

## ***Interactive comment on “Exploration of PM<sub>2.5</sub> sources on the regional scale in the Pearl River Delta based on ME-2 modeling” by Xiao-Feng Huang et al.***

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General comments: This study apportioned the sources of fine particles in the Pearl River Delta (PRD) region of China using both PMF version 5 and ME-2 methods. The authors found that ME-2 model could produce better results than the PMF model. Ten sources of PM<sub>2.5</sub> were found in the PRD region including secondary sulfate (21%), vehicle emissions (14%), industrial emissions (13%), secondary nitrate (11%), biomass burning (11%), SOA (7%), coal combustion (6%), fugitive dust (5%), ship emission (3%), and aged sea salt (2%). Furthermore, authors identified the source contribution from both local and regional emissions.

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In general, the scientific content in this manuscript is good for publication. However, I have some comments that I hope it could help author improve their manuscripts.

Major comments:

1, Line 109: The authors assumed OM/OC is 1.8. This ratio seems too high for me. According to He et al. (2011), the OM/OC is 1.6 for the urban areas. Could the author explain for this ratio? In addition, why do you use the OM, not OC as the input variable in the model? I think OM/OC ratios should vary following the sampling days. Therefore, if you input the OM instead of OC in the model, it will cause more uncertainties. How did the authors calculate the uncertainty for the OM?

Reply:

We agree that the OM/OC should vary to some extent from sample to sample, although this ratio is difficult to measure and usually fixed at a constant. However, an advantage of fixing the OM/OC at a constant is that additional uncertainty can be avoided in the transformation from OC to OM, since the columns of G (factor time series) are normalized in the model calculation process (Paatero et al., 1994). Thus, it is the same using OM or OC in the model. In previous aerosol mass spectrometry measurement for PM<sub>1</sub>, the OM/OC ratio was measured to be 1.6 for urban atmosphere (He et al., 2011) and 1.8 for rural atmosphere (Huang et al., 2011), we adopted 1.8 for the six sites (including urban, suburban, and background atmospheres) because it is assumed that the difference between PM<sub>1</sub> and PM<sub>2.5</sub> may contain more aged regional aerosol with higher OM/OC, which has been explained in the revised text.

2, PMF model vs ME-2 This study compared the PMF and ME-2, but I cannot find the information which shows how the authors conducted the PMF in details. I suggested that the author should write more about PMF version 5.0, what is difference between PMF v5.0 and ME-2. For example, in PMF v.5, they also have constrained factor functions, did the authors use this function to constrain the factor? In addition, the authors should write more how they select the number of the factors and optimize the

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PMF results. I would be grateful if the authors show correlations between the PMF and ME-2 results.

Reply:

More details of the PMF running have been provided as below: "After examining a range of factor numbers from 3 to 12, the 9-factor solution output by the PMF base run ( $Q_{true}/Q_{exp}=2.5$ ) was found to be the optimal solution, with the scaled residuals approximately symmetrically distributed between  $-3$  and  $+3$  (Fig. S6) and the most interpretable factor profiles (Fig. S7). The model-input total mass of the 18 species and the model-reconstructed total mass of all the factors showed a high correlation ( $R^2=0.97$ , slope=1.01) (Fig. S8). The factor of biomass burning was not extracted in the eight-factor solution, while the factor of fugitive dust was separated into two non-meaningful factors when more factors were set to run PMF."

More descriptions about the difference between PMF and ME-2 are added as below: "SoFi is a user-friendly interface developed by PSI for initiating and controlling ME-2 (Canonaco et al., 2013), and it can conveniently constrain multiple factor profiles. Although USEPA PMF v5.0 can also use some priori information (such as ratio of elements in factor) to control the rotation after the base run, it is not able to use multiple constrained factor profiles to control the rotation (Norris et al., 2014). Therefore, SoFi is a more convenient and powerful tool to establish various constrained factors for source apportionment modeling."

A comment is added for the comparison between PMF and ME-2 results in Section 3.2: "Although these nine factors of the ME-2 modeling generally showed high correlations ( $R^2=0.81-0.97$ ) with the corresponding factors of the PMF modeling in terms of time series, it is easy to see that the ME-2 modeling provided a better..."

Line 164:  $Q_{true}/Q_{exp}=2.5$ . Could the authors explain why they use the  $Q_{true}/Q_{exp}$  ratio of 2.5 to optimize the solution? I think the ratios depend on the number of factors and the uncertainties. Did the author add the extra uncertainty in the PMF model?

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Reply:

Yes, the  $Q_{true}/Q_{exp}$  ratio depends on the number of factors and the uncertainties. Ideally, if the model entirely captured the variability of the measured data and all uncertainties were properly defined, a  $Q_{true}/Q_{exp}$  value of 1 would be expected. We did not intend to say the  $Q_{true}/Q_{exp}$  ratio of 2.5 is the best value, but intend to monitor this value and compare it to that of the ME-2 solution. In result, the ratio of the ME-2 solution (1.2) is closer to 1.0, indicating that the species residuals had decreased and the ME-2 solution should be more reasonable. Extra uncertainties in this study were added as below: "The uncertainties of  $SO_2$ ,  $NH_3$  and all metal elements, which have scaled residuals larger than  $\pm 3$  due to the small analytical uncertainties in Table S3, need to be increased to reduce their weights in the solution (Norris et al., 2014)." The above points have been clarified in the revised manuscript.

Other minor comments:

1, Line 172: Please define "EV"

Reply: Suggestion taken.

2, Line 205-206: I think the much lower concentration of PM2.5 at DP because this sampling site near the sea therefore the air pollutants are more diluted. I am not really clear why low PM2.5 concentration at DP indicate the large contributions of pollution transported from outside region? Could the author explain for this?

Reply:

To make the point clearer, we have rephrased the text as below: "The DP background site had little local emission and was hardly influenced by the emissions from the PRD under both southerly flow and northerly flow. Thus, its air pollution reflects the large-scale regional air pollution. The average PM2.5 concentration at DP was as high as  $28 \mu\text{g}/\text{m}^3$ , indicating that the PRD had a large amount of air pollution transported from outside this region."

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3, Line 227-230: The authors compared the PM<sub>2.5</sub> between the cities. This comparison is not meaningful to me because the authors compared the levels at different time periods. For example the PM<sub>2.5</sub> levels at Beijing and Tianjin were measured in 2012-2013, while the PM<sub>2.5</sub> concentration measured in this study was in 2015. Please note that after 2012, the PM<sub>2.5</sub> trends at Beijing and Tianjin also showed a huge decrease under the “Control Action Plan”. I suggest the author should update the PM<sub>2.5</sub> level in the Table 4.

Reply:

Updated with more recent data available in the literature.

4, Line 252-256: Could the author explain “high OM concentration was considered to present the LV-OOA” and “high OM concentration was considered to represent SVOOA”? Could you please discuss more about that: why the (NH<sub>4</sub>)<sub>2</sub>SO<sub>4</sub> associated with LV-OOA and NH<sub>4</sub>NO<sub>3</sub> and SV-OOA shared same source?

Reply:

The following discussion has been added into the revised manuscript. “In this study, secondary organic aerosol (SOA) did not appear as a single factor, even if we run the ME-2 with ten or more factors. SOA can usually be described by low-volatile oxygenated organic aerosol (LV-OOA) and semi-volatile oxygenated organic aerosol (SV-OOA), based on the volatility and oxidation state of organics (Jimenez et al., 2009). In previous studies (e.g., He et al., 2011; Lanz et al., 2007; Ulbrich et al., 2009), the time series of LV-OOA and SV-OOA were highly correlated with those of sulfate and nitrate, respectively, implying that LV-OOA and sulfate (or SV-OOA and nitrate) cannot be separated easily in cluster analysis, especially when there is no effective tracer of SOA. In this study, the high OM concentration in the secondary sulfate-rich factor was considered to represent LV-OOA, while the high OM concentration in the secondary nitrate-rich factor was considered to represent SV-OOA (Yuan et al., 2006b; He et al., 2011).”

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5, Figure 8: Regarding the aged sea-salt factor, the contribution of this factor at QA and HS sites from the northerly flow was higher than those from the southerly flow. Could you explain for that?

Reply:

The following discussion has been added. “The spatial distribution of aged sea salt among the different sites was a complex result of the site locations relative to the sea and meteorological conditions, e.g., wind and tide. A relatively high level of aged sea salt was observed at the Qi-Ao Island (QA), especially in the northerly flow, which can be attributed to that the QA site was surrounded by the sea and had lower wind speeds in the northerly flow (in Table 3).”

6, Line 527: A typo-mistake “theMe-2”.

Reply: Corrected.

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