

Reviewer #1

General comments:

The Pearl River Delta (PRD) region is an important area for air quality research. This study provides a year-long observation of aerosol species using aerosol mass spectrometry. The paper's focus is given to secondary organic aerosols which account for about a quarter of submicron particle mass concentration. In general, the paper is well organized and clearly written. Presenting such an extensive observational dataset itself serves as a contribution to the atmospheric chemistry community. I only have a few minor comments on the current paper.

Responses: We thank the reviewer for acknowledging our efforts and for all the valuable comments.

Minor comments:

Comment: On the Q-ACSM data analysis: Previous studies used to have different categories for SOAs. For example, some studies have separated an aq-SOA (aqueous-processing SOA) component. Can the authors provide more information on the assumption and reasons for their choice of SOA categories?

Response: We learned from this comment. We did not find any literatures have resolved aqSOA factor on the basis of Q-ACSM measurements. However, we do find the aqSOA factor was resolved from ToF-ACSM measurements which differ with Q-ACSM mainly in resolution of spectrometer, and the aqSOA was characterized by high fraction of m/z 29 (CHO^+) and high correlations with sulfate, for example the square of correlation coefficient between sulfate and aq-SOA reached as high as 0.94 in Lei et al. (2021). The square of correlation coefficients between both LOOA and MOOA with sulfate in this study is below 0.5, thus do not support directly if they are related with aqueous phase reactions. To make readers aware of this, we added following discussions in Sect. 2.2: “Note that a aqSOA factor (called aqOOA in these references) was previously resolved using the aerosol mass spectrometer measurements (Sun et al., 2016; Zhao et al., 2019) or time-of flight ACSM measurements (Lei et al., 2021), and the factor was resolved as aqSOA because of its high fraction of m/z 29 (CHO^+) and high correlation with sulfate.

Both two resolved SOA factors in this study showed relatively weak correlations with sulfate (Fig.1), do not support directly if they are related with aqueous phase reactions.”

Comment: Fig.3 & Fig.5: Earliest studies reported fast formation of sulfate at haze episodes, which seems to be different from the measurements shown here. Can the authors show the reasons?

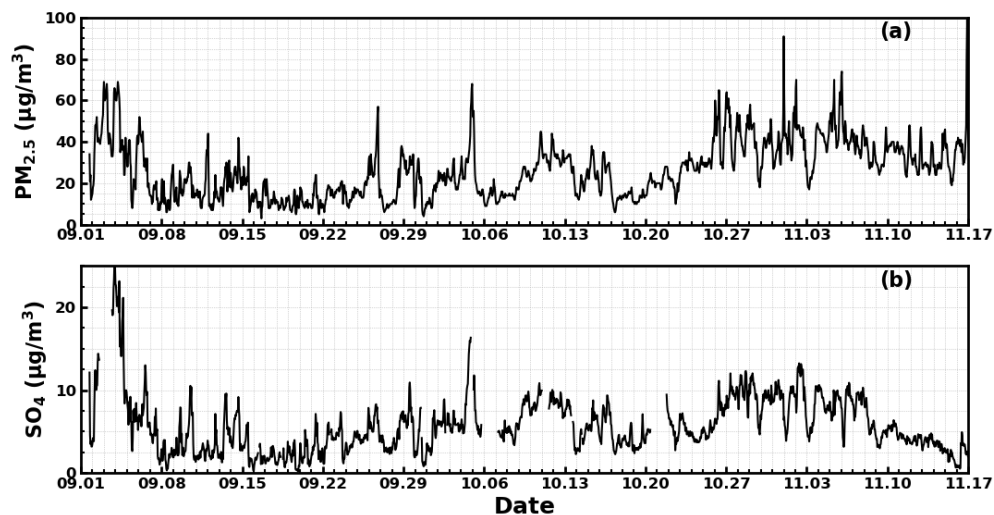


Figure 1. Time series of PM_{2.5} (a) and sulfate (SO₄) (b) during autumn of 2020.

Response: Thanks for your comment. Sulfate is not the focus of this study, however, this is quite an interesting question and made us noticed that the sulfate evolution during the observations of our study differed much with results shown in previous studies. First, results shown in Fig.3 and Fig.5 do not reflect the formation rate of sulfate. Sulfate contribute most to submicron aerosol mass during haze episodes of autumn, therefore the time series of sulfate and PM_{2.5} are shown in Fig.1 as an example. It can be seen that both PM_{2.5} and sulfate rarely show accumulation characteristics that last several days, and shown strong diurnal variations, and fast sulfate formation can be observed in daily scale. This is quite different with results shown in Chen et al. (2021) as shown

in Fig.2, and the results shown in Chen et al. (2021) was observed also at Guangzhou urban area in autumn of 2018. During the observations of Chen et al. (2021), the continuous accumulation of sulfate in some haze episodes is remarkable and totally different with those in observations of this study. Results of these studies highlight the seasonal variations of aerosol chemical compositions might differ much among years. And the reasons behind this might be related with the changes of meteorological conditions and emission conditions. And for the difference between observations of Chen et al. (2021) with this study, the changes of meteorological conditions might play the dominant role, because relative small emissions changes might be expected from 2018 to 2020. To make this point clear to readers, the following discussions was added the Sect 3.2 of the revised manuscript:

“Note that seasonal variations of aerosol chemical compositions might differ much among years due to different meteorological conditions and emissions. For example, the evolution of sulfate during autumn in this study (Fig.S9) have remarkably different

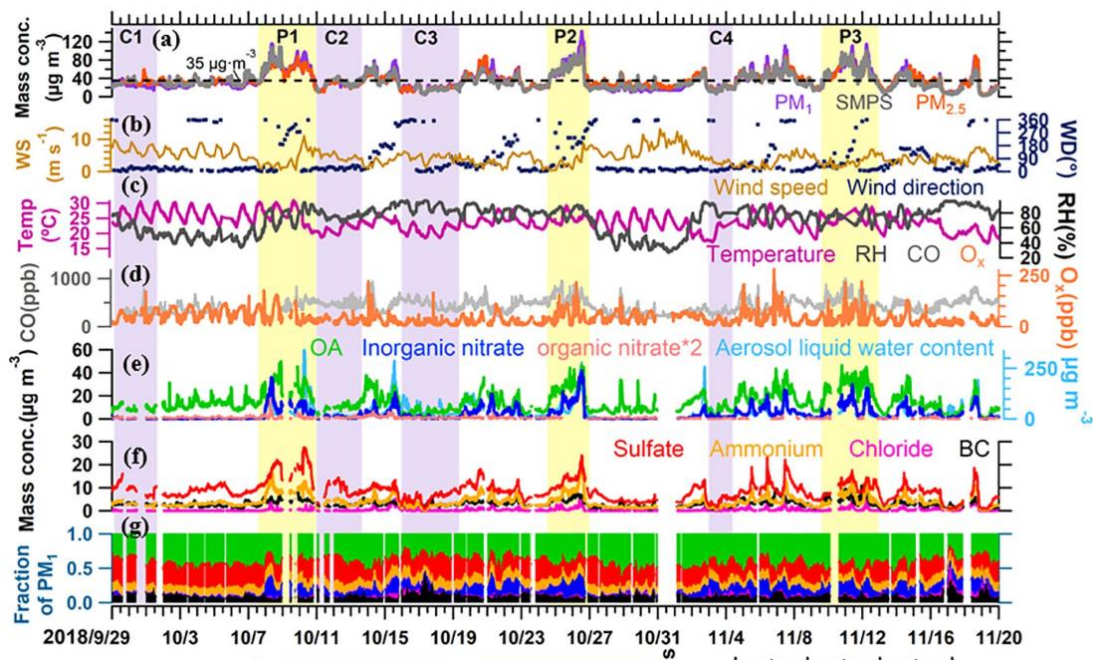


Figure 2. Time series of different parameters in Chen et al. (2021).

accumulation characteristics with those observed in autumn of 2018 as shown in Fig.1 of Chen et al. (2021a). Even so, SOA play significant roles in haze formations of Guangzhou urban area in all seasons hold based on results of existing literatures (Zhou

et al., 2020b).”

In addition, sulfate formation mechanism is a hot spot in recent years, however, most investigations on sulfate formations mechanism were done on the North China Plain. Synthesized research on the formation mechanisms of sulfate in the PRD region remain lacking, and might be one of our focuses in next years.

Comment: The variations of boundary layer height may affect the interpretation of the formation mechanisms of aerosol species. Can the authors provide more quantitative results on its influence?

Response: We agree with the reviewer that the variations of boundary layer height might affect significantly on the interpretation of the formation mechanisms, however, the observations of parameters that relate to planetary boundary layer (PBL) height evolutions is lacking. Reanalysis data of PBL might be available, however, my experiences told me that climatological analysis using reanalysis datasets might be ok, however not accurate enough for resolving hourly scale processes such as SOA formation within several hours. Therefore, in the data analysis of SOA formation, we only tried to qualitatively interpret the results and give clues on future studies. We thought about using CO to scale the observations as done in other studies, however, we found that only small decrease was observed in CO although other parameter such as POA is decreasing substantially, demonstrating that the CO variations reflect poorly PBL evolutions in strong CO source regions.

Chen, W., Ye, Y., Hu, W., Zhou, H., Pan, T., Wang, Y., Song, W., Song, Q., Ye, C., Wang, C., Wang, B., Huang, S., Yuan, B., Zhu, M., Lian, X., Zhang, G., Bi, X., Jiang, F., Liu, J., Canonaco, F., Prevot, A. S. H., Shao, M., and Wang, X.: Real-Time Characterization of Aerosol Compositions, Sources, and Aging Processes in Guangzhou During PRIDE-GBA 2018 Campaign, *Journal of Geophysical Research: Atmospheres*, 126, e2021JD035114, <https://doi.org/10.1029/2021JD035114>, 2021.

Lei, L., Sun, Y., Ouyang, B., Qiu, Y., Xie, C., Tang, G., Zhou, W., He, Y., Wang, Q., Cheng, X., Fu, P., and Wang, Z.: Vertical Distributions of Primary and Secondary Aerosols in Urban Boundary Layer: Insights into Sources, Chemistry, and Interaction with Meteorology, *Environmental science & technology*, 55, 4542-4552, 10.1021/acs.est.1c00479, 2021.