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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Time dependent source apportionment of submicron organic aerosol for a rural site in an alpine valley using a rolling PMF window" *by* Gang Chen et al.

Anonymous Referee #2

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Review comments, This manuscript reports the analysis of one year of ACSM mass spectral data obtained from a polluted rural Swiss site using ME-2 implemented in the time-rolling scheme. Five factors were resolved, including two POA factors (traffic-related HOA and BBOA), one local OA (LOA) factor, and two oxygenated OA factors (a less oxidized LO-OOA and a more oxidized MO-OOA). The diurnal cycles, seasonal variations, sources and processes of the OA factors were discussed and the statistical and rotational uncertainties for the modelled OA factors were assessed. In addition, the rolling PMF analysis results were compared to the results from the conventional PMF analysis on the data segregated by seasons and the source apportionment of offline AMS filter samples. This is a valuable study that demonstrates the utility and

strength of applying rolling PMF analysis to the long terms ACSM data. This paper is an important contribution to the field of aerosol source apportionment and should be accepted for publication after the following review comments are addressed.

This study is extensive and the amount of information given in the manuscript is massive. However, the texts can sometimes be a bit hard to follow and confusing. I suggest more efforts are made to organize the contents more effectively, streamline the discussions, and improve the paper's general readability. For example, the descriptions of various aspects of the PMF analysis are lengthy and somewhat lack of coherence. Compiling a summary table of the key information and parameters used in the analysis could be useful. Many figures in the manuscript and the supporting information are hard to read and the figure captions are ineffective. Font size should be increased to be legible under normal page view. Figure captions should be sufficiently detailed to make the figures understandable.

The LOA factor appears to be an artifact arising under certain instrumental condition, thus is not a real ambient OA factor per se. Calling it Local OA implies that it is an OA component associated with certain local emission. This could be misleading, so are the discussions about the ambient behaviors of this factor.

The effort to determine statistical and rotational uncertainties for the OA factors is commendable. One question however is since the PMF solutions are selected, the average a-value is calculated to be fairly low (around 0.2). The errors for the OA factors are then determined based on the selected PMF solution. Aren't this approach somehow circular?

Specific comments:

Line 22, Change "cite" to "site".

Line 21 - 22, it is not appropriate to claim this study "the first ever application of rolling PMF analysis for a rural site . . ." The data analyzed by Parworth et al. (2015) came

from a rural site in Central United States and that study was the first to report the application of rolling window PMF analysis on long term ACSM data.

Section 2.2, it is useful to provide information about ACSM operation and quality control measures, such as the ACSM measurement time resolution, the detection limits for NR species.

Line 148, change “doesn’t” to “don’t”

Line 179, what’s the reason for the ion signals at m/z 12 and 13 being mostly negative? Is this an issue specific to this study?

Line 179 -180, why is m/z 15 “affected by high biases due to potential interference with air signals”? ACSM determines particle signals as the difference between the filter-off and filter-on modes. So, aren’t the air influences on the ACSM diff signals removed from the diff signals?

Line 251, the meaning of “within a range of 0.4” is vague. Spell out the range. What exactly does “random” mean in “a random a-value”?

Section “2.6.1 Window settings”, many discussions within this section do not seem closely related to the topic of setting the proper rolling window size.

Line 254- 255, it is mentioned that different a-values were chosen to constrain the fitting of LOA. is there any significance with the specific value?

Line 257, does this sentence suggest surface ionization enhances the production of N-containing ions? Are there reference(s) to support this?

As the LOA factor appeared after a filament change, it is a factor associated with a certain instrument condition. ME-2 analysis that constraints the LOA profile may lead to the retrieval of this factor may be forced even when it was not supposed to be present.

Line 277, what is eBCtr ?

Line 282, isn’t the measurements done with Q-AMS, how was it known that m/z 43 is C2H3O+?

Line 287 – 288, what do 4th and 5th position reference to?

Line 316, define BLH

Line 325-328, this sentence is awkward and difficult to understand. Consider to revise.

Line 398, the small seasonal differences in HOA% is interesting. What’s the explanation? Since HOA is mainly a POA factor whose concentration should be influenced strongly by BLH, it loading tends to be much higher during winter than in summer. In contrast, stronger photochemistry tends for lead to higher SOA in the summer. So the seasonal difference in HOA% is expected to be strong in winter.

Line 428, what are the standard deviations for the a values?

Line 483, is dimethyl disulphide sufficiently low volatility to be in the particle phase? Are there HR-AMS or other analytical results to support the presence of this compound? As pesticide application is usually seasonal, did you see evidence from this perspective?

Line 519 – 521, are there references to support this statement?

Fig 3, what do BCtr, BCwb stand for?

Fig. S6, explain the error bars in the caption

Fig S8, how exact was the probabilities calculated?

Fig. S9, the key is difficult to understand, what are the dots exactly, what does it mean “clouds of measured f44 vs. f43 in SOA factors”?

Fig. S10, figure caption hard to understand. What are the “Missing time points”?

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