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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Highly time-resolved chemical characterization of atmospheric submicron particles during 2008 Beijing Olympic Games using an Aerodyne High-Resolution Aerosol Mass Spectrometer" *by* X.-F. Huang et al.

X.-F. Huang et al.

hely@pku.edu.cn

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General comments: This paper reports the characteristics of submicron aerosol in Beijing during the 2008 Olympic Games. The authors use an Aerodyne High-Resolution Time-of-Flight AMS to measure the chemical composition of aerosol particles. The dataset is unique and may be useful for assessing the effect of strict emission controls on the air quality during this time period. However, my impression is that this paper is a preliminary study. It needs important revisions before publication. Major comments are summarized below.

Major comments: - AMS measurements: One of the major conclusions of this paper is that the average PM1 concentration in the summer of 2008 was lower by 31% than that in 2006. Is this number well above the absolute accuracy of the AMS measurements? Is the AMS measurement in this study comparable to that by Sun et al. (2010)? I have a critical question about the AMS collection efficiency. In Figure 1, the ratio of AMS mass to TDMPS volume (i.e., density) often exceeds 2 (sometimes close to 3), which is unlikely the case with ambient aerosol. In addition, I have a critical question about the uncertainty associated with the AMS size cut. Previous studies observed relatively large contributions of supermicron particles to the accumulation mode in the Beijing atmosphere (van Pinxteren et al., JGR, 2009; Guo et al., ACP, 2010). A subtle change in the ambient size distribution may result in substantial difference in the mass concentration detected by the AMS. The authors should address these points and reconsider the significance of their conclusion (31% difference). Reply: The authors admit that the cutoff size of AMS is limited. As mentioned in section 2.2.1 in the paper, particles with vacuum aerodynamic diameters of 1 μm particles are transmitted through the inlet at an efficiency of $\sim$30-50% depending on exact details of the lens assembly and sampling pressure (Jayne et al., 2000; Liu et al., 2006). Therefore, the AMS measurement is typically referred to as PM1. Although van Pinxteren et al. (2009) and Guo et al. (2010) reported supermicron particles existing in the accumulation mode, their size distributions are at ambient RH conditions and thus the size distributions represent those of wet particles. When sampled by AMS, these wet supermicron particles are measured under vacuum, where they will lose most of water to shrink their sizes to some extent. The systemic comparison between vacuum aerodynamic diameter (by AMS) and aerodynamic diameter (by MOUDI) is complex and needs more information on particle density and shape, as suggested by Takegawa et al. (2009). Therefore, it is not feasible to compare the two types of diameters in this study. The scatter plot of the AMS mass vs the TDMPS volume gives their mean relationship as below: AMS mass=2.07*TDMPS volume R2=0.84 However, we do not think that this relationship can indicate an aerosol density of 2.07, because what TDMPS measured is between

3∼600 nm and what AMS measured is about PM1. So, the higher ratio (2.07) than normal aerosol density could be related to the particles between 600∼1000 nm and unmeasured crustal materials. The main purpose of the comparison between AMS and TDMPS is only to confirm their time trends. The conclusion of the reduction of PM1 mass in southerly flow from 2006 to 2008 is not a major conclusion of this paper. In both the studies in 2006 and 2008, the comparison between AMS and SMPS (or TDMPS) results was good, indicating the effectiveness of each measurement. A difference of 30% between 2006 and 2008 is an observational result and could be influenced to some degree by the difference of the two instruments used, which has been pointed out in the revised paper as below: "...in the Olympic campaign was largely decreased by 31%, suggesting possible pollution control effects during the Olympic period. However, this difference could also be influenced to some degree by the agreement of the different AMS instruments used." Reference Takegawa, N., Miyakawa, T., Watanabe, M., Kondo, Y., Miyazaki, Y., Han, S., Zhao, Y., Pinxteren, D., van, Bruggemann, E., Gnauk, T., Herrmann, H., Xiao, R., Deng, Z., Hu, M., Zhu, T., Zhang, Y., 2009. Performance of an aerodyne aerosol mass spectrometer (AMS) during intensive campaigns in China in the summer of 2006. Aerosol Science and Technology 43, 189–204.

- Cooking related organics: The COA mass spectrum extracted in this study was compared with those of chicken and hamburger cooking. Why chicken and hamburger? Are these foods representative in this region? The peaks associated with the lunch and dinner times are plausible, but the comparison of the mass spectra seems to me very superficial. Reply: Our group ever made AMS measurement of four typical types of Chinese cooking emissions. The comparison between the MS of COA in this study and those of Chinese cooking shows high similarity with respect to both the correlation coefficients of MS and most prominent ions (m/z 41 and m/z 55). For more details about this comparison, please refer to our recent publication as below. For the present paper, we have discarded the comparison between the MS of COA and those of chicken and hamburger cooking, but added the following sentences into the revised paper. "The MS of the COA is characterized by most prominent ions of m/z 41 (mainly

C3H5+) and m/z 55 (mainly C4H7+), which indicates large presence of unsaturated organic compounds (e.g., unsaturated fatty acids) and is well consistent with the MS characteristics measured for primary Chinese cooking emissions (He et al., 2010). For more details about the comparison between the MS of the COA and primary Chinese cooking emissions, please refer to another our recent publication (He et al., 2010)." He, L.-Y., Lin, Y., Huang, X.-F., Guo, S., Xue, L., Su, Q., Hu, M., Luan, S.-J., and Zhang, Y.-H.: Characterization of high-resolution aerosol mass spectra of primary organic aerosol emissions from Chinese cooking and biomass burning, Atmos. Chem. Phys. Discuss., 10, 21237-21257, doi:10.5194/acpd-10-21237-2010, 2010.

- OOA-1 and 2: The authors say that the mass spectra of OOA-1 and OOA-2 are similar. What is the importance of the separation of OOA-1 and OOA-2 in this case? Without other supporting data, I do not believe that the two types of OOA correspond to organic aerosols from different source regions. Reply: In fact, Table 1 gives the information why we have selected the 4 factor-solution that separated OOA into OOA-1 and OOA-2. In the 3 factor-solution, factor time trends, diurnal cycles, and spectra appear mixed with each other due to too few factors and the MS of HOA and COA seem not to be "clean" enough. In the 4 factor-solution, the two OOA factors have similar O/C ratios of 0.48 (OOA-1) and 0.47 (OOA-2), indicating they are not a result of large differences in volatility or oxygenation level. Instead, OOA-2 correlated better with sulfate and nitrate than OOA-1, indicating that the source regions of OOA-2 were more similar to those of SO2 and NOx emissions. This can also be supported by Fig. 5 (the back trajectory plot), the OOA-2/OOA-1 ratio ranged largely from 0.39 (for the BT cluster NWN) to 0.94 (for the BT cluster SE). In general, OOA-2 correlated better with sulfate and nitrate than OOA-1, indicating that the source regions of OOA-2 were more similar to those of SO2 and NOx emissions, consistent with the back trajectory analysis in section 3.4. Therefore, we finally selected the 4-factor solution and stated that the difference between OOA-1 and OOA-2 is most LIKELY to exist in source regions. The above discussion has been included in the paper.

- Scientific significance of this study: There have been a number of publications discussing the characteristics of aerosol in Beijing (especially the CAREBeijing-2006 special issue in JGR) and a number of publications discussing the effects of emission controls on the air quality during the Beijing Olympic Games (e.g., Witte et al., GRL, 2009). The authors should cite these previous studies and clarify the scientific significance (new findings) of this study. Reply: The statements below have been added into the introduction part of the paper: "Several satellite-based studies have recently indicated significant reduction of air pollutants during the 2008 Beijing Olympic Games (Cermak and Knutti, 2009; Mijling et al., 2009; Witte et al., 2009). For example, based on analysis of aerosol optical thickness, Cermak and Knutti (2009) suggested that the magnitude of the aerosol load reduction during the Olympic period was at 10∼15% compared to that expected for without emission reductions. A modeling study also supported significant pollutant reduction during the Olympic period (Wang et al., 2010). More detailed studies, especially analysis of ground-level measurement results with high time resolution, are necessary to interpret in depth variation of surface air quality during the Olympic period." "In order to characterize in depth the processes and mechanisms of severe air pollution in Beijing on a regional scale, an international field campaign "Campaigns of Air Quality Research in Beijing and Surrounding Region 2006" (CAREBeijing-2006) was conducted in summer 2006. The publications of CAREBeijing-2006 about aerosol studies indicated that aerosol pollution in Beijing was a regional problem on a scale of up to 1000 km (Garland et al., 2009; Jung et al., 2009; Matsui et al., 2009) and high PM periods were usually associated with air masses from the south with high concentrations of sulfate, nitrate, and ammonium (Takegawa et al., 2009; van Pinxteren et al., 2009; Yue et al., 2009). However, high time resolution variations of PM were little demonstrated in CAREBeijing-2006. In addition, the complex organic aerosol was not classified into different types to explore their corresponding sources and formation mechanisms in CAREBeijing-2006." In addition, relevant previous publications of CAREBeijing-2006 have also been cited to compare or support conclusion at necessary places of the paper.

Minor comments: - Abstract and other sections: The authors suggest that northern air masses were influenced by local emissions. What was the spatial scale of the local emissions? Reply: As the north to Beijing is dominated by mountains, "local" just means the urban area of Beijing, which is about 750 km2. This information has been added into the first sentence of section 2.1 as "HR-ToF-AMS measurement of airborne fine particles was performed continuously between July 24 and September 20, 2008 on the campus of Peking University (PKU) in the northwest of the urban area of Beijing, which is about 750 km2."

- Table 1: Table 1 should be included in the supplemental material. This information is not useful for general readers. Reply: More and more AMS users are using PMF to analyze ambient AMS datasets and we think Table 1 is necessary for these readers to understand the solution of PMF analysis. So, we would like to keep Table 1 in the main part of the paper.

- Ion balance: What was the ion balance during the time period? Reply: The plot of ion balance, i.e., the measured NH4+ vs the NH4+ needed to fully neutralize sulfate, nitrate, and chloride (2SO42-+NO3-+Cl-), has a slope of 1.05 and a correlation coefficient of R2=0.95. This indicates good ion balance and neutralized particles during the campaign. This information has been added into the paper as below. "During the campaign, the measured NH4+ matched well the NH4+ needed to fully neutralize sulfate, nitrate, and chloride (i.e., 2SO42-+NO3-+Cl-), with a linear correlation coefficient of R2=0.95 and a slope of 1.05."

-Nucleation: I am curious to see if there was a nucleation event during the time period. Reply: The nucleation events during the campaign are reported in another paper in this ACP special issue. Yue, D. L., Hu, M., Zhang, R. Y., Wang, Z. B., Zheng, J., Wu, Z. J., Wiedensohler, A., He, L. Y., Huang, X. F., and Zhu, T.: The roles of sulfuric acid in new particle formation and growth in the mega-city of Beijing, Atmos. Chem. Phys., 10, 4953-4960, doi:10.5194/acp-10-4953-2010, 2010.

Please also note the supplement to this comment:
http://www.atmos-chem-phys-discuss.net/10/C7364/2010/acpd-10-C7364-2010-supplement.pdf

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