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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 #2

Received and published: 2 July 2019

Wu et al. focus on the CCN activity of particles with irregular shape through a combination of theory and experimental data. Although the study contains interesting material, I feel it cannot be published in its current form and unfortunately must recommend rejection to provide the authors enough time to prepare a modified manuscript.

Overall this study contains a very large amount of material that is not wrong, but is "textbook", often without reference to any of the large body of published literature on the subject. I strongly recommend that the authors give appropriate credit to the published work, but also keep only what is absolutely necessary. Reviewer #1 presents a few

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references, I can add also the textbooks of Seinfeld & Pandis, Pruppacher and Klett, as well as review of state-of-the art mechanistic parameterizations (e.g., from the groups of Ghan or Nenes) – which normally adopt formulations that use lognormals (e.g., Fountoukis and Nenes, 2005). There is now a rich literature based upon CCN spectral analysis to constrain hygroscopicity – considering shape factors, multiple charging and DMA transfer function effects (e.g., Cerully et al., 2011 and references therein) – even for well established calibration aerosol like sodium chloride and ammonium sulfate.

What I suggest is that the authors consider a resubmission, focusing on the results of the soot activation experiments, and also comparing them to existing literature. In their analysis it is nice that shape effects are considered, and even nicer that the fractal dimension is explicitly determined, because it allows then to explore other theories of activation such as adsorption-activation theory (e.g., Kumar et al., 2009; Laaksonen et al., 2016 and references therein) – alone or in combination with Kohler theory. It would be interesting to see how this other framework performs and if it can provide insight on the drivers of CCN activity for the particles studied.

The study also suffers from limitations that include the important DMA transfer function effects and multiple charging (although noted on Page 17 by the authors, they do not go through more in-depth analysis), which is considered routine in established groups.

Below some specific comments on aspects of the paper that also should be addressed: Page 2, Line 6: k-Kohler is not just for partially soluble particles, it's to address chemical complexity (mixtures of solutes of a wide range of molar masses) – and can account for partial solubility as well.

Page 2, Line 9: Kohler theory is formulate for a particle. It can (and is) extended to a size distribution easily, as mentioned in the above texts. I don't understand why the authors present this as an issue.

Page 2, Line 10: Sure, you can use - and is even elegant - to use Dirac functions to represent many particles of a given size as a size distribution. I understand where

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the authors come from when they say that Kohler theory cannot be applied to a broad size distribution - but I also find it quite narrow because it is equivalent to saying that a size distribution with a finite particle width cannot be described with a monodisperse particle distribution (which is sort of an obvious statement)! On the other hand, Kohler theory can be applied to size distributions, even in the narrowest sense considered by the authors, because it defines the "size" above which all becomes droplets (this of course, assuming the soluble fraction is sufficient, with all that this implies). In this sense, a characteristic size is linked to a characteristic saturation - which is the basis of Kohler theory and the concept of a critical point that comes out of it.

Page 2, Line 10: True, but then you need to use a theory that does not assume at least perfectly wettable particles. The work of Gorbunov, or adsorption activation theory can easily treat such situations – and is physically based. It would be nice if the authors actually refer to that work and consider it in their analysis.

Figures: uncertainties are sometimes included in the activation plots, but they are not propagated later on in any of the analysis. This would be nice to see – and also include uncertainty from the DMA transfer function, shape factor, multiple charges, etc.

Page 19, line 30: I don't understand why nanoparticles will not be in equilibrium. The relevant timescale is extremely small – unless if I misunderstood the point raised by the authors.

References Cerully, K.M., Raatikainen, T., Lance, S., Tkacik, D., Tiitta, P., Petaja, T., Ehn, M., Kulmala, M., Worsnop, D.R., Laaksonen, A., Smith, J.N. and A. Nenes (2011) Aerosol Hygroscopicity and CCN Activation Kinetics in a Boreal Forest Environment during the 2007 EUCAARI Campaign, Atmos.Chem.Phys., 11, 12369-12386

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