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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Influence of the dry aerosol particle size distribution and morphology on the cloud condensation nuclei activation. An experimental and theoretical investigation" *by* Junteng Wu et al.

Anonymous Referee #1

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Wu et al. present laboratory data and theoretical modeling to treat the influence of particle morphology on CCN activation spectra. Size distribution data and the k-Kohler parameterization are combined to predict the CCN number concentration as a function of supersaturation. Fractal dimension inferred from single particle transmission electron microscopy experiments is folded into the analysis. The method is tested using ammonium sulfate and soot particles from a miniCAST burner. K-values for soot as a function of ozone exposure are presented.

This manuscript is not suitable for publication in ACP, according to the journal standard

to publish studies "with general implications for atmospheric science". The paper does not reach clear new conclusions, mostly focuses on methodology and would be more suitable for AMT. More importantly, the manuscript lacks context of a large body of published literature, needs additional analysis folding in DMA transfer function models, and will require significant rewriting to become acceptable for publication.

Major comments

Eqs. (11) and (12), "the master equations" of the work are simply a cumulative size or supersaturation distributions that have been used in similar for decades in droplet activation schemes or closure studies (e.g. Abdul-Razzak and Gahn, 2000, Snider et al., 2003 and references therein).

In general, the work herein can be interpreted either as a laboratory CCN closure experiment or an improvement to constrain laboratory inferred k-values from data. No studies in that context are cited. For example, it has long been recognized that k-values derived from CCN data are "effective" or "apparent" k-values that have folded in effects of surface tension, solubility, and particle morphology (e.g. Poschl et al., 2009, Sullivan et al., 2010). CCN closure experiments that take size distribution data, composition and shape have been performed extensively on ambient aerosol. Studies that account for particle shape in the calibration of CCN instruments using DMAs and non-spherical particles are routine (e.g. Snider et al., 2006, Rose et al., 2008, Kuwata and Kondo, 2009). Using TEM to obtain particle shape may be novel here (this referee is not 100% sure), but the concepts described in Section 2 are well established.

The "master equation" (Eq. 11) cannot be applied to DMA distributions. The role of the DMA transfer function and of multiply charged particles needs to be taken into account (e.g. Petters, 2018). It is clear that the effect of multiply charge particles is not large as shown by the shoulder in Figure 6. However, the paper makes claims about kappa being a "Dirac delta distribution", i.e. a single value applied to a distribution. The question is whether the observed activation behavior is due to size distribution alone, or whether shape and composition heterogeneity increases the broadness. Therefore, Eq. (11) should be replaced with the DMA transfer function, including multiply charged particles, and the use unexplained discrepancies to identify the effect of shape and/or heterogeneity.

The influences of size distribution heterogeneity and particle shape on the cumulative CCN spectrum have been discussed in the literature (e.g. Petters et al., 2009, Kuwata and Kondo, 2009, Su et al., 2010, Cerully et al., 2011). The work of Su et al. is particularly pertinent to this paper.

The aging studies of miniCAST soot are interesting. However, it is important to place these results in the context of several similar studies that have investigated the CCN activity of (chemically aged) soot.

The framing around improving k-Kohler or Kohler theory in general is not justified. Kohler theory predicts the activation behavior of a single particle. If one applies that theory recursively to a size distribution with non-uniform composition, or a shaped particle, then those assumption need to be questioned, as it has been in previous studies. Statements that "[f]or all practical purposes, both theories operate under the hypothesis of Dirac delta distributions" don't really make sense in that context.

It is not clear what the main finding of the work is. Equations are presented and somewhat successfully tested against experimental data, but that type of exercise has been presented before. The novelty of the work and the scientific findings contributed need to be better articulated.

The writing of the manuscript needs to be improved significantly. Words are frequently used incorrectly. The writing lacks context of important prior literature. The framing of the work needs to be revised. Section 2 reads more like a tutorial than a scientific paper.

The data availability is inconsistent with the journal policy. The data should be de-

posited in a publicly available repository.

Other comments

"The modern parametrization of the classical theory of nucleation (k-KÓğhler theory) implicitly assumes a Dirac delta distribution to model the density of ideal spherical point size dry particles and droplets."

To my understanding Kohler theory isn't equivalent to "classical nucleation theory", which usually refers to homogeneous nucleation.

The second part of the sentence isn't quite parsable. Is this talking about density profiles within a particle? Or differences in density and composition within a distribution. Given the context of the paper, it seems to be the latter. k-KÓğhler treats the activation of a single particle. It therefore does not make any assumptions about the heterogeneity of the size distribution.

"To the present day" → please rephrase

"internally mixed nature" → internally mixed refers to a distribution property (i.e. two substances may be internally mixed or externally mixed across a distribution). A single particle is either pure or mixed. k-KÓğhler theory is one means to treat the activation of mixed particles.

"partially soluble" → please change to sparingly soluble. The state of dissolution is determined by the water content. A substance may be "partially dissolved" but it has a strictly defined solubility value.

"neither the classical nor the -KÓğhler theory account for the dry aerosol particle size distribution and morphology" → this is not the objective of those theories. This is a problem on how it is applied in practice.

"more specifically, the crossing of the Kelvin limit has been reported in several instances" → Please explain what is meant by that.

"is the master equation of this work" → this is an odd formulation

"geometric deviation" → geometric standard deviation

"and the fitting is implicitly assumed not to carry any physically meaningful parameter. " → not true. See eg. Cerrully et al. 2011, Figure 3.

"within 20% incertitude" → within 20% uncertainty

"soot emissions are emblematic of human activities" → please reword

"understanding the anthropogenic impact on the atmosphere." → please reword

"understanding the anthropogenic impact on the atmosphere." → please specify what "very representative" means. Is there a metric for this?

"univocally defined" → is defined.

"Young soot"; "mature soot" → Soot aren't biological entities. "Fresh soot" and "chemically aged soot" would appear to be better terms.

"k-Köhler theory is built around the idea that the soluble fraction only affects the heterogeneous nucleation " → this is not strictly true. k-Köhler can be used to parameterize CCN activity descriptively, even if the underlying mechanism is incorrect. Such parameterizations have been termed "apparent k" or "effective k" in the literature."

"k parameterizes all the information on the composition of the droplet approximated as an ideal water solution" → this is incorrect. K is a one parameter activity coefficient model that explicitly models the non-ideality at the point of activation.

"Incidentally, the effect of the dry particle electrical charge on the activation is not considered in this work." → This is unclear what this means. As written, it suggests that particle charge itself affects activation. To my knowledge there is no evidence for such a claim. If it is meant that the effect of particle charge on selected size is not considered, this is correct and needs to be addressed (see major comment).

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