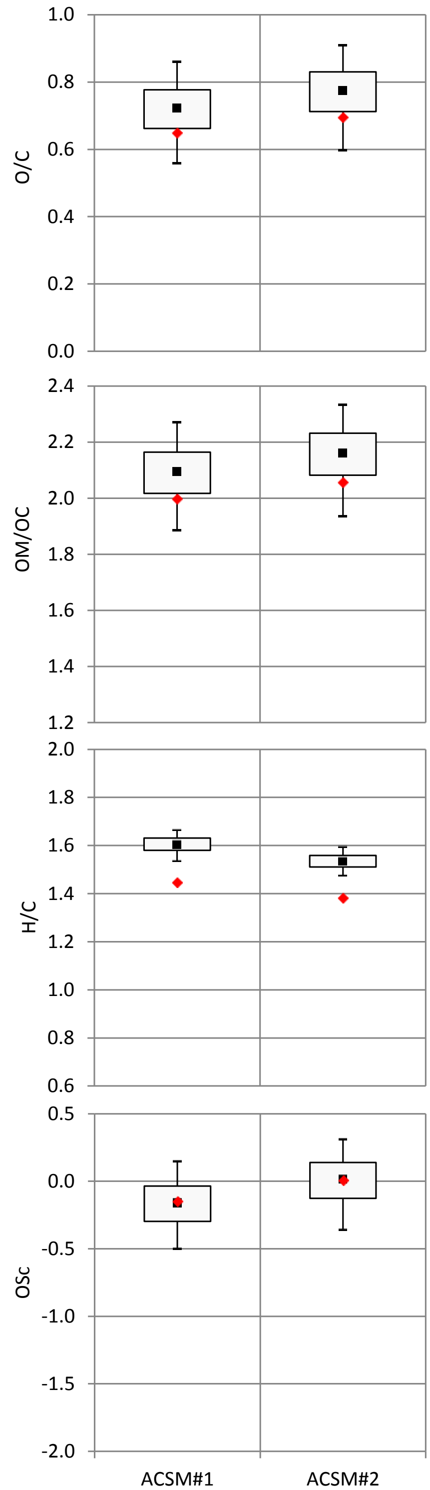

Figure S7. Comparison of O/C, OM/OC, H/C and OSc of ambient OA estimated for ACSM#1 and ACSM#2 from 20 June to 17 August 2013. Black: 5^{th}, 25^{th}, 50^{th}, 75^{th} and 95^{th} percentiles estimates following Canagaratna et al. (2015); red: median estimates following Aiken et al. (2008) for O/C and OM/OC, Ng et al. (2011b) for H/C and Aiken et al. (2008), Kroll et al. (2011) and Ng et al. (2011b) for OSc.

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