Comment from Referee #1:

Interactive comment on "Impact of in-cloud aqueous processes on the chemical compositions and morphology of individual atmospheric aerosols" by Yuzhen Fu et al. Anonymous Referee #1 Received and published: 26 May 2020

In this paper, the authors did a good job of presenting their results on the different morphologies and mixing states of activated and interstitial particles. I think that the analysis could be strengthened by further exploring the connection between the offline TEM-EDS analysis and the online SPAMS analysis.

We would like to thank the referee for the positive and valuable comments to improve our manuscript. We agree with the comments and have strengthened the analysis, in particular, the connection between the TEM/EDS and the SPAMS results. As suggested, the size distribution and mass spectral information were included to support the discussion. We have addressed the specific comments in the sections below and made the appropriate revisions to the manuscript. The referee's comments are in the black text followed by our response in the blue text.

As Prof. Surratt mentioned, it has been previously shown that SOA can form via in-cloud processes and the formation of SOA can result in core-shell phase morphology. As such, it would be beneficial to assess the SPAMS mass spectra for SOA products, especially given the increase in the number fraction of S-OM relative to S-rich particles in the RES v. INT particles.

Thanks for your comment. Indeed, we also hope to obtain more information about OM from SPAMS to explain and corroborate the results obtained from TEM/EDS. In the section 3.3, we use the ratios of relative peak area of organics to sulfate of OM particles during in-cloud (RES and INT) and pre-cloud (Ambient) periods from the data of SPAMS (Table S2), to help explain the in-cloud formation of OM found by TEM/EDS, which show that the ratios of relative peak area between organics and sulfate are similar

between the INT and particles before cloud event, whereas they are higher in the RES.

Additionally, the size distribution of different particle morphologies and mixing states could be assessed and compared to the size-resolved mass spectra from the SPAMS.

Thanks for your comment. We have added the size-resolved number fraction distributions of the RES and INT by TEM/EDS (Figure S8) and SPAMS (Figure S9). We also added simple description and comparison in the SI:

The size distribution of different particle types revealed that S-rich and aged soot particles were predominant in smaller size segments, and aged mixture particles in larger size segments (Figure S8). Likewise, the size-resolved number fractions of different particle types from the results of the SPAMS also showed that the BC-containing particles were mainly distributed between 0.1 and 1.3 μm, representing ~80% of the submicron RES and ~73% of the submicron INT population, respectively (Figure S9).

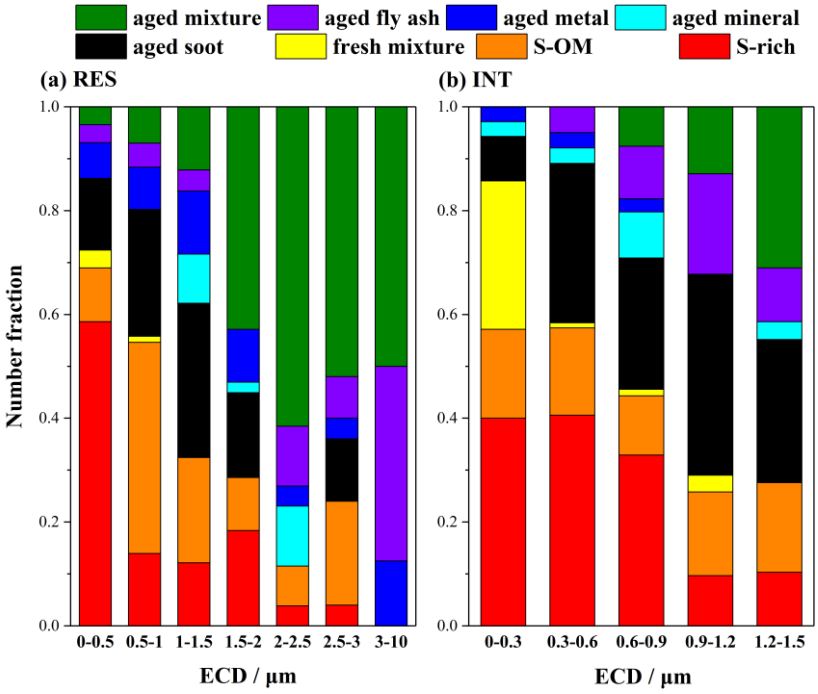

Figure S8. Size-resolved number fraction distributions of RES and INT by TEM/EDS.

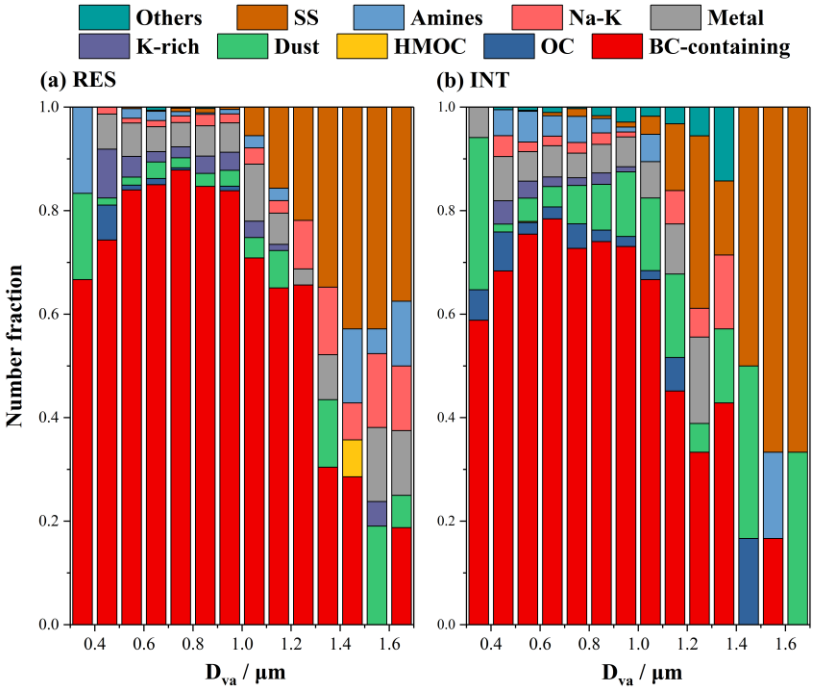

Figure S9. Size-resolved number fraction distributions of RES and INT by SPAMS.

Additionally, I would recommend that the authors include the EDS spectra that correspond to the TEM images presented in Figure 3.

Thanks for your suggestion. We have added the EDS spectra of the OM-containing particles corresponding to each TEM image in Figure 3 (that is Figure 5 now):

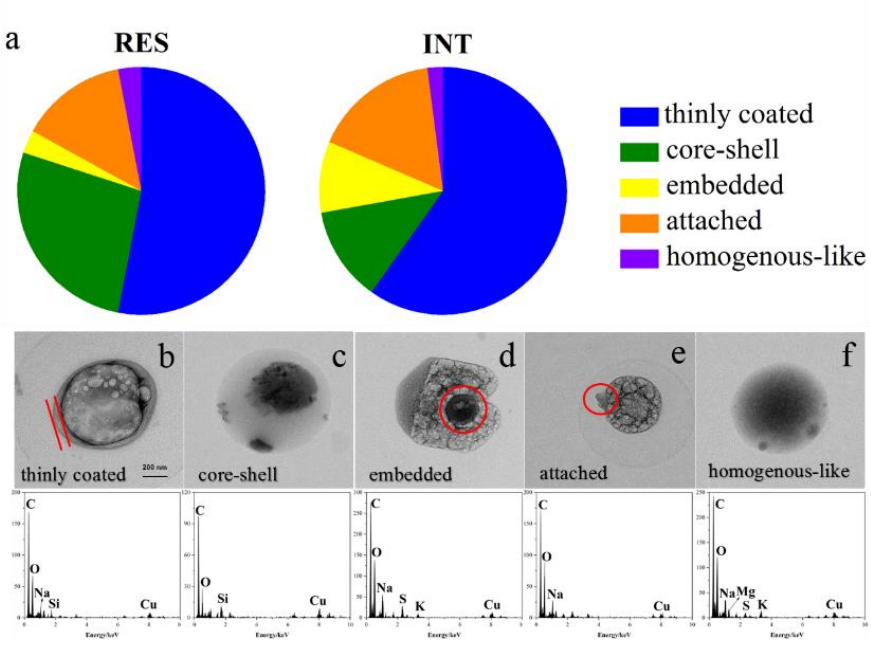

Figure 5. Number fractions of the OM-containing particles with different mixing structures in the RES and INT (a) and typical TEM images and corresponding EDS spectra of OM: thinly coated (b); core-shell (c); embedded (d); attached (e); homogenous-like (f) during cloud event #2 and #3.

In the SI, to clarify the difference between figures S1 and S3, I would recommend that the authors change the title of S1 to indicate that the fraction types correspond to EDS.

Thanks for pointing out this. We have changed the title of Figure S1 (that is Figure 3 now) from “Number fraction of different particle types in the RES during three cloud events.” to “Number fraction of different particle types in the RES during three cloud events measured by TEM/EDS.”.