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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Characterization of submicron particles by Time-of-Flight Aerosol Chemical Speciation Monitor (ToF-ACSM) during wintertime: aerosol composition, sources and chemical processes in Guangzhou, China" *by* Junchen Guo et al.

Anonymous Referee #1

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This manuscript investigated the aerosol composition and sources in Guangzhou, China, with a focus on ACSM measurements and subsequent PMF analysis. Routine analysis and plots were made. The conclusions are solid, but not exciting. However, latter part of the manuscript brought advance to the knowledge base by investigating the SOA sources and formation mechanisms from the perspective of RO2 chemistry. It is found that SOA has moderate correlation with RO2* in non-pollution days, but not in polluted episodes. Some conclusions are inferred by this analysis and I will discuss

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Discussion paper

more later in the comment. This analysis distinguishes this manuscript from previous studies. I would like note that this type of novel analysis is missing from most of previous studies in China that are based on AMS+PMF analysis. Overall, I recommend publication with major revisions.

Major Comment Even though the analysis on the relationship between SOA and RO2 is novel, some discussions can be improved and I hope the authors will consider the following comments to make the manuscript hopefully more impactful. The major issue in this analysis is that SOA and RO2 have dramatically different lifetime (days vs seconds). From this point of view, I find the moderate correlation between SOA and RO2 in figure 10a and b intriguing, but hard to explain. My hypothesis for this correlation is that both SOA and RO2 are controlled by the amount of oxidants available. Another hypothesis is that they have similar diurnal variation (as shown in figure 5). It would be helpful to de-trend the SOA and RO2 and then make the correlation analysis. For example, correlating SOA and RO2 for data points at the same hour of day. In addition, the lifetime issue is especially important in polluted days when the air is more stagnant, SOA lingers for a long time, but RO2 lifetime is short due to enhanced NOx concentration. This is likely the main reason for the lack of correlation between SOA and RO2 in polluted days (i.e., figure 10c and d, Page 17 Line 495). Overall, the different lifetimes of SOA and RO2 should be kept in mind when interpreting any results. Correlation between RO2 and O3. Assume RO2 is at steady-state, then we can approximate d[RO2]/dt=k[O3][VOC]-k[RO2][NO]=0 [RO2]âĹİ ([O3][VOC])/([NO]) If the [VOC]/[NO] ratio doesn't change much, a correlation between RO2 and O3 may be expected. The equation here is over-simplified, but my point is that many discussions may start from or be explained by this type of simple mathematical derivation. In figure S4, the correlation between RO2 and O3 for the whole campaign is shown. I suggest the authors also categorize all data points into sub-groups (non-pollution vs polluted, day vs night, as figure 10) and show correlation relationship. The reason I suggest this is that based on figure 10c, polluted days only account for a small fraction of all data points. Thus, the relationship between RO2 and O3 for polluted days are

not clearly shown in the assembly of all data points. Figure 11 and Page 17 Line 505. By eyeballing, the correlation between RO2 and SOA each sub-group is very week. The p-value for linear regression must be included. Page 2 Line 62 and Page 17 Line 518. It is not clear why the intercept represents the extent of other SOA formation mechanisms. I think the related conclusions are overblown. Page 17 Line 513: RO2 concentration can not represent the amount of gas phase oxidation products. Figure 8. Please include all data points in this plot, in addition to the binned-average. Figure 10 and 11 and S4. Please use orthogonal linear regression, which considers the measurement uncertainty in both x- and y-axis.

Minor Comments: Page 4 Line 102: I believe the authors mean “inevitable”, instead of “evitable”. Page 6 Line 175: Even though the measurement details of RO2* have been described in previous studies, it is still beneficial to briefly mention how the measurements were done. Page 9 Line 254: The selection of a-value should be justified. Page 9 Line 274: Even in non-pollution period, rush hour peaks in HOA are expected. Thus, the lack of diurnal variation in HOA in this study is alarming. It may be due to the low resolution of ACSM. Page 10 Line 307: Replace “high volatility” with “semi-volatile”. Page 16 Line 490: the trend between f44 and SOA concentration is not clear in figure 10. Regarding the claimed conclusion that f44 of SOA decreases with increase of SOA concentration, I suggest the authors to check if there is any “contamination” in PMF analysis. In other words, in polluted days, is some POA apportioned into SOA by PMF analysis? This can be done by either run PMF on non-pollution and polluted days separately and see if the fraction of SOA changed, or check the residual of POA characteristic ions during polluted days. The former approach is more reliable.

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