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Atmospheric Chemistry and Physics Discussions

The seasonal cycle of ice-nucleating particles linked to the abundance of biogenic aerosol in boreal forests

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Abstract. Ice-nucleating particles (INPs) trigger the formation of cloud ice crystals in the atmosphere. Therefore, they strongly influence cloud microphysical and optical properties, as well as precipitation and the life cycle of clouds. Improving weather forecasting and climate projection requires an appropriate formulation of atmospheric INP concentrations. This remains challenging, as the global INP distribution and variability depend on a variety of aerosol types and sources, and neither their short-term variability nor their long-term seasonal cycles are well covered by continuous measurements. Here, we provide the first year-long set of observations with a pronounced INP seasonal cycle in a boreal forest environment. Besides the observed seasonal cycle in INP concentrations with a minimum in wintertime and maxima in early and late summer, we also provide indications for a seasonal variation in the prevalent INP type. We show that the seasonal dependency of INP concentrations and prevalent INP types is most likely driven by the abundance of biogenic aerosol. As current parameterizations do not reproduce this variability, we suggest a new parameterization approach, which considers the seasonal variation of INP concentrations. For this, we use the ambient air temperature as a proxy for the season which affects the source strength of biogenic emissions and by that the INP abundance over the boreal forest areas. Furthermore, we provide new INP parameterizations based on the Ice Nucleation Active Surface Site (INAS) approach, which specifically describes the ice nucleation activity of boreal aerosols particles prevalent in different seasons. Our results characterize the boreal forest as an important but variable INP source and provide new perspectives to describe these new findings in atmospheric models.

1 Introduction

Cloud processes are of particular importance for the evolution of weather and climate, as they regulate the global distribution of precipitation and influence Earth's radiative budget (Hoose and Möhler, 2012; Murray et al., 2012). Ice-nucleating particles (INPs) trigger the formation of ice crystals in clouds (Pruppacher and Klett, 2010), and therefore influence cloud microphysical and optical properties, as well as the lifetimes of mixed-phase and ice clouds (Hoose and Möhler, 2012). However, cloud processes remain highly uncertain in weather forecasting and climate projections (Boucher et al., 2013), also due to a lack of understanding of the critical parameters that predict atmospheric INP concentrations. The proportion of aerosol particles, which can act as INPs generally increases with decreasing temperature, as the free energy barrier to nucleation is reduced. Early parameterizations therefore linked INP or primary ice formation in clouds solely to temperature without any link to aerosol properties (Cooper, 1986; Fletcher, 1962; Meyers et al., 1992). More recent studies suggest a dependence of the INP number concentration on aerosol concentrations in specific size ranges (DeMott et al., 2010; Tobo et al., 2013), air mass origin

https://doi.org/10.5194/acp-2020-683
Preprint. Discussion started: 31 July 2020

Atmospheric Chemistry and Physics Discussions

(McCluskey et al., 2018) and rain events (Huffman et al., 2013; Iwata et al., 2019; Prenni et al., 2013). Others suggest aerosol type-specific descriptions (Harrison et al., 2019; Ullrich et al., 2017; Wilson et al., 2015), e.g. by linking the ice nucleation ability of the aerosol type to the aerosol surface area (Harrison et al., 2019; Ullrich et al., 2017). Due to the abundance of diverse atmospheric INP types distributed over the globe, it is not possible to find a direct dependence of INPs on a single parameter, which could be used to describe and predict primary ice formation processes. Long-range transport of aerosol particles as well as local sources and sinks influence INP populations and are potentially in flux due to both anthropogenic and natural influences like seasonal cycles. To examine the impact of seasonal changes on the INP population, continuous long-term measurements are necessary, but currently lacking. Only a few studies report atmospheric INP data collected in different seasons and resolve seasonal trends (Hartmann et al., 2019; Schrod et al., 2016; Tobo et al., 2019; Wex et al., 2019), which partly show enhanced INP concentrations in the summer months (Tobo et al., 2019; Wex et al., 2019). However, a comprehensive analysis of a continuous long-term record at one location for a full seasonal cycle is not yet available.

The boreal forest ecosystem is one interesting environment for such long-term observations. Boreal environments are characterized by meteorological conditions, vegetation and radiation budgets with strong seasonal trends and a clear annual cycle. Boreal forests cover 15 million square kilometres, representing one-third of all forested land (Tunved et al., 2006). They are generally far from anthropogenic and dust sources and are characterized by high biogenic aerosol concentrations (Kulmala et al., 2013; Spracklen et al., 2008; Tunved et al., 2006). The vegetation in boreal forests emits primary biological aerosol particles (PBAPs) and biogenic volatile organic compounds (BVOCs), which are prone to form secondary organic aerosol (SOA) (Spracklen et al., 2008), and collectively constitute 'biogenic aerosol'. PBAPs are directly derived from biological organisms, for example spores, pollen, fungi and leaf litter, and are distinct from SOA particles that form via new particle formation (NPF) and grow in size by multicomponent condensation (Ehn et al., 2014; Kulmala et al., 2013). BVOCs are integral as precursors for the NPF events, which are frequently observed in boreal forests (Kulmala et al., 1998, 2001). The frequency of NPF events shows a seasonal variability with a bimodal distribution of peak frequencies in spring and in autumn (Dall'Osto et al., 2018; Kulmala et al., 2001; Nieminen et al., 2014). A similar seasonal trend is observed in PBAP concentrations (Manninen et al., 2014; Schumacher et al., 2013). Several biogenic aerosol types have been shown to have atmospherically relevant ice-nucleating abilities (Augustin et al., 2013; Creamean et al., 2013; Hader et al., 2014; Möhler et al., 2007; Morris et al., 2004; O'Sullivan et al., 2015, 2018; Pratt et al., 2009; Schnell and Vali, 1973), especially at temperatures above -15°C (Christner et al., 2008; Murray et al., 2012). Although their contribution to the total global INP abundance is thought to be rather low (Hoose et al., 2010), biogenic aerosol may contribute substantially at regional scales where biological aerosol sources are important. For example, Tobo et al. (2013), Prenni et al. (2009) and O'Sullivan et al. (2018) have observed biogenic aerosol in the INP populations of the forested environments in Colorado, in the Amazon basin and in rural areas in Northern Europe.

2 Methods

We systematically measured INP concentrations at the Station for Measuring Ecosystem-Atmosphere Relations SMEARII (Hari and Kulmala, 2005), which is located in the Finnish boreal forest (61° 50' 50.685"N, 24° 17' 41.206"E, 181 m a.m.s.l.). As the nearest city (Tampere) is located about 60 km west-southwest from the station (Sogacheva et al., 2008) the prevalent aerosol population is mainly influenced by the forest. An extensive set of permanent measurements at the SMEARII contributes to a well-characterized picture of the site including meteorological as well as general aerosol-related information, which are available on the open research data portal AVAA (Junninen et al., 2009). The first comprehensive ice nucleation campaign at SMEARII called HyICE-2018 took place from February 2018 to June 2018. First results from HyICE-2018 are published in Paramonov et al. (2020), who show INP measurements with a continuous flow diffusion chamber (CFDC) during the first part of the HyICE-2018 campaign from 19 February 2018 to 2 April 2018. In this paper, we present the results of filter-based INP

https://doi.org/10.5194/acp-2020-683
Preprint. Discussion started: 31 July 2020

Atmospheric Chemistry and Physics Discussions

measurements, which provide a continuous record from 11 March 2018 to 13 May 2018 with a consistent time resolution of 24 hours. After these two intensive sampling months during the HyICE-2018 campaign, the INP measurements were continued until 31 May 2019 with a time resolution of mostly 48 hours or 72 hours, and only in a few cases with sample time intervals of up to 144 hours. By this, we obtained a continuous long-term record of INP temperature spectra from 11 March 2018 to 31 May 2019.

2.1 Aerosol filter sampling

Ambient aerosol particles were collected on 47 mm Whatman nuclepore track-etched polycarbonate membrane filters with a pore size of 0.2 µm. The filter sampling line and the filter holder are made of stainless steel and were installed in a cottage in the forest, with a rooftop PM10 inlet connected to the other sampling components installed indoors. A vacuum pump in combination with a critical orifice ensured a constant sampling flow rate of about 11 std l min^{-1}. Because the PM10 inlet provides a precise 10 µm cut-off size for a flow rate of about 16 std l min^{-1}, it is possible that larger particles have also been collected. However, the deviation of total aerosol number and surface concentration from the PM10 concentrations is < 1%, as it is shown in the Appendix Fig. A1a. We therefore refer to PM10 number concentration and PM10 surface concentration, when comparing the INP concentrations and calculating INAS (Ice Nucleation Surface Site) densities. Filters were pre-cleaned with 10% H_2O_2 and rinsed with deionized water that was passed through a 0.1 µm Whatman syringe filter, before being dried for use in sampling. After sampling, the filters were stored in sterile petri dishes, wrapped in aluminium foil and frozen until the sample were analysed for their INP content.

2.2 INSEKT

The INP content of the collected aerosol samples was quantified using the INSEKT (Ice Nucleation Spectrometer of the Karlsruhe Institute of Technology) method described in Schiebel (2017). The INSEKT is based on an ice spectrometer developed at the Colorado State University, which is described in Hill et al. (2016). INSEKT measures INP concentration as a function of activation temperature in the immersion freezing mode between about 247 K and 268 K. For the INP analysis, the collected aerosol particles are washed from the filter membranes by immersion in 8 ml of nanopure water, which was passed through a 0.1 µm Whatman syringe filter. The sample solution was spun on a rotator for approximately 20 min, and subsequently the aerosol suspension is diluted with 15- and 225-, or 10- and 100-fold volumes of filtered nanopure water. Small volumes of 50 µl are pipetted into two 96-well PCR plates. The wells are partitioned into different groups, including a group for the undiluted suspensions, the two diluted samples, and for the filtered nanopure water that serves to determine background freezing levels. Filter handling and suspension preparation always occurs in a clean flow cabinet using tweezers, which have been pre-cleaned in the same manner as the filter membranes. The filled PCR plates are then placed into the INSEKT instrument, which consists of two aluminium blocks, each with openings for holding a 96 well PCR plate. The aluminium blocks are connected to a chiller (LAUDA Proline RP 890), which pumps ethanol cooling liquid through the aluminium blocks at a constant cooling rate of 0.25 K min^{-1}. Eight evenly distributed temperature sensors measure the temperature distribution inside the blocks with a 2 Hz resolution. The aluminium blocks are placed in a PVC box insulated with 2 cm of Armaflex insulation material. The upper part of the PVC box is equipped with an antireflection and depolarized glass pane covering the PCR plates and preventing contamination from the ambient air. In order to avoid condensation on the glass the interior of the PVC box and the upper side of the glass pane are continuously flushed with particle free synthetic air at a constant flow rate of about 80 l h^{-1}. A camera with a 60 cm focal distance detects brightness changes in the small suspension volumes that are related to freezing during the cooling process. LabView software is used to control and monitor the cooling rate, temperature and brightness changes. Using this setup the frozen fraction of the small aerosol suspension volumes are determined as a function of temperature. From the fraction frozen, the INP concentration per standard litre of sampled air is calculated, binned on a 0.5 K grid and corrected by the background from the filtered nanopure water, using the procedure

https://doi.org/10.5194/acp-2020-683
Preprint. Discussion started: 31 July 2020


Atmospheric Chemistry and Physics Discussions

described in Vali (1971). Error is estimated by determination of 95% confidence intervals using the Wilson score interval (Wilson, 1927) in the form described by Agresti and Coull (1998). Data points with a ratio of upper to lower confidence interval higher than 8 are considered insignificant and neglected. A systematic error due to the preparation process and flow measurements is added, with 4% for the undiluted suspension, 5% for the first dilution step, 8% for the third dilution step and 11% for the fourth step, if needed. In addition, INP concentrations derived from handling blank filters, which were collected without flowing air through the membranes, are subtracted.

Heat treatment tests of the collected aerosol samples provide additional information about the heat sensitivity of the containing INPs and have been applied in various previous INP studies (Hill et al., 2016; O'Sullivan et al., 2018; Wilson et al., 2015). For these tests, a test tube filled with 2 ml of the aerosol suspension is kept in boiling water for approximately 20 min. Afterwards, the treated sample is analysed with the INSEKT in the same way as previously described.

2.3 Additional Instrumentation at SMEARII

For characterizing the sampled aerosol particles further, atmospheric aerosol size distributions were continuously measured with a Differential Mobility Particle Sizer (DMPS). The covered size range was 3 nm - 1000 nm in electrical equivalent diameter with 10 min time resolution (Aalto et al., 2001) with a closed loop flow arrangement (Jokinen and Mäkelä, 1997). The instrument was operated following guidelines from Aerosols, Trace Gases, and Clouds Research Infrastructure (ACTRIS) (Wiedensohler et al., 2012). The aerosol sample was taken from 8 m height inside the canopy through a total suspended particle (TSP) inlet. The super-micron aerosol size distribution was determined with a TSI Aerodynamic Particle Sizer (APS) model 3321 for the size range 0.5 - 10 µm in aerodynamic diameter. The sample was drawn through a vertical sampling line to avoid particle losses. The inlet is at a height of 6 m above the ground and consists of a total suspended particle inlet (Digitel Inc.). The inlet was heated to 40°C to prevent condensation and to ensure that fog droplets are evaporated and the RH remains below 40 %.

The intense measurement period during the HyICE-2018 campaign also provides additional aerosol instrumentation, like a long time-of-flight aerosol mass spectrometer (L-ToF-AMS) to measure the aerosol chemical composition and a wideband integrated bioaerosol sensor (WIBS) (Kaye et al., 2000; Savage et al., 2017) to derive information about biogenic fluorescent aerosol particles. The WIBS provides information about particle size, particle asymmetry and fluorescence emission intensity for particles in the size range 0.5 – 10 µm in diameter. Fluorescence excitation is achieved by two lasers with wavelengths of 280 nm and 360 nm. Two channels record the fluorescence emission in the ranges of 310 nm - 400 nm and 420 nm - 650 nm. A detailed description of the set-up and working principle of the L-Tof-AMS during the HyICE-2018 campaign can be found in Paramonov et al. (2020).

3 Results and Discussion

3.1 INP temperature spectra and time series

All INP temperature spectra measured from 11 March 2018 to 31 May 2019 are shown in Figure 1 in a monthly representation. The INP concentrations range from about 10^{-4} std l^{-1} to 10^{-2} std l^{-1} at the highest and from about 10^{0} std l^{-1} to 10^{2} std l^{-1} at the lowest temperatures. These concentration values fall within the range of INP concentrations measured during previous globally distributed field studies, which are summarized in Kanji et al., (2017). This indicates that primary ice formation in boreal forest areas is comparable to other regions on Earth, despite the lack of anthropogenic and dust sources. In our study, we observe both INP concentrations and spectral shape to be highly variable from day to day and to show clear seasonal trends. INP temperature spectra in March, December, January and February constitute the lowest of the entirety of INP temperature spectra, whereas the INP temperature spectra with highest INP concentrations are recorded in May and September.

https://doi.org/10.5194/acp-2020-683
Preprint. Discussion started: 31 July 2020

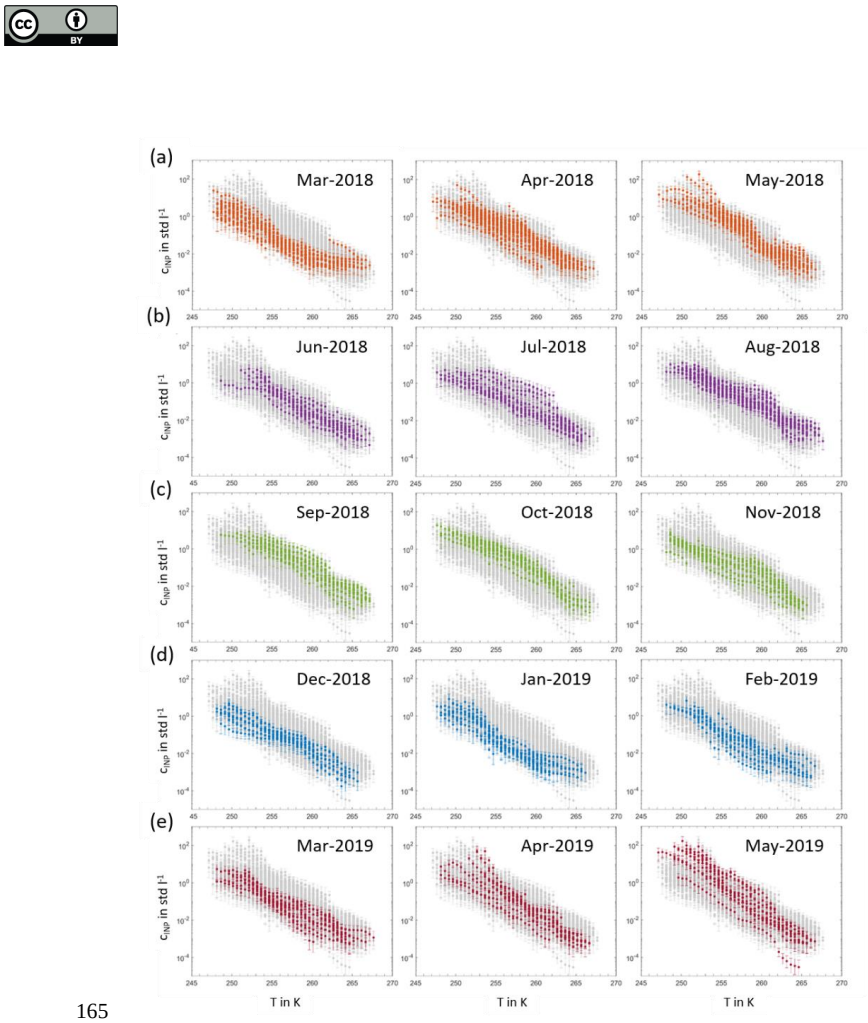

Figure 1: Monthly overview of INP temperature spectra. Each panel shows the entirety of INP temperature spectra measured from 11 March 2018 to 31 May 2019 (grey) with the spectra of the specific month highlighted in colour.

Figure 2a depicts the full time series of INP concentrations measured as a function of activation temperature with a time resolution between 24 h and 144 h. The colour contours represent the seasonal cycle of INP concentrations, which are lowest in wintertime from December to March and highest in the summer months, especially during May and September, as it was already observed in Fig. 1. An additional peak is found in the beginning of July. The INP concentrations in the middle temperature range around 257 K show the most distinct seasonal cycle. The variability of INP concentrations at the lower and upper end of the temperature range is less pronounced. Figure 2b shows the time series of INAS densities, which are calculated by normalizing the INP concentration measured by INSEKT with the aerosol surface concentration of atmospheric PM10 aerosol particles derived from DMPS and APS. The INAS densities show the similar seasonal trend and annual variability as the INP concentrations. For comparison, the time series of PM10 aerosol number concentrations and PM10 aerosol surface concentrations are shown in the Appendix Fig. A1b.

https://doi.org/10.5194/acp-2020-683
Preprint. Discussion started: 31 July 2020

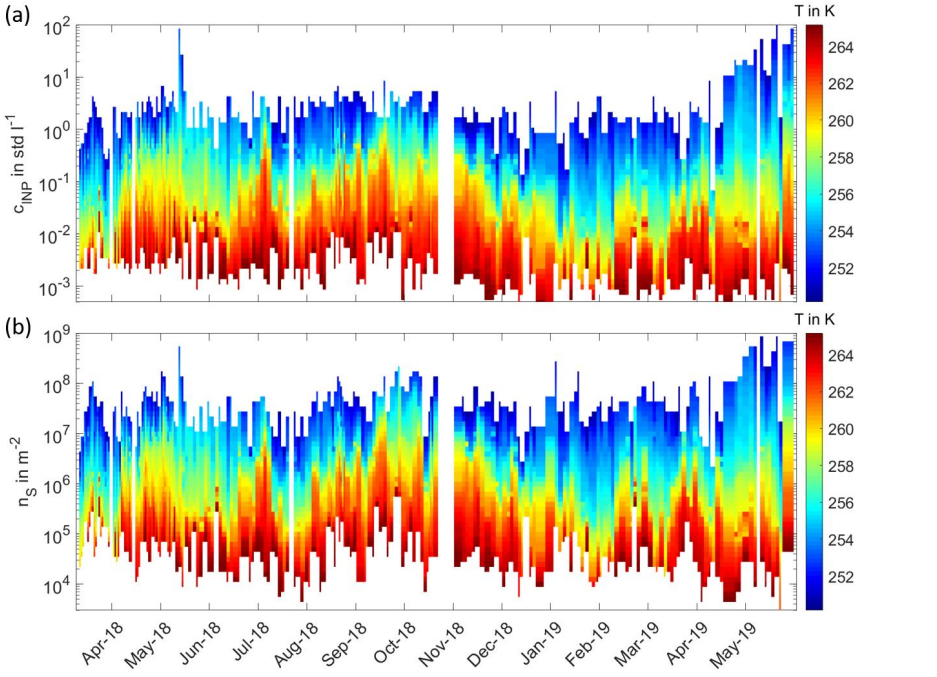

Figure 2: Long term record of INPs from 11 March 2018 to 31 May 2019 in the Finnish boreal forest. Panel (a) shows the time series of INP concentrations c_{INP} with a general temporal resolution between 24h and 144h. The colour code indicates the corresponding activation temperatures in K. In panel (b) the time series auf INAS densities n_S is displayed in the same manner as for the INP concentrations in panel (a).

3.2 Comparison to meteorology and aerosol properties

To investigate factors that influence the abundance of INP and might explain the daily and seasonal variability of INP concentrations, the INP time series are compared with other data sets like meteorological and aerosol data. As the seasonal trends and variability is most pronounced at activation temperatures around 257K, the INP time series at 257 K is used for this comparison. In Fig. 3a, the monthly averaged INP time series at 257 K is compared with the monthly fraction of NPF event days and snow coverage measured alongside the INP measurements at SMEARII, as well as with the averaged concentrations of pollen and other PBAP. The analysis of NPF events is based on permanent measurements at SMEARII and was provided by Simo Hakala, University of Helsinki. As there were no simultaneous direct measurements of pollen and other PBAP available for the period of our INP measurements, we compare to pollen and PBAP measured in 2003 and 2004 by Manninen et al. (2014) at SMEARII. The observed INP peak in spring coincides with the peak in pollen concentrations, whereas the peak in September is found to correlate with enhanced concentrations of other PBAP. Maxima in NPF event fractions are recorded in spring and in autumn, which has also been observed in many previous years back to 1996 (Dall'Osto et al., 2018; Kulmala et al., 2004; Nieminen et al., 2014). Snow-free periods are characterized by relatively high INP concentrations, whereas complete snow cover yields low concentrations.

Figure 3b shows the INP time series at 257 K during the intensive measurement period of the HyICE-2018 campaign from March to May 2018. After a period of rather constant INP concentrations in March, we observe a steady increase of INP concentrations in April, which comes along with the snowmelt period at SMEARII. After the snowmelt, INP concentrations are again on a rather constant but higher level. During the HyICE-2018 period, more comprehensive aerosol characterization was done, including measurements with a L-ToF-AMS and a WIBS. Both, the organic aerosol mass concentration measured by L-ToF-AMS and the number concentration of PM10 fluorescent particles measured by WIBS, show clear increases during the snowmelt period. Here, we define the number concentration of PM10 fluorescent particles as the number concentration of

https://doi.org/10.5194/acp-2020-683
Preprint. Discussion started: 31 July 2020

Atmospheric Chemistry and Physics Discussions

PM10 aerosol particles, whose fluorescence emission intensity produces a fluorescent signal in all three laser-channel combinations of WIBS. The time series of the number concentration of PM10 aerosol particles with a fluorescence signal in other laser-channel-combinations is shown in the Appendix in Fig. A2.

Besides a significant correlation of INP concentrations to the snow depth, the ambient air temperature measured in 4 m above ground level also showed a significant correlation. Figure 4a depicts the related time series, which clearly shows the INP time series at 257 K following the course of the ambient air temperature. Only for ambient air temperatures about > 15°C, the deviation of the two time series is higher. However, the general seasonal trend in the ambient air temperature is the same as we observed in the INP time series. Figure 4b shows a direct comparison of the INP time series with the measured snow depth. Periods of decreasing INP concentrations clearly overlap with snowmelt periods, whereas an increasing snow depth comes along with increasing INP concentrations.

In wintertime, complete snow cover seems to suppress biogenic particle emissions, resulting in comparably low INP concentrations. Such a correlation is also supported by the peaks of pollen and PBAP concentrations in snow-free periods in spring and in autumn, and by the increases of the organic aerosol mass concentration and fluorescent particle numbers observed in spring. Fluorescent particles are expected to be primarily of biological origin except for a few percent, which could arise from non-biological materials (Pöhlker et al., 2012; Savage et al., 2017). No day-by-day relation of NPF events and INP peaks are identified, but this is not unexpected given that particles formed during NPF events are initially smaller than 5 nm in diameter and events are more likely when condensation sinks are low (Dada et al., 2017). However, we consider the enhanced NPF event frequency as an indicator of generally higher biological activity in the forest. Enhanced biological activity means more biogenic INP emissions from the vegetation, which agrees with the seasonal dependencies from the previous findings. Any direct impact of NPF events on the boreal forest INP abundance remains uncertain and requires more investigation.

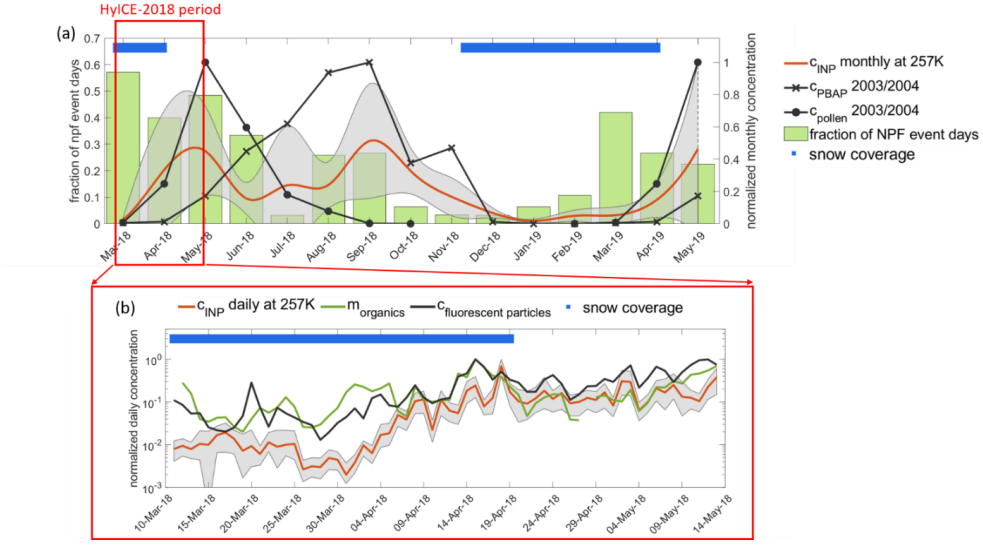

Figure 3: Factors co-varying with INP concentrations. In panel (a), monthly averaged INP concentrations at 257 K (red, standard deviation grey shading) are compared to the monthly fraction of NPF event days (green bars) from March 2018 to May 2019. Monthly averaged concentrations of PBAP and pollen (black dots and crosses) measured in 2003 and 2004 by Manninen et al. (2014) are additionally displayed. The panel (b) inset shows the HyICE-2018 period including a comparison of daily INP concentrations at 257 K (red, error bar area grey shading) to the mass concentration of organics (green) and to the concentration of fluorescent PM10 aerosol particles with a fluorescence signal in all three WIBS laser-channel-combinations (black). In both panels, the blue bar is indicative of snow coverage.

https://doi.org/10.5194/acp-2020-683
Preprint. Discussion started: 31 July 2020

Atmospheric Chemistry and Physics Discussions

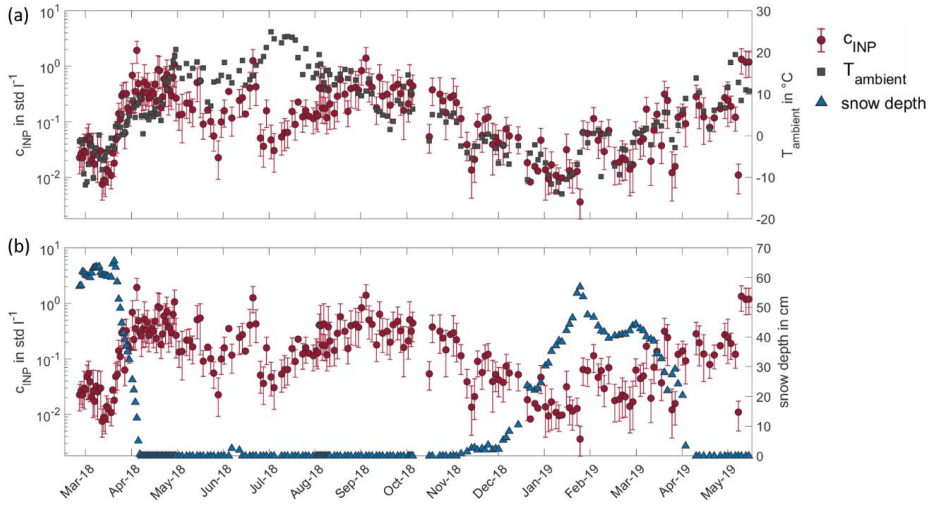

Figure 4: Time series of INP concentrations compared with the time series of ambient air temperature and snow depth. In panel (a), the time series of INP concentrations at 257 K (red dots) is plotted with the ambient air temperature measured at 4 m above ground level and averaged over the INSEKT filter sampling time intervals from 11 March 2018 to 31 May 2019 (grey squares). In panel (b), the INP concentrations at 257 K are replotted and compared to the measured snow depth, also averaged over the INP sampling intervals (blue trangles).

3.3 Heat treatment tests

The exemplary INP spectra in Fig. 5 demonstrate that the INP concentrations show an exponential trend with activation temperature (approximately linear shape of the log(c_{INP})-T-spectra) during wintertime, whereas summertime spectra show enhanced concentrations at around 260 K resulting in curvature in the spectra. After heating the INSEKT samples in boiling water for approximately 20 min, the resulting INP spectra are shifted towards lower concentrations by one to two orders of magnitude throughout the temperature range. However, the characteristic bulge in the summertime spectra is conserved or even more pronounced after the heat treatment. The observed shift of INP spectra after heat treatment reveals the presence of heat-labile INP types, which hints to particles of biogenic origin containing ice active proteinaceous material (Hill et al., 2016; Morris et al., 2004). The characteristic differences in the shapes of the INP spectra further indicate that different aerosol types dominate the INP populations in winter- and summertime. These differences are consistent for the observations in 2018 and in 2019, suggesting a systematic seasonal behaviour.

https://doi.org/10.5194/acp-2020-683
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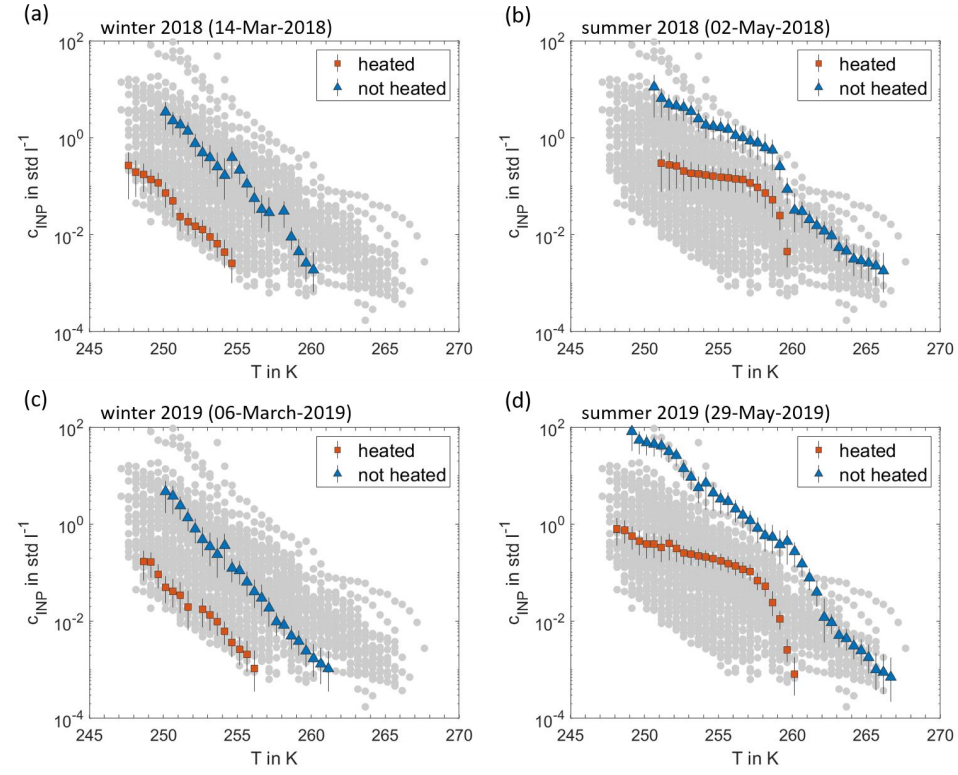

Figure 5: Effect of heat treatments. INP temperature spectra of the non-heated aerosol samples (blue triangles) are compared to the spectra of the heat-treated samples (red squares). Examples of two days in 2018 typical for winter and summer (panels (a) and (b)) and two days in 2019 also typical for winter and summer (panels (c) and (d)) are shown. In each panel, the grey dots display the entirety of non-heated and heat-treated samples.

3.4 Parameterizations

The observations presented in Fig. 1-5 indicate that the INP populations in boreal environments are dominated by biogenic emissions from the vegetation in the forest. We provide evidence that INP concentrations experience a seasonal cycle, which we link to seasonal trends in biogenic aerosol. The observational data we have presented poses new challenges for quantitative INP predictions, as it is essential to incorporate seasonal trends to achieve accurate descriptions. In Fig. 6a-d the measured INP concentrations are plotted versus the INP concentrations predicted by current parameterizations (DeMott et al., 2010; Tobo et al., 2013; Ullrich et al., 2017). DeMott et al. (2010) and Tobo et al. (2013) have developed temperature-dependent parameterizations that use the number concentration of aerosol particles with diameters > 0.5 µm (Fig. 6a and b). Tobo et al. (2013) provide an additional temperature-dependent formulation using the FBAP (fluorescent biological aerosol particle) concentration (Fig. 6c). Ullrich et al. (2017) use the measured aerosol surface area concentration to formulate a temperature-dependent parameterization of the INAS density of mineral dust (Fig. 6d). Among the selected parameterizations, Tobo et al. (2013) reproduce most of the data points by predicting 63% (Fig. 6b) and 80% (Fig. 6c) of the measurements to within one order of magnitude. The aerosol-specific parameterization of Ullrich et al. (2017) best matches the temperature trend, but overestimates the measured INP data, reproducing only 23% of the data points to within a factor of 10. This is not surprising, as the boreal forest aerosol is not dominated by mineral dust. The predictions of DeMott et al. (2010) and Tobo et al. (2013) overestimate INP concentrations especially in wintertime, as they do not include seasonal dependencies. These comparisons emphasize the need for a parameterization that accounts for seasonality.

https://doi.org/10.5194/acp-2020-683
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Atmospheric Chemistry and Physics Discussions

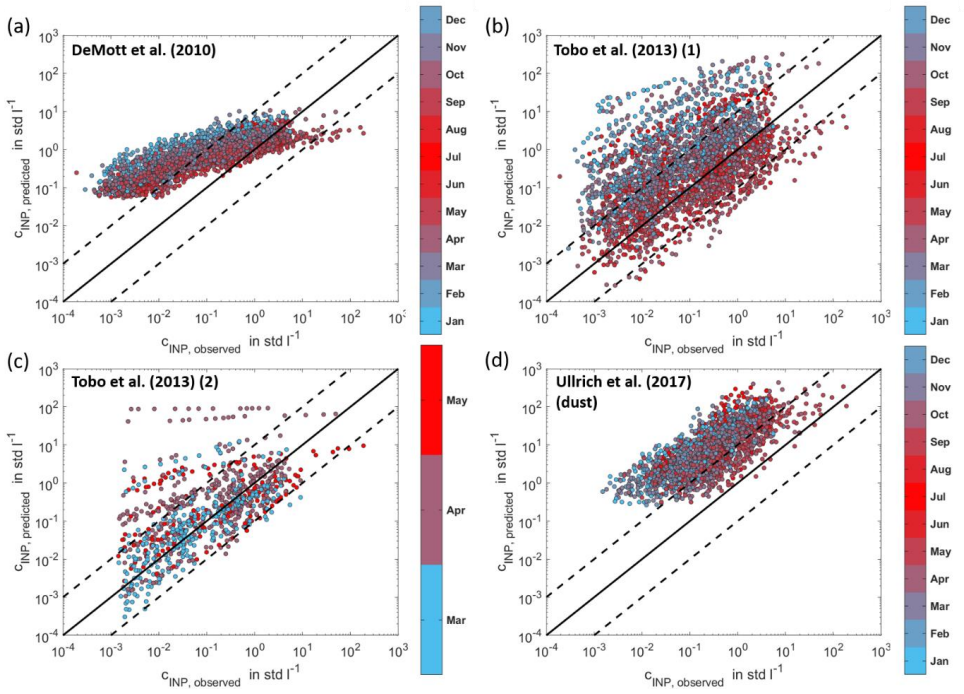

Figure 6: Comparisons to INP parameterizations. The measured INP concentrations are compared to INP concentrations predicted by parameterizations from DeMott et al. (2010) (a) and Tobo et al. (2013) (b) both using the number concentration of aerosol particles with diameter > 0.5 µm. Further, the measured concentrations are compared to the predicted concentrations by the second parameterization of Tobo et al.,(2013) using the number concentration of FBAP (c) and to the parameterization by Ullrich et al. (2017) for the temperature-dependent INAS density of mineral dust (d). In Tobo et al. (2013), FBAP concentrations were measured using an excitation wavelength of 355 nm and detecting the fluorescence emission in the range of 420 – 575 nm. The black lines show the 1-1-line (solid) and the 1-10-line and 10-1-line, respectively (dashed).

Given the need to represent seasonality, we suggest two new formulations for describing the seasonal variability of boreal forest INPs. Our first new non-aerosol specific approach assumes that the atmospheric INP concentration is predominantly determined by the boreal forest as the major INP emitting source with a magnitude that naturally changes with the seasons. Using ambient air temperature averaged over the aerosol filter sampling times as a proxy for the seasonal cycle, the observed INP concentration is quite well overlaid (see Fig. 4a). This clear relationship motivates us to use this parameter for formulating the new parameterization. The measured INP temperature spectra between 250 K and 265 K were used to create a least squares fit between the activation temperature T in K and the natural logarithm of INP concentrations c_{INP} in std l^{-1}. This describes the activation behaviour of INPs in temperature space. To account for seasonal dependencies in this formulation, the linear relation between the ambient air temperature T_{amb} in K and the natural logarithm of the time series of INP concentrations c_{INP} in std l^{-1} was used to establish a prefactor which shifts the parameterized INP temperature spectra to higher or lower INP concentrations depending on the ambient air temperature. The resulting parameterization is

$$c_{INP} = 0.1 \cdot exp\,(a1 \cdot T_{amb} + a2)\ \cdot exp\,(b1 \cdot T + b2) \quad (1)$$

with a1 = 0.074 ± 0.006, a2 = -18 ± 2, b1 = -0.504 ± 0.005, b2 = 127 ± 1 and with the activation temperature T and ambient air temperature T_{amb} in K. This new parameterization is able to reproduce 97.22% of the data to within a factor of 10. 88.21% and 49.79% are reproduced within a factor of 5 and 2, respectively. In Fig. 7a measured INP concentrations are compared to those predicted by the new parameterization underlining the good agreement. With this new approach, we do not directly describe the INP concentration in the atmosphere in a specific environment, as it was common in previous studies (DeMott et

https://doi.org/10.5194/acp-2020-683
Preprint. Discussion started: 31 July 2020

Atmospheric Chemistry and Physics Discussions

al., 2010; Tobo et al., 2013). Rather we have found a way to describe the boreal forest as an important INP emitting source. We suggest this formulation for application in atmospheric models to describe the source concentration of INPs abundant at ground level, which can then be further transported to cloud-relevant altitudes within the models. Here, it needs to be considered that INPs might undergo changes in their size distribution and chemical composition, when they are transported from their source to higher altitudes, which could affect their ice nucleation ability.

Our second formulation is aerosol-specific and describes the ice nucleation efficiency of boreal forest aerosol types using the INAS approach (Ullrich et al., 2017; Vali, 1971). As the seasonal cycle of INAS densities (see Fig. 2b) and the heat test results described earlier (see Fig. 5) indicate a seasonal change in INP types, we suggest specific parameterizations for different seasons. For the INAS density parameterization for boreal forest INPs, an exponential relation as suggested by Ullrich et al. (2017) is assumed and has the form

$$n_S = exp(a1 \cdot T + a2) \quad (2)$$

The INAS density n_S in m^{-2} is calculated by normalizing the measured INP concentration by the total surface area concentration derived from DMPS and APS size distribution data. We adjusted the parameters in Eq. (2) to our measured INAS densities considering the data set in three different periods corresponding to winter, summer and transition periods. The periods are defined by the measured snow depth s (s = 0 cm corresponds to summertime, s > 10 cm corresponds to wintertime and 0 < s ≤ 10 is the transition period.). The new fits yield a1 = -0.543 ± 0.007 and a2 = 154 ± 2 for summertime, a1 = -0.495 ± 0.008 and a2 = 141 ± 2 for wintertime and a1 = -0.49 ± 0.01 and a2 = 140 ± 3 for the transition period. In Fig. 7b the INP concentration calculated with these new parameterizations are compared to the measured concentrations. In summertime 92.57% (79.40%, 41.32%) of the data is reproduced by the INAS density fit within a factor of 10 (5, 2), in wintertime 97.32% (86.30%, 47.80%) and in the transition period 99.11% (95.56%, 56.89%) are reproduced.

4 Conclusions

This study provides a unique dataset of continuously recorded INP concentrations for more than one year at the SMEARII station located in the Finnish boreal forest. The observations illustrate that the boreal forest is an important source of biogenic INPs with resulting concentrations comparable to other environments (Kanji et al., 2017). We observe a clear seasonal cycle of INP concentrations and INP types in the boreal forest and conclude that this cycle is linked to the prevalence of biogenic aerosol particles. We suggest that these particles are primarily particles emitted by the forest vegetation, but are also correlated with the variable biogenic activities in the forest which appear to contribute both to INPs and to NPF. Current parameterizations do not represent the newly observed seasonal INP variability or concentrations. Thus, we suggest two new approaches for formulating and quantifying the annual cycle of INPs over boreal forest areas. The first considers the boreal forest as an aerosol emission source, including INPs, which appears to vary strongly with the seasonal changes. The second formulates season specific INAS parameterizations for boreal forest aerosol particles. The new formulations of both approaches reproduce almost all the data points of our long-term record of INP concentrations to within a factor of 10 and provide a basis for models to assess the global or regional importance of boreal forest INPs. This study shows that the ice nucleation activity in the atmosphere is highly variable depending on the surrounding conditions and reinforces the need to examine different characteristic regions on Earth on longer time scales with the aim to establish an overall picture of the global INP abundance and variability.

https://doi.org/10.5194/acp-2020-683
Preprint. Discussion started: 31 July 2020

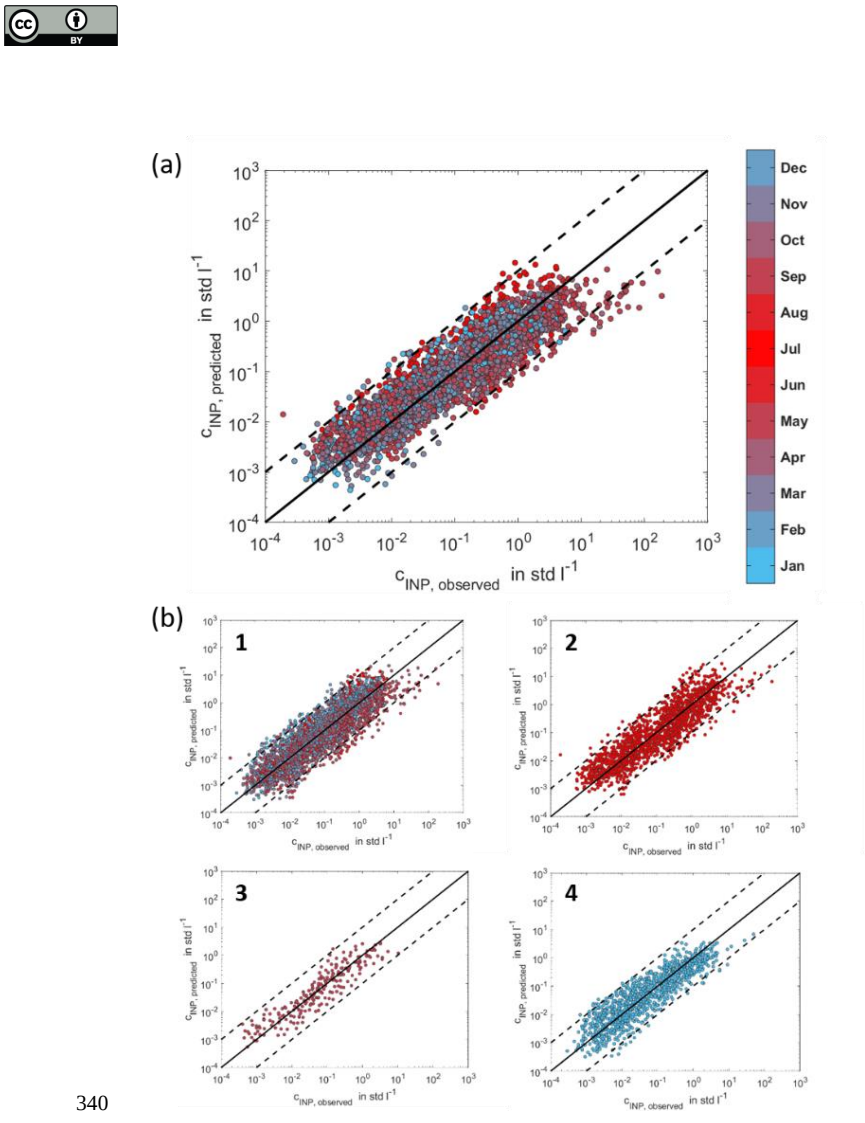

Figure 7: New INP parameterizations for boreal forest INPs. Panel (a) shows INP concentrations predicted by our new parametrization using ambient air temperature compared to measured INP concentrations colour-coded with the corresponding month. In panel (b), the measurements are compared to the predictions of the new INAS density parametrization. The four panels show the new INAS parameterization for the boreal forest INP over the whole year (1), only describing summertime INPs (2), the parameterization for the transition period (3) and the formulations for boreal INPs in wintertime (4).

https://doi.org/10.5194/acp-2020-683
Preprint. Discussion started: 31 July 2020

Atmospheric Chemistry and Physics Discussions

Appendix A

Figure A1. Comparison of PM10 aerosol population to total aerosol population. Panel (a) shows the percentage deviation of the number (blue dashed) and surface area (red solid) concentrations of the total aerosol from the number and surface area concentration only considering PM10 aerosol particles for the time period from 11 March 2018 to 31 May 2019. In panel (b), the time series of PM10 number concentrations (blue dashed) and of PM10 surface area concentration (red solid) are displayed for the same time period.

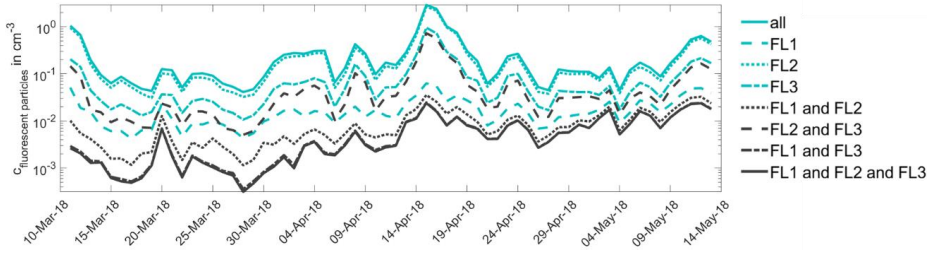

Figure A2: Time series of fluorescent particle number concentrations. The number concentrations of fluorescent particles measured by WIBS in different laser-channel-combinations are shown from 11 March 2018 to 13 May 2018 (FL1: λ_{ex} = 280 nm, λ_{em} = 310 – 400 nm; FL2: λ_{ex} = 280 nm, λ_{em} = 420 – 650 nm; FL3: λ_{ex} = 370 nm, λ_{em} = 420 – 650 nm).

Data availability. The measurement data used in this study is available via KITopen data repository under https://doi.org/10.5445/IR/1000120666 (Schneider et al., 2020).

Author contributions. JS wrote this manuscript, did the INP data analysis and part of the filter analysis. KH and OM both strongly supported the analysis and the writing. OM, MK, TP, JD and DM initiated and planned the HyICE-2018 campaign. JS, KH, BB, TS, NSU, FV, PB, PH, ZB, MPA, BJM, EST, DC, KK, YW, DM and JD conducted the measurements within the HyICE-2018 campaign and contributed to the data analysis and the interpretation. SH provided the analysed NPF event data. LH provided the analysed L-ToF-AMS data. JK, TP and TL contributed to the discussion and interpretation of the data and the manuscript writing.

Competing interests. The authors declare no competing interests.

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Preprint. Discussion started: 31 July 2020

Atmospheric Chemistry and Physics Discussions

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