Anonymous Referee #2

General comments:

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).

[*Response*]: We thank the reviewer's comments and suggests. We agree that the study period of this paper was relatively short and limited its representative to air quality in Beijing; related statement was added in the revised version. However, it should be mentioned that this kind of work was not frequent as mentioned by reviewer 1, especially in megacities of China like Beijing. Furthermore, we hold opinion that the value of this study was to provide ready source emission information based on high-resolution particle size distribution and particle chemical composition data, which could not be achieved in the previous studies conducted in Beijing that based on filter sampling data. As the reviewer suggested, we added more statements in supplementary materials for the description of the meteorological condition during the study period, the statistics of various meteorological parameters in August from 2007 to 2011 was also provided in supplementary materials.

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
[*Response*]: Suggestion taken. See page 2, line 27-30.

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).
[*Response*]: Suggestion taken. See page 4, line 3-4.

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.

[*Response*]: We had examined the representative of the IAP site before this study started. In the year of 2008, the Beijing-Tianjin-Hebei Atmospheric Environment Monitoring Network was established by the Institute of Atmospheric Physics (IAP), Chinese Academy of Sciences. The network monitored atmospheric pollutants in real-time and provided early warnings of atmospheric pollution in the region from June to October 2008 (Xin et al., 2010). The network include 17 monitoring sites, and 5 of them located in the Beijing area, which named the 325 m mast of IAP (monitoring site in this study, suburban site), Olympic Village (between the north third ring road and north fourth ring road, suburban site), Shuangqing Road (near the north fifth ring road, suburban site), Longtan Lake (near the south second ring road, downtown site), and Yangfang Town (close to the north sixth ring road, suburban site). Monthly average $PM_{2.5}$ mass concentrations of these five sites in August 2008 were $62.2 \pm 52.5\,\mu g/m^3$, $60.6 \pm 49.1\,\mu g/m^3$, $57.7 \pm 44.5\,\mu g/m^3$, $60.8 \pm 47.6\,\mu g/m^3$ and $50.8 \pm 48.1\,\mu g/m^3$, respectively. Only minor differences were found between the IAP site and the downtown site (Longtan Lake site). Furthermore, hourly variation of $PM_{2.5}$ between IAP site and Longtan Lake site was highly correlation (r=0.91, p<0.001). Hence, the IAP site could be assumed as representative for the Beijing urban environment. We agree that local sources of the specific air pollutant like restaurants should be considered when selected environmental monitoring site, however, considering the restaurant industry in Beijing developed rapidly and the number of large-scale catering enterprises with main business income over 2 million yuan each year in Beijing exceeded 2,000 in the end of 2011 (source: Beijing Statistical Information Net), the number of small and medium sized catering enterprises was estimated to exceed 70,000. 80% of these restaurants located within the fifth ring (source: Beijing Statistical Information Net), it is very hard to select a monitoring site that far away from restaurants, especially in the urban area of Beijing. So the distance between our monitoring site and the restaurants may be too close, but it actually reflects the typical urban area atmospheric environment in Beijing.

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.

[*Response*]: The sheath flow of SMPS and APS were 3.0 L min^{-1} and 4.0 L min^{-1}, respectively. Operational parameters (flows and high voltage values) of the SMPS and APS were checked and calibrated before and after the one month intensive

observation period. Missing data were recorded for two hours during the one month observation period due to power failure. Missing number concentration values were replaced by interpolated values of the determined values within the same size bin. The related discussion has been added into the text. See page 5, line 8-11, and page 7, line 3-5.

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).

[*Response*]: We thank the reviewer's comments and suggests. We added a section in the supplementary materials to briefly describe the SMPS and APS data merging procedure. The data merging procedure was scripted in the statistical language of CRAN R and applied to data frames containing the hourly averages for the SMPS and APS size bins. Missing data were replaced with interpolated values (two hours data in this study). Concurrent hourly average spectrum-pairs were then merged and entered into a data frame with a common range of particle size bins (d^t_B = 0.014 to 2.5 μm) together with an estimated value for the transitional-regime effective density. The value of transition-regime effective densities ranged from 0.75 to 2.15 g/cm^3 which corresponded to merged spectra with overlapping regions covering up to 9 size bins. The related discussion has been added into the supplementary materials.

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

[*Response*]: We thank the reviewer's comments. The full name of abbreviations HR-ToF-AMS is high-resolution time-of-flight aerosol mass spectrometer, and the full name of abbreviations IAP is Institute of Atmospheric Physics, The related information was added into the corresponding position in the revised version. See page 4, line 19, and page 5, line 24-25.

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."

[*Response*]: We thank the reviewer's comments and suggests. It should be mentioned that the uncertainties calculated from equation 1 were for the measured values like number concentrations as well as chemical composition and gaseous pollutants data. Uncertainties for the missing data, not mentioned in the ACPD version, however, were assumed as 3 times of the mean as suggested in previous study (Gu, et al., 2011). The related information was added into the revised version, see page 7, line 15-16. As the reviewer suggested, we provided a more detailed description for choosing the right number of factor in the revised version. Briefly, The PMF was run several times for the Fpeak values between -3 to 3 in steps of 0.2. The Q-value versus Fpeak plot shows a typical "U" shape with the lowest Q-value corresponding to Fpeak of -0.4, which is selected in this study. The theoretical Q (Q_{exp}) is estimated by using the equation: nm-p(n+m), where n is the bins number of the profile of the particle number size distribution or chemical composition and gaseous pollutants data, m is the number of samples in the data set, and p is the number of factors fitted by the model. If the assumption of the source profile and the estimation of the errors in the input data are accurate, solutions with numbers of factors that give Q/Q_{exp} near 1 should be obtained (Ulbrich et al., 2009). It was observed that an 8 factor solution provided the most meaningful results, with the value of Q/Q_{exp} close to 1 and a correlation coefficient (R^2) of 0.98 between the modeled and experimental concentrations. In total, the modeled particle number concentration could explain 99% of the observation data. The related discussion has been added into the text, See page 7, line 21-30, and page 8, line 1-4.

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.atmoschem-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.

[*Response*]: We thank the reviewer's comments and suggests. We agree that the number of data points assigned to a single wind sector might be too small and insufficient to be statistically significant. We recalculated the CPF results and added more statements in the revised version. See page 8, line 13-20.

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 40nm 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.

[*Response*]: We thank the reviewer's comments. We agree that there should be difference of particles size distribution from cooking sources between indoor and outdoor environments. We added a reference in the revised version to discuss this kind of difference (Diapouli, et al., 2011). Still, the distance between outdoor and indoor in that study was only three meters and not long enough to explore the size distribution evolution of cooking particles during nucleation and accumulation processes. Since this kind of study is still rare, we weakened the comparison mentioned by reviewer 2 and discussed possible deviation of this comparison in the revised version. Beside that kind of comparison, as reviewer 1 suggested, we checked specific markers in the HR-AMS data for cooking emissions. More statements were added in the revised version to discuss the relationship between factor 1 and these cooking emissions related markers(e.g., $C_5H_8O^+$, $C_6H_{10}O^+$, and $C_7H_{12}O^+$). See page 9, line 1-10, and line 17-21. We agree that the correlation of factor 2 and NO_X cannot be seen as high positive and we deleted this statement. Instead, we provided more statement to discuss the relationship between factor 2 and NO_X. See page 10, line 2-15.

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.

[*Response*]: Suggestion taken. We re-wrote this sentence, see page 10, line 15-17. We agree that factor 2 may not be specifically associated with diesel particles since the diurnal pattern of Factor 2 do not display a marked daily trend as the heavy diesel trucks. Considering its size distribution mode and chemical composition characteristics, we think factor 2 is most likely associated with aged vehicular exhaust, which are formed by condensation and coagulation from primary vehicle exhaust during the dilution process to the receptor site (Canagaratna, et al., 2010).The related discussion has been added into the text. See page 10, line 24-30.

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.

[*Response*]: We thank the reviewer's comments. The relevance of factor 4 and "regional sources" was not primarily derived from the CPF plots, but from the particle size distribution and particle chemical composition. Factor 4 had classic accumulation mode and explained most of chemical composition that existed in droplet mode, all of this provided a clear relationship between factor 4 and "regional sources". We agree that CPF analysis seemed confused in the current analysis. As the reviewer suggested, we reevaluated the CPF result and deleted those high CPF values with low values of total number of wind occurrences. High CPF values associated with factor 4 were reduced to southeast sector by the modified CPF analysis, and moderate CPF values from northeast sector were also found in the modified CPF analysis, suggesting a background regional source. Based on the modified CPF analysis, the contribution from northeast sector was negligible to the factor 7. The related discussion has been added into the text. See page 12, line 18-20, and page 14, line 17-24.

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.

[*Response*]: We thank the reviewer's comments above. We agree that the NOx-VOCs photochemical reaction should associated with factor 6 and secondary organic aerosol (SOA) should be an important contributor to factor 6 as we discussed in section 4.3. As the reviewer suggested, if the NOx-VOCs photochemical reaction could be the dominant factor, then the diurnal variation of ozone, the product of NOx-VOCs photochemical reaction, should be in strict accordance with factor 6. However, as Fig, S3 showed, the maximum concentration of ozone appeared at about 2 pm, two hours earlier than the maximum value of factor 6 which peaked at 4 pm. Consequently, other gas phase photochemical oxidation should also contribute to the increase of factor 6 after 2 pm. The diurnal variation of sulfate showed a peak value at about 3 pm, its gaseous precursor SO_2 showed a peak value at 12 am, and declined until 7 pm, suggesting the conversion of SO_2 to sulfate via in-cloud or aerosol droplet process and gas condensation process at afternoon. In a previous study conducted in summertime of Beijing, Guo et al.(2010) found that the majority of measured sulphate existed in droplet mode that attributed to in-cloud or aerosol droplet process, and his result showed the gas-to-particle conversion was also an important formation pathway for sulfate in summer of Beijing. In this study, factor 6 contains a large amount of organic matter and sulphate, most of the appointment organic existed in Aikten mode (24.2%), and most of appointment sulphate existed in Aikten mode (28.8%) and droplet mode (28.2%). Considering both organic matter and sulphate contributed to factor 6, we

renamed factor 6 as a mix of SOA and secondary sulfate from gas phase photochemical oxidation. The related discussion has been added into the text. See page 13, line 23-29.

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.

[*Response*]: We thank the reviewer's comments above. We agree that this assumption would bring a lot of uncertainty when compared volume contribution in this study with mass contribution in other studies. To address this problem, we proposed a new method to estimate mass concentration of different sources from their volume concentration in this study. The theoretical foundation of the new method is that particles emitted or formed from specific sources should be associated with one or several of the known chemical composition; for example, particles emitted from cooking are mainly organic matter. Then particle density of the known chemical composition was assigned to each source and their mass concentrations were estimated. It should be mentioned that, although the new method is much better than the directly equivalent of volume concentration to mass concentration, yet uncertainty of the new method cannot be ignored. We agree that the comparison of annual average to one month observation in this study was improper, so we changed the previous references and added two new references in which source apportionment results were conducted from relatively short period (one month or one season). We kept the work of Pey et al. (2009) in Table 3 though the study period was much longer than our study. Since few study discussed particle number contribution from different sources, we think the comparison of this study to that of Pey et al. (2009) would help us to know the source contribution to particle number concentration in different mega cities. The related discussion has been added into the text. See page 15, line 29-30, and page 16, line 1-17.

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: : :).

[*Response*]: We thank the reviewer's comments above. We modified that sentence in the revised version. See page 16, line 25.

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.

[*Response*]: We thank the reviewer's comments and suggests. We agree that no study made a specific discussion on the separation of sulfate from local or remote origin, however, similar conclusion can be found in a few studies. For example, using size distributions associated with each source, Ogulei, et al. (2006) attributed the secondary sulfate source in Baltimore to local origin for this source associated mainly with particles smaller than 0.2 μm. He provided a new perspective that local and remote sources were able to distinguish by using size distributions associated with each source. Using PSD data, Pey et al. (2009) also showed two kinds of secondary sulfate sources in Barcelona. One associated to photochemical nucleation particles indicating local origin and the other related with the regional and/or urban pollution. Since few study focused on the separation of sulfate from local or remote origin based on PSD data, we added more statement to discuss the uncertainty of this kind of separation in section 4.2 of revised version, which would benefit for the future source appointment work to give more specific separation on secondary sulfate sources from local or remote origin. See page 17, line 28-30, and page 18, line 1-7.

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.

[*Response*]: As we discussed in section 4.2, local sources were generally characterised by unimodal or bimodal number distributions consisting mostly of particles less 0.1 μm in diameter. So all the appointment factors except factor 4 in this study can be seen as local origin, and then particles associated with factor 4 can be seen as regional contribution that transported from the surrounding areas of Beijing. As it shown in Table 2, percent of nitrate and sulphate attributed to factor 4 was 42.1% and 37.3% for droplet mode, and 8.1% and 17.9% for condensation mode. On average, 36.2% of measured nitrate and 31.9% of measured sulphate in this study was attributed to factor 4. In other word, more than one-third of the secondary nitrate and nearly one-third of secondary sulfate in Beijing was transported from the surrounding areas of Beijing. The related discussion has been added into the text. See page 18, line 11-16.

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.

[*Response*]: We thank the reviewer's comments above. We re-wrote this sentence and added more statement about the quantification contribution of fugitive dust (road dust in ACPD version) and power plant. See page 19, line 22-30, and page 20, line 1-6.

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

[*Response*]: We thank the reviewer's comments above. We modified this section in revised version.

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