

Short Comment submitted by Dr. Prashant Kumar to Atmospheric Chemistry and Physics on January 21, 2014.
Manuscript commented: Garimella, S., Huang, Y.-w., Seewald, J. S., and Cziczo, D. J.: Cloud condensation nucleus activity comparison of dry- and wet-generated mineral dust aerosol: the significance of soluble material, Atmos. Chem. Phys. Discuss., 13, 31041-31078, doi:10.5194/acpd-13-31041-2013, 2013.

This study investigates the Particle Size Distribution (PSD) and Cloud Condensation Nuclei (CCN) activity of dust generated from Arizona Test Dust (ATD), and naturally occurring Illite and Sodium Rich Montmorillonite (NaMon) mineral samples. Wet and dry generated samples are considered; in the latter, a new method (that differs from e.g., Herich et al., 2009; Kumar et al., 2011a) is used. The study also investigates the presence of soluble ions present by use of Ion Chromatography (IC) and Particle Analysis by Laser Mass Spectrometry (PALMS), while elemental analysis (using Energy-Dispersive X-ray or EDX) complements the measurements. The authors compare their dry-generated results against dry-generated CCN activity results of Kumar et al. (2011a) and Herich et al. (2009). In their analysis, charge and shape corrections are also considered. The conclusions and discussions focus on the ability of κ -Köhler Theory (KT) or Frenkel, Halsey and Hill Adsorption Activation theory (FHH-AT) to describe the mineral aerosol CCN activity.

This is a largely well-written study with a diverse set of measurements. I commend the authors for their efforts. In reading the manuscript however, a number of issues with the analysis, discussion, and conclusions were identified that require extensive revision. I focus exclusively on the dry generation method and its results, as the authors show what previous studies have also shown that wet generation for mineral aerosol introduces biases in measurements of dust/mineral CCN activity.

General comments

- The main conclusion of the study is that κ -KT is preferred over FHH-AT, especially since the former is deemed “simpler”. The suggestion seems at first logical and practical, given that κ -KT is a standard approach to parameterize CCN activity. The important consequences of adopting fundamentally different activation physics for mineral dust in calculations of cloud droplet number (CDN) are however overlooked.

FHH-AT particles requires little water uptake to reach the critical diameter, compared to KT particles with the same critical supersaturation (Kumar et al., 2009b). This means KT particles can deplete considerably more water vapor than FHH-AT particles in cloudy updrafts (even if the both theories are applied so that the CCN spectra computed are identical). This can affect the maximum supersaturation that develops (i.e., cloud droplet number that forms) and the sensitivity of CDN to aerosol variations, as demonstrated by Kumar et al. 2009b with parcel simulations. The disproportionately strong impact of large particles (following KT) on CDN and the associated Aerosol Indirect Effect was shown in the global modeling study of Morales and Nenes (2013); these strong impacts virtually disappear when FHH-AT is applied to dust aerosol.

Therefore, using KT to parameterize CCN activity of dust/mineral aerosol can get the CCN number correctly, but will by design bias the CDN and its sensitivity to aerosol, and hence the Aerosol Indirect Effect in regions of freshly emitted dust. I strongly recommend the authors to revise their manuscript to caution the readership of these issues.

- κ -KT is not “simpler” than FHH-AT. Both follow a simple power law of s_c vs. D_{dry} (with different exponent). I can argue they exhibit the same degree of complexity when applied to dust: κ -KT requires two parameters,

the kappa of the soluble fraction, and, its virtual variability with size; FHH-AT requires A_{FHH} and B_{FHH} (which can be approximated with one set of values). Given this and that a CDN parameterization to account for the correct theory has been developed and used in GCMs (and available to the community for general use) for dust aerosol, I strongly recommend the authors to revise these statements accordingly.

- Often in the text, κ -KT is deemed a better framework than FHH-AT (e.g., Page 31058, Line 19-24, “Since both...value of κ ”). These statements are not justified with sufficient physical/theoretical arguments or a hygroscopicity closure based on measured soluble fraction (as done by Kumar et al. 2011b); I strongly recommend the authors to tone down such claims throughout the text. This is especially true since there seem to be errors in the hygroscopicity parameter used for the analysis of Figs. 3, 4, and 9 (see technical comments for details). Further, an exponent analysis applied similar to the Kumar et al. (2011a,b) leans support for FHH-AT, not κ -KT.

Specific/Technical concerns:

1. CCN measurements and their interpretation

The κ lines shown in Figs 3, 4, and 9 by Garimella et al. (2013) for $\kappa = 0.1, 0.2, 0.02,$ and 0.01 seem to be incorrect. “ $\kappa = 0.1$ ” should read “ $\kappa = 0.01$ ”, etc. The κ lines upon which the results of Kumar et al. (2011a,b) and Herich et al. (2009) plotted also seem incorrect. This impacts the comparisons and discussions made by authors.

To facilitate the review, I took the liberty to re-plot the data, superimposing the Kumar et al. (2011a) measurements and displaying the results in the following figure; power law fits to the data were also made, and are presented in attached supplement. In interpreting the data, support for each theory is evaluated based on its ability to reproduce x_{exp} (Kumar et al. 2011a). According to KT, particles with appreciable hygroscopicity exhibit $x = -3/2$. For KT, as κ decreases from $\kappa = 0.05$ to $\kappa = 0$ (Fig. 2 of Kumar et al., 2011a) particles becomes less hygroscopic causing a decrease in the exponent from $x = -1.5$ to $x = -1.0$. KT can only give a different exponent if there is an appreciable soluble fraction that changes strongly with size (e.g., Kumar et al., 2009b). In FHH-AT, x depends on the value of A_{FHH} and B_{FHH} but generally ranges between -0.80 and -1.20 (Kumar et al., 2009a). Shape factors can account for the location of the critical supersaturation vs. dry diameter lines but to a much lesser extent than the exponent (Kumar et al., 2011a). Please see attached supplement for discussion below.

- Visually, the data of Kumar et al. (2011a) is not considerably different from Garimella et al (2013), despite the strong difference in aerosol generation technique and all the other potential artifacts discussed in the text. It is very unlikely that size-dependence of soluble fraction in the particles across the studies could be the same; if the soluble fraction were the source of hygroscopicity (the basis of κ -KT) then I would have expected to see much more variability in the observed CCN activity. The data seem to “fall” on the same FHH-AT isoline regions.
- Four different measurements for dry corrected ATD are provided by Garimella et al. (2013); $x_{\text{exp}} \sim -0.67$ was calculated which is inconsistent with κ -KT ($x_{\kappa} \sim -1.5$) for the range of κ considered (0.01 to 0.1), unless if there is a strong size-dependence in the soluble fraction. Combining the experimental data sets of Kumar et al. (2011a) and Garimella et al. (2013), an $x_{\text{exp}} \sim -0.73$ is observed. FHH fits to these combined data points yield $x_{\text{FHH}} = -0.84$ for one set of parameters.
- For dry generated Illite, Garimella et al. (2013) provide two measurements resulting in the $x_{\text{exp}} \sim -0.41$, which neither κ -KT nor FHH-AT can easily explain (Kumar et al. 2011a). From only two data points however it is

questionable if one can conclude which theory is better. By combining experimental data points of Kumar et al. (2011a) with Garimella et al. (2013), $x_{\text{exp}} \sim -0.95$, which is close to $x_{\text{exp}} = -0.92$ from Kumar et al. (2011a), and follow well the corresponding FHH-AT fit ($A_{\text{FHH}} = 1.02$ and $B_{\text{FHH}} = 1.12$).

- A similar analysis holds true for dry generated NaMon aerosol where CCN activity is better described by FHH-AT based on exponents fit.
- Dry generated ATD for Garimella et al. (2013) compare well with those of Kumar et al. (2011a). Some differences are seen for dry generated Illite and NaMon, which could be attributed to differences in aerosol generation method between the two studies.

Page 31058, Line 25-26: “In the case of ATD.....size is corrected”. I agree that there is a discontinuity in the κ for the entire spectrum. For this reason, Kumar et al. (2011a) fit FHH-AT to dry generated ATD, and found that FHH-AT agrees with experimental measurements for the entire spectrum of dry diameters which is not possible with a single-set of values for κ -KT.

2. FHH Adsorption Activation Theory

Eq. 4 is the Brunauer, Emmet, and Teller (BET) Adsorption, not the Frenkel, Halsey and Hill (FHH) Adsorption isotherm (Sorjamma and Laaksonen, 2007). The two isotherms models should not be mixed in the discussion, as their formulations are very different. The BET model, as presented here, requires one parameter (“ c ”) to define the relationship between surface coverage, θ and Saturation ratio, S . The FHH model is a two parameter model based on A and B in Eq. 5 (where A defines interaction of adsorbed molecule between the surface and first layer, and B defines interactions between surface and subsequent layers) so that $S = \exp(-A/\theta^B)$, where $\theta = (D - D_{\text{dry}})/2D_w$, D is the droplet diameter, D_{dry} is the dry diameter of the particle, and, D_w is the diameter of a water molecule. It is my request to the authors to make this correction.

For completeness, the authors should provide a summary of available FHH-AT data on dust/mineral aerosol activation, just like they have provided mineral/dust aerosol κ for κ -KT.

Page 31047, Line 12: “asserts” seems too strong, as FHH-AT can explain equally well (if not better) the data. “suggested” might be a better term.

3. Particle Generation and Charge Correction

Kumar et al., 2011b found that using a minimum two in-line Silica gel diffusion dryers is required to maintain Relative Humidity (RH) to less than 5% for the wet generation method. Has the inline RH been measured to ensure conditions are truly dry for the particles generated via wet-generation?. A number size distribution data comparing blank (DDI) versus wet-atomized mineral aerosol would be good to show here as well.

Page 31052, Line 12 - 15: “This method....mineral dust”. Authors suggest that automatic application of charge corrections does not correct size distributions for dry-generated aerosols, where distributions do not span the entire range of scans. This is true. Authors, however do not provide a charge-correction procedure to correct this artifact which is observed for dry-generated mineral aerosols other than what is applied by the BMI software based on method of Wiedensohler et al. (1998).

Kumar et al. (2011a) showed that charge corrections must be applied carefully as well. I (and co-authors of Kumar et al., 2011a) were very careful to use an impactor at the largest size bin, to ensure that only singly-

charged particles flow through, and apply the charge-correction procedure which was clearly laid out by Moore et al. (2010). Please indicate how charge corrections were applied for dry generated aerosols and, ideally, an example of how the particle size distribution looked like before and after the correction.

Kumar et al. (2011a) found that dust particles generated with the dry method (soft saltation) were highly charged, and required a significant amount of neutralizing – one Kr neutralizer was not enough for the particles to acquire a charge distribution in equilibrium. Did Garimella et al. (2013) carry out such tests? This may be one reason why the particles in the dry generation experiments may have less singlets than thought. Please include this in your discussions.

4. Results

Page 31056, Line 17-22:

- Ion Chromatography analysis determines concentrations of ions (cations and anions) in terms of mmol/kg. Please provide a detailed analysis on procedure used for determining resulting particle size equal to 40 – 50 nm that corresponds to smaller peak of wet-generation method for ATD and Illite.
- Kumar et al. (2011b) Table 3 indicated that typical density of salts found in mineral aerosol can range from 1.8 g cm⁻³ for calcium nitrate to 2.66 g cm⁻³ for potassium sulphate. Similarly, Ca²⁺, NO₃⁻¹, K⁺, SO₄²⁻ are the ions measured in Garimella et al. (2013). Please clearly describe your assumption of using single density equal to 2.3 g cc⁻¹ in your calculations.
- There is no description of carbonates (CO₃²⁻) in the IC analysis even though it is well known that calcite (CaCO₃) can make up to 30% of dust composition (Kumar et al., 2011b). Please revise your IC analysis to include CO₃²⁻ in measurements and discussions.

Page 31057, Line 13-14: “Accounting for.....droplet activation (Fig. 3).” Please provide inversions (as a new Figure) to illustrate the effect of multiple charge on supersaturation at a given dry diameter.

5. Discussion

Page 31058, Line 18: The authors claim that the FHH fits from Kumar et al. (2011a) may be biased by the size artifact, and no evidence is provided to indicate that the smaller particle were corrected in Kumar et al., 2011a. This is incorrect. An example of the impact of multiple charging is shown in Fig. 3 of Kumar et al. (2011a), where one can clearly see that the multiple charge correction in this case increased D_{dry} from 170 nm to 247 nm. Kumar et al. (2011a) also conclude that the effect due to multiple charge correction is further pronounced at lower critical supersaturations (equal to 0.15% and 0.20%). Further, Kumar et al. (2011a) describe the procedure utilized for correcting the D_{dry} due to multiple charges as well as reference the procedure of Moore et al. (2010) to account for the uncertainty in the activation efficiency due to counting statistics uncertainty and flow rate variability. Please revise this incorrect statement used by the authors in this study regarding Kumar et al. (2011a)

Minor Comments

Following are comments on sections of the paper (denoted by the heading).

1. Introduction

Page 31045, Line 6-11:

- Please provide a reason as to why Illite, Sodium-rich Montmorillonite (NaMon), and Arizona Test Dust (ATD) represent a good approximation for mineral dust aerosol.

- Please provide dust sources rich with above mentioned minerals with references.
- Please provide references for previous studies that authors mention to have previously studied CCN activation of these minerals, for example: Koehler et al., 2009; Herich et al., 2009; Sullivan et al., 2009, 2010, Kumar et al., 2011a,b.
- There are other minerals such as Calcium-rich Montmorillonite, Quartz/Silica, Calcite, and Kaolinite that also represent dust aerosol. These minerals are not considered and it is fine that it is out of scope of this manuscript. But the information should be mentioned in this manuscript.

The authors provide a detailed explanation for the two different theories in Section 2. However, a brief introduction for the two different frameworks i.e. κ -KT and FHH-AT must be provided in the Section 1: Introduction to help understand why different frameworks were developed and used. This link is missing when going to Section 2.

Introduction should also discuss why CCN activation results for dust and mineral aerosols studied in literature and this manuscript are compared to different theories (κ -KT and FHH-AT).

Lab aerosol generation method for dust aerosol has been previously studied in the literature. The authors should provide a more detailed introduction on the previously studied aerosol generation method as it forms an important basis of this study where dry and wet generation methods are discussed. The authors should then link previous literature of Herich et al.(2009), Koehler et al.(2009), Sullivan et al. (2009, 2010) and Kumar et al.(2011a,b) to these methods.

The authors do not introduce the charge and shape effects in Introduction, even when it is importantly discussed through the text of this paper. Please provide references for literature that have i.) Quantified shape of dust particles, for example, Okada et al. (2001), Chou et al. (2008), and Kumar et al. (2011a), and ii) Quantified charge and shape of aerosols including dust particles to CCN activation, for example, Lance et al., (2006), Rose et al. (2008), and Kumar et al., (2011a).

The authors should also provide past literature that has measured/quantified the effect of soluble portion of dust/mineral aerosol to CCN activation, for example, Kumar et al. (2011b) and Sullivan et al. (2010). This analysis should form the basis for introducing IC and PALMS in this study.

Page 31044, Line 2: Please revise to read as "...are amongst the important factors..."

2.2 FHH Adsorption Activation Theory

Page 31047, Line 9: Please replace "dust particles" with "mineral aerosols" after CCN activity to read as "...describe CCN activity of mineral aerosols..."

3.1 Samples

Authors provide a comparison on CCN activation potential of ground and unground ATD in Table 1 and conclude that grinding process does not affect the activation behavior for Illite and NaMon. Please include corresponding data for Illite and NaMon in Table 1.

Does grinding of particles affect the particle shape? This is implicitly assumed throughout the study. It is suggested to present relevant data, in Table 1, for ground and unground ATD, Illite, and NaMon.

Authors suggest that the grinding process increases the number of particle in 100-1000 nm range (Page 31048, Line 6). It would be useful to show corresponding number size distributions for ground and unground ATD, Illite, and NaMon.

3.2.1 Particle Generation

Page 31049, Line 20: Please provide more details about in-line dryer. What dryer was used and number of dryers used in series.

Page 31049, Line 3: Please clarify if it is Teflon Bars or Teflon Beads. Text says bars while Fig. 1 says beads.

In Fig.1 for dry generation method (shown in red) please indicate the amount of excess flow sent to hood.

In Fig.1 for wet generation method (shown in blue) please indicate the amount of excess flow sent to hood.

3.2.2 Particle Sizing and Counting

Page 31050, Line 2: Please provide radioactivity and units of Polonium-210. Also please provide total number of strips used in the setup.

The authors set DMA sheath flow rate to 5 L min^{-1} such that DMA sheath to aerosol ratio (SAR) equals 5:1.36. Please provide an explanation as to why DMA SAR equal to 5:1.36 is used in this study. Also compare this ratio to previous studies on mineral aerosol CCN activation, for example, Herich et al., 2009; Koehler et al., 2009; Sullivan et al., 2009,2010; and Kumar et al., 2011a,b.

Kumar et al. 2011a found that a minimum of three Kr-85 neutralizers in series were required to completely neutralize the surface charges and attain the Boltzmann equilibrium distribution. Please provide data to ensure that sufficient strips of Polonium-210 were used to attain Boltzmann equilibrium distribution in these experiments.

3.2.3 CCN Counting

Page 31051, Line 1: Please define a new symbol for supersaturation, as S has been previously used by the authors to define Saturation Ratio.

Page 31051, Line 6 - 8: The value $S = 0.05$ is close enough to $S = 0.07$ in Chamber A. Similarly, value $S = 0.71$ is close enough to $S = 0.70$ in Chamber B. It is unclear what values of S authors are referring to as “outside typical operational ranges of CCNC”.

What was the CCNC Sheath to Aerosol Ratio? Also compare this ratio to previous studies on mineral aerosol CCN activation, for example, Herich et al., 2009; Koehler et al., 2009; Sullivan et al., 2009, 2010; and Kumar et al., 2011a,b.

3.2.4 Ion Chromatography

Kumar et al. (2011b) and Sullivan et al. (2010) are two studies that performed IC measurement on regional dust and mineral aerosols. Please compare your list of anions and cations with literature studies that have performed IC on dust and/or mineral aerosols.

Carbonates are expected to be dominant soluble ions in dust aerosol (Kumar et al., 2011b). It is surprising authors have not accounted for carbonates in their IC analysis measurements.

Further, it is strongly suggested to provide hygroscopicity closure based on IC to determine κ from IC as performed by Kumar et al. (2011b). This will provide a clear indication on the applicability of κ -KT or FHH-AT to mineral aerosol CCN activity.

3.3.2 Shape Correction

Page 31054, Line 11: Authors claim that Wenk and Bulakh (2004) have indicated that clay minerals have a layered structure with flat or sheet-like morphology. Please provide specifics for the clay minerals investigated by Wenk and Bulakh (2004). Please indicate if same minerals (i.e. ATD, NaMon, Illite) were considered in that study. If not, please provide an explanation on how Wenk and Bulakh (2004) results can be extended to minerals considered in this study (i.e. ATD, Illite, and NaMon)? This will be a useful comparison.

Page 31054, Line 20 - 24: I would request to re-write this statement to clearly explain that $D_{me} = 200$ and 400 nm were chosen for dry-generated ATD, Illite, and NaMon; $D_{me} = 50, 100,$ and 400 nm were chosen for wet-generated ATD and Illite. However, only $D_{me} = 50$ and 400 nm were selected for wet-generated NaMon.

Please provide a better explanation as to how Eq. 7 fits into Eq. 8. If Eq. 7 is not needed for calculation of D_{se}/D_{me} in Table 2, please remove Eq. 7 for clarity and brevity.

4. Results

Page 31055, Line 21: Some clarification is needed on the statement “..., the peak concentration is shifted to a smaller size.”. As far as I am concerned, the results actually show that lower concentration of particles is observed at the same D_{me} for cyclone-impacted dry generated particle when compared to dry-generated particles. This is different from peak concentration shifting to smaller size. Kindly revise if true.

Page 31056, Line 5-6: There is some ambiguity in this statement. Please state that the first peak in wet generation method is in part a result of soluble non-mineral particles. Other factors may also be contributing to this.

Page 31057, Line 7-8: “The images...mobility size”. How did the authors reach this conclusion just based on visuals? There is no EM scale provided in Fig. 4 for particle images to be compared to D_{me} . Please provide more information.

5. Discussion

Page 31058, Line 12 - 14: “Impaction...size (Figs. 4 and 8)”. This statement holds true for ATD where CCN activity for dry corrected is comparable to dry w/impactors (Fig. 4). The authors provide results for Dry-w/impactor for Illite and NaMon at 200 nm (Fig. 4) but do not provide results for dry corrected Illite and NaMon at 200 nm. Please provide these points for completeness.

6. Conclusions and Atmospheric Implications

Page 31059, Line 22: “introduce a soluble mode”. In my opinion there is not enough evidence based on IC analysis that only soluble materials contribute to this more hygroscopic peak in wet generation. Please revise this statement to include the word “potential” or “possible”.

Page 31059, Line 25-26: “The effective...of the dust”. There is no analysis comparing κ of wet generated dust to dry generated dust in this paper and this is introduced in conclusions. I request that authors either should discuss this in the main results or not mention this for the first time in conclusions.

Page 31060, Line 17-18: “For representation...0.01-0.1”. The values of κ suggested by authors needs to be revised, as some of the κ values in Figs. 3, 4, and 9 do not seem correct.

Editorial Comments

1. Introduction

Page 31043, Line 2: Please replace “Their” with “Aerosols” to read “Aerosols influence...”

Page 31043, Line 20: Please include “and growth” to read “...they facilitate the nucleation and growth of droplets.”

Page 31043, Line 26: Please remove quotes.

Page 31043, Line 27: Please replace “in” with “that alters the” to read as “...factor that alters the Earth’s...”

Page 31043, Line 28: Please include “a” before CCN to read as “...serve as a CCN and...”

Page 31044, Line 4: Please include “and their analytical analysis” to read as “...aerosol particles and their analytical analysis indicate...”

Page 31044, Line 9: Please include “a” before CCN to read as “...as a CCN in the...”

Page 31045, Line 13: Please provide the following references for κ -Köhler Theory (KT): Köhler, 1936; and Petters and Kreidenweis, 2007.

Page 31045, Line 13: Please provide the following references for FHH Activation Theory (FHH-AT): Sorjamaa and Laaksonen, 2007; Kumar et al., 2009a,b.

2.2 FHH Adsorption Activation Theory

Page 31047, Line 10: Please provide references for FHH-AT after “dust particles”, for example, Sorjamma and Laaksonen, 2007; and Kumar et al., 2009a, b.

3.1 Samples

Page 31048, Line 12: Please also add shape in addition to surface roughness for statement to read as “..such as surface roughness and shape, ...”. Artificial grinding of particles will not only affect surface roughness but will also affect particle shape.

3.2.1 Particle Generation

Page 31049, Line 2: Please replace “mineral dust” with “mineral aerosols”. The samples used by authors are not mineral dust, but mineral aerosols that are representative of atmospheric dust aerosol.

Page 31049, Line 4: Please replace “dust” with “mineral samples” or something similar, and be consistent throughout.

Page 31049, Line 11: Please replace “dust” with “mineral samples” or something similar, and be consistent throughout.

3.2.2 Particle Sizing and Counting

Page 31050, Line 4: Please define Z_p in Eq. 6.

Page 31050, Line 14: Give full form of SMPS.

3.2.5 Electron Microscopy

Page 31052, Line 4: Please provide more details about the conventional impactor.

Page 31052, Line 5: Define MOUDI

Page 31052, Line 7: Define MIT

Page 31052, Line 8: Define JEOL

Page 31052, Line 8: Define TEM

3.2.1 Shape Correction

Page 31053, Line 22: Please provide references after "...mobility as the particle."

Page 31054, Line 7: Please provide references after "... , with $\chi \geq 1$."

Page 31054, Line 10: Please provide reference after "...are non-spherical."

Page 31054, Line 15: Please provide reference for Eq. 7.

4. Results

Page 31055, Line 24: Please complete your statement "...the peak of the wet distribution is shifted to a smaller size." What is this relative to? Is it relative to dry-generated NaMon or cyclone-impacted NaMon or dry/wet generated ATD/Illite?

Page 31056, Line 4: For ATD and Illite, the first peak due to wet-generation appears closer to 35 – 40 nm. Please provide a range from 30 – 50 nm.

6. Conclusions and Atmospheric Implications

Page 31059, Line 12: Please include "cloud droplet nucleation" between "...their..." and "... in the..".

Page 31059, Line 15: "three mineral dusts...". These are representative mineral samples and not mineral dusts. Please revise throughout the text.

Figure 1, Page 31069:

- Text says bars and Fig. says beads. Please be consistent.
- Please provide excess flow rate to hood shown in blue and red.

Figure 2, Page 31070:

- Define SMPS in text.

Figure 3, Page 31071:

- Please indicate what diameter is being referred to in x-axis. Is it D_{me} or D_{se} or D_{ve} ?
- Please correct kappa lines for $\kappa = 0.01, 0.02, 0.1, \text{ and } 0.2$. They seem incorrect.

Figure 4, Page 31072:

- The sub-figures are extremely difficult to follow. Please provide these in different pages and increase resolution.
- Please correct kappa lines for $\kappa = 0.01, 0.02, 0.1, \text{ and } 0.2$. They seem incorrect.

Figure 4, Page 31073:

- Please correct typo. Illite is a non-swelling clay, not swelling clay. NaMon is a swelling clay, not non-swelling clay.
- ATD is not a non-swelling clay. It is an industry produced samples that behaves like a non-swelling clay.

Figure 5, Page 31074:

- Please include unground Illite to be consistent with ATD and NaMon.

Figure 7, Page 31076:

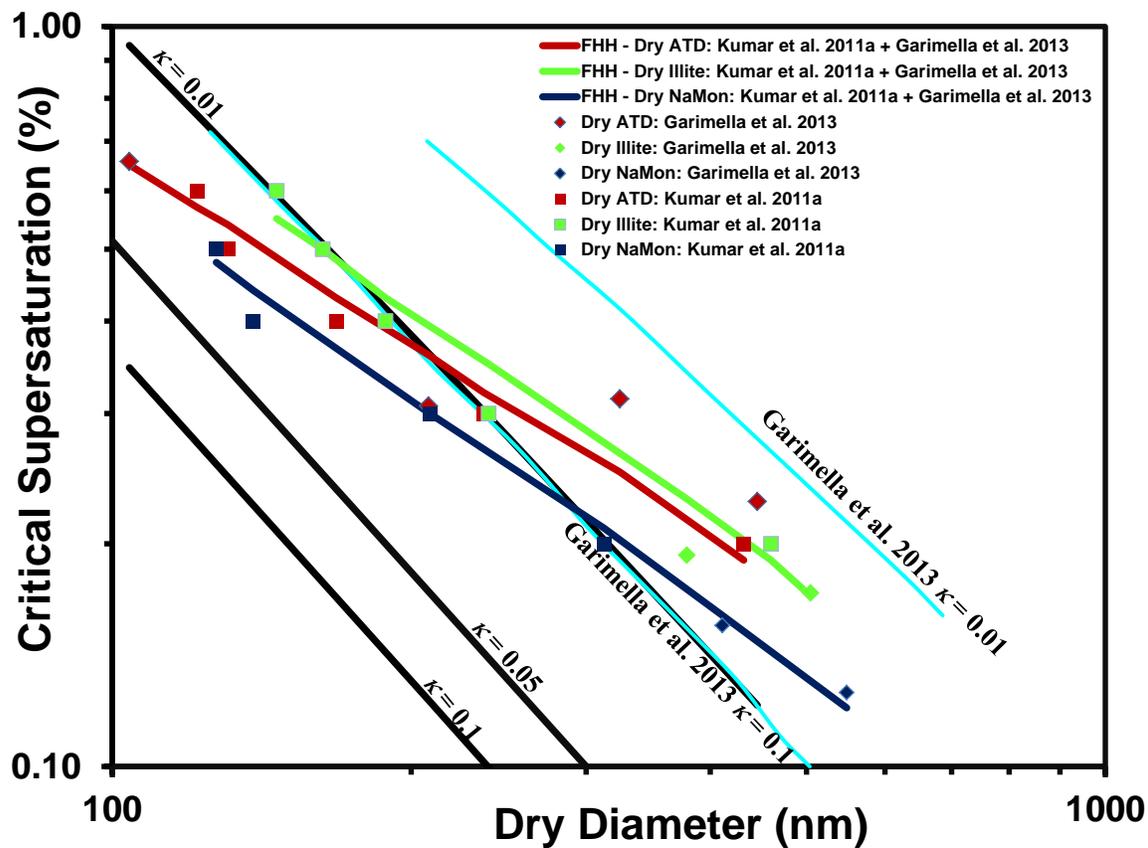
- Please mention in Fig. caption that PALMS peaks were collected on 400 nm particles.

Figure 9, Page 31078:

- Please correct kappa lines for $\kappa = 0.01, 0.02, 0.1, \text{ and } 0.2$. They seem incorrect.

References

- Chou, C., Formenti, P., Maille, M., Ausset, P., Helas, G., Harrison, M., and Osborne, S.: Size distribution, shape, and composition of mineral dust aerosols collected during the African Monsoon Multidisciplinary Analysis Special Observation Period 0: Dust and Biomass-Burning Experiment field campaign in Niger, January 2006, *J. Geophys. Res.*, 113, D00C10, doi:10.1029/2008JD009897, 2008.
- Kumar, P., Sokolik, I. N., and Nenes, A.: Parameterization of cloud droplet formation for global and regional models: including adsorption activation from insoluble CCN, *Atmos. Chem. Phys.*, 9, 2517–2532, doi:10.5194/acp-9-2517-2009, 2009a.
- Kumar, P., Nenes, A., and Sokolik, I. N.: Importance of adsorption for CCN activity and hygroscopic properties of mineral dust aerosol, *Geophys. Res. Lett.*, 36, L24804, doi:10.1029/2009GL040827, 2009b.
- Lance, S., Medina, J., Smith, J. N., and Nenes, A.: Mapping the operation of the DMT continuous flow CCN counter, *Aerosol Sci. Tech.*, 40, 242–254, 2006.
- 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.
- Morales Betancourt, R. and Nenes, A.: Understanding the contributions of aerosol properties and parameterization discrepancies to droplet number variability in a Global Climate Model, *Atmos. Chem. Phys. Discuss.*, 13, 31479–31526, doi:10.5194/acpd-13-31479-2013, 2013.
- Okada, K., Heintzenberg, J., Kai, K., and Qin Y.: Shape of atmospheric mineral particles collected in three Chinese arid-regions, *Geophys. Res. Lett.*, 28(16), 3123–3126, doi:10.1029/2000GL012798, 2001.
- Rose, D., Gunthe, S. S., Mikhailov, E., Frank, G. P., Dusek, U., Andreae, M. O., and Pöschl, U.: Calibration and measurement uncertainties of a continuous-flow cloud condensation nuclei counter (DMT-CCNC): CCN activation of ammonium sulfate and sodium chloride aerosol particles in theory and experiment, *Atmos. Chem. Phys.*, 8, 1153–1179, doi:10.5194/acp-8-1153-2008, 2008.
- Sorjamaa, R. and Laaksonen, A.: The effect of H₂O adsorption on cloud drop activation of insoluble particles: a theoretical framework, *Atmos. Chem. Phys.*, 7, 6175–6180, doi:10.5194/acp-7-6175-2007, 2007.
- Sullivan, R. C., Moore, M. J. K., Petters, M. D., Kreidenweis, S. M., Qafoku, O., Laskin, A., Roberts, G. C., and Prather, K. A.: Impact of particle generation method on the apparent hygroscopicity of insoluble mineral particles, *Aerosol Sci. Tech.*, 44, 10, 830–846, 2010.



Mineral Type (Dry Generated)	x_{exp} (Garimella et al., 2013)	x_{exp} (Kumar et al., 2011a)	x_{exp} (Kumar et al., 2011a + Garimella et al. 2013)	x_{FHH}	$x_{\kappa=0.1}$	$x_{\kappa=0.01}$
ATD	-0.67	-0.82	-0.73	-0.84	-1.5	-1.4
Illite	-0.41	-0.92	-0.95	-0.93	-1.5	-1.4
NaMon	-0.72	-0.93	-0.92	-0.93	-1.5	-1.4