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Comment on amt-2022-188

Anonymous Referee #2

Referee comment on "A multiple-charging correction algorithm for a broad-supersaturation scanning cloud condensation nuclei (BS2-CCN) system" by Najin Kim et al., Atmos. Meas. Tech. Discuss., <https://doi.org/10.5194/amt-2022-188-RC1>, 2022

The manuscript presents a novel approach for reducing the multiple charging effects from the CCN activity measurements using a BS2-CCN system. Kim et al. (2022) discusses the application of a new probability density function for reducing multiple charging effects from measurements obtained using the BS2-CCN. A prominent advantage of the method explained in Kim et al. (2022) is that applied the charge correction is applied to number concentrations measured at each particle size. Therefore, the effect on larger multiply charged particles on the size-resolved number concentrations becomes easier to detect and eliminate.

Overall, the work is new and the method to apply multiple-charging corrections for the BS2-CCN instrumentation has not been done before. The work extends on previously published works (e.g., the kernel function is added to the Kim et al 2021 multiple charge correction) with the BS2-CCN data. In general, the main concern is that multiple factors affect CCN measurement (aerosol shape, aerosol aggregation, viscosity, volatility, solubility, surface activity) and these effects are confounded; it is difficult to isolate the effects of multiple charging alone. However, the data collected from the BS2-CCN counter and subsequent analysis will be important for understanding CCN spectra of atmospheric aerosol and thus the work warrants publication. The following questions and comments address ideas that maybe unclear to the reader in the manuscript.

- Composition of aerosols does not affect the probability of multiple charging. However, the morphology of the aerosols changes the probability of charging. Are the ambient aerosols spherical?
- Was there any contribution from other physical factors (e.g., mixing state surface activity, viscosity, non-spherical morphology) on the uncertainties in ? Did the authors take any measures to control the contribution from the aforementioned and other

sources of uncertainty in ?

- Page 4 – The authors did a good job of describing the formulation of the new kernel function for multiple charge correction in the CN and CCN number concentrations measured using the DMA 3080. Traditionally, the charge correction for the CN and CCN measurements from the DMA 3080 is done using the Weidensohler (1988) method in the SMCA. Was any significant difference observed between the number concentrations obtained from the 2 charge correction methods? On Line 79, Is charging theory -Wiedensohler 1988 applied? Some clarification on this part of the text could elucidate differences and similarities in theories applied.
- How much is the overall improvement in the size-resolved activation ratio of the aerosol compared to the traditional SMCA approach and is this difference statistically significant?
- Page 6 line 160 – The authors mention that calibration results obtained using the charge correction algorithm may be closely replicated with a minimized influence of multiply charged particles, if “experimental control is performed well”. What does that mean? Were the experimental conditions varied across different calibration procedures? What type of experimental control would be required to obtain high quality calibration without the use of charge correction algorithms?
- The authors mention that the hygroscopicity parameter was derived from the formulation given by Petters and Kreidenweis (2007). This suggests that the uncertainties in the at the point of activation (which result from there being multiply charged particles in the population corresponding to the dry activation ratio) will directly relate to the uncertainties in . Moreover, the uncertainties due to different multiply charged particles will likely have different magnitude. How do these uncertainties in the size-resolved activation ratio translate to the uncertainties in the aerosol ? Furthermore, is there any correlation between the uncertainties due to specific multiply charge particles and the charge that they carry?
- For the test on the ambient aerosols – What was the chemical composition of the ambient aerosols which were analyzed using the new algorithm? The quantified uncertainties in was helpful, however did the authors verify what proportion of these uncertainties in were due to the multiple charging problem?
- Figure 2(b) is confusing. What are the sizes of the particles that carrying doubly and triply charges? Are they the particles with the same mobility of the singly charged particles, or the probability of the size of the particles being doubly or triply charged? Are the doubly and triply charged particle sizes in the 95% of the Gaussian distribution? It is suggested to mention that fraction of doubly and triply charged particles depends on the number concentration of the larger particles.