Comment on acp-2021-1069
Anonymous Referee #3

Referee comment on "Molecular Characteristics, Sources, and Formation Pathways of Organosulfur Compounds in Ambient Aerosol in Guangzhou, South China" by Hongxing Jiang et al., Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2021-1069-RC3, 2022

In this work, the authors identified the key factors (e.g. relative humidity, oxidation of biogenic volatile organic compounds via ozonolysis, and NOx-related nitrooxy organosulfateformations) and processes (e.g. heterogeneous secondary reactions) for the molecular variation of OrgSs in Guangzhou. The paper is well written. The results are clearly presented and discussed. The data analysis is solid and sound. I have a few minor comments/suggestion for authors' consideration.

line 105, "A total of 55 PM2.5 samples were used for negative ESI-FT-ICR MS analysis and each sample was ultrasonic extracted with methanol in a cold-water bath (Jiang et al., 2021a)." What would be the extraction efficiency of OSs with methanol in a cold water bath? Would there be any potential artifacts when the samples were extracted with methanol?

line 122, "We assume that the different OSs may have similar ionization efficiency (Bateman et al., 2012), because the sulfate functional group are readily ionized during the ESI process (Lin et al., 2012). However, the ionization efficiencies may vary among different OSs compounds for other reasons, such as surface activity on ESI droplets (Kuang et al., 2016)." Can the authors elaborate what would be the uncertainties in determining the concentrations if we assume different OSs have the same or similar ionization efficiency?

line 155, "Our estimates of OrgSs mass to organic matter mass are in the range of 0-30%, which are comparable to the 30% observed in PM10 organic mass over Hungary (Surratt et al., 2008), and in the range of 5-50% estimated in several sites for fine
particulates." Can the author elaborate how they obtain these numbers?

line 254, "These results probably show that although combustion sources can emit numbers of OrgSs, the low abundance of primary low-oxidative and aromatic OrgSs in ambient samples but abundant in source samples probably suggested that the OrgSs in Guangzhou suffered little or indirect influence from primary emissions (e.g., secondary formation via the combustion-emitted precursors)." Can the authors elaborate why the primary aromatic OrgSs are low oxidative? What the atmospheric stabilities of other OSs?

line 385, "We noted that RH is an important driver associated with the seasonal distribution of OrgSs composition, as RH and temperature are clustered at the negative end of the first dimension," In addition to aerosol composition and environmental and meteorological factors, would the physical state of the aerosols (e.g. solid, liquid, semi-solid, liquid-liquid phase separation) affect the formation and transformation of OSs?