

Response to reviewer 1:

We are very grateful to the reviewer for the constructive suggestions and for the proposed corrections to improve our paper. Here, all the issues raised had been addressed. Accordingly, the manuscript had been modified.

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General comments:

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*1. Many of the methods used in this study are over-simplified, which by itself would not be a problem if the interpretation of the results would be made accordingly. However, the authors seem to take the results almost at face value, discuss each individually, and do not try to combine all the results into any kind of larger picture which would increase our understanding of NPF. The conclusions in practice state that two different NPF events were observed, with the condensing vapors changing over time in both, and that some methods indicate strong H<sub>2</sub>SO<sub>4</sub> involvement, while some indicate the opposite. This is not enough to warrant publication in ACP, and the analysis should be expanded and improved. The authors measured growth rates during two NPF events. Then the composition of 10-50 nm particles is estimated using three different methods: 1) Calculating the contribution to growth from H<sub>2</sub>SO<sub>4</sub> and coagulation. 2) Measuring the changes in hygroscopicity of 25-45 nm particles. 3) Measuring the changes in PMI composition. During neither event do these methods agree. During the first event (Oct 12), method 1) suggests marginal contribution from sulfate, while 2) and 3) suggest a more pronounced contribution. On Oct 14, 1) shows that sulfate could explain the entire growth (though there was in fact none), while 2) shows that organics might be more important, and 3) is undecided. The authors add some speculation after each method, but there is practically no discussion on the discrepancies between the methods. Possible errors rising from the methods, calculations or interpretation need to be addressed for the reader to know if indeed any of the methods are actually reliable/applicable for getting information on the composition of 10-50 nm particles.*

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**Response:**

Considering the reviewer's comments, we reorganized the manuscript. The discussion on AMS measurements was deleted because the AMS cannot detect the chemical composition of newly formed particles. In the modified manuscript, we focused on the comparison between the water soluble fraction of newly formed particles derived from HTDMA measurements and that estimated assuming the H<sub>2</sub>SO<sub>4</sub> condensational growth. The main goal is to investigate whether the water soluble fraction of newly formed particles can be fully explained by H<sub>2</sub>SO<sub>4</sub> condensation. The error analysis was added into the revised manuscript. Another method (Nieminen et al., 2010) for calculating particle growth contributing by H<sub>2</sub>SO<sub>4</sub> condensation was adopted instead of that in Stoltzenburg et al (2005). This new method is more straightforward. In addition, a Lagrangian study was performed to estimate the particle growth rate by following the reviewer's suggestion. The detailed modifications are given in below.

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2. As a clear example of the lack of error analysis in the paper, at the end of section “2.5. Estimation of H<sub>2</sub>SO<sub>4</sub> concentration”, the authors claim that due to the high radiation during the events, the “proxy is accurate”. Later the growth rate by H<sub>2</sub>SO<sub>4</sub> during Oct 12 is reported to be 0.28 nm/h without any uncertainties discussed. Mikkonen et al (2011) gave an overall “relative error” of about 40% for the proxy, and also concluded that the proxy was worst at predicting H<sub>2</sub>SO<sub>4</sub> concentrations at another mid-level mountain site in Germany. Based on Fig. 7 in that paper, one could conclude that the error in the proxy might be up to a factor of 5 at times.

**Response:**

In order to investigate the similarity between our measurement site and the Meteorological Observatory Hohenpeissenberg, (HPB, the mountain site in Mikkonen et al.’s paper), we compared the key parameters, which were used to construct a proximity measure of H<sub>2</sub>SO<sub>4</sub> concentration in Mikkonen et al.’s study at our research station with those at HPB. The summary of key variables for our field campaign is given in the following table. It shows that these key variables in our measurement site are more similar to other stations rather than HPB. Therefore, the H<sub>2</sub>SO<sub>4</sub> estimated by the proxy proposed by Mikkonen may not produce a factor of 5 in our study. In the revised manuscript, we took the uncertainty of 40% in the estimation of H<sub>2</sub>SO<sub>4</sub> concentration.

	[SO <sub>2</sub> ] ppb (>0.1 ppb)	CS 10 <sup>-3</sup> s <sup>-1</sup>	Radiation W m <sup>-2</sup> (> 1 W m <sup>-2</sup> )	RH %	[O <sub>3</sub> ] ppbv	Temp K
Mean	0.81	5.8	144.3	88.0	21.2	282.7
sd	0.70	3.3	178.4	13.3	10.0	277.5
Median	0.57	5.1	62	94	21.0	281.9
5-95%	0.23-2.0	1.8-12.4	9-555	57-98	6.8-39.2	274.6-289.3
Similar to	Melpitz	San Pietro Capofiume (SPC), Italy	Hyytiälä 2003	On average, RH at Goldlauter is higher than other sites	Hyytiälä 2007	Hyytiälä 2007 and HPB

Modifications in the MS:

“The H<sub>2</sub>SO<sub>4</sub> concentration is estimated according to the proxy reported by Mikkonen et al. (2011):

$$[H_2SO_4] = 8.21 \times 10^{-3} \cdot k \cdot Radiation \cdot [SO_2]^{0.62} (CS \cdot RH)^{-0.13} \quad [4]$$

where  $k$  is the reaction rate constant, which is calculated according to the equation [3] in Mikkonen et al. (2011) and is scaled by multiplying it with 10<sup>12</sup>. CS is the condensation sink, which determines how rapidly molecules will condense onto pre-existing aerosols (Kulmala et al., 2005). CS in s<sup>-1</sup> is calculated according to Dal Maso et al., (2005) under dry condition. Radiation is global radiation in W m<sup>-2</sup>, RH is relative humidity in %. SO<sub>2</sub> is the sulfur dioxide concentration in molecules cm<sup>-3</sup>.

In Mikkonen et al.'s study, they pointed out the proxy is the worst for predicting  $\text{H}_2\text{SO}_4$  concentration at a mid-level mountain site in Germany (The Meteorological Observatory Hohenpeissenberg, HPB). However, we compared the key variables, which were used in Eq. [4], measured at our measuring site with different sites in Mikkonen et al.'s paper. It was found that these key variables in our measurement site are more similar to other stations (Melpitz and Hyytiälä) rather than HPB. Therefore, the  $\text{H}_2\text{SO}_4$  concentration estimated by this proxy may not produce an extreme high error. Here, relative error in  $[\text{H}_2\text{SO}_4]$  estimation using equation [4] is considered as 40% (Mikkonen et al., 2011).”

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3. The homogeneity of the surroundings is a strongly underlying assumption throughout the paper, and this should be discussed in more detail, starting from a more detailed site description. To me, a mode of 25 nm particles appearing, remaining without growing for 4 hours and then disappearing, suggests inhomogeneous surroundings causing particle formation and growth. And if this is indeed the case, the NPF event on Oct 12 may also be influenced, and the observed GR may be an over- or underprediction of the real particle GR. Here the authors should make more use of the actual aim of the campaign, and do Lagrangian studies. The three stations are relatively close, but as the average wind speeds were around or below 1 m/s during both events, differences should be visible between the size distributions. This would give an independent GR measurement which should add valuable information to the interpretation. Additionally, I am extremely confused about Fig. S1. I am aware that the color scale is different, as well as the y-axis scale for some reason, as compared to Fig. 1, but should not the top two plots in Fig. 1 be the same as in Fig. S1? The modal diameter during Oct 12 seems to be clearly larger in Fig. 1 than in Fig. S1. Also, the top and bottom right plots in Fig. S1 are identical!

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**Response:**

This idea is really great. We reanalyzed the data step by step. The discussions are given below.

**(1)** A model of the terrain is shown in Fig. 1 (Herrmann et al., 2005). The Thüringer Wald Mountain extends in south-easterly direction. As indicated in Fig. 1, three research stations were employed: Schmücke (the summit of the mountain, 937 m above sea level), Goldlauter (605 m a.s.l.), and Gehlberg (732 m a.s.l.). The surrounding of stations is covered by forest. The dominant trees in this region are Norway spruces (8–23 m).

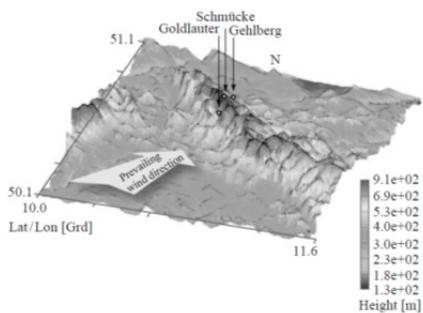


Fig. 1: Map of Central Germany with the marked area of investigations (Copy from Herrmann et al. (2005)).

**(2)** The particle number size distributions measured at three research stations are given in Fig. 2. In the old supporting material (Fig. S1), we made a mistake. The minimum particle size measured by SMPS is 10 nm. We have already corrected in the new version. From Fig.2, we can find that the evolution of the particle number size distributions during the NPF events at Goldlauter and Schmücke are very similar, and quite different from that at Gehlberg. The NPF event at Gehlberg is relatively weak and started with a larger size of newly formed particles. Next step, we will investigate the meteorological parameters at three research stations.

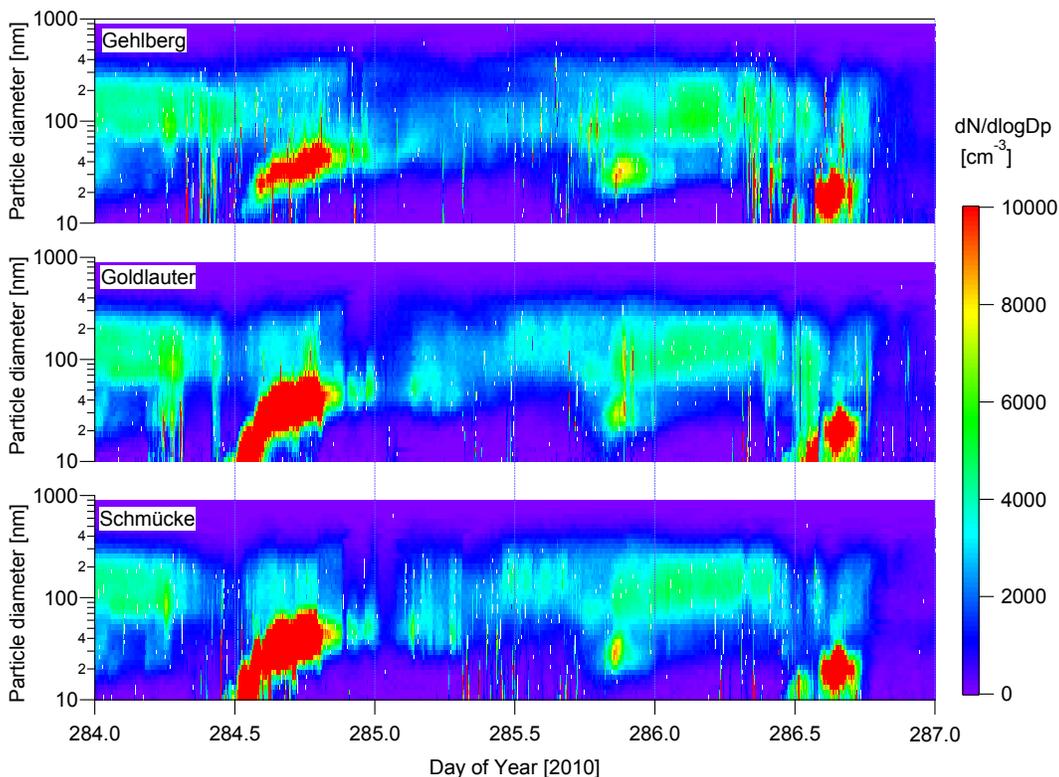


Fig. 2: The particle number size distributions at three research stations.

**(3)** The relative humidity (RH) on October 14 and 12, 2010 are presented in Fig. 3. One can see that the RH at Gehlberg station is 90%, which is much higher than that on other two stations. It could be foggy at Gehlberg. Sometime, it is visible at Gehlberg station, as photo shown in below. Therefore, we will only consider the Goldlauter and Schmücke research stations.

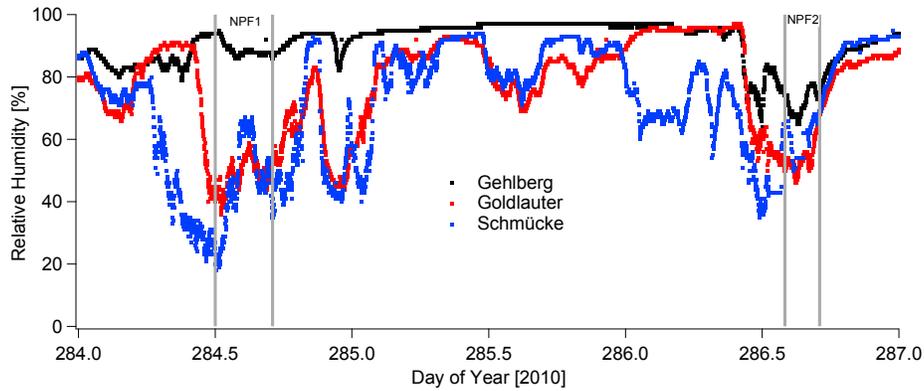


Fig. 3: RHs at three research stations.



The photo taken in Gehlberg station

**(4)** Before performing the Lagrangian study, the air flow conditions are examined. Except for the meteorological parameters, such as wind direction, the number concentration of 100 nm particle with longer life-time in the air is used as an additional tracer to examine the air flow connection between two research stations. As shown in Fig. 4, the number concentration of 102 nm particles -- being representative for the Aitken particle mode, shows a covariation at the two sites during 12:00-18:00. (We chose 102 nm particles because this part of the size distribution belongs to the part that is least influenced by particle formation or particle deposition processes; statistical analyses for the overall HCCT campaign (work in progress) showed that this particle size range is the most suited for a consideration of connected flow.) The covariation of 102 nm particles is

one important factor suggesting that there was a good flow connectivity between the Schmücke and Goldlauter research stations.

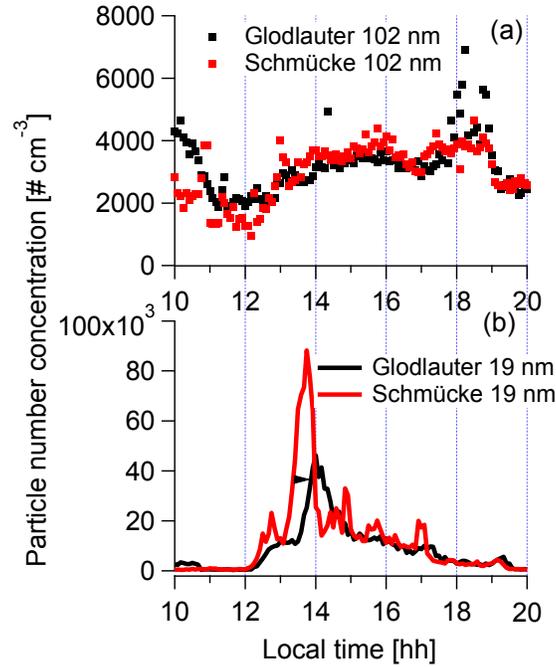


Fig. 4: The time series of particle number concentration during NPF event on October 12.

The particle growth rates  $GR_{Lag}$  is now calculated on the basis of a Lagrangian consideration. The calculation is based on the picture that particles are formed in the upwind region of the HCCT area, and subsequently transported across the measurement sites.

If there is a good airflow connectivity between two sampling sites (Here upwind site A and downwind site B), the change in particle size can be observed when air mass transported from A to B. Hence, the Lagrangian growth rate ( $GR_{Lag}$ ) can be estimated:

$$GR_{Lag} = \frac{D_{g,B} - D_{g,A}}{\Delta T} \quad [7]$$

where  $D_{g,A}$  is the GMD of particle number size distribution measured in site A at time T, and  $D_{g,B}$  is the GMD in site B at time T+ $\Delta T$ .  $\Delta T$  is the time interval during which air parcel travelled from A to B.

The detailed discussions see below:

## **Modifications in MS**

### **“4.4 Comparison between $GR_{obs}$ and $GR_{Lag}$ ”**

The particle growth rate  $GR_{Lag}$  is now calculated using Lagrangian consideration. The calculation is based on the picture that particles are formed in the upwind region of the HCCT area, and subsequently transported across all three measurement sites. For the methodological comparison of growth rates, the NPF event on October 12, 2010 is analyzed. For the methodological comparison of growth rates, the NPF event on October 12, 2010 is analyzed. Due to it is foggy at Gehlberg station, here Schmücke and Goldlauter stations are considered only (see supporting material).

Calculating  $GR_{Lag}$  requires an analysis of the atmospheric flow across the sites Schmücke (upwind) and Goldlauter (downwind). Besides pure meteorological considerations involving temperature, wind speed and wind direction, we chose the number concentration of 102 nm particles ( $N_{102}$ ) as a preferential tracer for air flow connectivity. One particular reason is that an examination of particle size distributions during HCCT suggested that this particle size range was the least influenced by any particle formation, deposition, or cloud activation processes during transport. (Details of this topic are not shown here; they will be part of a forthcoming paper in this special issue.)

Fig. 1 shows that the wind direction at Schmücke straddled around  $45^\circ$  (northeast) on October 12. The average wind speed was  $2.3 \text{ m s}^{-1}$  during the NPF event. Fig. 5a shows similar levels of  $N_{102}$  at Schmücke and Goldlauter between 12:00 and 18:00 LT. These arguments indicate a direct flow connection between Schmücke and Goldlauter. At this wind speed, the travel time of the air parcel was estimated to be 25 min. This travel time agrees with the time delay in the number concentration of 19 nm particles ( $N_{19}$ ; Fig. 5b), i.e. the observations are consistent with the picture of particle formation upwind the HCCT sites and subsequent transport of that air parcel across the study area. For the evaluation of Eq. 7 we assume we assume this observed time difference  $\Delta T$  of 25 min

We subsequently evaluated differences in the nucleation mode particle size between Schmücke and Goldlauter by comparing the lognormal modal diameters obtained from a least-squares fitting routine. The size difference  $GR_{Lag}$  could be calculated at the time resolution of the SMPS measurements, i.e. every 5 min. Averaging over the time period 13:00-18:00 LT yields a mean particle growth rate  $3.8 (\pm \dots) \text{ nm h}^{-1}$  obtained from this inter-site data comparison. During this period,  $GR_{obs}$  calculated from Eq. 6 is  $5.0 (\pm \dots) \text{ nm h}^{-1}$ , i.e. 24% higher than  $GR_{Lag}$ . In view of the considerable uncertainties involved in both

methods (we estimate the uncertainty to amount to roughly a factor of two), we consider both values consistent.

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4. Another opportunity to make better use of the acquired data is to go further in calculating the “soluble fraction”. The use of a “soluble fraction” is questionable, but it should be pointed out in the text that the main benefit of that term is that it is independent of particle size and the RH at which the GF was measured. The authors could try to relate the GF of the organics in the AMS to the measured GF based on O:C ratios (according to e.g. Massoli et al., 2010, *Geophys Res Lett*, Relationship between aerosol oxidation level and hygroscopic properties of laboratory generated secondary organic aerosol (SOA) particles).

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**Response:**

We mentioned the benefit for using the soluble fraction term in the modified manuscript. The relationship between O:C ratio and growth factor of organics was investigated, as shown in Fig. 5. Because this relationship is not closely relevant to the current topic, we did not show it in the modified manuscript. It will be discussed in another manuscript.

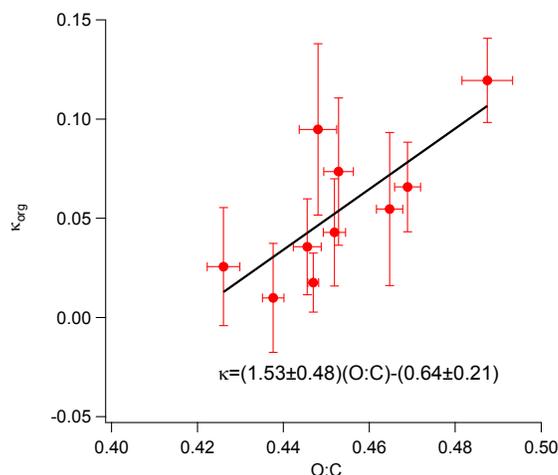


Figure 5: Relationship between O:C and k<sub>org</sub>.

Modifications in MS:

“The advantage of using soluble fraction term is to allow us analyzing 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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5. Section 2.6: I had to look up Stoltzenburg et al (2005) to understand what calculations were actually made based on this section. As written now, the section is confusing, at least for readers who have not

done these specific calculations themselves. Several equations are listed without a good explanation how one ties into the other, and the actual important steps are left unmentioned. The flow chart in Fig. A1 in Stoltzenburg et al (2005) is an extremely efficient way of showing what is actually done, and something similar would fit better into this paper than listing some equations that can be used to calculate different quantities. The authors should clarify this section, either by rephrasing the text and perhaps adding a flow chart, or as I would recommend, remove at least equations 5, 7 and 8, and just shortly explain the important steps and refer the interested readers to Stoltzenburg et al (2005). Lacking major points that definitely should be explicitly mentioned are that modes were fit to the SMPS data, and that the GR from H<sub>2</sub>SO<sub>4</sub> is calculated based on the H<sub>2</sub>SO<sub>4</sub> proxy. Finally, I do not understand the end of the section, where intramodal coagulation "can contribute to sulfate fraction". Intramodal coagulation should not change any fractions inside the mode.

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### **Response:**

Thanks for the comments. In the revised version, a new method for estimating particle growth rate contributed by sulfuric acid developed by Nieminen et al., (2010) will be used instead of the one presented in Stoltzenburg et al (2005). Firstly, the vapor concentration required for growth rate of 1 nm/h in certain particle size ranges is calculated according to (Nieminen et al., 2010):

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

here  $x_0$  and  $x_1$  are the ratios of the vapour molecule diameter ( $d_v$ ) to the initial and final particle diameter, respectively. Then the growth rate contributed by sulfuric acid during the time period used for the determination of GR $x_0$ - $x_1$  is calculated directly as:

$$GR_{H_2SO_4} = [H_2SO_4]_{\text{det}} / C_{GR=1\text{nm/h}, H_2SO_4} \quad [2]$$

Where  $[H_2SO_4]_{\text{det}}$  is the median value from the measured sulfuric acid concentration during the timeframe for the determination of GR $x_0$ - $x_1$ .

### **Modifications in MS:**

#### **“3.4 Particle growth contributed by H<sub>2</sub>SO<sub>4</sub> condensation**

Theoretically, the vapor concentration required for growth rate of 1 nm/h in certain particle size ranges can be calculated according to (Nieminen et al., 2010):

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

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 H<sub>2</sub>SO<sub>4</sub> applied in this study are 135 amu and 1650 kg/m<sup>3</sup>, respectively, corresponding to hydrated H<sub>2</sub>SO<sub>4</sub> molecules (Kurtén et al., 2007). It should

note that equation [8] was developed specially for particle with diameter of 3-7 nm. For larger particles (>10 nm, the case in this study), this method give similar results to that calculated using the Fuchs-Sutugin approach (Nieminen et al., 2010). The calculated  $C_{GR=1nm/h, H_2SO_4}$  may be an underestimate because it is assumed that every  $H_2SO_4$  molecule colliding with the particle is attached to it which is not necessarily the case.

Then the growth rate contributed by  $H_2SO_4$  during the time period used for the determination of  $GR$  is calculated directly as:

$$GR_{H_2SO_4} = [H_2SO_4]_{det} / C_{GR=1nm/h, H_2SO_4} \quad [9]$$

where  $[H_2SO_4]_{det}$  is the median value from the measured  $H_2SO_4$  concentration during the timeframe for the determination of  $GR$ .

For a spherical particle, its volume change ( $\Delta v$ ) due to condensational growth within the time interval of  $\Delta t$  can be simply calculated:

$$\Delta v = \frac{\pi}{6} \cdot (Dp_t^3 - Dp_0^3) = \frac{\pi}{6} \cdot (Dp_t - Dp_0) \cdot (Dp_t^2 + Dp_t Dp_0 + Dp_0^2) \quad [10]$$

$$\text{Here } Dp_t - Dp_0 = GR_{obs} * \Delta t \quad [11]$$

Where  $Dp_t$  and  $Dp_0$  are the GMDs at time  $t=0$  and  $t=\Delta t$ , respectively.

The observed growth rate can be presented as the sum of the growth rates due to  $H_2SO_4$  ( $GR_{H_2SO_4}$ ) and organic vapors ( $GR_{org}$ ) condensation (Paasonen et al., 2010):

$$GR_{obs} = GR_{H_2SO_4} + GR_{org} \quad [12]$$

By combing equations [10-12], the overall change of particle volume concentration can be separated into two fractions, contributing by  $H_2SO_4$  and organic vapors condensation. The fraction contributed by  $H_2SO_4$  can be presented as:

$$\Delta v_{H_2SO_4} = \frac{\pi}{6} \cdot (Dp_t^2 + Dp_t Dp_0 + Dp_0^2) \cdot GR_{H_2SO_4} \cdot \Delta t \quad [13]$$

We assumed that the original particle only consists of ammonium sulfate. Then, the volume fraction of  $(\text{NH}_4)_2\text{SO}_4$  in total particle volume ( $V_t$ ),  $\varepsilon_{(\text{NH}_4)_2\text{SO}_4}$ , can be estimated as:

$$\varepsilon_{(\text{NH}_4)_2\text{SO}_4} = \frac{v_0 + \Delta v_{\text{H}_2\text{SO}_4}}{\pi * Dp_t^3 / 6} \quad [14]$$

where  $v_0$  is the original particle volume at time 0. Here, assuming original particle as ammonium sulfate may not cause larger error due to the tiny mass compared to the particles after growing. One should keep in mind that the neutralization of  $\text{H}_2\text{SO}_4$  by  $\text{NH}_3$  could also lead to the increase in particle volume, which was not considered in equation [13]. Therefore, the  $\varepsilon_{(\text{NH}_4)_2\text{SO}_4}$  is lower estimated.”

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Specific comments:

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*The grammatical errors were too numerous to list in the technical corrections, and the language needs to be improved before publication in ACP is possible.*

Response:

We improved the language.

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*Page 11416, line 5: HCCT should be written out or just not mentioned in the abstract.*

**Response:** we wrote it out in the abstract.

“on Hill Cap Cloud Thuringia 2010 (HCCT-2010) field campaign”

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*11417, 6-17: In a list of instruments capable of measuring ultrafine particles, the NAMS should also be mentioned. See e.g. Zordan, C. A.; Wang, S.; Johnston, M. V. Time resolved chemical composition of individual nanoparticles in urban air. Environ. Sci. Technol. 2008, 42 (17), 6631–6636.*

**Response:** We cited this paper in the modified manuscript.

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*11418, 22-24: Why do the authors talk about upwind and downwind sites? Does this not imply a constant wind direction? Now Goldlauter is named an upwind site, but at least in Fig. 2a, it should be downwind of the other sites.*

**Response:** The definition of upwind and downwind sites is used in the identification of cloud events. Here, we will remove it from manuscript.

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*11419, 10: What is an "automatic" silica gel dryer?*

**Response:** An automatic regenerating adsorption aerosol dryer was used to keep the RH of inlet below 30%. The working principle of this dryer is described in detailed by Tuch et al. (2009).

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*11419, 17-18: If you are going to talk about DMA1 and DMA2, they need to be mentioned earlier. Overall a 2-3 sentence explanation of the working principle of an HTDMA would be useful.*

**Response:** We added some texts in the MS:

“Only a brief description is given here. The H-TDMA consists of three main parts: (1) A Differential Mobility Analyzer (DMA1) that selects quasi-monodisperse particles, and a Condensation Particle Counter (CPC1) that measures the particle number concentration leaving the DMA1 at the selected particle size; (2) An aerosol humidifier conditioning the particles selected by DMA1 to a defined relative humidity; (3) The second DMA (DMA2) coupled with another condensation particle counter (CPC2) to measure the number size distributions of the humidified aerosol.”

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11420, 2: Draxler

**Response:** We corrected.

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11420, 18 & 22: First the authors state that SOA is typically hydrophobic, and then that SOA typically has a growth factor  $> 1$ . The intent with the soluble fraction needs to be clarified, together with an improved discussion on the organic growth contribution.

**Response:**

The water soluble fraction is only an “equivalent” soluble fraction (i.e. assuming ammonium sulfate as the only soluble substance).  $\epsilon_{\text{soluble}}$  is therefore an upper estimate for the true soluble volume fraction. The advantage of using soluble fraction term is to allow us analyzing the particle hygroscopicity independently of differences in size. We clarified in the MS.

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11424, 6-7: Are the authors implying that the end of the event and increase in wind speed are related? If so, please state how. Otherwise rephrase.

**Response:** Yes, the increased wind speed resulted in a decrease in particle concentration, indicating the injection of clean air mass. The particle growth was not observed after air mass change.

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11424, 26-28: on Oct 14?

**Response:** Yes, we added the “October 14” in the sentence.

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11425, 8: decreasing

**Response:** It was corrected.

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11425, 15-16: If the mode was internally mixed, why does the GF at 30 nm differ so much from the rest?

**Response:**

We rechecked the GF-PDF during the NPF events. Around 16:00, the 30 nm-particles are internally mixed. Later, particles changed to be externally mixed around 16:45. It will be clarified in the manuscript.

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11425, 25: I recommend that you do not speculate on which species took part in the actual nucleation based on the soluble fraction of 25 nm particles. What does the Zhang 2004b citation refer to in this sentence?

**Response:** This citation was removed from MS.

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*11426, 1-4: Certainly all these reaction would decrease the GF of H<sub>2</sub>SO<sub>4</sub>, but they would probably increase the GF contribution of any reacted organics. Are the authors sure that these reactions would cause a net decrease in GF of a particle?*

**Response:**

We are not sure on this point. This statement will be deleted.

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*11426, 13: Why only use 12:00-17:00. Earlier it is stated that the event continues until 20:00. The period 17-20 may be more interesting as the calculated H<sub>2</sub>SO<sub>4</sub> is much lower (zero at the end of the event).*

**Response:**

We make it clear in the revised manuscript. The time period of 13:00-20:00 will be focused on.

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*11427, 13: The authors seem to state that the derivative of a constant “cannot be calculated”. It can, and the answer is zero, as also the authors deduce from their data.*

**Response:**

We agree. “Therefore, the observed growth rate cannot be calculated using equation [6].” was removed from MS.

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*11428, 17: I would not talk about “addition” when the rise in acidity is accompanied by a dramatic drop in concentration of all AMS species. Generally, the absolute concentrations should also be taken into account in the interpretation of the data.*

**Response:**

The discussions on AMS data were deleted from the manuscript.

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*Fig. 2: Although it can be figured out, the position of the station should be clearly marked in the figure. Also, there is no need to show all of Europe in these maps. The different trajectories could be made out much better if the maps were zoomed in to e.g. 0-25 long, 50-60 lat.*

**Response:**

We modified Fig. 2.

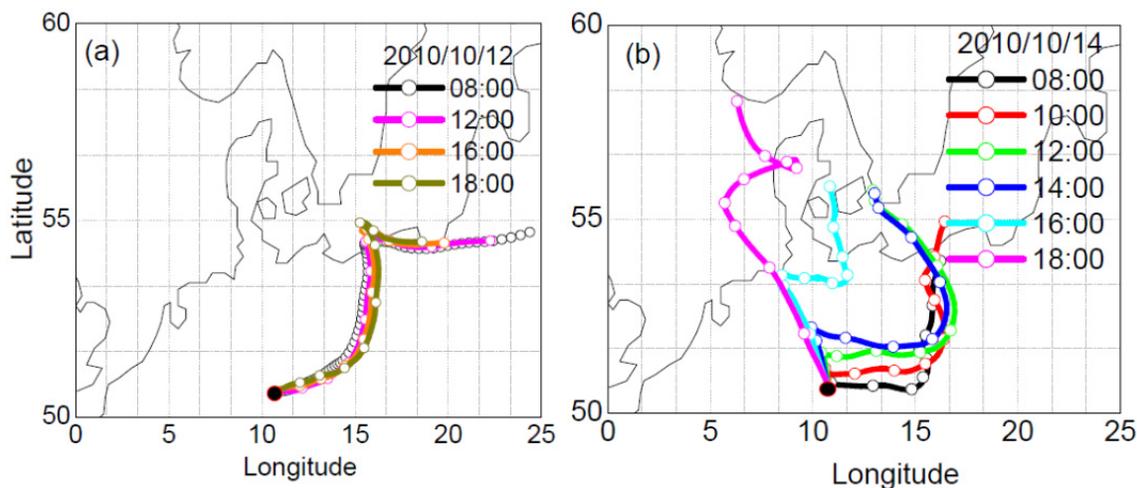


Fig. 2: 72-hour backward trajectory of the air mass (800 m above ground level) arriving at the sampling site during NPF events. The black dots indicate the research station.

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*Fig. 5 caption: The “ion molar ratio” should be explained here.*

**Response:**

This figure was deleted.

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**Reference**

Dal Maso, M., M. Kulmala, I. Riipinen, R. Wagner, T. Hussein, P.P. Aalto, and Lehtinen, K. E. J.: Formation and growth of fresh atmospheric aerosols: Eight years of aerosol size distribution data from smeer ii, hyytiälä, finland, *Boreal Environ. Res.*, 10, 323-336, 2005.

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