

We thank the referees for their comments, which helped improving the quality of our manuscript. A point by point response (in blue) to the reviewers' comments (in black, italics) will follow. Changes in the text are indicated in in black.

Anonymous Referee #1

The manuscript presents the results obtained from the off-line analysis of filter samples collected at 9 sites in central Europe with different exposure characteristics. The study is mainly focused on the identification of the different sources that contribute to the organic aerosol loadings using PMF analysis. The obtained results indicate that biomass burning is a major contributor to primary organic aerosol with 88% in the alpine valley and 43% north of the alpine crest. On the contrary, the sum of HOA, COA and BBOA contributes less at the sites north of the alpine crest than at the southern alpine valley. Secondary organic aerosol production is enhanced during summer due to the increased biogenic emissions with temperature. Finally, it is estimated that primary biological particles which cannot be resolved by PMF could contribute significantly to PM10 organic aerosol.

The manuscript is very well written, coherent and easy to follow. A lot of effort has been put into the selection of the PMF solutions and the subsequent sensitivity analysis in order to provide the most sane and justified selection of factors, given the statistical nature of the analysis. This paper can be recommended for publication after some minor corrections listed below:

1) As the title reads "Long-term chemical analysis and organic aerosol source apportionment.." I would expect a short description of the trends in the chemical species as well of the 9 sites. It would be helpful as in a subsequent section the correlation with ammonium and nitrates is mentioned.

We have mentioned in the manuscript (page 3, lines 11-14) that two papers regarding the offline analysis of this dataset are planned. This section has been modified as follows:

"... This paper focuses on the identification of the main factors influencing the OA concentrations at the different sites and the assessment of the associated uncertainties. In a second paper, we will investigate the site-to-site differences and general trends in the factor time series and their relationship with external parameters. ..."

In the corrected version of the manuscript we have added a discussion on the fraction of ammonium that can be attributed to nitrate and to sulfate, for different seasons. This section reads as follows (P13 L30-33):

"... Here, we have used ammonium as a proxy for aged aerosols affected by anthropogenic emissions, as WOOA correlates better with ammonium than with nitrate sulfate. We note that in winter, when WOOA is highest, 56% of ammonium can be attributed to nitrate, whereas in summer ammonium sulfate dominates (97% of ammonium can be attributed to sulfate). Therefore, WOOA correlates more with nitrate ($R^2 = 0.64$) than sulfate ($R^2 = 0.48$). ..."

2) The possibility that WOOA could partially originate from the oxidation of BBOA could also be mentioned. During BB events ammonium is in a significant excess compared to sulfate, and this could possibly explain the good correlation between WOOA and NH_4^+ .

As we have mentioned above, the excess of ammonium compared to sulfate in winter is attributed to ammonium nitrate, which is thermodynamically more stable under lower temperatures and

higher relative humidity. As both WOOA and nitrate originate from a similar process – i.e. oxidation of precursors during winter time – both species do correlate. However, we do not think that this correlation is sufficient evidence to support that WOOA could partially originate from the oxidation of BBOA. We indeed think that this is the case based on modelling results we have recently published (Ciarelli et al., 2017) and on the chemical analysis of the same samples on a molecular level, using ultra-high mass resolution spectrometric techniques. The latter results will be presented in an upcoming publication.

Minor corrections:

Abstract L21: ... smaller than 10 μm from 9 stations ...

The text has been adapted to ... smaller than 10 μm at 9 stations...

“... We present offline-AMS measurements for particulate matter smaller than 10 μm at 9 stations in central Europe with different exposure characteristics for the entire year of 2013 (819 samples). ...”

P16, L26: ... closely correlates with NH_4^+ .

The text has been corrected accordingly.

“...WOOA, dominant SOA category during winter, closely correlates with NH_4^+”

References:

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