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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Source apportionment of ambient fine particle from combined size distribution and chemical composition data during summertime in Beijing" *by* Z. R. Liu et al.

Anonymous Referee #2

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This manuscript presents the results of source apportionment analysis based on particle size distribution data combined with chemical composition data. The value of this study could be seen in adding additional information to the already existing dataset on source apportionment analysis, and not in its scientific or experimental novelty. Further limitation of this study is the relatively short time period (August 2011) which doesn't allow any general conclusions regarding the air quality in Beijing, especially as this point is not discussed in the manuscript. It remains unclear for the reader how typical from meteorological point of view was this study period for Beijing. The data are rather adequately and mostly sufficiently analysed, however the interpretation and discussion of the data in not adequate (see comments below).

Specific comments:

1. Introduction:

Please add some relevant studies on PM and health conducted in Beijing. Some examples: Leitte et al., EHP 2011, Breitner et al., STOTEN 2011, Leitte et al., EHR 2012, Liu et al., ER 2013

Page 1370: please add also to the reference list some studies done in the research group of Prof. Hopke who pioneered the source apportionment based on PSD data (Kim et al. 2004, Zhou et al., 2005, Ogulei et al., 2006, Ogulei et al., 2007).

2.1 Sampling site

How representative is the monitoring site for Beijing? Are there any measurements conducted for exploring the spatial variability (at several monitoring sites in parallel)? As the authors state on page 1374 that two main Chinese restaurants are located less than 100m northwest and southeast of the sampling site and as one of the main sources of UFP found in this study was cooking, the question arises whether it was the influence of the two restaurants. To my knowledge one of very important criteria for site selection is that in the vicinity of the site no other important local sources of the specific air pollutant should be present.

2.2 Data description

One of my major concerns is related to data quality and data treatment. No specific information on quality criteria is given for SMPS and APS Instruments. What were the criteria for sheath flow, blower output, DMA temperature and so on. How often were the parameters controlled? Are there any missing data? How were missing data treated in the calculation? Missing data can significantly influence the calculation of the "average" diurnal variation.

The authors stated that the DMPS and APS were merged according to the method of Beddows. Please give more details on that, which software was used, how the merging

routine was applied, was were the results (in the end the values of transition-regime effective densities, as the instruments measure different equivalent diameters which could be converted into each other only by using of an effective density).

Please explain the abbreviations HR-ToF-AMS and IAP (in general all abbreviations should be explained).

2.3 Positive matrix factorization

The authors replaced the missing number concentrations (as well as chemical composition and gaseous pollutants data) with interpolated values. However, it seems (from equation 1) that the uncertainties to these interpolated values were the same as good data. As the interpolated values are not measured (as the others), why same uncertainties were assigned instead of much higher uncertainties that were used in many previous studies? Does the number of factors are solely based on criteria of scaled residuals and Q-value? Please explain more explicitly how the number of factors was chosen. On the other hand, the method of choosing the "right" number of factors by PMF internal criteria is not sufficient. As indicated by Ulbrich et al (2009), "The ambient and synthetic data indicate that the variation of the PMF quality of fit parameter (Q, a normalized chi-squared metric) vs. number of factors in the solution is useful to identify the minimum number of factors, but more detailed analysis and interpretation are needed to choose the best number of factors."

2.4 Conditional probability function

The number of hourly meteorological data in this study is about 31*24=744. Moreover, a considerable amount of data with wind speeds <1 m/s have been excluded (as shown in fig. S1), thus the number of data points assigned to a single wind sector might be too small and insufficient to be statistically significant. The CPF results might be biased in some wind sectors and not reliable (Zhou et al., 2004, http://www.atmos-chem-phys.net/9/2891/2009/acp-9-2891-2009.html). There is no explanation on this throughout the manuscript. Clarifications should be made before CPF results were

used extensively in result section for identifying possible locations of factors.

3. Results

As already mentioned by Reviewer 1 the attribution of factor 1 to cooking is doubtful. Both studies cited by the authors (Li et al., 1993 as well as Buonanno et al., 2009) were conducted indoors and not outdoors. However, due to nucleation and accumulation processes the size distribution of particles from one specific source will change with the distance between the source and receptor site. Therefore, one should be very carefully by comparing the results between different studies, especially between indoor and outdoor microenvironments. Furthermore, I agree also with reviewer 1 that the 40 nm mode is also very typical of urban emissions from traffic. To sum up, I'm not convinced by the presented data, that this factor could be attributed to cooking activities.

Factor 2: the authors stated that this factor has a high positive correlation with NOx. Where could I see it? In the Table 2 the contribution of NO to this factor is 0 and of NO2 7.3, what is definitely lower as the contribution of NO2 to factors 3, 4, 5 and 7. I don't understand why the correlation between NOx (how was NOx estimated) is could be high.

In the next sentence the authors describe the factor 2 using following terms: "condensation mode", "droplet mode", "organic matter" and "sulphate". Whereas the two first terms describe physical properties, the other two describe chemical properties of particles. Please don't mix it in one sentence. I couldn't believe that this factor could be attributed solely to diesel particles. It is rather a mix of carbonaceous particles from different combustion processes. Please, be more cautious with naming of the factors.

Factor 4 and 7: Factor 4 peaks at 150 nm in number distribution and at 300 nm in volume distribution, and the authors name it "regional sources". The naming seems primarily derived from the CPF plots, which is a little confusing as CPF plot also points to the northeast region. The same is true for factor 7. Can it also be regionally transported? The CPF plot points to the southeast region, which is also the industrial area

in Hebei province. The evidence for the naming is not strong enough. Factor 6: Factor 6 contains high fraction (82%) of ozone, it is very likely related to the particles formed from gas phase photochemical oxidation. Based on its unique diurnal variation, the NOx-VOCs photochemical reaction should be the dominant factor, instead of oxidation of SO2.

4. Discussion

Page 1380: "However, because it is difficult to obtain the particle density from different sources, we assume the volume concentration is equivalent to the mass concentration in the present study, making the comparison possible" - this assumption doesn't make the comparison better, it means more reliable. I'm sorry, but volume concentration is not equivalent to the mass concentration, especially if the density changes.

I have some difficulties to follow the comparison of annual averages (Song et al., 2006) or even 5-years-average (Chen et al., 2012) with results obtained for ONE month only (this study). I couldn't believe that any conclusions coming from this comparison are valid.

Other sentence regarding the comparison: "The contribution from secondary nitrate is more or less the same with that of previous studies". The corresponding values in Table 3 are 16% (Song et al., 2006), 8% (Chen et al., 2012) and 5.6 % (volume concentration, this study). More precise description as "is more or less" is needed for a scientific journal (for example: comparable to the Chen study, but lower as observed by Song et al. . .).

Last by not least I can't follow the authors in their argumentation that the secondary sulfate factor 6 is only of local origin (section 4.2 Local and remote secondary aerosol). All previous source apportionment studies using PSD data have the same high temporal resolution of the physical information on particles, some of them have also in addition high time-resolves data on chemical properties of particles. No study discussed such a separation of sulphate particles. It is difficult for me to believe that sulphate particles

could be found in different modes depending on their origin (local generated or long range transported). It could be that the long-range transported particles increase during their journey due to accumulation, but in my opinion such complete separation of both kinds of sulfate particles as the authors postulate is not possible.

Page 1381: "More than one-third of the secondary nitrate and secondary sulfate in Beijing was transported from the surrounding areas of Beijing (Table 2)" - I don't see it in Table 2. Please explain it.

4.4 Implications for future abatement policy

The implication "This result suggests that cutting down emissions from road dust and power plant will effectively decrease the concentration of fine particle and improve the air quality in Beijing" without any further quantification is rather trivial. We already knew it without reading this manuscript. Every limitation of particle emission will lead to decrease of particle concentrations in the ambient air.

Also the implication regarding cooking is not supported by the presented data.

5 Conclusions

The conclusions section in this manuscript is highly generalized and is essentially a quick summary of the manuscript. The authors appear to simply repeat the findings of the work, and make only limited conclusions.

Interactive comment on Atmos. Chem. Phys. Discuss., 13, 1367, 2013.