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Interactive comment on “Highly time-resolved chemical characterization of atmospheric fine particles during 2010 Shanghai World Expo” by X.-F. Huang et al.

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This paper reports an online mass spectrometry study of aerosols in Shanghai during 2010 World Expo. Typical AMS data were obtained including highly time-resolved mass concentration of major non-refractory aerosol components, OM/OC ratio and oxidation levels of organic mass etc. SP2 was used to measure mass loading and mixing state of BC. This is the first AMS and SP2 measurement in Shanghai region and deserves to be presented to the society. However the manuscript needs to be revised before the following issues being addressed.

My two major concerns are: 1. As the authors mentioned that a series of air pollu-

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tion control measures were implemented during the World Expo, so the air quality in Shanghai was much better at that time. In other words, observations made in this sampling period might not be representative ones for the region. The authors should explain more about the importance of the study other than the first AMS measurement in the region. **REPLY:** Although the pollution control measures made the air quality during the World Expo better than before, this difference should be quantitative, and the basic pollution mechanisms of air pollution in Shanghai should not change. The purpose of this paper is to describe both the basic characteristics of fine particle pollution in this megacity and some unique features during the World Expo. For example, although biomass burning was ever found to be a significant source of fine particles in Shanghai, its contributions were found to be negligible in this study due to the pollution control measures. The purpose of this paper has been stated more clearly in the introduction part, as below: “This paper summarizes and analyzes the findings based on the advanced on-line measurement techniques during the 2010 Shanghai World Expo, in order to reveal both the basic characteristics of fine particle pollution in this mega-city and some unique features related to the emission control measures during the World Expo.”

2. The authors should take full advantage of the highly time- and particle-resolved information offered by AMS and SP2 and give more sophisticated data analysis. The results and discussion in the current manuscript are kind of simple compared with the previous papers (about the studies in Beijing and PRD region) from the same group. Besides, a lot of studies (only three of them were cited) have been carried out on the aerosols in Shanghai. More discussion and comparison regarding to the previous works are expected here. **REPLY:** The current discussion in the manuscript is a result after the authors’ careful consideration. We have strengthened some discussion according to the following detailed comments by the reviewer. More significant data analysis using the highly time-resolved data should be based on detailed meteorology analysis and modeling work, which are currently beyond the scope of this paper. After searching through the literature, it was found that the aerosol studies in Shanghai were not as

many as expected. Especially, the emphasis in this paper is about organic aerosol and its sources, but actually the previous source apportionment results of organic aerosol in Shanghai are very limited. After double checking, the authors believe that we have cited most of the comparable previous results in the literature.

Some specific comments more or less related to the above major concerns: 1. Page 1099, line 3: More technique details should be given for the calculation of PM1 mass concentration. Does the value include other refractory materials such as mineral dust?

REPLY: The relevant text has been modified to give more detailed information, as below. "Fig. 1a shows the time series of the PM1 mass concentration (calculated as the sum of organic matter (OM), SO₄²⁻, NO₃⁻, Cl⁻ and NH₄⁺ measured by AMS and BC measured by SP2) during the entire campaign."

2. Page 1099, line5: Is this the first reported PM1 measurement in Shanghai? If not, please compare the data with previous work. REPLY: After checking again in the literature, it was confirmed that this was the first time to report PM1 in the ambient air of Shanghai.

3. Page 1100, line 10: Comparison with previously reported OM/OC values by filtration studies in Shanghai are expected here. REPLY: The OM/OC ratio of organic aerosol is very hard to determine. To our knowledge, this ratio in Shanghai is reported in this paper for the first time.

4. Page 1100, line 14-19: How about the correlation between the time series of O/C ratio and sulfate or nitrate concentrations? Can any OC fragments be identified to give more specific explanation of the lower O/C ratio in China? REPLY: The correlation coefficient (R^2) between O/C and sulfate concentration is 0.111, and it is 0.018 between O/C and nitrate concentration. Both the two correlation coefficients are not significant enough, which indicates that the aging of organic aerosol is a much more complex process than the formation of sulfate and nitrate. As shown in Fig. 4a in the paper, due to the complexity of organic aerosol, the oxygen atoms almost exist in all ion frag-

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ments, therefore, the low O/C values in China are determined by a large amount of oxygen-containing fragments rather than a few main fragments.

5. Page 1100, line 20: Different mixing states of BC are not fully discussed (as in Huang, 2010, JGR) here. REPLY: We have strengthened the relevant discussion, as below. “Fig. 1c also shows the time series of the number fraction of internally mixed BC (referred to as NIB later) detected by SP2 during the campaign. While freshly emitted BC is generally externally mixed with other aerosol materials, various processes in the ambient atmosphere especially condensation of secondary species would transform it to the internal mixing state. Although the internally mixed BC particles were ever reported to account for $\sim 70\%$ in biomass burning plumes (Schwarz et al., 2008b), the emissions from biomass burning were strictly controlled during the World EXPO and found to contribute little to fine particles by source apportionment analysis in section 3.3. Therefore, the NIB can be used to roughly distinguish fresh emissions and aged particles in this study. The NIB was observed to vary in the range of 16.0–71.7%, with an average of 41.2%, indicating that the bulk BC was dominated by fresh and thus local combustion emissions during the campaign. This result is similar to those previously observed in another mega-city in China, Shenzhen, with the average NIB of 40–46% (Huang et al., 2012). In addition, it is found in Fig. 1 that during the period of May 19–27, the PM₁, BC, and NIB maintained at high levels, indicating that aged air mass and thus regional transport played an important role in the formation of these high pollution days.”

6. Page 1101, line 13: The averaged diurnal pattern of sulfate could have buried the real variations as shown in Fig. 1. REPLY: Although the average process could bury some special variations of species in individual days, it would not bury the general features of the diurnal pattern. For example, in Fig.3, we see stale concentrations for sulfate but daytime low concentrations for nitrate. This difference should be attributed to the diurnal variation of meteorology and the chemistry of species rather than the average process.

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7. Page 1101, line 17-19: Does this mean all the inorganic acids were neutralized by ammonia all the time? Is there any particle acidity variation? REPLY: The aerosol species that AMS can measure include $(\text{NH}_4)_2\text{SO}_4$, NH_4HSO_4 , NH_4NO_3 and NH_4Cl , therefore, the NH_4 concentration is a combined result of the variations of the above species. The original sentence has been modified to make the information expressed more accurate, as below: “The diurnal pattern of ammonium observed was generally the combined result of the variations of $(\text{NH}_4)_2\text{SO}_4/\text{NH}_4\text{HSO}_4$, NH_4NO_3 and NH_4Cl .” Actually, AMS cannot directly distinguish SO_4^{2-} and HSO_4^- . Although by comparing the amounts of anions (SO_4^{2-} , NO_3^- and Cl^-) and cation (NH_4^+) we can infer the ion balance and thus the particle acidity, the recent findings indicate that the measurement of NH_4^+ by AMS can be potentially influenced by organonitrates (Farmer et al. PNAS, 6670–6675, 2010). Therefore, we believe it is not suitable to discuss particle acidity by AMS measurement in this study.

8. Page 1103, line 21-29: Is there any chemical kinetic explanation for the time scales here (3h delay between HOA and SV-OOA peaks and 2h delay between SV-OOA and LV-OOA peaks)? Are the time delays same with the measurements in different sites? REPLY: Since organic aerosol (HOA, SV-OOA, LV-OOA) is a mixture of various compounds, the chemical kinetics of its formation are not well understood yet. Therefore, more chemical kinetic explanation for the time delay among HOA, SV-OOA and LV-OOA peaks may be over-interpretation. These time delays were also found in New York (Sun et al., 2011). In the revised paper, we have cited the result in New York to support our finding, as below. “Similar phenomena were also observed previously in the diurnal patterns of HOA, SV-OOA and LV-OOA in New York (Sun et al., 2011). This interesting time delay of the daytime peaks of the three organic components could imply a photochemical oxidizing process from HOA (and/or concurrently emitted gaseous organic pollutants) to SV-OOA to LV-OOA, which is well consistent with the recent finding that the aging of organic aerosol is a continuum in the atmosphere (Jimenez et al., 2009), and the responsible mechanisms need to be explored by modeling work in future.”

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9. Fig.3: SO₄, NO₃ and NH₄ should be SO₄²⁻, NO₃⁻ and NH₄⁺ respectively. REPLY: Corrected.

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