PAUL SCHERRER INSTITUT

PSI

LABOR FÜR ATMOSPHÄREN-CHEMIE

Wir schaffen Wissen – heute für morgen

ME-2 and rolling PMF for analysis of long-term aerosol chemical speciation data

Full-year ACSM data analysis

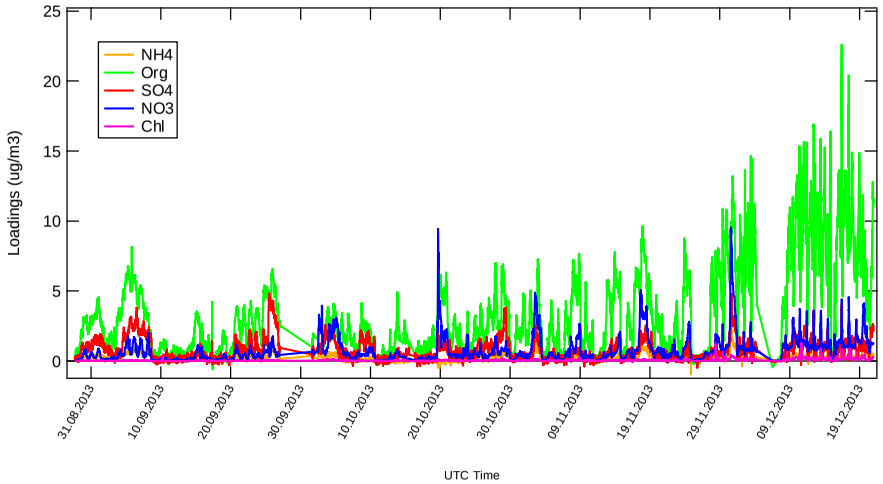
- Automatic rolling PMF algorithm to apportion sources of organic aerosols in a rural site south to the Swiss Alps:
  - ACSM and NABEL station in Magadino
- ACSM measurements in Sept 2013 – Oct 2014
- Auxiliary gas-phase measurements and meteorology
- Filter measurements

- Despite rural location, quite high PM levels in winter when specific topography of the valley favors accumulation of pollutants (annual average PM_{10} in 2008/09 of 20.9 $\mu g\ m^{-3}$)*

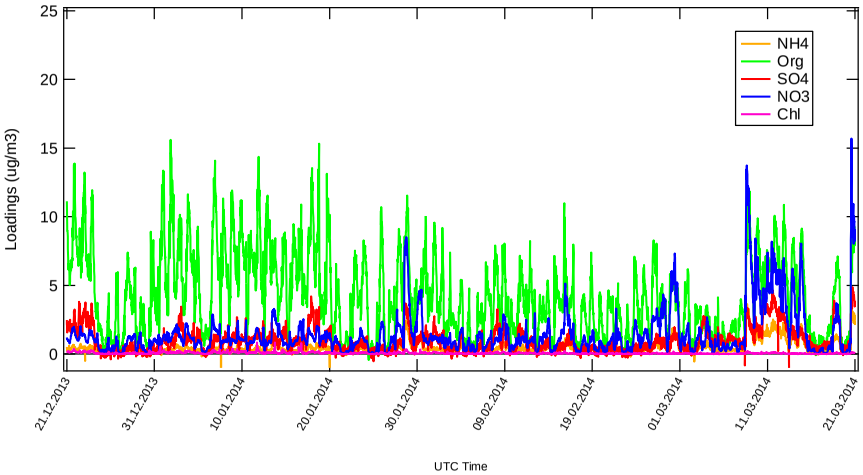
[*] Gianini, et al. (2012) Atmos. Env. 54, 149-158

ACSM measurements in Magadino 13/14

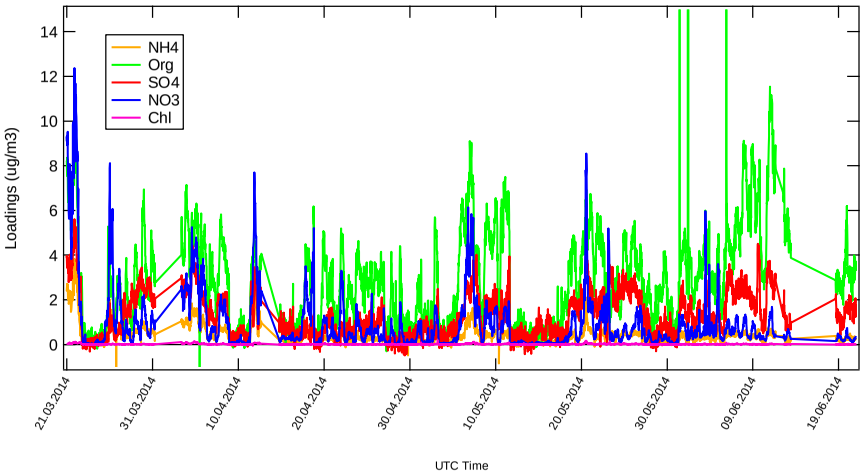
Time Series Fall 2013



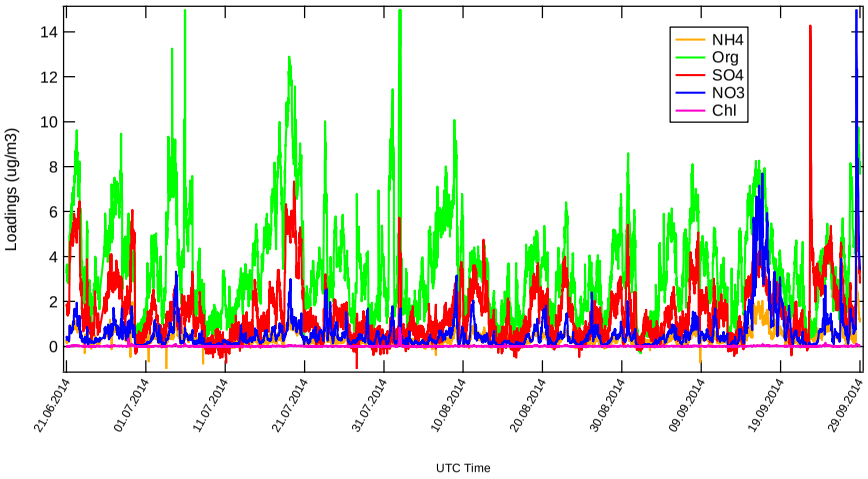
Time Series Winter 2013 / 2014



Time Series Spring 2014

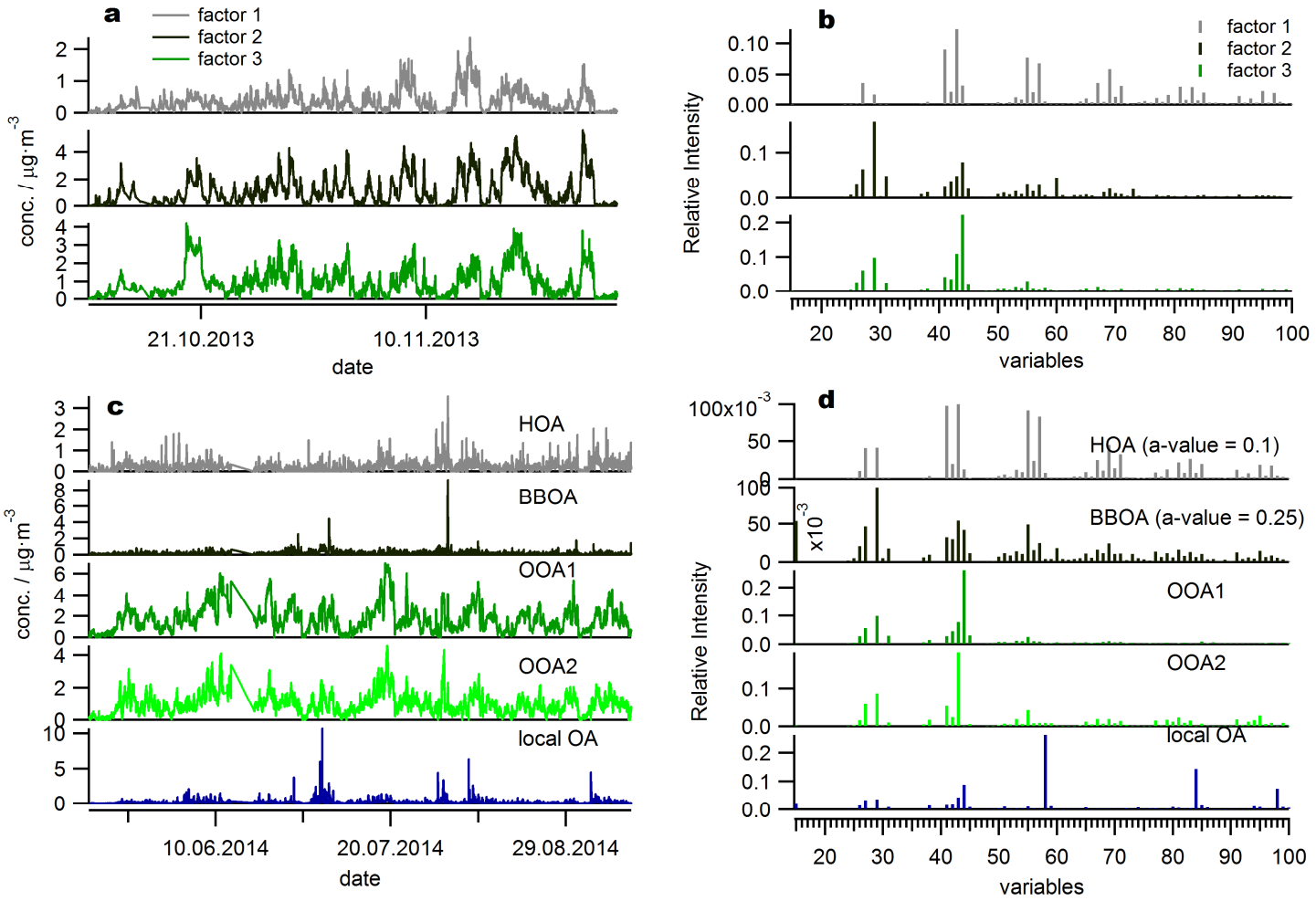


Time Series Summer 2014



Full-year ACSM data analysis

Pre-analysis: (Un)constrained PMF on different seasons to estimate the number of factors, perform sensitivity tests on constraining parameters, elucidate special features, *etc*.



Automatic rolling SoFi

- Rolling PMF algorithm to account for seasonal and/or meteorological variations in organic aerosol sources
- **PMF window ➔** a short subset of data (1-2 months) where PMF is run while aerosol sources stay constant.
- **Automatic part ➔** To determine the number of SOA factors:
- $\sqrt{\left(I_{44}(\text{LV-OOA}) - I_{44}(\text{SV-OOA})\right)^2 + \left(I_{43}(\text{LV-OOA}) - I_{43}(\text{SV-OOA})\right)^2} \geq 0.05$
- ➔ To estimate a goodness of PMF solutions resulted from all model iterations using selection criteria
- **Rolling part ➔** PMF window shift by a short-time interval (½ to 8 days).
- AuRo-SoFi is a combination of an automatic rolling PMF window with a graphical user interface SoFi[1] that runs multilinear engine[2].

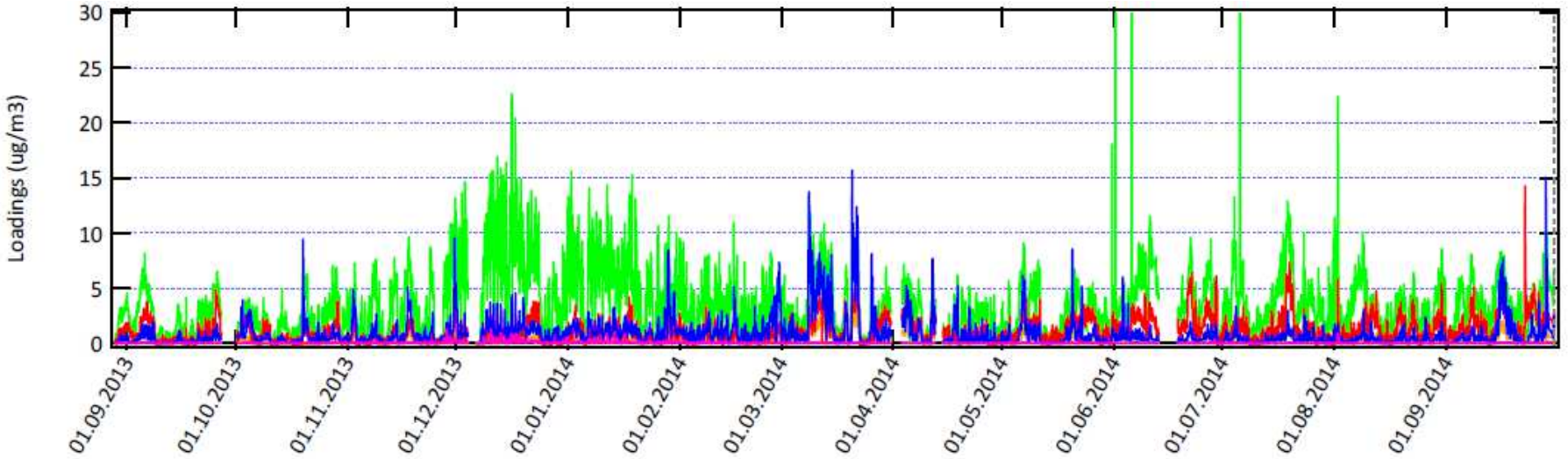
[1] Canonaco F, et al. (2013) Atmos. Meas. Tech. 6(12), 3649-61 [2] Paatero P (1999) J. Comput. Graph. Stat. 8(4), 854-88

Selection criteria for PMF solutions

	Factor	Correlation	Criterion
Primary OA	HOA	Time series, diurnal cycle	R^2(HOA, NO_x)
			R^2(HOA, $BC_{traffic}$)
	BBOA	Time series, diurnal cycle	R^2(BBOA, $BC_{wood\_burning}$)
Secondary OA	SV-OOA	Time series, diurnal cycle	R^2(SV-OOA, NO_3^-)
			R^2(SV-OOA,Temperature)
	LV-OOA	Time series, diurnal cycle	R^2(OOA, NH_4^+)
			R^2(LV-OOA, SO_4^{2-})

- For each of *n* PMF windows → *k* solutions with *a*-value $0 \leq a_{ij} \leq a_{max}$
- Pearson's correlation coefficients between the factor solutions and inorganic species measured by ACSM or an external parameter
- The solution(/s) that maximize(s) the total score (e.g. $\sum R^2$) is to be chosen.

AuRo-SoFi for a full-year source apportionment



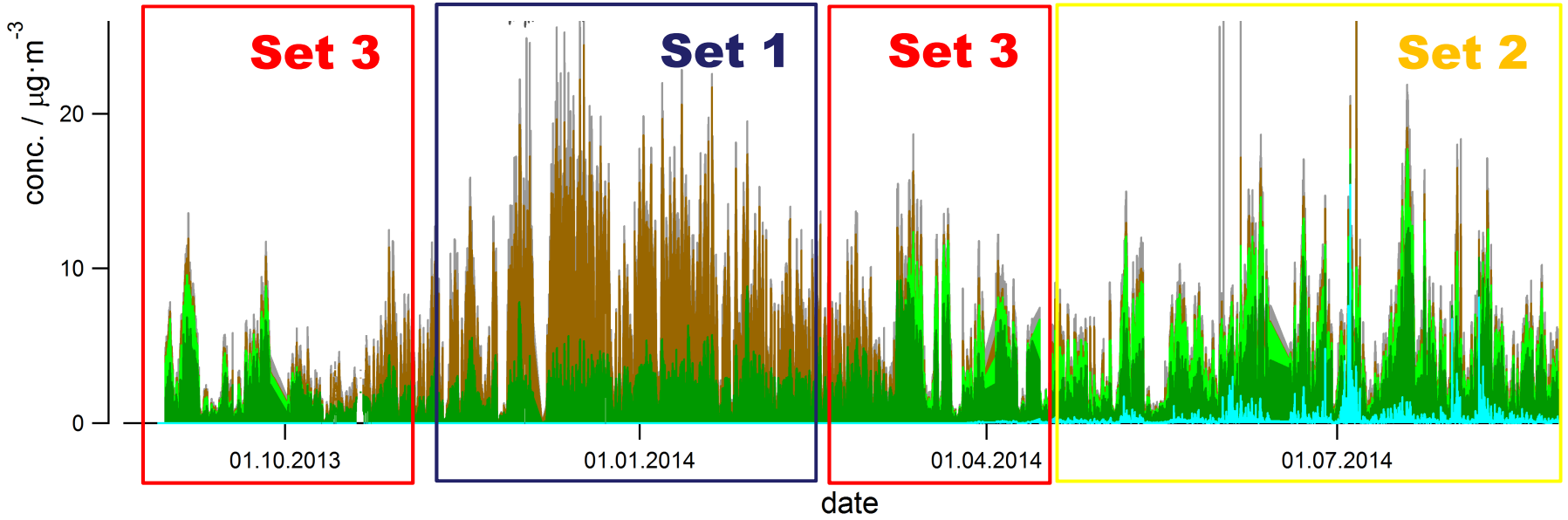
- AuRo-SoFi model parameters:
- PMF window = 1 month, 25 a-value iterations, window shift 1 days
- HOA reference profile from literature, a-value = [0, 0.6]
- BBOA reference profile from literature, a-value = [0, 0.6]
- Local OA (light blue trace) from PMF on summer data, a-value = [0, 0.05]

3/4 and 4/5-factor solutions

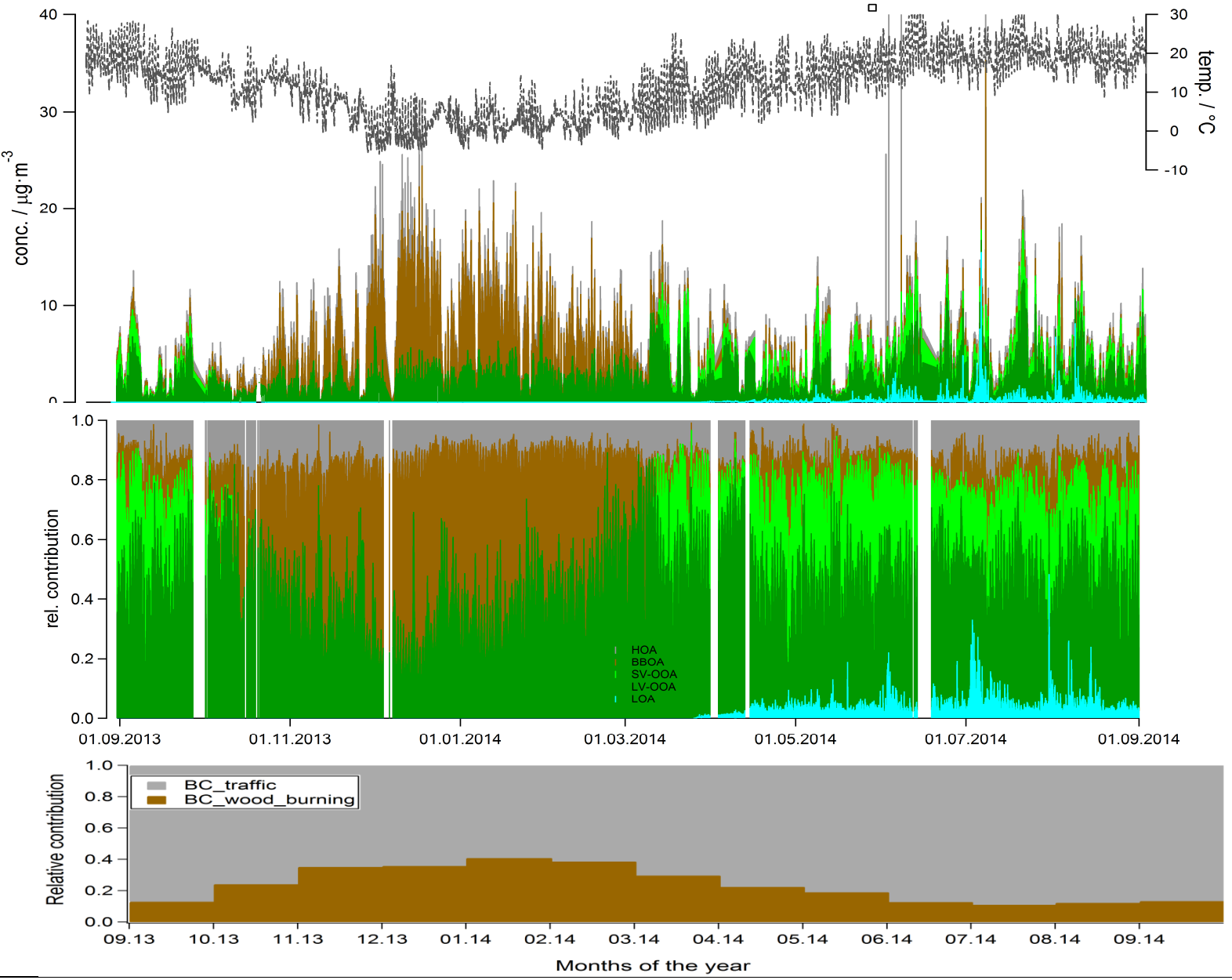
	Correlation of time series							Correlation of diurnal cycles					
Tracer	NOx	BC_{tr}	BC_{wb}	NO_3	T	SO4	NH4	NOx	BC_{tr}	T	NO_3	SO_4	NH_4
Factor	HOA		BBOA	SV-OOA		LV-OOA	OOA	HOA		SV-OOA		LV-OOA	OOA
Set #1	1	1	1	0	0	1	1	1	1	0	0	0	0
Set #2	1	1	0	1	1	1	1	1	1	1	1	1	1
Set #3	1	1	1	0	0	1	1	1	1	0	0	1	1

4/5-factor solution

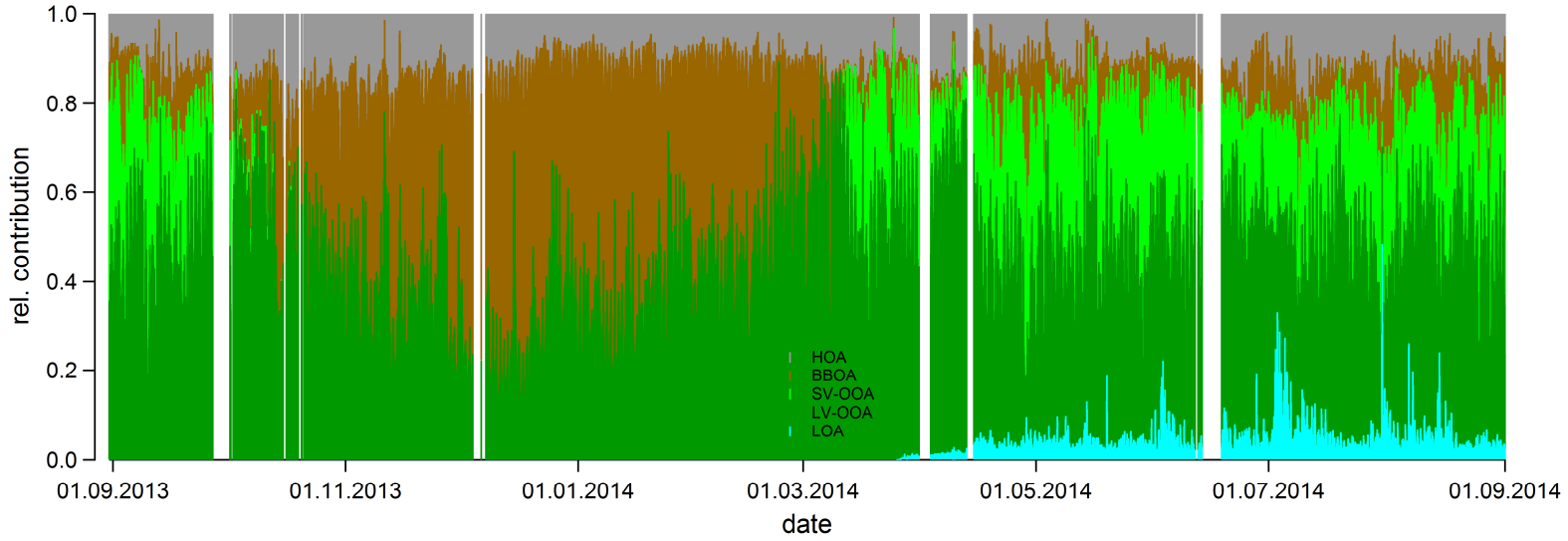
25 *a*-value iterations, window shift of 1day



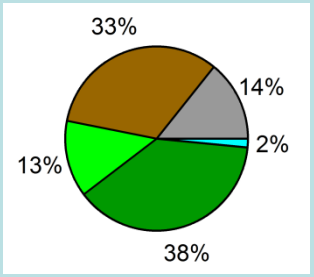
Relative contribution of OA sources and BC

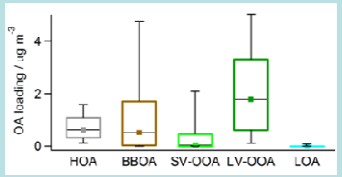


AuRo-SoFi results: source apportionment



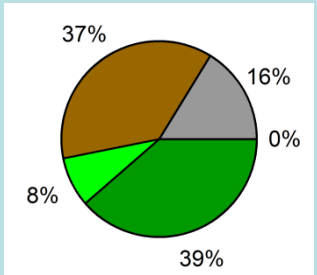
Full-year average

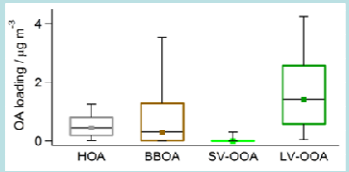




5.4 µg/m³

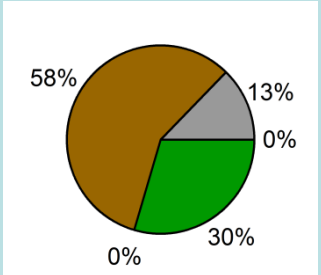
Fall 2013 (SON)

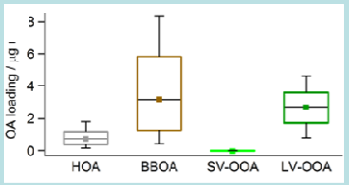




3.8 µg/m³

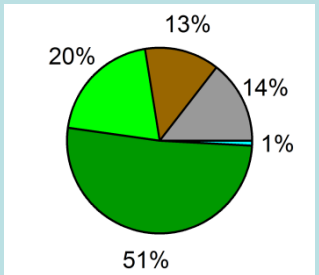
Winter 13/14 (DJF)

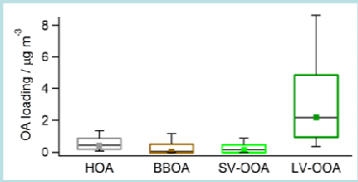




7.5 µg/m³

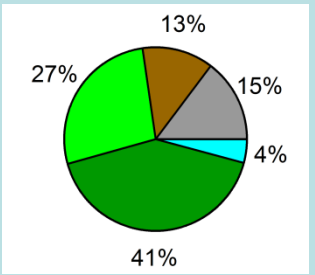
Spring 2014 (MAM)

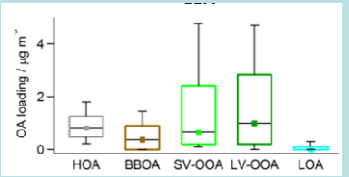




4.5 µg/m³

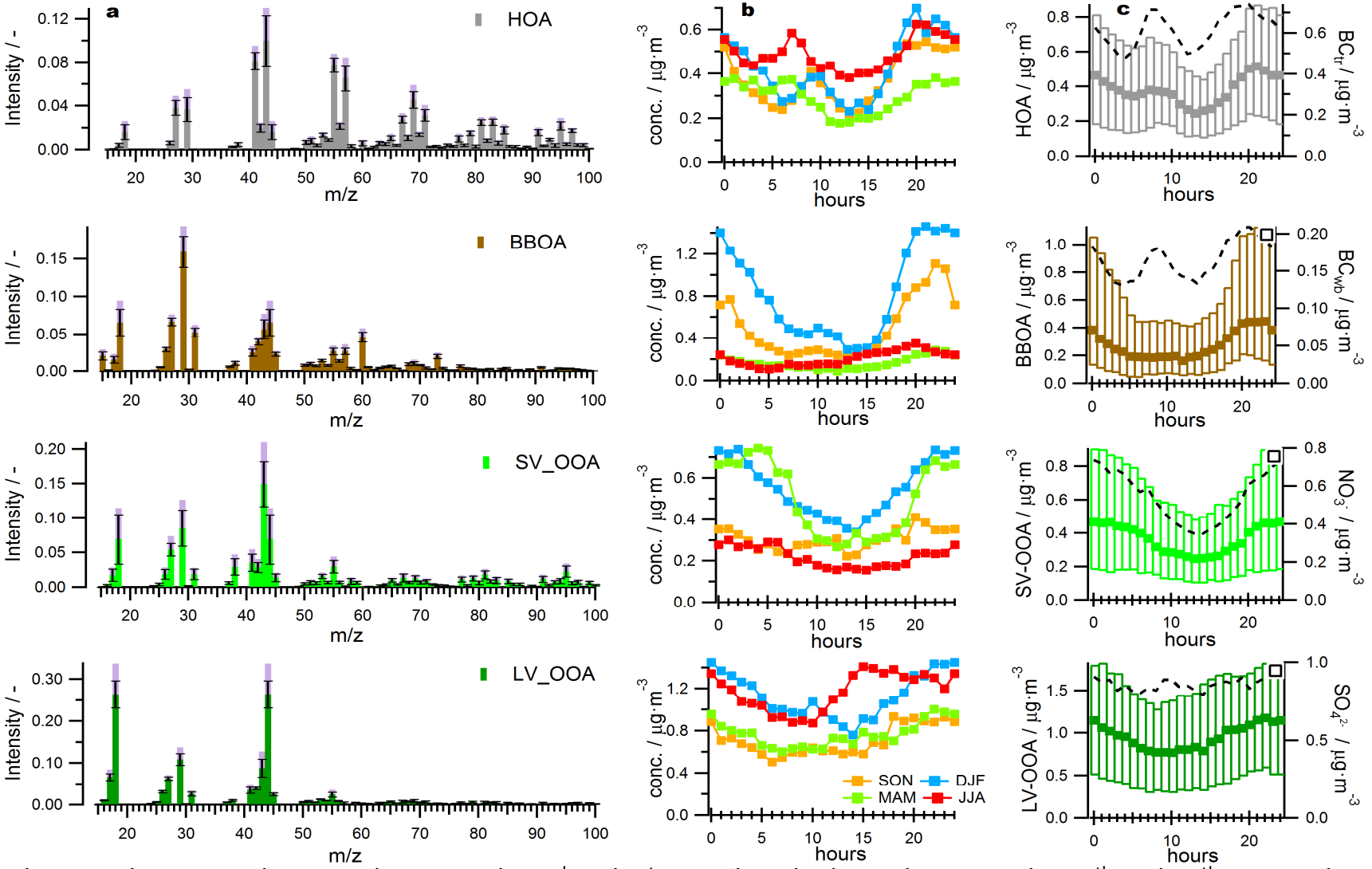
Summer 2014 (JJA)





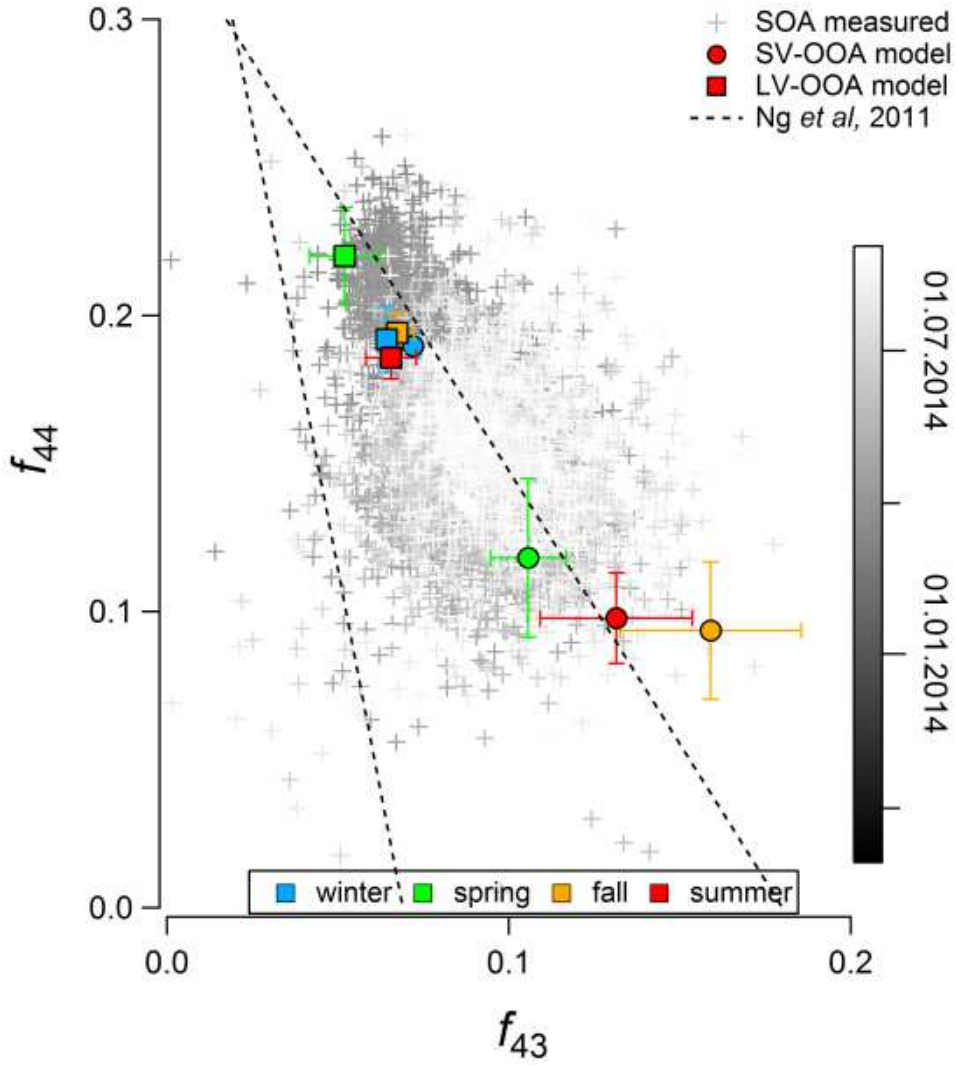
5.4 µg/m³

Profiles and diurnal cycles of main OA sources



The error bars span the range between the 3rd and 1st quartiles; the bars - between the 90th and 10th percentiles.

Seasonal variability of secondary OA factors



Outlook for AuRo-SoFi

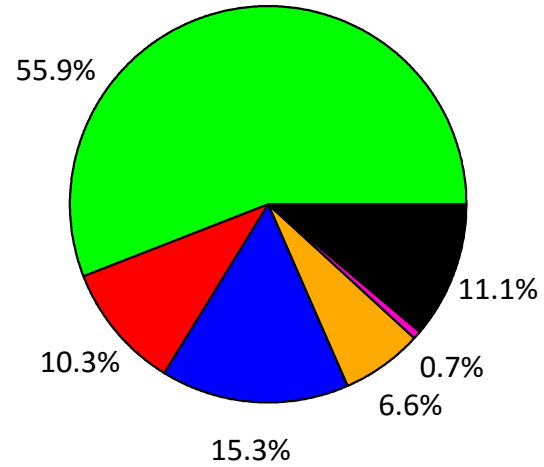
- Implement an AuRo-SoFi panel into the latest version of SoFi-6 source apportionment software
- Implement post-analysis criteria-based selection procedure for the cut-off of the group of the "best" PMF solutions (within SoFi-6)
- Investigate the seasonal dependence of the selection criteria

Conclusions

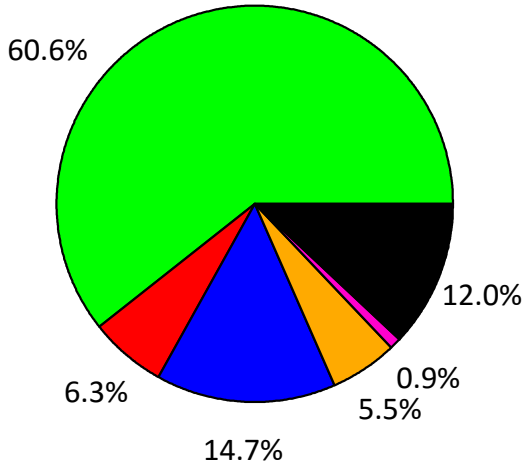
- There is a clear seasonality in NR-PM_1 measured by ACSM, and in BBOA factor retrieved by PMF, with an increase in winter.
- The total contribution of the OOAs increases in summer.
- The two oxygenated OA factors show high seasonal variability of their profiles.
- BBOA contributed on average 58% in winter, which is lower compared to the offline analysis.

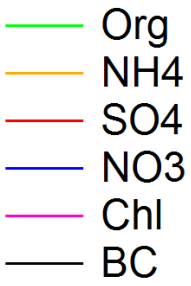
ACSM measurements in Magadino 13/14

SON
Average total concentration: 7.37 ug/m3

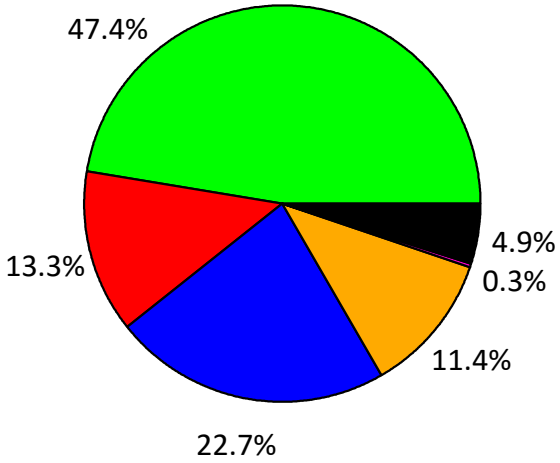


DJF
Average total concentration: 13.85 ug/m3

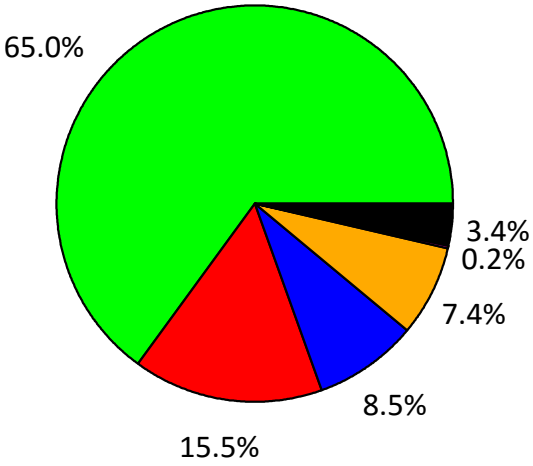




MAM
Average total concentration: 9.79 ug/m3



JJA
Average total concentration: 9.68 ug/m3



full year
Average total concentration: 10.20 ug/m3