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**Size-dependent  
activation of aerosols**

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# Size-dependent activation of aerosols into cloud droplets at a subarctic background site during the second Pallas Cloud Experiment (2nd PaCE): method development and data evaluation

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## Abstract

In situ measurements of aerosol water uptake and activation of aerosols into cloud droplets provide information on how aerosols influence the microphysical properties of clouds. Here we present a computational scheme that can be used in connection with such measurements to assess the influence of the particle chemical composition and mixing state (in terms of the water uptake) on the cloud nucleating ability of particles. Additionally, it provides an estimate for the peak supersaturation of water vapour reached during the formation of the observed cloud(s). The method was applied in interpreting results of a measurement campaign that focused on aerosol-cloud interactions taking place at a subarctic background site located in northern Finland (second Pallas Cloud Experiment, 2nd PaCE). A set of case studies was conducted, and the observed activation behavior could be successfully explained by a maximum supersaturation that varied between 0.18 and 0.26% depending on the case. In these cases, the diameter corresponding to the activated fraction of 50% was in the range of 110–140 nm, and the particles were only moderately water soluble with hygroscopic growth factors varying between 1.1 and 1.4. The conducted analysis showed that the activated fractions and the total number of particles acting as CCN are expected to be highly sensitive to the particle hygroscopicity. For example, the latter quantity varied over a factor between 1.8 and 3.1, depending on the case, when the mean hygroscopic growth factors were varied by 10%. Another important conclusion is that size-dependent activation profiles carries information on the mixing state of particles.

## 1 Introduction

Largest uncertainties in the current predictions regarding the climate change arise from the influence of aerosols on the microphysical properties of clouds (IPCC, 2007). This underscores the need to improve our knowledge on the connections between the atmospheric aerosols and clouds (Ghan and Schwartz, 2007; Lohmann et al., 2007).

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To address this issue, several advances in theoretical understanding of the cloud formation have been made during the recent years, and many of the results have been incorporated into large-scale climate models as computationally efficient parameterizations (Chen and Penner, 2005; McFiggans et al., 2006 and references therein).

5 However, further experimental studies involving in-situ measurements of aerosols and their activation into cloud droplets are still warranted to refine microphysical models as well as the resulting parameterizations (Ghan and Schwartz, 2007).

Ground-based measurements of activation of ambient aerosols are typically based on a separate determination of “interstitial” and “residual” fractions of aerosols. The former refers to those particles that did not participate in the cloud formation, whereas the latter acted as cloud condensation nuclei (CCN) and thus grew through condensation of water vapor to notably larger diameters, i.e. over an order of magnitude, than interstitial particles. Using such an experimental setup, the fraction of particles that are activated into cloud droplets can be inferred. Size-resolved measurements, if available, provide further insights into aerosol activation through allowing for investigating size-dependent particle activation characteristics (e.g. Svenningsson et al., 1994; Henning et al., 2002; Kompula et al., 2005; Targino et al., 2007).

Size-resolved measurements on the cloud scavenging of ambient particles can be complemented by determining the particle water uptake properties by using e.g. a Hygroscopic Tandem Differential Mobility Analyzer (H-TDMA) that is used to measure the hygroscopic growth of particles as a function of time and particle size (McMurry, 2000). H-TDMA measurements allow for investigating the influence of the particle water-solubility and they provide also information on the particle mixing state in terms of the water uptake. The importance of H-TDMA measurements is highlighted because the particle hygroscopicity and mixing state are expected to play a large role in the cloud nucleating ability of atmospheric particles (McFiggans et al., 2006). Furthermore, the maximum supersaturation reached during the formation of observed clouds can be estimated on the basis of activation and H-TDMA measurements by applying the Köhler theory (Svenningsson et al., 1994, 1997; Mertes et al., 2005). To be more

specific, a typical approach in these studies is to determine the fraction of particles capable of acting as CCN as a function of the supersaturation of water vapour, and then estimate the maximum supersaturation reached during the cloud formation by comparing the predicted size-dependent activation curves with the measurement results.

Several computational tools have been developed to aid interpretation of the experimental data on the water uptake properties of particles with varying physico-chemical properties (e.g. Rissler et al., 2004; Kreidenweis et al., 2005; Gasparini et al., 2006; Padró et al., 2007; Petters and Kreidenweis, 2007; Ziese et al., 2008). These frameworks have been motivated by CCN measurements where particles are exposed to a known water vapour supersaturation and are often used to interpret laboratory experiments where the particle chemical composition is more constrained compared to variability exhibited by atmospheric particles. On the other hand, the existing parameterizations of cloud droplet formation during an ascent of a convective air parcel require the updraft velocity of the air parcel as an input (Abdul-Razzak and Ghan, 2000; Co-  
hard et al., 2000; Nenes and Seinfeld, 2003; Ming et al., 2006), a quantity which is difficult to determine in ground based studies (Verheggen et al., 2007). To the best of our knowledge, no corresponding computational procedure that would be applicable to studies on ambient activation of particles using the above described experimental setup is presented in the literature.

The current study addresses the above-mentioned gaps in the literature by developing and applying a method that is suitable for analyzing size-resolved data on atmospheric aerosols, their activation characteristics and hygroscopic properties. To be more explicit, our goals are to: 1) present a simple computational procedure which allows for calculating the activated fractions of particles based on the Koehler theory and information on the particle hygroscopicity, 2) derive expressions for estimating the maximum supersaturation of water vapour and for calculating CCN spectra, and 3) apply the developed framework to interpreting results of a measurement campaign that focused on aerosol-cloud interactions taking place at a subarctic background site (Komppula et al., 2006).

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The paper is organized as follows. In Sect. 2, the computational procedure is derived and some additional issues that arise when applying the method are discussed. The relevant measurements carried out during the measurement campaign and some general features of the results are briefly described in Sects. 3 and 4, respectively. The results of the data analysis are given in Sects. 4, and the summary and conclusions follow in Sect. 5.

## 1.1 Model approach

## 1.2 General formulation

We consider a situation where there are size-segregated measurements for the particle number concentration and for the activated fraction of particles,  $CN_{\text{tot},1}$ ,  $CN_{\text{tot},2}, \dots$ ,  $CN_{\text{tot},N}$  and  $AF_1$ ,  $AF_2, \dots, AF_N$ , respectively. The corresponding particle dry diameters are denoted by  $d_{p,1}$ ,  $d_{p,2}, \dots, d_{p,N}$ . To start with, we assume that no entrainment took place during the measurements. We also assume that the hygroscopic properties of particles have been measured or estimated for each particle size class. For the purposes of this study, it is convenient to characterize the hygroscopic properties by the normalized cumulative distributions of the hygroscopic growth (HG) factors,  $GR_1$ ,  $GR_2, \dots, GR_N$ . The distributions can be obtained through a fitting procedure. It should be noted that the approach does not involve any assumptions regarding the shape of the humidified distributions, and thus it allows for considering a case where particles with the same dry size exhibit a variety of water uptake properties. Also, the list of the key symbols used in this section is given in Appendix A.

The Köhler theory states that those particles/droplets whose critical supersaturation,  $s_c$ , is lower than the ambient supersaturation of water are activated, i.e. they grow spontaneously through condensation of water vapour (Seinfeld and Pandis, 1998). According to the Köhler theory, the normalized cumulative distributions of the fraction of

particles that are activated at a supersaturation  $s$ ,  $K_i(s)$ , can be formulated as follows:

$$K_i(s) = \frac{CN_i(s_c < s)}{CN_{\text{tot},i}}, \quad (1)$$

where  $CN_i(s_c < s)$  is the number of particles having a critical supersaturation below  $s$  in the size class  $i$ ,  $i=1, \dots, N$ . In order to estimate the maximum supersaturation reached during the formation of observed cloud(s),  $s_{\text{max}}$ , we derive an expression for the fraction of activated particles in the size bin  $i$ ,  $AF_i^*$ . Here we apply the Köhler theory by assuming that those, and only those, particles which have a critical supersaturation below a value  $s$  contribute to the droplet population of a cloud formed under maximum supersaturation  $s$ . Consequently,  $AF_i^*$  and  $K_i$  are equal, i.e.  $AF_i^* = K_i$ . The validity of this assumption will be discussed in Sect. 2.2.

Next we utilize information on the particle hygroscopic properties by expressing the distribution  $K_i$  in terms of  $GR_i$ . This is achieved by first relating  $GR_i$  and corresponding distribution of the particle water-soluble volume fraction,  $SF_i$ . In the next step, the Köhler theory is used to relate the water-soluble mass fraction and the particle critical supersaturation. To this end, the water uptake of particles is modeled by assuming that the dry particle mass consists of two components: water-soluble and water-insoluble fractions, and that the former component exhibits similar hygroscopic behavior than ammonium sulfate at subsaturated conditions. Accordingly the soluble volume fraction of particles,  $\varepsilon$ , is related to the measured hygroscopic factor,  $GR_{\text{meas}}$ , as follows (e.g. Swietlicki et al., 1999):

$$\varepsilon = \frac{GR_{\text{meas}}^3 - 1}{c(a_w)}, \quad c(a_w) = GR_{AS}^3(a_w) - 1. \quad (2)$$

Here  $a_w$  is the water activity at which the hygroscopic growth measurements were conducted and  $GR_{AS}(a_w)$  is the growth factor of ammonium sulfate at the water activity  $a_w$ . From Eq. (2), we obtain a relation between  $GR_i$  and  $SF_i$ :

$$SF_i(\varepsilon) = GR_i[g_i(\varepsilon)], \quad g_i(\varepsilon) = (c_i\varepsilon + 1)^{1/3}, \quad c_i = c(a_{w,i}). \quad (3)$$

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Here  $a_{w,i}$  is the water activity at which the hygroscopic properties of particles in the size bin  $i$  were measured or estimated. On the other hand, the Köhler theory provides a link between the particle water solubility, parameterized here with  $\varepsilon$ , and the particle critical supersaturation,  $s_c$  (Seinfeld and Pandis, 1998):

$$s_c \approx k \varepsilon^{-1/2} d_p^{-3/2},$$

$$k = 2 \left( \frac{4}{3RT} \right)^{3/2} \left( \frac{M_w}{\rho_w} \right) \left( \frac{M_s}{v\phi\rho_s} \right)^{1/2} \sigma^{3/2}. \quad (4)$$

Here  $d_p$  is the dry particle diameter,  $\sigma$  is the particle surface tension,  $M_s$ ,  $v\phi$ , and  $\rho_s$  are the molecular weight, the “effective” number of ions into which a solute molecule dissociates (the van’t Hoff factor) and the density of ammonium sulfate, respectively. In addition,  $R$  is the ideal gas constant,  $T$  is the temperature, and  $M_w$  and  $\rho_w$  are the molecular weight and density of water, respectively. The relation linking  $AF_i^*$ ,  $K_i$  and  $GR_i$  is now obtained from the equality of  $AF_i^*$  and  $K_i$  (see above), Eq. (3) and Eq. (4):

$$AF_i^*(s) = K_i(s) = 1 - GR_i [h_i(s)],$$

$$h_i(s) = \left( c_i k^2 d_{p,i}^{-3} s^{-2} + 1 \right)^{1/3}. \quad (5)$$

The function  $h_i$  maps a supersaturation  $s$  to the HG factor of particles having a critical supersaturation  $s$  and a dry diameter  $d_{p,i}$ . In deriving Eq. (5), it was assumed that  $\sigma$  and  $i$  are independent of the particle size. However, if the particle water-soluble fraction contains surface-active molecules,  $\sigma$  depends on the solute concentration and hence also on the particle size (e.g. Adamson and Gast, 1997; Padró et al., 2007). Similarly the van’t Hoff factor may also exhibit sensitivity to the particle size. For the sake of brevity, detailed discussion on this topic is postponed to Sect. 2.2. Explicit expressions for  $GR_i$  in cases where the humidified particle can be described with a Gaussian or log-normal distributions or a sum of them are presented in Appendix B.

The “effective” maximum supersaturation,  $s_{\max}$ , reached during the formation of observed cloud(s) can be estimated using Eq. (5) by minimizing the difference between the calculations and observations with respect to a chosen norm. We use the term

“effective” to highlight the underlying assumption according to which the observed air masses share the same supersaturation history whereas atmospheric observations may represent an ensemble of different supersaturation histories. The assumption is necessary, however, given the experimental setup considered here. In the following, we estimate  $s_{\max}$  using the Euclidean norm and thus minimize the following function:

$$G(s) = \sqrt{\sum_{i=1}^N [AF_i^*(s) - AF_i]^2}. \quad (6)$$

The resulting activated fractions  $AF_i^*(s_{\max})$  can be compared with the measurements to evaluate the agreement between the observations and the results of the estimation procedure.

Equation (5) can also be used to calculate the CCN spectra:

$$CCN_{tot}(s) = \sum_{i=1}^N AF_i^*(s) CN_{tot,i}. \quad (7)$$

In particular,  $CCN_{tot}(s_{\max})$  can be compared with the observational number of activated particles to test the agreement between the measurements and calculations. Independent measurements of CCN spectrum, if available, can be used in a further closure study (see Snider et al., 2003, for example). Finally, Eq. (7) can also be applied to investigate the sensitivity of CCN concentrations to the particle chemical composition and/or mixing state by varying the values of the parameters which determine the distributions  $GR_j$ .

### 1.3 Further considerations

#### 1.3.1 Size dependent surface tension and Van’t Hoff factor

Here we extend the above-presented method to a case where the surface tension and/or van’t Hoff factor exhibits dependence on the particle size that arises from the

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presence of the water-soluble component of aerosols. Presenting a detailed treatment that addresses these issues is beyond the scope of this study, and it suffices to note that several frameworks for calculating the van't Hoff factor and surface tension exist (Li et al., 1998; Nenes et al., 1998; Abdul-Razzak and Ghan, 2004; Sorjamaa et al., 2004). To maintain flexibility in this respect, we present an approximative scheme that can be used in connection with any chosen thermodynamic model.

First we note that certain thermodynamic methods require that the soluble fraction of particles is specified for each particle size class (e.g. Abdul-Razzak and Ghan, 2004). Since our approach is based on describing the particle water-solubility using continuous distributions (see Eq. 4), a representative value for the particle water-solubility is needed for each size class. An intuitive choice in this respect is the soluble fraction corresponding to the median growth factor, in particular when the humidified distributions are monomodal and relatively narrow (see Eq. 2). Once the model for calculating the surface tension and/or for the van't Hoff factor has been chosen along with the representative values of the needed parameters, the following scheme can be applied.

1. Calculate the particle critical diameter as well as the surface tensions and surface excesses of the water-soluble component,  $\sigma_i$  and  $\Gamma_i$ , respectively, at the representative critical supersaturation for each particle size class. If size dependent dissociation is accounted for, also the van't Hoff is calculated separately for each size class.

2. Substitute  $k$  in Eq. (4) with the following expression:

$$k_i = 2f_i^{-1/2} \left( \frac{4}{3RT} \right)^{3/2} \left( \frac{M_w}{\rho_w} \right) \left( \frac{M_s}{v_i \phi_i \rho_s} \right)^{1/2} \sigma_i^{3/2}. \quad (8)$$

Here  $f_i$  is the fraction of water-soluble matter that partitions into the bulk solution contained by particles in the size class  $i$ , and it can be calculated using  $\Gamma_i$  (e.g. Abdul-Razzak and Ghan, 2004). The parameter  $f_i$  is introduced because the surface/bulk partitioning of the surface-activate solute influences the Raoult's term in the Köhler equation and hence also the particle critical supersaturation (Sorjamaa et al., 2004). To account for this, we decrease the water-soluble volume fraction of particles by the

amount of the water-soluble matter residing on the particle/droplet surface. If the surface/bulk partitioning is not taken into account,  $f_j$  is set to the unity. The obtained expressions  $k_j$  are then substituted for  $k$  in Eq. (5) and the subsequent calculations are carried out as previously.

### 5 1.3.2 Entrainment

Another potentially important issue neglected above is the entrainment of cloud-free air into the air parcel where measurements were conducted (Noone et al., 1992; Mertes et al., 2005). However, the method can be extended to investigate the influence of inhomogeneous entrainment by invoking some approximations. Let us assume that the  
10 activated fractions were measured in an air parcel containing a cloud air and cloud-free air with volume fractions  $1-x$  and  $x$ , respectively. Provided that both fractions contained the same aerosol population, the activated fractions in the cloud air,  $AF_{c,i}$ , are related to the measured activated fractions  $AF_i$  as follows,  $AF_{c,i} = xAF_i$ . The corresponding supersaturation can be estimated as previously with the exception that  $AF_{c,i}$  are used  
15 instead of  $AF_i$ . When comparing the resulting predictions with measurements, the calculated activated fractions should be multiplied with the factor  $x$ . It should be kept in mind, however, that the resulting estimates of  $s_{\max}$  refer to the undiluted fraction of the sampled air. Also, unless the extent of entrainment can be assessed with an independent method, sensitivity studies with varying value of  $x$  can be performed to  
20 find the value range of  $x$  which is consistent with measurement results.

### 1.3.3 Kinetic limitations

An assumption behind the approach is that all particles having a critical supersaturation below  $s_{\max}$  are incorporated into cloud droplets. Atmospheric cloud formation is, however, a dynamical process during which the water vapor concentration changes over  
25 relatively short timescales and therefore particles may not have enough time to grow to cloud droplet sizes (Chuang et al., 1997; Nenes et al., 2001). In order to quantify

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possible errors associated with these so-called kinetic limitations, the cloud formation has to be simulated explicitly with a dynamic model that requires information on the updraft velocity as an input (Pruppacher and Klett, 1997). Determining the updraft velocity near the cloud base is problematic, however, when operating in ground based platforms (Verheggen et al., 2007). Furthermore, the aim here is to provide a simple computational tool for interpreting data rather than present a complex numerical model. For these reasons, extending the method to cover the effects of kinetic limitations is not feasible. Nevertheless, we acknowledge that kinetic limitations may bias results of a data evaluation done based on the method presented here.

## 2 Experimental methods

The second Pallas Cloud Experiment (2nd PaCE) took place at the Pallas Global Atmosphere Watch station in northern Finland from 16 September 2005 to 6 October 2005 (location 67° 58'N, 24° 70'E, see Komppula et al., 2005, for details). The station was inside the cloud cover for around one third of the campaign time and a wealth of data on the relationships between aerosols and cloud droplets was gathered. An overview of the experiment and its main results has been described in Komppula et al. (2006) and therefore only measurements that are directly relevant to the current study are described below.

The particle size distributions were measured with a dual-inlet setup that allowed for determining the activated fraction of particles during cloud events. The first inlet sampled the total air which contained both residual and interstitial particles, whereas the second inlet was equipped with a PM<sub>2.5</sub> cyclone to measure only the interstitial particle population. The fraction of activated particles was determined as a function of the particle size by comparing the particle concentrations from these two inlets (Komppula et al., 2005). Similar Differential Mobility Particle Sizer (DMPS) systems were used at both inlets for particle sizing at the dry particle diameter range 7–500 nm, and also similar Condensation Particle Counters (CPC, model TSI 3010) operated at both inlets.

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A log-normal fit with two modes, corresponding to the Aitken and accumulation modes, was performed to the particle size distributions, and the number of activated particles in these two modes was inferred separately.

Hygroscopic properties of both the dried interstitial aerosol population, sampled with the  $PM_{2.5}$  inlet, and the total aerosol population, sampled with the total inlet, were measured with an H-TDMA instrument (Joutsensaari et al., 2001). The humidifier was kept at about 90% relative humidity, and the measured dry diameters were 30, 50, 80, 100 and 150 nm. The relative humidity of the aerosol flow was kept about 2–3% lower than that of the sheath flow. A log-normal function was fitted to each humidified distribution and the performance of the fit was evaluated. The comparison showed that the chosen fit describes well the measurement data for the periods considered here and hence the results based on the fit are used in the following analysis.

The presence of clouds was verified from the relative humidity and visibility measurements (Vaisala FD12P weather sensor) as well as from photographs of the site taken automatically every 30 min for daytime periods. Moreover, the air mass history was evaluated by calculating the 5-day back trajectories for every three hours, or for every hour for cloud events lasting less than three hours, using the HYSPLIT model (Draxler and Rolph, 2003). The data were classified here according to the air mass history into three categories: continentally-influenced European (“EUR”), marine (“MARINE”) and “mixed” air masses that have spent considerable amount of time both over sea and land (“MIX”). In order to be classified as a continental or marine case, the trajectory path had to have a clear history over the continent or the sea, respectively. If any indication of mixing of these two sources was observed, the trajectory was classified as a mixed case. This was done to differentiate continental and marine cases so that that the results would not be biased by mixing of the two source types.

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### 3 General characteristics of aerosols and their activation

The measurement site was inside cloud for approximately 100 h during the experiment, and the data set contains 13 pronounced cloud events that lasted at least three hours. The average particle number size distributions for the cloudy periods, classified according to the air mass category, are shown in Fig. 1 and the corresponding average characteristics of the aerosols and their activation are displayed in Table 1. As can be seen, the average particle number concentrations were around  $1000\text{ cm}^{-3}$  in every air mass category. Air masses with a marine origin contained a pronounced Aitken mode, whereas high accumulation mode concentrations were measured in continental air masses. The latter is indicative of aged anthropogenic pollution, as supported also by the relatively high total particle volume concentrations. The average number concentration of activated particles varied between 212 and  $480\text{ cm}^{-3}$  with the lowest values observed for marine air masses. This is expected because the marine-influenced cases contained the smallest number of accumulation mode particles which activated with >50% efficiency during the experiment (Table 1). The contribution of the Aitken mode particles to the total number of activated particles was highest in the marine air masses (36%), and lowest in the European air masses (18%). The results imply that Aitken mode particles formed a minor but non-negligible source of cloud droplets during the experiment, even though their activation efficiency was low: between 10–27% of the Aitken mode particles activated to cloud droplets on average. Also, it can be seen that the aerosol properties measured in “MIX” air masses generally fell between those of “EUR” and “MARINE”, which lends confidence to the trajectory-based air mass classification.

The average HG factors for cloudy periods, classified according to the particle size and air mass history, are shown in Table 2. The average HG factors varied between 1.12 and 1.42, and generally increased with the increasing particle size. Size-wise, the average growth factors were 1.16, 1.23, 1.25, 1.30 and 1.38 for particles having a diameter of 30, 50, 80, 100 and 150 nm, respectively. The highest and lowest HG

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factors were measured in “EUR” and “MARINE” air masses, respectively, whereas the measured values in “MIX” fell between those for the other two air mass types. The size-averaged HG factors were 1.35, 1.18 and 1.28 in “EUR”, “MARINE” and “MIX” air masses, respectively. Here it is worth noting that the humidified particle distributions exhibited only a single mode, whereas two distinct hygroscopic modes, termed typically as less and more hygroscopic modes, are often observed in ambient measurements (e.g. Cocker et al., 2001; Ferron et al., 2005).

## 4 Data analysis

Four cloud events were chosen for a detailed case study using the approach described in Sect. 2. The aim of the case studies was to estimate the maximum supersaturation reached during the formation of observed cloud(s) and to provide more information on the role played by the mixing state and chemical composition in the cloud nucleating ability of atmospheric particles.

### 4.1 Selected cases

The initial data set was screened according to the following criteria to assure that the chosen cases would meet the assumptions behind the applied method: 1) no rainfall took place during the period and 2) the fraction of activated particles was >90% at the diameter range >400 nm. The former criterion was applied because raindrops scavenge interstitial particles and may thus bias the measurements (Komppula et al., 2005). The latter criterion was applied because smaller activated fractions are taken to be indicative of entrainment (Noone et al., 1992; Hallberg et al., 1998; Mertes et al., 2005) and there is no information on the extent of entrainment which complicates quantitative analysis (Sect. 2.2.2). Another factor that could explain a partial activation of large particles is the presence of insoluble black carbon or dust particles (Hallberg et al., 1998). This cannot be verified, however, since the hygroscopic measurements cov-

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ered only dry diameters up to 150 nm and thus associated uncertainties are avoided by applying the criterion. After the screening, four cases were selected from the remaining data set so that they cover the range of ambient conditions met during the experiment. Taken together, the cases represent different air masses, degrees of pollution as well as the scale of the observed aerosol and their activation characteristics (Table 3). These features and results of the case studies are discussed in detail in the following.

## 4.2 Hygroscopic properties

The dry diameters measured by the H-TDMA instrument and those measured by the DMPS set-up did not overlap, and therefore the hygroscopicity data were extra- and interpolated to the diameters at which the activation measurements were conducted. To this end, the available H-TDMA data for each case was averaged over the considered time interval (Table 3). Here the smallest measured particle size (30 nm) was not considered since particles with sizes <50 nm are not expected to participate in the cloud formation under conditions relevant to this study. As described in Sect. 3, the humidified distributions could be fitted accurately with a single log-normal function.

The hygroscopic growth measurements and the corresponding fits are shown in Fig. 2. The function used in the fitting is of the following form  $f(x)=ax^b$ , where  $x$  is the mean HG factor or the geometric standard deviation (GSD) of a humidified distribution, and  $a$  and  $b$  are the fitting parameters. As can be seen, the performance of the fits was good and the extrapolated values are in a reasonable range. It is also worth noting that in all cases, particles are less hygroscopic than pure ammonium sulfate particles which have a HG factor of  $\sim 1.77$  at the water activity of 0.90 (Tang and Munkelwitz, 1997).

One issue neglected so far is the fact that particles selected by the first DMA in an H-TDMA instrument do not have exactly the same size but consist of a width of sizes as determined by the transfer function of the instrument (Knutson and Whitby, 1975; Stolzenburg, 1988; Reischl, 1991). On the other hand, the considerations presented

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above assume that the only factor behind a spread in humidified particle distributions is the particle chemical composition. In order to quantify the relationship between the “real” GSD that corresponds to the humidified distribution of particles having a certain size  $d_p$  and the observational GSD, which may reflect the chemical composition of particles with varying sizes, a series of calculations was performed as follows. First, we calculated the transfer functions characteristic to the DMA used in the experiments and fitted a log-normal function to the resulting particle size distributions. The values of GSDs of particle distributions passing through the first DMA,  $GSD_1$ , were calculated to be 1.025, 1.026 and 1.028 for particles having a diameter 50, 100 and 150 nm, respectively. In the next step, the particle size distributions were treated as probability distributions and all size-selected particles passing through the first DMA were assumed to have the same chemical composition. Accordingly, it is easy to show that the “real” distribution of HG factors of particles with a size  $d_p$  is  $Y=X_1^{-1}X_2$  where  $X_1$  is the probability distribution determined by the transfer function and  $X_2$  is the distribution corresponding to the measured HG factors. We calculated the distribution  $Y$  using Monte Carlo methods for different values of  $GSD_1$ , the geometric standard deviation of the measured distribution,  $GSD_2$ , and the mean HG factor of the humidified distribution. The results show that errors in calculating the “real” GSD are below 0.01 (in absolute scale) when  $GSD_1 < 1.04$  and  $GSD_2 > 1.05$  and that they are insensitive to the mean HG factor. Moreover, the absolute errors in the “real” value of GSD reach a value of 0.014 when  $GSD_1 = 1.04$  and  $GSD_2 = 1.05$ . Keeping in mind that the values of  $GSD_1$  and  $GSD_2$  were consistently below 1.03 and above 1.05, respectively, in our case, the results show that the issue considered here can be neglected in the subsequent considerations.

### 4.3 Particle activation

The measured fractions of activated particles are shown in Fig. 3 for each case. The activation profiles share similar features with those of previous ground-based particle activation studies (Svenningsson et al., 1994, 1997; Martinsson et al., 1999; Mertes



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et al., 2005; Targino et al., 2007): the activated fractions start increase rapidly with the particle size at the size range 50–80 nm, reach >80% at the diameter range 150–200 nm and exhibit only slight increase at the diameter range >200 nm. In the considered cases,  $D_{50}$ , i.e. the diameter at which particles activate with 50% efficiency, was estimated to lie in the range 112–141 nm, being lowest in the case A and highest in D. In comparison,  $D_{50}$  varied between 88 and 215 nm during the whole experiment. It can be seen that the activated fraction fluctuated somewhat at the diameter range <70–80 nm. This is attributed to low particle concentrations at the smallest size bins and to relatively rapid coagulation scavenging of ultrafine particles by cloud droplets (Komppula et al., 2005).

The peak supersaturations reached in the observed cloudy air masses,  $s_{\max}$ , were estimated as described in Sect. 2.1. The values of  $s_{\max}$  as well as the predicted fractions of activated particles, calculated using Eqs. (5) and B1, and the total number of activated particles, calculated with Eq. (7), are shown in Fig. 3. The value of  $s_{\max}$  is seen to vary between 0.18 and 0.26%, being smallest in the case C and largest in the case A. This range compares well with the values reported from previous ground based particle activation studies (Sveningsson et al., 1994, 1997; Martinsson et al., 1999; Mertes et al., 2005). Overall, the predicted fractions of activated particles were within or close to the error bars, excluding the smallest sizes in which the activated fractions fluctuated slightly due to the reasons discussed above. The predicted and observational number of particles incorporated into cloud droplets were within 9.6, 0.8, 15.7 and 17.1% in the cases A, B, C and D, respectively, which shows also good agreement (see Table 3). By comparing the size-dependent trends in the particle activation it can be seen that the method can also explain the case-varying steepness of the increase in the activated fractions in the size range  $\sim 100$ –200 nm and the smaller, as compared to other cases, activated fractions of >200 nm particles in the case D. The latter feature results from the smallest HG factors exhibited by the particles in the case D (Fig. 2). However, the calculations tended to overpredict slightly the measured fractions of activated particles especially in the size range >150 nm. A possible reason is

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that the hygroscopic properties of particles were not measured at this size range but had to be extrapolated and are therefore uncertain (Fig. 2). Additionally, there were only a small number of particles at the large sizes which makes the results susceptible to fluctuations in the measured particle concentrations.

We note that the presented estimates of  $s_{\max}$  apply only to the measured air masses and may not be representative of the sampled cloud(s) as a whole. For example, in ground-based studies, the measurements system may be located so close to cloud base that higher supersaturations than the observations imply are reached higher in the cloud air.

#### 4.4 Sensitivity studies

A set of sensitivity calculations was performed to investigate how the particle activation behavior would change as a response to changes in the particle water uptake properties under the assumption that the peak supersaturation of water vapour is not affected. The calculations were done by varying the mean HG factors and/or GSDs while keeping the values of  $s_{\max}$ , estimated as described in the previous section, constant.

The first sensitivity study considered the importance of size-dependent variability in the particle hygroscopicity. This question has relevance, for example, to developing a simple and accurate parameterization of the cloud formation that can be used in atmospheric models (Broekhuizen et al., 2006; Medina et al., 2007; Kivekäs et al., 2008). Here we repeated the calculations based on Eqs. (5) and (7) but by using the size-averaged value(s) of the 1) mean HG factors, 2) GSDs of the humidified distributions, and 3) both mean HG factors and GSDs. The results are illustrated for the case A in Fig. 4 and the changes in the predicted number of activated particles are shown in Table 4. As can be seen, only small changes in the size dependent particle activation or in the total number of activated particles were predicted to occur as a result of size averaging. The relative changes in the total number of activated particles remained <10% even for the case A which was most sensitive case in this regard.

The second sensitivity study focused on the impact of hygroscopicity to the particle

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cloud-nucleating ability. The calculations were motivated by several recent studies that have addressed this topic (McFiggans et al., 2005; Dusek et al., 2006; Ervens et al., 2007; Hudson et al., 2007) and they were performed by increasing or decreasing the fit-based mean hygroscopic growth factors (Fig. 2) by 5 or 10% and repeating the activation calculations using Eq. (5). In comparison, 5 and 10% changes in the HG growth factor translate to changes of ~16 and 33% in the water-soluble volume fraction. The resulting size dependent activation is illustrated for case A in Fig. 5 and the number of activated particles are shown in Table 4 for each case. Figure 5 shows that the size-dependent particle activation is highly sensitive to the particle hygroscopicity: even 5% change in the mean HG factor resulted in clear deviations from the base case calculations. Overall, the largest changes are seen in the size range 100–200 nm, and the value of  $D_{50}$  is predicted to vary between ~90 and 150 nm. As seen further from Table 4, large changes in the particle activation behavior were also predicted to occur in other cases. The calculated number concentrations of activated particles varied up to a factor of 2.8, 1.6, 1.8 and 3.1 in the cases A, B, C and D, respectively. In addition, the cases D and B exhibited the largest and smallest sensitivity to the particle hygroscopicity, respectively, and these cases also featured the largest and smallest measured hygroscopic growth factors, respectively, among the considered cases (Fig. 2). Taken together, these results suggest that the CCN ability of a particle population having relatively small or moderate HG factors, as the case was here, is highly sensitive to the particle hygroscopicity.

The third and last sensitivity study investigated the influence of the particle mixing state on the particle activation, which is also a topic of active research (Rissmann et al., 2006; Stroud et al., 2007). The influence of the mixing state was quantified by increasing the values of the fitted GSDs, shown in Fig. 2, by 5 or 10% as well as by decreasing them by 5%, and carrying out the calculations as previously. Larger decreases are not considered here because they would lead to unrealistically small GSD values. The rationale behind this approach is that wider humidified distributions reflect more heterogeneous chemical composition of particles whereas the opposite holds for

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narrower distributions. The predicted size-dependent activation behavior is illustrated in Fig. 6 for case A and the number activated particles are shown in Table 4. Most notable feature displayed by Fig. 6 is that the heterogeneity in the chemical composition is reflected in the steepness of the activation curves: when only a narrow range of critical supersaturations are present in each particle size, the activation curve resembles a step function whereas more gradual changes are seen when each particle size contains a spread of critical supersaturations. All curves are also seen to intersect at the point corresponding to  $D_{50}$  since the shape of the distribution of critical supersaturations does not play a role at 50% activation efficiency. Furthermore, Table 4 shows that the total number of activated particles is insensitive to the value of GSD. This is because the activated fractions of particles with sizes smaller and larger than  $D_{50}$  respond in an opposite manner to changes in GSD. To conclude with, non-negligible changes in size-dependent particle activation are predicted to take place in response to changes in the particle mixing state, and thus the steepness of the “S”-shaped activation profiles may carry information on the particle mixing state, as suggested previously by Martinsson et al. (1999) and Mertes et al. (2005).

It is worth noting that  $s_{\max}$  was not adjusted to changing aerosol properties but was kept constant in all sensitivity scenarios. For example, if particles are more hygroscopic, they will deplete more water vapor during the cloud formation and thereby increased particle hygroscopicity leads to smaller peak supersaturations and hence smaller number of activated particles. As discussed in Sect. 2.2.3, however, accounting for such “dampening” dynamical responses is beyond the scope of the method and therefore the presented estimates on the importance of the considered factors represent probably upper limit estimates.

## 5 Summary and conclusions

We derived an algorithm for analyzing the influence of the particle chemical composition and mixing state (in terms of the water uptake) on the cloud nucleation ability of

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atmospheric particles on the basis of size-resolved measurements of particle activation and hygroscopic growth. The method provides also a quantitative estimate of the peak supersaturation of water vapour reached during the formation of the observed cloud(s). No assumptions regarding the shape of the humidified particles distributions are required, but if particle diameters for which activation and water uptake measurements were conducted are not the same, inter- and/or extrapolation of the measurement data is necessary.

The method was applied in interpreting results of a measurement campaign that focused on aerosol-cloud interactions taking place at a subarctic background site. A set of case studies was conducted, and the size-dependent activation trends could be successfully explained by a maximum supersaturation that varied between 0.18 and 0.26% depending on the case. There was no indication on the presence of organic films on particles that would be able to suppress condensation of water vapour nor any clear evidence on variations in the peak supersaturation, in contrast to the results of a study by Mertes et al. (2005). Furthermore, the conducted analysis showed that the activated fractions and the total number of particles acting as CCN are expected to be highly sensitive to the particle hygroscopicity; for example, the latter quantity varied by a factor of 1.8–3.1, depending on the case, when the mean hygroscopic growth factors were varied by 10%. Here it should be noted that the particles were only moderately hygroscopic with hygroscopic growth factors varying between 1.1 and 1.4. Another important conclusion that can be drawn from the case studies is that the size-dependent activation profiles may carries information on the mixing state of particles. This was demonstrated by a sensitivity study where the parameter describing the spread in the water-solubility of particles having the same dry size was varied. The results show that the steepness of the calculated activation curves changes notably in response to changes in the mixing state.

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## Appendix A

### 5 List of the key symbols used in Sect. 2

$AF_i$  – The measured fraction of activated particles in the size class  $i$ .

$AF_i^*$  – The predicted fraction of activated particles in the size class  $i$ .

$CN_{\text{tot},i}$  – The measured particle number concentration in the size class  $i$ .

10  $CN_i(s_c < s)$  – The number of particles having a critical supersaturation below  $s$  in the size class  $i$ .

$d_{p,i}$  – The dry diameter of particles in the size class  $i$ .

$GR_i$  – The normalized cumulative distribution of the hygroscopic growth factors of the particles in the size class  $i$ .

15  $K_i$  – The fraction of particles in the size class  $i$  that are activated according to the Köhler theory at a supersaturation  $s$  ( $K_i(s) = CN_i(s_c | s) / CN_{\text{tot},i}$ ).

$s_{\text{max}}$  – The maximum supersaturation, estimated using the approach described in Sect. 2, that is reached in the observed cloud(s).

20  $SF_i$  – The normalized cumulative distribution of the water-soluble volume fraction of particles in the size class  $i$ .

## Appendix B

### Expressions for the cumulative distributions of hygroscopic growth factors

25 The humidified particle size distributions measured by the second DMA in H-TDMA instrument can be often described by a log-normal or Gaussian distribution(s). In the

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former case, the function  $GR_i$  in (5) is the following:

$$GR_i[h_i(s)] = \frac{1}{2} + \frac{1}{2} \operatorname{erf} \left[ \frac{\ln[h_i(s)] - \ln(g^*)}{\sqrt{2} \ln(\sigma_g)} \right], \quad (\text{B1})$$

where  $g^*$  and  $\sigma_g$  are the mean growth factor and the geometric standard deviation of the mode, respectively. When the humidified distribution has a Gaussian profile,  $GR_i$  has the following functional form:

$$GR_i[h_i(s)] = \frac{1}{2} + \frac{1}{2} \operatorname{erf} \left[ \frac{h_i(s) - g^*}{\sqrt{2} \sigma_g} \right] \quad (\text{B2})$$

Equations (B1) and (B2) can be generalized to cases where the particles exhibit multi-modal growth behavior by summing the corresponding cumulative distributions and weighting them with the corresponding fractional particle concentrations. Furthermore, it should be noted that if the diameters which are selected by the first DMA in H-TDMA instrument do not match the diameters at which the activated fractions are determined, the observational values of  $g^*$  and  $\sigma_g$  need to be extra- or interpolated to these diameters. An example of such fitting procedure is given in Sect. 5.

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**Table 1.** Time-averaged aerosol and cloud activation characteristics according to the air mass history. Shown are the temporal length of the data sets, total particle volume ( $V_{\text{tot}}$ ), total particle number concentration ( $N_{\text{tot}}$ ), number of Aitken mode particles and the corresponding fraction of the total number concentration ( $N_{\text{ait}}$  and  $N_{\text{ait}}/N_{\text{tot}}$ , respectively), the number of activated particles and the average activated fraction ( $N_{\text{act}}$  and  $N_{\text{act}}/N_{\text{tot}}$ , respectively) and finally the fractional contribution of the Aitken and accumulation mode particles to the total number of activated particles ( $A_{\text{ait}}/N_{\text{act}}$  and  $A_{\text{acc}}/N_{\text{act}}$ , respectively).

Air mass	No. of h	$V_{\text{tot}}$ ( $\mu\text{m}^3\text{cm}^{-3}$ )	$N_{\text{tot}}$ ( $\text{cm}^{-3}$ )	$N_{\text{ait}}$ ( $\text{cm}^{-3}$ )	$N_{\text{ait}}/N_{\text{tot}}$ (%)	$N_{\text{act}}$ ( $\text{cm}^{-3}$ )	$N_{\text{act}}/N_{\text{tot}}$ (%)	$A_{\text{ait}}/N_{\text{act}}$ (%)
EUR	19	7.1	1041	324	31	480	46	18
MARINE	22	1.1	961	745	78	212	22	36
MIX	58	4.1	939	551	59	298	32	20

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**Table 2.** Average hygroscopic growth factors of particles during the cloudy periods of the experiment.

	Diameter				
	30	50	80	100	150
Air mass					
EUR	1.22	1.33	1.37	1.36	1.42
MARINE	1.12	1.14	1.15	1.22	1.26
MIX	1.18	1.21	1.24	1.31	1.42

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**Table 3.** Some general features of the analyzed cases. Shown are the date and time period of the cases, the air mass type, the total particle number concentration and total volume ( $N_{\text{tot}}$  and  $V_{\text{tot}}$ , respectively), Aitken mode number concentration and the corresponding fraction of the total number concentration ( $N_{\text{ait}}$  and  $N_{\text{ait}}/N_{\text{tot}}$ , respectively) as well as the number of activated particles and the fraction of activated particles ( $N_{\text{act}}$  and  $N_{\text{act}}/N_{\text{tot}}$ , respectively). Furthermore, the last two columns show the fractional contribution of Aitken mode particles to activated cloud droplets ( $A_{\text{ait}}/N_{\text{act}}$ ) and hygroscopic growth factors of particles having a 100 nm dry diameter.

Case	Date	Time	Air Mass	$N_{\text{tot}}$ ( $\text{cm}^{-3}$ )	$V_{\text{tot}}$ ( $\mu\text{m}^3/\text{cm}^3$ )	$N_{\text{ait}}$ ( $\text{cm}^{-3}$ )	$N_{\text{ait}}/N_{\text{tot}}$ (%)	$N_{\text{act}}$ ( $\text{cm}^{-3}$ )	$N_{\text{act}}/N_{\text{tot}}$ (%)	$A_{\text{ait}}/N_{\text{act}}$ (%)	HGF 100 nm
A	23.9.	04:00–07:00	MARINE	1834	1.8	1514	83	228	12	50	1.12
B	28.9.	23:00–24:00	MIX	930	4.6	457	49	399	43	16	1.31
C	29.9.	07:00–08:00	EUR	479	1.6	280	58	140	29	27	1.26
D	29.9.	19:00–20:00	MARINE	279	0.4	225	81	36	13	52	1.13

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**Table 4.** The total number of activated particles in different sensitivity scenarios and cases.

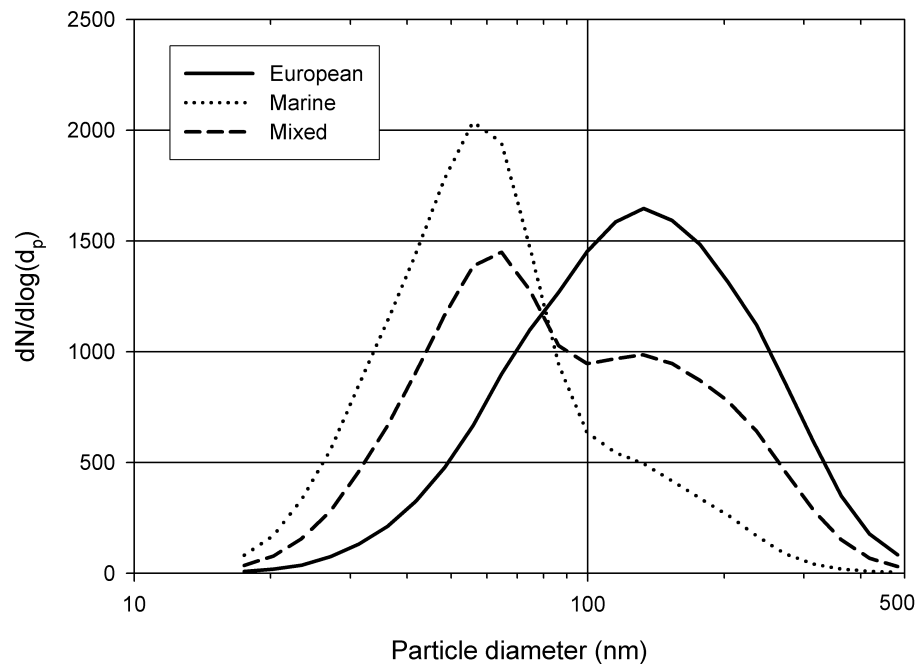
	Case			
	A	B	C	D
Base case	224	402	118	29
Size averaged mean HGF	228	401	117	28
Size averaged GSD	233	403	118	30
Size averaged mean HGF & GSD	241	403	116	29
1.1xHGF	338	475	138	43
1.05xHGF	280	440	157	37
0.95xHGF	170	357	97	22
0.9xHGF	119	305	75	14
1.1xGSD	250	387	119	30
1.05xGSD	232	393	121	31
0.95xGSD	232	409	119	31

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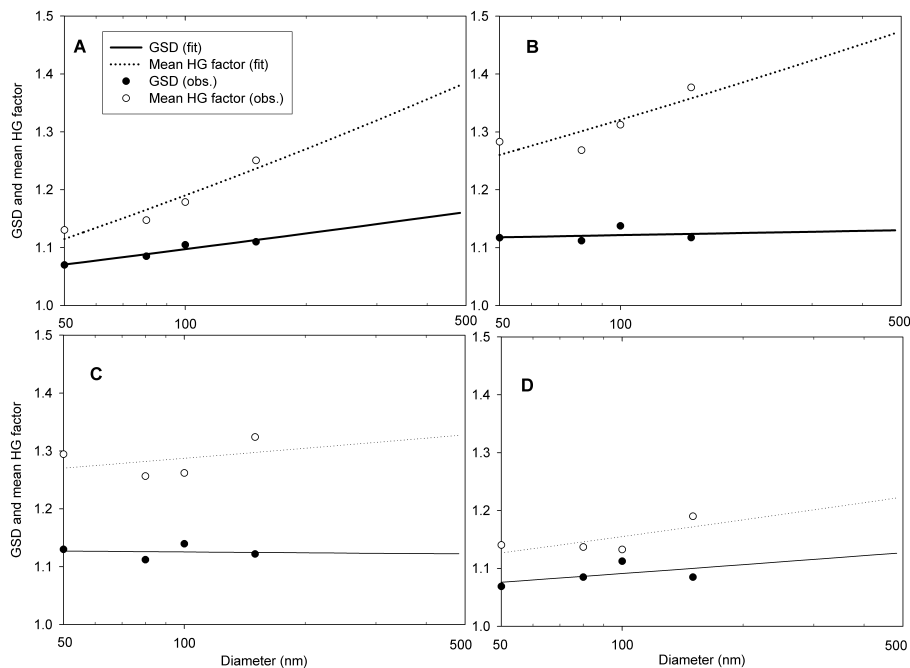


**Fig. 1.** Time-averaged dry particle distributions, classified according to the air mass, during cloud events.

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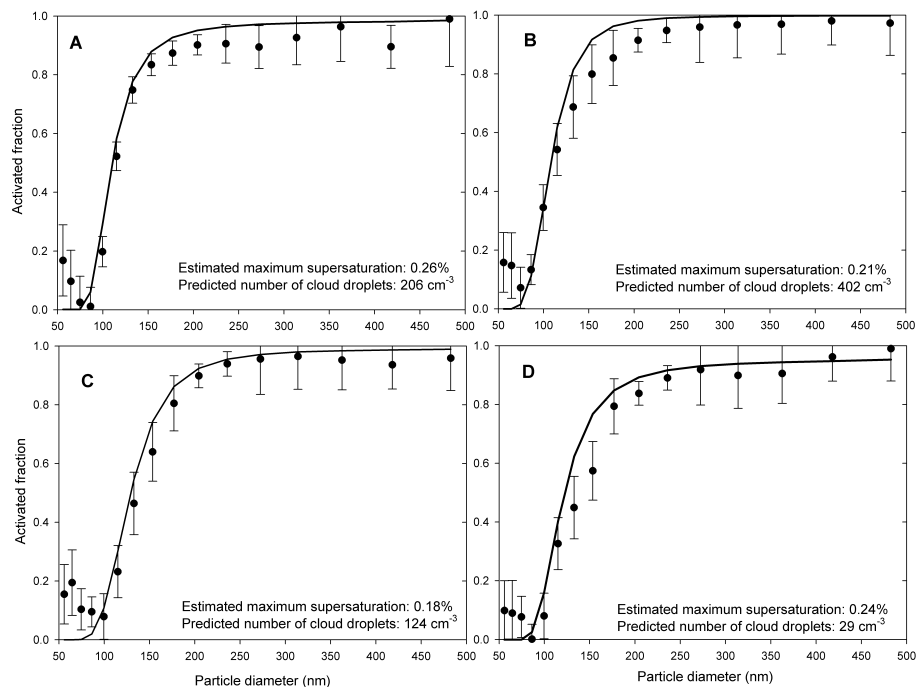


**Fig. 2.** The mean hygroscopic growth factors and their geometric standard deviation (GSD) based on the observed distributions (hollow and solid symbols, respectively) and the corresponding fits (dashed and solid lines, respectively).

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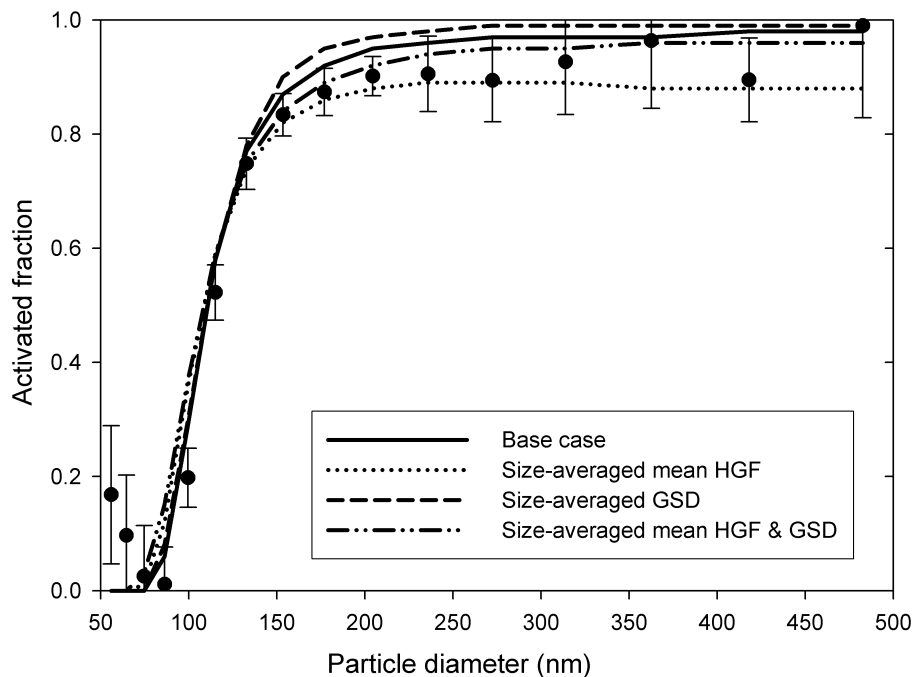


**Fig. 3.** The measured fractions of activated particles (symbols) and the predicted activated fractions (solid lines) for the considered cases. The error bars are based on the size-segregated concentration difference observed between the DMPSSs measuring at different inlets during the non-cloudy period prior and after the cloud event. Also shown are the estimated maximum supersaturations and predicted number of cloud droplets for each case.

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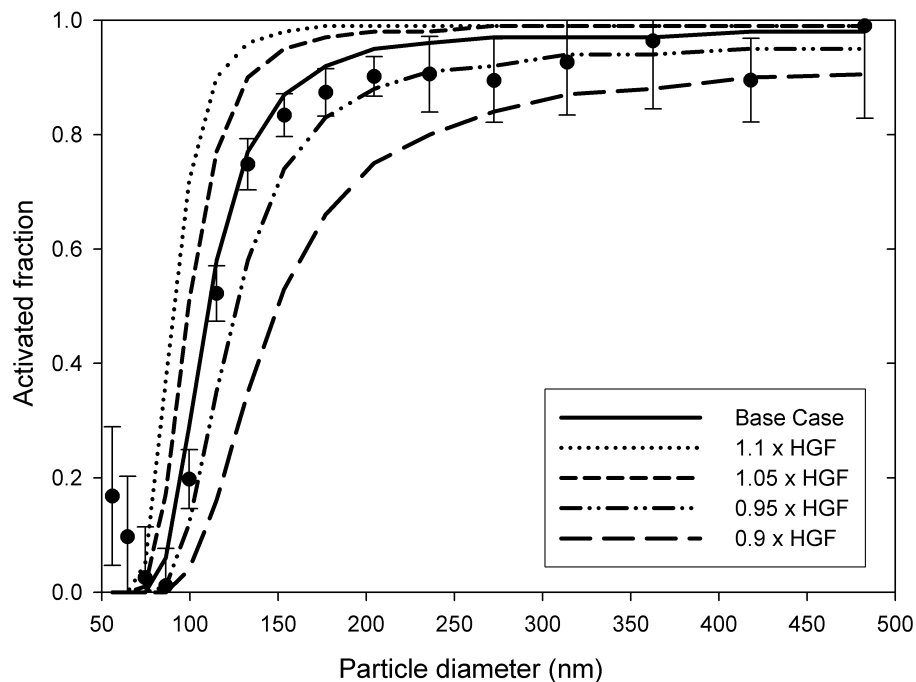


**Fig. 4.** The influence of size-averaging the particle hygroscopic properties on the particle activation for the case A. The measurements are displayed with symbols and the calculations are showed with lines as indicated in the legend.

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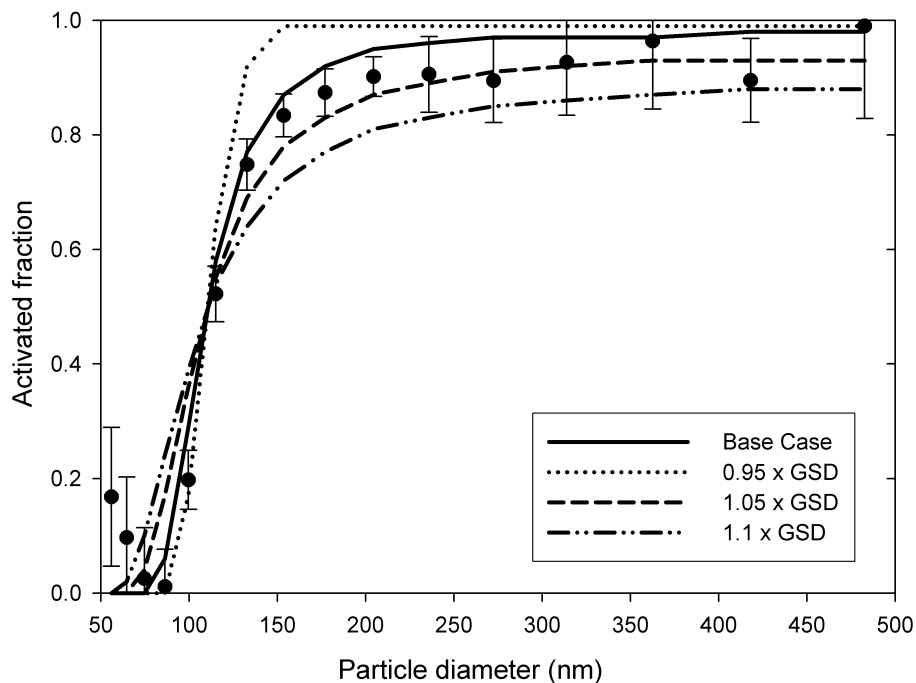


**Fig. 5.** Sensitivity of the particle activation to the mean HG factors for the case A. The sensitivity calculations were performed by multiplying the fit-based mean HG factors by a constant varying between 0.9 and 1.1 (shown in the legend).

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**Fig. 6.** Sensitivity of the particle activation to GSDs of the HG factor distributions for the case A. The sensitivity calculations were performed by multiplying the fit-based mean HG factors by a constant varying between 0.95 and 1.1 (shown in the legend).

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