



The condensing  
vapors driving new  
particle growth to  
CCN sizes

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# Some insights into the condensing vapors driving new particle growth to CCN sizes on the basis of hygroscopicity measurements

Z. J. Wu<sup>1,2</sup>, L. Poulain<sup>2</sup>, W. Birmili<sup>2</sup>, J. Größ<sup>2</sup>, N. Niedermeier<sup>2</sup>, Z. B. Wang<sup>3</sup>, H. Herrmann<sup>2</sup>, and A. Wiedensohler<sup>2</sup>

<sup>1</sup>College of Environmental Sciences and Engineering, Peking University, 100871, Beijing, China

<sup>2</sup>Leibniz Institute for Tropospheric Research, 04318, Leipzig, Germany

<sup>3</sup>Multiphase Chemistry Department, Max Planck Institute for Chemistry, Mainz 55128, Germany

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Correspondence to: Z. J. Wu (zhijunwu@pku.edu.cn)

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## Abstract

New particle formation (NPF) and growth is an important source of cloud condensation nuclei (CCN). In this study, we investigated the potential chemical species driving new particle growth to the CCN sizes on the basis of particle hygroscopicity measurements carried out at the research station Melpitz, Germany. The predicted CCN number concentrations using  $\kappa$ -Köhler theory were analyzed to assess the contribution of NPF to possible CCN. Three consecutive NPF events occurred during summertime were chosen as examples to perform the study. Hygroscopicity measurements showed that the  $(\text{NH}_4)_2\text{SO}_4$ -equivalent water-soluble fraction respectively accounts for 20 and 16 % of 50 and 75 nm particles during the NPF events. Numerical analysis showed the ratios of  $\text{H}_2\text{SO}_4$  condensational growth to the observed particle growth were 20 and 13 % for 50 and 75 nm newly formed particles, respectively. Both hygroscopicity measurements and numerical analysis confirmed that organic compounds were major contributors driving particle growth to CCN sizes. The critical diameters at different supersaturations estimated using AMS data and  $\kappa$ -Köhler theory increased significantly during the later course of NPF events. This indicated that the enhanced organic mass fraction caused a reduction in CCN efficiency of newly formed particles. Our results implied that the CCN production associated with atmospheric nucleation may be overestimated if assuming that newly formed particles can serve as CCN in case they grow to a fixed particle size, which was used in some previous studies, especially for organic-rich environments. In our study, the enhancement in CCN number concentration associated with individual NPF events have been 63, 66, 69 % for supersaturation 0.1, 0.4, and 0.6 %, respectively.

## 1 Introduction

The formation of new particles from gaseous precursors and their subsequent growth represent a key stage in the lifecycle of atmospheric aerosol particles. This new parti-

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cle formation (NPF) process represents an important source of atmospheric particles and possibly also for the number concentration of potential cloud condensation nuclei (CCN) (Spracklen et al., 2008; Wiedensohler et al., 2009; Wang and Penner, 2009; Laaksonen et al., 2005; Yue et al., 2011; Kazil et al., 2010; Sotiropoulou et al., 2006; Laakso et al., 2013). NPF has thus the potential to influence climatologically important processes such as precipitation patterns and Earth's energy balance (Paasonen et al., 2013). The contribution of atmospheric nucleation to the global CCN budget remains the single largest uncertainty, which, together with our general poor understanding of aerosol-cloud interactions, results in major uncertainties in the radioactive forcing by atmospheric aerosols (Kerminen et al., 2012). Recent model studies (Spracklen et al., 2008; Merikanto et al., 2009; Westervelt et al., 2014) have attempted to elaborate on the connection between NPF and CCN production, a process that is sensitive to a number of environmental factors.

Freshly formed particles are about several nanometers in diameter, and they must grow tens of nanometers in order to serve as a potential CCN (Dusek et al., 2006; Kerminen et al., 2012). Apparently, the nucleation rate, the particle growth and the rate by which growing particles are removed by coagulation or deposition greatly influence the CCN production associated with atmospheric nucleation (Kuang et al., 2009; Kerminen et al., 2004). From the point of view of chemical species, both sulfuric acid and organics contribute to the subsequent particle growth after nucleation (Smith et al., 2004; Pierce et al., 2011; Ehn et al., 2007; Kulmala et al., 2004; Brus et al., 2011; Kulmala et al., 2006; Sipilä et al., 2010; Zhang et al., 2004; Kiendler-Scharr et al., 2009; Wang et al., 2010; Ristovski et al., 2010). The contribution of sulfate and organics in the particle growth seems to be strongly depending on the location (e.g. Yue et al., 2010; Boy et al., 2005). For example, sulfuric acid fully explains the particle growth observed in the polluted areas, Atlanta, USA (Stolzenburg et al., 2005), while it represents only 10 % in Boreal forest area (Boy et al., 2005). Sulfuric acid and organics, which fraction is the leading component in particles after growing to CCN size strongly affects their CCN efficiency (e.g. Dusek et al., 2010).

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Due to the differences in hygroscopicity of sulfuric acid and/or its ammonium salts and secondary organic compounds (Virkkula et al., 1999; Varutbangkul et al., 2006; Tang and Munkelwitz, 1994), hygroscopicity measurements during a NPF events can provide insight into the changes in condensing vapor properties and chemical composition of newly formed particles (Hämeri et al., 2001; Ehn et al., 2007; Ristovski et al., 2010). In this study, we investigate the potential chemical species driving new particle growth into CCN sizes by using experimental data on particle hygroscopicity and chemical composition measured in Melpitz, Germany. In addition, the production of potential CCN associated with the NPF event will be evaluated.

## 2 Experiments

Atmospheric measurements were performed at the research station Melpitz, Germany (51.54° N, 12.93° E, 86 m above sea level). The atmospheric aerosol observed at Melpitz can be regarded as representative for Central European background conditions (Birmili et al., 2009). An account of the NPF process at Melpitz and its relationship with precursor gases and meteorology can be found in Größ et al. (2015).

The data of this study were collected during the European Integrated Project on Aerosol Cloud Climate Air Quality Interactions (EUCAARI, Kulmala et al., 2009) intensive field campaign in 2008. All instruments were set up in the same container laboratory and utilized the same air inlet. This inlet line consisted of a PM<sub>10</sub> Anderson impactor located approximately 6 m a.g.l. and directly followed by an automatic aerosol diffusion dryer (Tuch et al., 2009) that maintained the relative humidity in the sampling line below 30 %. Particle hygroscopicity, particle number size distribution, and chemical composition of non-refractory PM<sub>1</sub> were determined using a hygroscopicity tandem differential mobility particle analyzer (H-TDMA), a Twin Differential Mobility Particle Sizer (TDMPs), and a High Resolution Time-of-flight Aerosol Mass Spectrometer (HR-ToF-AMS), respectively.

## 2.1 Particle hygroscopicity measurements

The H-TDMA used in this study has been illustrated in previous publications in detail (Wu et al., 2011; Massling et al., 2003), and complies to the instrumental standards prescribed in Massling et al. (2011). The H-TDMA consists of three main parts: (1) a Differential Mobility Analyzer (DMA1) that selects quasi-monodisperse particles at a relative humidity below 10 %, and a Condensation Particle Counter (CPC1) that measures the particle number concentration leaving DMA1 at the selected particle size, (2) an aerosol humidifier conditioning the particles selected by DMA1 to a defined relative humidity (RH), (3) the second DMA (DMA2) coupled with another condensation particle counter (CPC2) to measure the number size distributions of the humidified aerosol. DMA2 and the aerosol humidification are placed in a temperature-controlled box. Hygroscopicity scans with 100 nm ammonium sulfate particles were performed frequently to analyze the stability of the relative humidity of 90 % in the second DMA. Hygroscopicity scans with a deviation of more than 3 % in relative humidity from the set-point of 90 % were not considered for further analysis.

The hygroscopic growth factor (HGF) is defined as the ratio of the particle mobility diameter,  $D_p(\text{RH})$ , at a given RH to the dry diameter,  $D_{p_d}$ :

$$\text{HGF}(\text{RH}) = \frac{D_p(\text{RH})}{D_{p_d}} \quad (1)$$

The TDMA<sub>inv</sub> method developed by Gysel et al. (2009) was used to invert the H-TDMA data. Dry scans (RH < 10 %) were used to calibrate a possible offset between DMA1 and DMA2 and define the width of the HTDMA's transfer function (Gysel et al., 2009).

## 2.2 Particle chemical composition

The Aerodyne HR-ToF-AMS (here simply referred to as AMS) (DeCarlo et al., 2006) was typically operated with a time resolution of 5 min. Due to the 600 °C surface tem-

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perature of the vaporizer, the AMS only analyzes the non-refractory chemical composition of the particles. Soot, crustal material, and sea-salt cannot be detected. Therefore, based on the transmission efficiency of the aerodynamic lenses and the detected compounds, the AMS can provide the size-resolved chemical composition of sub-micrometer non-refractory aerosol particle fraction (NR-PM<sub>1</sub>) (Canagaratna et al., 2007). The vacuum aerodynamic diameter for AMS measurements was converted to mobility diameter by division of AMS vacuum aerodynamic diameter by the estimated particle density (1600 kg m<sup>-3</sup>). Hereafter, the mobility diameter is used in AMS data below.

### 2.3 Particle number size distribution

A TDMPS was deployed to measure particle number size distributions from 3–800 nm mobility diameter with a time resolution of 10 min (Birmili et al., 1999). The system consists of two Differential Mobility Analyzers (DMA, Hauke-type) and two Condensation Particle Counters (CPC, TSI model 3010 and TSI model 3025). The sheath air is circulated in closed loops for both DMAs. Evaluation of particle number size distributions includes a multiple charge inversion, the CPC efficiency and diffusional losses in the DMA and all internal and external sampling lines according to the recommendations in (Wiedensohler et al., 2012).

## 3 Methodology

### 3.1 Derivation of the soluble particle fraction

Based on the Zdanovskii–Stokes–Robinson (ZSR) method (Zdanovskii, 1948; Stokes and Robinson, 1966), the HGF of a mixture can be estimated from the sum of HGF<sub>*i*</sub> of a pure component (*i*) time their respective volume fractions,  $\varepsilon_i$  (Malm and Kreidenweis,

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1997):

$$\text{HGF}_{\text{mixed}} = \left( \sum_i \varepsilon_i \text{HGF}_i^3 \right)^{1/3} \quad (2)$$

Here, we assume that two groups including soluble and insoluble fractions contribute to the particle growth (also refer to Ehn et al., 2007; Swietlicki et al., 1999). The soluble fraction is assumed as ammonium sulfate and the insoluble fraction as organic compounds. Then,  $\varepsilon$  of soluble fraction can be calculated by:

$$\varepsilon_{\text{soluble}} = \frac{\text{HGF}_{\text{measured}}^3 - 1}{\text{HGF}_{(\text{NH}_4)_2\text{SO}_4}^3 - 1} \quad (3)$$

where  $\text{HGF}_{\text{measured}}$  is the HGF of particle measured by HTDMA, and  $\text{HGF}_{(\text{NH}_4)_2\text{SO}_4}$  is the HGF of pure  $(\text{NH}_4)_2\text{SO}_4$  particle with the same size. When calculating  $\text{HGF}_{(\text{NH}_4)_2\text{SO}_4}$  in different diameters, the parameterizations for  $(\text{NH}_4)_2\text{SO}_4$  water activity developed by Potukuchi and Wexler (1995) and the density reported by Tang and Munkelwitz (1994) are used. The Kelvin term was considered in the calculation.

The assumption of insoluble organic fraction may lead to overestimate the soluble fraction because atmospherically relevant secondary organics typically have a growth factor larger than 1 (e.g., Varutbangkul et al., 2006). This implies that in the presence of several classes of hygroscopic substances,  $\varepsilon$  derived from Eq. (3) is only an “equivalent” soluble fraction (i.e. assuming ammonium sulfate as the only soluble substance).  $\varepsilon_{\text{soluble}}$  is therefore an upper estimate for the true soluble volume fraction. The advantage of using the soluble fraction term is to be able to analyze the particle hygroscopicity independently of differences in size. The uncertainty of the estimated soluble volume fraction is around 5%, which was derived from the measurement uncertainty of HGF (2.5%) according to the error propagation function.

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## 3.2 Calculation of CCN number concentration

According to the  $\kappa$ -Köhler theory (Petters and Kreidenweis, 2007), the single hygroscopicity parameter,  $\kappa$ , of a given internal mixture can be predicted by a simple mixing rule on the basis of chemical volume fractions  $\varepsilon_j$ :

$$\kappa_{\text{chem}} = \sum_i \varepsilon_i \kappa_i \quad (4)$$

Here,  $\kappa_i$  and  $\varepsilon_i$  are the hygroscopicity parameter and volume fraction for the individual (dry) component in the mixture with  $i$  the number of components in the mixture. We derive  $\varepsilon_j$  from particle chemical composition measured by AMS. Here, the size-resolved particle chemical composition is used to estimate the  $\kappa_{\text{chem}}$ .

The critical diameter ( $D_{\text{pcrit}}$ ) at which 50 % of the particles were activated is calculated from  $\kappa$ :

$$D_{\text{Pcrit}} = \left( \frac{4A^3}{27\kappa_{\text{chem}} \ln^2 S_C} \right)^{1/3} \quad (5)$$

$$A = \frac{4\sigma_{s/a} M_w}{RT\rho_w} \quad (6)$$

Here,  $\kappa_{\text{chem}}$  is calculated from size-resolved AMS data using the ZSR mixing rule.  $D_{\text{Pcrit}}$  is the critical diameter at which 50 % of the particles were activated at the supersaturation,  $S_C$ .  $\sigma_{s/a}$  is the droplet surface tension (assumed to be that of pure water,  $\sigma_{s/a} = 0.0728 \text{ N m}^{-2}$ ),  $M_w$  the molecular weight of water,  $\rho_w$  the density of liquid water ( $1000 \text{ kg m}^{-3}$ ),  $R$  the universal gas constant,  $T$  the absolute temperature.

The CCN number concentration is estimated by integrating the particle number size distribution from the critical diameter to the maximum diameter detected by TDMPMS (800 nm, above which the particle number concentration is generally negligible). As aforementioned, the size-resolved chemical composition was used to calculate the

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$\kappa$  values. The critical diameters, corresponding to supersaturation 0.2–0.7 %, roughly span from 50 to 110 nm in mobility diameter. Therefore, the chemical composition of 50–120 nm particles was calculated by integrating AMS mass size distribution data and used to estimate  $\kappa_{\text{chem}}$ . In the same way, the chemical composition of 150–200 nm particles is used to calculate  $\kappa_{\text{chem}}$  for the critical diameter of around 170 nm, which corresponds to a supersaturation of 0.1 %.

### 3.3 Estimation of H<sub>2</sub>SO<sub>4</sub> concentration

H<sub>2</sub>SO<sub>4</sub> concentrations were estimated using a modified version of the chemical mass balance model introduced by Weber et al. (1997), driven by solar radiation as a source of OH:

$$[\text{H}_2\text{SO}_4] = B \frac{[\cdot\text{OH}][\text{SO}_2]}{\text{CS}} \quad [\text{cm}^{-3}] \quad (7)$$

Here,  $[\cdot\text{OH}]$  is the hydroxyl radical concentration estimated from Eq. (8) in  $\text{cm}^{-3}$ .  $[\text{SO}_2]$  is the measured sulfur dioxide concentration in  $\text{cm}^{-3}$ .  $B$  is a constant related to the reaction rate of the two species. CS is the condensation sink (Pirjola et al., 1999) in  $\text{s}^{-1}$  calculated from the particle number size distribution adjusted to ambient relative humidity. For this adjustment, an empirical growth law based on one year of hygroscopicity measurements at Melpitz was used. The term  $B[\cdot\text{OH}][\text{SO}_2]$  represents the production term of H<sub>2</sub>SO<sub>4</sub>, and CS is referred to represent the loss rate of H<sub>2</sub>SO<sub>4</sub> on the pre-existing particles.  $B$  was derived by correlation analysis of measured and estimated  $[\text{H}_2\text{SO}_4]$  for 9 days during EUCAARI-2008 during which the data capture was satisfactory. Linear regression analysis yielded a value of  $27.49 \times 10^{-13} \text{ cm}^3 \text{ s}^{-1}$  for  $B$ .

$$[\cdot\text{OH}] = A \cdot \text{Rad} \quad [\text{cm}^{-3}] \quad (8)$$

where Rad is the global solar radiation flux in  $\text{W m}^{-2}$ .  $A$  was derived by linear regression of these parameters for the EUCAARI-2008 data set, yielding a value of  $6166 \text{ m}^2 \text{ W}^{-1}$

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for A. The calculation of  $\text{H}_2\text{SO}_4$  concentration was done within the works of Größ et al. (2015), with details provided therein.

### 3.4 Particle growth contributed by $\text{H}_2\text{SO}_4$ condensation

Theoretically, the vapor concentration required for growth rate of  $1 \text{ nm h}^{-1}$  in certain particle size ranges can be calculated according to (Nieminen et al., 2010):

$$C_{\text{GR}=1 \text{ nm h}^{-1}} = \frac{2\rho_v d_v}{\gamma m_v \Delta t} \cdot \sqrt{\frac{\pi m_v}{8kT}} \cdot \left[ \frac{2x_1 + 1}{x_1(x_1 + 1)} - \frac{2x_0 + 1}{x_0(x_0 + 1)} + 2 \ln \left( \frac{x_1(x_0 + 1)}{x_0(x_1 + 1)} \right) \right] \quad (9)$$

here  $x_0$  and  $x_1$  are the ratios of the vapour molecule diameter ( $d_v$ ) to the initial and final particle diameter, respectively. The mass ( $m_v$ ) and density ( $\rho_v$ ) of sulfuric acid applied in this study are  $135 \text{ amu}$  and  $1650 \text{ kg m}^{-3}$ , respectively, corresponding to hydrated sulfuric acid molecules (Kurtén et al., 2007). It should be mentioned that Eq. (9) was developed specially for particles with diameter of  $3\text{--}7 \text{ nm}$ . For larger particles ( $> 10 \text{ nm}$ ), this method give similar results to that calculated using the Fuchs–Sutugin approach (Nieminen et al., 2010). The calculated  $C_{\text{GR}=1 \text{ nm h}^{-1}, \text{H}_2\text{SO}_4}$  may be an underestimate because it is assumed that every sulfuric acid molecule colliding with the particle is attached to it which is not necessarily the case.

Then the growth rate contributed by sulfuric acid during the time period used for the determination of GR is calculated directly as:

$$\text{GR}_{\text{H}_2\text{SO}_4} = [\text{H}_2\text{SO}_4]_{\text{det}} / C_{\text{GR}=1 \text{ nm h}^{-1}, \text{H}_2\text{SO}_4} \quad (10)$$

where  $[\text{H}_2\text{SO}_4]_{\text{det}}$  is the median value from the measured sulfuric acid concentration during the timeframe for the determination of GR.

The observed growth rate can be presented as the sum of the growth rates due to  $\text{H}_2\text{SO}_4$  ( $\text{GR}_{\text{H}_2\text{SO}_4}$ ) and organic vapors ( $\text{GR}_{\text{org}}$ ) condensation (Paasonen et al., 2010):

$$\text{GR}_{\text{obs}} = \text{GR}_{\text{H}_2\text{SO}_4} + \text{GR}_{\text{Org}} \quad (11)$$

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By combining Eqs. (9)–(11), the overall particle volume change can be separated into two fraction contributing by  $\text{H}_2\text{SO}_4$  and organic vapors condensation.

## 4 Results

### 4.1 Particle formation and growth

5 The NPF events consecutively took place from, 5 to 7 June 2008, as displayed in Fig. 1a. The bursts in number concentration of 3–10 nm particles were observed associated with increasing ambient temperature, decreasing relative humidity, and increasing in estimated  $\text{H}_2\text{SO}_4$  concentration (shown in Fig. 1b). The particle formation rates ( $J_{3-25\text{nm}}$ ) were 13.5, 6.1, 9.3  $\text{cm}^{-3}\text{s}^{-1}$  on 5, 6, and 7 June, respectively. The highest  
10 formation rate was observed on 5 June corresponding to the highest  $\text{H}_2\text{SO}_4$  concentration. This was consistent with previous studies showing that  $\text{H}_2\text{SO}_4$  was a key species of atmospheric nucleation (Kuang et al., 2008). The newly formed particle grew to around 100 nm in diameter within 24 h. On average, the particle growth rates were respectively 2.8, 3.6, and 4.4  $\text{nm h}^{-1}$  for NPF events on 5, 6, and 7 June 2008. One  
15 can note that the new particles continued growing during nighttime when sulfuric acid concentration was close to zero. This indicated that other species, most likely, organic compounds contributed to the particle growth during this time period.

### 4.2 Hygroscopicity of newly formed particles

The size-resolved particle HGFs at  $\text{RH} = 90\%$  were measured during the NPF events. As shown in the Fig. 1c, during the particle formation period, the particle hygroscopicity increased significantly, independent on particle sizes. This was because that  $\text{H}_2\text{SO}_4$   
20 was produced by photochemistry reactions and condensed onto the pre-existing particles. The  $\text{H}_2\text{SO}_4$  and its salts (such as  $(\text{NH}_4)_2\text{SO}_4$ ) are highly water soluble compared to the secondary organic aerosols. At the same time, the increasing ambient

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temperature could drive the semi-volatile organic species in particle phase to partition to gas phase. Both processes resulted in a decreasing of organic fraction in particle phase, as shown in Fig. 1d, thereby enhancement in particle hygroscopicity. The significant decline in particle hygroscopicity took place after 15:30 LT when sulfuric acid concentration decreased significantly. During this time period, sulfuric acid condensation played a minor role in particle growth. Simultaneously, ambient temperature decreased to 10 °C. Lower temperature facilitates the condensation of semi-volatile organic vapors onto the particles. This was confirmed by AMS measurements showing that organic fraction increased extremely during nighttime. As a result, the organic mass fraction increased significantly, leading to an evident decline in particle hygroscopicity.

Table 1 summarizes the water soluble fraction of newly formed particles when these particles grew to 35, 50, and 75 nm, respectively. On 7 June, the water soluble fraction of 35 nm newly formed particles was 34 %. It decreased to 23 % when particle grew to 50 nm, further, reduced to 17 % when particles reach to 75 nm. On 5 and 6 June, the hygroscopicity of newly formed particles decreased with increasing particle size, as well. It implies that a large fraction of species contributing to particle growth was organics, which are typically less water soluble. This can indirectly be confirmed by AMS measurements showing that organic fraction in particles increased at the later stage of NPF events (see Fig. 1d) considering that the chemical species contributing to particle growth was similar to those condensing onto the larger pre-existing particles which can be detected by AMS. The contribution of H<sub>2</sub>SO<sub>4</sub> condensation to particle growth was estimated using the method introduced in Sect. 3.3 for different particle sizes listed in Table 1. The ratios of H<sub>2</sub>SO<sub>4</sub> condensational growth to the observed particle growth ( $F_{GRH_2SO_4} = GR_{H_2SO_4} / GR_{obs}$ ) are given in the Table 1. For example, on 7 June,  $F_{GRH_2SO_4}$  was 30 % for 35 nm particles, meaning that H<sub>2</sub>SO<sub>4</sub> condensation only contributes 30 % of the observed particle growth. With increasing particle size, the contribution of H<sub>2</sub>SO<sub>4</sub> condensation decreased, as shown in Table 1. This was consistent with the changes in the water soluble fraction of newly formed particles. Both

particle hygroscopicity measurements and numerical analysis showed that organics were major potential contributors to the particle growth.

### 4.3 Enhancement in CCN concentration during the NPF events

The critical diameters and CCN number concentrations at different supersaturation during NPF events are displayed in the Fig. 2. The critical diameters at different supersaturation decreased during the first several hours of the NPF events and enhanced at the latter stage of the events. This was consistent with the variations in particle hygroscopic growth at RH = 90 % above-mentioned. As shown in the Fig. 2b, the CCN number concentration clearly increased significantly during the NPF events. The minimum in CCN number concentration was observed during the period of particle formation and the maximum appeared at the end of the NPF events.

During three consecutive NPF days, the CCN number concentration step-wisely enhanced due to the accumulation processing. The NPF events occurred on 5, 6, and 7 June were typical regional cases. The enhancement in CCN number concentration caused by atmospheric nucleation was evaluated by comparing the average CCN number concentrations over two hours prior to the beginning of the event (the period t1 marked in Fig. 2) with the same time period before the end of the events (the period t2 marked in Fig. 2). The ratios of average CCN number concentration over t2 to t1 were respectively 1.9, 2.0, and 1.5 for 0.1, 0.4, and 0.6 % SS. On average, the enhancement ratios in CCN number concentration associated with individual NPF events were 63, 66, 69 for 0.1, 0.4, and 0.6 % SS, respectively. The absolute increases in CCN number concentrations associated with each event were 162, 931, and 756 # cm<sup>-3</sup>, on average.

Several previous studies reported that the enhancement in CCN number concentration associated with atmospheric nucleation varied significantly in different environments. At the Finnish sub-Arctic Pallas station, a 210 ± 110 % increase in the number concentration of 80 nm particles was observed from the beginning of a nucleation event to the end of the event (Asmi et al., 2011). At a forested site (SMEAR II station in Hyytiälä) in Southern Finland, nucleation enhanced CCN number concentration by 70

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to 110 %, varying with the supersaturation level (Sihto et al., 2011). In a polluted urban area, Beijing, China, the average CCN enhancement factors were between about 1.5 and 2.5 (Yue et al., 2011; Wiedensohler et al., 2009). In Boulder, CO, Atlanta, GA, and Tecamac, Mexico, the pre-existing CCN number concentration increased on average by a factor of 3.8 as a result of new particle formation (Kuang et al., 2009). Overall, the enhancement in CCN number concentration associated with atmospheric nucleation varied significantly in different environments. Please note that the methods for defining the enhancement factors used in the existing literature were very different. Therefore, a general conclusion on how significant NPF and growth process contributes to CCN budget cannot be drawn, currently.

## 5 Discussion

The above field observations clearly showed that newly formed particles had the ability to grow into CCN sizes within several hours at Melpitz. The particle hygroscopicity measurements strongly suggested that organic compounds were the major contributors driving particle growth into CCN sizes. The previous studies performed in clean atmosphere also showed that the newly formed particles mainly consist of organics. For examples, sulfuric acid is able to account for roughly 30 % of the growth rate of newly formed particles in the rural atmosphere of Hohenpeissenberg, Southern Germany (Birmili et al., 2003), and only around 10 % in the boreal forest area of Finland (Boy et al., 2005). However, In the polluted atmosphere of Atlanta, USA, the available amount of sulfuric acid was sufficient to explain all of the observed particle growth (Stolzenburg et al., 2005). At Melpitz, biological activities produced a lot of biogenic volatile organic compounds (BVOCs) and lead to an organic-rich environment during summertime. The oxidation products of BVOCs may be responsible for the new particle growth.

We note that the condensation of organics lead to a rapid particle growth when sulfuric acid concentration was close to zero during nighttime, as shown in Fig. 1. The

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organic condensing materials with low hygroscopicity reduced CCN efficiency of the new particles, as indicated by critical diameters given in Fig. 2. Such phenomenon was also reported by Dusek et al. (2010). They showed that enhanced organic mass fraction caused a reduction in CCN efficiency of small particles during the new particle formation. These results implied that the CCN production associated with atmospheric nucleation may be overestimated if assuming that new particles can serve as CCN in case they grow to a fixed particle size (such as Asmi et al., 2011), especially for organic-rich environments.

## 6 Conclusions

In this study, the particle number size distribution, particle hygroscopicity, and particle chemical composition during three regional NPF events were measured to investigate the new particle growth process and its effects on CCN activity. The particle formation rates ( $J_{3-25\text{nm}}$ ) were 13.5, 6.1, 9.3  $\text{cm}^{-3}\text{s}^{-1}$ , and the particle growth rates were 2.8, 3.6, and 4.4  $\text{nm h}^{-1}$  for NPF events on 5, 6 and 7 June 2008, respectively. The observation showed that sulfuric acid is a key species of atmospheric nucleation.

The newly formed particles grew to CC sizes within one day. The particle hygroscopicity and chemical composition measurements and numerical calculation confirmed that organic compounds are major contributors driving particles growth to CCN sizes. The  $(\text{NH}_4)_2\text{SO}_4$ -equivalent water-soluble fraction accounted for 20 and 16 % of 50 and 75 nm newly formed particles, respectively. The step-wised increase in CCN number concentration during three consecutive NPF events was observed. On average, the enhancement ratios in CCN number concentration associated with individual NPF events are 63, 66, 69 for 0.1, 0.4, and 0.6 % SS, respectively.

We found that the new particles hygroscopicity decreased significantly with condensational growth of organic compounds, which are generally less water soluble. Correspondingly, the critical diameters at a certain supersaturation decline, indicating that enhanced organic mass fraction caused a reduction in CCN efficiency of newly formed



particles during the new particle formation. Our results implied that the CCN production associated with atmospheric nucleation may be overestimated if assuming that new particles can serve as CCN in case they grow to a fixed particle size, which was used in some previous studies, especially for organic-rich environments.

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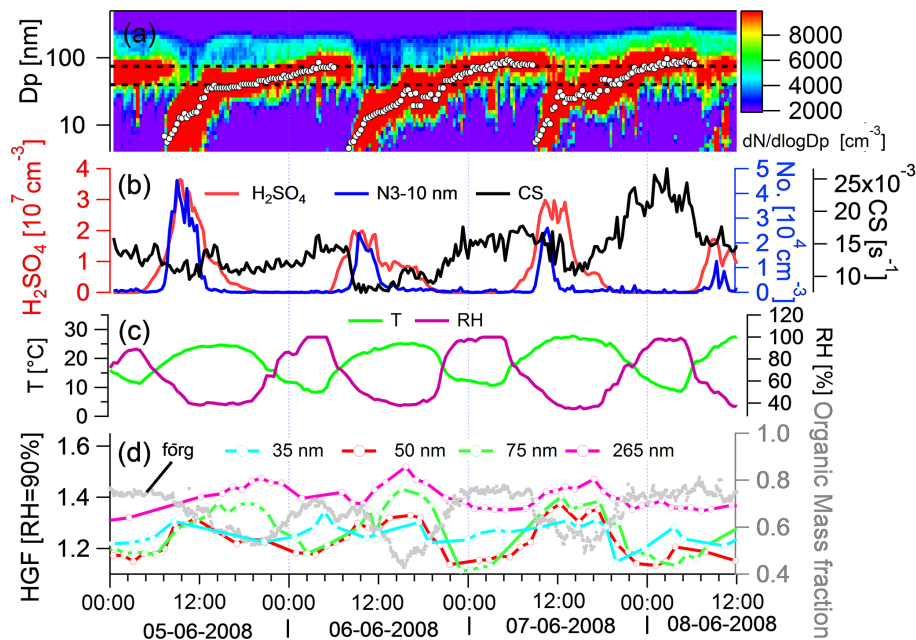
**Table 1.** The water soluble fraction of newly formed particles and the ratios of  $\text{H}_2\text{SO}_4$  condensational growth to the observed particle growth.

$D_p$ Date	35 nm		50 nm		75 nm	
	$\varepsilon$	$F_{\text{GRH}_2\text{SO}_4}^*$	$\varepsilon$	$F_{\text{GRH}_2\text{SO}_4}$	$\varepsilon$	$F_{\text{GRH}_2\text{SO}_4}$
5 Jun 2008	–		24 %	23 %	20 %	15 %
6 Jun 2008	25 %	23 %	14 %	17 %	10 %	11 %
7 Jun 2008	34 %	30 %	23 %	20 %	17 %	13 %

\*  $F_{\text{GRH}_2\text{SO}_4} = \text{GR}_{\text{H}_2\text{SO}_4} / \text{GR}_{\text{obs}}$ : the ratio of  $\text{H}_2\text{SO}_4$  condensational growth to the observed particle growth.

## The condensing vapors driving new particle growth to CCN sizes

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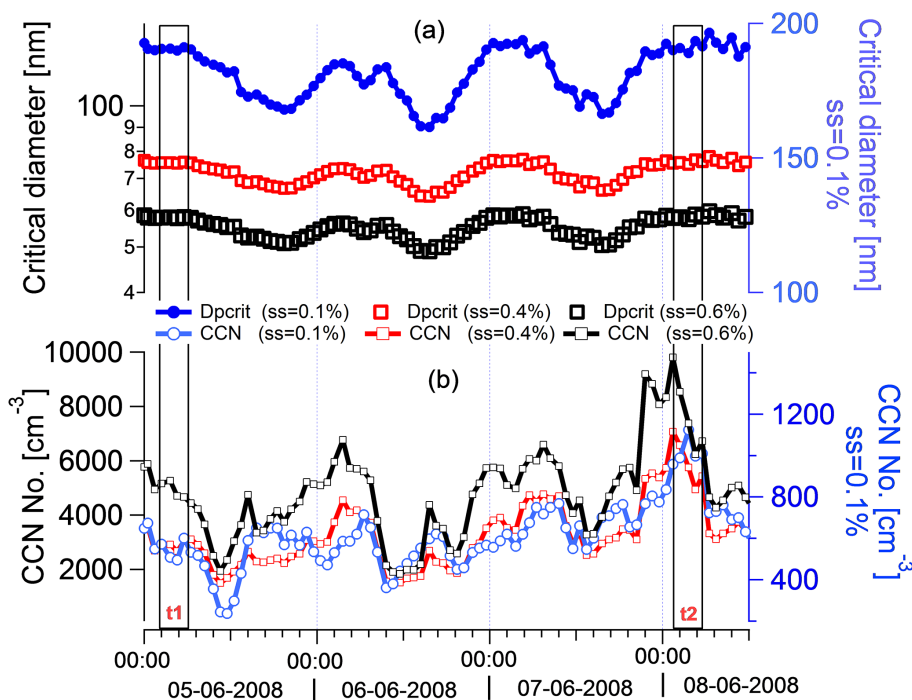
**Figure 1.** Particle number size distribution, 3–10 nm particle number concentration, H<sub>2</sub>SO<sub>4</sub> concentration, condensation sink (CS), temperature (*T*) and relative humidity (RH), size-dependent particle hygroscopic growth factors at RH = 90%, and mass fraction of organic compounds (*f*<sub>org</sub>) during the new particle formation events.

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**Figure 2.** Critical diameter and CCN number concentration during NPF events.

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