Anonymous Referee #3

General comments:

In this manuscript high-time resolution chemical and optical properties of aerosols were studied in the urban area of Shanghai, China. This information was useful to obtain he particle properties in Shanghai. The data collected in this study was limited, only 3-day data, but since there were no earlier studies in this region, these measurements provided the basic data set of aerosol properties in Shanghai. However, the data analysis in this study was not deep enough. Only correlations and comparisons were performed, which yielded little on the relationship between aerosol chemical and optical properties. New findings were adequately mentioned in the conclusions of this manuscript. Some statements were not persuasive and seemed to conflict with each other. Many corrections and clarifications were necessary to improve the manuscript. I suggest that the manuscript should be substantially revised before publishing in ACP.

We thank the reviewer for the valuable comments on this manuscript. We accept most of this reviewer's suggestions and made substantial revisions. We reorganized Part 3.3 and tried to clarify the impacts of chemical mixing state on the aerosol optical properties. We rewrote the conclusions focusing on the new findings of this work. Below are our point by point responses.

Specific comments:

1. Introduction: There have been many studies to investigate the relationship between aerosol chemical and optical properties, such as IMPROVE, and there have also some studies in Beijing and PRD region. However, those previous studies were not included in the introduction part.

We accept this suggestion by adding the following description of the previous studies at the end of Paragraph 1 in the introduction:

"Many studies have investigated the relationship between aerosol chemical and optical properties. For example, the Interagency Monitoring of Protected Visual Environments (IMPROVE) algorithm involving different chemical components with known mass extinction efficiency and hydroscopicity has been adopted by the U.S. Environmental Protection Agency (EPA) for estimating light extinction (Pitchford et al., 2007). Studies in China, such as in Beijing (Roger et al., 2009; Huang et al., 2010) and the Pearl River Delta (PRD) region (Garland et al., 2008), also contribute to the understanding of aerosol chemical and optical properties."

2. Page 31958, Line 10-15. Besides field study there are many important lab studies on the optical properties of soot aging. The introduction should give a complete picture of this area, from fieldwork to the lab work. In addition, many of the previous studies have showed that aged soot can effectively enhance aerosol optical properties. Those aspects should be clarified in this introduction. Slowik et al. Aerosol Sci. Technol. 2007, 41, 734 Qiu et al. Sci. Technol. 2012, 46, 4474-4480 Xue et al., Environ. Sci. Technol. 43, 2787–2792 (2009). Pagels et al., Aerosol Sci. Tech. 43, 629–640 (2009) Khalizov et al., J. Phys. Chem. 113, 1066–1074 (2009). Xue et al., Phys. Chem. Chem. Phys. 11, 7865-7875, DOI:10.1039/b700001a (2009). Khalizov et al., J. Geophys. Res. 114, D05208, doi:10.1029/2008JD010595 (2009).

We accept this suggestion by adding the discussion of lab studies on the optical properties of soot aging and citing the related literatures at the end of Paragraph 2 in the introduction:

"A series of laboratory studies have been carried out to investigate effect of coating soot aerosol with different coating materials, such as sulfuric acid (Zhang et al., 2008; Khalizov et al., 2009a; Khalizov et al., 2009b; Pagels et al., 2009), organic acids (Slowik et al., 2007; Xue et al., 2009a, b) and secondary organic coating (Qiu et al., 2012; Khalizov et al., 2013). These coatings were observed to increase soot particles' hygroscopicity and to effectively enhance their optical properties."

3. Page 31958, Line 18: severe air pollution? Use numbers, like PM concentration increase, energy consumption increase, vehicle use increase.

We add several sentences in the revised manuscript using numbers to describe how severe the air pollution is in China:

"With rapid economic growth and urbanization, mega-cities in China have experienced severe air pollution problems (Chan and Yao, 2008). The energy consumption of China is increasing at an annual growth rate of as high as 10%. During 2000-2010, the installed capacity of thermal power plants and the vehicle population increased 195% and 300%, respectively. By the end of 2010, the vehicle population in China exceeded 200 million (Wang and Hao, 2012). Tropospheric NO_2 over eastern China, especially above the industrial areas, increased with the fast economic growth, e.g. the growth rate of NO_2 column in Shanghai was 20%±6% per year (van der A et al., 2006). The annual average of PM_{10} (particulate matters with diameters smaller than 10 μm) concentrations in 113 key cities in China was 82 μg m^{-3}, which is about 4-6 times that in the developed countries (Wang and Hao, 2012)."

4. Page 31958, Line 23-25. Summarize the results of previous studies.

We don't quite understand this comment since we have summarized the results of previous studies in the manuscript as "Sulfate, nitrate, ammonium, organic matter, and elemental carbon have been identified as the major chemical components in fine particles in Chinese mega-cities and contribute over 90% of the extinction coefficient (Cao et al., 2012; Wang et al., 2012; Zhang et al., 2012a)."

5. Page 31959, Line 1-7. This part should be included in the experiment part, not here.

Since this part has little connection with the topic of this manuscript, we removed this part in the revised manuscript.

6. Page 31959, Line 10-18. Rewrite this part. Summarize your work: what did you want to do, and how did you do?

We accept this suggestion and rewrote this part as follows:

"We used the Aerosol Time-of-Flight Mass Spectrometer (ATOFMS) to measure the size, chemical composition, and mixing state of individual particles. A Monitor for AeRosols and GAses (MARGA) was also run, to quantitatively measure the bulk concentration of particulate inorganic ions. The aerosol optical properties (including scattering and absorption coefficients and single scattering albedo) were monitored by a home built Cavity Ring-Down Spectrometer (CRDS) and an integrating nephelometer. As the fine particles significantly affect atmospheric visibility (Seinfeld and Pandis, 2006), we will emphasize the influence of the evolution of chemical species in fine particles on their optical properties. This is the first report of highly time-resolved variations of aerosol optical properties associated with chemical components of single particles in Shanghai."

7. Page 31961, Line 15. 0.8 inch? Or unit missing?

We add "inch" in the revised manuscript.

8. Page 31963, Line 2. 6 m long stainless steel pipe. Did you calculate the particle loss in the sampling inlet, since 6 m inlet could result in a particle loss.

Yes, we have compared the particle number concentration measured through this 6-m long sampling pipe with the direct sampling result. With the help of the cyclone pump, particle loss in the sampling inlet is negligible.

9. Page 31963, Line 10. Where was the Shanghai Meteorological Bureau? How far was it from the sampling site? Can this data represent the condition at the sampling site?

The meteorological data in this work were obtained at the station 2.6 km west of the Fudan campus. Parallel measurements of PM_1 and $PM_{2.5}$ mass concentrations at both sites since May, 2012 confirm that data collected at this station can represent the conditions at the sampling site.

10. Page 31963, Line 19. From Fig. 1, except 14th Oct. I don't think the wind data were consistent with back trajectories. In addition it made no sense to use three figures to present back trajectories in three days, instead of three periods.

The wind data shown in Figure 1 are presented in a 30-min time resolution, while the back trajectories are shown with 6-hr time resolution. When the time resolution is increased to 1-hr for the back trajectories, they look quite consistent with the wind data (as shown in Figure R1). We used Fig.1 to present the background meteorological information, while the division of three periods was discussed later according to the particulate pollution.

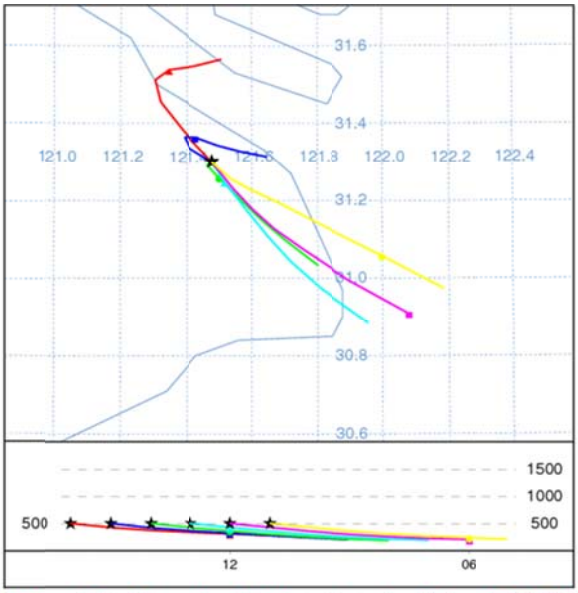

Figure R1. 1-h resolved back trajectories during 19:00-24:00 in Oct 13, 2011.

11. Page 31963, Line 20. "almost the entire day…" avoid using this ambiguous expression. There were many this kinds of expressions in the manuscript.

We change "almost the entire day" to "the first 19 hours" in Page 31963, Line 20.
We remove the "almost" in Page 31968, Line 23.
Page 31974, Line 9-11: We rewrote the sentence as below:
Particles in Period 2a had almost the same mass fraction of sulfate (37%), nitrate (38%) and ammonium (25%) with those of Period 2b (37%, 39%, and 24%) while they had distinct extinction behaviors.

12. Page 31964, Line 16: I am confused by the classification of the three periods. I don't think the period 2 should be from 23:00, 13 Oct. to 10:00 15 Oct. From back trajectories, the air mass origin definitely changed at some time during 14 Oct.

We defined the three periods based on the particulate pollution level. Period 2 was referred to as the highly polluted period. During Period 2, all the air masses came in from the west side of Shanghai passing through industrial areas. Indeed, the 48-hr back trajectories changed during Period 2, which might be the reason for the differences between Period 2a and Period 2b. We add an extra discussion in Part 3.3 about this possibility in the revised manuscript.

13. Page 31964, Line 25. Was there any overlap between ECOC particle and OC particle? Describe the criteria how to define the particle type.

There was no overlap between ECOC particle and OC particle. Since the particles were grouped by clustering, they were considered one class or another, but cannot be both. As shown in Fig. S2, ECOC particles show both significant carbon signals, such as $^{12}C^+$ and $^{36}C^+$, and organic carbon signals, such as $^{27}C_2H_3^+$ and $^{43}C_2H_3O^+$, while OC particles show more organic carbon signals, such

as $^{27}C_2H_3^+$, $^{37}C_3H^+$, $C_3H_3^+$and $^{43}C_2H_3O^+$ and much weaker intensity of carbon signals.

14. Page 31966, Line 13-15. The new particle formation and dust event were not directly measured, but in the statement from previous part, the author made an affirmative statement about this. Also, provide a definition of new particle formation in the present study (i.e., Zhang, Science 328, doi:10.1126/science.1189732, 1366-1367, 2010; Chem. Rev. 112, 1957-2011, 2012).

Indeed, the new particle formation was not directly measured in this work. We asserted that there was a new particle formation process simply based on the "banana" shape of the SMPS result. In the revised manuscript, we add the definition of the new particle formation when the term first appears in Part 3.1 with the citation of the suggested literatures as below:

"At midday on Oct 15, a new particle formation event was identified based on the typical "banana" shape in the two-dimensional contour plots (Fig. 2a) of SMPS measurement versus time (Heintzenberg et al., 2007). New particle formation occurs in two distinct stages, nucleation to form a critical nucleus and subsequent growth (Zhang, 2010; Zhang et al., 2012c)."

The identification of a dust event was based on a sudden decrease of the PM_1/PM_{10} ratio together with high concentrations of Ca^{2+} and Mg^{2+} and the fact that the air mass was transported over a long distance.

15. Page31966, Line 19. If it was from local origin, it should have spikes on the contrary.

We agree with this comment. We rewrote this sentence in the revised manuscript and add extra discussions about metal-containing particles in the supplementary materials (requested by the other reviewer) as below:

"The temporal profile of the Na-K-rich particle type presented no significant spikes during the whole sampling period, suggesting regional rather than local origins. Discussion of metal-containing particles is given in the supplementary materials."

In the supplementary materials:

"Metal-containing particles consist of several sub-groups including Fe-, Na-Al-K-Mn-, V- and Pb-containing particles, accounting for 4.3%, 2.0%, 0.7% and 1.2% of total particles, respectively. The Fe-containing particles can be further divided into three sub-types as well (Na-K-Fe-, 0.9%; Fe-S-N-, 1.7%; Fe-N-, 1.7%), according to the mass spectral patterns and temporal variations. Pb- and Fe-S-N-containing particles had similar temporal variations in number fraction (both having peaks during Period 2), indicating their sources were from the northwest of Shanghai. V-containing particles occurred only when the wind blew from the east suggesting ship emissions as their source (Ault et al., 2010). Fe-N- and Na-Al-K-Mn-containing particles had a nearly constant number fraction, indicating regional sources. Na-K-Fe-containing particles only occurred in Oct 13 with two spikes at around 11:00 and 16:00, possibly originating from a local source."

16. Page31967, Line 20. This expression would be misleading since the ratio of sulfate and nitrate to ammonium was called particle acidity, but it is not. Personally I don't like this method to estimate particle acidity and it cannot reflect particle acidity very well. However, it was really difficult to decide particle acidity. The author should clarify the uncertainty of this method. For my knowledge, the thermo dynamic model ISSORPIA was a better way to calculate acidity. Even if the authors did not use this to calculate acidity and re-analysis, this should be pointed out. Nenes, A. et. Al, Aquat. Geochem. 1998, 4 (1), 123-152. Guo, S. et. al, Environ. Sci. Technol., 2012, 46(18), 9846-9853

We agree with this comment that the ratio of sulfate and nitrate to ammonium is not a quantitative measure of particle acidity. At both urban and rural sites in east China, the sum of sulfate, nitrate and ammonia typically constituted 40%–57% of $PM_{2.5}$ mass (Yang et al., 2011). As the major acids and the only base in the atmosphere, sulfate, nitrate, and ammonium play dominant roles in determining the acid-base balance. The ratio of sulfate and nitrate to ammonium was used as an indicator of aerosol acidity in many previous studies (He et al., 2012; Zhang et al., 2012b). In the ATOFMS field, the ratio of the sum of absolute peak areas of $^{62}NO_3^-$ and $^{97}HSO_4^-$ to that of $^{18}NH_4^+$ has been used as an indicator of single particle acidity (Denkenberger et al., 2007; Pratt et al., 2009; Wang et al., 2010) for a while. However, this qualitative index has never been compared with other highly time-resolved quantitative measurements. In this work, we presented a good linear fit between qualitative and quantitative results and for the first time demonstrate the significance of this ratio as a semi-quantitative indicator of single particle acidity.

We take this reviewer's suggestion, rephrase this part of discussion clarifying that the ratio of sulfate and nitrate to ammonium is only an indicator of acidity, and refer the ISSORPIA model as the classical method to calculate particle acidity, as follows:

"One of the classical methods to calculate aerosol acidity is the thermodynamic model ISSORPIA (Nenes et al., 1998; Fountoukis and Nenes, 2007; Guo et al., 2012), which considers a thermodynamic stable state of the main inorganic ions (i.e. Na^+, K^+, Mg^{2+}, Ca^{2+}, NH_4^+, NO_3^-, SO_4^{2-}, Cl^-) in the particle phase."

17. Page31968, Line 7. It made no sense to use average size, because it cannot indicate particle distribution. Usually mass medium aerodynamic diameter (MMAD) or medium aerodynamic diameter (MAD) were used to describe the particle size distribution

Usually, particle size distribution sampled by ATOFMS follows the Normal Distribution. The average size is equal to median aerodynamic diameter (MAD) here.

18. Page31968, Line 25. During period 3, if there was dust, the acidity should be low. This result conflicted with your previous statement. In addition, particles from regional transport should be more aged, and have higher acidity than the local particles. There were also a few other conflicted results between particle origin and acidity.

During the dust event in this experiment, most of the dust particles were in coarse mode, so they had little impact on the concentration of fine particles. PM_1 concentration stayed low and no significant increase of dust particle number in this size range was recorded by the ATOFMS. So the statement of high acidity of fine particles in Period 3 didn't conflict with the dust event.

Regional transported particles are not necessarily more acidic than the local particles. Mega-cities in China, such as Beijing and Shanghai, have been observed to have acidic particles in urban areas due to deficiency of ammonia in the atmosphere (Pathak et al., 2009; Wang et al., 2009; He et al., 2012; Zhang et al., 2012b). In this work, with the help of online chemical analysis, we tried to test the above observation by showing the particle acidity change when regional transport brought more ammonium salt to urban Shanghai.

19. Page 31969, Line 25. This explanation was not persuasive. The SSA depended on the ratio of scattering to extinction.

We agree with this comment. The rain washed out the particulate matter in the atmosphere, especially those water-soluble scattering materials like sulfate, nitrate and ammonium. The decreased scattering coefficient should be the major reason for the low SSA value. We replaced the sentence with "This could be explained by the decreased ratio of water-soluble scattering materials like sulfate, nitrate and ammonium which were washed out more effectively by rain."

20. Part 3.3.1: The authors used two paragraphs to discuss the relationship between PM1 and optical properties, but there was no clear point or new findings here.

We agree with this comment that these two paragraphs provide little new findings. The original purpose of this part of the discussion is to show that the PM_1 mass concentration cannot explain all the variations of extinction coefficient and chemical composition must be considered. In the revised manuscript, we removed Part 3.3.1 and just left a few sentences to point out the inconsistency between PM_1 mass concentration and extinction coefficient. We added two more references here (Bergin et al., 2001; Garland et al., 2008) to support the statement that "In field studies, PM_1 has been reported to contribute over 80% of light extinction".

21. Part 3.3.2: Re-define periods really confused some readers. This part also compared some trends and showed some correlation that lacked integrated and quantitative analysis of the particle chemical and optical properties. This is really confusing and made it difficult to figure out the points of this part.

Actually, we didn't redefine the periods, but rather only subdivide them based on higher time-resolution data. Subdividing periods was aiming to facilitate the interpretation of one of our main topics that only when the particle mixing state stays stable, the method using PM mass extinction efficiency to assess visibility is meaningful. What we try to point out is: 1) When a quick change of aerosol chemical components occurs, simple correlation between PM mass concentration and extinction coefficient could lead to incorrect conclusions about the aerosol optical properties. 2) Analysis solely based on meteorology and bulk chemical measurements is

not always reliable, since different particle mixing states could cause distinct extinction coefficient (as shown in Fig. 5). The main focus of this part is: With the help of online single particle analysis, we can identify a time period with constant chemical composition and mixing state of the particles and ensure more precise interpretations of aerosol optical properties (mass extinction coefficient in this work).

22. Conclusion: it was more like a summary, not a conclusion. There were too few new findings here.

We rewrite the conclusion part focusing on the new findings of this work.

"Using high-time-resolution instruments, we investigated the evolution of the chemical and optical properties of PM_1 in the Shanghai urban area during a 72-hour sampling period. The air mass which originated from northwest of Shanghai brought a high concentration of particulate matter (PM) containing an increase in ammonium, nitrate and organic carbon species. The ratio of the sum of the absolute peak area of NO_3^- and HSO_4^- to that of NH_4^+ in the ATOFMS was used as an indicator of particle acidity and compared with the equivalent charge ratio of SO_4^{2-} and NO_3^- to NH_4^+ from MARGA data. The temporal profiles of both values showed that particle acidity decreased when regionally-transported air mass were present, and revealed that urban areas in mega-cities like Shanghai have more acidic particles due to a deficiency of ammonia in the atmosphere. The excellent correlation ($R^2 = 0.82$) between the ATOFMS particle acidity and the quantitative measurement by MARGA confirms the validity of the ATOFMS ratio of sulfate and nitrate to ammonium as a semi-quantitative measure of particle acidity.

The mass extinction coefficient was used to characterize aerosol optical properties in this work. Our highly time-resolved data showed that the method using PM mass extinction efficiency to assess the atmospheric visibility is only valid when the particle mixing state remains stable. With the help of the ATOFMS, we can identify time periods with constant chemical composition and mixing state of the particles and ensure more precise interpretations of aerosol optical properties. Chemical analysis at the single particle level showed that nitrate and ammonium played important roles in the particle scattering efficiency in Shanghai, while the addition of organic material decreased the scattering efficiency of particles containing scattering materials. In the current study, all the measurements were carried out under dry conditions. The influences of relative humidity on aerosol mixing state and the resulting optical property changes should be considered in future studies."

Reference:

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