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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Spatiotemporal variations of ambient PM_{10} source contributions in Beijing in 2004 using positive matrix factorization" *by* S. Xie et al.

S. Xie et al.

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We sincerely appreciate your carefully reading our paper and detailed comments, which we believe will improve the quality of this paper. The responds to your comments are as below:

1. This paper examines seasonal and spatial variations of ambient PM10 sources in Beijing by applying Positive Matrix Factorization. While this receptor models have been used worldwide to study source characteristics of PM10, this paper is very much similar to a companion paper Song et al. [AE, 2006, 40, 1526-1537] which was to apply PMF to a PM2.5 dataset in Beijing. Conclusion drawn in this paper shows little beyond our understanding based on the study of Song et al.

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Discussion Paper

I was also one of the authors of the mentioned paper Song et al. (2006). It was our first attempt to analyze PM2.5 sources in Beijing in 2000, through which we obtained elementary experiences in applying PMF model. Considering controlling the high PM10 concentration level in Beijing is pretty difficult, a special project was funded by the Beijing government to investigate the PM10 sources and the corresponding control measures. Based upon the forgoing studies and the characteristics of PM10 mass concentration distribution, we set up six sampling sites to investigate PM10 sources. This study was different from PM2.5 observation in 2000, with great improvement in selecting sampling sites, collecting particle samples and application of the PMF model. Furthermore, great differences existed for PM sources in 2004 and 2000, as well as for PM10 and PM2.5 sources. Two examples are, the two dust related sources in this study accounted for far more than those in PM2.5 in Song et al. (2006); industrial source were identified in Song et al. (2006), but was not found in PM10 apportionment in 2004.

Another point that should be mentioned is the sizes of the data sets for previous PMF analysis in Beijing are very small, e.g. 100 samples from 5 sites in Song et al. (2006), 115 samples from 1 site in Yuan et al. (2008), 26 samples from 1 site in Cao et al. (2004), and 61 samples from 2 sites in Jing et al. (2007). Since PMF need large number of data points to obtain stable solutions, the results in these studies might be not reliable. With more than 240 samples in this study, we identified the spatiotemporal distributions of PM10 source contributions over Beijing, especially the spatial differences. This was different from Song et al. (2006).

2. Before applying PMF for source analysis, it is good to give some details in the manuscript about some of the basic findings for PM10 in Beijing. What were the ranges of mass concentration of each species observed? What were the seasonal variations? What were the correlations between different PM species? How do these observations compare to those in other large cities around the world? Information like this would help the reader to appreciate the extent and quality of the data, as well as the differences

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and/or similarities with other regions of the atmosphere.

We accept these comments and will try our best to revise according to them. We would like to mention that we did consider presenting the PM10 basic findings when preparing this manuscript. But considering our theme as PMF source apportionment and the appropriate size of the paper, we decided to briefly introduce some basic characteristics of PM10 in Beijing in section 3.1, with Fig. 2 presenting the seasonal variations of PM10 major constituent concentrations. In order to control the paper length, we did not present the variations for each species, nor did we compare to other large cities. Instead we focused on specifying and reasoning the PMF findings. Based on these comments, we will add a figure presenting the species concentrations in PM10 in our revised manuscript.

3. The most important thing of doing PMF is to select the number of sources that could best explain the measured pollutant concentrations. Although mentioning a number of parameters that could justify the PMF performance, it is not conveyed clearly at all how the seven source categories were determined and why they came up with the ones they did. Like many, the authors tend to over-interpret the factors found PMF. They keep increasing the number of factors as long as they could find some information in the publication pool to justify their results. With only 40 samples available, there is not enough variability present in so few data points to be able to pull out as many as seven factors. A good example of how to use PMF for atmospheric data sets, I believe, was described by Lanz et al. [ACP 2007, 7, 1503-1522]. These authors systematically increased the number of factors in a PMF analysis (of a particle composition data set) from 2 to 6 and showed the results from all the different analyses. As a result, the reader gets a good idea of (1) why so many factors were needed to describe the data, and (2) how robust the contribution from the major factors is as a function of the number of factors. Therefore, detailed information regarding the model performance is a must when reporting PMF results.

Thanks for these reasonable comments, which in fact were also some open questions

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for PMF analysis as a method of source apportionment. We determined the possible factor numbers by referring to the values of Q, eij/σij, and the coefficients of multiple linear regression; as well as by assessing physical meaning of the factors. Then we get final factor numbers, namely the possible number of sources. Considering our focus as obtaining the spatiotemporal distributions of PM10 source contributions based on PMF results and the length of the paper, we did not specify the process of determining factor numbers. We will do this by referring to Lanz et al. [ACP 2007, 7, 1503-1522] in our revision. As for some parameters used in PMF analysis, such as romat, although there have been some studies using them to determining the factor numbers (Lee et al., 1999; Lanz et al., 2007), their applicability need to be further investigated (Paatero, 2000). The rotation problems for the PMF resolved factors and the uncertainty analysis of the results are still unsolved questions and, factually, the focuses of PMF study (Rizzo and Scheff, 2007 AE). Considering there were some existing papers discussing such topics and our major topic, we did not put them in our paper.

4. Therefore, it is not difficult to understand why there are many abnormal findings associated with the PMF results which the authors ignored to discuss. In source 2, significant amount of Na far larger than Al, Ca, and Fe is associated with the crustal soil, and this does not appear in Figure 4. Arsenic (As), a typical tracer for coal combustion, appears in the vehicle emission source rather than in coal combustion. In source 5, ion balance is apparently not reached with NH4 concentration around 1/100 of NO3 concentration. In source 7, OC concentration is almost ten times EC in vehicle emissions. Considering the typical OC/EC ratio of 1-2 in vehicle emissions in China (e.g. Cao et al. [AE, 2003, 37, 1451-1460]; Louie et al. [AE, 2005, 39, 1695-1710]), we may believe for sure that significant secondary OC is mixed in this source, etc.

These problems mentioned above have been noticed in our study and we tried to find more reasonable solutions by numerous trials. At last, we chose the results in this paper which was proved to be the best in our trials. In general, the resolved profiles

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accorded well with the corresponding source characteristics, so we thought them to basically reflect the characteristics of PM10 sources in Beijing. As for the existing different points from previously reported source profiles, e.g. the high Na+ in source 2, unbalanced NH4+ and NO3- in source 5, OC as 10 times of EC in source 7, as well as the element As appearing in vehicle emission source, which in fact was found by Song et al. (2006), too. We thought that more investigations on source profiles of vehicle emission, coal combustion, crustal soil, and urban fugitive dust were needed to achieve nearer form the truth. Meanwhile, quality control in aerosol sampling and analysis, as well as improvement of the PMF method will also help.

5. Specific comments:

(1)Line13, pp577, K should not be included in the calculation of crustal elements. K, as measured by IC, is present in the ion form rather than element form in the particles.

Accept. This was a mistake in writing this paper. K was factually measured by ICP-MS, not IC, and therefore included in crustal elements.

(2)Line10, pp578, the reference Hua et al. 2006 is not present in the reference list.

Accept. Thanks for reading carefully. We will put Hua et al. (2006) in the reference list.

(3)Line4, pp580, the fact that only 3-4% of waste in Beijing is treated y incineration does not necessarily indicate that the overall contribution from waste incineration is negligible. This source might be associated with significant PM emission intensity.

Accept. We will revise this. According to the apportionment results, waste incineration characteristics were found in source 7. This was the first time to identify its possibly important influence in Beijing, but we still need more evidences.

(4)Line8, pp580, it would be good if the authors could provide the percentages of fuel usage of gasoline and diesel in Beijing to justify their apportionment results.

This sentence in Line8, pp580, " For diesel emissions, EC was higher relative to OC

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than in the gasoline vehicle source profile." meant generally EC/OC is higher for diesel emissions than gasoline emissions, but did not mean EC is higher than OC in the vehicle emission source profile in Beijing. In order to eliminate this possible misunderstanding, we will do some revision here. Meanwhile, we accept the suggestion to provide the percentages of fuel usage of gasoline and diesel in Beijing to justify our apportionment results.

(5)Line27, pp583, the reference Duan et al. (2004) seems to be quoted irrelevant to the preceding discussion. The main point here is that non-local biomass burning might be transported to Beijing together with crustal soil in March and April. As most of the crustal soil was blowing to Beijing by the northerly/northwesterly wind bypassing the dessert area to the N/NW of Beijing, those straw burning emissions in the provinces to the south of Beijing could not make any contribution to the ambient PM in Beijing at this time.

Accept. We will revise it. Here we were indeed talking about contributions from nonlocal biomass burning. In March and April, there might be influences by forest fires and other biomass burning from the north, and in other months, there might be influences from the south.

(6)As more and more attention is paid to the source characteristics of PM2.5 as it is more related to human health, a comparison of source contributions between PM10 in this study and PM2.5 in Song et al. 2006 is suggested, although the data are collected in different years. More in-depth understanding about source characteristics could be achieved through this comparison.

Accept. We will do such comparison with Song et al. 2006 in our revision to get more in-depth understanding about source characteristics for aerosols in Beijing.

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