The Importance of Adsorption for CCN Activity and Hygroscopic Properties of Mineral Dust Aerosol

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Abstract

This study uses published data on dust-water interactions to examine the importance of including water adsorption effects when describing the hygroscopic and cloud condensation nuclei (CCN) behavior of mineral dust aerosol. Adsorption activation theory (AT) better represents fresh dust-water interactions than Köhler theory (KT), as *i*) a consistent set of adsorption parameters can describe the hygroscopic behavior of dust (under both sub and supersaturated conditions), *ii*) the dependence of critical supersaturation, s_c, with particle dry diameter, D_{dry}, is closer to observations. The long adsorption timescale could also contribute to the large differences observed between dry and wet generated dust hygroscopicity. If KT and AT are consistently applied to the same dust size distribution, KT predicts up to tenfold higher CCN and 40% higher droplet number concentration than AT. This profoundly different behavior between the theories suggests that both may be required for a comprehensive description of atmospheric dust CCN activity.

1. Introduction

Mineral dust is ubiquitous in the atmosphere and represents a dominant type of particulate matter by mass. Dust particles can act as cloud condensation nuclei (CCN), giant CCN (GCCN) (e.g., Rosenfeld et al., 2001; Levin and Cotton, 2008), or ice nuclei (IN) (e.g., DeMott et al., 2003; Field et al., 2006) affecting cloud microphysics, albedo, and lifetime. Despite its well-recognized importance, assessments of dust impacts on clouds and climate are highly uncertain. In this study, we address the role of dust as CCN with the goal to provide an improved representation of dust CCN activation in the climate models.

Dust CCN activity is currently described by Köhler theory (herein KT; Köhler, 1936), which is based solely on the contribution of the solute and curvature effects upon water equilibrium vapor pressure. KT implies that dust particles devoid of any solute would require very high ambient supersaturations (dictated by the Kelvin equation) to act as CCN. It is well known however that adsorption of water on insoluble particles (especially clays) can lead to hygroscopic growth similar to deliquescent salts (e.g., Schuttlefield et al., 2007). Past studies have demonstrated that calcite ($CaCO_3$) (a mineral with very low solubility compared to deliquescent salts) and Arizona Test Dust (ATD) can interact with water vapor and adsorb multiple layers of water under subsaturated conditions (Gustafsson et al., 2005; Vlasenko et al., 2005; Hatch et al., 2008). This interaction implies that dust mixtures and individual minerals with hydrophilic insoluble surfaces can affect water activity of aerosol (especially when the solute fraction of particles is low) with largely ignored implications for predicted CCN activity. Henson (2007) and Sorjamaa and Laaksonen (2007) recognized this gap, and developed

adsorption activation theory (AT) to describe the activation of hydrophilic insoluble CCN. The Sorjamaa and Laaksonen (2007) formulation is based on the FHH (Frenkel, Halsey and Hill) adsorption model (and constrained by two adjustable parameters, A_{FHH}, B_{FHH}). Kumar et al. (2009) incorporated FHH-AT into a droplet activation parameterization for use in regional and global models, assuming that the aerosol constitutes an external mixture of "soluble" (KT) and "insoluble" (AT) particles.

Even if constrained by the same CCN activity or hygroscopic growth data, predicted CCN concentration and cloud droplet number, N_d, can differ between using KT and FHH-AT because: *i*) the relationship between particle critical supersaturation, s_c, and dry diameter, D_{dry}, differs between theories, resulting in a different predicted CCN spectrum even if the same size distribution is used, and, *ii*) KT particles require substantially more water to activate than FHH-AT particles with the same s_c (Kumar et al., 2009). Competition for water vapor in a cloud parcel during activation of KT particles can thus be more intense than for FHH-AT particles, leading to a different parcel maximum supersaturation, s_{max}, and droplet number.

In this study, we substantiate the importance of considering water vapor adsorption effects on the activation of mineral dust particles. This is done by fitting published CCN activity and hygroscopic growth data to the KT and FHH-AT, and examining whether each theory can *i*) describe subsaturated hygroscopic growth and CCN activity with one set of water-interaction parameters, and, *ii*) reproduce the observed dependence of s_c with respect to D_{dry}. Finally, we evaluate the differences in the CCN number and droplet number concentrations predicted by KT and FHH-AT, using the consistent parameters and the same aerosol size distribution.

2. Comparison of Köhler and Adsorption Activation Theories

KT provides a relationship between the equilibrium vapor pressure of an aqueous droplet as a function of its wet diameter and exhibits a maximum value termed as critical supersaturation, s_c, at a characteristic critical wet diameter, D_c. Particles exposed to ambient supersaturation above s_c typically activate into cloud droplets (Nenes et al., 2001). In KT, s_c depends on the amount of solute in the dry particle, which is related to its chemical composition and size. Petters and Kreidenweis (2007) parameterized the solute term of KT in terms of a hygroscopicity parameter, κ, which was derived from the relationship between D_{dry} and s_c. κ can be used to directly compare the hygroscopicity of aerosol over a wide range of composition, with $\kappa \rightarrow 0$ for completely insoluble particles (for which $s_c \sim D_{dry}^{-1}$) to $\kappa \rightarrow 1.4$ for the most hygroscopic atmospheric aerosol (for which $s_c \sim D_{dry}^{-3/2}$). According to KT, a constant value of κ should be able to describe both aerosol subsaturated water uptake (where relative humidity, RH, is below 100%) and predict CCN activity (RH > 100%).

FHH-AT is similar to KT, except that the solute term is replaced with an adsorption term modeled by the FHH isotherm (Crittenden and Thomas, 1998). The adsorption parameter B_{FHH}, strongly affects the shape of the equilibrium curve, and largely determines the existence and value of s_c and D_c (Kumar et al., 2009). As with KT, s_c in FHH-AT can be related to D_{dry} as $s_c = CD_{dry}^{x}$. Particles with an appreciable soluble fraction follow KT, and $x \sim -1.5$ when $\kappa > 0.2$. In FHH-AT, x varies between -0.8 and -1.5, depending on A_{FHH}, B_{FHH} (Kumar et al., 2009).

3. Evidence for Adsorption Activation

Figure 1a shows published data (symbols) of s_c as a function of D_{dry} (Koehler et al. 2009; Sullivan et al., 2009) for different dust types and individual mineral particles generated in the lab either with the use of a dry fluidized bed, or via wet atomization from an aqueous suspension of dust particles. The CCN activity data are fitted to a power law expression, $s_c = CD_{dry}^{x}$, from which the "experimental" exponent, x_{exp}, is determined (Table 1). A_{FHH} and B_{FHH} and the corresponding exponent, x_{FHH}, were determined from fitting the FHH-AT model (Figure 1a, lines) to the experimental data via least squares minimization. The KT fits to the data (expressed in terms of κ) are given by Koehler et al. (2009) and Sullivan et al. (2009), from which the corresponding KT exponent, x_κ, is computed. The values of the exponents, adsorption parameters (A_{FHH}, B_{FHH}), and κ (determined by Koehler et al., 2009, and Sullivan et al., 2009) are presented in Table 1.

In Figure 1b, x_κ (circles) and x_{FHH} (squares) are plotted against x_{exp} for all dust samples and individual minerals. With the exception of $CaCO_3$ and $CaSO_4$ (calcium sulphate) (where $x_\kappa \rightarrow$ -1 because of the very low κ), $x_\kappa \sim$ -1.5. $CaCO_3$ (representing fresh unprocessed dust) and $CaSO_4$, $CaC_2O_4.H_2O$ (calcium oxalate monohydrate or COH) (representing atmospherically processed mineral dust) are better described by FHH-AT, as x_{FHH} is in perfect agreement with x_{exp}. For wet-generated ATD, Owens Lake (OL), Canary Island Dust (CID), and oxalic acid ($C_2O_4H_2$), x_{FHH} lies closer than x_κ to the 1:1 line. x_κ for dry Saharan Dust (SD), ATD and wet $Ca(NO_3)_2$ are closer to x_{exp} than x_{FHH}; this is expected for $Ca(NO_3)_2$ because it is highly soluble (deliquescence RH = 49%; Fountoukis and Nenes, 2007), but not for dry ATD and SD. The large scatter ($R^2 < 0.7$

for the s_c-D_{dry} data for dry ATD) and potential size-dependant composition (for SD) may explain this.

Another indication that KT may be an incomplete description of the dust CCN activity presents itself in the value of wet-dust κ parameters, and the implications thereof. If the aerosol can be considered as a mixture of a soluble salt with molar volume (M_s/ρ_s), effective van't Hoff factor ν_s, and volume fraction ε_s, then $\kappa = \left(\frac{M_w}{\rho_w}\right)\left(\frac{\rho_s \nu_s}{M_s}\right)\varepsilon_s$, where (M_w/ρ_w) is the molar volume of water. Assuming that the hygroscopic fraction of dust behaves like ammonium sulfate gives $\left(\frac{M_w}{\rho_w}\right)\left(\frac{\rho_s \nu_s}{M_s}\right) = 0.61$ (Petters and Kreidenweis, 2007). Therefore, the dust κ parameters can be used to infer an "equivalent soluble volume fraction", computed as $\varepsilon_s = \frac{\kappa}{0.61}$. If KT indeed applies, then ε_s should reflect the true soluble fraction of dust. From the values of κ reported in Table 1, ε_s= 0.58, 0.65-1.78, and 0.43 for wet ATD, OL, and CID, respectively. Such a large soluble fraction in fresh dust is much larger (or even impossible if larger than unity) than the expected 2% soluble mass fraction in ATD (Vlasenko et al., 2005) and 3-37% in OL (Koehler, 2008). Koehler et al. (2009) attribute this enhanced hygroscopicity to redistribution of the soluble material among the insoluble dust cores, particularly in the smaller size range. Given that KT implies $s_c \sim \varepsilon_s^{-0.5} D_{dry}^{-1.5}$, ε_s will have to scale with $D_{dry}^{0.3}$ for KT to yield $x_\kappa = x_{exp} \sim$ -1.36. This means that ε_s varies more than 60% over the diameter range (40 – 200nm) reported in the Koehler et al. (2009) experiments, so that the soluble fraction at the high s_c should be close to unity. This is certainly possible; the

hygroscopicity parameter, however, does not seem to change considerably when subsets of the activation data (especially in the higher supersaturation range) are separately considered. This implies that KT may not completely describe the CCN activity of dust, so that other processes, such as physisorption, could contribute to the water activity depression required to yield the observed CCN activity. The long equilibration time (minutes or more) associated with adsorption (e.g., Schuttlefield et al., 2007) may explain why the hygroscopicity of dry and wet generated dust are so different. The residence time of dust particles in the instrumentation is typically less than a minute, limiting the amount of water that can adsorb and bias the observed hygroscopicity below its equilibrium value. Wetting the dust particles prior to measurement would maximize the amount of adsorbed water and express the full extent of its hygroscopicity. One approach to modeling this system is using one value for A_{FHH}, B_{FHH}, combined with a variable uptake coefficient (that is very low during formation of the monolayer, and progressively increases with the number of adsorbed layers). Another explanation is the swelling of clays; during complete wetting, more surface area could be exposed for interaction, which would enhance dust hygroscopicity compared to a dry particle. Future work should focus on the existence and mechanism of adsorption/desorption transients.

4. Reconciling Dust Hygroscopicity under Subsaturated and Supersaturated Conditions.

Herich et al. (2009) measured water uptake on mineral dusts and different clays under subsaturated (with a hygroscopicity tandem differential mobility analyzer; HTDMA) and supersaturated (with a cloud condensation nuclei counter; CCNc) conditions. The hygroscopic growth factors (GF) were measured with a HTDMA, and the

CCN activity was measured using a CCNc. A poor correlation in experiments (deviation larger than ±50%) was found between κ derived from the HTDMA and CCNc. Herich et al. (2009) attributed this to resolution limitations in the HTDMA GF. Alternatively, KT may not adequately represent dust-water interactions, so that a single value of κ is not expected to describe the subsaturated water uptake and CCN activity for mineral dust aerosol. If FHH is more appropriate, then one set of A_{FHH} and B_{FHH} (neglecting the potential non-equilibrium artifacts) should reproduce both subsaturated and supersaturated properties of mineral dust aerosol, and is attempted below.

Gustafsson et al. (2005) studied the subsaturated hygroscopic uptake of ATD particles generated from suspensions in distilled water. Surface coverages as a function of RH were measured using a thermogravimetric analysis, during which multilayer adsorption (the number of water molecule layers, $\theta \sim 3 - 4$) were observed for RH greater than 50%. Under such conditions, the FHH adsorption isotherm is applicable and is fitted to the data. The optimal values for A_{FHH} and B_{FHH} are 1.16 and 0.88, respectively, versus 0.85 and 0.88 from CCN activation experiments (Table 1). Vlasenko et al. (2005) measured subsaturated hygroscopic growth of dry ATD; fitting a FHH adsorption isotherm to the growth data for RH > 70% gives A_{FHH} = 0.19 and B_{FHH} = 0.98 (RMSE = 0.035), which are very close to the FHH parameters obtained from CCN activation experiments for the same compound (A_{FHH} = 0.27 and B_{FHH} = 0.79; Table 1). Fitting FHH isotherms to the Gustafsson et al. (2005) and Hatch et al. (2008) measurements for $CaCO_3$ (different type from Table 1) gives A_{FHH} = 0.83-1.00 and B_{FHH} = 0.76. All the above suggests that the adsorption parameters for similar samples are indeed consistent across different studies.

5. Impact of KT and AT on CCN and Droplet Number

In this section, differences in predicted CCN concentrations and droplet number concentrations from application of KT and FHH-AT are estimated. For this, we use a single-mode lognormal aerosol obtained from in-situ measurements of SD during the NAMMA field campaign (Twohy et al., 2009) (with geometric mean diameter, D_g = 0.10 μm, standard deviation, σ_g = 1.6, and total particle concentration, N_0 = 225 per cm^3). The CCN spectra computed with KT and FHH-AT (using κ, A_{FHH}, and B_{FHH} listed in Table 1 and the lognormal CCN spectra formulations of Kumar et al., 2009) are presented in Figure 2a. For supersaturations between 0.05% and 0.5% (a range relevant for cumulus and stratocumulus clouds), application of KT results in 8-12 times larger CCN that when applying FHH-AT. This is a direct consequence of $x_\kappa < x_{FHH}$, which tends to yield a larger activation fraction for KT-derived CCN spectra. For supersaturations greater than 0.5%, most aerosol in both distributions activate, so CCN computed by KT and FHH-AT converge.

The larger CCN concentrations (at a given supersaturation) associated with use of KT suggests that the calculated droplet number, compared to using FHH-AT, will be larger. KT however requires more water to activate particles than FHH-AT (Kumar et al., 2009), so the competition for water vapor in the former particles is stronger, potentially impacting s_{max} and N_d. For example for s_c = 0.05%, the ratio of water volume at D_c in KT against FHH-AT ranges from 4.83 (dry ATD) to 15.43 (wet ATD). Hence for the same size distribution, the droplet number difference from application of each theory depends on two competing factors: *i*) the stronger competition of KT CCN for water vapor, and *ii*) the typically larger activation fraction associated with KT. These factors are

203 comprehensively accounted for in droplet number calculations carried out with the
204 Kumar et al. (2009) parameterization. In all droplet number calculations presented, the
205 parcel is assumed adiabatic, with initial temperature, 273 K; pressure, 600 mbar; and
206 updraft velocity, w ranging from 0.1 ms^{-1} to 10 ms^{-1}. For each dust type, the respective κ
207 and FHH parameters (A_{FHH} and B_{FHH}) from Table 1 are used.

208 Figure 2b shows the ratio of total CCN that activate to cloud droplets using KT, N_d^{κ},
209 to that from FHH-AT, N_d^{FHH}, as a function of parcel updraft velocity (symbols) for four
210 different dust types. The corresponding parcel s_{max} is also shown (solid lines). For wet
211 CID and wet ATD, $\frac{N_d^{\kappa}}{N_d^{FHH}}$ is largest (~ 1.3 - 1.4) at w ~ 0.1 ms^{-1} and approaches 1.0 for w
212 > 1 ms^{-1}. This is because the parcel s_{max} < 1% for all w < 1 ms^{-1} (Figure 2b), where
213 $\frac{F_k}{F_{FHH}} > 1$ (Figure 2a) and droplet differences are dominated by the larger activation
214 fractions associated with KT. Similarly, $\frac{F_k}{F_{FHH}} > 1$ for dry ATD and SD and w < 1 ms^{-1}.
215 However, for w > 1 ms^{-1}, the competition of water vapor from KT particles is sufficiently
216 strong so that $\frac{N_d^{\kappa}}{N_d^{FHH}} < 1$. At very high updrafts (> 3 ms^{-1}), all particles activate,
217 and $\frac{N_d^{\kappa}}{N_d^{FHH}} \to 1$.

218 **6. Conclusions.**

219 In this study, we compared Köhler theory (KT) against FHH adsorption activation
220 theory (FHH-AT) when applied to the activation of mineral dust aerosol. Based on

published data, a number of potential issues were found with KT, suggesting it may not fully represent CCN activity of mineral dust aerosol, since *i*) a consistent set of FHH-AT adsorption parameters can be found that describe both the subsaturated hygroscopic growth and CCN activity, and, *ii*) the critical supersaturation vs dry diameter exponents determined for FHH-AT are often closer to observations, than those from KT. Application of KT and FHH-AT leads to the differences in predicted CCN and cloud droplet number concentrations, even if consistent hygroscopicity and adsorption parameters (i.e., derived from the same experimental data) are used. For the dust samples considered here, CCN concentrations can differ by a factor of 10, and results in a 40% difference in predicted cloud droplet number concentration. Thus, a comprehensive description of CCN activity of mineral dust aerosol throughout its atmospheric lifetime may require a combination of both KT and FHH-AT.

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298 Generation of Submicron Arizona Test Dust Aerosol: Chemical and Hygroscopic
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300 **Table 1:** FHH parameters for different mineral dusts and dust related compounds
301 composites. FHH adsorption activation fits to the experimental CCN activity data
302 obtained from Koehler et al. (2009) and Sullivan et al. (2009).

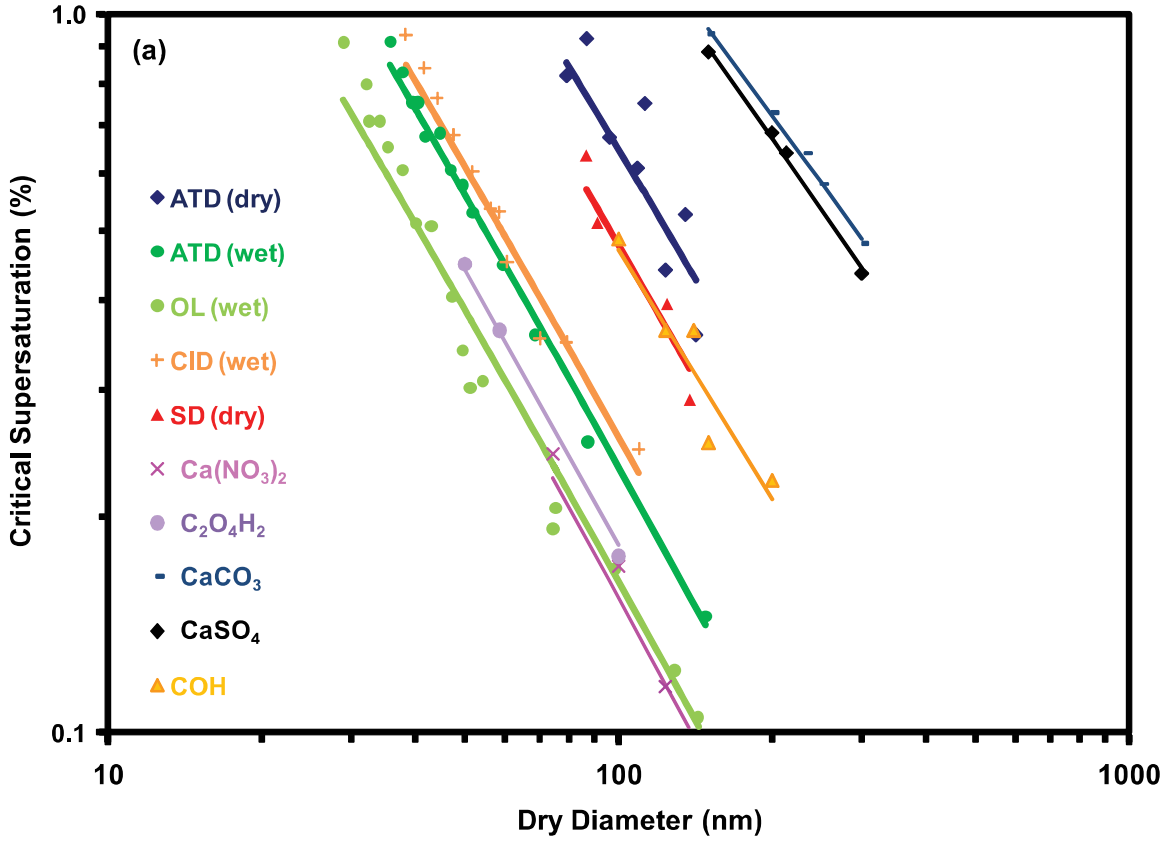
Description (Acronym)	**Generation***	κ	A_{FHH}	B_{FHH}	x_{κ}	x_{FHH}	x_{exp}
Arizona Test Dust (ATD)	Dry	0.025	0.27	0.79	-1.43	-1.20	-1.39
Arizona Test Dust (ATD)	Wet	0.35	0.85	0.88	-1.49	-1.26	-1.36
Owens Lake (OL)	Wet	0.39-1.07	1.14	0.91	-1.50	-1.25	-1.36
Canary Island Dust (CID)	Wet	0.26	0.80	0.88	-1.49	-1.24	-1.33
Saharan Dust (SD)	Dry	0.054	0.42	0.83	-1.47	-1.23	-1.42
Calcium Nitrate ($Ca(NO_3)_2$)	Wet	0.51	1.13	0.90	-1.50	-1.30	-1.59
Oxalic Acid ($C_2O_4H_2$)	Wet	0.50	1.02	0.90	-1.50	-1.27	-1.35
Calcium Carbonate ($CaCO_3$)	Dry	0.0011	0.25	1.19	-1.18	-0.96	-0.96
Calcium Sulfate ($CaSO_4$)	Dry	0.0016	0.10	0.91	-1.21	-1.02	-1.02
Calcium Oxalate Monohydrate (COH or $CaC_2O_4.H_2O$)	Dry	0.048	0.57	0.88	-1.47	-1.15	-1.16

303 *"Dry" refers to dust particles generated with a fluidized bed; "Wet" refers to atomization from an
304 aqueous solution/suspension

Figure Captions

Figure 1. (a): FHH adsorption activation fits (lines) to the observed CCN activity (points) for dust types presented in Table 1. Data obtained from Figure 7.1 (pp 154) and Figure 5 from Koehler et al. (2009) and Sullivan et al. (2009), respectively. "Dry" refers to dust particles generated with a fluidized bed, and "wet" refers to atomization from an aqueous suspension. **(b):** Comparison between x_{exp}, x_{κ} (circles) and x_{FHH} (squares). Color scheme identical to (a). Dashed lines represent ±7.5% deviation from 1:1 line.

Figure 2: (a): Ratio of CCN spectrum given by Köhler theory to that given by FHH adsorption activation theory as a function of supersaturation. Numbers noted on each curve refer to the ratio of water volume required by KT over FHH-AT to activate a CCN with s_c = 0.05%. **(b):** Ratio of parameterized activated fraction (points) for different dust types as a function of increasing updraft velocity in a cloud parcel. Also shown are the corresponding parcel s_{max} (lines) for each dust type. Color scheme identical to (a). Dust types defined in Table 1.



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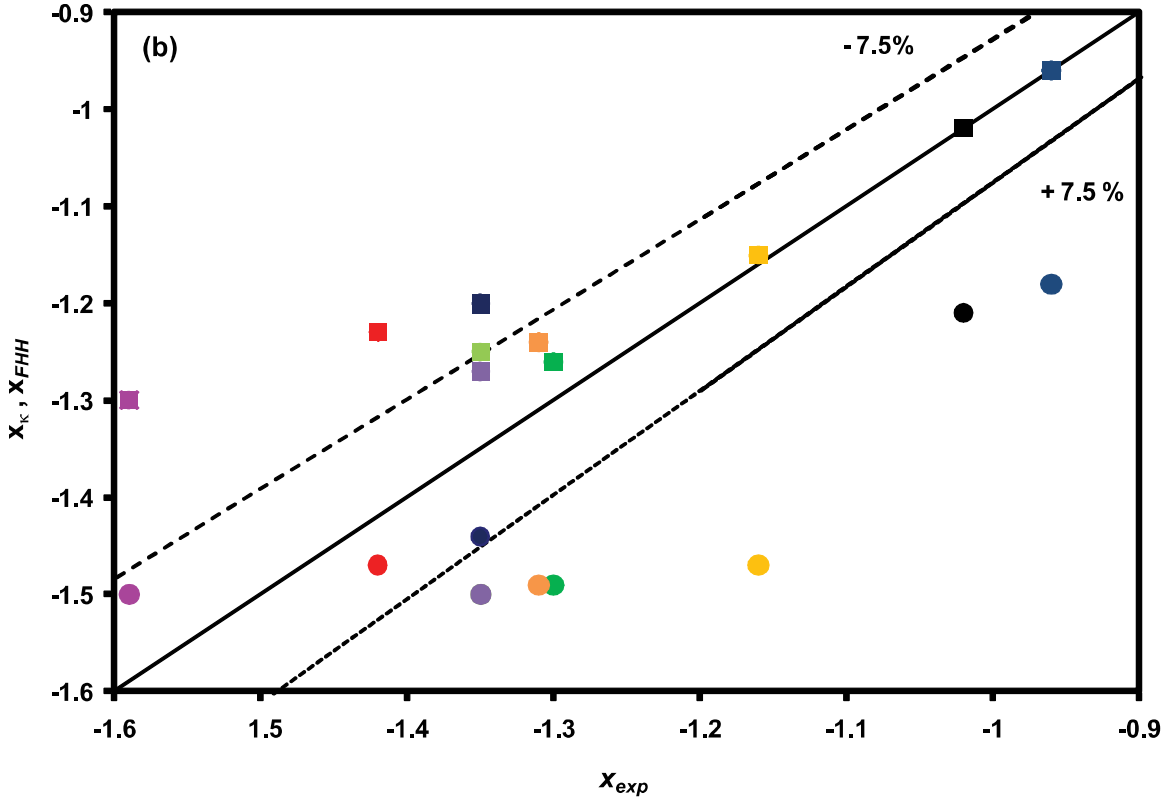
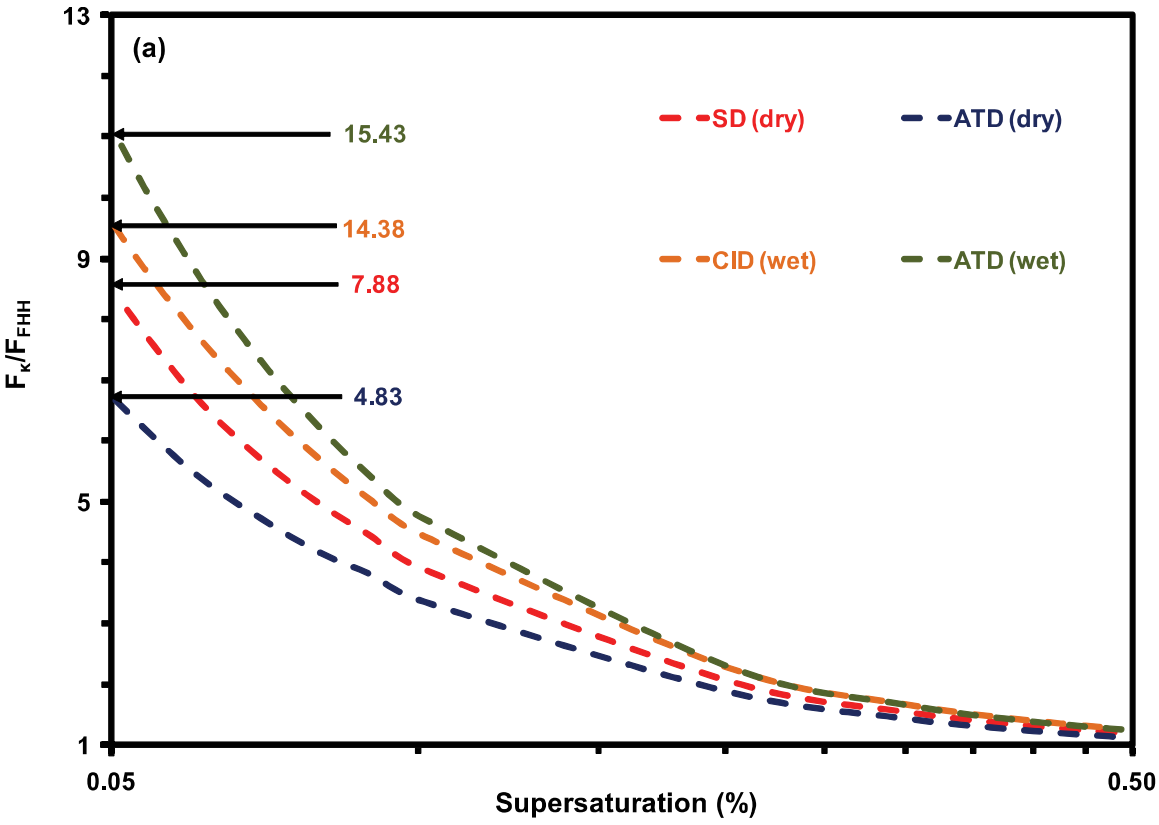


323
324 **Figure 1**


(a)
SD (dry)
ATD (dry)
CID (wet)
ATD (wet)
15.43
14.38
7.88
4.83
13
9
5
1
0.05
0.50
Supersaturation (%)
F_κ/F_{FHH}

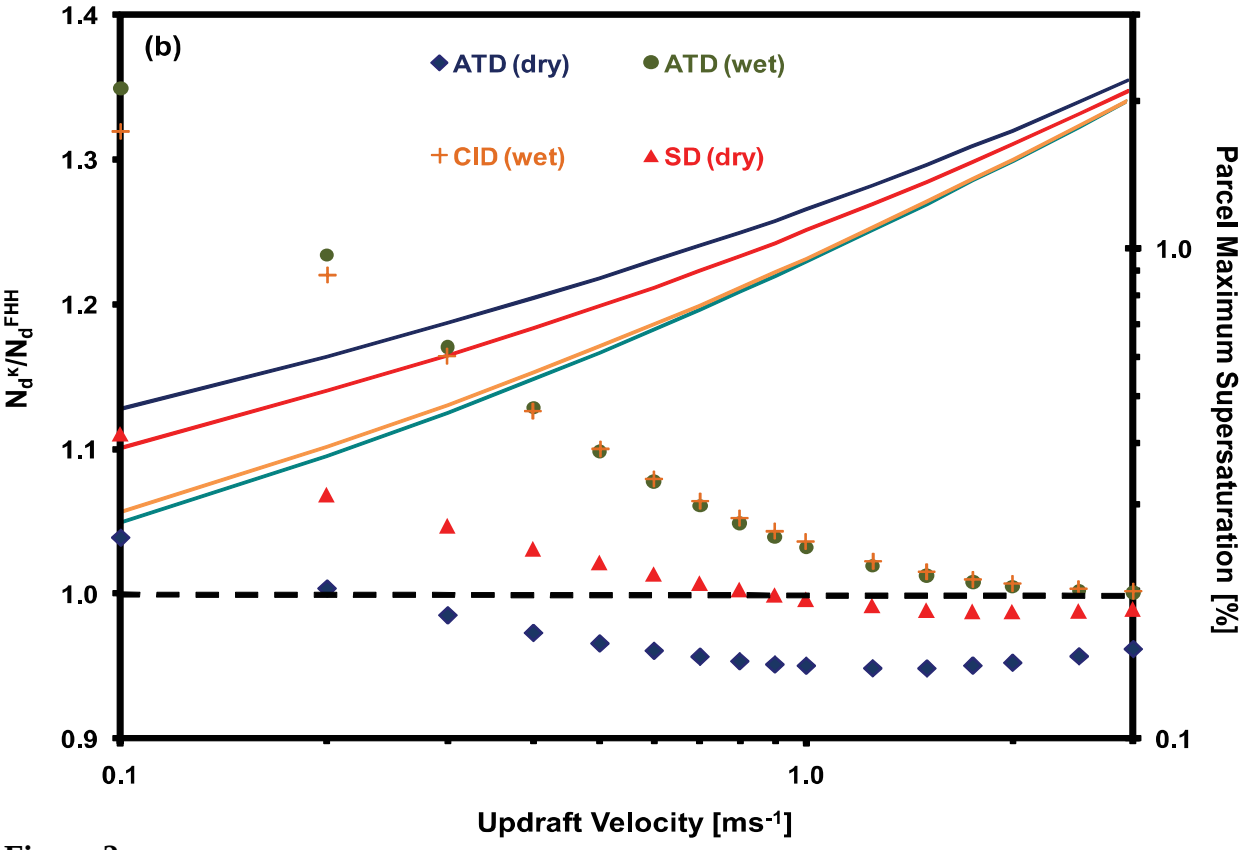

(b)
ATD (dry)
ATD (wet)
CID (wet)
SD (dry)
1.4
1.3
1.2
1.1
1.0
0.9
0.1
1.0
Updraft Velocity [ms^{-1}]
N_d^κ/N_d^{FHH}
Parcel Maximum Supersaturation [%]

Figure 2