

We thank the Dr. Lance for the helpful comment on our manuscript regarding the conclusion section. In the revise manuscript, we have substantially strengthened this section as suggested in ACP guidelines for authors as follows. The revised parts of the manuscript are in blue.

“AqSOA molecular composition and processing in cloud episodes were studied using online molecular information obtained by EESI-ToF-MS at a high-mountain site in China. Cloud processing substantially influences OA composition, resulting in large differences among distinct cloud episodes. Organics in cloud droplets had an average molecular formula C_{9.95–12.92}H_{14.53–21.78}O_{5.15–6.02}N_{0.32–0.42}S_{0–0.01}Si_{0–1.29} for the selected four cloud episodes. CHO compounds contributed predominantly to OA in cloud droplets. CHON was enhanced markedly in cloud droplets compared with cloud-free aerosol particles and interstitial aerosol particles in most cloud episodes. The majority of CHON compounds were likely organonitrates, highlighting the enrichment of organonitrates compounds in cloud processing. OA in cloud droplets contained higher numbers of C, O, and N atoms, exhibited a CH₂-based homologous series, and showed an enrichment of higher-molecular-weight compounds compared with adjacent cloud-free aerosol particles, collectively highlighting the importance of accretion reactions in cloud processing of OA at the molecular level. We identified several compounds significantly enriched in cloud droplets, including typical aqSOA tracers such as oxalic acid. The new aqSOA tracers, such as C₆H₁₂O₆ and C₉H₂₂N₂O₄, could help future studies identify cloud processing aqSOA.

This study provides direct molecular-level evidence for the contribution of accretion reactions during cloud processing of OA. Although previous cloud observations using FT-ICR-MS reported the presence of oligomers in cloud samples, these studies could not distinguish whether such compounds originated from cloud processing or aqueous aerosols, as no concomitant aerosol samples were collected for comparison (Zhao et al., 2013; Cook et al., 2017). By directly comparing OA composition in cloud droplets with that in cloud-free aerosol particles, our results clearly demonstrate that accretion reactions occur within cloud droplets. It has been assumed that HMWC are predominantly formed in aerosol liquid water rather than cloud water, owing to the lower reaction rates of accretion reactions in the more dilute cloud-water environment (Ervens et al., 2011). In contrast, our study provides direct molecular-level evidence that such compounds can also be formed in cloud water, extending earlier observations by Cook et al. (2017). These findings highlight that accretion reactions should be considered when modeling aqSOA formation in clouds.

The HMWC formed via accretion reaction may have implications for the environment and climate. Due to the increase in the HMWC, accretion reactions likely reduce the volatility of organics and could potentially enhance OA mass concentration and alter the aerosol size distribution after cloud evaporation. The formation of HMWC can also modify physicochemical properties, such as lifetime, oxidation state, viscosity, and hygroscopic properties, which may further influence the cloud

30 activation of these aerosols. In addition, the formation of N-containing compounds in cloud droplets, such as organonitrates, pyrrole, and imidazole, may also affect the physicochemical properties of aqSOA, e.g., contributing to brown carbon and thus 35 affecting regional radiative forcing.

Based on the measurement of high time resolution (~20 s), we find that the concentrations of individual organic compounds were highly dynamic in clouds, which is likely due to the turbulence in clouds. Such a highly dynamic nature in 40 clouds poses difficulties in extracting the influence of chemical processes on individual compounds for instrumentation with low temporal resolution. Therefore, our results highlight the necessity of high time resolution measurements (< 1 h), especially online systems achieving minute-level resolution to investigate the chemical processes in clouds, considering dynamic variations of compounds in clouds due to turbulence in clouds and alterations in air masses.

It should be noted that this study provides molecular formulas only, while detailed structural information is warranted to 45 better constrain the sources, formation mechanisms, and climate impacts of aqSOA in clouds. In addition, sources of compounds enriched in cloud droplets will be investigated in future studies.”