

# ***Interactive comment on* “Source apportionment of PM<sub>2.5</sub> in Shanghai based on hourly molecular organic markers and other source tracers” by Rui Li et al.**

## **Anonymous Referee #1**

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### General comments:

This paper presented source apportionment and chemical characteristics of fine aerosols in Shanghai, China. This work was performed based on highly time-resolved measurements during a near one-month campaign in winter, along with PMF analysis. The combination of different aerosol compositions, including OC, EC, water-soluble ions, organic markers, and some other tracer elements was applied to constrain the PMF analysis. I agree such a methodology could help to further understand PM<sub>2.5</sub> sources and its processes in the atmosphere. However, I don't support that the data collected from the two different sites could be reasonably combined together into the

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PMF analysis. This data treatment strategy can lead to a huge uncertainty in PMF results. Therefore, I don't suggest this paper can be accepted, if the authors wouldn't fully address the uncertainty generated from the two databases in PMF analysis. When carefully reading this paper, one can find it's not well written, and it's hard to follow. Moreover, some discussions could not be directly supported by the results. It should be suggested that all (or major) authors should carefully review/validate this manuscript before submitting.

Comments in detail:

Lines 14-21: Abstract should be concise. These sentences can be improved to only briefly describe what necessary and importance of this study are, as well as to briefly address scientific question of this study.

Line 19: Measurements at the two different sampling sites were conducted in this work, which should be addressed here. This is a very important information in PMF analysis. Aerosol chemical composition, especially organics, can be substantially different at different sites even in a same city region. Again, I don't believe this methodology is reasonable unless the authors can prove it.

Lines 21-22: This sentence can be improved. And what's the difference between "molecular makers" and "organic makers" here? Please rewrite this sentence.

Lines 23-24: Not clear. Please rewrite it.

Line 24: "The three" -> "These three". Another question, why did the authors select those sources for the quantification discussion here? Why not others included, since you have identified many source factors?

Line 25: It has already been well known about "the secondary source is an important source of atmospheric pollution" in eastern China. Better to rewrite this sentence.

Lines 26-27: It's a confusing sentence (Grouping different sources . . . , respectively). Please rewrite it. And what is the difference between SOA and SOC mentioned here

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together? Maybe only needing to show any individual of the two terms in the abstract section, if you don't need to explain the different things between SOA and SOC. In addition, SOC and POC should be defined before using them.

Lines 33-35: Maybe a region scale should be given here, for instance, e.g., in eastern Asia or China?

Lines 35-36: This sentence should be revised to be better read. In addition, “Zhang et al., 2009;.” -> “Zhang et al., 2009).”

Lines 36 – 42: These sentences should be revised. As written here, aerosol climate and environmental impacts are not well described. They can be improved better for easy follow. For example, the sentence (lines 38-40) is discussing about “climate impact of aerosols”, while in the hereafter sentence, you conclude “importance for air quality”. I mean, in this paper, it may be not needed to highlight aerosol climate aspects in too much detail, but it can be discussed with a very brief version. In addition, in the first sentence (lines 33-35), you already have addressed the situation of PM2.5 pollution and it's important. But, in the sentences (lines 40-42), you highlighted the similar information again. Overall, I would suggest that more concise sentences can be made here to better describe importance and necessity that are associated with the work of the present study.

Lines 43-44: Again, this information has been declared in the above paragraph, no? Is the sentence of lines 44-45 expressing a similar knowledge as the lines 35-36?

Lines 46-47: What are “. . . other methods”? They could be briefly discussed here, since you would highlight some advantages of PMF model, especially you will need apply it with a unique PMF input dataset.

Lines 48-52: It may be not only due to this reason (as you discussed above) why PMF has been widely applying in aerosol source apportionment, as reported by many previous studies. Meanwhile, it would not be necessary to list too many references

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here. If I understood well, the authors would like to highlight the advantage of PMF compared to other receptor models, and why PMF was applied in the present study.

Line 55: Filter measurements can be also available for short-term source apportionment study. Not sure if this sentence is necessary/clear here with the current way.

Lines 58-62: These sentences are not clear enough. What are advantages of the observation methods in the present study, compared to AMS techniques? One could argue that the combination between AMS and organic tracers could be also a very good strategy for source apportionment of organic aerosols (e.g., Huang et al., 2014 Nature).

Lines 63-64. One transmission sentence between the two sentences is needed, otherwise one could understand that many previous studies performed in Shanghai could be only due to a large population. I guess you would express that Shanghai is a megacity, representing for a typical economic zone in China and with a large population there, while air pollution issues are complex due to unclear enough aerosol sources that have to be better understood to further improve air quality.

Lines 64-71: Following the similar comments above, I would suggest the author to improve this paragraph.

Lines 67-68: It is not true that offline techniques are limited in air pollution event studies.

Line 77. As I mentioned above, the reasonable application to combine the two data sets from the two different sampling sites for PMF analysis needs to be further proved.

Line 95. More detail in methods of organic compounds data analysis (including more descriptions in instrumental information) and data quality control should be described here, since such data set is a major part in the present study.

Line 131: “1/2” can be understood by “1 or 2” or “0.5”. Same comment on line 132: “5/6”.

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Line 134. A reference or acknowledgement when you use trajectory data and tool should be given.

Lines 142-143. Some references should be given to support this statement. And what the link between this sentence and the ones hereafter? It seems that further discussion on potential impacts of the meteorological conditions on PM<sub>2.5</sub> pollution observed in the present study is needed. Wind-dependent analysis of different aerosol sources can be applied here, I think.

Line 167: Define the terms BS, DISP and BS-DISP.

Line 169: Please show specific results and/or further prove the cases when the factors were mixed based on those base runs.

Lines 172-173: Figure 3 presents results resolved by the MM-PMF analysis. This information should be given in this sentence, since two PMF methods have been applied and hereafter you did a lot of discussion on Figure 3.

Line 173: “Secondary nitrate factor (F1) was identified by high concentrations of nitrate and ammonium...”. The PMF factor profile is described by contributions of each variable, but not their absolute concentrations. I think it might be enough when you directly discuss their contributions in factor profiles. Same comment on the similar statements.

Lines 177-179: what’s difference between SOA and SOC? Please show results about “... which can be seen from the correlation of the respective species with OC.”

Lines 179-181: Those sentences could be read by: “Figure 4 presents the relative contributions of various source factors to PM<sub>2.5</sub> and OC based on MM-PMF analysis, respectively. The F1 and F2 contributed 33.5% and 15.6% to the total PM<sub>2.5</sub> mass concentration, respectively.”

Lines 181-182: What does SOC contribute to, PM<sub>2.5</sub> or OC?

Lines 182-183: Such sentences would be better discussed step-by-step for each factor,

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something like: "As shown in Fig. 5, the diurnal variation of the secondary nitrate factor shows high concentration during nighttime (e.g., 21:00 – 04:00), while its low concentration is observed during daytime (e.g., 11:00-14:00). etc. The F1, a secondary sulfate-rich factor, didn't present obvious diurnal pattern, which is however characterized by limited variations across the day. etc. ." This is only an example. The authors don't need to exactly follow it, but clearer discussions are needed. In addition, influence of air temperature on the nitrate-rich factor should not be ignored, due to volatile properties of particulate ammonium nitrate.

Line 186: please show evidence to further demonstrate influence of regional transportation on the F1, when you addressed it here.

Lines 187-189: the diurnal variations of RH and temperature can be plotted to support "high RH condition during the day". But it's not convinced to address that no obvious diurnal variation of sulfate-rich factor could be mainly due to the reason here. Other reasons should be further discussed. In addition, the conclusion of the sentence (lines 188-189) is weak, which is not sufficiently supported by the results.

Lines 192-194: This sentence could be modified by "The factor 3 (F3) profile presents a high abundance of EC, OC, Ca, and Cu, as well as it contains some organic tracers (PAHs and organic acids, etc.), . . .". Lines 192-194: In addition, what does contribute to 11.3% of the total PM2.5 mass on average? An information to be needed to transfer this sentence to the next, to indicate this factor could be linked to vehicle exhaust. Otherwise it's hard to understand if this factor 3 is associated with vehicle exhaust source when you start to discuss it in the next sentence. What does this "accounting for 16.2% to OC" refer to, the total vehicle exhaust emissions or only POC?

Lines 194-196: This sentence is not clear. Please revise it.

Line 197: what is "daily variation"?

Lines 197-198: In addition, "xxx" has high correlation with NO<sub>x</sub> and CO, respectively,

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...? Mistakes in “R=67” and “R=58”.

Lines 198-201: Needs to revise these sentences for better understanding.

Line 204: “loads” -> “loadings. Were you discussing on profile in this sentence? Please revise this sentence.

Line 207: The “F4” needs to be defined.

Lines 207-209: I think this sentence can be better improved.

Line 210: a mistake in “R=49”.

Lines 209 – 210: I don’t understand why you discuss on “F2” in the first phase of this sentence, then you get conclusion about “F4”?

Line 210: “source.The” -> “source. The”. What does “the factor” refer to?

Lines 210-213: More discussions to be needed to further support “suggesting the benefit of the online high-resolution measurement.”.

Line 213: “measurement.The” -> “measurement. The”

Lines 213-214: “are often used in industrial processes” -> “are often used as industrial emission markers”. But I am not sure if you did this meaning? In addition, the figure that you are discussing should be pointed out.

Line 215: a mistake in “R=59”.

Line 215: what did you discuss in this sentence “No diurnal variation was observed.”? This is not clear at all.

Line 215: “The factor” refer to the factor 5?

Lines 216-217: This sentence can be improved for better reading.

Lines 217-218: how could you prove your factor 6 that mainly comes from ship transportation? Further discussion/evidence to be needed.

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Line 218: again, this sentence is not clear: “The diurnal contribution at night was greater than that during daytime.”. Please the authors carefully check such similar issues over the entire manuscript.

Lines 219-220: to me “the contribution (10.7%)” is relatively high, but I don’t think “is not negligible” is an appropriate use here.

Lines 228-229: Mistakes in “R=59” and “R=56”.

Line 230: “such as PAHs present in the source” -> “such as PAHs presented in the source profile”? or a figure you discussed should be linked here. “The results are different” -> “These results are different”

Lines 232 – 233: the two factors should be discussed separately.

Lines 234 – 236: literatures should be cited here to support this statement. In addition, “K” needs to be defined.

Line 236: “This factor, contributed 4.9% and 5.7% of total PM2.5 and OC mass on average, respectively.” -> “On average, this factor contributed 4.9% and 5.7% to the total PM2.5 and OC mass concentrations, respectively.”

Line 237: “biomass burning contains high-loading” -> “the source profile of biomass burning factor contains high loadings . . .(Fig. ?)”.

Line 238: “(including coal combustion, biomass burning, etc.)” -> “(including coal combustion and biomass burning, etc.)”.

Lines 239-241: To be suggested to discuss the two factors, biomass burning and cooking, separately.

Lines 242 – 243: which figure/table did you discuss in this sentence?

Line 243: This value “14.7%” is not a major fraction, but it can be called as a relatively high fraction.

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Lines 244 – 245: Do the authors mean that this SOA factor could be associated with a mixture SOA factor from anthropogenic and biogenic sources? One could argue very small fraction of biogenic emission during wintertime in Shanghai region. In addition, it is not sufficient by the similar diurnal variations between nitrate and SOA to indicate that they have similar formation processes and interaction between anthropogenic-biogenic sources. Further discussion is needed to clearly explain such a factor and its potential formation processes.

Lines 258 – 264: those sentences should be discussed associated with your results when you need them to support something. Or if only for a summary purpose here, those contents could be moved into the introduction section.

Line 270: “It can be seen from Fig.7 that the composition varies with the source of the air mass.” -> “As shown in Fig. 7, the PM<sub>2.5</sub> sources vary evidently associated with different air mass origins.”

Lines 273-275: those sentences can be improved to be better read.

Line 276: cluster 1 is not associated with North China, but it could be more reasonable coming from eastern China.

Lines 283 – 284: could be possible regional transport contributing to dust observed at this receptor site? Wind analysis together with dust source could help to further investigate this point.

Lines 287 – 292: the clusters 3 and 4 could be associated with relatively clean air masses originated from ocean areas (see Fig. 6). The differences between clusters 3 and 4 are a long-range and a short-range transportation characterization, respectively. More locally-formed pollutants could be expected observed within cluster 3 air masses, while more regional pollutants could be linked to cluster 4.

Lines 297-299: Please rewrite this sentence.

Lines 299-300: Please revise this sentence for better reading.

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Line 312: why the authors used the different HYSPLIT model versions in this paper? It should be declared in the main text as well.

Lines 330-331: do the authors have any evidence to support this statement: "secondary sulfate factor may also be affected by primary emission of coal combustion."?

Lines 331-333: this sentence can be better improved.

Lines 333-335: give a link (e.g., figure or table) to your results what you are discussing.

Lines 335-336: a same comment as the above, give a link. In addition, which air mass trajectories are shorter, to support your statement "the dust source is more affected by local and short-distance air mass transport."? It can be additionally/more directly helped to check wind dependence of your source factors (especially for local factors), in addition to air mass analysis.

Lines 342-335: very weak evidence from the present study to support this sentence.

Lines 345-346: again, do you have evidence to support this? If yes, please show that.

Line 347: I guess this section might be better moved to methodology section or at beginning of section 3.1. Moreover, I guess it would be also worthful for doing correlations (with R and slope values) of individual component (e.g., nitrate, sulfate, ammonium, POC, SOC, EC, Ca<sup>2+</sup>, and K<sup>+</sup>) between the two different PMF analysis.

Lines 357-358: This sentence can be divided into two sentences to address the two different things. In addition, line 357 "is shown" -> "are shown"; line 358 "are show" -> "are shown".

Line 359: "Table S7.Compare" – "Table S7. Compare"

Lines 384-402: I think this conclusion section needs a major revision to highlight the key findings and further atmospheric implications from the present study, in addition to some technical corrections what I have listed below:

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Line 385: “a typical city” -> “a megacity”

Line 388: what does “contribution of secondary pollution sources” contribute to?

Line 392: Please rewrite this sentence.

Lines 400-402: Very common statements, since they are well-known knowledges and have been reported by many previous studies. As I commented above, the authors would be able to better introduce what the key findings/implications supported by the present study.

Comments on figures:

Figure 1 should be improved by adding a larger scale map to indicate where Shanghai is located in China or eastern Asia.

Figure 2 and Figure 8 should be merged together by a single figure. There is no need to repeat PM2.5 temporal variations again in the main text. If you wanted to highlight its relation with wind direction, you can add wind direction in Figure 2.

In figure 3, I think it's better to draw the time series of those sources with lines than using solid-filled areas, since we cannot see clearly if there is a missing gap for the sampling.

Figure 4: Again, without proving your reasonable PMF analysis using the data from different sites, it isn't convinced to accept these quantitative results.

Figure 5: why did the authors show two-hour resolution diurnal variations instead of using hourly, since you have hourly data as mentioned in previous section? I suggest to use hourly.

Figure 6 and Figure 7: I believe that it can be more visualized to merge the two figures within a single one, e.g., different clusters associated with different pie charts.

Figure 9: a same as Figure 4.

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Figure 11: The mass concentrations of those source factors can be also presented here to better understand the differences between the different PMF analysis methodologies.

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