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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Characterization of submicron aerosols at a rural site in Pearl River Delta of China using an Aerodyne High-Resolution Aerosol Mass Spectrometer" *by* X.-F. Huang et al.

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General comments: This manuscript reports the chemical characteristics of submicron aerosol particles measured by high-resolution time-of-flight aerosol mass spectrometer (HR-ToF-AMS) at a rural site in the Pearl River Delta (PRD) of China. Since the sampling site located at the downwind of the central PRD area, the field data reported help to better understand how the rapid urbanization and industrialization of PRD influencing the regional air quality. This field study captured active biomass-burning (BB) activities during the later period of study but there are a few points to be clarified. To the best of my knowledge, it is the first HR-ToF-AMS field dataset reported in the PRD

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region and the authors discussed the observations with enough details. Overall, I suggest the publication of this work in ACP with minor revision. The specific comments are shown below.

Specific comments: 1. This work clearly shows the significant increase of biomass burning aerosols concentration after November 12. As shown in Figure 2d, BB event generated a huge amount of organic aerosols. To better compare with the literature, I suggest showing the average mass concentrations of BB and non-BB data individually in Figure 2g. It also help to know the background levels of each PM1 components at Kaiping before BB period Reply: Following this comment, we have divided the PM1 composition in Figure 2g into two cases, i.e., BB period and non-BB period. The comparison with the literature is now based on the non-BB case. The relevant text was updated accordingly.

2. In Section 3.3, the average diurnal pattern of N/C ratio has a large peak in the late afternoon. The authors suggest that this is due to the continuous accumulation of biomass burning aerosols generated in daytime. From Figure 4a, the daily peak position of N/C ratio looks matching quite well with that of O/C ratio in October, which means that the N/C ratio may be enhanced by the active photochemistry in noontime. It would be useful to average the BB and non-BB N/C ratio separately to see if there is any significance difference between them. Conducting the same diurnal analysis for other ratios and PM1 components is highly recommended. Reply: There is still significant difference between the diurnal patterns of O/C and N/C. The diurnal pattern of O/C starts to increase at 9 am and reaches the peak at 3 pm, which is generally consistent with photochemical activity. However, the diurnal pattern of N/C starts to increase at 6 am and reaches the peak at 5 pm, which is generally consistent with the farmers' daily activities. The N/C ratios for the non-BB and BB periods are 0.018 and 0.029, respectively, which clearly indicates that high N/C ratios are possibly related to biomass burning, as mentioned in the manuscript. Moreover, the correlation between N/C and O/C during the campaign gives a poor correlation coefficient of R2=0.002,

ACPD

10, C12307–C12311, 2011

further indicating that the N/C increase may not be closely related to photochemical activities in this study. The above information has been clarified in the modified paragraph as below. “Figs. 4e and 4f present the average diurnal variations of H/C, N/C, O/C and OM/OC. Since the ratios are influenced by relative organic constitutions rather than absolute organic concentrations, their diurnal patterns should be mostly attributed to the diurnal changing of relative importance of different sources. Both the O/C and OM/OC ratios start to increase at 9 am in the morning, and reach a peak at 3 pm in the afternoon when photochemistry is the most active to produce secondary organic aerosol with high O/C ratios. The H/C ratio shows a reverse diurnal pattern to those of O/C and OM/OC as expected. The diurnal pattern of N/C starts to increase at 6 pm in the early morning and reaches a peak at 5 pm in the late afternoon, which is generally consistent with the daily activities time of local farmers. This may imply that the accumulation of biomass burning-related aerosols in the atmosphere due to daytime burning events could be responsible for the continuous daytime N/C ratio increase.” In addition, the reason why we did not make diurnal patterns separately for BB and non-BB periods is that, the data of the BB period is only for 7 days, which cannot be used to obtain stable 24 h diurnal patterns to compare with other long periods validly.

3. It is recommended to highlight the BB data on the Van Krevelen diagram because the characteristics of freshly formed BB aerosols can be very different to the aged SOA. Reply: Following this suggestion, we have highlighted the data points for the BB period with blue in the V-K diagram. The following sentences have been added into the text to describe the characteristic. “The highlighted data points for the biomass burning period in Fig. 4d were mostly distributed in the upper left area, suggesting that the primary biomass burning orgnic aerosols usually had relatively lower O/C ratios but higher H/C ratios. This is consistent with the elemental composition of the BBOA component extracted from PMF analysis of OA in the next section.”

4. According to the PMF analysis, the N/C ratio of BBOA component is about 0.06, which is much higher than those of their laboratory-generated BB aerosols (N/C <0.02).

ACPD
10, C12307–C12311, 2011

What are the possible reasons causing such large difference? Also, it would be worth to compare the N/C ratio of BBOA identified in previous studies. Reply: We have given explanation about high N/C ratio in section 3.3 as below. “However, the direct HR-ToF-AMS measurement of burning plumes of rice straw and five other biomass materials in laboratory by our group did not find N/C ratios of above 0.02 (He et al., 2010). Thus, the high N/C ratios observed near the end of the campaign are inferred to be possibly due to secondary reactions of NH3 and NOx with organics in biomass burning plumes and/or simultaneous burning of high N-containing soil materials in the open field. Another possibility is that, biomass burning in PRD may also include other high N-containing biomass materials that were not tested by He et al. (2010), since Laskin et al. (2009) have found high abundance of N-heterocyclic alkaloids in biomass burning aerosols in test burns of ponderosa pine in the US.” In the MILAGRO project in Mexico City, the HR-ToF-AMS aircraft measurement dataset identified a BBOA component with N/C of 0.013 (DeCarlo et al., 2010) and the HR-ToF-AMS ground measurement dataset identified a BBOA component with N/C of 0.01 (Aiken et al., 2009). But just as analyzed above, different regions may have different biomass materials to burn, which is a possible reason for different emission levels of N-containing organics.

5. There are contradictory descriptions about the origin of BB aerosols. In Section 3.1, the authors mention that fresh BB aerosols are the major contributor to organic mass with smaller particle size, implying that there is a significant local BB aerosols emission. However, in Section 3.5, they conclude that regional transport from central PRD rather than local emission was the major origins of BBOA component based on the meteorological information. Please clarify. Reply: We have clarified this point by adding the following sentences in section 3.5. “Higher BBOA concentrations were mostly associated with wind speeds of <2 m/s, denoting the dominant role of local burning events. However, there are still some higher BBOA concentrations corresponding to wind speeds of >2 m/s, suggesting that the regional transport of BBOA also existed.”

6. Page 25854, L7-10: Revised the following sentence “. . .with LV-OOA correlating

ACPD

10, C12307–C12311, 2011

best with nitrate due to the common semi-volatility and SV-OOA correlating best with sulfate due to the common low volatility" to ". . .with SV-OOA correlating best with nitrate due to the common semi-volatility and LV-OOA correlating best with sulfate due to the common low volatility". Reply: Correction made.

7. The HR-ToF-AMS can measure PM1 non-refractory components. However, I expect that the MAAP measured BC in PM2.5 instead of in PM1 in this study. If this is the case, it is not very appropriate to describe the measured BC as PM1 component in Section 3.2-3.3 and Figure 2. If not, please clarify in the section of experimental methods. Reply: Yes, the MAAP measurement was for PM2.5. The purpose was to use the MAAP BC result to give a more comprehensive composition of PM1, which certainly can lead to overestimation of BC in PM1. We have clarified this point to the readers by adding the following sentences in the text. In section 2.2, we have revised the sentences to "For a better mass closure measurement of fine particles in the campaign, a Multi-Angle Absorption Photometer (MAAP, Model 5012, Thermo) coupled with a PM2.5 cyclone was used for simultaneous measurement of refractory black carbon (BC), which cannot be detected by HR-ToF-AMS." In section 3.1, we have revised the sentences to "Ammonium, nitrate, BC, and chloride accounted for the rest 14.0, 10.7, 6.7, and 1.1% of the PM1 mass, respectively. It should be noted in this paper that the BC mass fraction in PM1 is likely overestimated because BC was measured for PM2.5 by MAAP in the campaign."

8. It is necessary to make Figures 2, 3 and 7 with higher resolution. Some axis labels and legends are not clearly shown. The wind speed data is totally covered by the wind direction data in Figure 2. Reply: Figures 2, 3, and 7 have been improved accordingly.

Interactive comment on Atmos. Chem. Phys. Discuss., 10, 25841, 2010.

ACPD

10, C12307–C12311, 2011

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