

Interactive comment on “Exploration of PM_{2.5} sources on the regional scale in the Pearl River Delta based on ME-2 modeling” by Xiao-Feng Huang et al.

Anonymous Referee #1

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General comments:

Huang et al. investigate the sources of PM_{2.5} in the Pearl River Delta (PRD) region of China, determining whether the sources are local or regional and how they vary under different meteorological conditions based on six sites representing urban, suburban, and background locations. The authors present detailed chemical composition results from data collected at each of the sites for approximately one month during each the main four seasons to represent the variability during a full calendar year. Both PMF and ME-2 were applied to the data to identify potential sources of PM_{2.5} in the area, which were subsequently correlated with meteorological conditions, such as monsoons, to

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further identify the importance of each of the sources including local versus regional nature and temporal significance. The authors compared the findings from this work with previous studies in the same areas as well as putting the results into a global context. Ultimately, the authors were able to identify key emission sources and locations that should be targeted in future pollution control measures.

Although the scientific quality of the work is good, the authors do not obviously highlight the uniqueness of this study. The data presented are new thus add to the scientific knowledge and understanding of the PRD and the methods used, particularly ME-2, appear to be novel in that they are applied to a unique dataset. If this is the case, the authors should include a sentence or two in the appropriate places within the manuscript (e.g. abstract). The scientific methods and assumptions are valid and the results are generally sufficient to support the interpretations and conclusions although some additional evidence or explanation is needed (see specific comments). Numerous pie charts are presented in the manuscript; the authors should consider moving some of these to the supplementary material to reduce the length of the manuscript or use a different style plot to distinguish between the different types of results being presented. The figures currently in the supplementary material need to be greatly improved in terms of clarity of the images as well as the addition of legends where possible. The manuscript generally flowed well but it could do with some slight reordering, especially the section describing the meteorological conditions, to make the manuscript flow even better.

Despite there being some major points that need to be revised, the overall quality of the work presented and manuscript itself are good; an interesting and enjoyable read. Once the revisions above and the comments below are addressed, I recommend this manuscript be published in ACP.

Specific comments:

Abstract, line 21: It is stated that the ‘regional annual average PM_{2.5} concentration was

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determined'. This is misleading as there were only ~4 months of measurements spanning the year, with samples taken every other day. Although those four months may be representative of the main pollution conditions, it should be mentioned or clarified that a full year of data was not obtained to determine/estimate this annual average.

Introduction, lines 50-51: Why are these noteworthy provinces? Please consider adding a few words as to why these are being highlighted.

Introduction, line 62: It is stated that the previous studies in the PRD provinces 'lacked the extensive representation of the PRD'. Please qualify this statement e.g. is it because only single locations were studied and that is the uniqueness of this study as several locations are studied at the same time.

Introduction, lines 65-67: Despite some of PMF's limitations, it is the first step for the application of ME-2 to a dataset. Further, as PMF does not require a prior information, new sources could be identified as a result both in terms of newly identified as a source in a given location or a newly identified emission source overall. Please add a sentence or two to acknowledge that PMF is usually the first step in factor analysis using ME-2, especially as the a priori information used for running ME-2 typically uses the factor profiles identified from PMF and/or identifies a number of factors that should be considered when running ME-2.

Introduction, lines 69-74: As mentioned, organic aerosols have been successfully apportioned using ME-2 via SoFi. As this study uses both organic and inorganic species it would be good to point this out as being novel. If it is the first study of its kind to apply the model to this dataset (in terms of the species and/or measurement period and location) then this should be highlighted in the manuscript in the appropriate places such as the abstract and later in the introduction. If this is not a unique case then something along the lines above should be mentioned in any case along with a citation of similar cases for comparison.

Section 2.1, lines 93-94: The current way in which the sampling periods are described

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are misleading as 'January-February' could be interpreted as being two full months whereas in fact it is a period of one month spanning two months. Add a few words clarifying that each sampling period for the seasons is one month and refer to table 2, where the exact sampling dates are noted.

Section 2.1, lines 100-101: 'two different types of samplers sampled' – clarify that it is the two samplers that were used in this study that were compared. The results of the inter-instrument comparison 'yielded a relative deviation of less than 5% for PM2.5 mass concentrations'. How many samples were obtained for this comparison? How was the 5% calculated/determined? Please consider adding something to the manuscript on this.

Section 2.1, in general: There is no mention of the exact number of samples that were obtained and whether there were any issues with any of them. Are all ~15 samples from each season valid and run as intended? What QA/QC was performed on the samples (standard laboratory QC and overall QA)?

Section 2.2, lines 146-147: Please provide examples of the 'sources of uncertainty that contributed little to the total uncertainty'.

Section 2.2, lines 159-160: Please expand on why a factor of 2 was applied to the estimated uncertainties. Specifically, please explain why a factor of 2 was chosen. If this is this a typical factor to apply, please provide a reference.

Section 2.3, line 168: It is not typical to refer to later sections in a manuscript. Perhaps consider summarizing what is in the later sections here or rephrase this sentence so that Section 3.2 can be referenced but the reader does not have to read that section at this point.

Section 3.1., lines 211-215: Figure 3b does not show that the seasonal variations in the major PM2.5 components were correlated with monsoon characteristics. Please expand on this, clarify, and/or provide additional evidence for this statement. Similarly,

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please expand on/clarify how figure S1 shows that the northern monsoon prevails in winter and the southern monsoon prevails in summer.

Section 3.2, lines 251-257: Please comment on why it might not have been possible to separate the secondary sulfate and LV-OOA as two separate factors as well as SV-OOA and secondary nitrate. Having a mixed factor is something the authors note as being a downfall of the PMF results so it needs to be acknowledged that even with SoFi there is a mixed factor. To confirm LV-OOA/OOA-1 and SV-OOA/OOA-2 factors in other studies, the time series is correlated with that of sulfate and nitrate, respectively. Perhaps the time series in this study are so similar that it was not possible to separate each of them into individual factors, although this is surprising when OM is a large contributor to PM2.5 at all of the sites.

Section 3.2, lines 278-279: Please expand on exactly how the SOA is calculated here. Is it a percent of each of the sulfate and nitrate fractions based on the contribution of OM to each factor?

Section 3.2, line 292: What are the unidentified sources? Is it the residual from ME-2?

Section 3.3, lines 309-313: In other locations e.g. in Europe, secondary sulfate is typically a regional source so perhaps comment on whether it is typical in this area for sulfate to be a more locally influenced source. Also, regarding the correlations with meteorological conditions, mention that temperature also plays a role, especially in influencing ammonium nitrate concentrations.

Section 3.3, lines 324-326: This statement emphasizes the importance of running PMF as a first step for identifying a range of possible sources, both typical and atypical sources. It would be worthwhile indicating this in the manuscript.

Section 3.3, lines 330-332: Although in Tao's study, the ship emissions study may not be a pure primary source, it could still be representative of ship emissions in general even if it's more of a limitation of the PMF output. In contrast, in this study ME-2 was

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used so sources are better separated yet the secondary sulfate and secondary nitrate factors comprise some organics. However, they are still likely secondary sources as the OA component is likely secondary also. Perhaps the naming of the secondary sulfate and secondary nitrate factors should be re-considered or clearly described in the text as being a predominantly secondary sulfate factor, for example.

Section 3.4, in general: The first part of this section, up to and possibly including Table 6, should be moved to earlier in the manuscript. Perhaps add it as a sub-section in the methods as a description of the different meteorological conditions and the links between wind direction, season, and monsoon. This would significantly help interpretations of the data earlier in the manuscript.

Section 3.4, lines 381-386: The average concentration of the ship emission source was similar between the two flows though. It is important to note this as the sources are referred to in terms of the average for the whole region during the different seasons/monsoons at other points in the manuscript.

Section 3.4, lines 417-419 and 443-444: The authors mention that the spatial distributions and source characteristics of secondary sulfate and secondary nitrate also reflected the corresponding characteristics of LV-OOA and SV-OOA, respectively. This is a circular point. The likely reason that each of the two sets of factors are not separated into individual factors is because the characteristics are similar between secondary sulfate and LV-OOA, for example. Temporally they will likely be the same and time-series are one of the main inputs for factor analysis. The sentences do not really make sense as the factor is a combination of the two, so of course they will show the same characteristics as there is only one output representing both sources. Please re-phrase and expand on this point. Similarly to an earlier comment on this, perhaps considering re-naming these factors would help reduce any confusion surrounding there not being separate LV-OOA and SV-OOA factors.

Section 3.4, lines 459-464: Have the authors considered the influence of residential

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biomass burning? In other locations few/no coal-fired power stations, the biomass burning factors are typically associated with residential space heating and other residential activities. Are the coal-powered power stations here so dominant that residential biomass burning is negligible or is such an activity not typical in this region?

Figures and Tables:

Table 3, page 6: Please reduce the spacing of the factor names so that it's clear there are only four factors as it currently reads like there are six.

Figure 3a, page 8: Please add a line to the figure caption explaining the differences in size of the pie charts. Also, clarify that the number in brackets next to each of the abbreviated site names is the concentration; currently the figure caption only details the units.

Table 4, pages 8-9: There have been more recent studies in some of the locations detailed in the table e.g. the ClearfLo project in London spans several years, with 2012 being the main year of measurements, and there are several publications from this project alone. Perhaps other projects in these locations could be cited in the main text. Further, please explain and maybe add a sentence in the text as to why the particular studies are listed in the table for comparison e.g. the studies use similar methods and/or present similar results (in terms of the species measured) to allow for a better comparison with the current study.

Table 5, pages 12-13: Please rearrange this table so that the comparable studies in the same locations are next to each other i.e. group the studies in the table that were performed in Guangzhou etc. In addition, perhaps a small comment/note would be good on how the traffic source in the Huang et al study compares to the vehicle emissions source in this study (e.g. does one include tire/brake wear and the other doesn't).

Figure 9, page 17: Please add some more information such as a key or legend to the figure. For example, what are the triangles? A scale and N arrow would be useful

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also. In the figure caption the 'shaded area' is noted as indicating the 'key emission area'. Firstly, perhaps a pattern could be used instead of red shading so as to prevent confusion that it represents the secondary sulfate source emission area. Secondly, please clarify if the 'key emission area' is for multiple different sources and reference the text in the manuscript where this is described further. Finally, the authors may wish to either move this figure to earlier in the manuscript or refer to it earlier in the text such as around line 380.

Supplement:

Table S2: Where did the 'PRD-annual' column come from? How were the numbers determined? It does not appear to be an average of the enrichment factors from the six sites listed in the rest of the table. Also, if the final column is based on the data collected in this study, then please add a note that the 'annual' is an estimation based on the four months of data collected during the study as opposed to 12 full months of measurements.

Figure S1: The image quality needs to be significantly improved. A key/legend, scale, lat/long details, and a N arrow should be added where possible and a couple of sentences explaining if the colors represent certain time periods, for example the purple/blues are for older dates and yellows are for newer dates (if a color-time scale is not available). Information in the caption needs to be added regarding the details of the trajectories themselves – are they 24-hour trajectories; were there any particular criteria entered for running them.

Figures S3 and S4: These figures need to be significantly improved to be clearer (currently they are fuzzy), include additional information such as the dates each of the six boxes represent and legends and scales. Some of these may be included in the small text boxes in the top left of each grid but these are currently not clear.

Figures S5 and S6: Similarly to the above, these figures need to be improved by sharpening the quality and clarity of the figures as well as including keys and scales where

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possible.

Minor and technical corrections:

Abstract, line 15: Possible typographical error as the meaning of 'ever experience severe PM2.5 is not clear. Please rephrase.

Abstract, line 28: A space is needed between the end of 'burning' and the percentage '(11%)'.

Section 2.1, line 108: DRI has a new model analyzer so if possible please add a model number for the instrument used in this study assuming it is the older analyzer.

Section 2.2, line 122: A space is needed between 'F' and 'are'.

Section 2.3, line 172: Please define 'EV'. It is defined later in the manuscript but this is the first occurrence.

Section 3.1, line 202: Here it is stated that 'trace elements accounted for 6.2%' but figure 2 indicates that trace elements contribute only 1% and 'others' contribute the 6.2%. Which is the correct number?

Section 3.1, line 211: Possible typographical error as the meaning of 'the dominant northeastern wind the year' is not clear. Please address.

Section 3.1, line 225: Please explain what yellow label vehicles are.

Section 3.1, line 232: Please provide example references to the studies performed in each of the cities listed and/or refer to table 4 where there are references.

Section 3.3, line 323: A space is needed between 'years' and '(People's Government'.

Section 3.4, lines 355-356: Please briefly comment on the other types of flows e.g. easterly flow?

Section 3.4, lines 390-393: Is there something that can be used as further evidence or to reference the road construction noted here or is it based on local knowledge?

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Section 3.4, lines 426-427: Show the coal-fired power plants on the map in figure 9.

Section 3.2, line 444: possible typographical error: this is meant to be SV-OOA instead of LV-OOA.

Section 4, line 519: Was this meant to read 'in recent decades' i.e. plural decades?

Interactive comment on Atmos. Chem. Phys. Discuss., <https://doi.org/10.5194/acp-2018-257>, 2018.