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TELLUS

The natural aerosol over Northern Europe and its relation to anthropogenic emissions—implications of important climate feedbacks

By P. TUNVED[1]*, J. STRÖM[1], M. KULMALA[2], V.-M. KERMINEN[2], M. DAL MASO[2], B. SVENNINGSON[3], C. LUNDER[4] and H.-C. HANSSON[1], [1]*Department of applied environmental science (ITM), Air Pollution laboratory, Frescativägen 54, SE-106 91 Stockholm, Sweden;* [2]*University of Helsinki, Gustav Hällströmin katu 2, FIN-00014, Helsinki, Finland;* [3]*Lund University, Division of Nuclear Physics, SE-22100 Lund, Sweden;* [4]*Norwegian Institute for Air Research (NILU), Instituttveien 18, P.O. Box 100, NO-2027 Kjeller, Norway*

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ABSTRACT

We use a recently developed parametrization to estimate the regional particle field in the summer time troposphere over Scandinavia that would result if the forest were the only source of particles. The calculated field is compared with available observational data. It is concluded that the needle leaf forest above 58°N alone is capable of producing aerosol mass concentrations corresponding to 12–50% of today's values in the boundary layer over Scandinavia. We also demonstrate that the forest itself could produce up to 200 CCN per cubic centimetre on average over Scandinavia and further show that an increase in temperature by 5.8 °C compared to today's average temperature could increase this CCN population by 40%. The study shows that we are able to approximate the natural aerosol field resulting from biogenic emissions over the boreal forest in the northern hemispheric region. This information provide an important contribution in the evaluation of the climate effect caused by anthropogenic emissions of particles over the forest and also opens the possibility to better address the climate feedbacks believed to be associated with the boreal region.

1. Introduction

Aerosols in the atmosphere are likely to have a large impact on climate due to both the indirect effect via the change of the microphysical properties of clouds and the direct scattering of incoming solar radiation (Twomey, 1974; Anderson et al., 2003). The radiation forcing resulting from aerosols (negative) and emission of greenhouse gases (positive) in the atmosphere is comparable in magnitude. Changes in particle emissions could therefore have a substantial impact on earth's climate. Recent findings suggest that decreasing anthropogenic particle emissions could motivate a substantial upgrade of the future global warming, from currently estimated maximum of 6.4° (IPCC, 2007) to 8° until 2100 (Andreae et al., 2005). Still, both particle sources and particle disperison in the atmosphere are poorly quantified. Although progress has been made in quantifying the climate effect of the atmospheric particle loadings (IPCC, 2007), it is still not possible to make precise estimates of the impact of these particles on the radiation balance of the atmosphere. The organic carbon containing fraction of the atmospheric aerosol has a central role in the atmosphere. Carbon compounds make up 10–90% of the fine particle mass, depending on location and time of year. The single most important source of Volatile Organic Compounds (VOC's) is natural sources. Natural sources contribute with 85% of the total amount of VOC's emitted to the atmosphere (Kiehl and Rodhe, 1995).

The most important natural organic carbon emitter in the northern hemisphere is the boreal forest. Consisting of mainly needle leaf trees, such as pine and spruce, the boreal region emits large quantities of terpenoids to the atmosphere. For the Scandinavian region, it has been shown that natural biogenic emissions of monoterpenes ($C_{10}H_{16}$) dominate VOC concentrations (Simpson et al., 1999; Tarvainen et al., 2007). Monoterpene emissions from pine and spruce are mainly controlled by temperature, and pool dependent emissions are exponentially dependent on temperature (Guenther et al., 1995). The lifetime of monoterpenes in the mid-latitude summer atmosphere is at

*Corresponding author.
e-mail: peter.tunved@itm.su.se
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most a couple of hours (Hakola et al. 2003). Once emitted, they are rapidly oxidised by common hydroxyl radicals, nitrate radicals and ozone in the atmosphere, resulting in numerous different oxygenated organics such as mono- and di-carboxylic acids (Kavouras et al., 1998). There is increasing evidence that several of these compounds partake in gas-to-particle partitioning (e.g. Kavouras et al., 1998).

Over Scandinavia, Tunved et al. (2006a, b) have shown both qualitatively and quantitatively that the monoterpene emissions from the boreal forest at high latitudes in Europe potentially contribute to particle generation with an apparent yield in the range of approximately 5–10%. Oxidation products from monoterpenes have been identified in particles over the forest (Cavalli et al., 2006). Several investigations of new particle formation events during the biologically active seasons indicate that biogenic aerosol precursor gases are necessary to support the observed mass increase during particle formation events (Kulmala et al., 1998; Mäkelä et al., 1997). Together, these results indicate that biogenic emissions (e.g. monoterpenes) significantly contribute to the evolution of the aerosol number size distribution during transport of clean marine air over northern parts of Fenno-Scandinavia. The biogenic emissions of organic compounds are not regarded to directly contribute to new particle formation, but instead provide the condensable species required for the freshly formed particles to grow. Laboratory experiments have also indicated that monoterpene oxidation result in particles with good CCN capability (e.g. Hartz et al., 2005). Thus, the results so far imply that the boreal region could contribute to both direct and indirect climate forcing over the northern hemisphere. Furthermore, increasing temperatures enhance, at least initially, pool dependent terpeneoid emission rates since the emission rate of these compounds is exponentially dependent on temperature. Monoterpene emissions likely contribute to generation of particle number over the boreal region via condensation growth of freshly formed particles. It is therefore possible that increasing monoterpene emissions could alter the amount of potential CCN's over the forest by changing size and abundance of particles. Therefore, the forest could provide additional cooling in times of a warming climate. This feedback mechanism could also be even more complex due to elevated CO_2 concentrations (see e.g. Kulmala et al., 2004). Like other natural systems, however, the boreal region is prone to be influenced by climate change. The boreal region may play an important role in the radiative balance of the atmosphere. Retreat of boreal forest in some areas and expansion of boreal forest in others could be the result of global warming. The forest may act as a sink of carbon dioxide thereby counteracting global warming, but furthermore, changes in the ecosystem may lower the earth's albedo, and thereby providing a positive feedback to global warming. Studies by Betts (2000) suggest that forestation in boreal regions may even supersede the negative forcing caused by carbon dioxide sequestration by boreal forests. Changes in the boreal ecosystem will likely also result in changed emission patterns of biogenic volatile organic compounds (BVOC), and therefore possibly also the particle production potential of the forest. In turn, this could impact the radiation budget of the northern hemisphere. Lathière et al. (2005) showed, using a coupled emission parametrizations and global dynamic vegetation model, that the net effect from temperature increase, elevated CO_2 concentrations and changes in ecosystems would be a 51% increase in monoterpene emissions in the Northern Hemisphere. The temperature increase during last 30 yr has been calculated to increase BVOC emissions by 10% during this period, and an additional global emission increase of 30–45% could possibly be expected from further increase in temperature within this century (Peñuelas and Llusiá, 2003).

In this study we adopt a statistical approach to quantitatively and qualitatively estimate how increasing temperatures could contribute to the increased particle concentrations over northern parts of Europe. We will make use of the simple but efficient parametrization of particle formation potential resulting from pool dependent BVOC emission represented by monoterpene emission calculations as described by Tunved et al. (2006a). From this parametrization we investigate how the changes in these pool dependent emissions as the result of increased temperature, could affect the particle loads over the boreal forest.

2. Methods

The goal of the current study is to estimate the regional aerosol over Scandinavia resulting from biogenic emissions. The key method is to use a previously developed parametrization described in Tunved et al. (2006a). This parametrization links monoterpene emissions to both particle size distribution and mass over the Fenno-Scandinavian boreal forest. The parametrization will be tested against field measurements in Scandinavia, and we will use this parametrization to estimate the change in aerosol loads over the forest due to an increase in temperature. The approach aims to minimize the influence from other sources than the forest in this region, thereby providing a lower concentration limit for the natural aerosol over Scandinavia. The different methods and measurements used are outlined in detail below.

2.1. Size distribution data

Size distribution observations from four stations distributed over Scandinavia are used in the study. The stations are distributed over the Fenno-Scandinavian boreal zone, with Aspvreten (58.8°N, 17.4°E) and Birkenes (58.39°N, 8.5°E) at the southern rim of the boreal regions and Hyytiälä (61.85°N, 24.28°E). The sub arctic region is represented by the station in Värriö (67.77°N, 29.59°E) (see Fig. 4). All stations use DMPS systems covering a size range from a few nm up to

approximately half a micron. The instruments are built and calibrated using identical methods. Since the size ranges covered by the different instruments vary slightly, the size distribution data from the different stations are spline fitted using identical size distribution intervals in order to create a uniform dataset for all stations. A more detailed description of the stations can be found in e.g. Laakso et al. (2003) or Tunved et al. (2003).

2.2. Establishing the parametrization

In order to establish the parametrization that will be used to relate monoterpene emissions to aerosol properties over Scandinavia, it is necessary to have both a large number of size distribution observations, an equally large number of individual trajectories and estimates of the monoterpene emissions. The Hyytiälä station with its long record of size distribution measurements serves as an excellent basis for development of the parametrization. During the years 1997–2005, 120 h trajectories arriving 100 m above ground level (m a.g.l.) were calculated for each hour using the HYSPLIT4 model (Draxler and Hess, 1997). Each individual endpoint of the trajectories is associated with relative humidity, ground level temperature, precipitation and pressure. The ground level temperature was estimated from the temperature at the actual height of each endpoint assuming a lapse rate of 0.65 °C (100 m)$^{-1}$.

Trajectories arriving from north in a 180° transport sector 90°W–90°E relative Hyytiälä were selected, but only trajectories spending their first 5 h over ocean were considered and used to establish the parametrization. The method is similar, but not identical, to the method described by Tunved et al. (2006a). An imaginary box travels along the trajectory where the height of the box is equal to the average calculated mixing layer height for that particular trajectory. This approach differs slightly from the approach described in Tunved et al. (2006a) where the mixing layer height was fixed at 1000 m. Also, regarding the selection of the sector, the current study adopts a wider section for the study. Since also the new sector covers mainly boreal forests we assume that we improve the statistics of the current study without influencing the basic assumptions behind the parametrization. This may easily be motivated by the assumption that the gas-to-particle formation is similar over the boreal region, also over larger scales. These changes in approach compared with the previous study (i.e. Tunved et al., 2006a) are likely to induce a change in the result, mainly as a result of the change in mixing layer height. The temperature used for the emission calculations is the temperature along each trajectory. Monoterpene emissions were estimated using the relation $F = \varepsilon D \gamma$ where F represent the total flux of monoterpenes from the forest in μg m^{-2} h^{-1}, ε is the emission potential (μg g(dry weight)$^{-1}$ h^{-1}), D is the foliar biomass density in g (dry weight) m^{-2}, and γ is an environmental correction corrections factor accounting or temperature and light dependency of the emission. In the analysis we only account for temperature dependent pool emissions, therefore $\gamma(\text{pool}) = \exp[\beta(T - T_s)]$, $\beta = 0.09^{\circ\ -1}$ with $T_s = 303.15$ (Guenther, 1995). An emission potential of 1.5 μg g(dry weight)$^{-1}$ h^{-1} (Steinbrecher et al., 1999) was used. The foliar biomass density of pine and spruce species in the trajectory fetch areas was estimated by combining the latitude dependent biomass densities derived for Finland (Laurila and Lindfors, 1999) with a 1/4 degree resolution fractional land cover classification of needle leaf species (Loveland et al., 2001). The emissions are assumed to be confined below the calculated average mixing layer height during transport. The mixing layer height is averaged for each trajectory individually, and the previously calculated ground level temperature was used in the emission calculations. By being exposed to the difference in transport time over land, differences in foliar biomass density of coniferous species and difference in land cover and temperature, each trajectory arrives to the station with a unique pattern of emissions, and thus yields a range of emissions for all different trajectories that arrives to the stations. The calculated emission along each trajectory is related to observed size distributions in order to evaluate the dependence on particle size distribution properties from monoterpene emissions. Size distribution data covering ±1 h around the arrival of each trajectory was extracted. After removal of faulty or missing data, a total of 7800 cases described transport in the selected sector and were associated with size distribution measurements at the receptor station Hyytiälä. With the current model, accumulated monoterpene emissions (in units of concentration, μg m^{-3}) in the conceptual box varied between a few micrograms up to 60 μg m^{-3}. The median was found to be 11.4 μg m^{-3} (varying between 6.2 – 23.0 μg m^{-3} as 25th–75th percentiles, based on the variation of monoterpene emissions). The arithmetic mean was 17.7 μg m^{-3}.

The particle mass was integrated for each size distribution between 0.01 and 0.45 μm. The integrated particle mass and emissions were binned according to monoterpene emission increments of 0.5 μg m^{-3}. A straight line was fitted to the resulting data as aerosol mass versus accumulated monoterpene emissions, giving intercept of 0.92 μg m^{-3} at zero terpene emissions (Fig. 1). The slope was found to be 0.047. Given the short atmospheric lifetime of monoterpenes, the slope corresponds to aerosol mass gain per oxidised unit mass of monoterpenes (Tunved et al., 2006a). Thus, the slope is an indirect measure of the yield (Odum et al., 1996). However, since the parametrization omits other sources and excludes the role of sinks, the yield must be considered as an apparent yield only. Tunved et al. (2006a) calculated the apparent yield to be on average 7.5%, ranging from 5 to 10%. The lower value found in the current study is related to the fact that we are using a more realistic representation of the mixing layer height in the current study, that is, on average a slightly lower mixing layer height as compared to the 1000 m used in the previous study. The choice of mixing layer height along the trajectories is in fact directly correlated to the calculated apparent yield due to the assumptions upon which the parametrization is based.

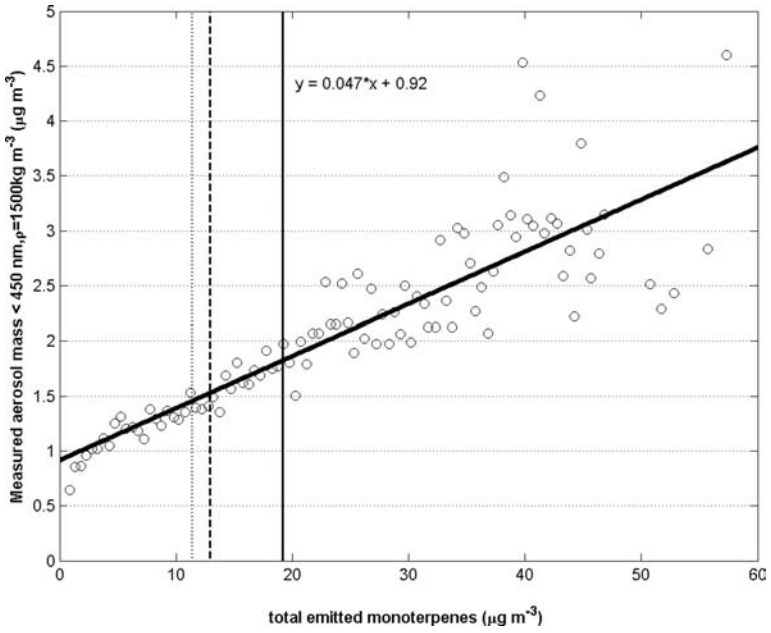

Fig. 1. Estimate of the particle mass gain per emitted mass of monoterpenes. The slope corresponds to the apparent mass yield. Hyytiälä April–September, 1997–2004. The vertical dashed lines correspond to the increase in monoterpene emissions. Intersect with solid black line represent resulting predicted aerosol mass. Response in mass by increase of temperature with 1.4° (dotted, vertical), 5.8° (dashed, vertical) and 8° (solid, vertical) considered.

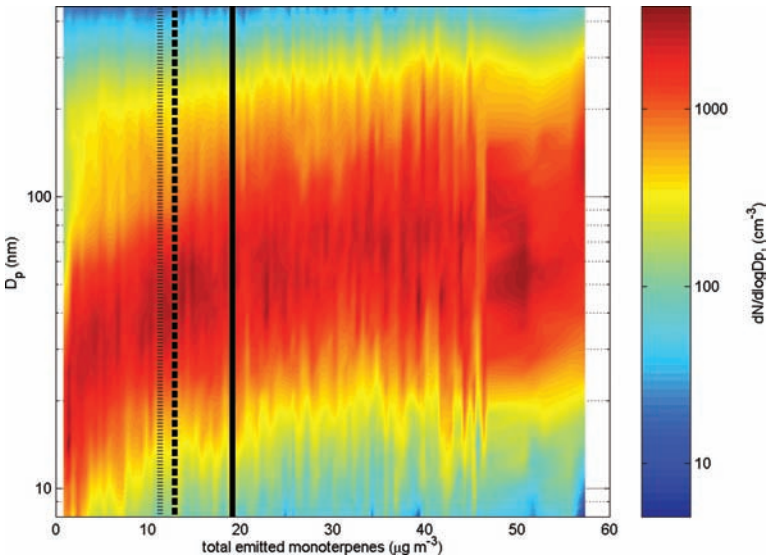

Fig. 2. Size distribution evolution as $dN/d\log D_p$ (cm^{-3}) as function of total mass of emitted monoterpenes ($\mu g\ m^{-3}$). Hyytiälä April–September, 1997–2004. The vertical dashed lines correspond to the increase in monoterpene emissions. Intersect with solid red line represent resulting predicted aerosol mass. Increase of temperature by 1.4° (dotted), 5.8° (dashed) and 8° (solid) considered.

Based on the equation of particle mass versus total amount of emitted monoterpenes presented in Fig. 1, the average particle mass resulting from transport over the boreal forest is thus 1.46 $\mu g\ m^{-3}$. This value is derived from the equation in Fig. 1 and calculated for today's calculated average emissions of monoterpenes, 11.4 $\mu g\ m^{-3}$.

The size distribution evolution as function of emitted monoterpenes is shown in Fig. 2. The mechanisms leading to accumulation of particle mass and number seem strongly connected to emissions of monoterpenes. This qualitatively means that the forest give the major contribution to the aerosol in the air transported also from northern Russia. One should however note that this does not directly suggest that monoterpenes (or equivalent compounds) are directly responsible for the creation of particle number. Instead, the emissions from the forest likely provide condensable material that allow recently formed particles to grow to a size range were they may survive as individual particles for longer time. These freshly formed particles likely have their source in aggregation of sulphuric acid, among other inorganic compounds. The presented correlation does not rule out the possibility that nucleation occurs frequently over the ocean as well, but instead suggests that it is when the air reaches the land based sources, that the concentration of condensable material reaches high enough levels to allow the particles to grow. What further can be seen in Fig. 2, is that the variability of the size distribution as it evolves with increasing estimated

emission is somewhat higher than for the evolution of the mass. This reflects the fact that the processes shaping the size distribution is far more complex than the processes shaping the mass evolution. Factors that are of importance are e.g. condensation sink, availability of nucleating gases (e.g. sulphuric acid), among others. These results are in agreement with Tunved et al. (2006a). Although not presented as an equation, Fig. 2 and the data associated with it, may be used to estimate the number and size of particles for a range of monoterpene emissions at different background sites in the boreal region Scandinavia

2.3. Test of parametrization at a different station

In order to test how the parametrization actually performs at another station inside the grid we apply it to the Birkenes station located at the Norwegian South-coast (58.35°N, 8.3°E). A northerly sector was drawn up from this point covering a range of 40° to −60° relative the station (0° represents north). Trajectories for the period 09/2002 to 08/2005 (this interval coincide with the availability of aerosol size distribution measurements) were calculated every 6 h and selected for analysis if they spent more than 70% in this northerly sector. The use of 70% in this case allows for more trajectories to arrive to the station, as its location (i.e. southwestern Norway) does not favour transport from the boreal region to the same extent as, for example, Aspvreten and Hyytiälä. Size distribution data was extracted around the arrival of these trajectories and the resulting size distribution was evaluated. Concurrent with this analysis we also calculated the monoterpene emissions along the path of trajectories, in the same way as described under Section 2.2. Using the parametrizations for mass and size distribution properties the calculated and measured size distribution were compared. The result is presented in Fig. 3. It can be seen that the agreement between our simple model and measured data at Birkenes captures the main features of the measured average size distribution fairly well. The parameterized distribution shows a more bimodal structure and underestimates the observed distribution in the range 70–150 nm diameter. The agreement between particles of larger sizes is however excellent. The mass agreement is perfect. As medians, the integrated mass at Birkenes is 1.32 μg m^{-3} (0.52–2.63 as 25th–75th percentiles) and the modelled mass is 1.32 μg m^{-3} (1.08–2.19 as 25th–75th percentiles, stemming from the 25th to 75th percentiles of the accumulated emissions. This range in turn, is the result of differences in transport pattern, average mixing layer height and temperatures resulting from different transport conditions). Thus, the parametrization overpredicts low end mass concentrations, but in turn underpredicts values in the high end of mass concentrations. Nevertheless, the results indicate that the model satisfactorily can be used to describe also the average aerosol properties in northerly air masses at locations close to, but outside, the boreal region.

3. Results and discussion

3.1. A simple test of the response to changes in temperature

As a first test we will investigate how the parametrization would respond to an increase in temperature. IPCC predict in a previous report that the temperature will increase with 1.4–5.8 °C until 2100 (IPCC, 2001). We assume a static scenario where neither the boreal ecosystem nor the emission potential of monoterpenes will change. We further assume that the change in temperature within this interval affects qualitative behaviour of our model

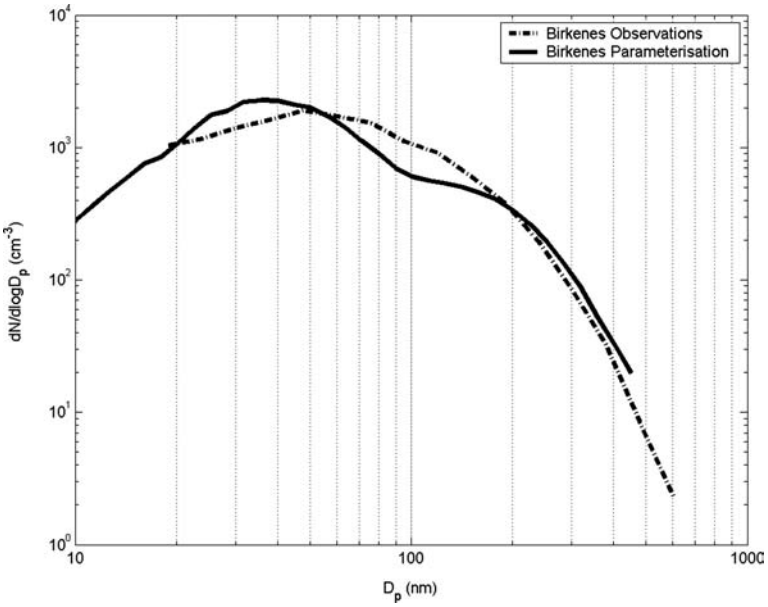

Fig. 3. Comparison of estimated and measured size distributions observed at Birkenes in air arriving in a northerly sector 40° to −60° relative the station (0° represent North). Data collected from 09/2002 to 08/2005 during months April–September. In total ∼950 hourly observations make up the data.

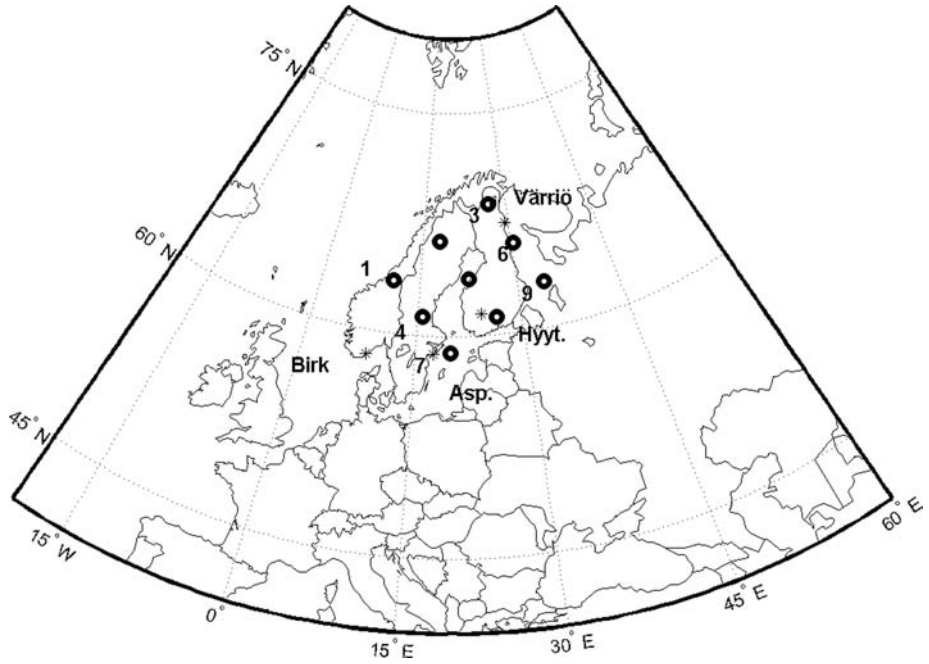

Fig. 4. The selected grid system. Värriö, Birkenes, Hyytiälä and Aspvreten indicated in the figure.

to small degree. By constraining the result by making these assumptions, we can easily calculate the increase in emissions in the selected sector. The response in both mass and size distribution properties to temperature increase can then easily be approximated from Figs. 1 and 2.

By increasing the temperature by 1.4° on average, the geometrical mean emission of monoterpenes in the selected sector would increase up to 12.9 μg m^{-3} (7.1–26.0 μg m^{-3} as 25th–75th percentile). This would result in a median mass concentration of 1.53 μg m^{-3}, that is, a 4.8% increase in aerosol mass during the selected transport conditions. With the highest estimate from IPCC, that is, a 5.8 °C increase in global average temperature, the emissions would increase to 19.2 (10.58–38.70 as 25th–75th percentile range) μg m^{-3}. Leaving all other parameters unchanged, this would result in a 25.1% increase in particle mass concentration at Hyytiälä during the selected transport conditions. The effect on number concentration will be treated in following section.

3.2. Regional estimate of natural aerosol loadings

The established relation between monoterpene emissions and aerosol properties has been evaluated and tested on large spatial scale previously (Tunved et al., 2006a). Together with the current study, it seems clear that that the mechanisms leading to aerosol formation in clean marine air masses over the boreal forest in Fenno-Scandinavia is similar over large areas of the boreal forest in Northern Europe. This assumption allows us to approximate the aerosol properties in marine air masses that transforms and arrive at all locations over the boreal areas in Fenno-Scandinavia, even at sites other than the one the relation has been derived for. This is especially evident from the evaluation of the parametrization at the measurement station Birkenes. Trajectories were calculated at 9 receptor locations 100 m a.g.l. evenly distributed over Scandinavia (Fig. 4). These trajectories were calculated with 6 h resolution through years 1999–2005. Each receptor point corresponds to the centre of a 450 × 450 km grid (Fig. 4). Along each trajectory, the monoterpene emissions were estimated and the accumulated emissions were calculated. However, in contrast to the trajectories arriving Hyytiälä, all transport directions were considered. The trajectories that arrive at the centre points of the grid system were allowed to accumulate monoterpene emissions from needle leaf sources above 58°N. No other source contribution was considered. This means that the only sources of monoterpenes in the air arriving to the different grids are needle leaf forests (i.e. spruce and pine) located in Scandinavia and northern parts of Russia.

It is clear from Fig. 1 that the mass concentration at low monoterpene emissions is small. If no other sources were present during transport over land, this mass concentration would be representative for the marine air arriving to the boreal zone. We now make the assumption that this background particle concentration is representative for the air entering the boreal zone from the marine environment and that the size distribution averaged for low monoterpene emissions could be interpreted as a steady state marine aerosol. In context, this approach means that all air reaching the boreal zone as defined in this study initially contain an aerosol with a mass concentration corresponding to the mass at zero monoterpene emissions (compare Figs. 1 and 2). This approach thus neglects all other sources except those

constituted by the forest above 58°N. This would conceptually be the same as if the forest was an island surrounded by oceans. The method will allow us to examine how the aerosol would evolve if no other sources than the forest were present, if a marine type background aerosol would enter the boreal zone.

By using the predefined background aerosol as starting point for evaluation of the aerosol properties at the different locations in the grid, we can estimate the particle production potential of the forest in a scenario where no continental particle sources are present. This will give us an estimate of the lower limit of the regional particle concentration in an entirely natural and needle leaf forest dominated source region. This will allow us to compare estimated natural aerosols with observed mixed natural anthropogenic aerosols, and allow for an estimate of the contribution from natural sources to the aerosol today.

As reported by Tunved et al. (2006a) there is a discrepancy in baseline values of both mass and number (e.g. the measured mass intersect with zero emissions is lower at northerly stations compared to southerly stations). This difference is most likely attributable to anthropogenic emissions as well as other natural emissions affecting the aerosol during transport. The farther south the station is located, the higher the probability is that the measured aerosol will contain particles influenced by anthropogenic sources. The lowest mass observed using the same approach as adopted in this study has been found to be 0.5 μg m^{-3} at Hyytiälä, and 0.25 μg m^{-3} at the northerly stations Värriö and Pallas (Tunved et al., 2006a). Thus, the true marine aerosol most likely contains a sub-450 nm particle mass in the range of 0.25 μg m^{-3}. Although anthropogenic sources are sparse, we cannot completely exclude their influence. In order to evaluate the role of biogenic sources we need to scale the parametrization accordingly. The simplest way to do this is to reduce the baseline value by a factor of 2. As we cannot be sure in what size range these emissions predominantly partition we adjust the whole number size distribution by a factor of 0.5, i.e. allow an aerosol of a total mass of 0.25 μg m^{-3} to enter the boreal zone. This assumption may appear a little crude, but is the best guess based on the current data set and methodology. In using this approach, we do also assume that the initial aerosol used in the parametrization is close to the marine type aerosol at high latitudes. This aerosol could be interpreted as a steady-state marine aerosol number and mass size distribution reflecting the balanced effect of both primary (e.g. sea spray) and secondary marine, for example, aerosol production as well as sinks (i.e. dry and wet deposition). The number concentration is higher than typically observed in marine air at high latitudes (e.g. Heintzenberg et al. (2000)) suggesting a value of $\sim$160 particles cm^{-3} in the Aitken size range with $\delta_{\mathrm{Aitken}} = 1.5$ and $Dg_{\mathrm{Aitken}} = 45$ nm The accumulation mode is represented by $N_{\mathrm{Acc}} = 60$ cm^{-3}, $\delta_{\mathrm{Acc}} = 1.6$, and $Dg_{\mathrm{Acc}} = 170$ nm in the accumulation mode size range. Our study suggests as a starting point a size distribution with modal parameters given by $N_{\mathrm{Aitken}} = 780$ $\delta_{\mathrm{Aitken}} = 1.9$ and $Dg_{\mathrm{Aitken}} = 25$ nm and $N_{\mathrm{Acc}} = 46$ cm^{-3} $\delta_{\mathrm{Acc}} = 1.40$ $Dg_{\mathrm{Acc}} =$ 0.177 nm. This result indicates that the accumulation mode is typically observed when only small amounts of monoterpenes have been emitted fairly well correspond to marine aerosols, whereas the Aitken mode contains far more particles. As discussed in Tunved et al. (2006a), the establishment of a number population in the smaller particle range over the forest is rapid as marine air is transported over the boreal region. This means that it is unlikely to find an aerosol with an Aitken mode number concentration in the same range as reported by Heintzenberg et al. (2000) in the area around Hyytiälä. Nevertheless, the size of the mode (23 nm) still indicates that these particles are recently formed.

Making the assumptions described above, the monoterpene mass accumulated during transport to the grid points in Fig. 4 were related to the parametrized mass increase and size distribution evolution. The analysis considers the time period April–September only since this is the time of the year when emissions of monoterpenes are assumed to reach non-negligible levels. As can be seen in Fig. 2, the size distribution evolution as function of monoterpene emissions is quite variable over small increments in monoterpene emissions. In order to avoid the bias resulting from this variation, an interval of ± 5 μg m^{-3} was extracted and averaged relative to each grid's calculated average monoterpene emissions. The result is presented in Fig. 5. An averaged number size distribution resulting from estimated monoterpene emissions is presented for each grid square (solid blue lines). As comparison, we add also observed size distribution data for the available stations within three of the grid squares as given by the dotted lines in Fig. 5.

The observed data represents the average observed number size distribution regardless of transport direction. The circles correspond to calculated size distribution without scaling factor and the solid lines correspond to the parametrized size distribution applying a scaling factor of 0.5. Värriö station is located in grid no. 3, Aspvreten in grid no. 7 and Hyytiälä in grid no. 8. By investigating the relation between measured overall and estimated, biogenic, aerosol size distribution at the stations Värriö, Aspvreten and Hyytiälä (corresponding to frame 3, frame 7 and frame 8 in Fig. 5, respectively) it is clear that the parametrization compares best with measurements at the northerly locations. The agreement between measurements and estimates gets worse the farther south the station is located. While the measurement includes also the anthropogenic component, the parametrization is assumed to represent the aerosol evolution as a function of monoterpene emissions only. Given the location of the stations, this result is consistent with natural sources dominating the aerosol at northern locations in the grid since the northerly locations are farther away from anthropogenic sources. The agreement between measurements and parametrized data in grid is good, although the average aerosol observed at Värriö during April–September contains more accumulation mode particles, as well as fewer sub-80 nm particles than derived with the parametrization. This is likely the result of the combined effect

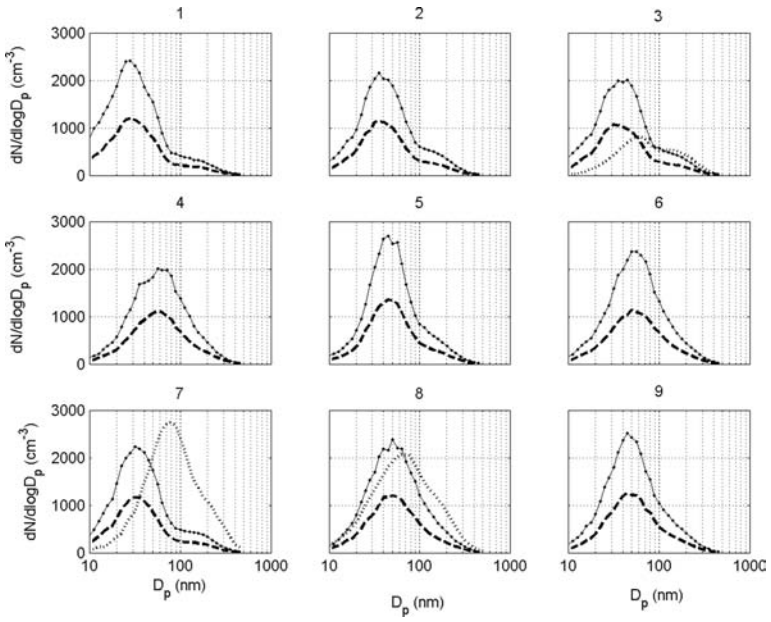

Fig. 5. The estimated size distribution resulting from monoterpene emissions from needle leaf sources above 58°N alone for each of the grid centre points shown in Fig. 1. Two scenarios assumed, one beginning with ∼0.5 μg m^3 of aerosol (circles) and one with a starting aerosol of 0.25 μg m^{-3} (dashed black) (see text for details). Observational data for Värriö (frame 3), Aspvreten (frame 7) and Hyytiälä (frame 8) (dotted lines) are added for comparison. All transport directions considered. Time period evaluated is April–September, years 1999–2004.

Table 1. Calculated mass and number concentrations for temperatures of today, for an increase of 1.4, 5.8 and 8 °C at the different grid points shown in Fig. 4

	Today		IPCC2001 1.4 °C		IPCC2001 5.8 °C		ANDREAE 8 °C	
Grid no.	N^* (cm^{-3})	m (μg m^{-3})	N (cm^{-3})	m (μg m^{-3})	N (cm^{-3})	m (μg m^{-3})	N (cm^{-3})	m (μg m^{-3})
1	767	0.61	766	0.63	766	0.72	760	0.77
2	771	0.86	789	0.92	812	1.14	833	1.29
3	772 (627)	0.81 (1.57)	772	0.86	828	1.05	814	1.18
4	797	1.43	774	1.56	792	2.1	758	2.45
5	828	1.11	828	1.19	733	1.55	781	1.79
6	813	1.34	794	1.46	787	1.95	719	2.27
7	760 (1900)	0.72 (5.07)	763	0.75	808	0.9	789	0.99
8	812 (1750)	1.28 (3.48)	809	1.39	762	1.85	719	2.15
9	827	1.24	813	1.34	746	1.77	734	2.06
Ave.	794	1.04	790	1.12	782	1.45	768	1.66

Note: Average mass and number concentrations are shown in last row. Values within brackets indicate the average of the measured data at the stations Värriö, Aspvreten and Hyytiälä, respectively. *N corresponds to integrated number concentration 10–450 nm.

of both other natural sources as well as anthropogenic emissions contributing to the measured, average aerosol at Värriö. Worst representation is found at Aspvreten (grid 7). This is likely the result of a substantial influence from continental sources at this station, an influence that is not included in the parametrization. Grid 8, represented by Hyytiälä, falls in the range between Aspvreten and Hyytiälä.

This fact has already previously been demonstrated (e.g. Tunved et al., 2003). On average the mass (<450 nm) observed at Aspvreten between April and September is 5.07 μg m^{-3}. The Estimated contribution from the forests alone given typical transport and temperature conditions is 0.72 μg m^{-3}. The observed average number concentration for all transport directions at Aspvreten during April–September 1999–2004 is 1900 cm^{-3}. The parametrization suggests a typical number concentration of 760 cm^{-3} (see Table 1 for further details). Corresponding values for the Hyytiälä grid is 3.48 and 1.28 μg m^3 for measured and calculated mass, respectively. The number concentrations correspond to 1750 and 812 cm^{-3} for measured and estimated 'boreal' number concentrations. At Värriö the measured mass is 1.57 and calculated mass is 0.81 μg m^{-3}. The number concentrations at Värriö correspond to 627 and 772 cm^{-3} for measured 'boreal' and estimated "boreal" number concentrations. This means that the forest above 58° alone could alone amount to

12.6, 36.8 and 52% of the mass observed given today's anthropogenic influence. The large difference between the observed average aerosol number size distributions at the stations could most likely be attributable to the fact that when polluted air is transported over forested areas, it has left the large continental sources behind and the rest of the transport is characterised by typically much smaller source strength. This in turn means that the sinks will be larger than the sources. As the air is transported away from these comparably large sources, the mass will be reduced until a new balance between sources and sinks is reached. This will be noticeable as an apparent reduction of particle mass (and number of large particles) when comparing measurement sites going from south to north. The parametrization is derived for situations where air with low concentrations of aerosol enters the boreal zone. Nucleation then rapidly establishes high number concentrations in the nuclei-Aitken size range. In the case of continental air that is transported northwards the situation is different. Formation of small particles does not occur (or have too short lifetimes to be observed) in these air masses. Instead mass is accumulated on the existing particles.

Regarding the aerosol number concentration, it seems that a typical number concentration that is supported by the forest is somewhere close to 800 cm^{-3} (Table 1) This number is fairly consistent, regardless of location in the map of Fig. 4, which in turn suggests that the forest alone is able to support an aerosol number population of approximately 800 cm^{-3}. Furthermore, the consistency between the grid points indicates that this number concentration is established rapidly. Once established, this number concentration seems to be fairly constant throughout the transport over the forest. The number concentration seems to reflect the typical number concentration that the forest can sustain. Once this number is established, subsequent new particle formation will be quenched; No new particle number is created and the removal of particles in the Aitken size range due to coagulation or deposition is comparably slow. The stable number concentration is contrasted by the mass concentrations observed in the different grids, which seem to vary with a factor of at least two, depending on the location of the receptor over Scandinavia.

3.3. Estimated changes in aerosol population and CCN concentration using different temperature scenarios

The estimate of the aerosol over the entire grid is displayed in Fig. 6. Since the emissions of monoterpenes exponentially depend on temperature it is easy to test the response of the parametrization resulting from a temperature increase. In order to investigate this trajectories were allowed to accumulate monoterpenes under three different scenarios: temperature is increased by 1.4°, 5.8° and 8°, in the following referred to IPCC14, IPCC5.8 (IPCC, 2001) and Andreae et al. (2005), respectively. The procedure used in this evaluation is identical to the evaluation of the aerosol in the separate grid points as described under Section 3.1. In a changing climate many conditions may change and influence the aerosol characteristics at any given place and time. This may be a changed monoterpene saturation vapour pressure, a change in transport patterns, a change in precipitation, or a change in forest cover as well as other such relevant parameters. Here we isolate the effect of a change in temperature only and assess the magnitude of the impact on aerosol characteristics. From Fig. 6 it is evident that the changes in temperature and thereby emissions have an effect on the resulting aerosol. This could have implications for the radiative balance due to the direct effect mediated by aerosol particles, although this effect is believed to be small under typical conditions in over Scandinavia. More important is the effect on the availability of particles

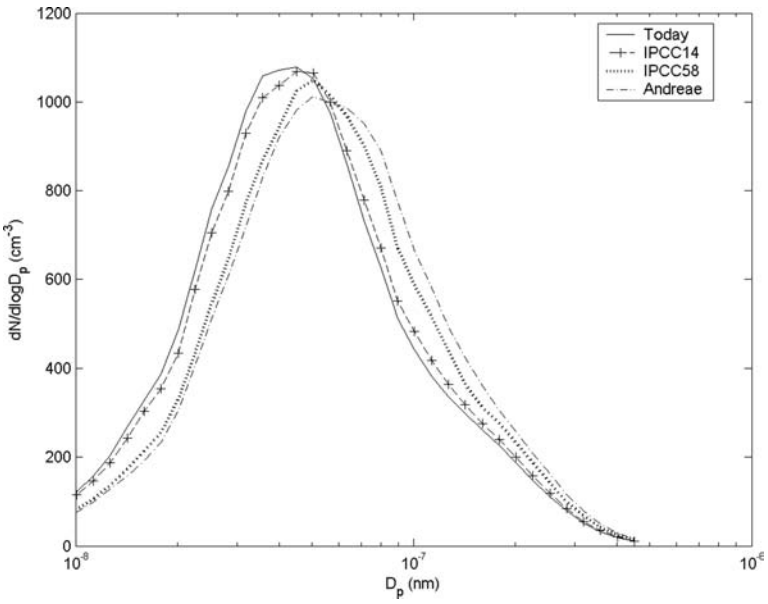

Fig. 6. Estimated regional average size distribution resulting from needle leaf forest sources alone over the northern Fenno Scandinavian region. Solid line represents the typical size distribution at current. Increase of temperature by 1.4° (+-signs), 5.8° (dotted) and 8° (dash–dotted) considered.

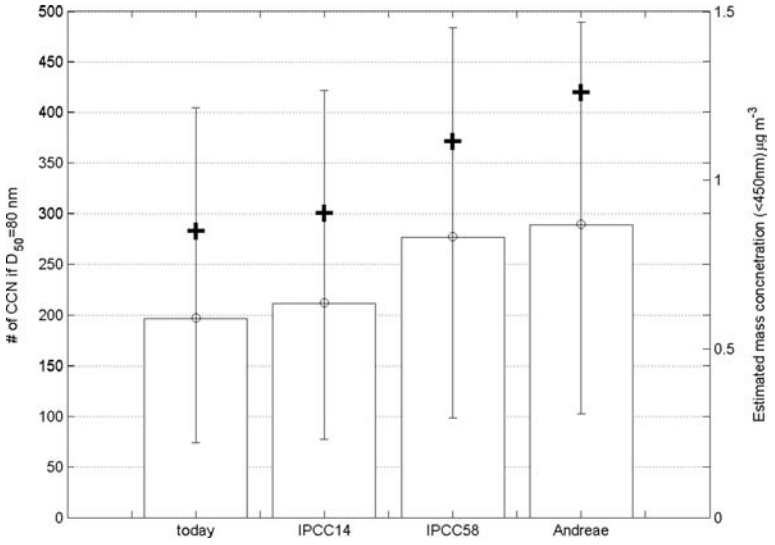

Fig. 7. Estimated average regional CCN concentration assuming $D_{50} = 80$ nm (bars) and mass concentration (crosses) at different temperature scenarios with all other parameters held constant. The range given in the figure corresponds to different scenarios assuming a CCN threshold of 50 nm as lowest possible activation diameter and 128 nm as the highest possible activation diameter, on average.

that might act as Cloud Condensation Nuclei (CCN), and therefore also the indirect effect on the radiation budget of the atmosphere. Controlled studies investigating the CCN capability of the organic aerosols as reported by Hartz et al. (2005) show that biogenic SOA derived from selected monoterpenes act as excellent CCN, with an average activation diameter around 50 nm at 1% supersaturation. A previous study by Komppula et al. (2005) report that the average activation diameter (D_{50}, i.e. diameter of which 50% of the particles activate) is on average 80 nm, varying between 50 and 128 nm, at a subarctic site close to Värriö. Also Kerminen et al. (2005) have shown based on observations that new particle formation really leads to cloud droplet formation. Using these thresholds as a lower limit of the particle size that may act as CCN, we simply estimated the number of potential CCN's by summing up the total number concentration above 50, 80 and 128 nm to get the maximum, average and minimum number of potential CCN's that would result from a temperature increase alone. The result is displayed in Fig. 7 together with predicted increase in aerosol mass. The corresponding mass for each scenario (today, IPCC14, IPCC5.8, Andreae) was found to be 0.85, 0.90, 1.12 and 1.26 μg m^{-3}. Corresponding concentration of potential CCN (assuming $D_{50} = 80$ nm) concentration was found to be 197, 212, 277 and 289 cm^{-3}. For the lower limit (assuming $D_{50} = 128$ nm), corresponding numbers would be 74, 78, 98 and 103 cm^{-3}. For the lowest 'CCN threshold' ($D_{50} = 50$ nm), the resulting potential CCN concentrations would be 404, 421, 483 and 489, respectively for the different temperature scenarios.

4. Conclusions

The natural particle loadings constitute one of the major uncertainties in addressing the climate impact resulting from anthropogenic emissions of climate active gases such as carbon dioxide and methane. The secondary production of fine mode particles from biogenic VOC's is comparable in magnitude to secondary fine particle production resulting from anthropogenic sulphur emissions (Kiehl and Rodhe, 1995). Monoterpenes are believed to play a major role in the global production of secondary organic carbon aerosols. One of the central problems associated with estimates of the climate effect induced by anthropogenic particle emissions is that the natural, unperturbed, particle system is very hard to describe. In this study we have utilized and slightly modified a parametrization for particle formation associated with emission of monoterpenes in clean, marine air masses to explore the role of naturally produced particles in relation to anthropogenic influenced particle loadings over northern Scandinavia. The results show that under present conditions, the boreal forest alone can sustain an aerosol population in the summertime atmosphere over Scandinavia that by mass corresponds to at least 12–50% of the today's mixed anthropogenic-natural aerosol system, depending on geographical location in the Fenno-Scandinavian Region. Based on previous findings by Komppula et al. (2005) and Kerminen et al. (2005), we further concluded that the forest itself can sustain a regional CCN population of approximately 200 cm^{-3}. This number is double the amount of CCN encountered under unperturbed marine conditions. It was further shown that the sensitivity to particle production resulting from changes in temperature was high. By modifying the monoterpene emissions through a change in the temperature following scenarios presented by IPCC2001 and Andreae et al. (2006) we showed that increase in temperature substantially could alter the particle production over the forest, thereby possibly increasing the CCN concentration by 46% due to a 8 °C temperature increase (7 and 40% increase for a temperature increase of 1.4° and 5.8°, respectively). The scenario assumes that all other parameters except temperature are constant. However, as climate change most certainly will affect

the characteristics of the worlds ecosystem, and thus also the boreal forest, these predictions must be treated with caution. One parameter that is believed to alter the emission potential of monoterpenes is the actual concentration of CO_2. A recent study by Arneth et al. (2007) for example suggests that increasing CO_2 concentrations actually can reduce isoprene emissions. Therefore, in order to accurately address the change in emissions of monoterpenes and their effect on the particle loads in the atmosphere we need to improve our understanding of the change in emissions from the forest resulting from a changed chemistry of the atmosphere and change in forested area and species distribution, besides the change in temperatures. Nevertheless, it seems clear that the forest could facilitate a very important climate feedback that could help slow warming over forested regions, based on temperature increase alone. Our results agree qualitatively with recent estimations by Kurten et al. (2003).

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