

## ***Anonymous Referee #1***

We thank this reviewer for helpful comments and suggestions. Below we provide a point-by-point response to this referee's comment. The page numbers refer to the original version published in ACPD.

### **1. Comment**

In this manuscript the long-term measurement of particle number size distribution was reported in Beijing. Back trajectory analysis was used to investigate the impacts of air mass origins on particle concentrations. PMF model was applied to apportion the particle sources. Particle number concentration and size distribution are important to human health, air quality, weather, and climate. It is crucial to investigate the particle number size distribution in a long-term to have a better understanding of the particle pollution and its impacts. As host of Olympics, Beijing implemented several emission control measures to reduce the air pollution. To evaluate the effectiveness of control measures, it is important to investigate not only the reduction of mass but also the reduction of particle number. Source apportionment can provide direct evidence of the reduction of various sources. However, the data analysis in this study was rather shallow. Some of the conclusions conflicted with the previous studies, but the authors did not offer convinced explanations. The English should be well polished. There are many grammar errors and inappropriate sentences. Insufficient writing would reduce the readability of the manuscript. I only list some of those language errors. I suggest that the manuscript should be major revised before publication can be considered in ACP. Abstract: some results and conclusions conflicted with the previous studies. More convinced explanations are needed. e.g., line 15, air mass origin is always an important role for particle pollution; Line 19, the secondary pollution was enhanced in August 2008. In addition, the sources that different factors stand for are needed to be well interpreted.

### **[Response](#)**

Thanks for your comment. We have improved both the language and data analysis of our manuscript. Please find the response to each comment below.

## **2. Comment**

I am a bit disappointed that the authors completely ignored the discussions of the fine particle mode (i.e., 3-20 nm). This mode is essential to the total aerosol number concentrations (i.e., Atmos. Chem. Phys. 10, 4953, 2010; Chem. Rev. 112, 1957, 2012; Science 328, 1366, 2010). Since their measurements did cover the particle size down to 3 nm, it would be highly desirable that the trend of fine particle mode is discussed during the period of 2004-2008, which has not been previously reported.

### Response

Yes, it is true that nucleation mode (3-20 nm) is the essential to the total particle number concentrations. However, its number concentration varies greatly depend on the occurrence of new particle formation event. In the revised manuscript, we have added the discussions of both increasing nucleation event and the reduction of nucleation mode particles:

“The numbers of new particle formation (NPF) event days were 8 and 6 during 2004-2007 and 2008, respectively. However, the higher NPF event frequency in August 2008 did not result in the higher number concentration for nucleation mode particles, which was reduced by 46% compared with the August periods 2004-2007. This phenomenon might be ascribed to two reasons: Firstly, The various actions taken during the Olympic periods reduced the emissions of gaseous precursors of newly formed particles such as SO<sub>2</sub> and VOCs (Wang et al., 2009). Hence, even we observed more NPF events in 2008, the strength of nucleation event decreased in 2008. Secondly, considering the PKU site is the representative of urban environment of Beijing. The nucleation mode particles are not only produced during the atmospheric nucleation process, but also by the freshly traffic emissions. Therefore, like the Aitken mode particles, the traffic emission is also a major source of nucleation mode particle at PKU site. Several temporal measures, like those that about half of the private vehicles were not allowed on the road, had been taken during the

summertime in 2008, resulting in the significant reductions both on nucleation and Aitken modes particles.”

### **3. Comment**

Introduction: the introduction is deficient. The authors need to summarize not only what the previous work did but also what their results and main conclusions.

#### *Response*

Thanks for pointing this out. We have improved it in the revised manuscript.

### **4. Comment**

Page 5168 Line 7: inappropriate word, use “negligible” Page 5168 Line 29: grammar error “had been”

#### *Response*

Corrected.

### **5. Comment**

Session 2.2: two different instruments were used, so inter-comparison is needed.

#### *Response*

Sorry for the misunderstanding. We used the same instrument TDMPS to measure ambient particle number size distributions from 2004 to 2008. The only differences before and after 2007 was the particle size range we measured.

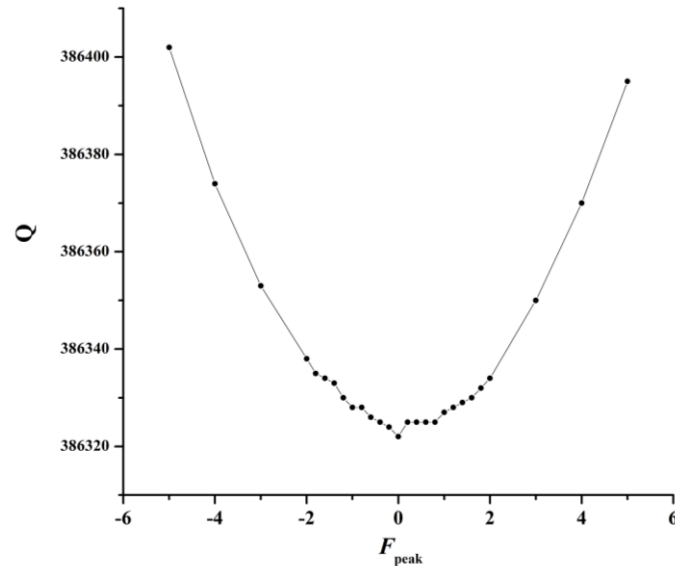
### **6. Comment**

Session 2.3.2: statistic results of the PMF model are needed. Why to choose five factors. How results will change when use different F-peak value and initial input. What are the sensitivity analysis results?

#### *Response*

In PMF analysis, if the assumption of the source profile and the estimation of the errors in the input data are accurate, The value of  $Q/Q_{\text{exp}}$  should be close to 1 (Ulbrich et al., 2009). Hence, the factor number was chosen as 4 because the value of

$Q/Q_{\text{exp}}$  is 0.96. Meanwhile, the  $Q$  value versus  $F_{\text{peak}}$  plot show a typical “U” shape with the lowest  $Q$  value corresponding to  $F_{\text{peak}}$  of 0, as the figure shown below. We have clarified these in the revised version.



## 7. Comment

Page 5174 Line 9: “Meteorological conditions did not: : :” It is not true. The meteorological conditions in August were quite different from 2004 to 2008.

### Response

Thanks for pointing that. We have deleted this sentence in the revised manuscript.

## 8. Comment

Session 3.1: As an important session of this manuscript, particle size distribution should be well discussed here, not only to show some numbers but comparing and explaining the differences.

### Response

In the revised manuscript, we have added some sentences to describe the particle size distributions. Meanwhile, followed the comment 2, we have compared the variations of nucleation mode particles and provided the possible explanations.

## **9. Comment**

Page 5175 line 9-10: It makes no sense. The air quality has nothing to do with vehicle population but vehicle in use. This sentence is of no use. In addition, the detailed control measures during Olympics should be briefly described here to make it clear to audiences who are not familiar with the control policy.

### **Response**

Yes, we do agree your comment. Here we have deleted this sentence (Line 9-10). Meanwhile, we have added one paragraph in the revised manuscript to describe the controls taken during Olympics.

“Several temporary measures had been taken to improve the air quality in August 2008. Since July 1st, total 30000 diesel and heavy polluting vehicles were not allowed to drive in Beijing. Meanwhile, only those vehicles meeting emission standards equivalent to Euro 2 were allowed to enter Beijing. From July 20, the even and odd-numbered license plates rule was performed, which might controlled 45% of total vehicles. During Olympic Games periods (8-23 August ), the extra 20% of governmental cars were not allowed to drive. In addition, the outdoor construction activities were stop and some gas stations were closed.”

## **10. Comment**

Page 5176 line 15-19: It cannot conclude that the air mass was not the key factor. Actually, meteorological condition is always an important factor for air pollution. The results of this work only can imply besides meteorological condition, other factors can also impact the air quality. In addition, wind speed is also an important factor. The authors should do the cluster analysis separately of 2004-2007 and 2008. Comparison of these results can probably give better explanation.

### **Response**

Thanks, the conclusion we had made is unfair. In the revised version, 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 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.

Meanwhile, the aim of this study is to compare the variations of particle number and volume concentrations. Hence, we did the cluster analysis together from 2004 to 2008. We appreciate that the referee's suggestion to do the cluster analysis separately. However, the results might not be comparable between 2004-2007 and 2008.

## **11.Comment**

Page 5177 line 4-6: Again, it's not true. Page 5177 line 26-27: "suggesting : : : " I don't really know the meaning of this sentence.

### [Response](#)

We have sharpened our conclusion: "The results pointed out so far that the improved air quality in August 2008 might not be only ascribed to the influence of different meteorological condition such as air mass history. The reductions of source emissions is also a major factor which could not be ignored....".

We want to explain that the vehicle exhaust emission may produce some gas phase pollutants, which could nucleated to form new particles.

## **12.Comment**

Session 3.3: the sources that each factor represents for should be better analyzed: Factor 1, in spite of exclusion of the new particle formation events, the nucleation process is always happening in the atmosphere. The diurnal variation of factor 1 does not have rush hour peak. To the contrary, factor 1 was higher in daytime, indicating it may relate to photochemical process. Thus factor 1 probably represents the growth of newly formed particles.

### Response

Thanks for your comment. It is true that the nucleation process is always happening in the atmosphere (Kulmala, 2003; Zhang et al., 2012). However, the previous study showed that in the polluted case, these newly formed particles are captured by the pre-existing particles, which cause the NPF event is not observed in the Beijing urban (Wang et al., 2011). Considering that we have excluded all the NPF event days in the dataset, the factor 1 should not contributed by the gas to particle nucleation.

As we have declared in the manuscript, PKU site could represent the urban environment of Beijing. The traffic emission is always a major source around the sampling site. The vehicle exhaust emission may produce some gas phase pollutants, which could nucleated to form new particles. Besides that, we have compared with other previous studies (Zhou et al., 2004; Zhou et al., 2005; Yue et al., 2008), the similar particle size distribution implied that the factor 1 should represent the local traffic.

### **13.Comment**

Factor 2, the first peak appears at noon time, but how to explain the second peak? Previous studies indicated there is no evening peak in Beijing. Even if there is evening rush hour, the particles from traffic cannot be aged after sunset when there is no sun light.

### Response

The factor 2 was composed of two parts: 1. the aged traffic-related particles from more distant areas of the sampling site. 2. the direct emission from the diesel and gasoline engine vehicles. The first peak of factor 2 might combined with these two sources. However, the evening peak might only contributed by the direct emission. Because as the referee said, the small particles from traffic such as factor 1 cannot be aged after sunset when there is no sun light.

### **14.Comment**

Page 5178 line 21:” besides traffic” did the author mean traffic emission is also

included in this factor?

**Response**

Sorry for the misunderstanding. The traffic emission is also one of the combustion sources, here we want to say that that factor 3 is mainly contributed by the other combustion source, not traffic emission.

**15.Comment**

Page 5179 line 1-10: Factor 4: several studies confirmed that in August 2008, the secondary formation was enhanced. The result of this work conflicts the previous results. Explain why.

**Response**

In previous studies, they only implied the relative fraction of secondary aerosols in PM<sub>2.5</sub> was enhanced, which is due to the decreasing of primary pollutants. In fact, both of the concentrations of primary and secondary aerosols were decreased. In this study, we found the concentration of factor 4, representing the secondary aerosols, was shown the decreasing trend as the other three factors, which is not conflict with the previous studies.

**16.Comment:**

Page 5179 line 10: “oxidation capacity” does the authors mean oxidability? Page 5179 line 16: what’s the meaning of “steady” here? Line 27: “significant cluster”? Page 5180, line 1: “The results show: : :”, line 13: “we also applied: : :” grammar error.

**Response:**

Corrected

**17.Comment**

Figure 2 is in consistent with figure 5. The size distribution in figure 5 has a strange shoulder, as well as figure 4.

**Response**

Sorry, we confused the dataset. The figures have been corrected in the revised version.



Thank you.

Kulmala, M.: How particles nucleate and grow, *Science*, 302, 1000-1001, 2003.

Ulbrich, I. M., Canagaratna, M. R., Zhang, Q., Worsnop, D. R., and Jimenez, J. L.: Interpretation of organic components from Positive Matrix Factorization of aerosol mass spectrometric data, *Atmos Chem Phys*, 9, 2891-2918, 10.5194/acp-9-2891-2009, 2009.

Wang, M., Zhu, T., Zheng, J., Zhang, R. Y., Zhang, S. Q., Xie, X. X., Han, Y. Q., and Li, Y.: Use of a mobile laboratory to evaluate changes in on-road air pollutants during the Beijing 2008 Summer Olympics, *Atmos Chem Phys*, 9, 8247-8263, 2009.

Wang, Z. B., Hu, M., Yue, D. L., Zheng, J., Zhang, R. Y., Wiedensohler, A., Wu, Z. J., Nieminen, T., and Boy, M.: Evaluation on the role of sulfuric acid in the mechanisms of new particle formation for Beijing case, *Atmos Chem Phys*, 11, 12663-12671, 10.5194/acp-11-12663-2011, 2011.

Yue, W., Stolzel, M., Cyrus, J., Pitz, M., Heinrich, J., Kreyling, W. G., Wichmann, H. E., Peters, A., Wang, S., and Hopke, P. K.: Source apportionment of ambient fine particle size distribution using positive matrix factorization in Erfurt, Germany, *Sci. Total Environ.*, 398, 133-144, DOI 10.1016/j.scitotenv.2008.02.049, 2008.

Zhang, R., Khalizov, A., Wang, L., Hu, M., and Xu, W.: Nucleation and Growth of Nanoparticles in the Atmosphere, *Chemical Reviews*, 112, 1957-2011, 10.1021/cr2001756, 2012.

Zhou, L. M., Kim, E., Hopke, P. K., Stanier, C. O., and Pandis, S.: Advanced factor analysis on Pittsburgh particle size-distribution data, *Aerosol Sci. Technol.*, 38, 118-132, 2004.

Zhou, L. M., Kim, E., Hopke, P. K., Stanier, C., and Pandis, S. N.: Mining airborne particulate size distribution data by positive matrix factorization, *J Geophys Res-Atmos*, 110, -, Artn D07s19  
Doi 10.1029/2004jd004707, 2005.