Anonymous Referee #1

General comments:

This paper describes the results on sources affecting fine and ultrafine particles in Beijing, over a one month summer campaign carried out in 2011. The authors of the manuscript did the source apportionment analysis by combining HR-ToF-AMS and SMPS data, by using PMF. This is not the first time this kind of work is presented, but at the same time this is not frequent. The authors have shown in the manuscript a fairly good review of previous similar studies carried out worldwide. The authors have found that 8 sources are explaining most of the variability of the particle and volume concentration between 15 nm and 2.5 μm. Most of the combustion sources are evident in the ultrafine modes, whereas those arising from mechanical processes such as road dust, or the regional component are found in accumulation and droplet modes. Although the manuscript is well organized, the figures are clear, and the results are in general satisfactory, there are some points that need a review.
[*Response*]: We thank the reviewer's comments.

Specific comments:

1) Pag 1368, line 7: solid mode exhaust. I particularly don't like this name. It's quite ambiguous. Maybe you may label this factor as "primary vehicular exhaust emissions", or some similar term.
[*Response*]: We agree with the reviewer's comments above. To make it clear and consistent with the analysis in section 3, we relabeled factor 2 as "aged vehicular exhaust emissions". See the changes throughout the paper.

2) Pag 1368, line 17: Add "," after "Overall"
[*Response*]: Suggestion taken. See page 2, line 17.

3) Pag 1368, line 19: Find an alternative word to "population".
[*Response*]: Suggestion taken. We found an alternative word "size distribution" to replace "population". See page 2, line 19.

4) Pag 1369, line 1: Please replace as follows: "Since particles in ambient air: : :.to particle conversion, a better understanding of the source attribution of particles is needed for investigating the associations between specific particle sources and health, but also for policy makers to introduce effective abatement strategies.
[*Response*]: Suggestion taken. See page 2, line 30, and page 3, line 1-5.

5) Pag 1369, line 25: Please remove from ": : :would be expected: : :" to : : :measured spectra", since this is an assumption not necessarily true and not important here.
[*Response*]: Suggestion taken. See page 3, line 24-26.

6) Pag 1370, line 8: Please replace as Oguleia by Ogulei; add Pey et al., 2009
[*Response*]: Suggestion taken. See page 4, line 6.

7) Pag 1371, line 2: Please add some information about the inlet system (diameter, flow inside, material, length). Was some correction applied? It is strange because it seems that a unique inlet was used, but individual cutoff inlets were used for SMPS-APS ad AMS systems. Even brief this information should be provided.

[*Response*]: We agree with the reviewer's comments above. Information about the inlet system was added as follow: ambient air was sampled into the SMPS and APS from a 0.5 inch (outer diameter, 2.0-m) stainless steel tube with a low flow PM_{10} inlet. The total flow through the stainless steel tube was 5 L min^{-1}, out of which about 0.3 L min^{-1} and 1.0 L min^{-1} were sampled by the SMPS and APS from two 0.25 inch (outer diameter, 0.5-m) stainless steel tubes, respectively. See page 5, line 2-6. Briefly, a unique inlet was used by SMPS-APS; another inlet was used by AMS. To make it clear, we re-wrote the last sentence in section 2.1 and added more information about the inlet system in section 2.2. See page 4, line 24-25. We calculated size-dependent diffusional and gravitational losses for the inlet line using the empirical functions given by Willeke and Baron (1993). The diffusion loss was estimated to be ~20% for the smallest measureable particles of 14.5nm, diffusion loss could be negligible for particles between 100nm to 1000nm, and estimated to be ~3% for particles of 2.5 μm. The corresponding correction was made for SMPS and APS data. The related discussion has been added into the revised version. See page 5, line 14-18.

8) Page 1371, line 16. Given that this point is very important in the context of this study, how was the fitting between APS and SMPS data? Did you any correction applied to fit both dataset? Maybe you should include something in the supplementary materials.

[*Response*]: We agree with the reviewer's comments above. We added a section in the supplementary materials to briefly describe the data merging procedure. Two kinds of correction were applied to fit both APS and SMPS data before the merging procedure. One was for the diffusion losses for the inlet line which was discussed above; the other one was for the drop efficiency for the APS measuring particles with aerodynamic diameters below 0.7 μm. This problem is addressed by merging the data twice in order to derive and then apply a correction curve for the APS. During the first cycle, the spectra are selected which have a sufficient overlap (>3 size bins) to exclude the lower size bins and merge the spectra onto a smooth continuous curve measured against the stiff cubic spline. Once fitted, the counts of the excluded size bins are compared to counterpart values of the spline fitted through the SMPS data and a row of correction values are calculated for the APS spectrum. The related discussion has been added into the supplementary materials. The related discussion has been added into the supplementary materials.

9) Page 1372-1373: PMF section. You talk about missing values replacement but you

don't mention the amount of missing values in the database. Please provide this information.

[*Response*]: Suggestion taken. 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 or estimated by linear interpolation of the measured concentration value for the chemical composition and gaseous pollutants data. The related discussion was added in the revised version. See page 7, line 3-5.

10) Page 1374, line 10. Even that cooking emissions may be relevant, are you completely sure about the attribution of this factor to cooking? The 40 nm mode is also very typical of urban emissions from traffic. As far as I can see in the manuscript, this source is that experiencing the largest daily variations, with two peaks (as you state at midday and in the evening). This source is also linked to NO2. I am afraid that this is the largest source to particle number in Beijing. During the Olympic Games in 2008 the authorities prohibited emissions from many sources but I am pretty sure that cooking was not prohibited. And air quality was significantly improved: : : I suggest checking specific markers in the HR-AMS data for cooking emissions, and show them in the manuscript given the magnitude of your findings.

[*Response*]: We thank the reviewer's comments and suggests. We agree that the current analysis was not enough to show factor 1 associated with cooking emissions. As suggested, we checked specific markers in the HR-AMS data for cooking emissions. In previous AMS study, cooking organic aerosol (COA) shows most significant correlation with a few $C_xH_yO_1^+$ ions (e.g., $C_5H_8O^+$, $C_6H_{10}O^+$, and $C_7H_{12}O^+$), which were suggested to be used as spectral markers for COA (Sun et al., 2011). We added more statements in the revised version to discuss the relationship between factor 1 and these cooking emissions related markers. See page 9, line 17-21. Figure 1, which was re-designed in revised version, shows the diurnal variation of $C_5H_8O^+$, $C_6H_{10}O^+$, and $C_7H_{12}O^+$, to provide direct evidence to support factor 1 be associated with cooking emissions. Furthermore, cooking organic aerosol was also identified by positive matrix factorization (PMF) using the high resolution mass spectra of HR-AMS data collected at the same period. That result will present in another paper, in which COA on average account for 20.4% of OA mass concentrations, and on average account for 3.3% of $PM_{2.5}$ mass concentration, close to the value of factor 1 contributed to particle volume (3.8%) in this study.

11) Page 1375, line 8-15. You state that this factor has a positive correlation with NOx to support your interpretation as a traffic factor. But association with NOx is even better for the previous one (cooking factor). You also mention the relation with truck traffic in the ring-road, only allowed from 00:00-06:00. I think this factor is related with traffic, but not specifically with truck traffic since it should display a marked daily trend and it's not. In my opinion the trend could be related with dilution processes rather than traffic patterns.

[*Response*]: We thank the reviewer's comments and suggests. We agree that the correlation between factor 2 and NO_X was not so strong like the other traffic related factor (factor 3), so we deleted this statement in the revised version. However, it's important to note that the emission source of factor 2 was mainly existed during night-time, in which time NO_X could be transformed to nitrate via the heterogeneous hydrolysis of N_2O_5 during summertime of Beijing (Pathak, et al., 2011). Considering the low wind speed during night-time (<0.5m/s), average arrival time of vehicular exhaust to the receptor site during night-time was estimated to be 10 minutes, would sufficient for the heterogeneous hydrolysis of NO_2 at the air-water interface under the condition of relative humidity closed to 80% (Finlayson-Pitts, et al., 2003). As a result, in the dilution process of vehicular exhaust to the receptor site, gaseous pollutant like NO_X gradually transformed into nitrate and particulate pollutant like organic matter remained unchanged. Consequently, it would be reasonable that factor 2 was chemically consistent with vehicular particle but inconsistent with gaseous character of vehicular exhaust during summer nighttime of Beijing. We agree that factor 2 may not be specifically associated with truck traffic. Considering its size distribution mode and the relevance of organic matter and sulfate, we renamed factor 2 as aged vehicular exhaust emissions. Correspondingly, we renamed factor 3 as fresh vehicular exhaust emissions. The related discussion has been added into the text. See page 10, line 2-15, and line 24-29.

12) Page 1377, line 8. Why this factor 4 (regional pollution) shows minimum concentrations in the afternoon?

[*Response*]: We think there are two reasons causing the minimum concentrations of factor 4 in the afternoon. One was for the greater wind speed in the afternoon during study period (Fig. S4), which benefiting for the spread of air pollutants and lower down the concentration of air pollutants, the same for the regional pollution. The other one was rainy events. There were five rainy days during the study period; three of them began in the morning and last to the noon or the afternoon. It is expected that concentrations of air pollutants would be significantly reduced by wet deposition, partly reducing the contribution of regional pollution in the afternoon. It should be mentioned that the diurnal pattern of factor 4 could be seen as almost flat and lack of a clear diurnal variation on the whole, consistent with diurnal pattern of regional sources suggested by Harrison, et al (2011). The related discussion has been added into the text. See page 12, line 11-18.

13) Page 1377, line 18. I totally agree with the explanation given in the text. In fact, the daily trend of factor 5 and factor 6 is opposed, and may be explained as follows: the volatilization of ammonium nitrate particles at noon release ammonia to the atmosphere, captured by HSO4 and SO42- to form particles.

[*Response*]: Suggestion taken. We added this discussion in the revised version. See page 13, line 15-18.

14) Page 1379, lines 5-19. I agree that this factor is related with road traffic but it's too

hard to say that is road dust, a coarse component built-up of mineral matter and metals essentially. Given that you didn't measure metals, neither mineral dust, and your size distribution finish in 2.5 um, I would change the name. In addition, warm areas usually show an important coarse nitrate component, not related with road dust but with Ca and Na (from soil dusts or sea salt). Such coarse nitrate particles, as yours, are indicative of ageing of air masses at local and regional scales.

[*Response*]: We agree with the reviewer's comments. The current analysis is not enough to show factor 8 was specifically associated with road dust though the relation with road traffic was possibility. As the reviewer mentioned, warm areas usually show an important coarse nitrate component. Guo et al., (2010) found that nitrate showed bimodal size distribution and the coarse mode nitrate on average contributed 42% to the measured nitrate during summertime observation in Beijing. When the relative humidity was lower than 50%, NH_4NO_3 usually dissociated and formed coarse mode nitrate by reactions of nitric acid with $CaCO_3$, K_2CO_3 or NaCl, the latter of which originated from soil dust or sea salt. In this study, we also found the relationship between factor 8 and nitrate. So it is likely that factor 8 was the mixture of soil dust and road dust, which have similar profiles. Based on the above analysis, we renamed factor 8 as fugitive dust. The related discussion has been added into the text. See page 15, line 7-15.

15) Page 1380, line 13. "Coal consumption" or "coal combustion"?
[*Response*]: It should be "coal combustion", we changed as "coal combustion" throughout the paper.

16) Page 1380, line 27. Change "diversification" by diversity/variety/mixture.
[*Response*]: Suggestion taken. We changed "diversification" as "diversity", See page 17, line 16.

17) Page 1381, line 5. You have to convince me about this very high cooking relevance.
[*Response*]: We added more statement in the revised version to further discuss the relevance of factor 1 with cooking emissions. See page 9, line 17-21.

18) Page 1381, line 21. Remove from ": : :, including: : :" to ": : :VOCs,"
[*Response*]: Suggestion taken. See page 18, line 21.

Reference:
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secondary formation, Atmos. Chem. Phys., 10, 947-959, 2010.

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