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**Size-resolved CCN
measurements**

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Technical note: A method for measuring size-resolved CCN in the atmosphere

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Abstract

We present a method to investigate cloud condensation nuclei (CCN) concentrations and activation efficiencies as a function of two independent variables, aerosol particle size and water vapor supersaturation. To date, most ambient CCN measurements have been made as the integral (total) CCN concentration as a function of water vapor supersaturation only. However, since CCN properties of aerosol particles are strongly dependent on particle size, as well as on chemical composition, which commonly varies with particle size, more detailed measurements can provide additional important information about the CCN activation. With size-resolved measurements, the effect of particle size on CCN activity can be kept constant, which makes it possible to directly assess the influence of particle chemistry.

The instrumental set-up consists of a differential mobility analyzer (DMA) to select particles of a known size, within a narrow size range. A condensation nuclei (CN) counter (condensation particle counter, CPC) is used to count the total number of particles in that size range, and a CCN counter is used to count the number of CCN as a function of supersaturation, in that same size range. The activation efficiency, expressed as CCN/CN ratios, can thus directly be calculated as a function of particle size and supersaturation. We present examples of the application of this technique, using salt and smoke aerosols produced in the laboratory as well as ambient aerosols.

1 Introduction

Most CCN measurements in the atmosphere until today have been made as integral CCN concentrations, i.e., total number of CCN per volume of air as a function of supersaturation (S), giving so-called CCN spectra ($N_{\text{CCN}}(S)$). The CCN concentrations are often normalized by the total number of aerosol particles, often-denoted CN (condensation nuclei), yielding CCN activation efficiencies or activated fractions (expressed as CCN/CN ratio). However, it is well known that the ability of an aerosol particle to act as

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a CCN depends primarily on the total amount of soluble material in the particle, which is a function of both its size and its chemical composition. Therefore, size-resolved measurements, which separate the effects of size from the effects of chemical composition, have important advantages: 1) The measured CCN activation efficiency for a given aerosol particle size can directly be compared and linked to independent measurements of chemical composition and/or hygroscopic properties of particles of the same size. This information can be used in closure and model studies to gain an improved understanding of the CCN activation; 2) The critical supersaturation for activation as a function of particle size can be determined; 3) Effective soluble volume fraction can be calculated from the critical supersaturation by using a model salt; 4) The state of mixture within one size class (chemical and morphological internal/external mixture) with respect to CCN activation properties can be studied; 5) If aerosol number size distributions are measured concurrently, CCN size distribution as a function of S can be determined; and 6) By integrating the CCN size distributions, integral (total) CCN concentrations as a function of S ($N_{CCN}(S)$) can be calculated.

Size-resolved CCN measurements have previously been used in laboratory experiments to study activation properties of specific aerosols of known chemical composition (e.g., Cruz and Pandis, 1997; Corrigan and Novakov, 1999; Prenni et al., 2001; Giebl et al., 2002; Raymond and Pandis, 2002, 2003; Kumar et al., 2003; Bilde and Svenningsson, 2004; Broekhuizen et al., 2004; Svenningsson et al., 2005; Dusek et al., 2006). A few size-resolved measurements in the atmosphere have been reported (Fitzgerald et al., 1982; Hudson and Da, 1996). Hudson have made additional measurements (not published) (J. G. Hudson, personal communication), but otherwise this method has not been applied widely in studies of mixed/unknown aerosols.

In this article, we describe the method of size-resolved CCN measurements, “3-D CCN measurements”, in more detail. We also discuss possibilities, limitations and variations with regard to some results from laboratory measurements of biomass burning smoke particles, and from field measurements.

2 Set-up and measurement strategies

The method of size-resolved CCN measurements (Fig. 1) includes the parallel use of a “3-D CCN counter” and an instrument for aerosol number size distribution measurements, e.g., a Scanning Mobility Particle Sizer (SMPS) (e.g. Flagan, 1998). The 3-D CCN counter consists of a Differential Mobility Analyzer (DMA), a CN counter (condensation particle counter, CPC) and a CCN counter. The DMA selects particles within a narrow size range and the CCN counter measures the CCN concentration in that size range as a function of supersaturation (S). The CN counter determines the total number of aerosol particles in the same size range, and the CCN activation efficiency as the CCN/CN ratio can thus directly be calculated. The CCN and CN concentrations must be corrected for multiply charged particles that penetrate the DMA, and a method for that is presented.

The measurements discussed in this article have been made using a 3-D CCN counter consisting of a TSI 3071 Electrostatic Classifier (DMA), which has been somewhat modified and changed to closed-loop arrangement (Jokinen and Mäkelä, 1997), a TSI CPC 3762, and two different CCN counters: the Mainz CCN counter (Roberts et al., 2001; Frank et al., 2006), and the Droplet Measurement Technologies (DMT) CCN counter (Roberts and Nenes, 2005). The Mainz CCN counter is a static thermal-gradient parallel-plate diffusion chamber (SDC) that can measure CCN concentrations in the range of 0.1–2%. The time resolution is in the range of 30–60 s, including a change of supersaturation in-between. The DMT CCN counter is a continuous-flow longitudinal thermal-gradient CCN counter that can measure CCN concentrations in the same supersaturation range. Data are collected at a time resolution of 1 Hz at constant supersaturation. However, it takes on the order of 2–4 min to change the supersaturation of the instrument. Both CCN counters were calibrated with respect to supersaturation and number concentration, using a method presented in Frank et al. (2006).

The 3-D CCN counter can be used in two different measurement strategies: the

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supersaturation scan and the size scan method. For the supersaturation scan, the DMA is first set to a given particle size and the CCN counter measures a CCN spectrum for that size (CCN concentrations as a function of supersaturation). This method is ideal for CCN counters that can perform fast supersaturation scans, such as the Mainz CCN counter.

For the size scan method, the CCN counter is set to a given supersaturation and the CCN concentration is measured as a function of particle size for that supersaturation. This method is ideal for use with a CCN counter where S changes require longer times, such as for the DMT CCN counter. The size can either be changed step-wise or continuously scanned. However, the data obtained by step-wise change is more straightforward to evaluate, and leads to less uncertainty. An advantage with continuous scanning is, however, that both CN and CCN size distributions can directly be obtained with the 3-D CCN counter itself, if the size-steps are reasonably small. The parallel SMPS is thus not needed.

We found that measuring CCN spectra at five different particle diameters, using five different and individually selected supersaturations for each diameter, using the supersaturation scan method, and vice versa, using the size scan method, gives sufficiently detailed information at a reasonable time resolution. The minimum time for a five-times-five scan is about 30–45 min for both methods, and is the highest time resolution possible with the present setup.

A drawback when measuring size-resolved CCN concentrations in the atmosphere is the low CCN concentration in a single size class, compared to integral measurements of all relevant sizes. Longer averaging times can therefore be necessary. The use of a continuous-flow CCN counter for size-resolved measurements is desirable, since the effective sampling time is at least an order of magnitude lower. When using the Mainz CCN counter to sample the background continental aerosol in central Europe, the concentration after the DMA was close to the lower detection limit of the CCN counter. To minimize the statistical variation, we averaged CCN spectra over 6 h, i.e., approximately 15–20 scans. However, when using the DMT CCN counter in the same

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or even in cleaner environments, a full five-times-five scan could be obtained in 30–45 min, with an acceptably low statistical variation. In laboratory measurements this problem is smaller, since the particle number concentration can be controlled and set to be high enough to measure with high time resolution.

5 The ratio between the aerosol flow and the sheath-air flow of the DMA determines the width of the size range that penetrates the DMA. It can therefore, to some extent, be used to control the particle concentration and thus improve the counting statistics. For example in laboratory experiments, where particle concentrations are high, a ratio of 1:10 is commonly used, but it is possible to increase this ratio to 1:3 for atmospheric
10 measurements. A change in the ratio from 1:10 to 1:3 increases the particle concentration by a factor of ~ 3.5 .

The finite width of the size range also introduces a slight instrumental broadening of the measured activation curves. An infinitely narrow size range would lead to an ideal step-function, if the measured particles were all of the same chemical composition. However, since the size range has a finite width, which depends on the aerosol
15 flow to sheath flow ratio of the DMA, this ratio also determines the broadening. The broadening effect will be discussed and compared to measured results in Sect. 3.

Since size-resolved measurements only determine the CCN and CN concentrations of particles in a few selected size ranges, concurrent measurements of aerosol particle
20 size distributions (CN size distributions) are necessary to derive the integral CCN concentrations. These measurements can for example be performed by using an SMPS (Fig. 1). By combining (multiplying) the size resolved CCN measurements at different S with the CN size distributions, CCN size distributions can be calculated for different S , see an example in Sect. 3 (Fig. 2d). These CCN size distributions can be integrated to
25 calculate the total number concentration of CCN as a function of S , which corresponds to the result normally obtained by integral CCN measurements.

The CN size distribution is also necessary for the correction for multiply charged particles that penetrate the DMA. When using a DMA for the size selection, the aerosol particles that penetrate the DMA are all within a narrow range in particle electrical

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mobility. The electrical mobility (Z) of a spherical particle with a diameter d is given by:

$$Z = \frac{i \cdot e \cdot C_c(d)}{3\pi \cdot \eta \cdot d} \quad (1)$$

where i is the number of electrical charges on the particle, e is the elementary charge, C_c the Cunningham correction factor, and η the viscosity of air. Particles with the same electrical mobility can have different diameters, d_1, d_2, d_3, \dots , corresponding to 1, 2, 3, ... charges. Most particles that penetrate the DMA have a diameter d_1 corresponding to singly charged particles, but particles with larger diameters corresponding to a higher number of charges are also present. In the CCN counter, these larger particles are normally activated at a lower supersaturation than the singly charged particles, and, since it is desirable to consider only the activation of singly charged particles, they will thus disturb the measurement. It is therefore necessary to correct for this bias, and the correction procedure is outlined here:

Before passing through the DMA, the particles pass through a bipolar charger where a defined charge distribution is imposed on the particles. The defined charge distribution, i.e., the probability of finding a particle of diameter d_j having a number of charges i , $P_i(d_j)$, can be calculated according to Wiedensohler (1988).

The number concentration of singly charged particles that penetrate the DMA ($CN(d_1)$) can be calculated by multiplying the total number of particles that penetrate the DMA (CN_{tot}), with the fraction of singly charged particles ($f(d_1)$). This fraction can be calculated using the size distribution $s(d)$, measured by the SMPS:

$$CN(d_1) = CN_{\text{tot}} \cdot f(d_1) = CN_{\text{tot}} \cdot \frac{s(d_1) \cdot P_1(d_1)}{\sum_i s(d_i) \cdot P_i(d_i)} \quad (2)$$

The CCN concentration of singly charged particles that activate at a certain supersaturation in the CCN counter, ($CCN(d_1)$), can be calculated by subtracting the multiply charged particles that activate at that same supersaturation from the total CCN con-

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centration measured (CCN_{tot}).

$$CCN(d_1) = CCN_{tot} - CN_{tot} \cdot f_a(d_{i>1}) = CCN_{tot} - CN_{tot} \cdot \frac{\sum_{i>1} s(d_i) \cdot P_i(d_i) \cdot a(d_i)}{\sum_i s(d_i) \cdot P_i(d_i)} \quad (3)$$

where $a(d_i) = CCN(d_i)/CN(d_i)$ is the activated fraction of particles for the respective diameters at the same supersaturation. The activated fraction $a(d_i)$ of the larger, multiply charged particles is estimated by combining all data in a five-times-five scan. If possible, the size and supersaturation ranges should be chosen in such a way that the largest particles measured are all activated or close to being all activated at all the supersaturations measured. Thus, for this size the assumption $a(d_{2,3,4,\dots}) = 1$ can be made. For the smaller particle sizes interpolations can be necessary to determine $a(d_{2,3,4})$ from the measured size-resolved spectra. In some special cases, such as in the case with peat smoke aerosols that will be briefly presented in the next section, additional assumptions have to be made.

3 Applications and discussion

To illustrate the potential of the size-resolved method we present and discuss a few examples from recent experiments. The 3-D CCN counter was first used in laboratory measurements as part of the project: "Impact of Vegetation Fires on the Composition and Circulation of the Atmosphere" (EFEU) (Wurzler et al., 2001; Chand et al., 2005; Dusek et al., 2005). Different biomass fuels, e.g., boreal and tropical wood fuels, savanna grass, and boreal and tropical peat, were burnt in a laboratory facility, and the properties of smoke particles were analyzed with different methods. The results from size-resolved CCN measurements of a spruce wood fire are presented in Fig. 2, as averages over a typical measurement period of about one hour. Figure 2a is a 3-D plot (CCN/CN ratio versus supersaturation and particle diameter) which clearly shows the three-dimensional data-set. At low supersaturation and small particle diameters, the

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CCN/CN ratio is low, whereas it is close to one at the largest particle sizes and the highest supersaturations measured. Figures 2b and c present the same data projected into two dimensions. In these two graphs the dependence on the size and supersaturation, respectively, can be studied in more detail. From the activation curves as a function of S (Fig. 2b), it can be seen that for the two largest particle sizes studied, nearly 100% of the particles are activated at all S studied. The critical S for activation for the 100 and 150 nm diameter particles can be estimated by determining the S at the CCN/CN ratio of 0.5. The critical S for activation of the 100 nm particles can be estimated to be close to 0.5%, and for the 150 nm particles to be close to 0.3%. By using a model salt, the size-resolved effective soluble volume fraction of these particles can be calculated with the Köhler equation (e.g. Pruppacher and Klett, 1997). The effective soluble volume fraction can be used in cloud droplet activation (cloud nucleation) parameterizations and models.

The results presented in Fig. 2c can be used to fit functions that can be multiplied with the CN size distribution, resulting in CCN size distributions for each supersaturation measured. In Fig. 2d we present the CN size distribution together with CCN size distributions at $S=0.23\%$ and 1.00% . The CCN size distributions are subsets of the CN size distributions and can be used to validate cloud activation models. From the CCN size distributions, the total number of CCN particles as a function of supersaturation can be integrated, yielding integral CCN spectra ($N_{\text{CCN}}(S)$) (i.e., the kind of data normally obtained in integral CCN concentration measurements).

CCN spectra from a fire with Indonesian peat, in which unusual features were observed for the larger particle sizes, are shown in Fig. 3. This is an example of how size-resolved CCN measurements can be used to distinguish external/internal mixtures with respect to CCN activation. The CCN properties from all wood fires studied in EFEU were similar (Fig. 2b), and the peat smoke particles also show a similar behavior for 50 and 100 nm particles (compare Figs. 2b and 3). However, for the 250 and 325 nm particles, the CCN/CN ratios are very different. For wood smoke, the CCN/CN ratio is close to one at all S studied, whereas for the largest peat smoke particles the CCN/CN

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ratio shows a slight dependence on S , but is never greater than 0.5. This indicates that for the larger size range, the peat smoke particles consist of an external mixture of two fractions: a minor fraction that is readily activated at supersaturations of 0.23% and higher, and a major fraction that is not activated even at supersaturations as high as 1.6%. Indeed, we found using Scanning Electron Microscopy (SEM) that a large fraction of the larger particles from the peat fires were hollow. We assume that these, for not yet fully understood reasons, behave hydrophobically. These results are discussed in more detail in Dusek et al. (2005), but here it is interesting to mention that this feature could not have been discovered with integral CCN concentration measurements.

The broadening of the activation curves can also give information about the state of mixing of the particles of a certain size. The theoretical instrumental broadening can be calculated with the Köhler equation, using the upper and lower diameters of the DMA transfer function. When comparing to measurements of CCN activation of single-compound aerosols, such as to the instrument calibration with pure ammonium sulfate particles, the broadening of the measured activation curve agrees very well with the ideal theoretical broadening (Fig. 4a). An aerosol to sheath flow ratio of 1:10 was used in the calibration, and it can be seen that these curves are very steep and relatively close to a step function. This shows the typical behavior of internally mixed particles, i.e., all particles of a given size have the same chemical composition and therefore the same activation behavior.

When investigating the activation curves of the smoke particles from the EFEU experiment (Fig. 4b) it can be seen that the broadening is much larger than for the ideal curves (aerosol to sheath flow ratio of 1:10). The particles of one size thus activates over a broad supersaturation range, indicating that the particles have very different morphology, and/or composition, spanning a wide range of soluble fractions. This indicates a more external mixture of the particles. Ambient aerosols measured in central Germany (Taunus Observatory at Kleiner Feldberg near Frankfurt am Main) as part of the Feldberg Aerosol Characterisation Experiment (FACE-2004) also show slight external mixing, but the state of mixture seems to be dependent on size (Fig. 4c). The

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activation curve of the 40 nm particles is very close to the ideal curve and indicates internally mixed particles, whereas the activation curves of 60 and 80 nm particles are broader than the ideal curves, and thus indicate some degree of external mixture.

4 Conclusions

5 A method for size-resolved CCN measurements has been developed and tested: the 3-D CCN counter. The instrument measures CCN concentrations and CCN/CN ratios as a function of supersaturation and aerosol particle size. The size-resolved measurements present an important improvement, as they make it possible to study the CCN properties of aerosol particles in more detail, in particular by separating the influences
10 of chemical composition and size distribution on the CCN activation. By this approach, these two factors can be studied individually, which is not possible from the common method of integral measurements of CCN concentrations. The size resolved results can be used to: (1) calculate the critical supersaturation for activation as a function of particle size; (2) calculate the effective soluble volume fraction using a model salt as
15 a function of particle size; (3) study the state of mixing (chemical and morphological) of particles in one size range; and (4) in combination with CN size distribution measurements, to calculate CCN size distributions and the integral CCN concentration as a function of supersaturation. These results can be used to validate CCN and cloud droplet prediction models.

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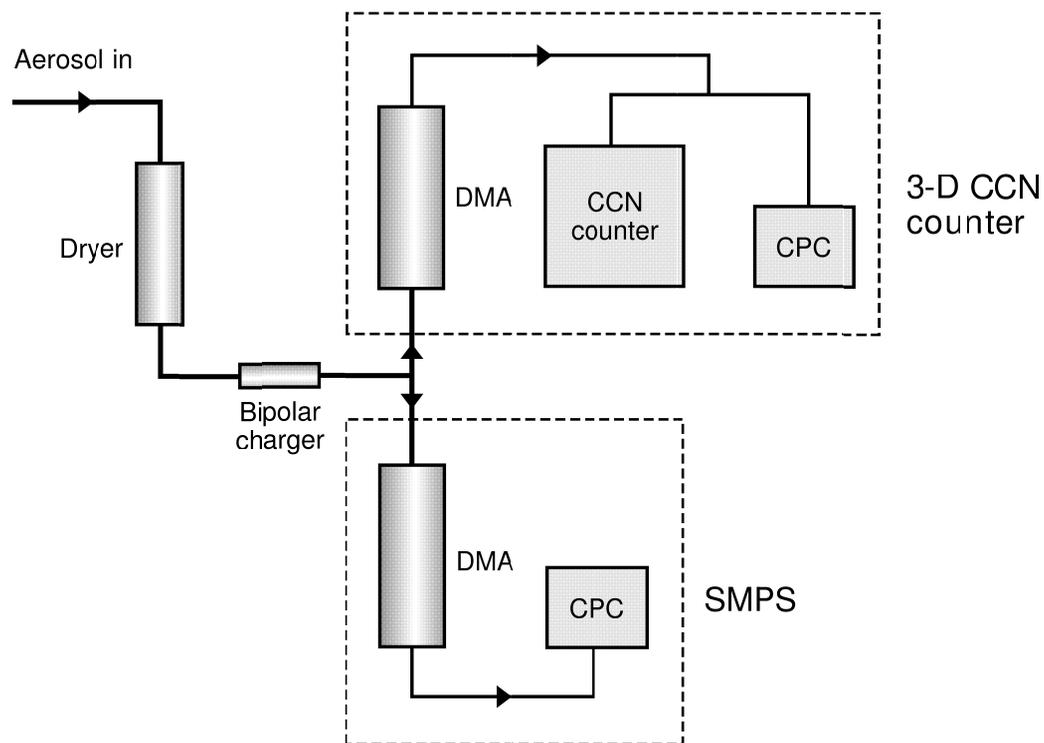


Fig. 1. The instrumental set-up, consisting of the 3-D CCN counter (DMA, CCN counter and CPC), and SMPS.

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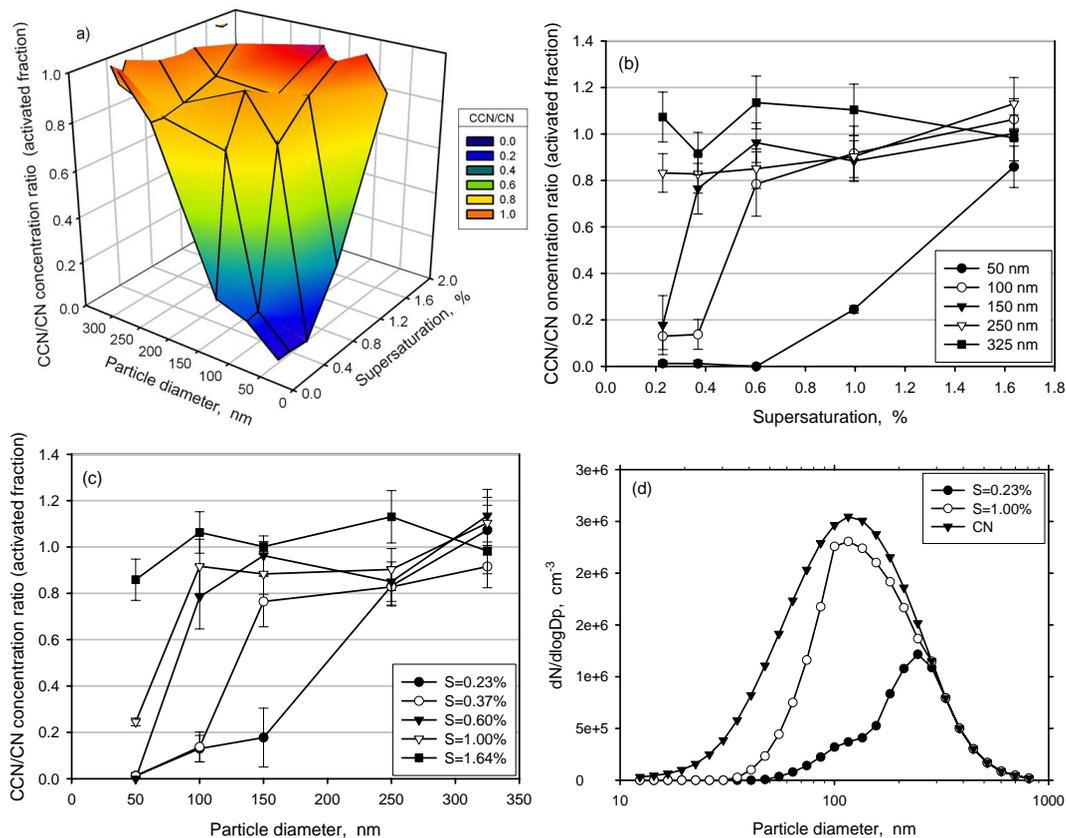


Fig. 2. Results from a spruce wood fire from the EFEU experiment. **(a)** Average CCN/CN ratios as function of supersaturation and particle diameter. **(b)** and **(c)** The same data projected and presented as CCN/CN ratios as a function of supersaturation (b) and as function of particle diameter (c). **(d)** Average CN and CCN size distributions.

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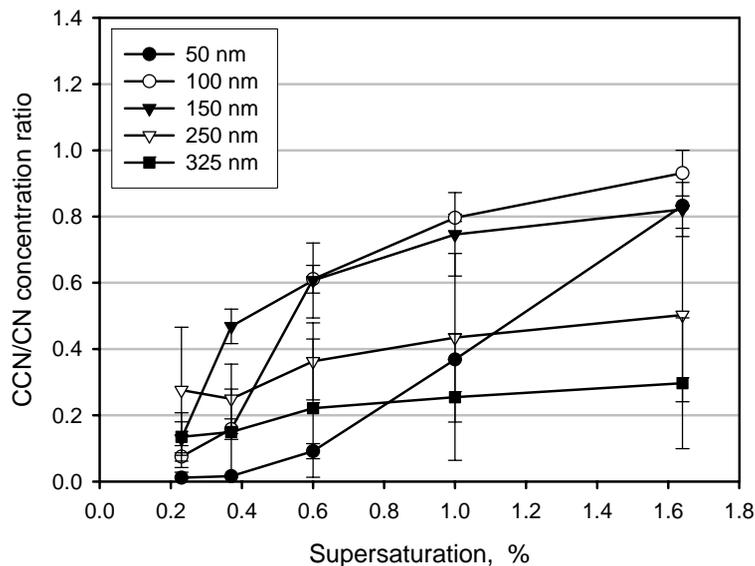


Fig. 3. Results from a peat fire from the EFEU experiment. Average CCN/CN ratios as function of supersaturation, compare with Fig. 2b. Sizes refer to particle diameter. The 50 and 100 nm particles show similar behavior to the wood fire, whereas the 250 and 325 nm particles behave different.

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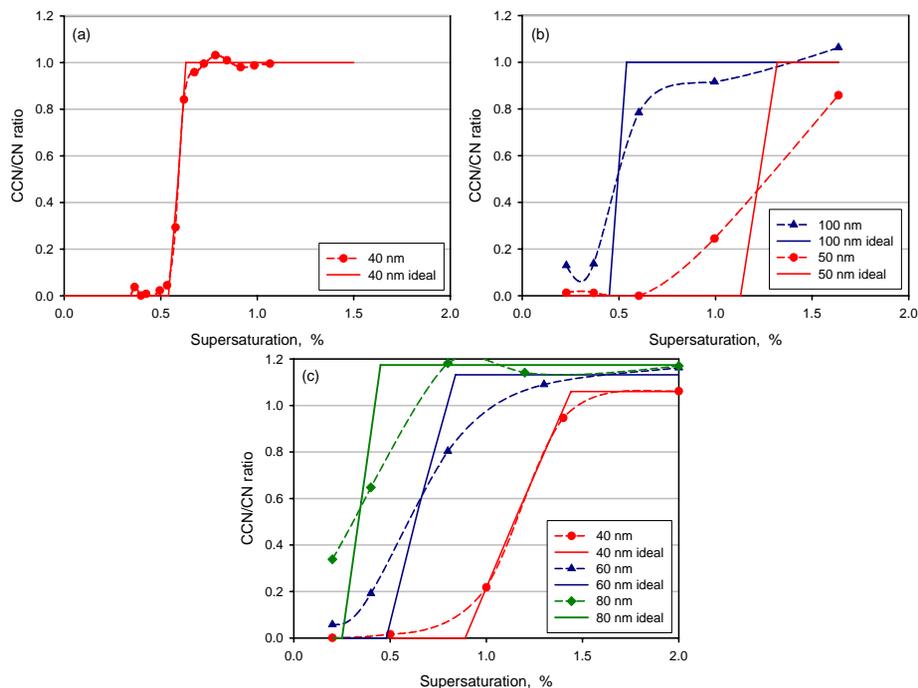


Fig. 4. Comparisons of the broadening of the activation curves with ideal (theoretical) instrument broadening. Sizes refer to particle diameter. **(a)** Pure ammonium sulfate particles, aerosol to sheath flow ratio 1:10; **(b)** smoke particles from a spruce wood fire, aerosol to sheath flow ratio 1:10; and **(c)** ambient aerosols sampled in central Germany, aerosol to sheath flow ratio 1:3.

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