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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Mixing state of individual submicron carbon-containing particles and their seasonal variation in urban Guangzhou, China" *by* G. Zhang et al.

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<General>

This paper presents characteristics of submicron carbon-containing aerosols in urban area of PRD during late spring and fall based on single particle measurements by a laser ablation mass spectrometer. The authors have classified the observed mass spectra using cluster analysis and found that the fraction of biomass burning particles significantly increased in fall. I think the paper is generally well-written and provides a good reference for the mixing state of submicron particles in this region. My major concern is the representativeness of the data presented here. The measurement periods

are too short to draw a general conclusion on seasonal variations. In my opinion, the paper would benefit by being more focused.

We would like to thank the reviewer for his/her useful comments and recommendations to improve the manuscript. The representiveness of the data was discussed in response to specific comment 2#. We also agreed with the comment that the study results provided here are limited to draw a general conclusion on season variations, and thus we clarified that this paper presents a case study of carbon-containing particles in spring and fall. The paper was revised to be more focus by improving the introduction and discussion sections, please refer to responses to the specific comments 1# and 2#. General and specific comments have been addressed below.

<Specific comments>

1. Introduction The importance of carbonaceous aerosols in the climate system is described in the introduction, but this is not directly linked with the major conclusions of this paper. Please focus on the current understanding of sources and processes of carbonaceous aerosols rather than their general importance. Please briefly review what is known and what is unknown regarding the mixing state of carbonaceous aerosols (not only in China but also other regions). Such a review is helpful to highlight new findings of this paper more clearly.

We agreed with the comment. Some of the description on the importance in the climate system of carbonaceous aerosols was removed, and we focused upon the introduction on the current understanding of sources and processes of carbonaceous aerosols accordingly. We have reviewed the previous studies on the mixing state of carbonaceous aerosols mainly using single particle mass spectrometer in Shanghai, China and also other regions, such as California, USA. Please refer to Lines 72-105 in the revised manuscript. According to the comments by Referee 2, results from PRIDE-PRD campaign on the physical and chemical properties of carbonaceous aerosol in the PRD region have also been added to the introduction section, please refer to Lines 61-68,

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and the sentence about Monsoon affects number fraction of carbon-containing particles as a function of aerodynamic diameter was deleted.

2. Experimental set-up The measurement periods (30 Apr to 22 May 2010 and 5-20 Nov 2010) are too limited to draw a general conclusion on seasonal variations. The authors mention that the instrument detected 700,000 particles for each period. What is the representativeness of the data? I would expect more particles in pollution episodes than average conditions. If the instrument detects 10 particles per second, for example, it needs only a day to obtain 700,000 particles. Please describe the representativeness of the data in more detail and clarify that this paper presents a case study rather than seasonal variations (the title should be also changed).

We agreed with the comment. The title has now been changed to 'Mixing state of individual submicron carbon-containing particles during spring and fall season in urban Guangzhou, China: a case study'.

The results for the comparison of carbon-containing particles between spring and fall were performed based on all the collected particles (i.e., 700,000 in number for each period). Typically, SPAMS detected 2000 particles per hour, and the number varied between several hundred during clean condition and several thousand within polluted episodes. Over the period of study, SPAMS measurements were nearly continuous for $\sim$21 and $\sim$15 days in the spring and fall periods, respectively. We believe that this amount of data for each period is sufficient to perform a statistical analysis, and to present representative results for this study. We have also reviewed some similar studies by other groups, and found that the number of analyzed particles, 600,000-700,000 for $\sim$25 days (Qin and Prather, 2006; Sullivan and Prather, 2007), was comparable or even less than those analyzed in our study. We have added the sentence "This amount of data should be sufficient to perform a statistical analysis, and thus to provide representative results in this study" to clarify the representativeness of data. Please refer to Lines 145-147 in the revised manuscript.

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3.2.2. Mixing state The discussion on nitrate aerosols in the last paragraph is too speculative. I recommend it should be removed.

We agreed with the comment and this paragraph has been removed.

3.2.3. Particle acidity I wonder how the relative acidity ratio depends on the variability of ionization efficiency due to the mixing state. If this parameter is only qualitative, I do not think it useful to show it in this paper. Laboratory experiments may be helpful to interpret this parameter.

This relative acidity ratio was first applied by Denkenberger, et al. (2007). Due to the matrix effect on the ionization process, it is believed that the relative acidity ratio may not be comparable between different particle classes. Nevertheless, it may still be applied to provide a comparison for particles with similar chemical pattern, and also provide an indication of the relative acidity of the particles with different sizes in this study. It is noted that it wasn't applied for quantitative results in this study. We only compared the relative acidity of carbon-containing particle types during spring and fall with different sizes, but with similar chemical components in this study.

4. Conclusions Again, the conclusions and implications should be more specific. I do not think the results of this paper could improve our understanding of atmospheric chemistry and reduce the uncertainty of climate modeling.

We agreed with the comment, and the conclusions and implications have been revised to be more specific. We have deleted the speculative conclusions, and revised the implication to "These results, by direct single particle observation, provide a reference of mixing state for calculating light extinction, or even modeling the climate forcing of aerosol in the PRD region with improved validity. They might also help to identify the source and to reveal the atmospheric processes of carbon-containing particles in the PRD region". Please refer to Lines 424-443 in the revised manuscript.

Reference

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Denkenberger, K. A., Moffet, R. C., Holecek, J. C., Rebotier, T. P., and Prather, K. A.: Real-time, single-particle measurements of oligomers in aged ambient aerosol particles, Environ. Sci. Technol., 41, 5439-5446, doi: 10.1021/es070329l, 2007. Qin, X. Y., and Prather, K. A.: Impact of biomass emissions on particle chemistry during the California Regional Particulate Air Quality Study, Intl. J. Mass. Spectrom., 258, 142-150, 2006. Sullivan, R. C., and Prather, K. A.: Investigations of the diurnal cycle and mixing state of oxalic acid in individual particles in Asian aerosol outflow, Environ. Sci. Technol., 41, 8062-8069, 2007.

Please also note the supplement to this comment:
http://www.atmos-chem-phys-discuss.net/12/C13575/2013/acpd-12-C13575-2013-supplement.zip

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Interactive comment on Atmos. Chem. Phys. Discuss., 12, 32707, 2012.