Comments on "Theoretical Framework for Measuring Cloud Effective Supersaturation Fluctuations with an Advanced Optical System" by Kuang et al.

Kuang et al. presents a theoretical framework for measuring cloud effective supersaturation fluctuations using an advanced optical system, which can improve understanding aerosol activation and cloud microphysics. The framework focuses on observing the critical activation diameter and hygroscopicity of activated aerosols through the scattering and water-induced scattering enhancement of interstitial and activated aerosols. It allows for minute- to second-level effective supersaturation measurements, capturing vital fluctuations for cloud microphysics studies. I think the manuscript, once revised to address the concerns outlined below, could be considered for publication.

Major comments:

(1) The theoretical framework introduced in this paper are mainly based on κ-Köhler theory, that is, supersaturation could be obtained with known dry diameter and hygroscopicity kappa. However, the application of κ-Köhler theory is under assumption of water surface tension and fully dissolution. Previous studies have uncovered the surface tension reduction (Gerard et al., 2016; Noziere et al., 2010; Ovadnevaite et al., 2017) and slightly soluble components (Ho et al., 2010) in atmospheric aerosol samples. So, if the apply the framework in field observation, the authors should add some discussion about the uncertainty originated from the above-mentioned assumptions.

(2) As the author mentioned in section 3.2 that the hygroscopicity parameter kappa for supersaturation prediction was $\kappa_{act,f(RH)}$ from aerosol light scattering enhancement factor $f(RH)$ by using humidified nephelometer under unsaturated condition. So, would it bring uncertainty to supersaturation prediction since there may be hygroscopicity deviations between unsaturated and supersaturated condition? Though the authors discussed a bias of 0.1 in κ only results in a 0.01% bias in supersaturation retrievals, but the retrievals supersaturation ratio was very low as it was shown in Fig 2c (the lowest value can be 0.02), so 0.01% uncertainty is comparatively large.

(3) As the author mentioned in the paper that we assumed aerosol populations remained unchanged during the 30-minute period (based on comparisons between PM1/PM2.5 and TSP inlets), which can sometimes introduce significant uncertainties in the size-

resolved AR calculations. Based on the authors observation experience, I wonder what is the frequency of the significant uncertainties' events. And 30-minute period was long and the assumption of constant aerosol populations may be not very appropriate, is there any possible improvement to decrease the time period?

(4) As the author mentioned that the supersaturation is effective ratio that make specific number or fraction of aerosol particles activated to CCN, rather than real environment supersaturation ratio. So, I am interested in how to use the "effective ratio" and detect new insight in observation or climate models. Can the author give a simple example or description about it?

Minor comments:

(5) Line 199-200: please added some description and references about how to accurately retrieve *D* by machine learning techniques

(6) References section: The format of the references is not consistent (e.g., some journal names are full but others are abbreviations)**.** Please revised carefully.

Reference

Gerard, V., Noziere, B., Baduel, C., Fine, L., Frossard, A. A., & Cohen, R. C. (2016). Anionic, Cationic, and Nonionic Surfactants in Atmospheric Aerosols from the Baltic Coast at Asko, Sweden: Implications for Cloud Droplet Activation. *Environmental Science & Technology, 50*(6), 2974-2982. https://doi.org/10.1021/acs.est.5b05809

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Noziere, B., Ekstrom, S., Alsberg, T., & Holmstrom, S. (2010). Radical-initiated formation of organosulfates and surfactants in atmospheric aerosols. *Geophysical Research Letters, 37*. https://doi.org/10.1029/2009gl041683

Ovadnevaite, J., Zuend, A., Laaksonen, A., Sanchez, K. J., Roberts, G., Ceburnis, D., Decesari, S., Rinaldi, M., Hodas, N., Facchini, M. C., Seinfeld, J. H., & O' Dowd, C. (2017). Surface Tension Prevails over Solute Effect in Organic-Influenced Cloud Droplet Activation. *Nature, 546*(7660), 637-641. https://doi.org/10.1038/nature22806