Anonymous Referee #3

Received and published: 1st March 2017

General Comments:

The authors demonstrate the use of positive matrix factorization (PMF) to the watersoluble, offline AMS spectra to reveal the contribution of the different organic aerosol (OA) sources (hydrocarbon-like OA (HOA), cooking OA (COA), biomass burning OA (BBOA), oxygenated OA (OOA), and an industry-related OA (INDOA) in Marseille, France. They also make comparison between online AMS and offline AMS source apportionment to further show the application of offline AMS measurements for OA source analysis. The paper is very well written, the experimental approach and the data analysis are very clear. I only have one question about the definition of watersoluble, offline AMS spectra.

The authors have a detailed description of how they extract the filters in the experimental section, page 5, line 11" One punch per filter 12 sample (from 5 to 25 mm diameter depending on the filter loading and on the number of 13 punches per composite sample) was prepared for analysis. Punches from the same composite 14 sample were extracted together in 15 mL of ultrapure water (18.2 Mï ¸ A°U cm, total organic 15 carbon < 5ppb, 25_C) in an ultrasonic bath for 20 min at 30_C. After extraction, filters were 16 vortexed for 1 min, and the resulting liquids were filtered with 0.45 ïA¸ m nylon membrane 17 syringe filters."

My questions are: How do we define water soluble AMS spectra? Will the water soluble AMS spectra strongly depend on the filer extraction method (e.g, the volume of water and the temperature used for extraction, and sonication time)? How would these factors affect the composition of aqueous extracts and the water soluble AMS spectra?

Without applying the same filter extraction approach, how could we compare the water soluble AMS spectra and source apportionment analysis in different studies? There is a possibility that the filters could be extracted in different ways in different studies.

Since the offline AMS measurements could be a very useful tool for OA source apportionment, as demonstrated in this work, the authors further elaborate and address these issues in the manuscript.

We thank Anonymous Referee #3 for the review and inputs.

Water soluble AMS spectra are defined as the mass spectra collected from the AMS analysis of the nebulized aqueous filter extracts. Indeed extraction conditions can affect the water soluble AMS spectra. Bozzetti et al. (2017) reported the comparison of water-soluble AMS spectra collected from the atomization of filter extracts using two different nebulizers. Results showed that the collected spectra were not different within the measurements repeatability; however more comparisons (between AMS spectra collected for filter extracted under different conditions) are required.

More importantly, the water extraction conditions may indeed affect the PMF factor water-solubility; therefore factor recoveries relative to filters extracted in different conditions might be different, and therefore should be re-determined from a comparison between the water-soluble OA source apportionment and a well-established OA source apportionment method, as in Daellenbach et al. (2016). Bozzetti et al. (2017) stated that in absence of a well-established OA source apportionment method to be adopted as a reference, the factor recoveries from Daellenbach et al. (2016) can be assumed as a first guess, but their applicability needs to be verified. Overall we do not expect a large factor recovery sensitivity to the water extraction conditions. This is confirmed by the results of the solubility analysis we conducted (see answer to comment 2 from anonymous reviewer #1). Results indicate the factor recoveries to vary by 10% on average when changing the water extraction conditions, where this bias is well within our factor recovery uncertainties. Not surprisingly, the factor recoveries estimated for offline-AMS

(Daellenbach et al., 2016; Bozzetti et al. 2016; Bozzetti et al., 2017) and from PILS-AMS (Xu et al., 2017) are in good agreement.

References (not already included in the main text):

Xu, L., Guo, H., Weber, R. J., and Ng, N. L.: Chemical Characterization of Water-Soluble Organic Aerosol in Contrasting Rural and Urban Environments in the Southeastern United States, Environ Sci Technol, 51, 78-88, 2017.