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Atmospheric Chemistry and Physics Discussions

# *Interactive comment on* "Long-term measurements of particle number size distributions and the relationships with air mass history and source apportionment in the summer of Beijing" *by* Z. B. Wang et al.

**Anonymous Referee #2**

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Review of "Long-term measurements of particle number size distributions . . ." by Wang et al (acp-2012-1045)

General comment This study presents some interesting data and a potentially useful analysis but is not, in my view, publishable in its current form. Firstly, the authors do not sufficiently justify yet another analysis of the reduction in pollution during the Beijing Olympics - brought about primarily by imposed restrictions on traffic and industrial emissions. Secondly, the analysis does not include a sufficiently quantitative assessment of the performance of the PMF model. Thirdly, the attribution of the PMF resolved



factors to various sources is poorly supported. Fourthly, while not a major point, the paper is not particularly well written and at times the prose becomes opaque. I realize that none of the authors are native speakers of English but I think they can do better than this. None of these factors, taken alone, are catastrophic but together they are daunting. Still, it is certainly conceivable that a sufficiently revised MS could be acceptable – but not this one. My specific comments in support of this evaluation are given below.

Specific comments

1. 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.

2. 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 im-

posed 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.

3. 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.

4. 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.

5. 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 $\sim$ 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.

6. 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.

7. P5175, Back Trajectory analysis The cluster analysis is interesting but is poorly pre-

sented. 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.

8. 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 $\sim 2$ – 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.

9. 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.

10. 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.

11. 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.

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