The paper presents a case study of three consecutive new particle formation events in a regional background station in Europe. Hygroscopicity measurements and AMS data are used to draw conclusions about the contributions of sulfuric acid and organic vapors to the growth of the newly formed particles to CCN sizes. CCN concentrations are calculated from size distributions and chemical composition data. The paper combines very useful measurements and calculations, but could make better use of its data and should be more rigorous in the presentation of the analyses. The following issues should be addressed before publication:

****

Thank you very much for spending a lot of time to give many good suggestions and comments. We will answer the following questions point by point.

****

Major comments:

1) No particle sources other than the NPF events seem to have been taken into consideration in the analysis. Melpitz is not a pristine environment, and even if the possible contribution of local sources can be excluded during the three-day period of observation (the paper does not state how or if this was done), wind direction can still change aerosol characteristics substantially within the time it takes for newly formed particles to grow to CCN sizes. How do the authors know that the results of their calculations pertain to aerosol particles specifically originating in the nucleation events when many hours have passed, as is the case for the calculated increases in CCN concentrations in section 4.3? How do the authors account for meteorological conditions, such as concentration/dilution due to boundary layer dynamics, when observing concentration changes (such as CCN concentrations)?

****

Response:

There are no local emission sources in the surrounding areas of the Melpitz research station. The possible primary emissions contributing to the atmospheric particles in Melpitz could be from the cities away tens of kilometers from the station via transportation. Typically, the primary particles are accompanied by trace gases, such as NO and SO_2 spikes. However, such phenomena were not observed in our measurements in Melpitz. As shown in Fig.1, in the early morning on 6 and 7 June, the slight enhancement of NO (a tracer for traffic related ultrafine particles (Janhäll et al., 2004)) concentration may be caused by the outflow of cities nearby Melpitz. We can note that the particle number concentration did not increased simultaneously. The small particles exhausted from car tailpipes in the cities may grow by condensation and coagulation and shift towards larger diameters and diluted by fresh air significantly with increasing distance from the roads (Zhu et al., 2002). As a result, the enhancement in particle number concentration was not observed in the rural site of Melpitz. Therefore, the instant impacts of primary emissions on atmospheric particles in Melpitz are not observed during the time period focused in our study. SO_2 from primary emissions contributes to the atmospheric nucleation after being oxidized to sulfuric acid by radicals. The new particle formation associating with enhanced SO_2 concentration was observed by many previous studies (e.g. Birmili and Wiedensohler, 2000).

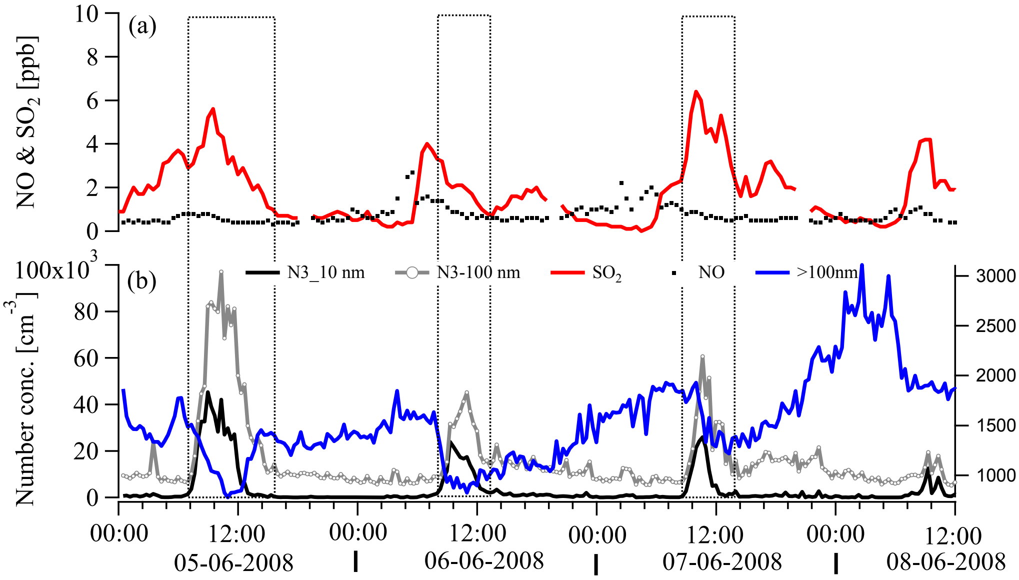

Fig. 1: The time series of SO_2 and NO concentrations (a) and number concentrations for particles in diameters of 3-10 nm, 3-100 nm, and larger than 100 nm (b).

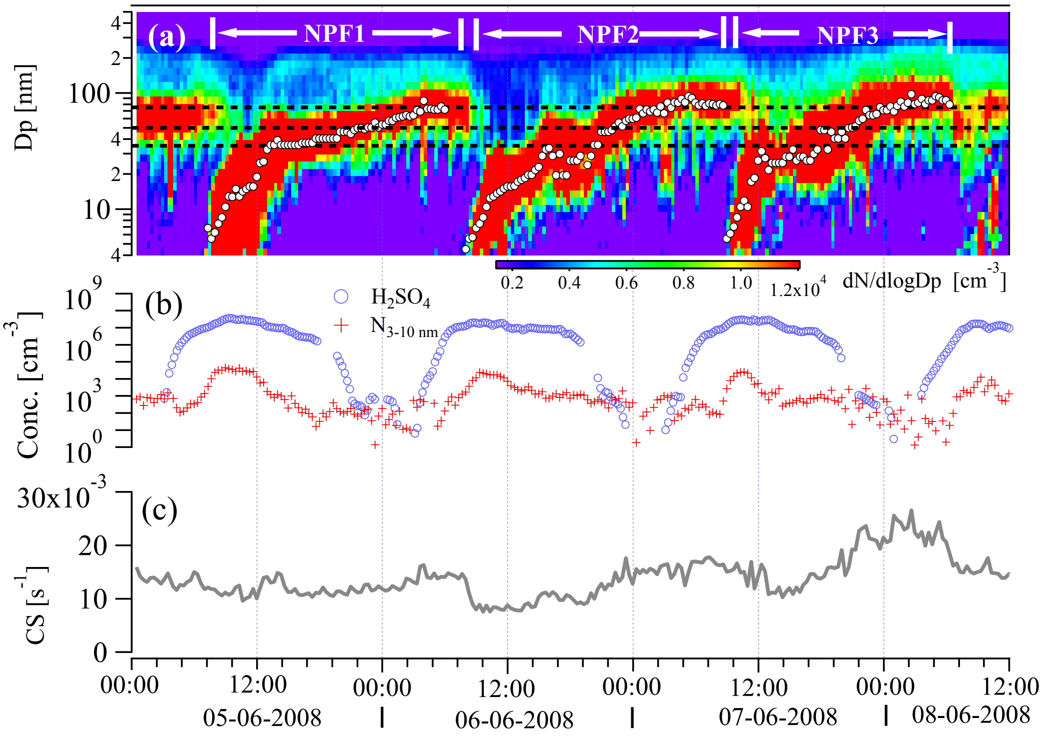

Fig. 2: Particle number size distribution, 3-10 nm particle number concentration, H_2SO_4 concentration, condensation sink (CS) during the new particle formation events.

From the regional scale point of view, the primary particles, such as soot particles are diluted after emission and gradually change into uninform distribution in the air with increasing distance from the emission sources. The lifetime of fine particles is around 1 week. The time period is 3 days. The primary particles could accumulate in the air and increase the CCN number. However, compared to the particle concentration contributed by nucleation and growth, the contribution of primary particles to CCN number can be ignored.

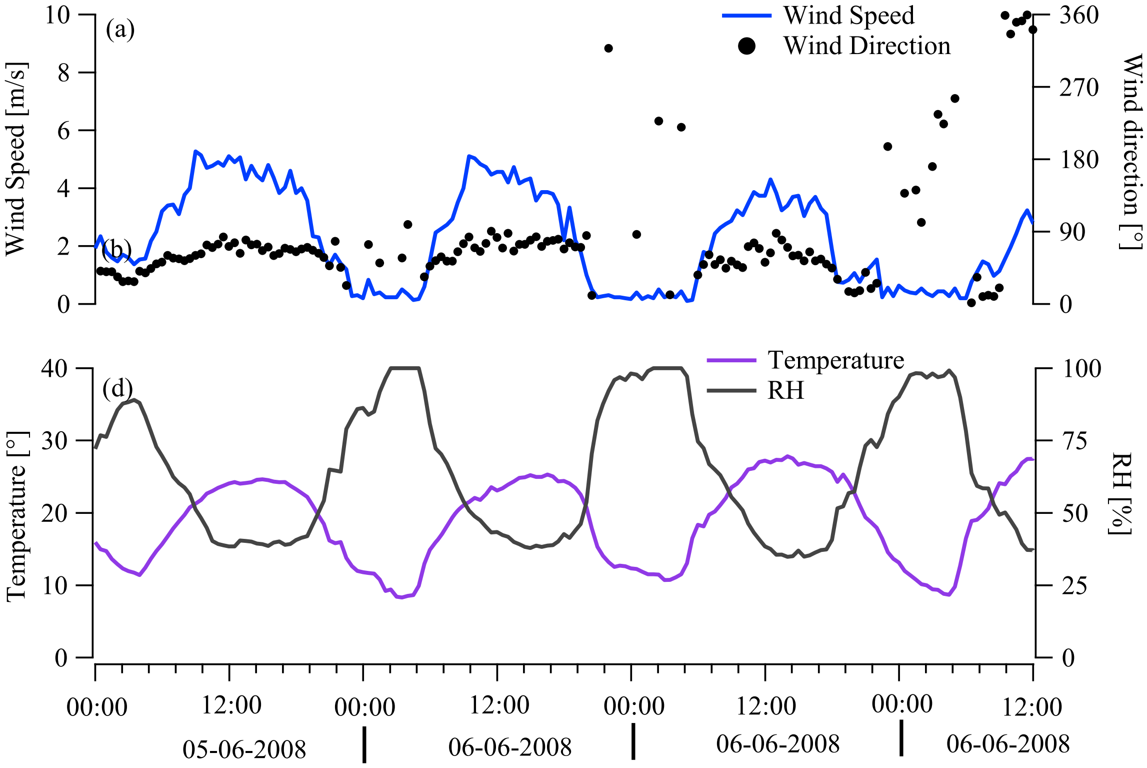

Fig. 3: The time series of wind speed, wind direction, ambient temperature, and RH during the NPF events.

As displayed in Fig.2, the particle number size distribution shows the new particle formed around 10 am and then grew versus time for more than 20 h. This shows that the NPF is a regional event (refere to Hussein et al., 2000) and could take place over a distance of a hundred kilometers. The Fig.3 shows the wind speed and wind direction during the NPF events. The wind showed a typical diurnal cycle. The wind speed is 4-5 m/s and kept a constant direction (south) during daytime. It is static wind during nighttime. No sharp change in wind direction and wind speed were observed.

Atmospheric boundary layer development and turbulent mixing will impact on NPF (Boy et al., 2006;Boy et al., 2003;Altstädter et al., 2015), and consequently on its CCN products. It is hard task to quantify the changes in CCN number due to boundary layer dynamics. In this study, 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. 4) with the same time period before the end of the events (the period t2 marked in Fig. 4). The weather conditions (see RH and T in Fig. 3) are quite similar during the three NPF days. Here we assume that the weather condition and boundary layer height are similar during two time periods. In addition, the enhancement factor of CCN number concentration is a ratio of CCN concentration during two periods and not an absolute value. To some extent, the ratio can weaken the influence of boundary layer dynamics. In addition, the ratio can exclude the effect of primary emission on CCN number. This is because that the contribution of primary particles to CCN number should be similar at the same time period on different days.

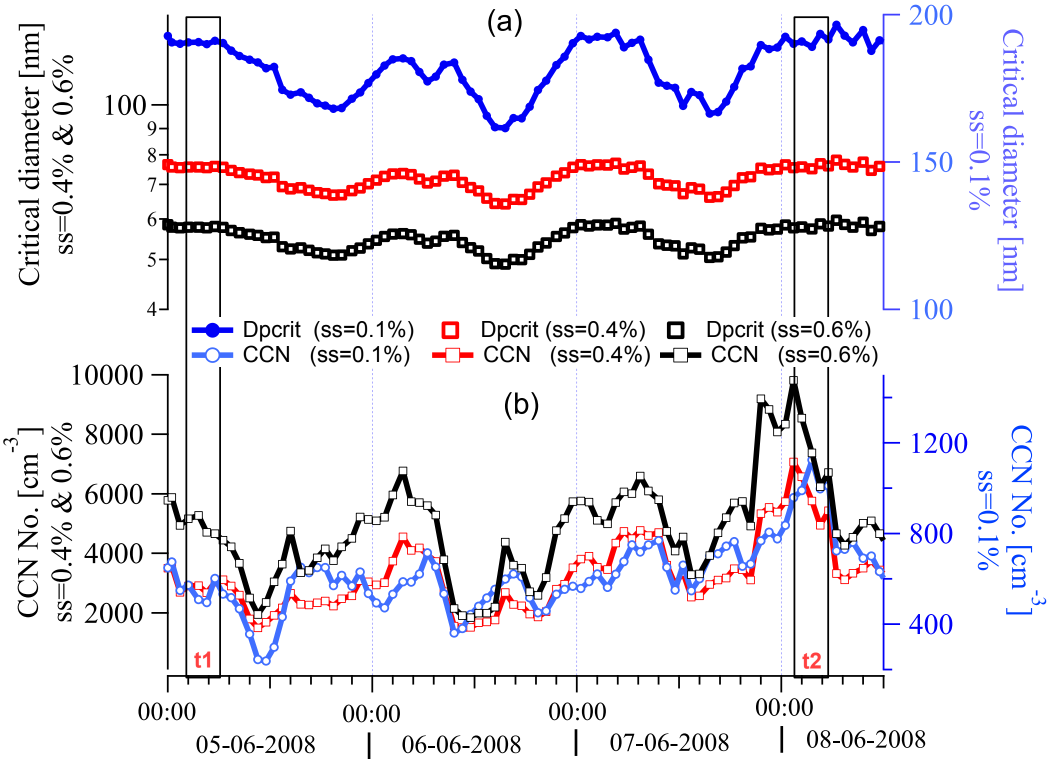

Fig.4: Critical diameter and CCN number concentration during NPF events.

According to the above-analysis, we can see that the new particle formation and growth is a major particle source at Melpitz. The NPF at a regional scale makes more meaningful for contributing the CCN number. The increased nucleation mode particle number concentration was followed with sulfuric acid concentration. This confirms that the particles mainly originated from atmospheric nucleation and subsequent growth, not from the primary emissions.

Modification in the MS

Modification in section “4.1 Particle formation and growth”

“The previous study on the basis of long-term observations showed that the NPF events take place frequently at Melpitz, especially on April, May, and June (Hamed et al., 2010). In present study, three NPF events, which consecutively took place from June 5 to June 7, 2008, as displayed in Fig. 1 (a), are selected for further analysis. These events are the best cases which showed clear particle bursts and subsequent growth process during the entire field campaign (from May 23rd to June 8th in 2008). The starting and ending time for each event were marked in the Fig.1 (a) as NPF1, NPF2, and NPF3. The bursts in number concentration of 3-10 nm particles were observed associated with increasing ambient temperature, decreasing relative humidity (shown in Fig. 2 (b)), and increasing in estimated H_2SO_4 concentration (shown in Fig. 1(b)). The condensation sink (CS) is between 0.01 and 0.02 s^{-1} during the NPF events. As marked in Fig.1 (a), the particle number size distribution shows the new particle formed around 10 am and then grew versus time for more than 20 h. This means that the NPF is a regional event (refere to Hussein et al., 2000) and could take place over a distance of a hundred kilometers. The Fig.2 (a) displays the wind speed and wind direction during the NPF events. The wind showed a typical diurnal cycle. The wind speed is 4-5 m/s and kept a constant direction (south) during daytime. It is static wind during nighttime. The particle formation rates ($J_{3\text{-}25nm}$) were 13.5, 6.1, 9.3 $cm^{-3}s^{-1}$ on June 5, 6, and 7, respectively. The highest formation rate was observed on June 5 corresponding to the highest H_2SO_4 concentration.

As indicated by the white circle in the Fig.1 (a), the mean geometric diameter (Dm) of log-normal ultrafine particle mode increased to around 100 nm within 24 hours. Over the time period from the beginning to the end of the NPF events as marked in Fig.1 (a), the average GR_{obs}s were respectively 2.8, 3.6, and 4.4 nm h^{-1} for NPF events on June 5th, 6th, and 7th, 2008. One 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.

There are no local emission sources in the surrounding areas of the Melpitz research station. The possible primary emissions contributing to the atmospheric particles in Melpitz could be from the cities away tens of kilometers from the station via transportation. Typically, the primary particles are accompanied by trace gases, such as NO and SO_2 spikes. However, such phenomena were not observed in our measurements at Melpitz. As shown in Fig.2 (c), in the early morning on 6 and 7 June, the slight enhancement of NO (a tracer for traffic related ultrafine particles (Janhäll et al., 2004)) concentration may be caused by the outflow of cities nearby Melpitz. The particle number concentration did not increased simultaneously. The ultrafine particles exhausted from car tailpipes in the cities may grow by condensation and coagulation and shift towards larger diameters and diluted by fresh air significantly with increasing distance from the roads (Zhu et al., 2002). As a result, the enhancement in ultrafine particle number concentration was not observed at the rural site of Melpitz. Therefore, the instant impacts of primary emissions on atmospheric particles are not observed during the time period focused in this study. SO_2 from primary emissions could contribute to the atmospheric nucleation after being oxidized to sulfuric acid by radicals. The new particle formation associating with enhanced SO_2 concentration was observed by many previous studies (e.g. Birmili and

Wiedensohler, 2000). Overall, the new particle formation and subsequent growth is the major source of particles, and thereby, CCN at Melpitz station. ”

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2) It seems to me that the available data should be exploited better. For example, size-resolved AMS data are available, but, as far as I can tell, are only made use of in the calculation of D(crit). In describing changes in the organic fraction during the NPF events, instead of using PM1, the size-resolved AMS data could be used to more accurately represent chemical composition of the smaller particle size range, thereby reducing the potential dominance of large particles (possibly from sources other than NPF) in the mass concentrations. Also, with those size-resolved AMS data available, why is all hygroscopicity and all condensational growth reduced to sulfate and organics? With HTDMA data available, it may be possible to derive a kappa (HTDMA), and use that for the calculation of the expected CCN concentrations – was there a specific reason the authors decided to use only the AMS chemistry + size distribution data?

****

Response:

Considering the accuracy of size-resolved particle mass concentration detected by AMS and the transmission efficiency of aerodynamic lenses (Canagaratna et al., 2007), the chemical composition of 30-100 nm particles is chosen to perform the analysis. Some discussions were added into the manuscript.

The HTDMA-derived kappa was not used in calculating the critical diameter. This reason is given as follow: The inconsistencies between HTDMA-derived kappa and CCNc-derived kapp have been reported in several previous studies (Good et al., 2010;Cerully et al., 2011;Irwin et al., 2010;Petters et al., 2009;Wex et al., 2009). Possible explanations are non-ideality effects in the solution droplet, surface tension reduction due to surface active substances, and the presence of slightly soluble substances which dissolve at RHs larger than the one considered in the H-TDMA (Wex et al., 2009). Due to these effects, is not necessarily constant and may vary with humidity. Extrapolating from HTDMA data to properties at the point of activation should be done with great care. In addition, the previous studies showed that critical diameters at different supersaturations can be well-predicted using AMS data and ZSR method. Therefore, the AMS data was decided to use to estimate the critical diameters instead of HTDMA-derived kappa.

Modification in the MS

As displayed in Fig.3 (c), the sulfate and ammonium were dominated in the inorganic mass fraction in 30-100 nm particles and obviously increased during the particle formation period (indicated by grey dashed line in Fig.1). While, the nitrate accounted for a minor fraction, which also observed by Zhang et al. in Pittsburgh (Zhang et al., 2004). They found nitrate contributed the least to the new particle growth. After 3:00pm on each day, the organic mass fraction increased and reached its maximum at midnight, indicating that organics played a key role in the particle growth at a relatively later time of the NPF event. The mass fraction of ion fragments m/z 44 and 57 in 30-100 nm particles are shown in the Fig. 3(b). The m/z 44 (CO_2^+ ion fragment) is a tracer for secondary organic aerosol，while m/z 57 ($C_4H_9^+$) is generally associated with primary organics from combustion sources (Zhang et al., 2004). The m/z 57 mass concentration is close to zero during the events. Compared m/z 57, the m/z 44 mass concentration are considerable, indicating that the organics contributing to particle growth was mainly secondary organic species.

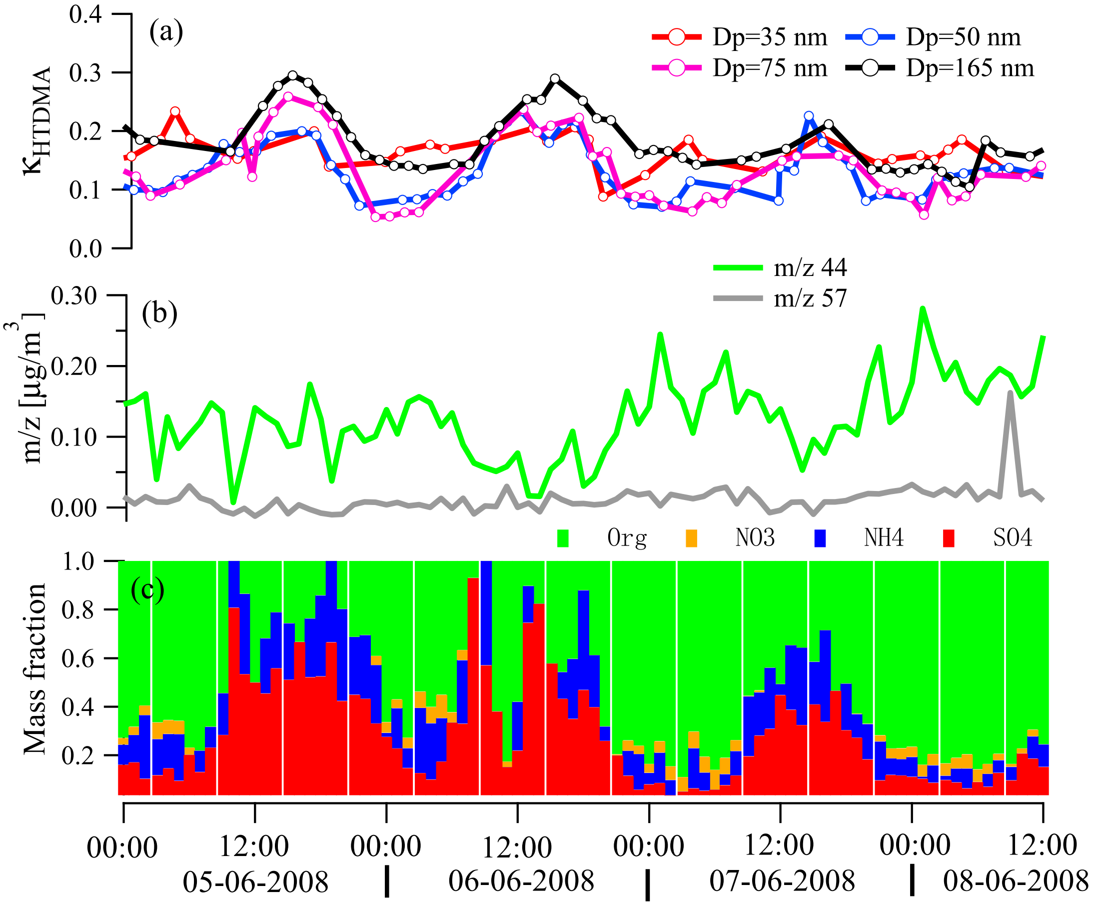

Fig.3: Size-resolved particle hygroscopicity and mass fraction of particle below 150 nm in mobility diameter.

Some texts were added into the section 3.2

“The HTDMA-derived κ was not used in calculating the critical diameter. This reason is given as follow: The inconsistencies between HTDMA-derived kappa and CCNc-derived κ have been reported in several previous studies (Good et al., 2010;Cerully et al., 2011;Irwin et al., 2010;Petters et al., 2009;Wex et al., 2009). Possible explanations are non-ideality effects in the solution droplet, surface tension reduction due to surface active substances, and the presence of slightly soluble substances which dissolve at RHs larger than the one considered in the H-TDMA (Wex et al., 2009). Due to these effects, κ is not necessarily constant and may vary with humidity. Extrapolating from HTDMA data to properties at the point of activation should be done with great care (Wu et al., 2013). In addition, the our previous study (Wu et al., 2013) showed that critical diameters at different supersaturations can be well-predicted using AMS data and ZSR method. Therefore, the AMS data was decided to use to estimate the critical diameters instead of HTDMA-derived κ. "

****

3) There is important information missing, such as: What were the kappa(i) values used that entered the kappa(chem) calculation? How and over what time period was GR(obs) calculated? How were the hygroscopic growth factors derived from the HTDMA data? The description of the results is also missing information: Increases/decreases are described without giving the time period over which increases/decreases were observed, or how large the increases/decreases were. How are the beginning, end, and duration of an NPF event defined? When stating that something was observed "during the events", what exact time frame does that refer to? Also, for which exact points during the particle growth were the soluble fractions calculated?

****

Response:

The information on the input parameters in the calculation will be added in the manuscript. The starting and ending time of PNF events were marked in the Fig. 1 (a).

Modification in the MS

In section 3.2:

"We use a simplified ion pairing scheme as presented in Gysel et al. (2007) to convert the ion mass concentrations to the mass concentrations of their corresponding inorganic salts as listed in Table 2."

Table 2: Gravimetric densities and hygroscopicity parameters κ used in this study.

Species	**NH_4NO_3**	**H_2SO_4**	**NH_4HSO_4**	**$(NH_4)_2SO_4$**	**Organic matter**
[kg/m^3]	1720	1830	1780	1769	1400
	0.67	0.92	0.61	0.61	0.1

In section 4.1:

"The previous study on the basis of long-term observations showed that the NPF events take place frequently at Melpitz, especially on April, May, and June (Hamed et al., 2010). In present study, three NPF events, which consecutively took place from June 5 to June 7, 2008, as displayed in Fig. 1 (a), are selected for further analysis. These events are the best cases which showed clear particle bursts and subsequent growth process during the entire field campaign. The starting and ending time for each event were marked in the Fig.1 (a) as NPF1, NPF2, and NPF3.”

“Over the time period from the beginning to the end of the NPF events as marked in Fig.1 (a), the average GR_{obs}s were respectively 2.8, 3.6, and 4.4 nm h^{-1} for NPF events on June 5th, 6th, and 7th, 2008.”

****

Comments on the writing:

1) There are multiple language errors, which should be corrected (singular/plural errors, missing/superfluous articles, wrong punctuations), as they can hamper the understanding of the material, e.g.: p. 8405, line 26 “sulfuric acid and organics, which fraction is the leading component in particles: : :”, p.8406, line “measurements during a NPF events”, p. 8406: “in the polluted areas, Atlanta, USA”, p. 8413 “took place from, 5 to 7 June 2008, “.

Response:

These errors were corrected in the MS.

2) There are many references in the text that are missing in the list of references, e.g. p. 8408, lines 21-23.

Response:

The MS was checked very carefully. The missing references were added in the MS.

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Specific comments by section:

Introduction:

p. 8405, line 8: “the single largest uncertainty” of what? In the statement by Kerminen et al. that is most likely cited here, nucleation is not stated to be “the single largest uncertainty” in the CCN budget.

Response:

“the single largest uncertainty” was modified to “The contribution of atmospheric nucleation to the global CCN budget spans a relatively large uncertainty range”.

p. 8405, line 9: “radioactive” should be "radiative"

Response:

It was corrected in the MS.

p. 8406, line 6: what are “potential chemical species”?

Response:

“Potential” is removed from the texts.

****

Experiments:

Perhaps the title “Measurements”, or “Experimental” would be better, as “Experiments” could raise the expectation of a controlled laboratory setup.

Response:

We agree. The “Measurements” was used instead of “Experiments”

p. 8406, line 15: Could the authors provide a little more information on the full dataset that this case study is a part of, and specifically, state why were those exact three days chosen? How many events were observed in the full dataset? Please also describe the origin of SO2 and Rad, used later in this study.

Response:

The particle number size distribution, gaseous pollutants, and meteorological parameters are routinely measured at Melpitz research station. The HR-Tof-AMS was operated only during the intensive field campaign from May 23rd to June 8th, 2008. The following table summarizes all instruments and measured parameters used in this study.

The previous study on the basis of long-term observations showed that the NPF events take place frequently at Melpitz, especially on April, May, and June (Hamed et al., 2010). In present study, three NPF events selected are the best cases which showed clear particle bursts and subsequent growth process during the time period with available AMS and HTDMA data. These NPF events are typical regional event, which spanned a larger spatial scale and therefore more significant impacts on CCN number concentration at a regional scale.

Modification in the MS

One table was added into the “Measurements” section and summarizes the instruments and parameters used in this manuscript.

Table: The summary of instrument and parameters used in this study

Instrument	Parameter
SMPS	Particle number size distribution
HTDMA	Particle hygroscopicity
HR-ToF-AMS	Size-resolved chemical composition
Monitor – APSA 360 Horiba Europe	SO_2 concentration
Kipp & Zonen CM6 Pyranometer	Global solar irradiance

An explanation was added into the manuscript to explain why only three events were selected in this study.

“The previous study on the basis of long-term observations showed that the NPF events take place frequently at Melpitz, especially on April, May, and June (Hamed et al., 2010). In present study, three NPF events, which consecutively took place from

June 5 to June 7, 2008, as displayed in Fig. 1 (a), are selected for further analysis. These cases are the typical regional events which showed clear particle bursts and subsequent growth process during the time period with available AMS and HTDMA data."

****

Section 2.1:

The HTDMA measurements should be described better: which dry sizes and RH's were chosen? What was the sampling schedule for the chosen parameters? How were the growth factors determined from the size distribution of DMA2?

Response:

The particles with dry sizes of 35, 50, 75, 110, 165, and 265 nm were measured by HTDMA at RH=90% with time resolution of 1 h. In this study, the HGFs of 35, 50, and 75 nm particles are used only. The HTDMA raw data were inverted using the TDMAinv method developed by Gysel et al. (2009). In Gysel's article, the TDMAinv method is introduced very detailed. Here, we will not give unnecessary details.

Modification in the MS

One sentence was added into the MS.

"In this study, the particles with dry sizes of 35, 50, 75, 110, 165, and 265 nm were measured by HTDMA at RH=90% with the time resolution of 1h. The HGFs of 35, 50, and 75 nm particles will be taken for further analysis. "

p. 8407, line 2: "described" is better than "illustrated"

Response:

It was changed in the texts.

****

Section 2.2:

Please provide a lower particle size detection limit of the AMS.

Response:

The aerodynamic lenses have 100% transmission efficiency down to 70 nm in a vacuum aerodynamic diameter (Canagaratna et al., 2007).

Modification in the MS:

One sentence was added into the MS: "The aerodynamic lenses have 100% transmission efficiency down to 70 nm in a vacuum aerodynamic diameter (Canagaratna et al., 2007)."

p. 8407, line 24: "typically": Since the investigated dataset is only three days, could the authors give more detail here?

Response:

The "typically" was removed from the texts.

p. 8408, line 8: How was the density value chosen?

Response:

We estimated the gravimetric particle density on the basis of measured chemical composition using the following equation:

$$\text{density} = \frac{[\text{Total}_{\text{AMS}} + \text{BC}]}{\frac{[\text{NO}_3^-]+\left[\text{SO}_4^{2-}\right]+[\text{NH}_4^-]}{1.75} + \frac{[\text{Cl}-]}{1.52} + \frac{[\text{Org}]}{1.2} + \frac{[\text{BC}]}{1.77}}$$

The mean particle density is 1.4 g cm^{-3} for May–June 2008. The detail description was given in (Poulain et al., 2014)

Modification in the MS:

"The particle density was calculated on the basis of measured chemical composition. The detail description about the calculation was given in Poulain et al. (2014)."

p. 8409, line 3: "two groups including soluble and insoluble fractions" – this is confusing. How many groups or fractions were there?

Response:

This sentence was rewritten in the texts.

"Here, the chemical compounds contributing to the particle growth are grouped into two fractions, e.g., soluble and insoluble fractions."

p. 8409, line 13 and following: Should the assumption of an insoluble organic fraction not underestimate the actual soluble fraction of the particle?

Response:

Yes, we agree. The assumption of an insoluble organic fraction may lead to overestimate the soluble fraction.

p. 8409, line 17: It would be helpful if the term "equivalent soluble fraction" was used consistently from this point onward.

Response:

The term "equivalent soluble fraction" was used consistently in the texts.

****

Section 3.2:

p. 8410, line 9: The size ranges of the AMS measurements, the chemical species used in the calculation, as well as the values used for their respective kappa(i) should be stated here.

Response:

A detail description is given in the manuscript.

Modifications in the texts:

"The AMS provides the particle mass size distribution of sulfate (SO_4^{2-}), nitrate (NO_3), and ammonium (NH_4^+) ions as well that of organic compounds. We use a simplified ion pairing scheme as presented in Gysel et al. (2007) to convert the ion mass concentrations to the mass concentrations of their corresponding inorganic salts as listed in Table 2."

Table 2: Gravimetric densities ρ and hygroscopicity parameters κ used in this study.

Species	NH_4NO_3	H_2SO_4	NH_4HSO_4	$(NH_4)_2SO_4$	Organic matter
ρ [kg/m^3]	1720	1830	1780	1769	1400
	0.67	0.92	0.61	0.61	0.1

p. 8410, line 15: "are activated", not "were activated" p. 8410, line 16: Please state which critical supersaturations were chosen for the calculation.

Response:

It was corrected in the text:

"DPcrit is the critical diameter at which 50% of the particles were activated at the supersaturation, Sc (0.1%, 0.4%, and 0.6 are chosen)."

p. 8410, line 19: Integrating the size distribution from D(crit) upwards: The implicit assumption of an internal mixture of the determined (bulk) chemical composition should be stated explicitly here.

Response:

The sentence "The CCN number concentration is estimated by integrating the particle number size distribution from the critical diameter to the maximum diameter detected by TDMPS (800 nm, above which the particle number concentration is generally negligible), assuming particles are internal mixture." was added into the texts.

p. 8410, line 22 and following: This discussion is confusing in its current location in the section: "The critical diameters" (p. 8411, line 1) could be interpreted as the critical diameters in this study, calculated from eq. 5, rather than a general statement on the approximate range of critical diameters, which, I believe, is what the authors mean. This description of the chosen AMS size ranges should be moved up, as stated in the initial comment on this section.

Response:

The texts in line 22 and following of p.8410 were moved up to the initial part of this section.

Modification in the MS:

"The CCN number concentration can be estimated by integrating the particle number size distribution from the critical diameter to the maximum diameter detected by

TDMPS (800 nm, above which the particle number concentration is generally negligible), assuming particles are internal mixture. The critical diameter (D_{pcrit}) at which 50% of the particles are activated is calculated from κ:

$$\mathrm{D_{Pcrit}} = \left(\frac{4A^3}{27\,\kappa_{\mathrm{chem}}\ln^2 \mathrm{S_C}}\right)^{1/3} \qquad [6]$$

Here, κ_{chem} is calculated from size-resolved AMS data using the ZSR mixing rule. D_{Pcrit} is the critical diameter at which 50% of the particles were activated at the supersaturation, S_c (0.1%, 0.4%, and 0.6% are chosen in this study).

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

$$\kappa_{\mathrm{chem}} = \sum_i \varepsilon_i \kappa_i \qquad [7]$$

Here, κ_i and ε_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 ε_i from particle chemical composition measured by AMS. Here, the size-resolved particle chemical composition is used to estimate the κ_{chem}. The size-resolved chemical composition was used to calculate the κ values. The critical diameters, corresponding to supersaturation 0.2-0.7%, roughly span from 50 to 120 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 κ_{chem}. In the same way, the chemical composition of 150-200 nm particles is used to calculate κ_{chem} for the critical diameter of around 170 nm, which corresponds to a supersaturation of 0.1%. The AMS provides the particle mass size distribution of sulfate (SO_4^{2-}), nitrate (NO_3), and ammonium (NH_4^+) ions as well that of organic compounds. We use a simplified ion pairing scheme as presented in Gysel et al. (2007) to convert the ion mass concentrations to the mass concentrations of their corresponding inorganic salts as listed in Table 2."

****

Section 3.4:

What were the time frames (and particle diameter ranges) that were actually used for the determination of the growth rates in this study?

Response:

The growth rate is calculated as follow:

The observed particle growth rate (GR) can be expressed as:

$$\mathrm{GR} = \frac{\Delta \mathrm{D_m}}{\Delta \mathrm{t}}$$

where Dm is a mean geometric diameter of log-normal ultrafine particle mode, which has been fitted to the number size distribution (Heintzenberg, 1994). GR means evolution of the mean diameter within a time period Δt.

In the Fig.1 (a), the Dm of ultrafine particle mode is indicated as the white circles. The time frames during which GRs were calculated is from the starting and ending time marked in the Fig.1 (a).

Modifications in the texts:

More texts were added into the texts to make the calculation of growth rate clear.

“The starting and ending time was marked in the Fig.1 (a).”

“As indicated by the white circle in the Fig.1 (a), the mean geometric diameter (Dm) of log-normal ultrafine particle mode increased to around 100 nm within 24 hours. Over the time period from the beginning to the end of the NPF events as marked in Fig.1 (a), the average GR_{obs}s were respectively 2.8, 3.6, and 4.4 nm h^{-1} for NPF events on June 5th, 6th, and 7th, 2008.”

p. 8412, line 19: Weren’t the H2SO4 concentrations calculated, rather than measured?

Response:

The H_2SO_4 concentration is calculated. The error was corrected in the texts.

Results:

p. 8413, line 9: How was the particle formation rate calculated?

p. 8413, line 18: average over what? What was the time frame of the GR determination?

It would be helpful is “GR(obs)” were used consistently.

p. 8413, line 20: define “particle formation period”

Response:

These three questions relating to the calculation of particle formation and growth rate will be answered together. To clarify the calculation methods, one section has been added into the MS to introduce the methods for calculating the formation rate and growth rate. The starting and ending time of NPF events were marked in the Fig. 1. The growth rate is calculated within the time frame between the starting and ending points as marked in the Fig.1. The formation period is defined as the time period during which an obvious increase in the number concentration of 3-10 nm particles was observed. The formation period is marked in the Fig.1.

Modifications in the MS:

“3.4 Calculation of particle formation and growth rate

“Assuming a constant particle source during a time period of t, the particle formation rate (Jnuc) can be expressed as (Dal Maso et al., 2005):

$$J_{nuc} = \frac{dN_{nuc}}{dt} + F_{coag} + F_{growth} \qquad [10]$$

In this study, N_{nuc} is the number concentration of nucleation mode particles ranging from 3 nm to 25 nm. F_{growth} is the flux of particles out of the specified size range (3-25 nm). The newly formed particles rarely grew beyond 25 nm before formation ended, and F_{growth} can be neglected. F_{coag} represents a loss of formed particles due to coagulation to the preexisting particle population. It can be calculated from the following equation:

$$F_{coag} = CoagS_{nuc}N_{nuc} \qquad [11]$$

where $CoagS_{nuc}$ is the coagulation sink of particles in the nucleation mode. The detailed calculation of coagulation sink is given in Deal Maso et al. (2005).

The observed particle growth rate (GR_{obs}) can be expressed as:

$$GR_{obs} = \frac{\Delta D_m}{\Delta t} \qquad [12]$$

where Dm is a mean geometric diameter of log-normal ultrafine particle mode, which has been fitted to the number size distribution (Heintzenberg, 1994). GR_{obs} means evolution of the mean diameter within a time period Δt."

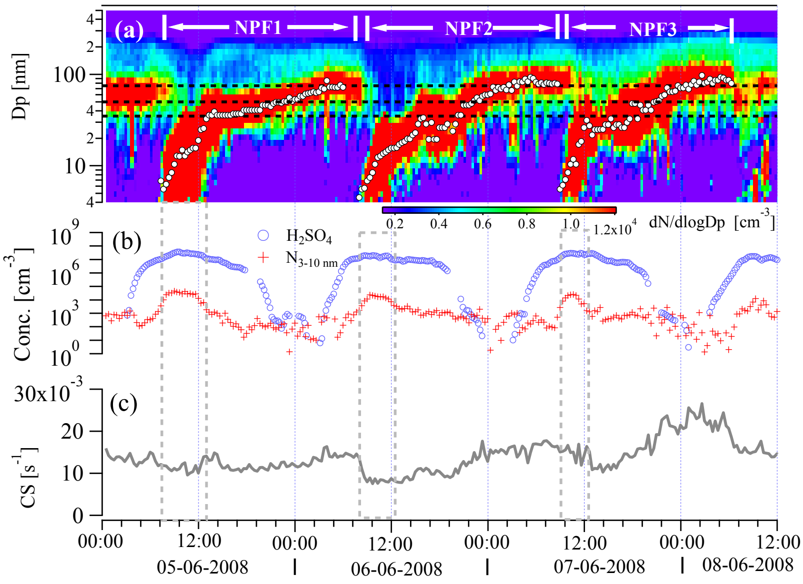

Fig. 1: Particle number size distribution, 3-10 nm particle number concentration, H_2SO_4 concentration, condensation sink (CS) during the NPF events. The starting and ending time of the events were marked in the upper place of panel (a) by NPF1, NPF2, and NPF3. The grey dashed lines indicated the time period of particle formation.

p. 8413, line 21: "This was because...": Has causality been established?

Response:

"This was because..." was removed. This sentence was rewritten.

p. 8413, line 23: "hygroscopic" would be better than "water-soluble"

Response:

"Hygroscopic" is used in the MS.

p. 8414, line 2: The processes were not observed directly. Rather, they can be inferred (to a degree) by the measurements. This should be made clear.

Response:

More texts were added into the MS

Modification in the MS:

"As shown in the Fig. 3(a), peak daily κs of 50, 75, and 110 nm particles occurred afternoon and minimum appeared in the midnight. The evolution of particle hygroscopicity was very similar to those of inorganic mass fraction (sulfate+nitrate+ammonium) in 30-100 nm particles. During the daytime, H_2SO_4 concentration increased and may condense onto the particles. At the same time, the increasing ambient temperature (see Fig. 2 (b)) could drive the semi-volatile organic species in particle phase to partition to gas phase. Both processes could result in an increasing of inorganic fraction in particle phase, thereby enhancement in particle hygroscopicity."

p. 8414, line 4: "Significant" should only be used when statistical significance is established.

Response:

"Significant" was removed from the texts.

p. 8414, line 6: Please substantiate the statement: "sulfuric acid condensation played a minor role in particle growth"? And what is the time period?

Response:

"sulfuric acid condensation played a minor role in particle growth" was removed from the texts.

p. 8414, lines 6 – 10: "Lower temperature facilitates [: : :] leading to an evident decline of hygroscopicity." It seems to me that the initial observation was the decrease in hygroscopicity and the increase in organic mass fraction, and the condensation of semivolatile compounds is a possible reason, rather than the reverse: stating a plausible process (condensation of semi-volatiles) and treating the observations as "confirming" it.

Response:

The sentences were rearranged.

Modification in the texts:

"Fig.3 displayed the size-resolved particle hygroscopicity (a), m/z 44 and 57 concentrations (b), and mass fraction of organic, sulfate, nitrate, and ammonium in 30-100 nm (mobility diameter) particles (c). As shown in the Fig. 3(a), peak daily κs of 50, 75, and 110 nm particles occurred afternoon and minimum appeared in the midnight. The evolution of particle hygroscopicity was very similar to those of inorganic mass fraction (sulfate+nitrate+ammonium) in 30-100 nm particles. During the daytime, H_2SO_4 concentration increased and may condense onto the particles. At the same time, the increasing ambient temperature (see Fig. 2 (b)) could drive the semi-volatile organic species in particle phase to partition to gas phase. Both processes could result in an increasing of inorganic fraction in particle phase, thereby enhancement in particle hygroscopicity. The decline in particle hygroscopicity took place after 15:30 (Local time) when sulfuric acid concentration decreased significantly. Simultaneously, ambient temperature decreased to 10°C. Lower temperature facilitates the condensation of semi-volatile organic vapors onto the particles. As a result, the organic mass fraction increased significantly during nighttime, as shown by AMS measurements (Fig.3 (c)), leading to an evident decline in particle hygroscopicity."

p. 8414, lines 9 and following: "when these particles grew to: : :" How were the hygroscopicity measurements at the different particle sizes attributed to the newly formed (rather than pre-existing) particles? How was the growth traced, and which exact hygroscopicity measurements were taken at which point in the growth process?

Response:

The equivalent water-soluble fraction 35, 50, and 75 nm particles are corresponding to the HTDMA measurement points at which the mean geometric diameter (Dm) of ultrafine particle mode reached 35, 50, and 75 nm.

p. 8414, line 17 and following: Would the size-resolved AMS chemistry data not be more helpful here, rather than the bulk organic fraction, which surely is more sensitive to larger particles?

Response:

We analyzed the size-resolved AMS data. The response is given above.

p. 8414, line 20: "considering that the chemical species contributing to particle growth was similar to: : :": This is an assumption and should be stated as such.

Response:

It was removed from the text.

p. 8414, line 18: please define "later stage"

Response:

"later stage" is changed to be "a relatively later time of the NPF event"

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Section 4.3:

p. 8415, line 12: ": : :..step-wisely enhanced due to the accumulation processing" It is not completely clear to me what is meant by that.

Response:

"During three consecutive NPF days, the CCN number concentration step-wisely enhanced due to the accumulation processing." was removed from the text.

p. 8415, line 17 "same time period before the end of the event": please define "end ofevent"

Response:

The starting and ending time of PNF events have been marked in the Fig.1 (a).

p. 8416, lines 22 – 24: Please substantiate the statement on BVOCs.

Response:

The VOCs measurements performed at Melpitz research station showed that VOCs are mainly biogenic volatile organic compounds. The following figure copied from a supplementary material of a recent publication (Mutzel et al., 2015) displayed the VOCs concentration during summertime at Melpitz station. It is very clear that the BVOCs are dominated in the atmosphere of Melpitz.

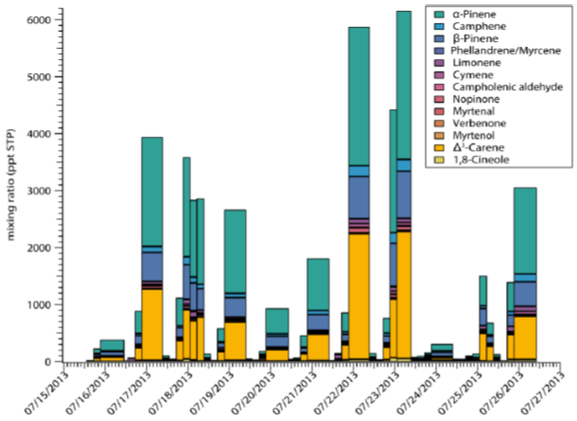

Figure S5. Time variation of the different VOC compounds sampled by TENAX cartridges. The width of each bar is covering the complete sampling period of each cartridge.

Modification in the MS:

At Melpitz, biological activities produced a lot of biogenic volatile organic compounds (BVOCs) (Mutzel et al., 2015) and lead to an organic-rich environment during summertime. The oxidation products of BVOCs may be responsible for the new particle growth.

p. 8417, line 14: "The observation showed that: : :." This does not follow from the observations presented in this study.

Response:

"The observation showed that sulfuric acid is a key species of atmospheric nucleation." was removed from the conclusions.

****

Comments on the Tables and Figures:

Table 1: What measurement points or what averages over which time frames are shown here?

Response:

In table 1, the equivalent water-soluble fraction 35, 50, and 75 nm particles is corresponding to the HTDMA measurement points at which the mean geometric diameter (Dm) of ultrafine particle mode reached 35, 50, and 75 nm.

Modifications in the MS:

One sentence was added into the MS “Here, the equivalent water-soluble fraction is corresponding to the HTDMA measurement points at which the mean geometric diameter (Dm) of ultrafine particle mode reached 35, 50, and 75 nm.”

Figure 1: The figure is overloaded. It is vital to be able to read details out of Figure 1 to understand the text, but everything is very small. My suggestion would be to make three Figures (total): one with size distributions, critical diameter, and CCN concentrations,
a second one with chemical composition and hygroscopic growth factors, and a third with the auxiliary meteorological and gas-phase data.
a) The plot should not be this saturated (in both the upper and the lower concentration limit); a lot of detail is lost. A logarithmic concentration scale would help tremendously.
What are the white circles? The diameter axis should have minor ticks (numerous references to specific particle sizes in the text). The color bar label is in an odd place.
b) What is the unit of H_2SO_4? c) The RH axis should not extend to 120%.
Figure 2: It would be helpful if the beginning and end of the nucleation event were marked in the plot, in addition to t1 and t2.

Modifications in the MS:

Figures are replotted and added into the MS:

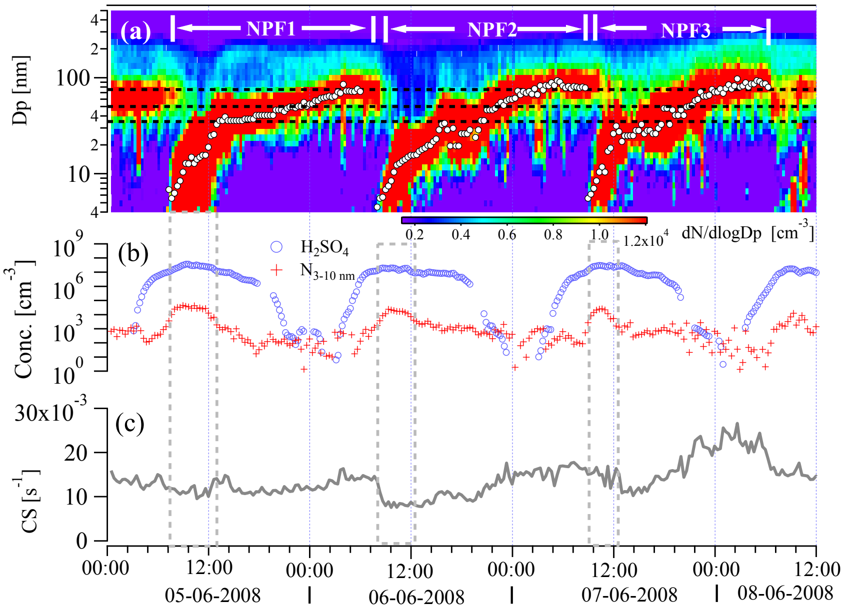

Fig. 1: Particle number size distribution, 3-10 nm particle number concentration, H_2SO_4 concentration, condensation sink (CS) during the NPF events. The starting and ending time of the events were marked in the upper place of panel (a) by NPF1, NPF2, and NPF3. The grey dashed lines indicated the time period of particle formation.

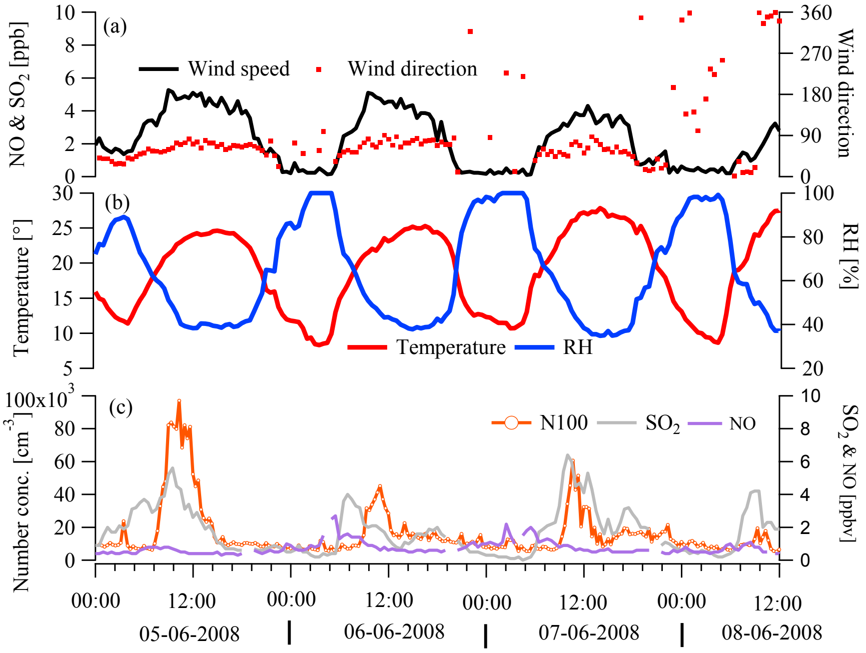

Fig. 2: The time series of wind speed and wind direction (a), ambient temperature and RH (b), and SO_2 & NO concentrations and number concentrations of particles in diameters of 3-100 nm (b).

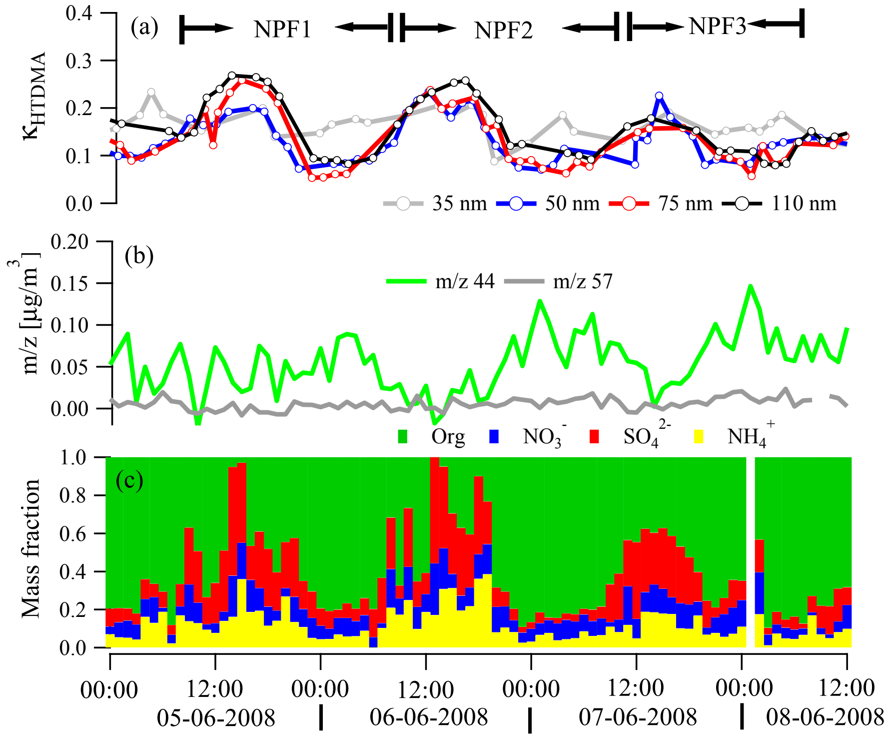

Fig.3: Size-resolved particle hygroscopicity (a), m/z 44 and 57 concentrations (b), and mass fraction of organic, sulfate, nitrate, and ammonium in 30-100 nm in mobility diameter (c).

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