New particle formation event forecasts during PEGASOS–Zeppelin Northern mission 2013 in Hyytiälä, Finland

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Abstract

New particle formation (NPF) occurs frequently in the global atmosphere. During recent years detailed laboratory experiments together with intensive field observations in different locations have provided insights into the vapours responsible for the initial formation of particles and their subsequent growth. In this regard, the importance of sulphuric acid, stabilizing bases such as ammonia and amines as well as extremely low volatile organics have been proposed. The instrumentation to observe freshly formed aerosol particles has developed to a stage where the instruments can be implemented as part of airborne platforms, such as aircrafts or a Zeppelin-type airship. Flight measurements are technically more demanding and require a greater detail of planning than field studies at the ground level. The high cost of flight hours, limited time available during a single research flight for the measurements and different instrument payloads in Zeppelin airship for various flight missions demanded an analysis tool that would forecast whether or not there is a good chance for a NPF event. Here we present a methodology to forecast NPF event probability at the SMEAR II site in Hyytiälä, Finland. This methodology was used to optimise flight hours during the PEGASOS-Zeppelin Northern mission in May–June 2013. Based on the existing knowledge we derived a method for estimating the nucleation probability that utilizes forecast air mass trajectories, weather forecasts and air quality model predictions. With the forecast tool we were able to predict the occurrence of NPF events for the next day with more than 90% success rate (10 out of 11 NPF event days correctly predicted). To our knowledge, no similar forecasts of NPF occurance have been developed for other sites. This method of forecasting NPF occurance could be applied also at other locations, provided that long-term observations of conditions favouring particle formation are available.

1. Introduction

Formation and growth of secondary aerosol particles has been observed in numerous locations and in different environments in the planetary boundary layer (for an overview see e.g. Kulmala et al., 2004; Kulmala and Kerminen, 2008). Numerous investigations have attempted to connect new particle formation (NPF) to atmospheric trace gas concentrations, atmospheric chemistry and meteorological processes (e.g. Weber et al., 1995; Riipinen et al., 2007; Paasonen et al., 2010). Most of the NPF observations are based on stationary ground-level measurements during which the sampled air masses and prevailing meteorological conditions are continuously changing. Typically the growth of the newly formed particles can be followed for several hours from these fixed point measurements, indicating that NPF usually occurs over large areas (Dal Maso et al., 2007; Hussein et al., 2009). In order to obtain more information on the spatial extent of NPF events both in the vertical and horizontal directions, measurements using aircrafts are needed. As part of the 4 year-long EU funded PEGASOS (Pan-European Gas-Aerosol-Climate Interaction Study) project, a Zeppelin NT (Neue Technologie) airship was performing atmospheric aerosol, trace gas and photochemistry measurement flights in Central Finland during May–June 2013. In order to most efficiently utilize the flight hours of the airship, it was necessary to prepare forecasts on the probability of NPF events in the coming days.

Most of the Zeppelin measurement flights during the campaign were directed to the vicinity of the University of Helsinki SMEAR II measurement station in Hyytiälä (Hari and Kulmala, 2005). Measurements of aerosol number size-distributions, trace gas concentrations and basic meteorological quantities were started at the SMEAR II station in January 1996. These long time-series records have been used extensively to characterise the conditions in which NPF occurs (or does not occur) in this boreal forest environment, based on both the local atmospheric conditions as well as the synoptic situation and air mass origins and transport route to the station (Boy and Kulmala, 2002; Boy et al., 2003; Lyubovtseva et al., 2005; Dal Maso et al., 2007; Sogacheva et al., 2008, Nieminen et al., 2014).

Field observations, laboratory experiments and theoretical considerations have shown that sulphuric acid is one of the key components in atmospheric NPF events, but in addition also trace amounts of other vapors such as ammonia, amines or oxidized organics are needed (e.g. Kulmala et al., 2013). Particularly the contribution of extreme low volatile organics seems to be crucial in the boreal forest environment (Kulmala et al., 1998; Yli-Juuti et al., 2011; Ehn et al., 2014). Proxies for the concentrations of these trace gases or their precursors have been developed based on campaign-wise measurements (Petäjä et al., 2009; Lappalainen et al., 2009). Based on the concentrations and emissions of these trace gases, several parametrizations have been developed to describe the occurance

and intensity of NPF (e.g. Buzorius et al., 2003; Bonn et al., 2008; Paasonen et al., 2010; Häkkinen et al., 2013).

In this work, we describe forecasts for the occurrence of NPF at the SMEAR II station. The forecasts are based on the above-mentioned long-term time series observations of the typical conditions during NFP days and non-NPF days, the air mass origins as well as weather and air-quality forecasts.

2. Materials and methods

The main objective of the NPF forecasts was to predict whether during the next three days NPF events were likely to occur at the SMEAR II station area. A time period of three days was chosen in order to have long enough time for preparing the measurement instruments needed on different flights while still maintaining reliability of the input data used in making the NPF forecasts. The final NPF forecast was always provided for the next day, as the Zeppelin measurement flights were typically planned one day in advance. All the NPF forecast results presented in this work refer to the final NPF forecasts, i.e. forecasts for the next day.

2.1 Predictions for trace gas concentrations, particulate matter and meteorology

Forecasts for concentrations of trace gases SO_2, O_3, NO_x, CO and OH as well as particulate matter (PM_{10}, comprising the total mass concentration of particles smaller than 10 μm in diameter) and relative humidity were obtained from the Finnish Meteorological Institute's SILAM (System for Integrated modeLling of Atmospheric coMposition) air quality model (Sofiev et al., 2006). This model provides predictions for the above mentioned variables for the next 5 days at several heights above the ground. Input information for SILAM includes anthropogenic emissions from the TNO-MACC data set, IS4FIRES information on wild fires, as well as emission calculations for sea salt, pollen, wind-blown dust, and natural volatile organic compounds. The weather forecast input data are obtained from the FMI HIRLAM model. The horizontal resolution of SILAM in the Scandinavian area is 6–7 km. All SILAM forecast data are freely accessible via internet (http://silam.fmi.fi/), and the forecast for the Northern Europe area is updated once per day. For the purpose of the current NPF event forecasts, we used predictions for the ground level (15 m above ground) during next three days from the model grid point nearest to Hyytiälä SMEAR II station with the time resolution of 1 hour.

As supporting data, we also used several “traditional” weather forecasts available on the internet (including forecasts by Finnish Meteorological Institute, Foreca and Norwegian Meteorological Institute), mainly to evaluate the probabilities of cloudiness and rain. During the campaign time, the

weather was rather variable and the forecasts were changing rapidly (even several times a day) from clear skies to partly cloudy and possibly rainy. All these conditions are known to affect directly the probability of NPF.

2.2 Air mass back-trajectories

Air mass arrival directions and source areas were forecast for 96 hours prior to the arrival of air at Hyytiälä using the HYSPLIT single particle Lagrangian transport model developed by NOAA and freely available on the internet (http://www.arl.noaa.gov/HYSPLIT.php). As input meteorological data for the model, we used the US National Weather Service's Global Forecasting System (GFS) weather forecast data which extends 192 hours forwards in time. The horizontal location accuracy of the air mass trajectory calculations using HYSPLIT has been estimated to be on the order of 10–30% of the total distance the air parcel has travelled (Stunder, 1996; Stohl, 1998; Draxler and Hess, 1998, 2010). We considered trajectories arriving each hour to Hyytiälä at 250-m height above ground calculated 96 hours backwards in time. Typically air masses travelled less than 1000 km during this time, meaning that the air mass source area predictions based on the back-trajectory calculations could be considered accurate within 100–300 km or better. Also, since we did not consider just individual air mass back-trajectories but rather took into account all the air masses that were to arrive during the morning and early afternoon (which is the typical time of NPF occurance in Hyytiälä), the effect of uncertainties in the position of individual trajectories was diminished.

2.3 NPF event forecasts and nucleation probability parameters

Typical conditions on NPF and non-NPF days in Hyytiälä are shown in Table 1 for May and June during years 1996–2012. The conditions are shown for the time window 8–11, which is the time when NPF typically starts in Hyytiälä. In a data-mining study of the SMEAR II station long-time series records of aerosol size distributions and meteorological parameters, Hyvönen et al. (2005) found that the condensation sink (describing the pre-existing aerosol surface area) and relative humidity were the two parameters most effectively separating NPF days from non-NPF days. Particle formation was occuring only on days with a low CS and low RH. On the other hand, photochemical production of vapors participating in nucleation and growth, namely sulphuric acid and oxidation products of organics, is more efficient in clear-sky conditions with high UV radiation intensity compared to cloudy conditions. Thus, our main criteria in forecasting NPF to occur was clear sky conditions, low condensation sink (in practise low PM_{10} concentration, which was obtained from SILAM) and low relative humidity in the early morning to noon-time, as this is the time when regional NPF events start in Hyytiälä (Kulmala et al., 2013). Note that in spring and summer time, days with low relative humidity are typically also warm and sunny, so these conditions are not necessarily independent of

each other. However, the difference between NPF days and non-NPF days is also seen in the absolute humidity (water vapor concentration, see Table 1).

The air mass source area and transport route to Hyytiälä were considered when making the NPF forecasts. In the long-time series analysis by Dal Maso et al. (2007), the occurrence of NPF in Hyytiälä was observed to be highly favorable in air masses originating from the Arctic Ocean and Northern Atlantic, and on the other hand suppressed in southern air masses. This is typically connected to clean air arriving from the west and more polluted air originating from Central and Eastern Europe, directly influencing the sink for newly formed particles. However, in air masses originating from south and south-east to Hyytiälä, SO_2 concentrations are typically higher than in westerly air masses, which would favor NPF due to a higher production rate of sulphuric acid (Riuttanen et al., 2013). Table 2 summarizes the criteria used for making the NPF forecasts. The flowchart representing the main decision making process for the NPF forecasts is shown in Figure 1. The threshold values for SO_2 and PM_{10} shown in the flowchart are based on the observed range of these variables on NPF and non-NPF days (Table 1).

We also developed several "nucleation parameters" to forecast the intensity of NPF. The parameters that worked best were either related to only the proxy concentration of sulphuric acid, or to proxies for both sulphuric acid and oxidation products of volatile organic compounds (such as monoterpenes). Paasonen et al. (2010) studied several different parameterizations for the formation rate of 2 nm particles, and found that at the Hyytiälä site nucleation rate could be mainly explained by the sulphuric acid concentration to the power of one or two.

The simplest nucleation parameter is described by the following equation:

$$NP_1 = \frac{[SO_2]\cdot[OH]}{PM_{10}\cdot RH} \quad (1)$$

where the sulphur dioxide concentration (SO_2), hydroxyl radical concentration (OH), particulate mass concentration (PM_{10}) and relative humidity (RH) are taken from the SILAM air quality forecasts for the grid point closest to Hyytiälä. The particulate mass concentration is available from the SILAM forecasts. In Hyytiälä, the PM_{10} concentrations correlate well with the condensation sink CS which describes the total sink of the newly formed particles due to the pre-existing aerosol population. The PM_{10} concentrations (in units $\mu g\ m^{-3}$) can be scaled to CS (in units s^{-1}) using the linear relationship $CS = 4.59\cdot10^{-4}\cdot PM_{10}$ (linear regression based on measurement data from Hyytiälä in 1996–2012 with correlation coefficient r=0.81). The relative humidity is included as RH^{-1} in Equation 1 in order to take

into account the observed anti-correlation between the relative humidity and particle formation intensity, mainly due to the fact that the highest sulphuric acid concentrations are limited to times of low ambient relative humidity (Hamed et al., 2011).

A nucleation parameter taking into account the oxidation products of monoterpenes, in addition to sulphuric acid, is described by the following equation:

$$NP_2 = \frac{[SO_2]\cdot[OH]}{PM_{10}\cdot RH}\cdot\frac{\exp(aT)\cdot\left(k_{OH}[OH]+k_{O3}[O_3]\right)}{BLH\cdot PM_{10}} \quad (2)$$

Here, the concentrations of sulphur dioxide SO_2, hydroxyl radicals OH and ozone O_3 (in units of cm^{-3}), particulate mass PM_{10} (in units μg m^{-3}), as well as relative humidity RH (in percentages) and temperature T (in units ºC) were obtained from the SILAM forecasts. The concentrations of monoterpenes were predicted based on the ambient temperature, as their concentrations have been shown to follow an exponential temperature dependence in Hyytiälä with the scaling coefficient a=0.078 $ºC^{-1}$ (Lappalainen et al., 2009). The OH and O_3 concentrations were used to calculate the proxy concentrations of the monoterpene oxidation products, and the reaction coefficients k_{OH}=7.5×10^{-11} cm^3s^{-1} and k_{O3}=1.4×10^{-17} cm^3s^{-1} are the averages of the reaction coefficients for individual monoterpene species weighted according to their typical concentrations observed in Hyytiälä (Hakola et al., 2003; Yli-Juuti et al., 2011). The modelled boundary layer height BLH is included in Equation 2 to take into account the dilution of monoterpene emissions into the developing boundary layer.

3. Results

3.1 Overview of the conditions during the campaign

The PEGASOS-Zeppelin Northern mission was a 40-day-long measurement campaign between 3 May and 11 June 2013. An overview of the meteorological conditions as well as trace gas and particle concentrations observed at the SMEAR II station during the campaign is shown in Figure 2. Most of the days were sunny with either clear or partly clear skies. Rain occurred on 13 days during the campaign. The air was rather clean from anthropogenic pollution, especially in the first and last week of the campaign. Occasionally, there were pollution episodes seen e.g from a ten-fold rise of the SO_2 concentration from its typical level of about 0.1 ppb. At the end of May, a longer period occurred during which more polluted continental air was transported from Central Europe to Hyytiälä.

Figure 3 shows the arrival routes of air masses to Hyytiälä during the period of our measurement

campaign. These trajectories were calculated for the 250-m arrival height above ground, and 96 hours backwards in time. From the beginning of the campaign until middle of May, approximately 17th May, the air masses originated mainly from over the Atlantic, and arrived at Hyytiälä either directly from the west over Scandinavia or from south-west making a turn over the Baltic sea. Air in Hyytiälä was relatively clean during this time, characterized by low particulate mass and trace gas concentrations. Especially SO_2 had very low concentrations during this time, with the exception of one pollution-related peak on 9 May. After mid-May, air masses turned to arrive mainly from east at Hyytiälä, originating either from over the Arctic Ocean or from the continental north-west Russia. During this time until early June, the condensation sink and PM_{10} concentrations were higher than in early May, indicating more polluted air. Also high concentration peaks in the trace gases SO_2 and CO were more frequent during this time. During the last weeks of the campaign in the beginning of June, air masses turned again to arrive at Hyytiälä from west over Scandinavia, resulting in cleaner air with low particulate matter and trace gas concentrations.

3.2 Performance of the NPF forecasts and nucleation parameters

Figure 4 shows the particle number size-distributions along with the forecasted NPF occurrence and the time-series of the nucleation parameters NP_1 and NP_2. In the beginning of the campaign, several strong NPF bursts occurred (high nucleation mode particle concentrations on 3, 6 and 8 of May), and our forecasts were able to capture these as well as the days with no new particle formation. Both of the nucleation parameters peaked on these three NPF event days, and were clearly lower on the days between NPF events, except NP_1 which had a relatively high value also on 4 May. During the beginning of the campaign time, air masses originated mainly from over the Atlantic Ocean and arrived at Hyytiälä after passing over Scandinavia. On some of these days, the air was remarkably clean, characterized by very low SO_2 concentrations (below 0.1 ppb), resulting in low sulphuric acid concentrations and weak or no NPF event on clear-sky conditions. The daytime peak value of 10^4 or higher for nucleation parameter NP_1 was typically associated with the occurance of NPF.

After mid-May until early June, the air masses arrived at Hyytiälä mainly from the east, either spending several days over continental Russia or, in some cases, coming more directly from over the Arctic Ocean via northwest Russia. The air mass circulation was driven by a persistent high-pressure system residing over Central Finland. This resulted in a rather unusual air mass transport pattern to Hyytiälä, and also made the NPF forecasting more challenging. During this time, there were situations when the polluted air masses resulted in a high condensation sink, preventing the occurrence of NPF. Also the SILAM forecasts for the SO_2 and PM_{10} concentrations were less accurate during the easterly air masses compared with air masses coming from the west or the south. This might be related to less accurate emission data for these species over the Russian area.

The nucleation parameter NP_2 started to have high values more frequently after the middle of May. One factor influencing this was the higher air temperatures during this time compared to the beginning of the campaign, as the emissions of monoterpenes are highly influenced by the ambient temperature. NPF events, however, were not as frequent during this time. On one hand, this period was influenced by the more polluted air masses arriving at Hyytiälä from the east. On the other hand this period included quite a few days (13 out of 22 days after 20th May) when a growing particle mode was observed to appear in Hyytiälä starting from sizes above 10–20 nm. These types of NPF events are typically observed during the summer time in Hyytiälä, and they might be connected to higher particle growth rates during the summer, leading to the observation of the newly formed particles after they have already grown for several hours (Buenrostro Mazon et al., 2009). Days on which the maximum value of the nucleation parameter NP_2 exceeded 0.02 started to be more likely and NPF event day rather than a non-event day.

The nucleation parameters NP_1 and NP_2 have a clear connection to the NPF: they represent the ratios between the source and sink terms for the newly formed particles. However, the numerical values for NP_1 and NP_2 and especially their uncertainty depend greatly on the weather forecast and air-quality forecast data taken from the SILAM model. As it is out of the scope of this work to evaluate the accuracy of the SILAM predictions for the various parameters used, the values of NP_1 and NP_2 presented in this study should be regarded as qualitative. When comparing the different days during the campaign, they did however provide useful information to support the NPF forecasting.

The particle number size distributions measured by the Differential Mobility Particle Sizer (DMPS) during the whole campaign are shown in the upper panel of Figure 4. Using the criteria developed by Dal Maso et al. (2005), each day was classified as either a NPF event, non-event or undefined day. On NPF event days a new mode of particles smaller than 25 nm is observed and these particles can be observed growing to larger sizes during several hours. NPF event days are further classified according to the possibility to reliably derive particle formation and growth rates (Class I) or not (Class II). The days when no new sub-25 nm particles were appearing were classified as non-NPF days. Undefined days are those days for which it was not possible to unambiguously determine whether NPF occurred or not. Table 3 shows the forecast and the corresponding event classification for each day. During the 40 day campaign, clear regional NPF events lasting for several hours were observed on 11 days in Hyytiälä. Six of these days were also forecast to be NPF days, and four to have a possibility of NPF to occur. The NPF day which we forecast to be a non-NPF day (9th June) was cloudy and had a possibility of rain according to weather forecasts, and the airmasses were forecast to originate from west, which is not the direction from where airmasses typically arrive to Hyytiälä on NPF event days (Dal Maso et al., 2007). On 10 days of the campaign there was no particle formation occurring in

Hyytiälä, and these were also forecast to be non-NPF days, except for two days (17th and 28th May) for which a possible NPF event was forecast. This was most probably caused by the very low SO_2 concentration. On only one of the days forecast to be non-NPF day, was there appearance and growth of new nucleation mode particles.

Comparison of the event classification and the event forecasts is shown in Table 4. We follow the method of Hyvönen et al. (2005) for calculating the score indices for the performance of the event forecasts on the 21 days classified as either NPF or non-NPF days (undefined days are removed from this comparison). Out of these 21 days our forecasts had two false NPF event days (non-event day forecast to be either event or to have a possibilty for event) giving a 10% false-event fraction, and one NPF event day forecast to be a non-event day giving a 5% missed-event fraction. The total error of the NPF forecasts (false and missed events) during the 21 classified days of the 40-day campaign was (2+1)/21 = 14%, which is comparable to the performance of the classification methods presented in the study by Hyvönen et al. (2005).

4. Summary and conclusions

Here we present a way to forecast new particle formation events. Being able to make such forecasts accurately is very important, for example, when airborne measurements are performed. As a summary, we made NPF forecast for 40 days. The forecasts were found to work reasonably well. Only one day when nucleation was forecast to occur was a non-nucleation event day. In total 24 days were predicted to be either NPF event days or probable NPF event days, and on 10 of them there was an NPF event, 11 were undefined (when it could not be reliably determined whether NPF occurred or not), and two were non-event days.

The main challenges in making the NPF forecasts were to obtain as reliable input data as possible from SILAM, HYSPLIT and weather forecasts. The methods utilized here are most likely also applicable to other locations where there is sufficiently long datasets available to characterise the conditions favourable for the occurrence of regional-scale particle formation. In urban areas and inside cities our methods are less likely to be applicable due to the day-to-day variation of emissions of vapours and particles from local anthropogenic sources.

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Tables and Figures

Table 1. Conditions observed at Hyytiälä during NPF and non-NPF days between 8:00 and 11:00 in months May–June 1996–2012. For each variable the median value is given and the interquartile range (25th and 75th percentiles) is shown in brackets. The median and interquartile values are calculated from all data at 30 min time resolution in the time window 8–11.

Parameter	**NPF day**	**Non-NPF day**
Temperature (°C)	11 (8–14)	13 (9–17)
Global radiation (W m^{-2})	560 (430–610)	230 (120–530)
Relative humidity (%)	45 (39–55)	76 (59–91)
H_2O (parts-per-thousand)	6.5 (4.9–8.0)	10.2 (8.6–12.4)
SO_2 (ppb)	0.12 (0.04–0.23)	0.09 (0.04–0.19)
O_3 (ppb)	39 (35–44)	34 (28–41)
Condensation sink (10^{-3} s^{-1})	2.2 (1.4–3.3)	3.9 (2.7–6.2)
PM_{10} (μg m^{-3})	3.7 (2.3–5.5)	6.6 (4.6–10.5)

401 **Table 2.** Criteria for the NPF forecasts (the source for each data is shown in parenthesis). All the
402 criteria within the category must be fulfilled, i.e. the criteria are combined with logical AND.

NPF forecast	**Criteria**
NPF day	• Sunny, clear skies (according to weather forecasts) • Low PM_{10} concentration, smaller than 3.7 μg m^{-3} (SILAM) • Low RH during the day, smaller than 45% (SILAM) • SO_2 concentration higher than 0.12 ppb (SILAM) • Airmasses originating from the Arctic Ocean or Northern Atlantic (HYSPLIT trajectories)
Weak NPF / Possibility of NPF / No continuous growth of nucleation mode particles	• Sunny day with some clouds, or partly cloudy (according to weather forecasts) • PM_{10} 3.7–6.6 μg m^{-3} and SO_2 > 0.12 ppb; OR PM_{10} > 6.6 μg m^{-3} and SO_2 > 0.23 ppb (SILAM) • RH during the day 45–76% (SILAM) • Airmasses not coming directly from the west–north-west, or passing over known areas of anthropogenic pollution (HYSPLIT trajectories)
No NPF	• Cloudy day, rain (according to weather forecasts) • High PM_{10}, higher than 6.6 μg m^{-3} (SILAM) • High RH, higher than 76% (SILAM) • SO_2 concentration smaller than 0.09 ppb (SILAM) • Airmasses originating from south (continental Europe) or east, or passing over known areas of anthropogenic pollution (HYSPLIT trajectories)

403 **Table 3.** NPF event forecasts (2nd column), and NPF event classification based on measured particle
404 size distributions (3rd column) for each day of the campaign. Class I and II NPF events refer to the
405 classification by Dal Maso et al. (2005). Remarks on the 4th column show the basis for the NPF event
406 forecast.

Date	Forecast	Classification	Remarks
3 May	Possibility of NPF	NPF (class II)	Airmasses coming from north to north-west. Low SO_2 and PM_{10}. Sunny the whole morning until mid-afternoon. RH drops to 25% during the day.
4 May	No NPF	Undefined	Airmasses coming from south-west. Elevated SO_2 and PM_{10} concentrations. Cloudy day with small chance of occasional rain.
5 May	No NPF	No NPF	Airmasses coming from south-west over southern Sweden, turning to west late in evening. Low SO_2 and PM_{10} concentration. Cloudy day with light rain throughout the day until evening. In the evening partly cloudy.
6 May	NPF day	NPF (class I)	Airmasses originating from west and circulating over western Finland. Relatively clean air, SO_2 and PM_{10} concentrations low during morning and increasing towards evening. Sunny until midday, afternoon clouds possible. RH dropping to 40% during the morning.
7 May	No continuous growth of nucleation mode particles	NPF (class II)	Airmasses coming from south-west over Denmark and southern Sweden. Elevated SO_2 and PM_{10} concentrations. Partly cloudy day. RH dropping to 55%.
8 May	NPF day	NPF (class I)	Westerly airmasses coming over Central Sweden. Low SO_2 and PM_{10} concentrations. Sunny day, warm temperatures (over +15°C). RH dropping to 35% during the morning.
9 May	No NPF	No NPF	Airmasses coming from southwest-south and circulating over southern Finland. Elevated SO_2 and PM_{10} concentrations. Cloudy and rainy day.
10 May	No NPF	No NPF	Airmasses coming from south over Baltic countries and southern Finland. Low SO_2 and somewhat elevated PM_{10} concentrations. Cloudy day with occasional light rain.
11 May	No NPF	Undefined	Airmasses coming from southwest over northern Germany, Denmark and southern Sweden. Low SO_2, somewhat elevated PM_{10} concentrations. Cloudy morning, partly cloudy in afternoon, small chance of rain. Clear skies in the evening.
12 May	Weak NPF	Undefined	Airmasses from south-west over northern France, Denmark and southern Sweden. Elevated SO_2 and relatively low PM_{10} concentrations. Partly cloudy, warm (over +15°C), RH dropping to 45%.
13 May	No NPF	Undefined	Airmasses from south-west over England, Denmark and southern Sweden. SO_2 concentration somewhat elevated, high PM_{10} concentrations. Partly cloudy day, early morning sunny. Warm (over +15°C), RH dropping to 45%.
14 May	Possibility of NPF	Undefined	Airmasses from south-west over Denmark and southern Sweden. Somewhat elevated SO_2 and relatively low PM_{10} concentrations. Possibility for clear sky in the morning, more clouds in the afternoon. Warm day (over +15°C), RH dropping to 40%.

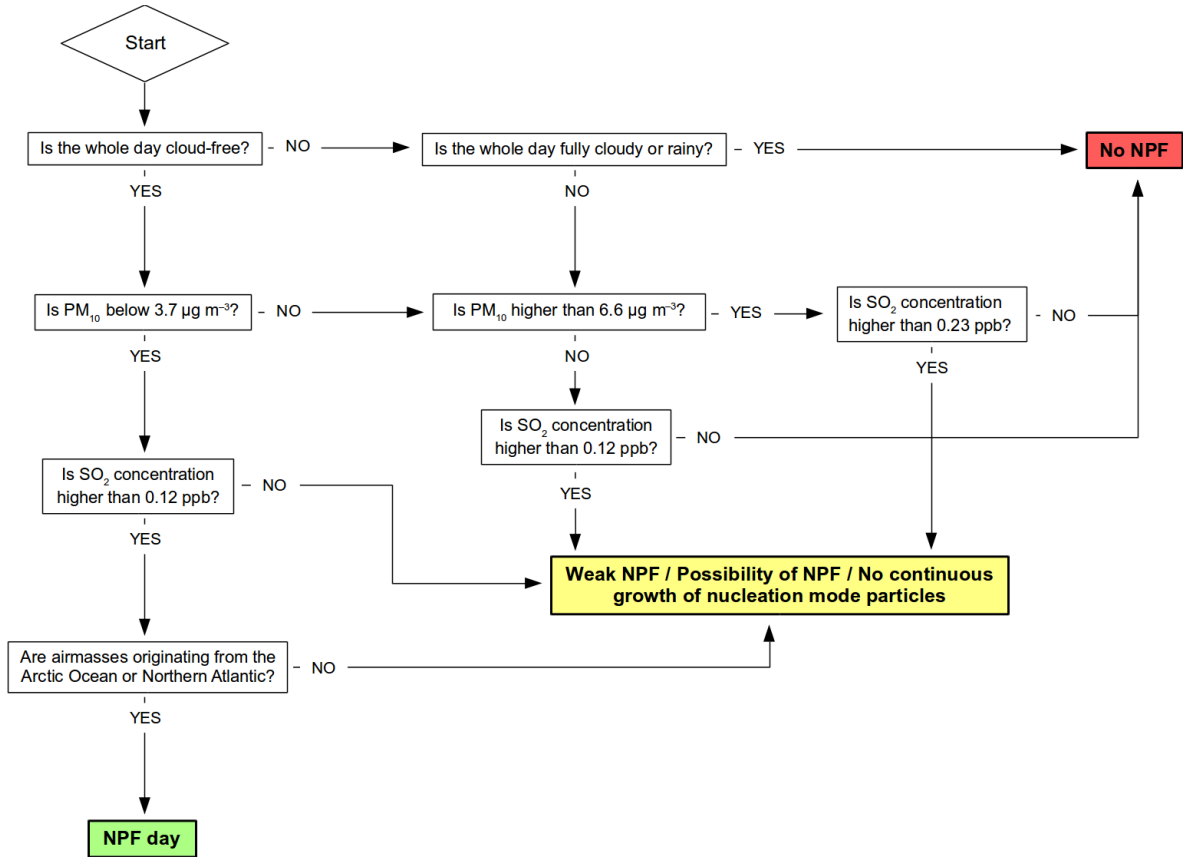
15 May	Possibility of NPF	NPF (class I)	Aimasses from south-west over England, Northern Germany and Southern Sweden. Somewhat elevated SO_2. Low PM_{10} concentrations. Partly cloudy early morning (possibility for clear sky), clear sky in the afternoon. Warm (over +15°C). RH dropping to 35% during the day.
16 May	NPF day	NPF (class II)	Airmasses from south-west over Northern Germany and Southern Sweden, circulating over Southern Finland. Low SO_2. Low PM_{10} concentrations. Clear sky in the morning, possibility of some clouds towards afternoon, warm (over +15°C). RH dropping to 35% during the day.
17 May	No continuous growth of nucleation mode particles	No NPF	Airmasses from south-west over England, Germany, and Southern Sweden, towards afternoon circulating over Northeast Russia. Low SO_2. Slightly elevated PM_{10} concentrations. Partly cloudy, very warm (over +20°C). RH dropping to 45% during the day.
18 May	No NPF	Undefined	Airmasses circulating over Finland and Northwest Russia. High SO_2, elevated PM_{10} concentrations. Cloudy, some rain, warm (over +15°C). RH dropping to 45% during the day.
19 May	No NPF	No NPF	Airmasses coming from east. Low SO_2 and PM_{10} concentrations. Cloudy and some rain, very warm (over +20°C). RH dropping to 50% during the day.
20 May	No NPF	Undefined	Airmasses coming from east. Low SO_2 and PM_{10} concentrations. Cloudy and some rain in the morning, very warm (over +20°C). RH > 90% during the day.
21 May	Possibility of NPF	Undefined	Airmasses coming from northeast. Elevated SO_2, low PM_{10} concentrations. Partly cloudy until afternoon (no continuous growth of nucleation mode particles), possibly clear skies in the evening. Very warm (over +20°C). RH dropping to 55% during the day.
22 May	Possibility of NPF	NPF (class II)	Airmasses coming from Arctic Ocean and circulating via northwest Russia. Elevated SO_2 and PM_{10} concentrations. Partly cloudy day, cloudier towards the afternoon (no continuous growth of nucleation mode) with a chance of rain. Warm (over +15°C). RH dropping to 45% during the day.
23 May	Possibility of NPF	NPF (class II)	Airmasses coming from Arctic Ocean and circulating over northwest Russia. Low SO_2 and PM_{10} concentrations. Partly cloudy in the morning, cloudy skies towards the evening (no continuous growth of nucleation mode).
24 May	No NPF	No NPF	Airmasses coming from Arctic Ocean and circulating via northwest Russia. Low SO_2 and PM_{10} concentrations. Cloudy day, chance of rain throughout the day.
25 May	NPF day	NPF (class II)	Airmasses coming from south-east and circulating over Central Finland. Low SO_2 and somewhat elevated PM_{10} concentrations. Sunny day with few clouds, very warm (over +20°C), RH dropping to 35% during the morning.
26 May	NPF day	NPF	Airmasses coming from east and circulating via Northern Finland to Hyytiälä. Low SO_2 and PM_{10} concentrations. Sunny day, some clouds in the afternoon, very warm (over +20°C), RH dropping to 35% during the morning.
27 May	No NPF	Undefined	Airmasses coming from east. High SO_2 and elevated PM_{10} concentrations. Cloudy and a chance of rain in the

			morning, partly cloudy in the afternoon.
28 May	No continuous growth of nucleation mode particles	No NPF	Airmasses coming from Arctic Ocean via northwest Russia to Hyytiälä. Low SO_2 and PM_{10} concentrations. Partly cloudy in the morning, clear skies towards afternoon. Very warm (over +20°C), RH dropping to 35% during the morning.
29 May	Possibility of NPF	Undefined	Airmasses coming from Arctic Ocean via northwest Russia to Hyytiälä. Low SO_2 and PM_{10} concentrations. Partly cloudy in the morning, clear skies towards the evening. Very warm (over +20°C).
30 May	Possibility of NPF	Undefined	Airmasses coming from Arctic Ocean via northwest Russia to Hyytiälä. Low SO_2 and low PM_{10} concentrations. Partly cloudy, with chances of rain in the evening. Very warm (over +20°C), RH dropping to 45% during the morning.
31 May	No NPF	Undefined	Airmasses coming from Arctic Ocean via northwest Russia to Hyytiälä. Low SO_2 and PM_{10} concentrations. Partly cloudy and chances of rainshowers during the day.
1 June	No NPF	No NPF	Airmasses coming from southeast. Low SO_2 and PM_{10} concentrations. Partly cloudy in the morning, more clouds towards afternoon, chances of light rain in the afternoon the day.
2 June	Possibility of NPF	Undefined	Airmasses coming from southeast. Elevated SO_2 and PM_{10} concentrations. Partly cloudy, very warm (over +20°C), RH dropping to 45% during the morning.
3 June	NPF day	Undefined	Airmasses coming from southeast. High SO_2 and elevated PM_{10} concentrations. Clear skies, some clouds in the afternoon, very warm (over +25°C), RH dropping to 35% during the morning.
4 June	Possibility of NPF	Undefined	Airmasses coming from southeast. Low SO_2 and PM_{10} concentrations. Partly cloudy morning, some rain in the afternoon, warm (over +20°C). RH dropping to 35% during the day.
5 June	Possibility of NPF	Undefined	Airmasses coming from southeast. Elevated SO_2 and low PM_{10} concentrations. Clear sky in the morning, partly cloudy starting from noon, possibility of rain in the evening, very warm (over +25°C). RH dropping to 25%.
6 June	Possibility of NPF	Undefined	Airmasses coming from east in the morning and turning to northwest in the afternoon. High SO_2 and elevated PM_{10} concentrations. Partly cloudy morning, rain and thunderstorms in the afternoon, warm (over +20°C).
7 June	No NPF	No NPF	Airmasses coming from northeast. Low SO_2 and elevated PM_{10} concentrations. Cloudy, rain and thunderstorms, moderate temperature (over +15°C).
8 June	NPF day	NPF (class II)	Airmasses coming from west, circulating over Sweden. Elevated SO_2 and low PM_{10} concentrations. Almost clear sky in the morning, partly cloudy in the afternoon, warm (over +20°C). RH dropping to 25% during the day.
9 June	No NPF	NPF (class II)	Airmasses coming from west. Low SO_2 and PM_{10} concentrations. Cloudy/partly cloudy, rain in the afternoon, moderate temperature (over +15°C). RH dropping to 45% during the day.

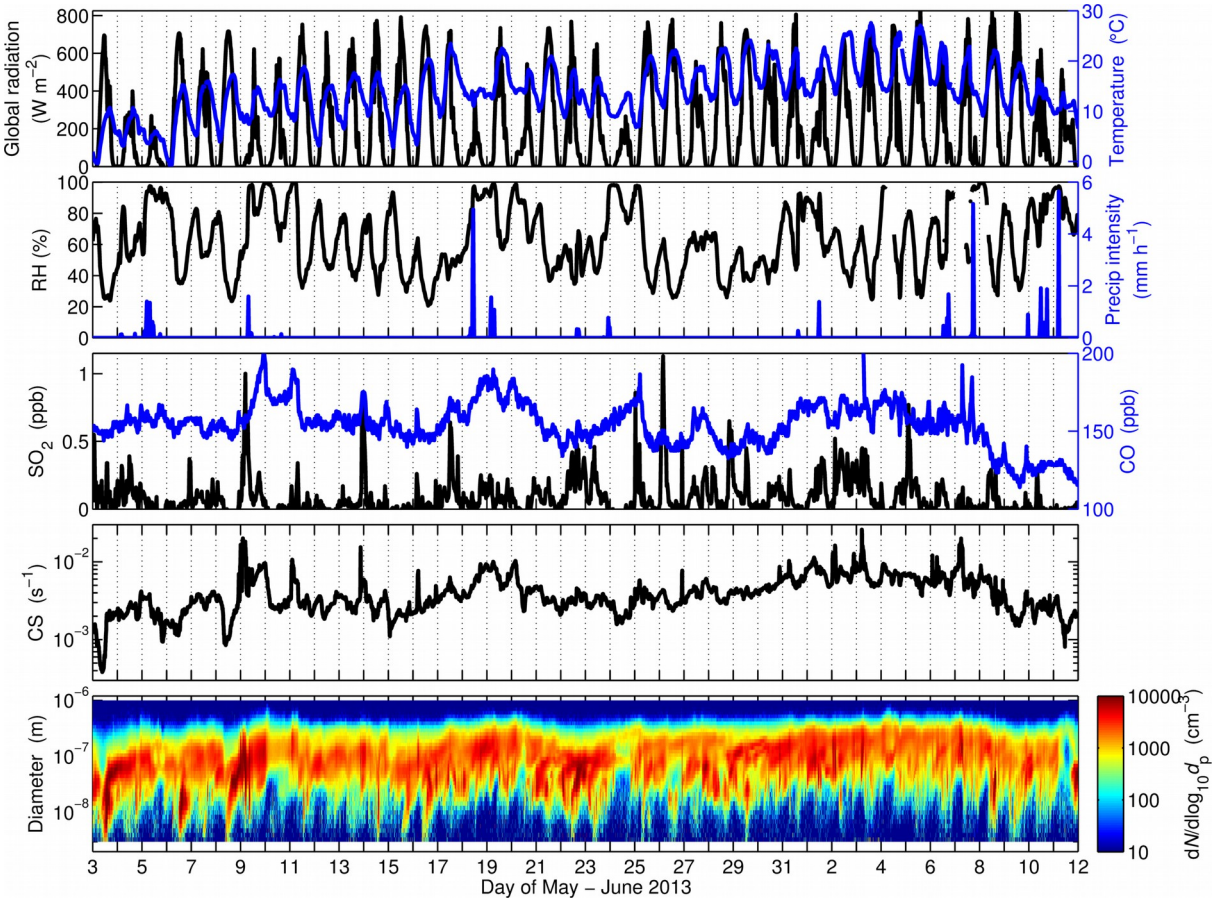
10 June	Possibility of NPF	Undefined	Airmasses coming from northwest. Low SO_2 and PM_{10} concentrations. Almost clear sky in the early morning, partly cloudy towards noon, chance of rain in the afternoon, moderate temperature (over +15°C).
11 June	No NPF	No NPF	Airmasses coming from northwest. Elevated SO_2 and PM_{10} concentrations. Partly cloudy, chance of rain, moderate temperature (over +15°C). RH dropping to 50% during the day.

Table 4. Comparison of the NPF classification based on DMPS data (rows), and the NPF forecasts (columns). On days marked with green color the forecasts were succesful in predicting whether NPF occurred in Hyytiälä or not, and on days marked with red the forecast was wrong according to observations. The days classified as undefined according to observations are left out of the comparison with forecasts.

	"NPF" forecast (8 days)	"Weak NPF / Possibility of NPF / No continuous growth" forecast (16 days)	"Non-NPF" forecast (16 days)
NPF day observed (11 days)	6	4	1
Undefined day observed (19 days)	2	10	7
Non-NPF day observed (10 days)	0	2	8



412 **Figure 1.** Flowchart of the decision making process for the NPF forecasts.



413 **Figure 2.** Overview of the meteorological parameters, trace gas concentrations and particle size
414 distributions during the campaign 3.5.–11.6.2013.

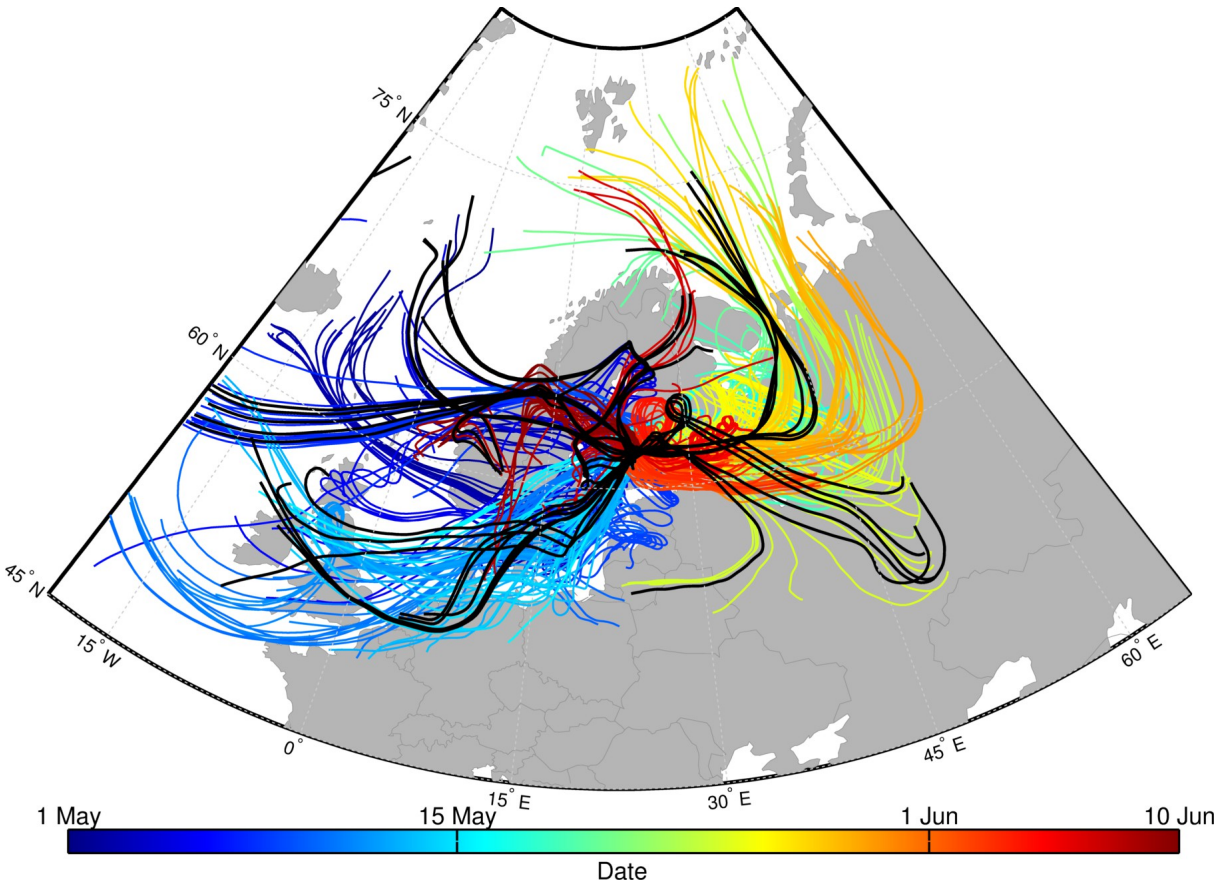
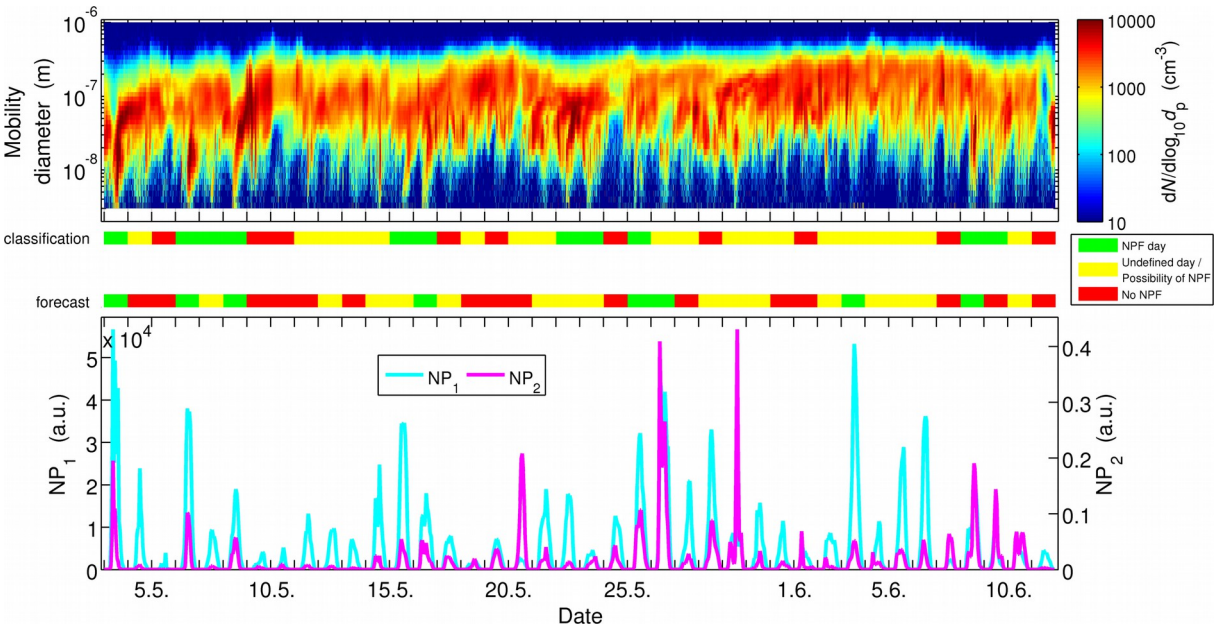


Figure 3. Airmass arrival trajectories to Hyytiälä 3.5.–11.6.2013 calculated using HYSPLIT model. The color indicates the arrival date and each trajectory represents airmass route during 96 hours before arrival. Airmass trajectories arriving on NPF days between 10–14 local time are marked with black lines.



419 **Figure 4.** Particle number concentration size distributions (top panel), and nucleation parameters NP_1
420 and NP_2 (bottom panel) during the campaign time 3.5.–11.6.2013. The colorbars between the panels
421 indicate the NPF forecast and classification: green for NPF days, yellow for weak or possible NPF
422 days, and red for non-NPF days (upper colorbar shows the NPF event classification based on the
423 DMPS data, and lower colorbar the forecast for each day).