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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Cloud condensation nucleus activity comparison of dry- and wet-generated mineral dust aerosol: the significance of soluble material" *by* S. Garimella et al.

Anonymous Referee #1

Received and published: 5 January 2014

General Comments for:

"Cloud condensation nucleus activity comparison of dry- and wet-generated mineral dust aerosol: the significance of soluble material" S. Garimella, Y.-w. Huang, J. S. Seewald, and D. J. Cziczo.

This paper evaluates key issues in the measurement and evaluation of the CCN activity of mineral aerosols. The argument is made that production of aerosols from aqueous slurries of mineral dust causes redistribution of soluble material and produces a size-

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mode of particles that is an artifact of the production method and not representative of atmospheric processes. The determination of the effective size and shape of the non-spherical mineral dust particles, a key parameter in CCN activity parameterizations, is also addressed. When particle size distributions have sharp drop-offs or only the tail of the distribution is measured, multiple-charging corrections are shown to have limited reliability. This was evident from electron microscopy, which is a major addition to previous work on this topic. Both Frenkel, Halsey, and Hill (FHH) adsorption activation theory and κ-Köhler agree in the size range of reliable measurements and likely atmospheric relevance, Dp >300nm, so κ-Köhler theory is shown to be suitable for even low-solubility materials such as mineral dust particles. Methods and results are well described and supported by thoroughly presented data. This manuscript presents valuable insight and is suitable for publication in ACP, pending response to the comments below.

The general issues associated with CCN activation measurements (effects of the shape of the size distribution on charge corrections, wet-dry generation) are clearly noted in the paper, but the broader implications as to previous measurements and future measurements alike could be more directly stated (broader implications if sub 300nm dust particles are not atmospherically relevant and/or cannot be correctly sampled or accounted for in measurements).

Specific Comments:

Please add to the text or to supplementary material a figure such as Fig. 3 from Kumar et al. (2011a) (activated fraction vs. dry diameter with sigmoidal fit) to more directly show the effects of charge and/or shape correction on the determination of the critical diameter.

31042:14-16 "κ-Köhler is a suitable framework less complex than FHH theory, to describe clay mineral nucleation activity despite apparent differences in with respect to size." This sentence must more clearly reflect its basis on the $\sim$300nm threshold above

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which the two approaches are known to give similar results. It seems the intent is to also suggest that if the proper corrections could be applied, then κ-Köhler theory would be suitable down to lower sizes, but this is not readily apparent.

Section 3.3.1 The application of charge correction is fairly standard, though this work, through electron microscopy results, shows limitations based on the shape of the particle size distribution. References to Moore et al. 2010, Kumar 2011a, Kumar 2011b would be appropriate, and if there are differences in the method of calculation, they should be noted.

31057:22-23 “Using cyclone impaction efficiently removes the larger particles before they enter the DMA”. This would more accurately be stated as: “Using cyclone impaction efficiently removes the larger particles before applying a Boltzman charge distribution.”

31058:4-5 “There is agreement in the charge- and shape-corrected activation results in this study and those found in Kumar et al. (2011a) and Kumar et al. (2011b) (Figs. 4 and 9).” This agreement is fairly clear in Fig. 4, but less so in Fig. 9, particularly for dry ATD. The fact that the data at Dp < 300nm is suggested to be questionable and most of the Kumar data is in this region makes the issue of agreement (both lying along the same line of constant κ) somewhat murky. It seems that the general sentiment of this paper is in disagreement with these previous results in terms of relevance (due to the size-range), and stating agreement, without conditions, blurs the arguments you seem to be making. This should be more clearly addressed. It might help to more directly compare results if FHH fits were made for your data, though that would involve fitting to data in the size region unlikely to be correctly sized, as displayed in Fig. 6.

31058:5-7 “The exception is an inability to reproduce the Kumar et al. (2011b) 100 % activation of the larger mode of wet-generated ATD.” Please clarify this statement, because this detail is not mentioned previously in the text.

31059:26-27 “This change in nucleation behavior using wet-generation is not observed

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when the dust is simply wetted and re-dried.“ This observation seems significant for interpretation of the conclusion regarding wet-generation of dust aerosols, because it speaks to the issue of atmospheric relevance in terms of the humidity-exposure during a particle’s lifetime. It should be expanded upon earlier in the text. The data supporting this statement could be added as supplementary material.

Fig 4: Caption. "n-lets" should be specifically mentioned and defined in the caption. Fig 4: Caption. “results form this study” should read: results from this study

Fig 6. It would be much clearer to have the ordering of elements the same for between the left (dry) and wet (right) columns. Then the “additional” elements found in the wet generation case would be segregated to the right side, and a clear line could be drawn to separate the consistent and “additional” elements. Trying to see the different elemental compositions from reading the bar labels makes the differences less obvious.

Fig. 5: Caption. Please make the caption clearer, such as: “Results from ion chromatography analysis for filtered supernatants of ground (magenta) and unground (black) samples of the three mineral dusts. In bottom right, unfiltered (black) and filtered (green) DI water control.” Fig. 5: Caption. “The filtered slurry supernatants show higher concentrations of soluble material.” It seems this refers to the control DI water, since all the mineral slurries were filtered. Please clarify this statement.

Fig 6: Caption. “(red boxes on x- axis)” It does not appear that the red boxes represent the additional elements but rather the 400nm particle results.

References:

Moore, R., Nenes, A., and Medina, J.: Scanning Mobility CCN Analysis – A method for fast measurements of size resolved CCN distributions and activation kinetics, Aerosol Sci. Tech., 44, 861–871, 2010.

Interactive comment on Atmos. Chem. Phys. Discuss., 13, 31041, 2013.