



## Aerosol hygroscopicity and cloud droplet activation of extracts of filters from biomass burning experiments

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[1] In this laboratory closure study, we compare sub- and supersaturated water uptake properties for aerosol particles possessing a range of hygroscopicity. Measurements for water sub-saturated conditions used a hygroscopic tandem differential mobility analyzer (HTDMA). Simultaneously, measurements of particle critical supersaturation were conducted on the same sample stream with a continuous flow cloud condensation nuclei (CCN) counter. For these experiments, we used filter-collected samples of biomass smoke generated in the combustion of two common wildland fire fuels, western sagebrush and Alaskan duff core. Extractions of separate sections of the filter were performed using two solvents, ultrapure water and methanol. The extracts were subsequently atomized, producing aerosols having a range of hygroscopic responses. HTDMA and CCN measurements were fit to a single-parameter model of water uptake, in which the fit parameter is denoted  $\kappa$ , the hygroscopicity parameter. Here, for the four extracts we observed mean values of the hygroscopicity parameter of  $0.06 < \kappa < 0.30$ , similar to the range found previously for numerous pure organic compounds. Particles generated from the aqueous extracts of the filters had consistently larger  $\kappa$  than methanol extracts, while western sagebrush extract aerosols  $\kappa$  exceeded those from Alaskan duff core. HTDMA- and CCN-derived values of  $\kappa$  for each experiment agreed within approximately 20%. Applicability of the  $\kappa$ -parameterization to other multicomponent aerosols relevant to the atmosphere remains to be tested.

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### 1. Introduction

[2] Organic carbon (OC) compounds comprise a substantial fraction of the ambient aerosol in many locations both at the Earth's surface and aloft [Hegg *et al.*, 1997; Murphy *et al.*, 1998; Carrico *et al.*, 2003; Malm *et al.*, 2004; Heald *et al.*, 2006]. It is known that the organic fraction of the atmospheric aerosol is a diverse mixture of many compounds possessing a range of chemical properties, including hygroscopicity [Saxena and Hildemann, 1996; Hemming and Seinfeld, 2001]. Organic material is often found mixed in individual particles with inorganic components such as potassium, ammonium, sulfate, and nitrate [Hudson *et al.*, 2004]. Through impacts on the aerosol total mass and number concentrations, mean particle size, and chemical composition-dependent properties (e.g., density, refractive

index, surface tension, and hygroscopicity), the presence of OC influences the radiative and cloud-nucleating properties of the aerosol [Kanakidou *et al.*, 2005].

[3] Previous studies investigated the hygroscopic and cloud nucleating properties of OC-inorganic mixtures of representative model compounds [Cruz and Pandis, 1998, 2000; Hansson *et al.*, 1998; Raymond and Pandis, 2002; Prenni *et al.*, 2003; Brooks *et al.*, 2004; Abbatt *et al.*, 2005; Henning *et al.*, 2005; Svenningsson *et al.*, 2006]. These studies suggest that, within experimental uncertainties, the hygroscopicity of internal mixtures is well described by the hygroscopicity of individual compounds and their respective volume fractions [Petters and Kreidenweis, 2007] (abbreviated here *P&K*). In some mixtures, a reduction or elimination was observed of the clear deliquescent phase transitions that might be expected for the inorganic-species alone [Hansson *et al.*, 1998; Prenni *et al.*, 2003; Brooks *et al.*, 2004; Badger *et al.*, 2006]. Particles composed of these mixtures generally exhibited water uptake weaker than the pure salts, with the exception of several organic acids [Cruz and Pandis, 1997; Prenni *et al.*, 2003]. Recently, more complex mixtures generated from atomization of solid phase solution extracts from filter samples [Gysel *et al.*, 2004; Dinar *et al.*, 2006; Wex *et al.*, 2007] were found to

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have fractions that range in hygroscopicity from that typical of secondary organic aerosol to that of inorganic salts [Virkkula *et al.*, 1999; VanReken *et al.*, 2005; Prenni *et al.*, 2007]. Field studies of carbon-dominated ambient aerosols likewise often find that deliquescence transitions are smoothed and overall hygroscopic response is weaker than in inorganic-dominated aerosols [Aklilu and Mozurkewich, 2004; Carrico *et al.*, 2005; Malm *et al.*, 2005; Lee *et al.*, 2006].

[4] Subsaturated hygroscopic growth is quantified by the diameter growth factor ( $GF$ ) at a given RH and typically utilizes measurements with a hygroscopic tandem differential mobility analyzer [Rader and McMurry, 1986; Brechtel and Kreidenweis, 2000a, 2000b]. Hygroscopicity also determines the relationship between the particle dry diameter and the critical supersaturation,  $s_c$ , when a particle activates into a cloud droplet. This determines the cloud condensation nuclei (CCN) activity of an aerosol that can be measured with a streamwise gradient CCN counter [Roberts and Nenes, 2005]. Hygroscopicities inferred respectively from  $GF$  and CCN measurements are related by the underlying water activity ( $a_w$ ) versus composition relationship, though discrepancies between the estimates from each technique may occur because they access different compositional regions (i.e., the solutions formed in CCN measurements are much more dilute than those formed in HTDMA measurements). A number of parameterizations predicting CCN activity from  $GF$  data have been proposed [Svenningsson *et al.*, 1992, 2006; Weingartner *et al.*, 1997; Swietlicki *et al.*, 1999; Brechtel and Kreidenweis, 2000a; Kreidenweis *et al.*, 2005; Gasparini *et al.*, 2006; Rissler *et al.*, 2006; Wex *et al.*, 2007], and these demonstrate predictive ability for many, but not all, internal mixtures. Discrepancies, for example as observed for secondary organic aerosol [Prenni *et al.*, 2007], warrant further studies of the general applicability of parameterizations linking  $GF$  with CCN measurements.

[5] In this study, we measured the diameter growth factors and CCN behavior of particles composed of multi-component organic-inorganic mixtures. The aerosols were generated by atomization of extracts from quartz filters that were loaded with biomass smoke. Distinct aerosol compositions were produced using extractions with two solvents, ultrapure water and methanol. The consistency of hygroscopicity estimates as determined independently from  $GF$  and CCN data was examined using the hygroscopicity parameter,  $\kappa$  [P&K]. Though the aerosols tested here were derived from extractions of biomass smoke loaded filters, no attribution of these same behaviors is implied for ambient biomass smoke.

## 2. Experimental Design

### 2.1. Filter Extracts and Aerosol Generation

[6] During November 2003, a laboratory combustion study examining the physicochemical and optical properties of fresh smoke from select biomass fuels occurred at the U.S. Department of Agriculture/U.S. Forest Service Fire Science Laboratory in Missoula, Montana [Engling *et al.*, 2006]. The experiments involved open burning of a known mass of fuel, typically 100–300 g. Smoke was sampled during a 2-h period directly from the enclosed combustion

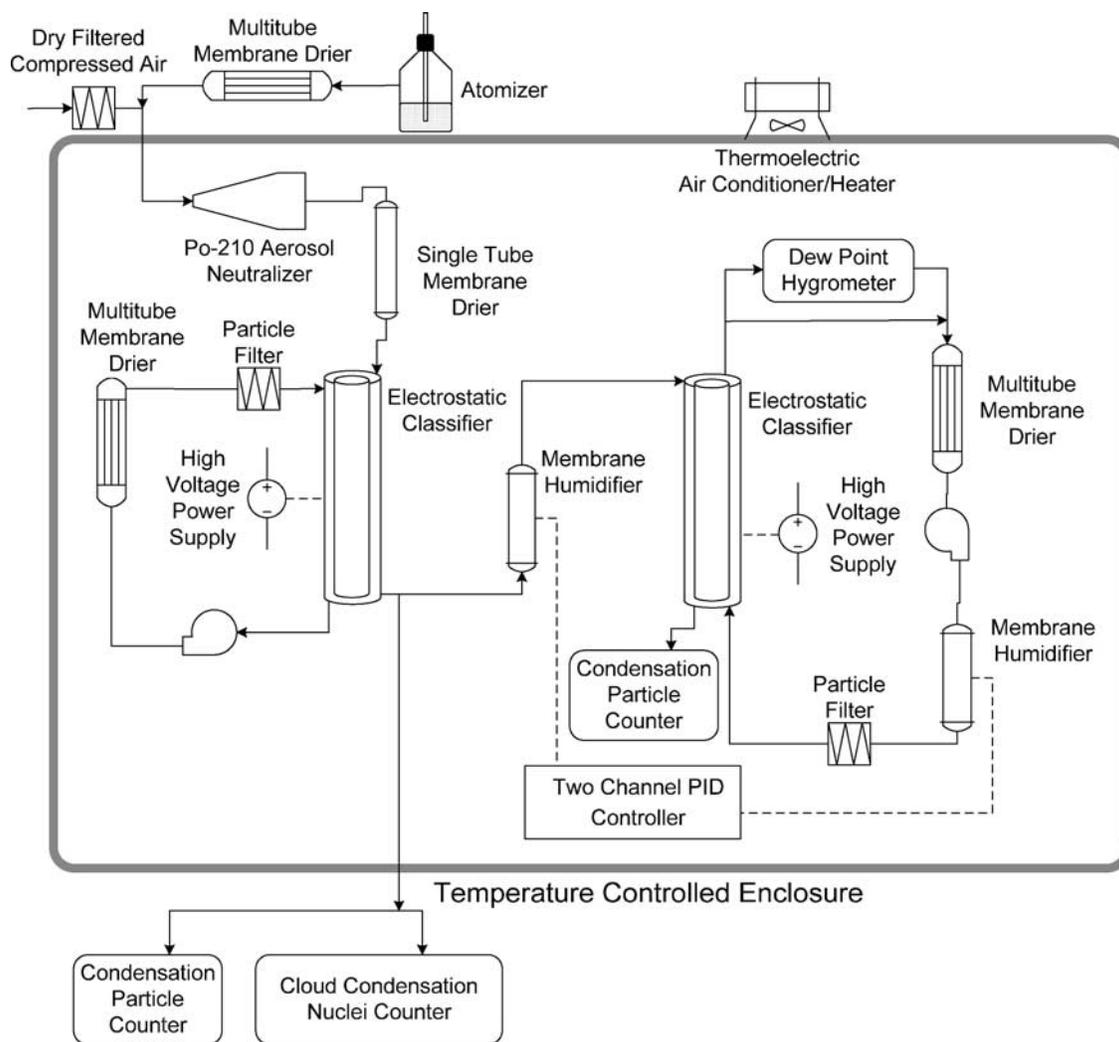
chamber measuring approximately 12.5 m  $\times$  12.5 m  $\times$  22 m high [Christian *et al.*, 2003]. PM<sub>2.5</sub> filter samples were collected on pre-baked quartz fiber filter substrates using high volume PM<sub>2.5</sub> samplers (Thermo Andersen, Inc. GI200-2.5 sampler using Whatman, Inc. QM-A filters prebaked at least 8 h at 600°C) [Engling *et al.*, 2006]. As is common practice, the filters were frozen after sampling and stored from November 2003 to August 2005 when portions of these filters approximately 5 cm  $\times$  5 cm were extracted into solution. Extractions involved submersion of the filter sections in an extraction solvent contained in pre-cleaned 1-liter glass laboratory flasks. The extraction solvent was 500 mL either of high purity methanol (Fisher Scientific, GC resolved methanol) or laboratory-generated ultrapure water (Barnstead, Inc. 18.2 M $\Omega$ -cm). The submerged filters were sonicated for two 15-minute periods in an ultrasonic bath.

[7] Next, aerosol was generated by atomization of the filter-extract solution (TSI Inc., Model 3076 atomizer). The aerosol sample was dried using three membrane driers in series (Permapure Inc., Model PD-100T) to RH < 5% (RH < 15% for water extract aerosols) as measured with a capacitive type sensor (Rotronic Inc., Hygroclip S). The membrane driers also served to selectively remove small polar organic vapor molecules, including methanol (www.permapure.com). Any residual methanol contained in the aerosol stream is thought to have been small, as discussed below in 3.1, but was not directly measured. Dry sheath air for the membrane filters and for subsequent aerosol dilution was provided by laboratory dry filtered air.

[8] Two smoke filter samples, Alaskan duff core and western sagebrush smokes, were selected as contrasts in fuel moisture, combustion phase, and chemical and hygroscopic properties [Chakrabarty *et al.*, 2006; Day *et al.*, 2006; Engling *et al.*, 2006]. Sagebrush (*Artemisia Tridentata*) was obtained from near Missoula, Montana and is a fuel common throughout the arid western U.S. lands, closely tied to fire ecology in western U.S. ecosystems [Obrist *et al.*, 2003]. Alaskan duff core is the peat-like material that collects on the forest floor in boreal forests and is composed of primarily leaves and twigs from the coniferous canopy and moss forming on the forest floor. It is a primary component of the fuel mixture in boreal forest fires [Kasischke *et al.*, 2005]. During combustion, higher moisture content duff core primarily smoldered whereas dry sagebrush had a dominant flaming phase, potentially contributing to the contrast in physicochemical properties.

[9] As validation of the measurement and analysis method, hygroscopic growth and CCN activation properties of single component aerosols were studied by atomizing pure salts dissolved in methanol or water solutions. The solutions were prepared by dissolving 0.2–0.5 g of reagent grade salts (NaCl and (NH<sub>4</sub>)<sub>2</sub>SO<sub>4</sub>, Fisher Scientific A.C.S. grade) in ultrapure methanol or water. Aerosol was generated and conditioned as described above.

[10] Previous work has examined comparisons of hygroscopic growth using  $\kappa$  for single component aerosols and simple mixtures [P&K]. The focus of this work is to test the inter-comparability of the two measurements of hygroscopic response for several multicomponent aerosols. The aerosols tested here were derived from extractions of biomass smoke loaded filters, though no attribution of these same behaviors



**Figure 1.** Schematic of the experimental setup.

is implied for ambient biomass smoke. Exploring the hygroscopic properties of ambient biomass smoke and the relationship to composition remains to be tested with more recent in situ measurements of biomass smoke.

## 2.2. HTDMA Measurements

[11] Measurements of aerosol hygroscopic growth were conducted with a newly constructed hygroscopic tandem differential mobility analyzer (HTDMA, Figure 1) [Rader and McMurry, 1986]. The HTDMA measures particle diameter growth factors under humidifying or dehumidifying conditions as a function of subsaturated relative humidity ( $RH < 95\%$ ) and is similar to configurations and protocols described previously [Brechtel and Kreidenweis, 2000b; Prenni et al., 2003; Carrico et al., 2005]. The aerosol sample was charge equilibrated with a neutralizer (Aerosol Dynamics Inc., ADI-100) containing four alpha-emitting  $^{210}\text{Po}$  strips (NRD, Inc. Staticmaster 2U500). The instrument selects a nearly monodisperse particle population with an electrical mobility classifier (TSI, Inc. Model 3081) and subsequently humidifies the sample before measuring with a second classifier interfaced to a condensation particle counter (TSI, Inc. Model 3010). The annular humidifiers

used on sheath and sample flows are custom built flow-through units constructed of stainless steel. They consist of a 0.0063 m OD aerosol sample flow separated by a water vapor-permeable membrane from a 0.0126 m OD liquid water jacket encased in a stainless steel shell. Humidity control is achieved by a dual-channel custom-built controller interfaced to resistance heaters on the water jacket. A sheath to sample flow ratio of 5:1 was used in both classifiers. These flow rates were chosen to facilitate humidity and flow control within the instrument in the size ranges of interest. They preserve equivalent flow ratios in each column as required for proper data inversion, though yield a broader transfer function than the often-used flow ratio of 10:1.

[12] HTDMA measurement and instrument control were conducted with a custom-built system interfaced to PC-based data acquisition software (National Instruments, Inc. Labview/Fieldpoint) and included voltage (Spellman, Inc. Model SL10N10), humidity (Rotronic, Inc. Hygroclip S), dew point temperature (Edgetech, Inc. Vigilant Series), dry bulb temperature (Instrulab, Inc. Model 3312A with thermistor probes), and pressure (Teledyne, Inc. Model 760S)

measurements. Relative humidity in the classifier column, used as the RH at which the GF data are reported, is calculated using the dew point temperature measurement at the outlet of the second classifier ( $\pm 0.2^\circ\text{C}$ ) and average dry bulb temperature from two thermistor probes ( $\pm 0.2^\circ\text{C}$ ) attached to the second electrostatic classifier column. We estimate the uncertainty in RH to be  $\pm 2\%$  at RH = 90%, similar to that found in previous studies [Carrico *et al.*, 2005]. The entire HTDMA measurement was conducted in a temperature-controlled enclosed chamber maintained at  $20 \pm 0.5^\circ\text{C}$  with a thermoelectric device (EIC Solutions, Inc. Model AAC-145-T-E-HC) (Figure 1).

[13] The second electrostatic classifier in the HTDMA measures a grown size distribution. The grown droplet diameter ( $D$ ) was obtained by fitting the size distribution to the TDMAFIT instrument transfer model [Stolzenburg and McMurry, 1988; Zhou *et al.*, 2002]. Measured hygroscopic growth factors ( $GF$ ) are taken as the ratio of the wet droplet diameter ( $D$ ) to the dry particle size ( $D_d$ ) such that  $GF = D(RH)/D_d$ . The uncertainty in the measurement of the growth factor using this technique is estimated as  $\pm 0.02$  in  $GF$  units [Carrico *et al.*, 2005].

### 2.3. CCN Measurements

[14] A portion of the quasi monodisperse aerosol selected with the first classifier in the HTDMA was delivered to a continuous flow streamwise gradient diffusion chamber used to determine the particle dry diameter-critical supersaturation relationships (Droplet Measurement Technologies, Inc., CCN-100 instrument, Figure 1, [Roberts and Nenes, 2005; Lance *et al.*, 2006]). The CCN counter supersaturation was calibrated with monodisperse  $(\text{NH}_4)_2\text{SO}_4$  particles [Petters *et al.*, 2006; Prenni *et al.*, 2007]. An optical particle counter placed at the outlet of the column measures the droplet size spectrum, and all droplets greater than  $D \sim 1 \mu\text{m}$  are counted as CCN. Final droplet diameters correlate with chamber supersaturation. For conditions here where supersaturations,  $s > 0.3\%$ , average droplet diameter exceeds  $3 \mu\text{m}$ , making it unlikely that many droplets remain too small for optical detection. Total condensation nuclei (CN) concentrations were measured in parallel (TSI, Inc. Model 3010 CPC). The ratio of CCN to CN concentration defines the droplet activation fraction as a function of supersaturation,  $s$ . Measured CCN response curves were fit to the following function:

$$f(x) = \frac{1}{2} \operatorname{erfc}\left(\frac{x}{\sqrt{2}}\right) \quad (1)$$

where  $x = (s - s_{50})/\sigma_s$ ,  $s_{50}$  is the supersaturation where 50% of the particles activate, and  $\sigma_s$  is the CCN response curve width. Multiply charged particles are present in the monodisperse aerosol stream exiting the first DMA. At the sizes investigated here (dry diameters  $< 120 \text{ nm}$ ), the fraction of multiply charged particles is insignificant and the aerosol is a monodispersion [Rader and McMurry, 1986]. The influence of multiplets would appear as a secondary peak preceding the main activation in a plot of the activated fraction versus dry diameter plot, which as illustrated later constitutes  $\sim 10\%$  or less of the total count.

[15] CCN measurements here used an “ $s$ -scan” where an aerosol population of a given dry diameter exiting from the

first classifier in the HTDMA was exposed to controlled supersaturation to determine critical supersaturations. Depending on the CCN activity observed in a given experiment, 8 to 20 supersaturation setpoints were selected in the range of  $0.1 < s < 1.5\%$ .

### 2.4. Linking HTDMA and CCN Measurements: The Hygroscopicity Parameter $\kappa$

[16] Both HTDMA and CCN data are fit to the model of  $P\&K$  to obtain a single hygroscopicity parameter,  $\kappa$ , for each data set. As reference, for pure compounds found in ambient aerosols,  $P\&K$  report a range of  $0.002 < \kappa < 0.4$  for select organic carbon compounds, while inorganic salts such as sodium chloride, ammonium sulfate and ammonium nitrate have  $0.5 < \kappa < 1.3$ , considering mean  $\kappa$  values derived from both HTDMA and CCN measurements. Here, we briefly review the concept behind this fitting.

[17] The saturation ratio,  $S$ , over an aqueous solution droplet is expressed in both sub- and supersaturated regimes by equation (2),

$$S = a_w \cdot \exp\left(\frac{4\sigma_{s/a}M_w}{\rho_wRTD}\right) \quad (2)$$

where  $\sigma_{s/a}$  is the solution surface tension ( $\sigma_{s/a} = 0.072 \text{ Jm}^{-2}$  for pure water is used here to produce a self consistent data set as suggested in  $P\&K$ ),  $M_w$  is the molecular weight of water,  $\rho_w$  is the density of water,  $R$  is the universal gas constant,  $T = 298.15 \text{ K}$  is the temperature, and  $D$  is the droplet diameter.  $P\&K$  suggest to parameterize water activity as follows:

$$a_w = \frac{D^3 - D_d^3}{D^3 - D_d^3(1 - \kappa)}, \quad (3)$$

where  $D_d$  is the dry particle diameter and  $\kappa$  is the hygroscopicity parameter. Combining (2) and (3) gives:

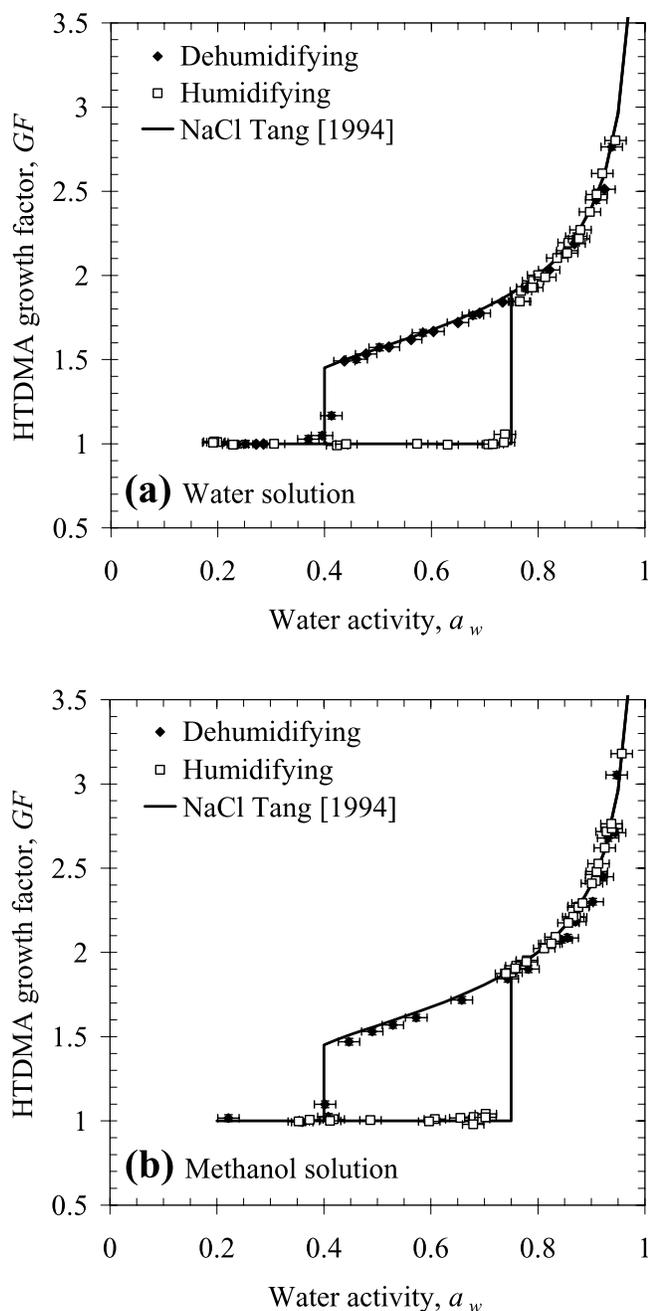
$$S(D) = \frac{D^3 - D_d^3}{D^3 - D_d^3(1 - \kappa)} \exp\left(\frac{\psi}{D}\right) \quad (4)$$

where  $\psi = 2.1 \times 10^{-9} \text{ m}$  is a constant. The critical saturation ratio for a selected dry diameter of a particle having hygroscopicity  $\kappa$  is computed from the maximum of the equation (4). To find  $\kappa$  for an observed pair of  $S_c$  and  $D_d$  we iteratively vary  $\kappa$  until the maximum in equation (4) is equal to the measured value of  $S_c$ . Our CCN calibrations use  $\kappa = 0.6$  for pure ammonium sulfate [Prenni *et al.*, 2007].

[18] HTDMA measurements yielded  $GF$ ,  $RH$  paired values which were converted to  $GF$ ,  $a_w$  pairs by dividing  $RH$  (expressed as a fraction) by the Kelvin term, the exponential term in equation (2). Growth factors, now given as  $GF$ ,  $a_w$  pairs are directly related to  $\kappa$  by rearranging equation (3), noting that  $GF = D/D_d$ , thus yielding:

$$\kappa = \frac{(GF^3 - 1)(1 - a_w)}{a_w}. \quad (5)$$

[19] For a given extract solvent-fuel combination,  $\kappa$  was calculated for each  $GF$ ,  $a_w$  pair from equation (5). The parameterization using  $\kappa$  suggested by  $P\&K$  is a fit to the



**Figure 2.** Comparison of HTDMA measured growth curves with theory for pure NaCl generated aerosol for dry size of 100 nm. Aerosol was generated in (a) water solution and (b) methanol solution. Uncertainties in  $GF$  ( $\pm 0.02$ ) are contained in the symbol size while uncertainties in  $a_w$  ( $\pm 0.02$ ) are shown as x-error bars.

data derived at  $a_w = 0.9$  for application to subsaturated aerosol growth. To include enough data points for inter-comparison, here we use data over the water activity range of  $0.85 < a_w < 0.95$ . Over this range,  $\kappa$  is nearly (though not exactly) constant for the aerosols generated in these experiments. At a specified  $a_w$ ,  $\kappa$  is a measure of the amount of bound water per unit volume of dry solute. For an ideal solution,  $\kappa$  is a constant depending only on the solute properties. Deviations from this idealized behavior cause

$\kappa$  to be a function of  $a_w$  causing discrepancies between sub- and supersaturated  $\kappa$ . However, as we show here, these deviations are sufficiently small to use  $\kappa$  from *P&K* to predict the particle behavior at different dilutions.

### 3. Results and Discussion

#### 3.1. Measurements With Laboratory-Generated Sodium Chloride and Ammonium Sulfate Particles

[20] To give assurance that the use of methanol did not confound the results, measurements of aerosols generated with sodium chloride dissolved in methanol were performed with both instruments. A dynamic shape factor of 1.08, corresponding to a cubic structure in a crystalline state, was used to correct the dry mobility equivalent diameter of the NaCl particles. The dynamic shape factor accounts for the increased drag of a nonspherical particle compared to that of a spherical particle with equivalent volume. In the transition regime, the slip flow correction factor is needed to convert the mobility equivalent diameter to the sphere equivalent diameter. For isometric particles, i.e., when the shape factor is independent of the orientation of the particle, the mobility equivalent diameter is adequate to approximate the slip coefficient of the nonspherical particle [*Kaspar, 1982*]. Since the slip flow correction factors of the mobility equivalent and volume equivalent diameters are not equal, the correction factor is  $\sim 1.04$  [cf. *Kreidenweis et al., 2005*] for particle sizes used in this study.

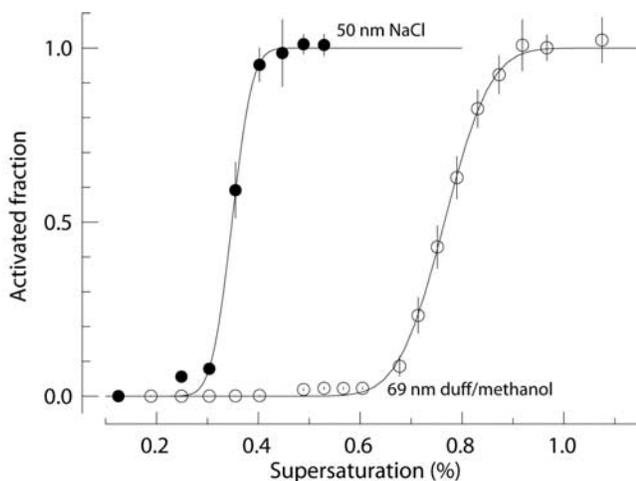
[21] For both methanol and water solutions of NaCl, measurements were completed for humidifying and dehumidifying conditions and show deliquescence and crystallization humidities of approximately  $a_w = 0.75$  and  $0.40$ , comparable to literature values [*Tang, 1980*] (Figure 2). Comparison of  $GF$  for NaCl particles generated from methanol and aqueous solutions are shown in comparison to literature data [*Tang and Munkelwitz, 1994*] (Figure 2). For  $(\text{NH}_4)_2\text{SO}_4$ , similar agreement with literature data was observed with deliquescence observed at  $a_w = 0.79$  and measured  $GF(a_w)$  as summarized in Table 1.

[22] As described above in the Methods section, CCN measurements with pure ammonium sulfate were used for instrument calibration. CCN measurements of the activated fraction as a function of supersaturation for NaCl particles, generated in methanol, with a dry diameter  $D_d = 50$  nm gave  $s_c = 0.35\%$  (Figure 3). A second experiment yielded  $s_c = 0.86\%$  for  $D_d = 26$  nm. For pure NaCl particles, *Kreidenweis et al.* [2005] report  $D_d = 51 \pm 3$  nm and  $27 \pm 2$  nm at  $s_c$  of  $0.3\%$  and  $0.8\%$ , respectively, from a range of published laboratory and modeling studies. The inferred values of the hygroscopicity parameter were  $\kappa = 1.13$  and  $\kappa = 1.34$ , respectively, for these two measurements of the methanol

**Table 1.** Comparison of HTDMA-Derived  $\kappa$  Values for Test Aerosol Generated From Pure Salts Dissolved in Methanol or Water Compared to Literature for These Same Inorganic Aerosols<sup>a</sup>

Test Aerosol	$GF$		HTDMA $\kappa$				
	( $a_w = 0.8$ )	( $a_w = 0.9$ )	Mean	Stdev	Min	Max	n
NaCl in water	1.99	2.40	1.414	0.104	1.218	1.595	14
NaCl in methanol	1.97	2.38	1.391	0.117	1.116	1.551	22
$(\text{NH}_4)_2\text{SO}_4$ in water	1.50	1.78	0.534	0.041	0.469	0.593	8
$(\text{NH}_4)_2\text{SO}_4$ in methanol	1.49	1.77	0.551	0.056	0.504	0.670	7

<sup>a</sup>Data are provided over the range  $0.85 < a_w < 0.95$ .



**Figure 3.** The measured activated fraction versus supersaturation measured with the CCN instrument for 50 nm pure NaCl and 69 nm duff/methanol extract aerosol. Vertical bars denote  $\pm$  one standard deviation of the [CCN]-to-[CN] ratio. The line is a best fit to the data based on equation (1).

generated NaCl particles. Using the Aerosol Inorganic Model, P&K report  $\kappa = 1.28$  for pure NaCl.

[23] We conclude that aerosol generation in methanol did not cause perceptible artifacts in sub- or supersaturated hygroscopic growth of these two salt solutions. Previous work has shown an impact from the atomization solvent on the atomized aerosol CCN properties, particularly those of organic compounds [Rissman *et al.*, 2007]. Despite our tests with known inorganic salts, such an effect cannot be unequivocally precluded here. However, rather than reporting the hygroscopic response of an ambient biomass smoke aerosol, the primary goal of this study is to examine closure between sub- and supersaturated growth of multicomponent aerosols representing a range of  $\kappa$  values.

### 3.2. Water Uptake of Extracted Filter Samples

[24] We next performed hygroscopic growth and cloud condensation nuclei measurements on aerosols generated from solvent extractions of smoke filter samples. Over the range  $0.85 < a_w < 0.95$ , arithmetic means and standard deviations of  $\kappa$  are calculated for comparison to the values measured with the CCN instrument as summarized in Table 2.

[25] For all four extracts, HTDMA  $GF$  measurements showed moderate, continuous water uptake beginning around  $a_w = 0.4$ . Subsaturated growth is characterized by a single growth mode, with little evidence of any sharp deliquescence transitions (Figure 4). The HTDMA hygroscopic growth results for  $a_w > 0.75$  are shown in Figure 4, where for a given

extract the point represents an individual HTDMA measurement of  $GF$  versus  $a_w$ . The plot represents measurements of  $GF(a_w)$  compiled over the entire range of dry diameters investigated ( $25 < D_d < 130$  nm). Superimposed are lines of constant hygroscopicity, corresponding to the minimum and maximum values of  $\kappa$  for HTDMA measurements as given in Table 2, with the gray-shaded area delineating intermediate values used for intercomparison with CCN measurements.

[26] Aerosols produced from extracts of duff core smoke samples were less hygroscopic than aerosols generated from sagebrush smoke extracts in the corresponding solvent. This was particularly evident for aqueous extracts, where HTDMA mean  $\kappa = 0.298$  for the sagebrush water extract versus 0.149 for duff core water extract (Figure 4 and Table 1). Methanol extracts followed the same trend though with lower hygroscopic growth,  $\kappa = 0.085$  versus 0.063 for sagebrush and duff core, respectively. As reference, in situ measurements of light scattering as a function of RH performed during the combustion experiments, Day *et al.* [2006] similarly showed that smoke from sagebrush had a stronger hygroscopic response than did smoke from duff core samples. The relative hygroscopicities are consistent with SEM imaging, which showed the dominance of organic carbon ‘tar balls’ (presumably less hygroscopic) in duff core smoke, whereas the sagebrush smoke samples contained sooty agglomerates and a greater prevalence of irregular and crystalline particles containing sulfur, chlorine, and potassium [Chakrabarty *et al.*, 2006].

[27] Comparing aerosols generated with each solvent extraction (water versus methanol) for a given fuel, substantially different hygroscopic behavior was observed (Figure 4). Non-hygroscopic organic compounds with long carbon chains and few functional groups are more soluble in methanol than in water and thus may be enriched in the methanol extracts. Inorganic compounds may be less soluble in methanol than water and thus may be enriched in the water extracts. However, solubility and hygroscopicity do not necessarily correlate. For example sodium chloride is not very soluble in water (36 g in 100 g water) but very hygroscopic ( $\kappa_{\text{ccn}} = 1.28$ ), while poly(acrylic acid) is infinitely soluble in water yet only weakly hygroscopic ( $\kappa_{\text{ccn}} = 0.05$ ). Thus it was unclear beforehand which extraction solvent should produce more hygroscopic particles. Here, we observed that particles generated from the methanol extracts exhibit lower growth factors than those generated from the water extracts.

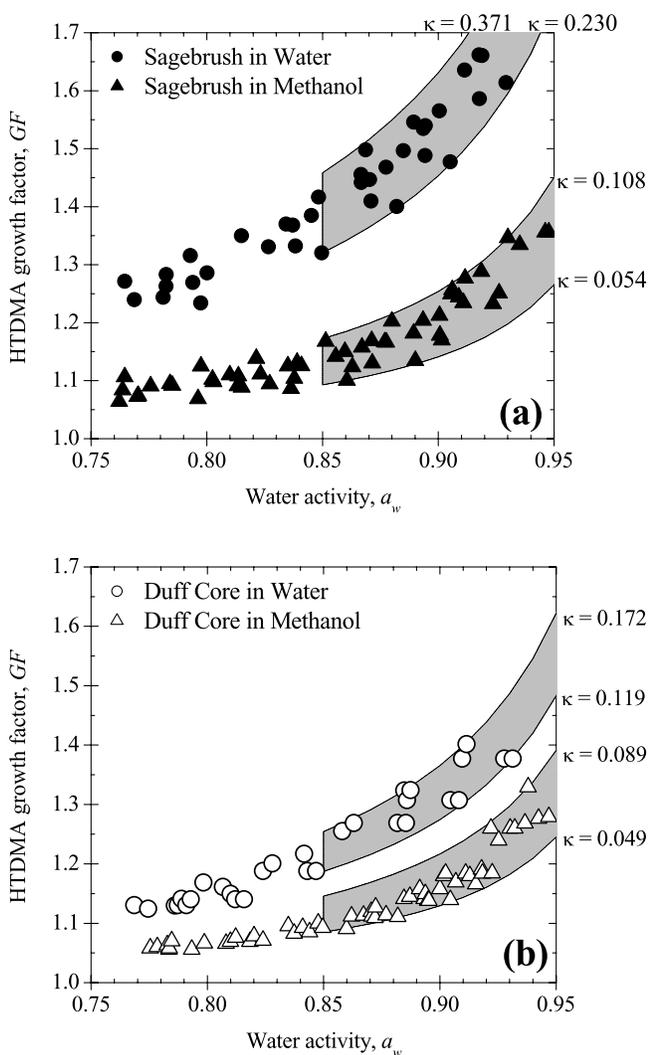
### 3.3. Comparison of CCN and HTDMA Measurements Using $\kappa$

[28] CCN measurements are shown as a plot of  $\log s_c$  versus  $\log D_d$  for aerosols generated from the four extracts (Figure 5). Superimposed are lines of constant  $\kappa$  plotted as

**Table 2.** Comparison of HTDMA-Derived and CCN-Measured  $\kappa$  Values for Water and Methanol Extraction of Filters From Duff Core and Sagebrush Burns<sup>a</sup>

Filter Extraction	$GF$		HTDMA $\kappa$					CCN $\kappa$				
	( $a_w = 0.8$ )	( $a_w = 0.9$ )	Mean	Stdev	Min	Max	n	Mean	Stdev	Min	Max	n
Sagebrush in water	1.30	1.56	0.298	0.040	0.230	0.371	21	0.252	0.049	0.206	0.327	5
Sagebrush in methanol	1.10	1.21	0.085	0.015	0.054	0.108	28	0.083	0.012	0.063	0.094	5
Duff core in water	1.16	1.36	0.149	0.020	0.119	0.172	13	0.148	0.004	0.145	0.151	2
Duff core in methanol	1.07	1.16	0.063	0.009	0.049	0.089	35	0.064	0.008	0.052	0.073	6

<sup>a</sup>Data are provided over the range  $0.85 < a_w < 0.95$ .



**Figure 4.** Hygroscopic growth curves for aerosol generated from water and methanol extracts of filter-deposited smoke from the combustion of sagebrush and duff core. Data points are growth factor ( $GF$ ) as a function of water activity ( $a_w$ ) from HTDMA measurements. Gray-shaded regions are the range of  $\kappa$  values found for HTDMA measurements as given by minimum and maximums in Table 1, excluding one outlier for sagebrush in methanol.

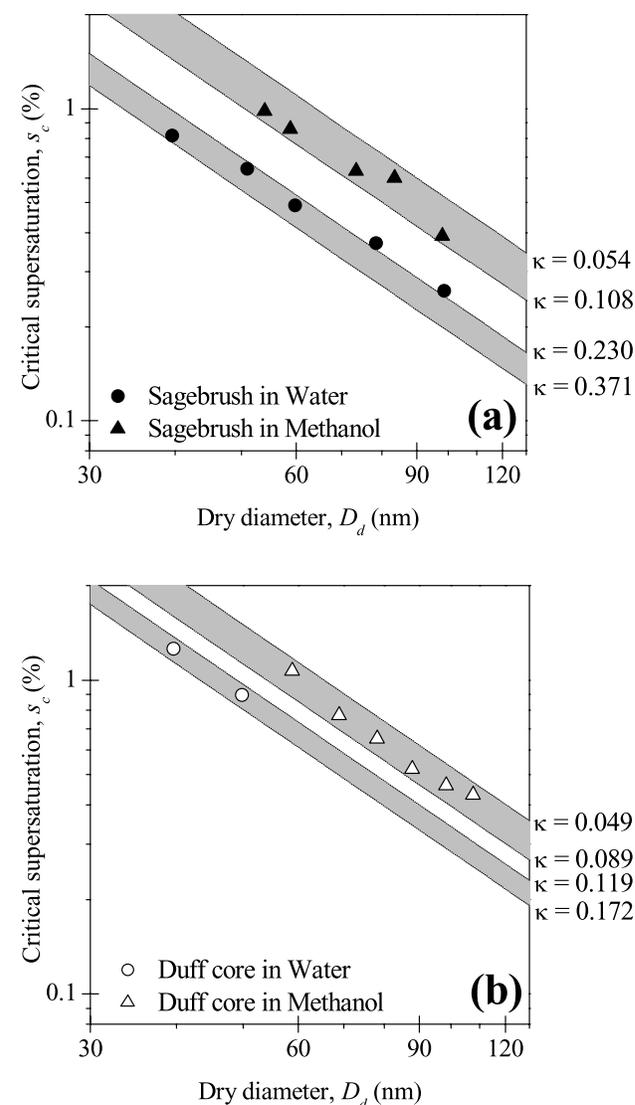
diagonal lines which form a power law with a slope of  $\sim -3/2$  [ $P&K$ ], along which the data generally fall. As before in Figure 4, the  $\kappa$  lines in Figure 5 represent the minimum-maximum range found from HTDMA measurements for each extract with intermediate values contained within the gray-shaded regions. Measurements of CCN activity of the four extracts followed the same hygroscopicity trend as HTDMA measurements, where  $\kappa$  for water extractions exceeded those for methanol extractions (Table 2). Likewise, for a given solvent, CCN measured  $\kappa$  for sagebrush extracts exceeded that of  $\kappa$  for duff core extracts. The latter trend is consistent with in situ measurements of aerosol light scattering as a function of RH as discussed previously [Day et al., 2006].

[29] Values of  $\kappa$  for CCN measurements for each extract are compared with corresponding HTDMA measurements

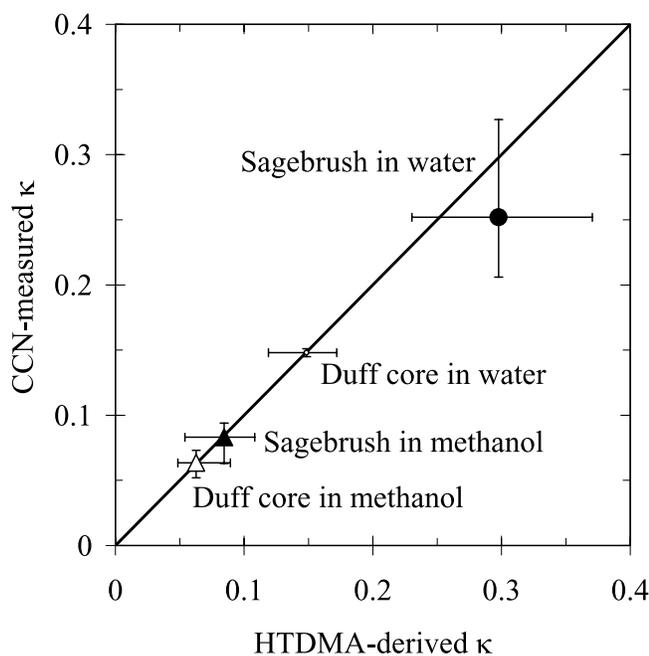
in Figure 6. Agreement for  $\kappa$  is within  $\pm 20\%$  for the four cases considered here. Applying  $\kappa$  derived from the HTDMA measurements to predict  $s_c$  yields a best fit with CCN measured  $s_c$  with linear least squares  $R^2 = 0.98$ , a slope of 1.08, and an intercept of 0.03 for  $n = 18$  individual data points for CCN  $s_c$  measurements.

#### 4. Summary and Conclusion

[30] We conducted a laboratory closure study to inter-compare water sub- and supersaturated hygroscopic growth properties of mixed component aerosols possessing a range of hygroscopic responses. The aerosols were produced from solvent extractions of biomass smoke collected on filters during previous laboratory biomass combustion experiments. Measurements of sub-saturated hygroscopic growth



**Figure 5.** Critical supersaturation ( $s_c$ ) values measured with the continuous flow diffusion chamber (datapoints) as a function of particle dry diameter ( $D_d$ ). Gray-shaded regions contain the range of  $\kappa$  values found for HTDMA measurements as given by minimum and maximums in Table 1, excluding one outlier for sagebrush in methanol.



**Figure 6.** Scatterplot comparison of mean hygroscopicity parameter ( $\kappa$ ) for HTDMA and CCN measurements. The error bars are the range of values from each data set, and the solid line is the one-to-one relationship.

using a hygroscopic tandem differential mobility analyzer were coupled with supersaturated-regime data from a continuous flow cloud condensation nucleus (CCN) counter. Comparisons of the two sets of measurements used a single parameter model that is quantified by the ‘hygroscopicity parameter,’  $\kappa$ .

[31] We first measured hygroscopicities of known aerosols, specifically pure NaCl and  $(\text{NH}_4)_2\text{SO}_4$  aerosols, for instrument calibration and verification. Next, two smoke filter samples were selected, that contrasted in terms of fuel moisture, combustion phase, smoke composition, and in situ hygroscopic properties: western sagebrush, typifying western U.S. shrublands, and Alaskan duff core typifying boreal forest floor materials. Aerosols were generated by extracting filter sections using either water or methanol and atomizing the resulting solutions.

[32] For the biomass smoke extracts, aerosols generated from water extracts of smoke samples were substantially more hygroscopic and CCN active as compared with those from methanol extracts. For the sagebrush case, means and standard deviations for the HTDMA  $\kappa = 0.298 \pm 0.040$  versus  $0.085 \pm 0.015$  for water and methanol extracts, respectively. As reference, corresponding diameter growth factors at RH = 90% were  $GF(90\%) = 1.56$  for a water extract versus 1.21 for the methanol extract of the sagebrush smoke.

[33] For either solvent extraction, the sagebrush smoke extracts exhibited stronger hygroscopicity than did the extracts of the duff core smoke samples. For example, for the water extract, HTDMA means and standard deviations were  $\kappa = 0.298 \pm 0.040$  versus  $0.149 \pm 0.020$ , for sagebrush and duff core, respectively. This result was consistent with separate measurements involving particle SEM imaging and in situ humidity-dependent scattering coefficient measurements of the fresh biomass smoke particles [Chakrabarty et

al., 2006; Day et al., 2006]. However, as we use a filter extraction method, we do not presume the same hygroscopic responses of these smokes in the ambient atmosphere.

[34] Using  $\kappa$ , these measurements show that sub- or supersaturated hygroscopic growth measurements can be used to predict the other within approximately 20% uncertainty. The successful closure between  $GF$  and CCN activity for these aerosols using a simple one parameter representation of hygroscopicity is encouraging for the future application of this technique to complex ambient aerosols. Further work is necessary to test the general applicability of the parameterizations to ambient smoke aerosols.

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