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Temperature influence on the natural aerosol budget over boreal forests

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Abstract

We investigated the natural aerosol evolution from biogenic monoterpene emissions among northern boreal forest area as function of temperature, using long-term field measurements of aerosol size distributions and back trajectories at two stations

- SMEAR I and SMEAR II in Finland. Particles were formed via nucleation through the clean ocean to land transition of air parcels, and continuously grew to larger sizes during air mass transport. Both the travelling hour over land and temperature influences the evolution of the particle number size distribution and the average aerosol mass yield from BVOC emissions. Average concentrations of nucleation mode particles were higher at lower temperatures, whereas the opposite was true for accumulation mode
- ¹⁰ higher at lower temperatures, whereas the opposite was true for accumulation mode particles, thus more CCN may be formed at higher temperature. The overall apparent aerosol yield derived from the aerosol masses against accumulated monoterpene emissions ranges 13–37 % with a minor yet complicating temperature dependence.

1 Introduction

- Natural aerosol particles, including sea spray, mineral dust, and primary and secondary biogenic particles, are central to our understanding of the Earth's climate system (Carslaw et al., 2010). This is first of all because the atmospheric concentration levels and properties of natural aerosols need to be known in order to determine which fraction of the atmospheric aerosol is of anthropogenic origin. Separating natural and
- anthropogenic aerosols and their precursors is extremely challenging, since they may interact with each other in complicated ways (e.g., Dentener et al., 1996; Hoyle et al., 2011). Secondly, natural aerosol particles are part of various climate feedback loops, as their future concentration levels are expected to change with changing climate conditions (Arneth et al., 2010; Carslaw et al., 2010; Korhonen et al., 2010; Quinn and Bates, 2011; Kulmala et al., 2004, 2013).





Biogenic Secondary Organic Aerosol (BSOA) originating from biogenic emission presents a major fraction to the global submicron aerosol budget (Kanakidou et al., 2005; Goldstein and Galbally, 2007). The BSOA affects climate directly by scattering solar radiation and indirectly by aerosol-cloud interactions (e.g., Makkonen et al., 2012).

- ⁵ The latter effect is tied strongly with atmospheric cloud condensation nuclei (CCN) production, and thereby also with atmospheric new particle formation and growth (Riipinen et al., 2011; Yu, 2011; Kerminen et al., 2012; Makkonen et al., 2012). Since potential BSOA precursor emissions from terrestrial ecosystems increase strongly with increasing temperatures (Guenther et al., 2012), the natural CCN production associated with
- BSOA may be enhanced in warmer future climate, which would lead to a negative climate feedback mechanism (Kulmala et al., 2004). Recent observations and model simulations give support for the enhanced BSOA formation due to higher temperatures (Day and Pandis, 2011; Leaitch et al., 2011; Miyazaki et al., 2012; Makkonen et al., 2012; Paasonen et al., 2013), yet the exact relation between the ambient temperature,
- BSOA formation and natural CCN concentration levels is far from resolved. A major reason for this is that the temperature influences not only the BSOA precursor emissions, but also their atmospheric oxidation, resulting gas-particle partitioning, and potential heterogeneous reaction in the particle phase (Kroll and Seinfeld, 2008; Hallquist et al., 2009; Donahue et al., 2012; Riipinen et al., 2012).

Boreal forests are believed to be globally one of the most important sources of BSOA precursors (Spracklen et al., 2008; Tunved et al., 2006). Field measurements have shown that clean air masses arriving at the boreal forest zone experience a notable BSOA and CCN concentration increase during the summer part of the year (Tunved et al., 2006; Lihavainen et al., 2009), with subsequent implications for aerosol-cloud interactions (Spracklen et al., 2008). Tunved et al. (2008) investigated future BSOA production over the boreal forest area by extrapolating the emissions with a projected temperature increase, but did not consider the complicated nature of the various temperature impacts.





BSOA formation is often treated by considering the aerosol mass yield, namely the fraction of precursor VOC that forms aerosol mass. Biogenic monoterpenes are the most commonly assumed aerosol precursors, while other precursor vapours are also considered to play a role (Mentel et al., 2009). However, large-scale models simulating

⁵ SOA formation on the global scale often use only monoterpenes as the organic precursor, especially for BSOA, even though the fraction of monoterpenes producing aerosol is not yet well-constrained (e.g., Spracklen et al., 2008; Makkonen et al., 2012).

In this study, we will investigate the influence of temperature on BSOA formation and associated aerosol dynamics empirically using long-term aerosol observations and air mass back trajectory calculations similar to Tunved et al. (2006). With these tools, we aim (a) to study the number and size of aerosol particles as they travel over the

Nordic boreal forest area, (b) to obtain a quantitative estimate of the apparent aerosol yield assuming that monoterpenes are the main precursors of aerosol over the boreal forest, and (c) to find out whether a temperature signal is identifiable in the current ¹⁵ measurement data, and how the potential temperature effect is coupled with the air mass transport and time evolution of the aerosol number size distribution.

2 Measurements and methods

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We use a previously developed (Tunved et al., 2006) and tested (Tunved et al., 2008) method, focusing on the observed temperature impacts at five ascending temperature
²⁰ bins to the natural aerosol loading in this study. The basis setup of the approach is to investigate the characteristic evolution of aerosol population in air parcels through clean marine to continental transition over northern boreal forest area. We study the particle size distribution and mass evolution as a function of time spent over land, and then we link them to the estimated cumulative monoterpene emissions during the air
²⁵ transport (Tunved et al., 2008).





2.1 Measurement description

The dataset was based on the observations at two Finnish SMEAR (Station for Measuring Ecosystem–Atmosphere Relations Hari and Kulmala, 2005) stations: SMEAR I (67°46' N, 29°35' E, 400 m a.s.l.) in Värriö, and SMEAR II (61°51' N, 24°17' E,

⁵ 170 m a.s.l.) in Hyytiälä, Finland. A 13 yr dataset from 1998 to 2010 at the SMEAR I station and a 15 yr dataset between 1996 and 2010 at the SMEAR II station based on continuous Differential Mobility Particle Sizer (DMPS) measurements have been used together with backward trajectory data.

At SMEAR II, the DMPS system measured atmospheric particle size distributions from 3 to 1000 nm during the whole observation period; whereas at the SMEAR I station, particles from sizes 8–460 nm were measured before April 2003, and the system was then extended to the same particle size range as in SMEAR II (Dal Maso et al., 2007). We used particle sizes smaller than 450 nm in all calculations to keep the data consistency. Particle size spectrums were divided into three size modes: nucleation

¹⁵ mode with particle diameters (Dp) < 25 nm, Aitken mode (25 < Dp < 100 nm), and accumulation mode (Dp > 100 nm). Particle number concentrations at each mode were calculated for every one-hour period, averaged from 15 min DMPS measurements. Hourly particle volume concentrations were integrated from the DMPS size distributions assuming spherical particles. Also, the hourly total aerosol masses were calculated from the total aerosol volume concentrations, applying a constant aerosol density ($\rho = 1500 \text{ kgm}^{-3}$, Kostenidou et al., 2007).

2.2 Backward trajectories

The hourly 96 h backward trajectories arriving at 100 m a.g.l. over the SMEAR I and SMEAR II stations were calculated from the HYSPLIT4 model (Draxler and Hess, 1997)

²⁵ during the whole measurement period. Each individual travel point of the backward trajectories included parameters of air temperature at travelling height, mixing layer height, solar flux, precipitation, pressure and relative humidity. The travelling time over





ocean or land for each trajectory was determined from topographical data. The ground level temperature was calculated from the air temperature of each trajectory travel point at the actual height, using a lapse rate of 0.65 °C per 100 m (Finlayson-Pitts and Pitts, 2000).

- ⁵ Trajectories from April to September each year were used. The reasons for that were to guarantee significant monoterpene emissions, to exclude seasonal dependence of the aerosol population evolution at the receptor sites, and to minimize anthropogenic influence which was significant in winter. A more detailed explanation of the approach can be found from the supporting text by Tunved et al. (2006). Several criteria, sim-
- ¹⁰ ilar to those in Tunved et al. (2008), were used to select back trajectories: (1) wide transport sections of back trajectories were chosen covering mostly clean boreal forest area without anthropogenic influence from Southwest Europe, which include one 180° transport sector 180° S–0° N relative to Värriö, and travelled only above 60° N at the SMEAR I station, and one 180° transport sector 90° W–90° E relative to Hyytiälä at the
- SMEAR II station, seen in Fig. 1. (2) Each trajectory has to spend its first five hours over ocean and travels at least 90 % of time inside the transport sectors. (3) Assuming an imaginary air parcel travelling along the trajectory, its travelling height should be continuously below the mixing layer height, when it travels over land. Compared with previous studies, we narrowed down our trajectory selections by adding the third criterion with the reason that this criterion will be in line with the following calculation of
- ²⁰ terion with the reason that this criterion will be in line with the following calculation of monoterpene emissions.

The total numbers of trajectories from extractions were 6481 arriving at the SMEAR I station, and were 5086 arriving at the SMEAR II station. For each selected trajectory, the average trajectory temperature was calculated from the estimated ground level

temperature at all endpoints travelling over land along the whole trajectory. To investigate the aerosol characteristics under different temperature conditions, we classified all the selected trajectories into five groups by separating their corresponding average trajectory temperature equally into 5 bins from 0 to 20 °C for both stations in this study.





2.3 Calculation of monoterpene emissions

Monoterpene emissions are strongly temperature dependent, and the total flux of monoterpenes was derived from the following equation (Steinbrecher et al., 1999)

 $F = \varepsilon D \gamma$

⁵ where *F* is the total flux of monoterpenes from boreal forest in $\mu g m^{-2} h^{-1}$, ε is the emission potential for coniferous species, with value 1.5 $\mu g (g(dryweight))^{-1} h^{-1}$. *D* is the foliar biomass density, and here we used a foliar biomass density of 54 % pine and 46 % spruce as a function of latitude in Northern European boreal forest zone (Lindfors et al., 1999). γ is an environmental correction taking into account the temperature dependency of monoterpene emission rate, which can be calculated from $\gamma = \exp(\beta(T - T_s))$, where $\beta = 0.09 \,^{\circ}\text{C}^{-1}$, and $T_s = 303.15 \,\text{K}$. We omitted the leaf temperature to simplify the calculation and used the estimated ground level temperature (*T*) from the meteorological profiles along each trajectory. Assuming monoterpene emissions were well confined within the well-mixed boundary layer, the concentration in-15 crement of monoterpene emissions per unit volume during transport over land (*E*) can be derived by E = F/L, where *L* is the boundary layer height. Monoterpene emissions from the meteorological profiles along each trajectory and the accumulated monoterpene emissions from the monoterpene emissions per unit volume during transport over land (*E*) can be derived by E = F/L, where *L* is the boundary layer height. Monoterpene emis-

sions from the ocean were set to zero, and the accumulated monoterpene emissions were calculated for each trajectory spent over land, disregarding the short lifetime of monoterpenes in air.

- 20 3 Results and discussion
 - 3.1 Formation of secondary organic aerosol

3.1.1 Aerosol mass gain with transport over land

We binned all the trajectories based on their travelling hours over land (T_{hol}) before arriving at the receptor sites, and calculated the average aerosol mass for each hourly



(1)

bin. The average aerosol mass was given only if there were at least ten trajectories travelling the same T_{hol} . The averaged aerosol masses as function of T_{hol} are depicted both for the SMEAR II station and the SMEAR I station in Fig. 2. The aerosol mass increased linearly with T_{hol} at these two stations, and the correlation coefficients R^2 between these two quantities were 0.75 at the SMEAR I station and 0.90 at the SMEAR II station. The linear fitting slopes indicating the average mass gain per hour at the receptor stations were 0.02 µgm⁻³ per hour at the SMEAR I station and 0.04 µgm⁻³ per hour at the SMEAR II station are specified at the SMEAR II station.

43% higher than the previously found for both stations by Tunved et al. (2006) (see the fitting slope comparisons with dash lines in Fig. 2), but the average aerosol mass increment per hour at the SMEAR II station is close to the result reported by Tunved et al. (2008).

The average air temperatures of trajectories with the same T_{hol} were averaged again for each value of T_{hol} . Colors of markers indicate the averaged air temperature in Fig. 2. ¹⁵ The average temperatures of trajectories arriving at SMEAR II tended to be higher with longer travel times. This tendency was more pronounced at the SMEAR I station. This tendency can be attributed to heat transfer from the continental land mass to the air parcel, and a possibility of the weaker correlation of T_{hol} and temperature could be explained by the passage over the Baltic Sea that air masses arriving at SMEAR ²⁰ II needed to undergo. Very quick transit to SMEAR II resulted in low temperatures

 $(T_{hol} < 18 h)$. Overall, we could reproduce the findings from Tunved et al. (2008).

3.1.2 Accumulated monoterpene emissions

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We classified all the trajectories by grouping them according to their average trajectory temperature into five temperature bins from 0 to 20 °C for both stations. The accumulated monoterpene emissions along trajectories as function of T_{hol} were calculated in each temperature bin, and their values vs. T_{hol} are plotted in Fig. 3 for both stations. The figure demonstrates the effect of T_{hol} and air temperature on the MT emissions, and one can clearly see the dependence between temperature and MT emissions.



The accumulated MT emissions at the SMEAR II station were slightly higher than at the SMEAR I station, but were of the same order of magnitude at these two stations. In addition, the data in the temperature bin 16–20 °C at the SMEAR I station were very limited, so we have neglected this temperature bin in the following analysis.

5 3.2 Aerosol dynamics and CCN formation

As can be seen from the results in previous section, the observed aerosol at our receptor stations depended on the time when the air parcel has travelled over the boreal forest, in which the temperature during this transport affects the amount of the assumed aerosol precursor, biogenic monoterpenes, which have been emitted into the air parcel.

¹⁰ Therefore, we studied the evolution of the average observed aerosol size distribution as a function of travelled time over land, and we also looked into the temperature dependence of this evolution. Additionally, we investigated the number concentrations of aerosol particles in three different size ranges.

3.2.1 Evolution of particle size distributions

- ¹⁵ We computed the average aerosol size distribution for each T_{hol} bin. The resulting average size evolution distributions in relation to T_{hol} in the first four temperature bins are depicted both for the SMEAR II station (Fig. 4), and the SMEAR I station (Fig. 5). For each individual particle size evolution sub-figure, a normal distribution function was fitted at each T_{hol} for all the three particle size modes, and the maxima at each fit-
- ting mode were then calculated, seen from the black circles in all sub-figures. A linear fitting to the hour over land corresponding to the maxima of particle number concentrations at dominant Aitken or accumulation mode was made, which gives the growth rate of particles determined by different *T*_{hol} in unit nmh⁻¹. The average growth rates in all sub-figures are listed in Table 1, together with the initial and ending diameter of particles during the growth. In addition, the initial and ending particle diameters were extrapolated to the same *T*_{hol} at Hour-10 for comparison.





Figures 4 and 5 show that new aerosol particles were formed when clean air masses entered the northern boreal forest area, and that the newly-formed particles grew to sizes of 20 nm, on average, after 10 h of T_{hol} , regardless of temperature. By looking at extrapolated particle sizes at $T_{hol} = 10$ h in the first four temperature bins in Table 1, we can see that the particle growth from the nucleation mode to Aitken mode clearly accelerates with increasing temperatures.

Continuous growth of the particles to the Aitken and accumulation mode as function of T_{hol} can be seen from all sub-figures at both stations, but new particles formation is not apparent in the average size distributions after 20 h of T_{hol} . The average growth rate of the Aitken and accumulation mode particles was $0.43 \text{ nm}\text{h}^{-1}$ at the SMEAR II station, and $0.56 \text{ nm}\text{h}^{-1}$ at SMEAR I station, which is similar to the reported growth rate value by Tunved et al. (2006). The growth rate increased clearly with the increasing temperature at both stations: from $0.31 \text{ nm}\text{h}^{-1}$ in the temperature bin $0-4^{\circ}\text{C}$ to $1.58 \text{ nm}\text{h}^{-1}$ in the temperature bin $16-20^{\circ}\text{C}$ at the SMEAR II station and from

- ¹⁵ 0.37 nm h⁻¹ in the temperature bin 0–4 °C to 0.84 nm h⁻¹ in the temperature bin 12– 16 °C at the SMEAR I station. Several previous studies (Dal Maso et al., 2005, 2007; Yli-Juuti et al., 2011) have shown that growth rates were higher during warmer season and our results are in line with these observations. The increase in the growth rate implicates an increase in the concentration of condensable vapours, as expected from
- the temperature dependence of the emissions of precursor organics. Therefore, our studies are in line with the assumption that BVOCs are the main gas precursors for aerosol growth. The average growth rate we observed is lower than growth rates observed from point measurements at single observation stations for particle formation events (Dal Maso et al., 2005; Yli-Juuti et al., 2011); This may due to the fact that our growth rates were averaged from the whole dataset, while growth rates in other stud-

ies were data taken only from selected days with clear new particle formation events, which were likely to be days with above-average condensing vapour concentrations.

The condensation sink (CS) determines the speed of precursor molecules condensing onto the pre-existing aerosol particles, and is strongly dependent on the aerosol





size distribution (Lehtinen et al., 2003). The median CS values were calculated from the evolution of aerosol size distributions as function of T_{hol} in all temperature bins for both stations, and their values are also included in Table 1. The CS increased with temperature, and was higher at the SMEAR II station compared with the SMEAR I station $_{5}$ in the same temperature bins.

3.2.2 Particle number concentrations at three size modes vs. monoterpene emissions

Nucleation mode particles are the seeds of BSOA. To investigate the response of newly formed nucleation mode particles to temperature along trajectories, the nucleation mode particle number concentrations against total accumulated monoterpene emissions in these five temperature bins are depicted both for the SMEAR II station (Fig. 6a) and the SMEAR I station (Fig. 6b). In both figures, each data point includes the averaged nucleation mode particle number concentration and accumulated monoterpene emission from trajectories travelling the same T_{hol} in each temperature bin, and marker

- ¹⁵ colors demonstrate temperature bins in all sub-figures. The nucleation mode particle number concentrations show a negative correlation with the accumulated monoterpene emissions at both stations. This is connected to the occurrence of new particle formation after longer travel times; it is likely that high values of CS resulting from particle growth caused by the monoterpene emissions that have been oxidised and grown the
- 20 particles through air mass transport. The average number of nucleation mode particles is lower when the average trajectory temperature is higher. However, when comparing the number of nucleation mode particles at similar levels of cumulative monoterpene emissions, no clear dependence on the trajectory temperature could be observed. The available data do not support the idea that air temperature itself would influence the nu-
- cleation mode particle production process; rather, a possible temperature dependence of nucleation mode particle concentration can be interpreted as a reflection of the feedback between the increased CS and the particle growth rate. New particles are formed mainly when CS is low; when CS is high, condensing vapour contributes to aerosol





mass formation but new particles are not formed. Overall, the nucleation mode particle number concentration at the SMEAR II station was about 2–3 times higher than at the SMEAR I station.

- Particles growing continuously to the Aitken mode and accumulation mode are cli-⁵ matically important, as they can be activated as CCN, and further form cloud droplets (e.g., Kerminen et al., 2012). In order to examine temperature impacts on CCN-sized particles formed along trajectories, particle number concentrations in the Aitken and accumulation mode vs. total accumulated monoterpene emissions in these five temperature bins are illustrated in Fig. 6 as the last four sub-figures for both stations. The average particle number concentrations in the Aitken mode did not change clearly
- with increased temperature, but rather scattered around the average in each temperature bin. Particle number concentrations in the accumulation mode show clear positive dependence on monoterpene emissions and temperature at both stations, but the slopes between accumulation mode particle number concentrations and ac-
- ¹⁵ cumulated monoterpene emissions became smaller with the increased temperature, and projected maxima of the accumulation mode particle numbers can be seen ca. 1500 cm⁻³ for both stations. Nevertheless, it is certain that more particles have grown to accumulation mode at higher temperature along transport, thus more particles may be activated as CCN at higher temperature. When investigating the particle concen-
- trations in light of both monoterpene emissions and temperature, temperature showed an indirect effect. For a given monoterpene concentration, the concentrations of Aitken and accumulation mode particles varied within the same bounds, with higher temperatures leading to higher particle concentrations in the studied size ranges. The effect was more pronounced at the SMEAR I station. Our data do not show clear evidence of
- temperature directly affecting particle concentrations via, for example, lowering saturation vapour concentrations of condensing species. Rather, the average concentration of particles in the larger size ranges seems to be determined by the amount of BVOC emitted into the air mass, which is dependent on temperature.





3.2.3 Particle loss from particle size distribution evolution

BSOA formation is often discussed in terms of aerosol yield (Hallquist et al., 2009; Odum et al., 1996), which is generally defined as the fraction of oxidized precursor entering the aerosol phase via condensation or equilibrium partitioning (Pankow, 1994;

⁵ Hallquist et al., 2009). In order to estimate the aerosol yield assuming monoterpene as the sole precursor in this study, we needed an estimate for the amount of aerosol mass produced during transport to the receptor stations. To achieve this, we needed an estimate of aerosol losses during transport. The removal mechanism of atmospheric particles includes two main processes: dry deposition and wet deposition. In the following, we try to estimate the losses of particles due to deposition during transport using a simple lifetime calculation. Aerosol loss as function of sizes in each T_{hol} can be derived from the following discrete deposition equation,

$$\frac{\mathrm{d}N_{i,\mathrm{loss}}(t)}{\mathrm{d}t} = \gamma N_i(t)$$

where γ is the size dependent particle deposition rate (h⁻¹), $dN_{i,loss}$ is the number of deposited particles at size bin Dp_i, and dt = 1 (h) in this case. We used the reversed aerosol lifetime in mixing layer (0–1 km) as the atmospheric aerosol deposition rate in this study, given by Williams et al. (2002), and interpolated it to the size range of our DMPS measurements. The total deposition loss of aerosol mass ($dM_{loss}(t)$) in each T_{hol} can be then estimated by the particle size distributions of deposited particles, assum-10 ing spherical particles and using the same particle density. Therefore, the summation

- of particle mass losses from all hours of T_{hol} ($\sum_{T_{end}}^{T_{start}} dM_{loss}(t)$) together with the measured total particle mass at the receptor stations reveals the total amount of BSOA formed during air mass transport. The total aerosol masses with deposition loss corrected against the measured aerosol masses are plotted in Fig. 7 for both stations.
- It is noticeable that about one third of the particle mass was lost during the transport.
 Hence it is of important to include aerosol losses in the next calculation of aerosol yield.

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3.3 Temperature influence on the apparent aerosol yield

The formation process of BSOA generally includes gas-phase oxidation of BVOCs and other compounds at first, and gas-particle partitioning secondly (Hallquist et al., 2009). Monoterpenes are highly reactive to the major atmospheric oxidants (e.g. ozone, OH,

- and NO₃), so it can be approximated that nearly all monoterpenes have undergone primary atmospheric oxidation before arriving at the receptor stations when the imaginary air parcel is being travelled by air mass transport. A certain fraction of the low-volatility oxidation products of monoterpenes can condense to the formed particles through gasparticle partitioning, which increases aerosol sizes and mass. Therefore, a simplified
- ¹⁰ aerosol mass gain in the imaginary air parcel can be linked to the certain fraction of the condensable monoterpene oxidation products accumulating along the trajectory. This certain fraction value, i.e. the ratio between the total aerosol mass and the cumulative monoterpene emission was defined as the the apparent aerosol yield from oxidation of monoterpene emissions along trajectories in this study.
- ¹⁵ The total aerosol masses corrected with deposition loss and the accumulated monoterpene emissions were calculated from the grouped trajectories in each temperature bin, and their values are illustrated in Fig. 8. The apparent aerosol yields were then obtained from linear fittings between these two quantities. The values of apparent aerosol yield in each temperature bin are depicted in Fig. 9 for both stations. At
- the SMEAR II station, the apparent aerosol yield decreased with temperature from the highest 37.1 % in the temperature bin 0–4 °C to 14.4 % yield in the temperature bin 16–20 °C. The average aerosol yield at the SMEAR I station differed from that at the SMEAR II station, where it was 13.7 % and 12.7 % in the first two temperature bins, and slightly increased to 18.5 % and 18.4 % in the next two temperature bins.
- ²⁵ The variation in the estimated apparent aerosol yields is quite large as a function of the simple temperature parameter and site, which clearly shows that using a simple parameterization based on monoterpenes is not a sufficiently accurate method to model background BSOA formation. Two features are evident in the yield estimations: firstly,





aerosol mass production in cooler temperatures is more efficient for the SMEAR II station, and the efficiency of VOC transformation is higher at this more southern station. At higher average trajectory temperatures, the apparent aerosol yield from monoterpenes at both stations started to approach values close to each other.

- ⁵ The difference between the SMEAR I and SMEAR II stations at low temperatures is difficult to attribute directly to any single cause. It is known from previous studies (e.g., Dal Maso et al., 2008) that particle concentrations followed a different annual cycle at these two stations, especially in the lower particle size range. Analysis of a damped temperature sum parameter has shown that the damped temperature sum value close
- to zero leads to elevated new particle formation and elevated small particle concentrations (Dal Maso et al., 2009), which was speculated to be caused by spring recovery of trees leading to VOC emissions with a higher potential to nucleate. As it has been shown that the emitted VOC mixture has a clear effect on BSOA formation rate, also in particle mass (Mentel et al., 2009), it can be argued that BVOC emissions at low temperatures have a different chemical mixture. However, as such an effect would be
- expected to be visible also for the SMEAR I station, this explanation cannot be generalized.

Due to its closer proximity to athropogenic sources, air masses arriving at SMEAR II were more likely to be influenced by anthropogenic sources. While our selection of the time period was performed so that winter- and spring-time periods which showed elevated SO₂ concentrations (Lyubovtseva et al., 2005) have been screened out, it is possible that emissions from eg. combustion activities add influence to aerosol mass formation during air mass transport.

The apparent aerosol yield in our study is higher than the previous found by Tunved et al. (2008), mainly because particle mass losses have been taken into account, yet is still higher than the BSOA yield from some plant chamber experiments, e.g., 1.9– 17.7 % by Hao et al. (2011), and 5–10 % from Mentel et al. (2009). However, model studies on SOA yield have shown much higher variations and uncertainties, ranging from several percentages up to 70 % (Hallquist et al., 2009).





Our estimate for the aerosol yield over a boreal forest area is to our knowledge the first attempt to obtain an average SOA mass yield assuming monoterpenes as the SOA precursor. The results, which show a large variation both between sites and as a function of temperature, demonstrate that applying a simple parameterization for the mass yield will not describe SOA formation adequately. The estimated apparent mass yield from monoterpenes ranged between 13 % and 37 %, with higher yields obtained for the SMEAR II station which is more influenced by anthropogenic sources.

4 Summary and conclusion

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We extended earlier studies to evaluate the potential temperature impacts on the natural aerosol dynamics and mass yield from biogenic momoterpene emissions among northern boreal forest area, using long-term field measurements of aerosol size distributions and back trajectories at two Finnish SMEAR stations. We saw that particles were formed via nucleation when the clean ocean air mass enters the northern boreal forest area. The newly formed particles continuously grew to the Aitken mode and fur-

ther to the accumulation mode during air mass transport, and the nucleation process ceased after certain travelling hours over land.

Our results suggest that the "averaged air parcel history" method can be used to obtain valuable information on the development of aerosol over boreal forest area. For the first time, we have shown that two main factors including the travelling hour over land and air temperature were dedicated to this natural aerosol evolution.

The nucleation mode particle number concentrations were higher at lower temperatures, but particle number concentrations in the accumulation mode slowly increased at warmer temperatures, suggesting more CCN could be formed from rising temperature. No clear evidence of temperature directly affecting particle production process can be observed, mostly that particle number concentrations were mainly determined by the

observed, mostly that particle number concentrations were mainly determined by th amount of BVOC emitted into the air mass, which was dependent on temperature.





The apparent aerosol yield derived from the aerosol masses against accumulated monoterpene emissions along trajectories was of the order of 13–37 % with a minor, yet opposite, temperature dependence between these two stations. Overall, our study suggests that the temperature dependence of BSOA production should be treated with ⁵ caution, and a simple parameterization should be especially avoided in predicting the aerosol production potential from rising temperature in future. Further investigation on the detailed processes of BSOA yield as function of temperature would be necessary to reveal the actual aerosol distribution and its climate effects.

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0–4	4–8	8–12	12–16	16–20	All
18.3	24.7	29.5	46.6	80.1	25.1
17.4	23.0	25.7	43.4	12.1	24.7
13	14	16	16	53	11
40.9	55.3	77.5	86.9	130.6	59.5
87	85	91	91	85	91
0.31	0.43	0.64	0.54	1.58	0.43
1.37×10^{-3}	1.44×10^{-3}	2.30×10^{-3}	2.88×10^{-3}	3.61×10^{-3}	1.82×10^{-3}
0–4	4–8	8-12	12-16	16-20	A II
		0 12	12 10	10 20	
19.6	28.0	32.7	55.0	-	24.3
19.6 20.0	28.0 27.6	32.7 30.8	55.0 39.9		24.3 23.7
19.6 20.0 9	28.0 27.6 11	32.7 30.8 13	55.0 39.9 28		24.3 23.7 11
19.6 20.0 9 48.8	28.0 27.6 11 60.9	32.7 30.8 13 82.8	55.0 39.9 28 107.3		24.3 23.7 11 69.2
19.6 20.0 9 48.8 88	28.0 27.6 11 60.9 92	32.7 30.8 13 82.8 90	55.0 39.9 28 107.3 90	- - - - -	24.3 23.7 11 69.2 91
19.6 20.0 9 48.8 88 0.37	28.0 27.6 11 60.9 92 0.41	32.7 30.8 13 82.8 90 0.65	55.0 39.9 28 107.3 90 0.84		24.3 23.7 11 69.2 91 0.56
	$\begin{array}{c} 0-4 \\ 18.3 \\ 17.4 \\ 13 \\ 40.9 \\ 87 \\ 0.31 \\ 1.37 \times 10^{-3} \end{array}$	$\begin{array}{c ccc} 0-4 & 4-8 \\ 18.3 & 24.7 \\ 17.4 & 23.0 \\ 13 & 14 \\ 40.9 & 55.3 \\ 87 & 85 \\ 0.31 & 0.43 \\ 1.37 \times 10^{-3} & 1.44 \times 10^{-3} \end{array}$	$0-4$ $4-8$ $8-12$ 18.3 24.7 29.5 17.4 23.0 25.7 13 14 16 40.9 55.3 77.5 87 85 91 0.31 0.43 0.64 1.37×10^{-3} 1.44×10^{-3} 2.30×10^{-3}	$0-4$ $4-8$ $8-12$ $12-16$ 18.3 24.7 29.5 46.6 17.4 23.0 25.7 43.4 13 14 16 16 40.9 55.3 77.5 86.9 87 85 91 91 0.31 0.43 0.64 0.54 1.37×10^{-3} 1.44×10^{-3} 2.30×10^{-3} 2.88×10^{-3}	$0-4$ $4-8$ $8-12$ $12-16$ $16-20$ 18.3 24.7 29.5 46.6 80.1 17.4 23.0 25.7 43.4 12.1 13 14 16 16 53 40.9 55.3 77.5 86.9 130.6 87 85 91 91 85 0.31 0.43 0.64 0.54 1.58 1.37×10^{-3} 1.44×10^{-3} 2.30×10^{-3} 2.88×10^{-3} 3.61×10^{-3}

Table 1. Growth rate fitting parameters and condensation sink values in the five temperature bins as well as from the whole dataset both at the SMEAR I and the SMEAR II stations.



Discussion Paper

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Fig. 1. The transport sectors of back trajectories at the two Finnish SMEAR stations. Area above the red line is the travelling zone of selected trajectories arriving at the SMEAR II station. The blue sector covers the transport area of trajectories arriving at the SMEAR I station. At both stations, each trajectory should spend at least 90% of travelling time inside the transport sectors.



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Fig. 2. Average aerosol mass (particle size smaller than 450 nm) vs. travelling hour over land of trajectories (a) at the SMEAR II station, and (b) at the SMEAR I station. Solid lines represent linear fits from data. Fitting slopes and their intercepts are written in the equations, together with their coefficients of determination (R^2). Marker colors indicate the averaged temperature at each hour over land. The dash lines are fitting slopes reported from Tunved et al. (2006) at these two stations.





Fig. 3. Accumulated total monoterpene emissions as function of hour over land **(a)** at the SMEAR II station, and **(b)** at the SMEAR I station. The marker colors of legends represent the same binned temperature in both sub-figures.









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Fig. 5. Aerosol particle size distributions as function of hour over land in the first four temperature bins at the SMEAR I station, Värriö. The black circles are the maxima of the normal size distribution function at each hour, and black line depicts the mode fitting.





Fig. 6. Nucleation mode particle number concentrations vs. total monoterpene emissions among the five temperature bins (a) at the SMEAR II station, and (b) at the SMEAR I station. Aitken mode particle number concentrations vs. total monoterpene emissions (c) at the SMEAR II station, and (d) at the SMEAR I station. Accumulation mode particle number concentrations vs. total monoterpene emissions (e) at the SMEAR II station, and (d) at the SMEAR I station. The marker colors of legends represent the same binned temperatures in all sub-figures.







Fig. 7. Total aerosol masses with deposition loss corrected vs. aerosol masses "observed" at **(a)** the SMEAR II station, and **(b)** at the SMEAR I station. The marker colors of legends represent the same binned temperatures. The dash line indicates 1 : 1 ratio, and the solid line is the 1.5 : 1 ratio.





Fig. 8. Total aerosol masses with deposition loss corrected vs. total monoterpene emissions among the five temperature bins (a) at the SMEAR II station, and (b) at the SMEAR I station. The marker colors of legends represent the binned temperatures. Solid lines depict fitting slopes between these two quantities at each temperature bin.





Fig. 9. Estimated aerosol yield (i.e. the direct fitting slope between aerosol mass and total monoterpene emission) in five temperature bins both at the SMEAR I station (blue bars) and the SMEAR II station (red bars). The black bars indicate 95% confidence intervals from the fitting slopes at both stations.

