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Discussions
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Interactive comment on "Insights into organic-aerosol sources via a novel laser-desorption/ionization mass spectrometry technique applied to one year of PM_{10} samples from nine sites in central Europe" *by* Kaspar R. Daellenbach et al.

Anonymous Referee #1

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Organic aerosol (OA) is a major component of the total aerosol. Thus, understanding the sources of OA is crucial for controlling aerosol pollution. In the past decade, the development of the aerosol mass spectrometer allows the direct quantification of organic aerosol mass, and source apportionment can be done by further applying PMF on such data set. The main weakness of AMS is that the hard ionization technique causes significant fragmentation, so that some sources are difficult to separate, such as secondary formation from anthropogenic or biogenic origins. To overcome this drawback, the authors, for the first time, explored the usage of LDI technique on offline samples, which causes less fragmentation and may give new insights of OA sources. The authors developed novel procedures for instrument calibration and uncertainty assessment, generated reference spectra for PMF from both specific environment (in the tunnel for traffic sources) and the lab (for wood burning sources), and carefully evaluated the results by comparing to the results from AMS technique. By doing so, the authors reported both the advantages and some potential issues of this technique, which makes this work easy to understand and follow. Although the ionization efficiency and the "matrix effect" of LDI technique remains poorly understood, which weakened the interpretability of some PMF factors, this work provided several new insights in OA sources and should be published in ACP after the following comments are addressed.

Minor comments:

Page 4, Line 16-17. "The observed ions are typically still fragments…". Some of the spectra (e.g. Fig.3d) reported in this work contains some high-mass peaks, which might be complete molecules. This work will be strengthened if you can identify them.

Page 5, Line 5-20 and Page 7, Line 5-10. The usage of $AgNO_3$ seems unnecessary to me. First, using UMR data does not require a high-quality mass calibration. Second, you mentioned that $AgNO_3$ may cause change in ionization physics, which might even hurt the mass calibration. If there are other important reasons for using $AgNO_3$, please specify. Otherwise, I think it is important to mention that spiking $AgNO_3$ should be skipped in future works.

Following this comment, the mass calibration accuracy (Fig.3c) is very unstable and too low under the mass resolution of 1000∼2000. Do you have explanations?

Page 5, Line 31. Remove "scales"

Page 5, Line 34. It is the first time you mention "WSOC", please mention the full name.

Page5, Eq1. Scaling LDI intensity to AMS intensity may cause problems. In later analyses, we know that LDI detection has bias on different OA sources, but AMS detection should be rather equal. If the fraction of different OA sources changed, for example, LDI-insensitive source increased but LDI-sensitive source decreased in the same magnitude, this change will be clearly seen as a decrease in LDI data, but NOT in AMS data. Under such circumstances, the scaling factor has a dependency on source distribution. This might (NOT necessarily) affect the overall PMF results, for example the seasonal variation of PMF factors and the comparison of LDI-PMF and AMS-PMF. Thus, I think it is necessary to double check the results using unscaled LDI data.

Page 7, Line 22-25. The fitting parameter in November is clearly different (Fig.2 red dots), though maybe not significantly.

Page 8, Line 33. "... of which 84, 94, 120, and 177 correlated with NOx...". 84amu is not marked in Fig.4a, and it does not show a strong correlation with NOx (R $\approx$ 0.2 as I read from the figure)

Page 9, Line 15. "Both traffic1 and traffic2 showed a relationship to NOx, only traffic2 correlated with eBCtr (Table S1)". What does the "relationship" mean? The correlation between traffic 1 and NOx is very weak. From Table S1, I think the traffic 1 has neither correlation with NOx, nor with eBC; while traffic 2 may correlate with both.

Page 9, Line 16-17. "This suggests that traffic2 is related to primary emissions from the combustion process and that traffic2 is also influenced by other processes, like e.g. secondary production." For people who are not familiar with traffic tracers, e.g. NOx and eBC, this interpretation is hard to follow. It is good to explain this a bit more.

Page 10, Line 31-33. "The contribution of the last remaining factor showed an exponential increase with temperature, similar to terpene emissions and biogenic SOA (Leaitch et al., 2011), suggesting this factor to be strongly influenced by biogenic SOA production. Therefore, this factor was termed bio-OA." Some other atmospheric processes, other than VOC emissions, are temperature-dependent, such as VOC oxidation and photochemistry. Is there additional evidence to validate the bio-OA factor, such as

higher fractional contribution in rural sites than in urban sites?

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