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Size-resolved aerosol composition and link to hygroscopicity at a forested site in Colorado

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Size-resolved aerosol composition
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Abstract

Aerosol hygroscopicity describes the ability of a particle to take up water and form a cloud droplet. Modeling studies have shown sensitivity of precipitation-producing cloud systems to the availability of aerosol particles capable of serving as cloud condensation nuclei (CCN), and hygroscopicity is a key parameter controlling the number of available CCN. Continental aerosol is typically assumed to have a representative hygroscopicity parameter, κ, of 0.3; however, in remote locations this value can be lower due to relatively large mass fractions of organic components. To further our understanding of aerosol properties in remote areas, we measured size-resolved aerosol chemical composition and hygroscopicity in a forested, mountainous site in Colorado during the six-week BEACHON-RoMBAS campaign. This campaign followed a year-long measurement period at this site, and results from the intensive campaign shed light on the previously reported seasonal cycle in aerosol hygroscopicity. New particle formation events were observed routinely at this site and nucleation mode composition measurements indicated that the newly formed particles were predominantly organic. These events likely contribute to the dominance of organic species at smaller sizes, where aerosol organic mass fractions of non-refractory components were between 70–90 %. Corresponding aerosol hygroscopicity was observed to range from $\kappa = 0.15–0.22$, with hygroscopicity increasing with particle size. Aerosol chemical composition measured by an Aerosol Mass Spectrometer and calculated from hygroscopicity measurements agreed very well during the intensive study with an assumed value of $\kappa_{\text{org}} = 0.13$ resulting in the best agreement.

1 Introduction

Whether or not a particle will act as a cloud condensation nucleus (CCN) at a given supersaturation depends on the particle's size and hygroscopicity, the latter referring to how readily the particle takes up water and grows to droplet size. For particles in

the accumulation mode, where the majority of atmospheric particle number concentrations are typically found, aerosol hygroscopicity can have large impacts on the number of available CCN (Petters and Kreidenweis, 2007), which in turn can impact cloud droplet number and size. Higher droplet number concentrations are expected to cause
5 changes in both cloud structure and lifetime, leading to enhancements in cloud reflectivity (Twomey, 1974), suppression of drizzle (Albrecht, 1989), changes in intensity and distribution of precipitation (Rosenfeld et al., 2008; Khain et al., 2005; Khain and Lynn, 2009; Storer et al., 2010; van den Heever et al., 2006, 2011; van den Heever and Cotton, 2007) and even changes in updraft and storm intensity (Lerach and Cotton, 2012).
10 It is thus essential to accurately represent the number and characteristics of CCN to model these important processes.

Hygroscopicity can be described by the hygroscopicity parameter, κ (Petters and Kreidenweis, 2007). Typical atmospheric aerosol components can be divided into three broad categories based on hygroscopicity. Inorganic salts, such as ammonium sulfate,
15 ammonium nitrate and sodium chloride, are highly hygroscopic with κ values ranging from ~ 0.6–1.2 (Petters and Kreidenweis, 2007). Laboratory studies of secondary organic aerosol (SOA) species have found κ values around ~ 0.1 (Prenni et al., 2007; Engelhart et al., 2008, 2011; Wex et al., 2009) although individual species, when separated by polarity, show a larger range (Suda et al., 2012). Black carbon and dust are
20 typically measured or assumed to have κ values of ~ 0–0.05 (Koehler et al., 2009; Yamashita et al., 2011). As a representative average value for the fine mode aerosol that dominates CCN number concentrations, continental aerosol particles are often assumed to be adequately represented by a hygroscopicity parameter of ~ 0.3 (Andreae and Rosenfeld, 2008; Pringle et al., 2010; Pöschl et al., 2009). However, in remote
25 locations, far from sources of anthropogenic sulfate and nitrate, organic species may represent the dominant fraction of the aerosol, leading to lower κ values (Gunthe et al., 2009). Biogenic secondary organic aerosol (BSOA) is of particular interest in remote regions where oxidized volatile organic compounds, emitted from biogenic sources, play a key role in aerosol formation. These oxidized biogenic compounds can condense

onto existing particles or contribute to new particle formation and growth (Kulmala et al., 2004a, b; Dusek et al., 2010) and have been shown to be a major source of organic aerosol mass (Kanakidou et al., 2005; Robinson et al., 2011).

Given the global significance of organic aerosol, particularly of biogenic origin, many laboratory studies have examined the hygroscopicity of various model BSOA species (Prenni et al., 2007; Petters et al., 2009b; King et al., 2010; Asa-Awuku et al., 2010; Massoli et al., 2010; Engelhart et al., 2011) while field studies have measured aerosol hygroscopicity in areas dominated by BSOA (Cerully et al., 2011; Gunthe et al., 2009; Paramonov et al., 2013; Levin et al., 2012; Sihto et al., 2011). These studies have consistently found average κ values below 0.3, as well as variability in aerosol hygroscopicity with size, the latter feature likely due to the differing sources and gas-to-particle conversion processes of the organic and inorganic aerosol components. For example, Levin et al. (2012) measured aerosol hygroscopicity ($14\,nm < D_p < 350\,nm$) from March 2010–May 2011 at a forested, mountainous site in Colorado as part of the on-going Bio-hydro-atmosphere interactions of Energy, Aerosols, Carbon, H_2O, Organics and Nitrogen (BEACHON) project. Although representative of a semi-arid, mountain ecosystem, the average κ value determined for the measured aerosol was 0.16 ± 0.08, similar to values measured in highly biologically active areas such as Amazonian (Gunthe et al., 2009; Pöschl et al., 2010) and boreal forests (Sihto et al., 2011). Levin et al. (2012) hypothesized that the low hygroscopicity measured during the BEACHON annual cycle period was due to a large mass fraction of secondary organic species, likely of biogenic origin, in the aerosol. However, chemical composition measurements were not available during the year-long BEACHON study. In this work, we report on size-resolved CCN concentrations measured during the subsequent Rocky Mountain Biogenic Aerosol Study (BEACHON-RoMBAS), and we compare our results to concurrent measurements of size-resolved aerosol chemical composition.

2 Methods

2.1 Measurement site and instrumentation

Measurements were made during two phases of the BEACHON project: from March 2010–May 2011 to study a full seasonal cycle, as previously reported (Levin et al., 2012) and during the BEACHON-RoMBAS intensive study which took place during July and August 2011. The measurement site for both periods was located in Manitou Experimental Forest (lat 39.10° N, lon 105.10° W, elevation 2370 m). Kim et al. (2010) and Levin et al. (2012) describe the site as well as typical transport conditions for air masses arriving at the site.

During both the year-long and BEACHON-RoMBAS campaigns, we made measurements of size-resolved CCN concentrations at 5 different supersaturation (s = RH − 100 %) conditions and used these data to calculate aerosol hygroscopicity following Petters et al. (2009a). The specific details of the instrument setup and data collection and processing were reported in Levin et al. (2012). Briefly, we used a differential mobility analyzer (DMA; TSI 3081) to select particles in 20 different size bins between ∼ 14 and 350 nm in diameter and then sent the quasi-monodisperse sample to a condensation particle counter (CPC; TSI 3010) to count particle number concentration and to a cloud condensation nucleus counter (CCNC; Droplet Measurement Technologies) to determine the number of particles that act as CCN under the given s conditions. This technique provides number size distributions of total aerosol and CCN concentrations, at a specified s, with ∼ 15 min temporal resolution. We stepped s inside the CCNC through 5 different set points between 0.15 and 0.95 %. Supersaturation at each CCNC instrument setting was determined via ammonium sulfate calibrations using the thermodynamic properties of ammonium sulfate aqueous solutions as prescribed by the Aerosol Inorganics Model (Clegg et al., 1998). Calibrations at each s setting were performed about once a day. Activated fractions were calculated from the size-resolved total aerosol and CCN data. The critical activation diameter (d_c), the diameter at which the aerosol was able to take up water and grow to cloud droplet size

at a given supersaturation, was defined as the diameter where the activated fraction = 0.5. Finally, we used the CCNC s setting and the d_c to determine the best-fit aerosol hygroscopicity parameter, κ.

During BEACHON-RoMBAS, sub-micron aerosol composition was measured with a High-Resolution Time-of-Flight Aerosol Mass Spectrometer (AMS, Aerodyne Research Inc.) (Canagaratna et al., 2007; DeCarlo et al., 2006; Drewnick et al., 2005). The AMS measures non-refractory material, which includes most organic species and inorganic salts other than NaCl, but not crustal material or black carbon. While the instrument nominally measures PM_1 it is most sensitive to particles with diameters between 35 and 600 nm (Zhang et al., 2004). The AMS was calibrated with size-selected ammonium nitrate every four days during the study. Following DeCarlo et al. (2006) we calculated lower detection limits for daily averaged data of 10 and 3 ng m^{-3} for organic and sulfate aerosol, respectively.

Nano-particle composition was also measured with a Thermal Desorption Chemical Ionization Mass Spectrometer (TDCIMS) (Smith et al., 2004; Voisin et al., 2003). The TDCIMS first charges the aerosol and then uses a nano-DMA (TSI 3085) to size select particles. The size-selected particles are then collected onto a charged wire via electrostatic precipitation. Once a suitable mass of aerosol has been collected, the wire is inserted into the chemical ionization mass spectrometer and gradually heated from room temperature to $\sim 600\,°C$, thus desorbing the sample. For the current study, the volatilized sample was then chemically ionized and detected with a high resolution time-of-flight mass spectrometer (HTOF, Tofwerk AG).

2.2 Aerosol composition and hygroscopicity

The κ value of a mixed-composition particle depends on its chemical constituents and can be calculated from volume-weighted aerosol composition (Petters and Kreiden-

weis, 2007)

$$\kappa = \sum_i \varepsilon_i \kappa_i \qquad (1)$$

where κ_i and ε_i are the hygroscopicity parameter and volume fraction of the individual components. For the Rocky Mountain region during the summer months, SOA and inorganic salts have been shown to be the main components of $PM_{2.5}$ (Levin et al., 2009). While black carbon can be a significant contributor to aerosol mass during the wildfire season in the western US (Park et al., 2007), during BEACHON-RoMBAS black carbon measured with a single particle soot photometer (SP2; Droplet Measurement Technologies) accounted for less than 5 % of fine mode aerosol mass (A. Aiken, personal communication, 2013), similar to values reported by Levin et al. (2009) during a non-smoke impacted time period in Rocky Mountain National Park. Dust particles may also be present in the aerosol, particularly during spring. However, dust particles typically have diameters larger than the 350 nm upper limit used here (Malm et al., 2009). Thus, for this study we assume that the accumulation mode aerosol was composed mainly of SOA and hygroscopic inorganic salts, predominantly ammonium sulfate. Gunthe et al. (2009) employed similar assumptions for field measurements made in the Amazon Basin during the rainy season and found good agreement between κ determined from size-resolved CCN measurements and calculated from AMS chemical composition measurements. In this work we follow a similar approach to compare two-component κ values calculated from chemical composition to κ determined from measured CCN activity. We also expand this analysis to show that a representative two component aerosol composition can be derived from κ and use the year-long BEACHON κ measurements to deduce seasonal changes in aerosol composition.

3 Results and discussion

3.1 Aerosol hygroscopicity

Figure 1 shows daily average κ values (colored circles) and standard deviations during BEACHON-RoMBAS at the five different CCNC s settings. Study averaged values (± 1 standard deviation) are also reported in Fig. 1. Overall, aerosol hygroscopicity was low during this study, more similar to values reported for SOA than ammonium sulfate, and, on average, below the typically assumed value of $\kappa_{continental} = 0.3$. Only at the lowest s value did κ values reach 0.3 during a few time periods. There was also little variation in κ during the study period, especially at higher s conditions. The values determined during the BEACHON-RoMBAS summer intensive were slightly higher than those measured during the summer months of the year-long BEACHON study (± 1 standard deviation of July/August means shown by gray shading in Fig. 1). However, averaged over the BEACHON-RoMBAS study, κ values at every supersaturation setting fell within 1 standard deviation of the values from the previous year.

The right axis of Fig. 1 shows activation diameters corresponding to the left axis κ values for each s setting. At lower s, particles of similar hygroscopicity must be larger to activate as CCN. Thus, by operating at five different s settings we are probing the hygroscopicity of particles at different diameters. While κ was low at all s settings, there was a small increase in κ with decreasing s, corresponding to increasing activation diameter. That is, there was an apparent change in aerosol composition with size, with particles larger than $\sim 100\,\text{nm}$ containing a larger fraction of more hygroscopic material, similar to what was observed during the year-long study (Levin et al., 2012). We must also note that the κ values presented here only represent the aerosol near the activation diameters. Particles larger or smaller than this range could have different chemical composition, and thus different κ.

3.2 Aerosol composition

Levin et al. (2012) suggested that the aerosol measured during the year-long BEA-CHON study was predominantly organic, based on the low κ values, but there were no direct chemical measurements available to corroborate this claim. The similarly low κ values determined during BEACHON-RoMBAS also suggest a highly organic aerosol. During BEACHON-RoMBAS, sub-micron aerosol chemical composition was measured with the AMS and TDCIMS. Contour plots of daily averaged organic and inorganic aerosol mass distributions determined from AMS measurements are shown in Fig. 2 and the study averaged organic mass fraction (X_{org}) as a function of particle diameter in Fig. 3. To compare AMS results with those from the CCNC system we converted vacuum aerodynamic diameter, measured by the AMS, to mobility diameter by dividing by the study averaged aerosol density, using the assumed values for the organic and inorganic fractions discussed below. Orange shading in Fig. 3 indicates the range in AMS X_{org} due to instrument uncertainty (Bahreini et al., 2009). Although the AMS can speciate a number of organic and inorganic compounds, we have binned the data into these two components to compare with the two component composition which can be estimated from size-resolved CCNC measurements. The AMS data indicate that the inorganic mass concentration consisted primarily of ammonium sulfate, calculated as the measured SO_4^{2-} mass multiplied by 1.37 assuming that all sulfate was in the form of $(NH_4)_2SO_4$. This method was used rather than using the AMS ammonium measurements since the precision of sulfate measurements was much higher than for ammonium in the particle time-of-flight mode. While acidic forms of sulfate could exist in the aerosol, AMS measurements, using the higher precision "mass spec" mode, indicated sufficient NH_4^+ mass to fully neutralize the aerosol during BEACHON-RoMBAS. Nitrate mass was extremely low, often below the detection limit, and when present was associated with organic nitrate aerosol (Fry et al., 2013). Other inorganic species were negligible in the AMS measurements during BEACHON-RoMBAS. As expected, the organic fraction was the dominant component of the aerosol throughout

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the study, with a few periods of increased inorganic mass concentration. Averaged over the whole study (Fig. 3), organics were the main aerosol component at all sizes with their relative contribution increasing at smaller sizes.

Extending composition measurements to smaller sizes, the TDCIMS provided composition data for particles 30–50 nm in diameter. These data are also shown in Fig. 3 (blue bar) with the width of the bar representing the range in X_{org} for a range of assumed organic aerosol molecular weights (100–500 g mol^{-1}). These data are consistent with the observed trends from the AMS, showing that these small particles were predominantly organic. Organic compounds appear to play a critical role in the growth of nucleation mode particles to CCN sizes (Dusek et al., 2010). The role of organic compounds in growth of small particles was supported by observations in the year-long BEACHON study, in which aerosol hygroscopicity appeared to be affected by shifts in size distributions typical of new particle formation (NPF) events, with κ over most of the measured size range decreasing as the newly formed particles grew to sizes large enough to activate as CCN (Levin et al., 2012). While newly-formed, organic particles dominated at small sizes, AMS measurements showed an increasing inorganic fraction at larger aerosol diameters. Given this enhanced inorganic fraction and the lack of local sulfate emissions, it is most likely that the inorganic aerosol was more aged and may have undergone some cloud processing (Fors et al., 2011; Hao et al., 2013).

3.3 Kappa composition closure

To compare the AMS measurements with those from the size-resolved CCNC, we calculated organic volume fractions (ε_{org}) from measured κ values by rearranging Eq. (1):

$$\varepsilon_{org} = \frac{(\kappa - \kappa_{inorg})}{(\kappa_{org} - \kappa_{inorg})} \qquad (2)$$

To convert from volume fractions to mass fractions, we assumed densities of 1.4 (Bahreini et al., 2005; Kroll and Seinfeld, 2005; Poulain et al., 2010) and 1.8 g cm^{-3}

(Tang, 1996) for the organic and inorganic fractions, respectively. We used a κ_{inorg} value of 0.6, assuming the inorganic aerosol fraction was predominately ammonium sulfate, and allowed κ_{org} to vary from 0 to 0.15. Studies have shown that κ of more oxidized secondary organic species can be as high as 0.22 (Chang et al., 2010). However, the lowest average κ value determined during BEACHON-RoMBAS was only 0.15, and so we use this value as an upper limit for the organic aerosol fraction. Assuming a higher value would lead to a physically meaningless, negative inorganic mass fraction, given the two component system. We also assumed that the range in κ_{org} did not change over the course of the study or as a function of size. Figure 3 shows the study-averaged organic mass fraction (X_{org}) calculated from κ as a function of study-averaged critical activation diameter. The gray shaded region shows the range in X_{org} resulting from the range in assumed κ_{org} values (0–0.15). Regardless of the assumption made about the hygroscopicity of the organic material during the study, Fig. 3 clearly shows that the organic fraction dominated aerosol mass at all measured sizes. There was also a decrease in X_{org} with increasing activation diameter, as expected from the higher average κ values at these sizes.

Averaged over the whole study, the best agreement (minimum χ^2) between X_{org} determined from κ values and AMS measurements was for a κ_{org} value of 0.13 (black line in Fig. 3). In the diameter range where the CCNC and AMS measurements overlap, the agreement in organic mass fraction using this value of κ_{org} is very good, indicating little size dependence in κ_{org} or any of the other assumptions inherent in these calculations. Further, the trend of decreasing X_{org} with increasing size is very similar between the two measurements.

We used the best fit value of size-independent $\kappa_{org} = 0.13$ as well as the other assumptions for κ_{inorg} and aerosol density discussed above to calculate daily averaged κ distributions from the AMS derived organic and inorganic mass fractions (κ_{AMS}). These values are shown in Fig. 4. We then selected the κ_{AMS} values at the diameters corresponding to the d_c for each CCNC s setting and plotted these in Fig. 1 to compare

with κ derived from CCNC measurements (κ_{CCNC}). The error bars for κ_{AMS} in Fig. 1 represent AMS uncertainty.

Overall, there is good agreement between κ_{AMS} and κ_{CCNC}. At the lowest s, largest d_c, there is some difference in κ at the beginning of the study, but then good agreement

5 after this time period. During the early time period (7/18–7/22), AMS measurements indicated an increased inorganic mass concentration, and thus higher κ, at sizes slightly larger than the largest CCNC d_c ($\sim 150\,nm$). There was also a sharp gradient in inorganic mass concentrations between 150 and 250 nm. The higher κ_{CCNC} values during this time period could be affected by larger, multiply charged particles appearing in

10 smaller DMA size bins. Although we correct CN and CCN number distributions for multiply charged particles (Petters et al., 2009a), size-dependent chemical differences in the aerosol are not accounted for in the inversion and could be impacting measured κ values.

From Fig. 2 it can be seen that the κ_{AMS} values shown in Fig. 1 were calculated at

15 diameters where mass loadings were very low, especially at the smaller d_c values. At smaller sizes, this resulted in more variable κ_{AMS} values, and in many cases we were unable to calculate κ_{AMS} because mass was below the AMS detection limit. At larger diameters, above the measureable range for the CCNC system, the AMS values are likely more certain, due to the higher mass concentrations at these sizes. In contrast,

20 the CCNC is a number-based measurement, and is thus most reliable at diameters with the highest number concentrations, so that comparing results from these two systems is challenging. Nevertheless, the general agreement between X_{org} and κ determined from these two measurement systems in this case supports the use of the AMS data to calculate hygroscopicity and the size-resolved CCNC data to calculate two component

25 aerosol composition. Further, while the AMS can only detect non-refractory particles, it appears to capture the important species for determining aerosol hygroscopicity in this size range at this site.

3.4 Seasonal aerosol composition

Based on the generally good agreement between aerosol composition measured by the AMS and calculated from CCNC data, we used the year-long CCNC data reported by Levin et al. (2012), and the same method applied to the BEACHON-RoMBAS data, to calculate monthly averaged, two-component aerosol composition distributions for a full year. These distributions are shown in Fig. 5 with the green and blue bars showing the organic and inorganic components, respectively. As before, the gray shaded region shows the range in calculated organic mass fraction for assumed κ_{org} values. However, the range in assumed κ_{org} had to be limited to $0 < \kappa_{org} < 0.12$ for the year-long data set to avoid a negative calculated X_{inorg} value during some months. Since a best fit value of $\kappa_{org} = 0.13$ was observed during BEACHON-RoMBAS this could indicate some seasonal changes in the composition of the organic aerosol at this site or the influence of other low κ aerosol components, such as dust or black carbon.

We observed a similar trend throughout the year-long study period as seen during the intensive BEACHON-RoMBAS campaign, with aerosol organic fraction decreasing with increasing diameter. There was, however, some seasonality in this pattern. Aerosol composition was least size-dependent during the late summer and early fall and then the variability with particle diameter steadily increased throughout the winter and into the spring. The largest change in organic fraction with size was observed during April, when X_{org} decreased to ~ 0.5 at the largest measured size. The seasonal change in X_{org} for particles at the largest measured size is consistent with the seasonal variability of volatile organic compound (VOC) emissions (Saarikoski et al., 2008), which are precursors for SOA production. Likewise, while NPF events occur year round, they were most common in the summer months at Manitou Experimental Forest (Levin et al., 2012), and could be a contributing factor to the increased X_{org} values during this time period. Less seasonality was observed in the inferred composition of the smallest measured particles, which remained almost entirely organic throughout the year-long study period.

4 Summary and conclusions

Atmospheric particles that act as CCN are important in controlling cloud and precipitation formation and lifetime, and can have considerable effects from local to global scales. High quality measurements of CCN concentration and aerosol hygroscopicity are needed to constrain modeled cloud and precipitation processes, and explore their subsequent impacts on water cycles and radiative transfer. In this paper we present aerosol composition and hygroscopicity data from a forested site in the eastern Rocky Mountains of Colorado made as part of BEACHON-RoMBAS. We used size-resolved CCN measurements to determine aerosol hygroscopicity and measured aerosol chemical composition with an AMS and TDCIMS and found consistency between measured hygroscopicity and aerosol composition. Likewise, we showed that CCN-derived κ values can be used to calculate aerosol composition, assuming conditions are met for a simple two-component system composed of organic aerosol of size-independent κ and an inorganic aerosol dominated by sulfate. Over the intensive summer study the best agreement between measured and calculated aerosol composition was achieved for an assumed value of $\kappa_{org} = 0.13$.

Aerosol chemical composition and hygroscopicity measurements showed that the aerosol measured at the BEACHON site was predominantly organic, and thus had low κ values. Averaged over the entire year-long BEACHON study, and all s settings, κ had an average value of 0.16 ± 0.08, similar to values determined in biogenic organic dominated locations such as a tropical forest in the Amazon during the wet season (Gunthe et al., 2009; Pöschl et al., 2010) and a boreal forest in Finland (Sihto et al., 2011), and lower than the commonly assumed value of $\kappa_{continental} = 0.3$. The relatively low κ values measured at these locations dominated by biogenic emissions suggest a predominance and potential importance of organic species, which generally have low hygroscopicity (Prenni et al., 2007; Petters et al., 2009b; King et al., 2010). It is known that highly biologically active areas such as boreal and tropical forests will have large contributions from organic aerosol leading to low κ values. However, results from this

study indicate that the aerosol in this Colorado, semi-arid, high altitude location is also dominated by organic species.

While organic species were dominant at all sizes, there were some changes in aerosol composition with size, with the organic fraction typically decreasing at larger diameters. The increased inorganic fraction of larger particles likely indicates that these particles were more aged and had likely undergone some cloud processing. The trend of decreasing organic fraction with larger diameters was observed year round, however, there was some seasonality observed during the year-long BEACHON study. During the winter months, there was a sharper decrease in calculated organic mass fraction with size, although even at the largest diameter ($\sim$ 150 nm) the aerosol was still predominantly organic. During the summer months, there was less size dependence in composition. We postulate that the increased organic fraction at larger diameters during the summer is due to increased biological activity during this time period, resulting in increased emissions of BVOCs, precursors of organic aerosol.

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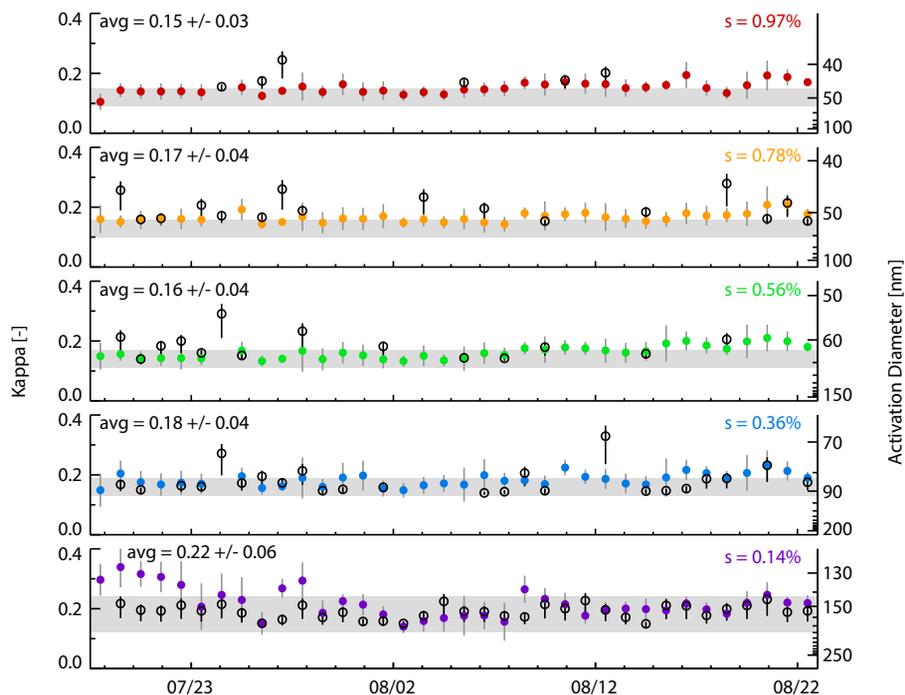
Fig. 1. Daily averaged κ values at each supersaturation setting during BEACHON-RoMBAS (colored circles) as well as calculated κ from AMS measurements (black circles). Gray bars show ± one standard deviation for measured κ values from July–August 2011, collected during the year-long BEACHON study. Right axis indicates activation diameter corresponding to the left axis κ values at the indicated supersaturation.
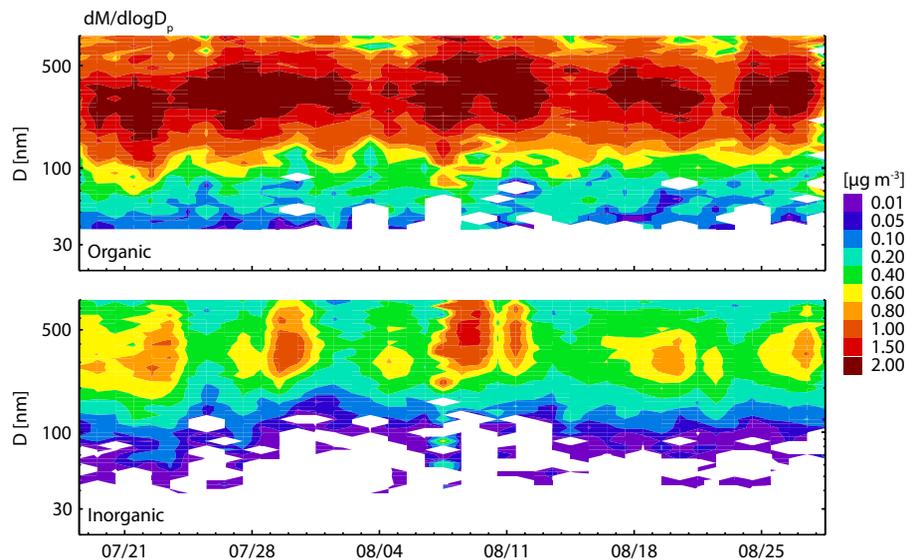
Fig. 2. Organic (top) and inorganic (bottom) mass distributions measured by the AMS during BEACHON-RoMBAS.

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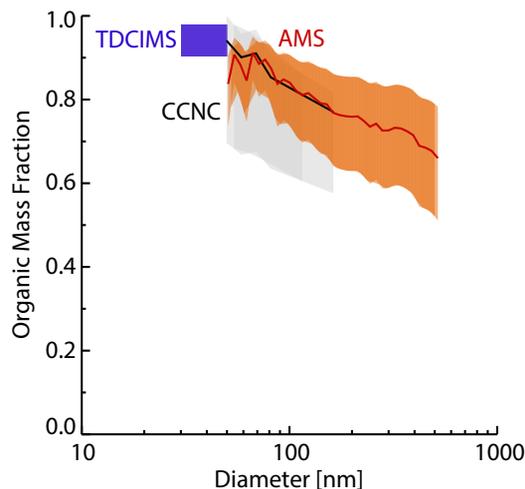
Fig. 3. Aerosol organic mass fraction (X_{org}) measured by an AMS (red) and TDCIMS (blue) as well as organic mass fraction calculated from size-resolved CCN measurements (black). Gray shading represents the range in X_{org} derived from CCNC data for values of $\kappa_{org} = 0$–0.15. Orange shading indicates AMS measurement uncertainty. The range in TDCIMS derived X_{org} reflects the uncertainty in the molecular weight of the organic aerosol.
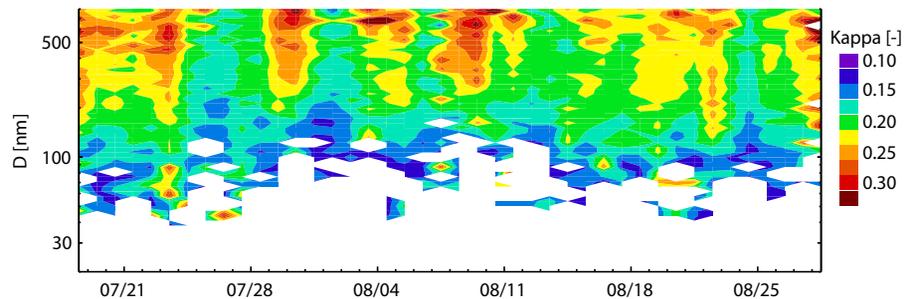
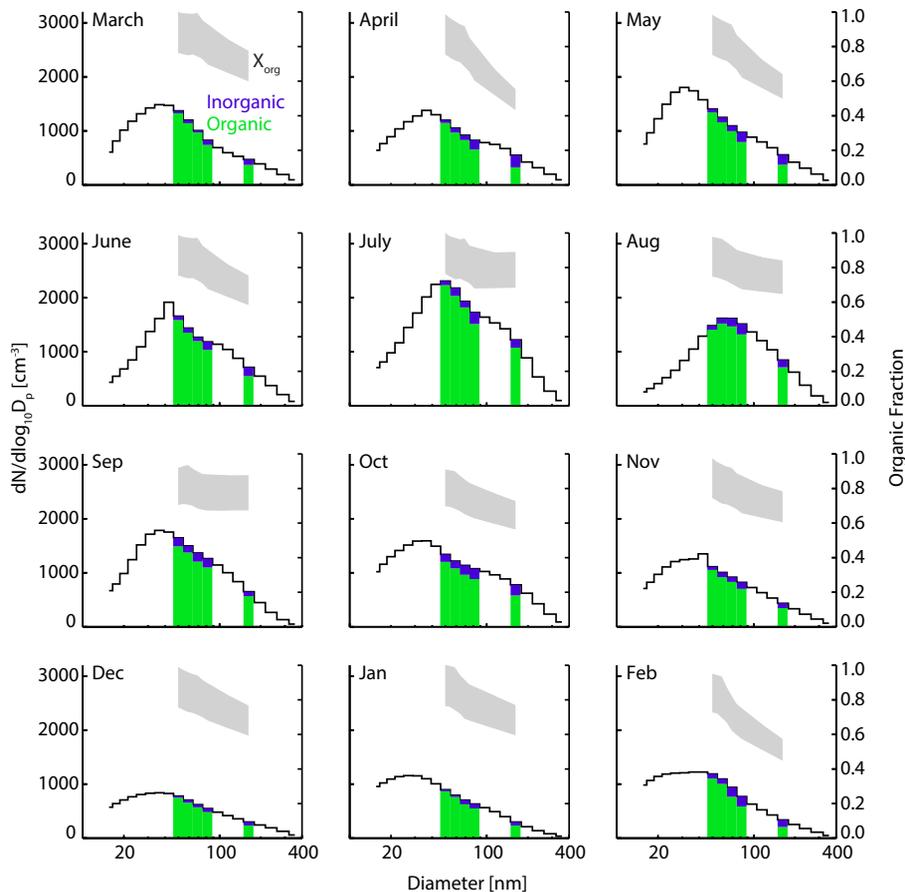**Fig. 4.** Daily κ distributions calculated from AMS measured aerosol composition.

Fig. 5. Monthly averaged aerosol size distributions (black) and two component chemical composition (green = organic, blue = inorganic) calculated from hygroscopicity measurements during the year-long BEACHON study. Gray shading shows the range in organic mass fraction (X_{org}) due to uncertainty in κ_{org}.

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