Anonymous Referee #2

We thank the reviewer for helpful comments and suggestions. Below we provide point to point response to each comment. The page numbers refer to the version published in ACPD.

1. Comment

Abstract, page 5167, lines 14-16. The authors assert here that air mass trajectories did not play a key role in the pollution reduction during the Games. Has someone claimed they did? The vast preponderance of previous studies (including those cited by the authors) attribute the reduction to imposed restrictions on industrial and traffic emissions. Given the assertion appears in the abstract, one would expect this to be an important issue, but there is no real debate on this point. Possibly the authors are thinking of the analysis by Gao et al (2011) since they cite this study in the text but they point out no contradiction between their results and those of Gao et al, and in fact I see nothing contradictory. Gao et al merely point out that trajectories played a role, not that they were "key" .This assertion should not be in the abstract without much more detailed discussion of the issue – and indeed the establishment that there IS an issue is in the text.

Response

Thanks for pointing this out. Now we admitted that this conclusion is confusion. In the revised manuscript, firstly we have admitted that both the previous and this studies showed that the air mass history do play a role on air pollution. The slow southern wind always indicating the polluted situation for Beijing case. However, the frequency of air mass from polluted southern direction (southwest and south) was 43% in 2008, which is 1.3 times higher than that (31%) in 2004-2007 (See Figure 3b). Although, the air mass condition in 2008 was not favorable to reduce of particle concentration, still, the lowest particle volume concentration had been observed (Figure 2b). This phenomenon indicated that the good air quality during the Olympic Games 2008 could not be only ascribed to the air mass condition.

2. Comment

P 5168, line24 through P5169, line23. The authors summarize previous work here and state their objectives, namely, to evaluate air quality during the Games using particle number distributions (PSD), and to assess which control strategies contributed most to reductions in particle number and volume concentrations. They also note that they will use "long-term" measurements dating to 2004. There are several issues which arise here. Firstly, previous studies (e.g., Wang et al, 2010; Wang and Xie, 2009; Zhou et al, 2010; all as cited in the MS) have already demonstrated that the reduction in pollution, including that in total particle volume (PM10), is attributable primarily to imposed reductions in construction and traffic. Why would one assume that the case would be different for particle number? Especially since numerous previous studies have presented data to show that a major source of particle number in urban airsheds – including Beijing - is traffic (e.g., Wu et al, 2008; Zhou et al, 2005; Kim et al, 2004; all as cited in MS). Of course, it is certainly possible that particle number and volume would be impacted differently by different sources but I see no reason to think this would be the case here and in fact the authors own analysis suggests that it is not. Secondly, there should be some exposition of why particle number as opposed to particle volume is important. Again, a case could be made, but it is NOT made, and leaves the reader at something of a loss as to why the study is being done. Finally, the authors should make a better case for the value of their "long-term" measurements. Do they feel, for example, that the comparison of Wang et al (2010, as cited in MS), has been biased by anomalous conditions in their much shorter term comparison? An issue should be clearly delineated here.

Response

In the introduction, we summarized several previous studies which have evaluated the air quality during the Olympic Games. However, these studies only concern the pollutant gases or particle mass concentration. The control measures focus on the traffic emission were the most important ones taken in August 2008. Previous studies

showed that the contributions of vehicle source to PM_{10} showed vary in a wind range, accounting for more than 30% in the megacities such as Milan, Madrid and Barcelona and less than 5% at some rural sites (Viana et al., 2008). In urban site of Beijing, the contribution of traffic source to mass concentration were only 6% (Song et al., 2006). However, the source related to traffic emission has a significant contribution to total particle number concentration, accounted for about 40% in northern Sweden (Krecl et al., 2008) and Pittsburgh (Zhou et al., 2004), even up to 78% in Erfurt (Yue et al., 2008). Hence, in this study, we want to focus on the particle number to evaluate the traffic control measures during Olympic Games, which has not been done before. The particle number can help us get a better understanding on the impacts of traffic control.

In this study, we respect the other shorter term comparison. However, we believed the "long-term" measurements might provide another perspective to assess the traffic control measures. In addition the shorter term comparison might be influenced by the different meteorological condition in each month. The comparisons between five years' August could avoid this impact as far as possible.

We have delineated above clearly in the revised version.

3. Comment

P5170, lines 12-15. The authors cite use of two CN counters with their DMA, a TSI 3010 and a 3025. Only the 3025 will measure down to the 3 nm cited by the authors as their lower size limit. Clarify.

Response

Thanks for pointing this out. We have clarified it in the revised MS: "The system is composed of two parallel Hauke-type Differential Mobility Analyzers (DMAs) that classify particles in the size ranges 3–80 nm and larger than 40 nm, counting using Condensation Particle Counters (CPCs, TSI Inc., St. Paul, MN, USA) models 3025 and 3010, respectively."

4. Comment

P5170, line 24 to P5171, line 5. The authors introduce the ToF-AMS here but do not discuss the species actually measured (other than the total mass – PM1.0). The only use of this data seems to be in Figure 7 (SNA+OOA). Since Figures 5 and 7 constitute the only "tracers" used to help identify sources, more should be done here. Which species were measured, with what certainty and with what time resolution? Additionally, these data were available only for the summer-early fall of 2008, in so far as I can see. Any source attribution associated with this data would be in principle only valid for this interval – scarcely long term. Some clarification would be useful here.

Response

We have added several sentences to describe the measurements of Tof-AMS: "The PM_1 particle mass concentration and chemical components (sulfate, nitrate, ammonium, chloride and organic matter) measurements were carried out using an Aerodyne High-Resolution Time-of-Flight Aerosol Mass Spectrometer (HR-ToFAMS) (Huang et al., 2010). The detection limits of sulfate, nitrate, ammonium, chloride, and organics were calculated to be 0.008, 0.004, 0.026, 0.004, and 0.033 $\mu g/m^3$, respectively. Meanwhile, four organic components including a hydrocarbon-like (HOA), a cookingrelated (COA), and two oxygenated (OOA-1 and OOA-2) organic aerosol were identified. All the data were averaged to 10 min in order to keep consistent with the particle number size distribution."

In addition, we have admitted that the AMS results was only available in August 2008 in this study. However, we only use the diurnal variation of (SNA+OOA) in the following analysis. Compared with the results in the wintertime (Sun et al., 2013) and other years (MS in the preparation). The diurnal variation of (SNA+OOA) did not show the significant differences.

5. Comment

P5172, Eq 1 and associated discussion As the authors note, the data matrix (Xij) in a PMF analysis is normally associated with different chemical species (the j index). Here the authors state it is associated with the PASD's. I think they need to be much

more specific here. Each of the j values must be the number concentration for a given size interval. What are these intervals? I do not see that even the number of such intervals is given. Furthermore, if the intervals are numerous (high resolution), then the detection limits will be rather low, particularly for the larger sizes. In this regard, the detection limits for the size channels (?) given in Figure 1 are not very informative once one gets above _ 20 nm. The authors state that the uncertainty for the size measurements was 15% for sizes smaller than 25 nm and 10% otherwise. This seems very unlikely given Poisson counting uncertainty and the rapid fall off in number concentration with size. Furthermore, no absolute uncertainties, i.e., the actual MDL's, seem to be given. These uncertainties, both absolute and mean normalized (MDL and error fraction), are a very important component of the PMF analysis. More information is necessary here.

Response

Sorry for the misunderstanding. In this study, we used the matrix of observed particle number size distributions as the input of PMF model. The *i* and *j* in Eq. (1) represent the number of samples (16170) and size bins (30).We have stated this in the revised manuscript:"Here a data matrix X_{ij} is observed particle number size distributions composed of *i* by *j* dimensions, in which *i* number of samples and *j* the size bins were measured. G_{ik} is the number concentration of particles from the k^{th} source associated with the i^{th} sample and F_{kj} is the size distribution profile associated with k^{th} source. E_{ij} is the residual for each sample."

The detection limit of the TDMPS was calculated by particle number concentration divided by raw counts of CPC for each size bins. In the revised Figure 1 (We have changed Y-axis to log scale), we could see the detection limit for each diameter. The larger detection limit were shown in small particles, which might be ascribed to the huge diffusion losses in the sampling tube. Based on the calculation of MDL, we followed the Eqs. (2-3) recommended by EPA to obtain the uncertainty.

6. Comment

P5172, lines 10-11 and 15-17. The exclusion of intense nucleation events is quite

reasonable but a bit of clarification is necessary. First, are the authors saying that they excluded 10% of their data due to such events? This seems to be the case but it should be clearly stated. Furthermore, the criteria used by Wu et al, (particle concentrations exceeding 104 cm-3 in the 3-10 nm range for 2.5 hr.) should be stated in the text. It would clarify that not all nucleation is being excluded by any means. More importantly, the implicit claim that the PSD's are stable and representative of the sources after one excludes such intense nucleation events is at least questionable and should be discussed. Not only do secondary aerosol appear in the "nucleation range" – and the authors have by no means excluded the impact of nucleation entirely from their PSD's but also in the accumulation mode via condensation of secondary precursor gas phase species (and through aqueous-phase production processes that inject modified aerosol into the air after cloud drop evaporation). This will be most apparent in measurements taken close to sources. I think that some discussion along the lines of that presented in Kim et al (2004, as cited in the MS) would be useful here, coupled with the sampling location for the authors' data.

Response

Thanks for pointing this out. We have added some sentences to clarify the criterion identify the nucleation event. Here, we excluded the nucleation event days because the particle number size distribution profiles during the nucleation event period are not constant, which is not fit to the basic assumption of PMF. In the PMF analysis, the factor with the peaks at 30 nm and 200 nm had been identified, which is coincide with the previous study (Kim et al., 2004). The high correlation between the daily pattern of factor 4 and secondary aerosol (SNA+OOA) was observed, indicating it could represent as secondary aerosols.

7. Comment

P5175, Back Trajectory analysis The cluster analysis is interesting but is poorly presented. For example, the authors highlight the large percentage increase in southern trajectories, suggesting that, because previous work has shown that it is the southern trajectories that are most polluted, changes in flow conditions could play no

role in the reduction in particle concentrations in 2008. The authors cite the previous study by Wehner et al (2008, as cited in MS) but the cluster wind sectors are not the same as those found in this study. For example, the E cluster of the authors corresponds most closely with the "local" cluster of Wehner et al. while the S cluster of Wehner et al encompasses both the SW and S clusters of the authors. Given this, one sees a somewhat different picture. For number concentration – which, after all, is the main topic of this analysis – the Wehner et al concentrations do not vary much by wind sector but, to the extent they do, slightly higher concentrations are from the North and East as compared to the South. (This is somewhat consistent with the authors' data as shown in Figure 4, which shows slightly higher particle concentrations coming from the NW and E than from the S.) Hence, the Wehner et al analysis suggests pollution is coming from the SW and E sectors of the authors, not just the S. Coupled with the fact that there are percentage decreases in the number of trajectories from the SW, NW and E in the authors' data (Figure 3), it is not at all clear that changes in air flow do not play some role in the reduction in number concentration. I think that the authors should simply emphasize, as per Figure 4, that the number concentrations are lower from all sectors in 2008 and that consequently, changes in air flow cannot be the entire answer to the pollution decrease.

Response

Thanks for your helpful suggestions. It is true that the back trajectory cluster in this study is different from the previous one (Wehner et al., 2008). Hence, we only considered the frequency of air mass from south is not fair. Hence, we have modified the manuscript like this:"On the contrary, more volume concentrations were observed when the air masses from southwest and south, with the mean values of 80 $\mu m^3/cm^3$ and 70 $\mu m^3/cm^3$, respectively, which are 2 times higher than those from other directions. This results consistent with the previous study that the slow southern wind controlled by high pressure systems always indicating the polluted situation for Beijing case (Wehner et al., 2008; Shen et al., 2011). The huge discrepancy of particle volume concentrations in five trajectories suggested that the air mass history has a significant impact on air quality. However, the frequency of air masses from the

polluted southern direction (southwest and south) was 43% in 2008, which is 1.3 times higher than that (31%) in 2004-2007 (See Figure 3b). Although, the air mass condition in 2008 was not favorable to reduce of particle concentration, still, the lowest particle volume concentration had been observed (Figure 2b). This phenomenon indicated that the good air quality during the Olympic Games 2008 could not be ascribed to the air mass condition."

Meanwhile, we have recognized that the conclusion: "air mass origins were not the key factor in the observed particle concentration reductions" is unfair. The air mass history do play a role on air pollution. The slow southern wind always indicating the polluted situation for Beijing case. However, in our analysis, we have realized that the air mass condition in 2008 was not favorable to reduce of particle concentration, still, the lowest particle volume concentration had been observed. Hence, we have modified the conclusion as: "This phenomenon indicated that the good air quality during the Olympic Games 2008 could not be ascribed to the air mass condition.", as the referrer suggests in comment 11.

8. Comment

P5177 and 5178, Section 3.3 The source apportionment by PMF is presented here in a somewhat unsatisfactory manner. The first step in such a presentation is typically to see how well the model reproduces variables in the Xij data matrix. What the authors present is how well total number concentrations are reproduced. This is not a very challenging test by itself. Furthermore, the text is unclear here. What do the authors mean by stating that the model-observational difference is 82% at 7 nm and 99% at 120 nm? As stated, it would indicate that there is a model-observational difference of a factor of _ 2 – very bad indeed. I suspect this is a linguistic problem since Figure 5a suggests much better agreement but the language must be cleared-up. More tellingly, the model-observational comparisons should be made for each size range (or at least a good selection of them covering the instrument measurement range) and in the form of linear regressions with R2, slope and intercept values all presented (again, this could be done in a Table in the SI and summarized in the text). As it stands, the model

bias is unclear and the prognostic value of the factor PSD's presented in Figure 5a is similarly unclear.

Response

Sorry for the misunderstanding. In the Figure 5a, we presented how well the mean modelled particle number size distribution to explain the observation. We have clarified this in the revised version: "The mean modelled profiles of particle number size distribution for each factor are presented in Figure 5a. The four factors are arranged in order of increasing peak diameter from factor 1 to factor 4. The modelled particle number size distribution (dashed line) is the sum of four factors. The correlation coefficient R^2 between observed and modelled particle number concentration in each size interval varied in a wide range, only about 0.5 in small diameter (< 10 nm) and up to 0.95 in larger size (> 15 mn). The weak correlation in small particles might due to the high uncertainty during the measurement. In total, the mean modelled total particle number concentration could explain 96% of the observation data, varying from 82% to 99% at 7 nm and 120 nm, respectively."

9. Comment

P5177, line 19 to P5178, line 29. The authors here try to interpret the factors in terms of sources. Of course, this is always a difficult area to deal with in PMF analysis and one must make allowances. Still, this is one of the weaker parts of the presentation. The presentation is very qualitative here and does not evoke much confidence. The attribution is based on three generic traits, the shape of the PSD, the diurnal variation in the factor strength, and the association with a few "markers, for different sources. None of these provide unique characteristics, i.e., they could be attributable to multiple sources. Ideally in such instances (indeed, ideally for ANY PMF analysis), all of the data would be incorporated into a single input matrix and the vector decomposition itself would yield a factor profile that could be easily (relatively) interpreted in terms of source characteristics. Since the input matrix must have homogeneous units, the present data set would require normalization, with number concentrations scaled by the total number concentrations, various particulate species

concentrations scaled by the total particulate mass (for example), gas phase species scaled by, say, the total gas phase speciated mass, etc. This would be a challenge but would have yielded a very nice, and novel, analysis. But this has not been done. The factor PSD shape, diurnal factor variance and a few "markers' have been compared external to the PMF analysis itself and qualitatively. To the extent possible, this must be rendered quantitative. Figures 6 and 7, which show the diurnal covariance of selected variables are useful but constitute an insufficient basis for source attribution. Additionally, a covariance matrix is necessary, in which all of the PMF factors (essentially the PSD deconvolution), the chemical concentrations of all measured species, and any other variance information the authors wish to use, are included (for example, a dimensionless index number to show the wind sector). One can then assess how unique the associations of this or that variable are with the other variables (i.e., how orthogonal the relationships really are). If particular variables in the matrix show high correlations, regressions can be done to further quantify the relationship. Such an analysis would provide much firmer support for the authors' interpretation of sources.

Response

We appreciate the referee's helpful suggestions. The determination of source is based on the similarity of particle number size distributions between derived from PMF analysis and source emission profiles. For example: the peak diameter of particle number size distributions from the fresh traffic emission was shown at ~15 nm, which is similar as the factor 1 in this study (Wehner et al., 2002; Gramotnev and Ristovski, 2004). This method had been commonly used in PMF analysis to obtain the apportionment of particle number size distribution (Kim et al., 2004; Zhou et al., 2004; Zhou et al., 2005; Yue et al., 2008). In this study, we used all the size distributions as input, the units are same. Hence we thought the normalization would not change the results.

The aim of this study is to evaluate the effects of traffic control measures during the 2008 Olympic Games. Hence, we have to assume that the source profile was consistent in August from 2004 to 2008. Unfortunately, we only have the complete data set during the CAREBeijing2008 intensive campaign. Hence, we are not able to

put other parameters like gaseous pollutants and chemical components as variables in the input matrix.

10.Comment

P5179, lines 8-9 The assertion that Factor 4 does not show a significant reduction while the other factors do seems poorly supported. Why is a 30% reduction considered insignificant while a 43% reduction is? What criteria are being used here? Furthermore, one would expect a secondary aerosol factor to be reduced since, presumably, emissions of precursor gas concentrations are also being reduced.

Response

We said that factor 4 does not show a significant reduction, this is only compared with the other three factors. We admitted this is not fair. We have revised this.

11.Comment

P5179, line 15 through the end of the paper This comment deals with the conclusions and is really a reiteration of earlier comments, especially comment 2, which questions the novelty of the conclusions reached in this analysis. For example, the authors conclude that imposed reductions in traffic and construction activity are the main reason why particle number and volume concentrations have been reduced. This scarcely differs at all from the conclusions previously reached by the numerous earlier studies cited by the authors themselves, and is completely expected. They also maintain (lines 10-12) that they demonstrate that air mass origins were not the key factor in the observed particle concentration reductions. If by "key" they mean "only" then I would agree but this does not really contradict earlier studies. For example, Gao et al (2011) only claim that the air flow patterns play a role and I see nothing in this study (as per my comment 7) to contradict this. I feel the authors, assuming that they can do it, must make a better case for the value of the work done here.

Response

See comments 2 and 7.

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