A comprehensive source apportionment data analysis for ACSM data

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Long-term online monitoring with the aerosol chemical speciation monitor (ACSM, Aerodyne Research, Inc.) and subsequent source apportionment analysis (Paatero 1994) reveals major aerosol sources. This information is relevant e.g. for selective political abatement strategies.

During the past decade the positive matrix factorization algorithm (PMF, Paatero 1994), which represents multivariate time series as a linear combination of static factor profiles (source profiles) and their time-dependent contributions, has been extensively used for the source apportionment of organic aerosol particles (Zhang et al 2011). PMF results suffer from rotational ambiguity (Paatero et al 2002), i.e. multiple PMF results possess the same goodness of fit. Therefore, the PMF solution space requires systematic investigation to retrieve environmentally reasonable solutions (Canonaco et al 2013). Past studies, e.g. Lanz et al 2008, Canonaco et al 2013, Crippa et al 2014 constraining factor profiles of known sources within the a-value technique (scalar value of the factor profile), performed only a limited exploration of the rotational ambiguity of PMF solutions.

Moreover, for long-term ambient aerosol studies the sources and their evolution vary as a function of the seasons (Canonaco et al. 2015). This change needs to be addressed when performing long-term source apportionment (SA) studies.

In this study we present a comprehensive SA analysis of ACSM data measured over more than one year in Zurich downtown with SoFi. The following crucial aspects were considered:

A. time-dependent source profiles to capture the variability of the sources over the year
B. thorough exploration of the rotational ambiguity and assessment of the rotational uncertainty
C. propagation of the statistical uncertainty of the measured data to the PMF result

Previous investigation of this dataset (Canonaco et al 2015) demonstrated three primary organic factors (POA) (traffic, cooking and biomass burning) and two secondary organic factors (semi-volatile oxygenated organic aerosol and a low volatility oxygenated organic aerosol) (OOA) throughout the year with small variations for the POA profiles and significant variations for OOA.

A) is addressed by performing PMF on small moving windows and allowing the algorithm to best adapt to the current data.

For point B) independent variation of the constrained primary factor profiles (traffic, cooking and biomass burning) are performed that create PMF results represented by a large three dimensional a-value matrix. The solutions are ranked based on user-defined criteria, e.g. the correlation between time series of the traffic factor and traffic markers (NO_x, black carbon), etc. and the best solution (set of solutions) are considered (see Fig. 1).

A small overlap of the moving windows leads to repeats, which are further used to address the statistical uncertainty of the measured data to the total uncertainty of the PMF result (point C).

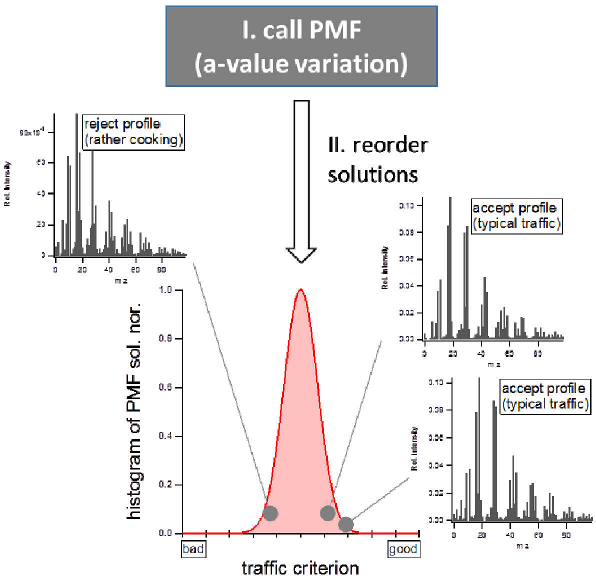

Fig. 1. PMF solutions reordered based on one hypothetical traffic criterion.

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