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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 #1

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The manuscript by Chen et al. describes the Organic Aerosol (OA) Source Apportionment results of one year long measurements carried out by an aerosol chemical speciation monitor (ACSM) in Magadino, a rural village located in the south of the Swiss Alpine region, one of the most polluted areas in Switzerland. The Authors applied positive matrix factorization (PMF) and identified well known OA factors: two primary OA factors (traffic-related hydrocarbon-like OA, HOA and biomass burning OA, BBOA), and two secondary factors (a less oxidized oxygenated OA, LO-OOA factor, and a more oxidized oxygenated OA, MO-OOA factor) plus a socalled local OA (LOA) factor. The main novelty of the study is the application (for the first time on a rural site) of a new "rolling" algorithm to account for the temporal changes of the source profiles in PMF.

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Thus, in addition to the description of the main components that characterize OA in the site under examination, the manuscript presents a detailed comparison between the more traditional methods of applying PMF and the new "rolling" method suggested as the best approach to improve the representativeness of the factors identified. The Authors also calculated the uncertainties (σ) for the modelled OA factors (i.e., rotational uncertainty and statistical variability of the sources) finding values ranging from a minimum of $\pm$4âĂĽ% (LOA) to a maximum of $\pm$40âĂĽ% (LO-OOA). The manuscript represents a huge effort with the aim of improving OA source apportionment PMF capability and it is of possible interest for a large audience of the atmospheric organic aerosol community. However, the text results quite hard to read and it is not completely clear in some parts because of a low-quality presentation and organization of figures and sections (both in the main text and in the SI). Moreover, the significance of the improvement provided by the new method should be clarified better (given the huge amount of analyses required to apply it). For this reason, my suggestion is to accept the paper only after a strong re-organization of the text and after the consideration of some major issues listed below.

Major issues and general comments:

The amount of data presented, the number of different methodologies and settings together with their evaluations make already complex to follow the discussion and understand the main results. Additionally the text is fragmentary, with too many mixing up with SI (sometimes with wrong references). All this makes too hard to follow the discussion and to find out the scientific relevance of the whole work. I strongly recommend to re-organize the paper, summarizing in the main text the most fundamental approaches and results and leaving technicalities to the SI (which should be better organized as well).

The rolling analysis is for sure an interesting attempt to deal with the temporal changes of the source profiles in PMF running over long-term datasets. On the other hand, it requires a huge amount of PMF runs (several thousands) which are very time con-

suming and expensive in term of calculation resources. This can be particularly true if someone hypothesize to extend the same approach to high-resolution AMS data or other datasets in which the number of variables can be substantially higher. Moreover, it is not clearly quantified the improvement of using the rolling approach with respect to the seasonal: I mean, if the the rolling is leading to estimations with uncertainties up to 40% for OOA factors and the improvement in quantification of the same factors is of the order of 5-10% someone can argue that the game is not worth of the effort. I'm not saying this is true, but in my opinion the Authors should add a more comprehensive (even if synthetic) description (motivated by numbers) of the advantages/disadvantages of the rolling approach together with recommendations on how and when the approach can be profitably used or not.

Specific comments

Abstract P2, L22: "cite" probably misspelled for "city"

P2, L27-28: the sentence is redundant: it was already introduced few lines above the distinction of OOAs in two main types.

Introduction P4, L83: here as well as along all the text, please revise the references: you should re-order them based on the years of publication.

P7, L144-146: where is the comparison? Reading this sentence, it seems in Fig. S1 we should find the comparison between different CE, but it's not the case. Please rephrase the sentence clarifying better what is presented in Fig. S1. Consider also that Fig. S1b is not introduced at all in the text neither explained in SI.

P11, L230 and all Section 2.6: there is a lot of confusion between this section and the Supplementary Section 2. It is difficult to follow and understand the steps and what is happening. For example, you mention here the socalled Local OA factor (LOA) but you introduce it much later in the text (and, since it is not a standard factor, it is difficult to understand the motivation of the described procedure). Moreover, you

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discuss figures of SI starting from Fig S6 here, and only later the previous ones. For this reason, please consider to reorganize substantially the text between this section and the corresponding SI section.

P16, L356: again, LOA factor is discussed but it is not introduced/described yet. Please introduce it before commenting on it: since it is not a standard factor you would need to add spectra and explanations before.

P17, L359-364: which kind of tests? It is not clear what you have done here. Consider to rephrase or to remove. Moreover, are you sure you are citing the right figure here? Fig S6 looks unsuitable to me here.

P17-18, L380-391: Consider to anticipate this introduction of LOA factor, as mentioned before. Moreover, why do you call this factor as "Local OA"? If I understood well it is clearly the result of an instrumental artifact: it is interesting that PMF can isolate also this kind of problems, but you should explain and put the emphasis on this.

P18, L394: Supplementary Section 4 is introduced and discussed before Section 3. The Supplementary Figure (Fig. S10) discussed here is instead postponed. This is misleading and create confusion.

P19, L422-424: why do you introduce Fig. 7 before Fig. 6? This sentence is anticipating the topic creating confusion. Please rephrase or remove it.

P20, L436-437: Could you please elaborate more on the peaking concentrations at 10-11am? Why the daily maximum is so late in the morning? I'm used to thinking that the diurnal evolution of PBL leads to more dilution and so lower concentrations of pollutants and that it starts earlier in the morning. I suppose there is a late sunlight illumination of the ground at the site, but it should be explained clearly because it sounds weird.

P23, L519-521: what do you mean with "more complex aging processes"? It is not clear and/or highly speculative. It is actually demonstrated by an increasing number of studies that OOA formation and ageing is complex also under low temperature and

dark conditions. If you mean that the higher variability at higher temperature could reflect different sources/precursors emissions enhanced at higher temperature (e.g., higher emissions of biogenic VOC and the subsequent mixing with preexisting OOAs or whatever) you should explain it better.

P24, L528-534: this is interesting. But the sentence is quite problematic. Can you elaborate more on it? Do you believe it can be also a question of selective water solubility of components?

Conclusions: expressions like "a somewhat better solution" or "more realistic results" are quite subjective and vague. "More realistic" based on what? What is the real improvement of using rolling PMF instead of more traditional (and less time-consuming) methodologies in term of identification and especially quantification of OA sources? You should clarify it better here in the conclusions. And possibly you should add a more comprehensive description (motivated as much as possible by numbers) of the advantages/disadvantages of the rolling approach together with recommendations on how and when the approach can be profitably used or not.

Figures General low quality, with too small font sizes making difficult to read labels of the axes and legends. Sometimes problematic also the choice of colors (e.g., Fig. 5 and 9a). Please check the readability of all the figures in the main text, in the Appendix A and in SI.

Figure 3: given that the x-axis (the time period) is common to all the panels, please consider to use only one or at least to make them of the same length (in order to improve the readability and the comparison between the different time trends)

Figure 5: all the labels (axes name and values, legend, etc.) are difficult to read. Especially graphs in panel b are completely not aligned, their legend is unreadable and the colors of the time series are misleading. Please increase the font size of all legends and labels and improve the general format of the figure.

Supporting Information Check if the Figures follow the order of presentation in the text: Figure S6 should be anticipated (because discussed before, at the beginning of Section S2). In order to improve the readability it is also important to put intervals and possibly titles between subsections or figures/tables referring to different tests/results/data.

Fig. S1b is not introduced at all in the main text neither explained here in SI. You need to explain what it is showing (for instance, what is the mini-denuder? Where it is introduced?)

Section 2 P5, L62-63: do you have any references for this? What do you mean with "more accurate estimations"?

P6, L88-91: it is hard to understand how do you use t-test. This is probably better explained in Canonaco et al. 2020, but it is important to spend some more words also here to improve the understanding of the readers.

P6, L96-99: The same comment, it is hard to understand the procedure. Please rephrase and explain better.

Fig. S5: labels of the axes are missing. Please add them and increase the font size of the color-legend.

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