Comment on acp-2021-704
Anonymous Referee #1

Referee comment on "Offline analysis of the chemical composition and hygroscopicity of sub-micrometer aerosol at an Asian outflow receptor site and comparison with online measurements" by Yange Deng et al., Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2021-704-RC1, 2021

This manuscript presents aerosol chemical composition and hygroscopicity from offline measurements and comparison with online analysis in Okinawa based on a combination of instruments. The RH-dependent hygroscopicity of sub-micrometer aerosols and their chemical components are investigated. The measurements and data have been made carefully and then the interesting results are presented, especially the comparison between the two methods is helpful to understand whether the offline analysis can be used as an alternative method for aerosol hygroscopicity studies.

However, I have some major concerns about the bias of the offline analysis. And it could be more cautious and thorough when interpreting some results. In addition, there are also some editing issues need that have to be addressed. The authors thus need to make a careful revision and correction to improve the overall quality of the paper for publication in the journal. I would recommend the editor to reconsider the papers only after a major revision by the authors.

Major Comments:

- Page 4, Line 30, the samples were stored in freezers after sample collection. How to ensure that compounds do not undergo some physical and chemical changes such as condensation and degradation during storage? Have you considered the offline analysis bias caused by storage?
- Page 9, the authors concluded good agreement between offline and online analyses from the high positive correlations of mass concentrations, but the $R^2$ of the mass fractions of sulfate, organics, ammonium, and EC (BC) from offline and online analysis were low. The authors should try to explain this. In addition, the average mass concentrations of sulfate, organics, and ammonium from online measurements were lower, but BC from online analysis was almost equal to those from offline analysis. What is the reason for this?
- Section 3.2, the authors should add more measured results from offline analysis and compare these with previous studies since many previous online analyses have tried to derive the hygroscopicity in a larger accumulation size above 300 nm. It is best to add
more discussions in these paragraphs.
- Page 13, Line 19, “Results obtained from … indicate that offline aerosol hygroscopicity analysis can be used as an alternative method”, but the coefficients of determination between offline and online results were low (Fig. 6), even less than 0.5. The authors should try to address this and make it convincing.
- Fig. 7, comparing the hygroscopicity parameter of WSOM and EOM between this work and previous experiments needs to be shown to be much more rigorous. Unless the authors can explicitly show that the experiment setup and estimation method were identical between the different experiments, they cannot make a like-for-like comparison between the different experiments. In addition, the RH conditions of these previous studies should be included in the figure.

Minor Comments:
- Page 5, Line 30, The residence time in the heated region should be compared with that for other systems.
- A schematic of the experimental set-up would help.
- The retrieval method of the HTDMA data should be included in section 2.2.
- Fig. 1, add the mass fractions from online analysis.
- Fig. S6, looks like one point is missing in the Fig. S6(c).
- I suggest adding more 'raw' measurement time series (e.g. in the supplementary information). For example, time series of hygroscopicity parameter over the experiment.
- Page 10, I was lost from line 7 to 11 when the aerosol concentrations and fractions are discussed. Is the discussion still based on Fig. 2 or other figures in the manuscript?