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The importance of accounting for enhanced emissions of monoterpenes from new Scots pine foliage in models - A Finnish case study

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ABSTRACT

Models to predict the emissions of biogenic volatile organic compounds (BVOCs) from terrestrial vegetation largely use standardised emission potentials derived from shoot enclosure measurements of mature foliage. In these models, the potential of new foliage to emit BVOCs is assumed to be similar, or up to twice as high, as that of mature foliage, and thus new conifers foliage is predicted to have a negligible to minor contribution to canopy BVOC emissions during spring time due to the small foliage mass of emerging and growing needles. Extensive observations have, however, recently demonstrated that the potential of new Scots pine needles to emit several different BVOCs can be up to about 500 times higher than that of the corresponding mature foliage. Thus, we build on these discoveries and investigate the potential impact of considering these enhanced emissions from new Scots pine foliage on estimates of monoterpene emissions and new atmospheric aerosol particle formation and their subsequent growth. We show that the importance of taking the enhanced monoterpene emission potential of new Scots pine foliage into account decreases as a function of season, tree age and latitude, and that new foliage could be responsible for the majority of the whole tree's foliage emissions of monoterpenes during spring time, independently of tree age and location. Our results suggest that annual monoterpene emission estimates from Finland would increase with up to ~25% if the enhanced emissions from new Scots pine foliage were also considered, with the majority being emitted during spring time where also new particle formation has been observed to occur most frequently. We estimate that our findings can lead to increases in predictions of the formation rates of 2 nm particles during spring time by ~75–280% in northern Finland and by ~130–870% in southern Finland. Likewise, simulated growth rates of 2–3 nm particles would increase by ~65–180% in northern Finland and by ~110–520% in southern Finland if the enhanced emissions of monoterpenes from new Scots pine foliage were explicitly considered. Since only one measurement study (Aalto et al., 2014), on which our work builds, has so far found highly pronounced emissions of monoterpenes from new Scots pine foliage compared to those of mature, we conclude that more spring time measurements of new conifers foliage are required for improving emission algorithms in biogenic emission models.

1. Introduction

Biogenic volatile organic compounds (BVOCs) form a large, heterogeneous group of organic atmospheric trace gases with wide varieties in chemical and physical properties. They are produced and emitted by vegetation due to many different reasons (Holopainen, 2004; Yuan et al., 2009; Holopainen et al., 2013; Tumlinson 2014), for example as a by-product of plant growth (e.g. Hüve et al., 2007; Aalto et al., 2014; Dorokhov et al., 2018) or in response to plant stress (Niinemets, 2010; Holopainen and Gershenzon, 2010; Faiola and Taipale, 2020). Emissions of monoterpenes ($C_{10}H_{16}$), an important class of terpenes, account for approximately 15% of the total global BVOC emissions from vegetation (Guenther et al., 2012). The fraction of assimilated carbon which is transferred back to the atmosphere in the form of a variety of BVOCs is usually around a few percent (Guenther et al., 1995; Bouvier-Brown et al., 2012), but can at times be more than 10% (Harley et al., 1996;

Llusiá and Peñuelas, 2000). Thus, BVOCs compose an important factor to consider in terrestrial plants' carbon balance. In the atmosphere, BVOCs influence the chemical composition (Mogensen et al., 2011, 2015), and impact formation (Donahue et al., 2013; Kulmala et al., 2014; Riccobono et al., 2014; Schobesberger et al., 2013) and growth (Ehn et al., 2014; Riipinen et al., 2012) processes of atmospheric aerosol particles. Since aerosol particles are known to influence our climate both directly and indirectly (Twomey, 1977; Albrecht, 1989; Charlson et al., 1992), reliable estimates of BVOC emissions into the atmosphere are crucial for predictions of climate change.

There exists several models to predict the constitutive emissions of BVOCs from terrestrial ecosystems into the atmosphere (e.g. MEGAN; Guenther et al. (2006, 2012), ORCHIDEE; Lathière et al. (2006), Messina et al. (2016), LPJ-GUESS; Smith et al. (2001), Sitch et al. (2003)), with MEGAN being the most popular one. Traditionally, these types of models have utilised emission potentials derived from shoot enclosure measurements of mature foliage. An emission potential, or emission factor, represents the emission rate of a compound at standard conditions (in this work at a temperature of 30 °C). As an increasing amount of studies have shown that the emissions of BVOCs depend on phenology (Guenther et al., 1991; Monson et al., 1994; Goldstein et al., 1998; Hakola et al., 2001; Petron et al., 2001; Karl et al., 2003; Räisänen et al., 2009; Aalto et al., 2014), attempts have been made to include this response in models. For example, in the ORCHIDEE model, leaf age now impacts emissions of isoprene and methanol (Messina et al., 2016). Though leaf age is not explicitly simulated in LPJ-GUESS, the emissions of isoprene from deciduous plant functional types are modelled to depend on seasonality (Arneth et al., 2007; Schurgers et al., 2011). In MEGAN v2.0 (Guenther et al., 2006), the emission rate of isoprene is modulated by the leaf developmental stages of deciduous land cover types. This has been further expanded in MEGAN v2.1 (Guenther et al., 2012), where the emission rates of more compounds (i.e. isoprene, methanol, 2-methyl-3-buten-2-ol, mono- and sesquiterpenes) from all plant species are assumed to be regulated by plant growth. Though it is assumed that leaf age impacts the emission rates of individual BVOCs differently, this dependency has not been treated to be tree species, or plant functional type, specific (Guenther et al., 2012). Since the majority of studies investigating the impact of leaf age on BVOCs emission rates have been conducted on deciduous isoprene emitting species, this might create a bias. For example, in MEGAN v2.1, the potentials of growing foliage to emit methanol, 2-methyl-3-buten-2-ol and monoterpenes are 3, 0.6, and 1.8 times that of mature foliage, respectively. However, measurements of Scots pine foliage have recently shown that the potential of new foliage to emit these BVOCs can be orders of magnitude higher than that of mature foliage (Aalto et al., 2014). This conclusion was drawn based on continuous enclosure measurements of three growing seasons (Aalto et al., 2014). Aalto et al. (2014) showed that the emission potentials of new foliage peak during spring and decrease significantly throughout the season, and hence depend far more on the time of year than that of mature foliage. Thus, it might also not be representative to use a fixed emission potential of new foliage in models. These findings can have substantial impacts on simulations of global BVOC emissions, since Scots pine is the most widely distributed pine species in the world; it is found across large parts of Europe, Canada, US and northern Asia, and within the Eurasian taiga, it is one of the most dominant evergreen tree species (e.g. Houston Durrant et al., 2016). For example, in Finland, Scots pine dominates ~65% of forest land (Finnish Statistical Yearbook of Forestry 2014). Due to lack of observations, we cannot exclude the possibility that also other evergreen species exhibit a similar phenological emission trait as found for Scots pine in Aalto et al. (2014).

Micrometeorological measurements of ecosystem scale fluxes are able to capture the contribution of all BVOC sources in the ecosystem, though without quantifying what those sources are. Unfortunately, such measurements are scarce, rarely continuous, and usually conducted during a limited period, which is most often in the summer, when the

very high emission potentials of new Scots pine needles have already significantly decreased (Aalto et al., 2014). Rinne et al. (2000) measured the ecosystem scale flux of monoterpenes from Scots pine dominated forests during two growing seasons, including May, but only for a few days in total, thus they reported the emission potential as a seasonal average. Räisänen et al. (2009) measured the ecosystem scale flux of monoterpenes from a Scots pine forest, in addition to the emissions from new and mature needles individually. Measurements of the ecosystem flux and chamber emissions of mature foliage were conducted from the end of June, while the detection of the emissions from new foliage was only started at the end of July. As the measurements were performed sporadically, only seasonally averaged potentials have been provided. The authors found that new needles have a higher potential to emit monoterpenes than mature needles by a factor of two, which is comparable to what is used in Guenther et al. (2012). However, these measurements did not cover the vital spring season. Taipale et al. (2011) and Rantala et al. (2015) measured the ecosystem scale flux of monoterpenes continuously starting from April or May until September, during four years. In both studies, the micrometeorological measurements were conducted on the same ~50 year old Scots pine forest at the SMEAR II station (Station for Measuring Ecosystem-Atmosphere Relations). The canopy, within an area with a radius of 200 m, is made up by Scots pine (~75%), Norway spruce (~15%) and deciduous species (~10%), mainly silver birch (Mäki et al., 2019). The potential of the forest to emit monoterpenes per ground area was in both cases shown to significantly decrease from spring and over the summer (Taipale et al., 2011; Rantala et al., 2015). Since the pines in that region carry about 2.5 needle age classes (Ťupek et al., 2015), the foliage mass is approximately 40% less in the spring than later in the season (i.e. about August onwards). Hence, the conclusion by Taipale et al. (2011) and Rantala et al. (2015) is further amplified if the potential to emit is considered per foliage mass.

If a model assumes that the emission potential of new needles is only slightly higher than that of mature foliage, then the influence of new coniferous foliage to canopy BVOC emissions is predicted to be very minor, since the mass of emerging and growing needles is small during spring time (Guenther et al., 2012). However, though the mass of new foliage is very small in the beginning of the growing season, correspondingly larger emission potentials of new foliage during spring time would change the conclusion of the contribution of new Scots pine foliage to Scots pine canopy BVOC emissions. In order to obtain a better understanding of the formation of new aerosol particles, it is especially crucial to investigate this importance of new Scots pine foliage to ecosystem BVOC emissions during spring time, since that is the time of year where new particle formation has been found to be most frequent (Vehkamäki et al., 2004; Dal Maso et al., 2005, 2007, 2008; Manninen et al., 2010; Vana et al., 2016).

The motivation of this study arises from the fact that Aalto et al. (2014) have shown that emerging and mature Scots pine foliage can have very different potentials to emit BVOCs. Such evidence naturally calls for a quantification of its potential atmospheric impacts. Thus, by making simple assumptions based on the existing data, and considering the contribution of the enhanced constitutive emission potential of new Scots pine foliage, we investigated the potential effects on the whole tree's emission potential. We examined this as a function of season, stand age and location in Finland, utilising published emission rates by Aalto et al. (2014) and models to predict the seasonal and yearly growth of Scots pine foliage. In order to analyse the potential underestimation of regional emissions when these enhanced emissions from new foliage is not accounted for, we upscaled our results to answer how many Gg of carbon could be underestimated in the predictions of constitutive monoterpene emissions from Finland. Finally, we estimated how this potential underestimation could impact forecasts of formation and growth of new small particles. Our ultimate objectives were to demonstrate the potential effects of monoterpenes from growing Scots pine needles on model predictions, question the current treatment of the

emissions of BVOCs from new coniferous foliage in biogenic emission models, and motivate readers to investigate whether new foliage of other evergreen species is also a very strong emitter of monoterpenes and other BVOCs.

2. Materials and methods

2.1. Yearly development of Scots pine needle mass

The yearly development of Scots pine needle mass was calculated for southern and northern Finland, by considering the total amount of needle age classes present in the stand and the maximum stand needle biomass. Hence, we defined that the stands carry 2.5 and 5.5 needle age classes in southern and northern Finland, respectively, which is based on observations from Finland (Korhonen et al., 2013; Wang et al., 2013; Ťupek et al., 2015). A maximum stand needle biomass of 5000 kg ha^{-1}, which is representative for southern and middle Finland (Ilvesniemi and Liu, 2001), was used for southern Finland, while 3500 kg ha^{-1}, which is representative for a relatively poor site in Lapland (Kulmala et al., 2019), was used for northern Finland. We utilised this foliage mass value for northern Finland, as the calculation results of northern Finland should serve as a lower estimate of the potential impact of the emission of monoterpenes from new foliage to the total stand emission. Finally, it is assumed that needle mass development follows a sigmoidal form (e.g. Mäkelä, 1997). Since tree foliage growth models usually omit simulating the growth of very young trees (e.g. Hari et al., 2008; Minunno et al., 2019), because of their low relevance with respect to e.g. biomass production, we likewise only modelled the growth of trees aged ≥10 years. The maximum stand needle mass in southern Finland is reached at the same time as the observed canopy closure at the SMEAR II station, Hyytiälä, southern Finland (e.g. Hari and Kulmala, 2005; Kulmala et al., 2001). It is assumed that the maximum is reached in northern Finland 15 years later, due to slower forest growth in the north (Fig. 1a). Since the stand foliage mass is higher in southern than northern Finland, and since fewer needle age classes prevail in the south, both the mass of new needles and the mass of senescing needles are significantly higher in southern than northern Finland (Fig. 1b and c). The mass of new needles is calculated as:

$$G_i^N = m_i^N - m_{i-1}^N + S_i^N \tag{1}$$

where G_i^N is the growth of new needles during year i (kgC), m_i^N is the maximum needle mass during year i (kgC) and S_i^N is senescence during year i. After canopy closure, $m_i^N = m_{i-1}^N$ and thus:

$$G_i^N = S_i^N = \frac{m_i^N}{I_j} \tag{2}$$

where I_j is needle longevity in the two locations. Since the foliage production rate is high in young stands (derivative of Fig. 1a), the fraction of new needles to the total stand needle mass is also higher in young than mature pine forest stands (Fig. 1d).
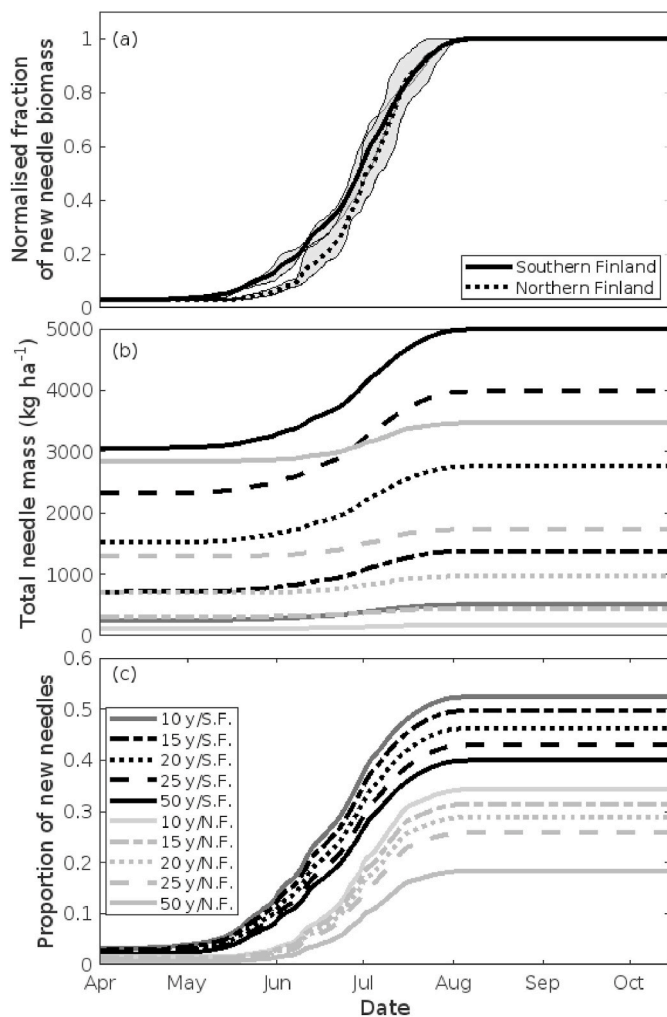
2.2. Seasonal development of Scots pine needle mass

The seasonal development of Scots pine needle mass was modelled with the CASSIA growth model (Schiestl-Aalto et al., 2015), where the daily growth of tree organs is driven by environmental variables, mainly temperature. Scots pine needles start elongating in spring simultaneously with the shoot, but shoot length growth is completed approximately one month before the growth of needles finishes. The model considers two parameters, which need to be estimated for the location of interest. Those are: time of growth onset and time of growth cessation. CASSIA has previously been parameterized using growth data measured in 2008 at the SMEAR II station, and the model has been shown to successfully predict the growth of needles (Schiestl-Aalto et al., 2015).
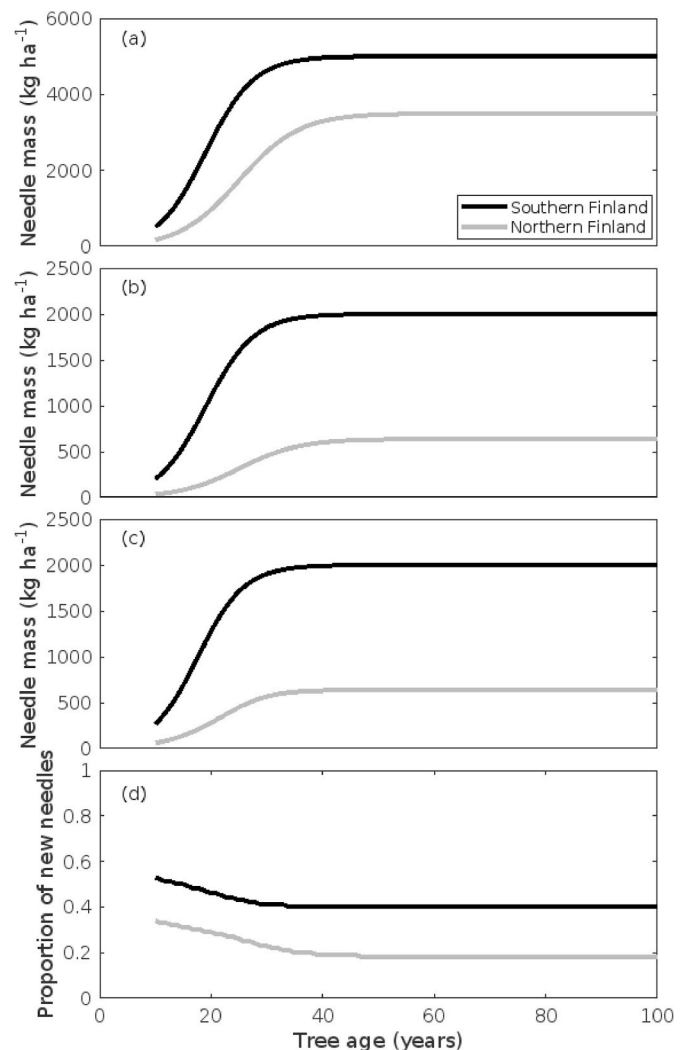
Fig. 1. Yearly Scots pine needle mass development. Values are given for the end of the growing season, assuming that the stand carries 2.5 (southern Finland) or 5.5 (northern Finland) needle year classes, respectively. **(a)** total stand needle mass before senescing needles fall off, **(b)** mass of senescing needles, **(c)** mass of new needles, **(d)** proportion of new needles to the total stand needle mass. Note the different scales on the y-axis.

We used this parameterization of time of growth onset and time of growth cessation to predict the seasonal development of Scots pine needles in southern Finland, while the corresponding growth in northern Finland was predicted utilising needle growth measurements conducted at the SMEAR I station in Värriö, Finnish Lapland, during the 2017 growing season. Furthermore, the model considers needle length by the end of the growing season as a yearly varying parameter. This parameter can be modelled if needed, but as the final needle length was measured at both stations during the years 2009–2011, we used the measured values. Additionally, the length of the needle primordia (i.e. the needles inside the bud) was set to 1 mm, and it was assumed that needle length is proportional to needle biomass (Aalto et al., 2014; Schiestl-Aalto et al., 2015, 2019). The relative needle mass per day was then calculated as L_d^N/L_{365}^N where L_d^N is the needle length on day d and L_{365}^N is needle length by the end of the growing season. Environmental data measured at the SMEAR II and SMEAR I station, respectively, during 2009–2011, were furthermore used as input to CASSIA. The resulting seasonal development of new Scots pine needles in southern and northern Finland is illustrated in Fig. 2a. Variations in the growth between the three investigated growing seasons are generally very

Fig. 2. Seasonal Scots pine needle mass development. **(a)** development of new needle mass in southern and northern Finland expressed as the normalised fraction of new needles out of the total new needle mass. Black curves are calculated as the mean during 2009–2011 in SMEAR I (northern Finland) and SMEAR II (southern Finland) conditions. The grey areas illustrate the variation between the model predictions for the three years. **(b)** total needle mass development for a Scots pine stand of several different ages throughout a growing season in southern (2.5 needle age classes) and northern (5.5 needle age classes) Finland. **(c)** proportion of new needles to the total stand needle mass throughout the season for different stand ages in southern (S.F.) and northern (N.F.) Finland. The legend shown in **(c)** is also valid for **(b)**. Note the different scales on the y-axis.

small, but greater in northern Finland, due to larger interannual fluctuations in ambient temperatures. The seasonal development of the total needle mass for Scots pine stands of different ages growing in southern and northern Finland is presented in Fig. 2b. This has been calculated by combining the behaviour shown in Fig. 2a with total stand needle mass values from Fig. 1a. The seasonal behaviour is also in accordance with observations (Rautiainen et al., 2012) before needles fall off. The fraction of new needles out of total stand needle mass for Scots pine stands of different ages growing in southern and northern Finland is provided in Fig. 2c. This has been calculated by combining the behaviour shown in Fig. 2a with new stand needle mass values from Fig. 1c.

2.3. Emissions of monoterpenes

2.3.1. Emissions of monoterpenes from new and mature Scots pine foliage based on Aalto et al. (2014)

We utilised measured emission rates of monoterpenes and chamber temperatures described and published in Aalto et al. (2014), hence we refer to Aalto et al. (2014) for details on the measurement set-up. In brief, the shoot exchange of monoterpenes was measured with an automated gas-exchange enclosure system and analysed by PTR-QMS (Proton Transfer Reaction - Quadrupole Mass Spectrometer) from one ~50 year old Scots pine tree located at the SMEAR II station during 2009–2011. Within one season, one mature shoot and one current year bud/shoot were measured, but during the next growing season, different shoots were chosen for the measurements. The shoot enclosures included parts of the shoots, i.e. both needles and the stem (see Fig. 1 in Aalto et al. (2014)). The reported emissions of VOCs from new foliage originated from buds in the beginning of the measurement period. In an elongating bud of Scots pine the stem develops first and growth of needles is very slow during the first ca. 5 weeks of the growth period (in southern Finland conditions, see e.g. Fig. 4 in Aalto et al., 2014). Thus, during the first weeks, it is likely that the reported emissions originate from the elongating (green) stem rather than from the needle primordia. It is additionally possible that some part of the emission, which is measured before the bud starts elongating, could originate from resin, which can be exudated even from healthy trees (Eller et al., 2013).

Only periods with data from both new and mature needles were considered. Since our analysis focused on emission potentials, we did not include exactly the same data as Aalto et al. (2014), because we were limited by occasional breaks in the measurements of chamber temperature. Though Ghirardo et al. (2010) and Taipale et al. (2011) have earlier demonstrated that a significant fraction of the total emissions of monoterpenes from Scots pine trees originates directly from *de novo* synthesis, the understanding of light-dependency on emissions from conifers trees is still very poor (e.g. Taipale et al., 2011), and thus the emission rates were standardised by Eq. (5) in Guenther et al. (1993) (T_s = 30 °C, β = 0.09 °C^{-1}) in order to also compare to literature values. We refrained from utilising varying β values (e.g. Hellén et al., 2018), since the temperature dependency is very sensitive to a low number of data points and any noise in the emission rate measurements. We consider our standardisation practice to be reasonable, since the ratio of the emission rates of new and mature foliage (Aalto et al., 2014) follows the same pattern as that of the emission potentials (Fig. 3).

The ratios of the emission potential of new needles to the emission potential of mature needles for the growing seasons in 2009–2011 are presented in Fig. 3. The subfigures in Fig. 3 have been cut due to clarity, but the excluded outliers are compiled in Table A1 together with information about the total amount of data points considered per one week average. As seen from the figure and also concluded by Aalto et al. (2014), new Scots pine needles can have a much greater potential to emit monoterpenes than mature needles. The difference in the potential to emit decreases throughout the season, but lasts until the lignification of the shoot is finalised. Hence, young shoots continue to have a higher potential to emit monoterpenes than mature needles until the end of August/beginning of September (Fig. 3f). Fig. 3 also illustrates why continuous measurements of VOC emissions are needed for providing sound emission potentials; (1) there is a large spread in the emission rates, even when standardised, thus having only a few measurement points might lead to biased emission potentials, and (2) emission rates, and hence potentials, are seasonally dependant, which has been shown already earlier for Scots pine, but also for other tree species (e.g. Hakola et al., 2001; 2006; Wang et al., 2017; Karl et al., 2003; Komenda and Koppmann, 2002). Additionally, it is clear that temperature is not always sufficient in explaining short term fluctuations, as there are large variations in the emission potentials within the one-week averages.
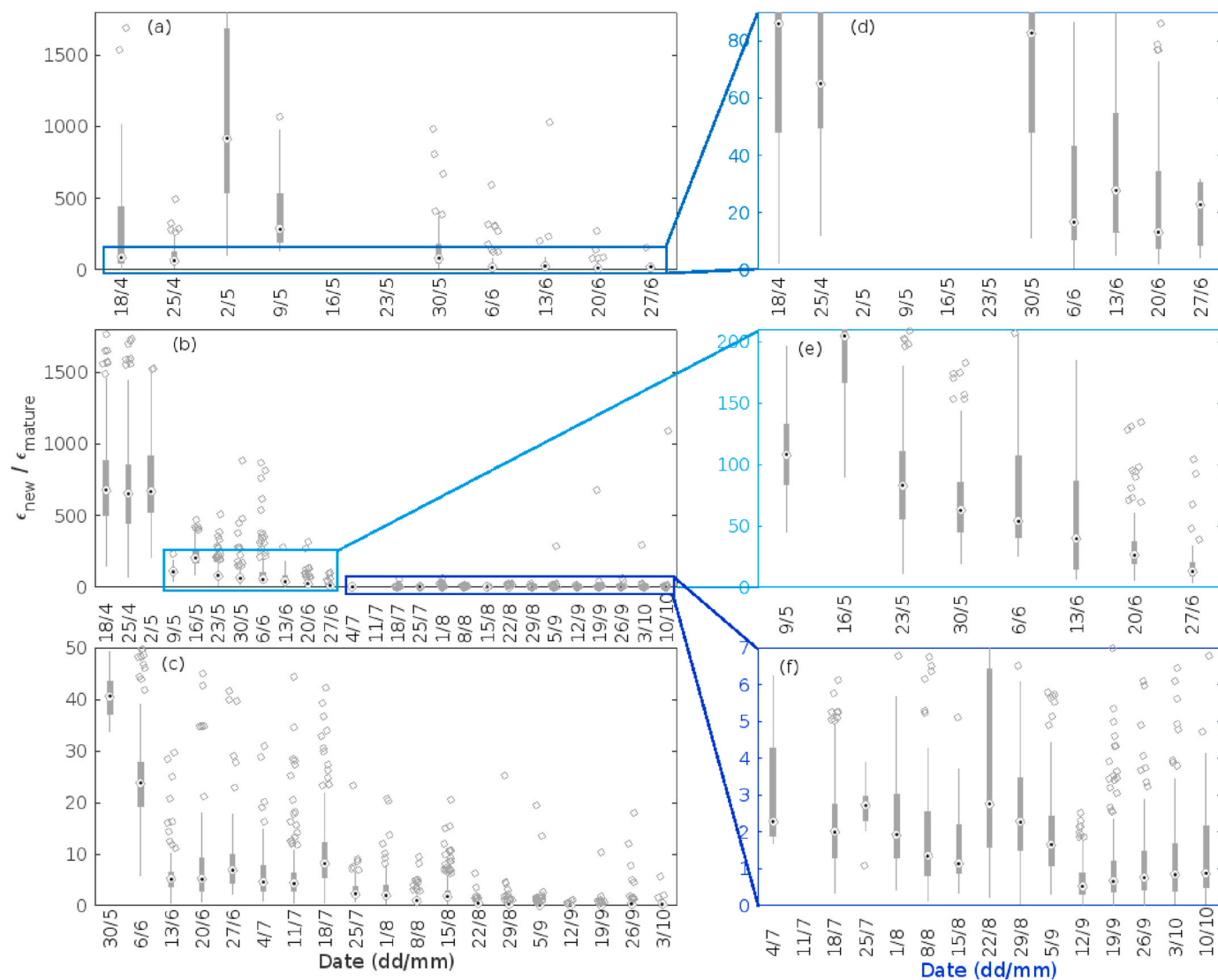
Fig. 3. Boxplot displaying the ratio of the emission potential of new needles to the emission potential of mature needles for years 2009 **(a, d)**, 2010 **(b, e, f)** and 2011 **(c)**. The date marks on the x axis indicate the middle points of the averaged periods. The subfigures have been cut due to clarity, but a list of the excluded outliers is found in Table A1. Note the different scales on the y-axis. The emission potentials are calculated based on the measurements presented by Aalto et al. (2014). Emission rates were obtained from one ~50 year old Scots pine tree at the SMEAR II station. Within one season, one mature shoot and one current year bud/shoot were measured, but during the next growing season, different shoots were chosen for the measurements. The emission potentials were standardised by Eq. (5) in Guenther et al. (1993) ($T_s = 30\ °C$, $\beta = 0.09\ °C^{-1}$). See Sec. 2.3.1 for more details.

2.3.2. Findings by Aalto et al. (2014) vs conclusions from other studies

In our analysis, we have only utilised data from Aalto et al. (2014) because there exists no other continuous long-term measurements of monoterpene emissions from different needle age classes simultaneously. However, measurement efforts have repeatedly demonstrated that there exist large intra-species variations in BVOC emission responses (e.g. Staudt et al., 2001; Bäck et al., 2012), and thus it is not certain that a similar seasonal pattern, as shown by Aalto et al. (2014), would be observed from other Scots pine individuals. Thus, in order to put findings by Aalto et al. (2014) into perspective and avoid drawing exaggerated conclusions in our study, the monoterpene emission potentials of new and mature Scots pine needles, calculated based on Aalto et al. (2014), are presented together with literature values, in Fig. 4. The literature values, which have also been standardised to 30 °C, represent different measurement years, locations, tree ages, needle ages, and measurement techniques (see Table A2). The requirement for including a study was that either the emission had been standardised to 30 °C or it was possible to (re)standardise it using the information provided in the paper. If the emission was not already standardised, a value of $\beta =$

0.09 $°C^{-1}$ was used as this is the most commonly used value in the literature for monoterpenes. The emission potentials used in MEGAN (Guenther et al., 2012) are not included in Fig. 4, because they have been standardised in a different way, and hence they cannot be directly compared to the potentials shown in the figure. For example, Langford et al. (2017) showed that the isoprene emission potential of oak might differ with up to a factor of four depending on which algorithm is used when standardising. Additionally, MEGAN provides emission potentials for plant functional types and not for individual tree species. According to Guenther (2013), the emission potentials of needle evergreen trees in MEGAN are partly based on literature values included in Fig. 4. Be aware that certain points in Fig. 4 represent only one measurement point, while most represent an average or median value based on a few measurement points, or e.g. in the case of Aalto et al. (2014), more than 100 or 200 data points.

The emission potentials of new foliage during spring and early summer, based on Aalto et al. (2014), are much greater than any other reported monoterpene emission potentials from Scots pine needles. The emission potentials, calculated from Aalto et al. (2014), of new needles
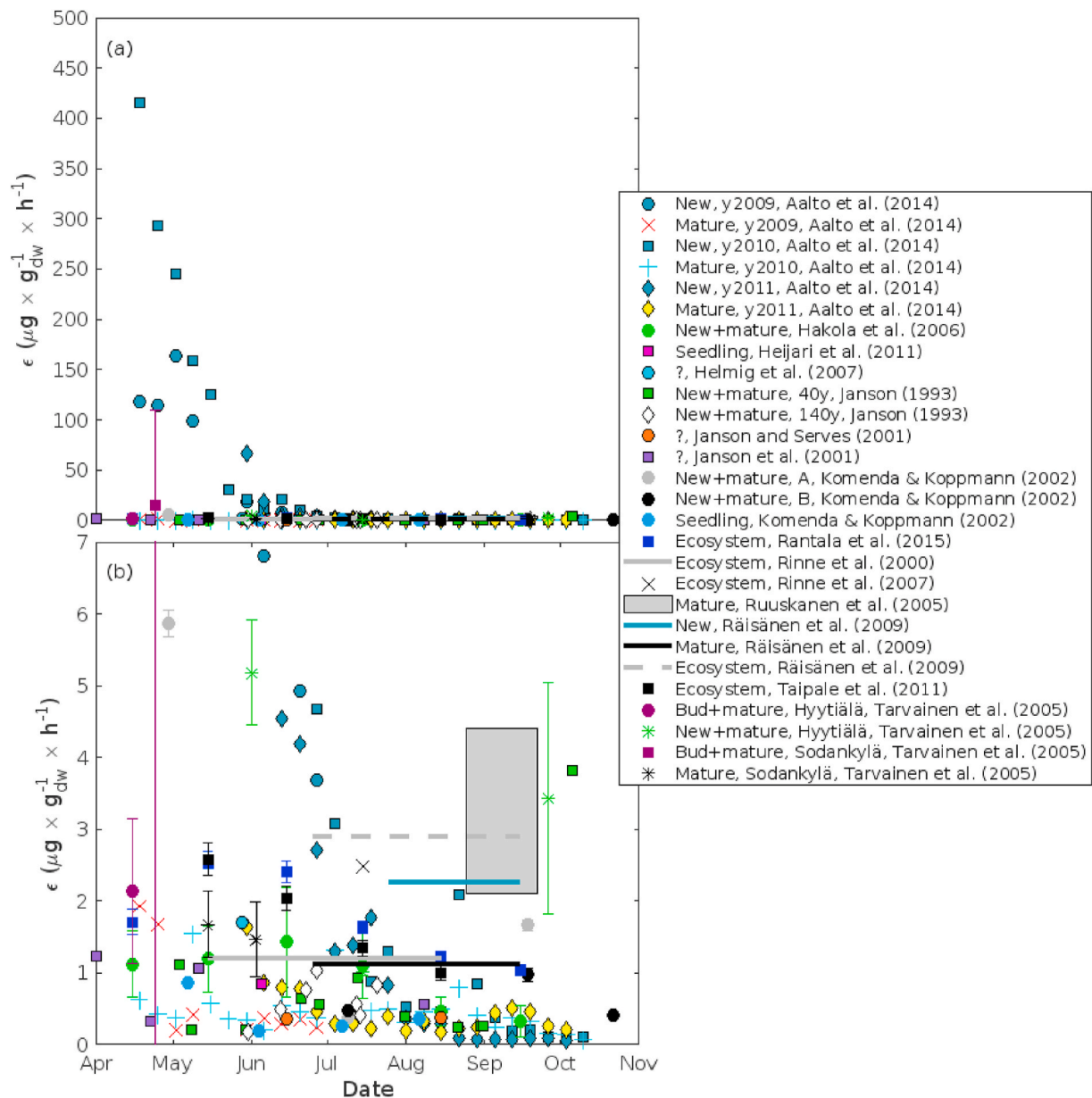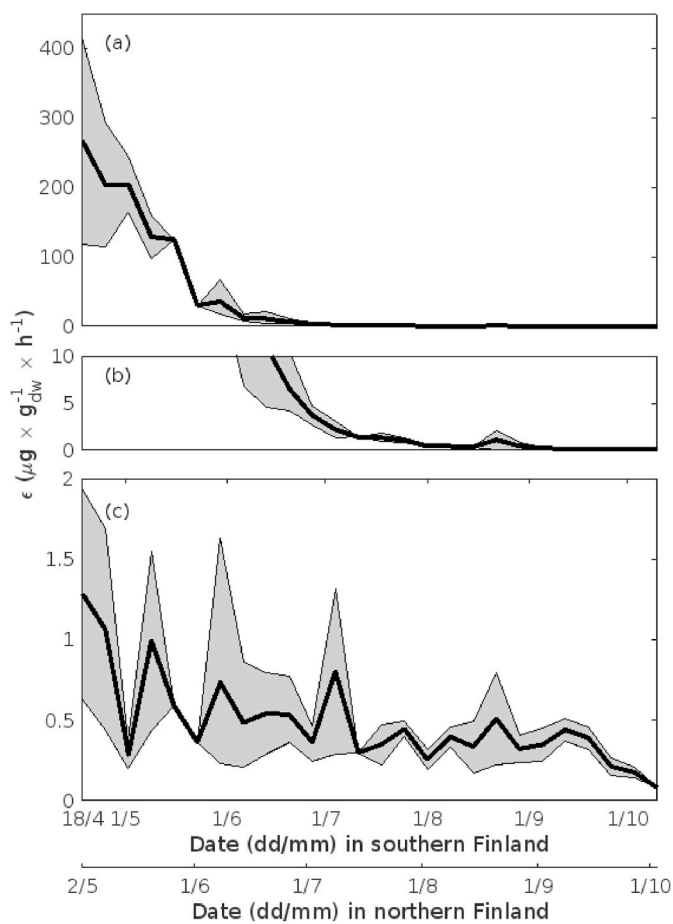
Fig. 4. The monoterpene emission potentials of Scots pine needles standardised to 30 °C (β = 0.09 °C⁻¹). **(b)** is a zoom of **(a)**, hence be aware of the different scales on the y-axis. Included in the figure are potentials calculated based on Aalto et al. (2014) together with other literature values (see Table A2). Literature values, which have been re-standardised to 30 °C, represent different years and locations (see Table A2). "New", "mature", "bud", "seedling" and "ecosystem" indicate that the emissions were measured from either new or mature needles, from buds or seedlings or as an ecosystem scale flux. A "?" indicates that no information was provided about the age of the measured needles, but it does not include measurements from seedlings nor the entire ecosystem. The added error bars to literature values are those that the respective authors reported. Sometimes error bars were not provided in the papers, and hence none are shown in the figure. Error bars are not added to the potentials calculated based on Aalto et al. (2014) due to clarity (see instead Fig. 3 for the variation). When the authors have only provided a seasonal emission potential, the value is indicated in the figure as a line that spans the period during which the authors measured the emissions. The emission potential reported by Ruuskanen et al. (2005) was reported as a range for the measured period, which is illustrated by the box in the figure. We refer to Table A2 for further details about the literature values used.

decrease throughout the season, while the corresponding potentials of mature needles stay largely the same, when they have decreased after the initial short peak (Figs. 4 and 5, Aalto et al. (2015)). Tarvainen et al. (2005) and Komenda and Koppmann (2002) also observed significantly higher monoterpene emission potentials from buds and new foliage, respectively, during spring, though not as large as Aalto et al. (2014). However, such a seasonal pattern is not detected in all studies (e.g. not in Janson, 1993 and Hakola et al., 2006). Räisänen et al. (2009), who measured the emissions from new and mature needles, individually, and without contributions from the woody parts of the branches, showed that the potential of new needles to emit monoterpenes is twice as high

as that of mature needles when calculated based on the dry mass of the needles. This is based on measurements from August–September, and is in accordance with findings by Aalto et al. (2014), who show that the difference in the potentials of the two needle age classes is about a factor of two in August (Fig. 3f). However, when Räisänen et al. (2009) determined their emission potentials based on needle surface, instead of needle dry mass, the authors did not find a significant difference in the emission potentials.

By far most literature values, which are based on enclosure measurements, are reported to be within ~0.1–2.3 μg g⁻¹ h⁻¹. This also includes the entirety of emission potentials of mature needles based on

Fig. 5. The monoterpene emission potentials of **(a)** new, and **(c)** mature Scots pine foliage as a function of the season in southern and northern Finland. **(b)** is a zoom of **(a)**. Note the different scales on the y-axis. Black curves are calculated as the means of the weekly medians from 2009 to 2011 (based on Aalto et al. (2014)). The grey areas illustrate the range of the emission potential. The lower and upper borders of the areas are calculated as the minima and maxima of the weekly medians of the three measurement years.

Aalto et al. (2014). A few points range up to ~6 μg g^{-1} h^{-1}, while only one measurement point results in a potential of ~15 μg g^{-1} h^{-1} when data based on Aalto et al. (2014) is not considered. These few high potentials are based on measurements during spring and autumn on branches where both new and mature foliage were present, or in one case, only mature needles (Ruuskanen et al., 2005). The exceptionally high value of ~15 μg g^{-1} h^{-1} originates from one measurement point of a mature shoot carrying buds (Tarvainen et al., 2005). The smallest reported potentials (~0.1 μg g^{-1} h^{-1}) are of new needles in the end of the growing season, and based on measurements by Aalto et al. (2014). The reported emission potentials of Scots pine seedlings are found in the lower end of the range (~0.2–0.9 μg g^{-1} h^{-1}), even though up to half of their needles are current year generation. However, the emissions from the seedlings were measured in the laboratory or in a research garden, and thus it is possible that the plants emit differently than plants growing in the field (Niinemets, 2010; Faiola and Taipale, 2020).

One reason for the discrepancy between the findings by Aalto et al. (2014) and previous chamber studies on Scots pines might be that other investigators, except Räisänen et al. (2009), have not measured the emissions from buds/growing needles and mature needles separately. And it might be very challenging to determine emissions from buds or growing needles, if the majority of needles inside the chamber are mature. Another reason might be that there is a larger uncertainty connected to the quantification of the biomass which was measured by

Aalto et al. (2014), since it is difficult to quantify the biomass of the stem and needles very accurately at any given point of time, when the elongating branch tips are measured continuously. Accurate biomass measurements would require the branch to be cut. As mentioned above, it is possible that the enhanced emissions observed by Aalto et al. (2014) do to some extent originate from the elongating (green) stem rather than from the needle primordia. If this is true, then the uncertainty connected to determination of the biomass within the closure is similar in other studies, since most other branch scale measurements have also included the stem tissue in the enclosures and also provide the emission rate per needle mass.

Five papers report ecosystem scale fluxes of Scots pine forests. Rinne et al. (2000) provide an ecosystem scale emission potential that is within the range reported from enclosure measurements (1.2 μg g^{-1} h^{-1}), while Rinne et al. (2007) and Räisänen et al. (2009) report values that are slightly higher than the general range (2.5 and 2.9 μg g^{-1} h^{-1}). The potential by Räisänen et al. (2009) is reported as a seasonal average (July - mid September) and is notably higher than the potentials based on Aalto et al. (2014) during the same time period. Canopy scale emission potentials by Taipale et al. (2011) and Rantala et al. (2015), which both measured in SMEAR II during separate years, are in a very good agreement with each other, though the micrometeorological method was different. Both studies observe a clear diminishment in the forest's potential to emit throughout the summer. The potential during April was, however, found to be less than during the summer months (Rantala et al., 2015), which can partly, but probably not fully, be attributed to the fact that the potential represents the entire month of April, while buds and new foliage are only contributing from mid April onwards.

2.3.3. Monoterpene emission potentials used in our calculations

We calculated the potential importance of new Scots pine foliage on total canopy monoterpene emission potential using the means of the weekly medians of the monoterpene emission potentials from 2009 to 2011, based on Aalto et al. (2014). In our investigations, we also considered the minima and maxima of the weekly medians of the monoterpene emission potentials from the three measurement years (Fig. 5). In order to conduct our analysis, we have to assume that this is representative for southern Finland. In order to approximate the influence of new Scots pine needles in northern Finland, we assumed that the potentials of needles to emit monoterpenes are similar in southern and northern Finland, but that they depend on timing of foliage growth. Since the foliage growth onset at the SMEAR I station is delayed by two weeks of that seen at the SMEAR II station, also the monoterpene emission values – both for mature and new foliage – were delayed accordingly (Fig. 5). Since needle growth has been observed to end about one week earlier in northern than southern Finland (Fig. 2), the seasonally dependent emission potentials of northern Finland have been modulated likewise, thus, the emission potentials have been adjusted to fit the more intensive, but (~three weeks) shorter period of growth in the north (Fig. 5). The presumption that the potential of the foliage to emit monoterpenes is similar in southern and northern Finland is naturally connected with some degree of uncertainty, since observations from new needles in the north are limited, but nevertheless supported by previous investigations on Scots pine (Tarvainen et al., 2005) and silver birch (Maja et al., 2015) in Finland. Finally, we assumed that all mature needles have the same potential to emit monoterpenes independent of their needle age class. Though Scots pine foliage preserves its ability to emit monoterpenes after a completed growing season (Vanhatalo et al., 2018), we only focus on the period of growth, as our interest lies in the difference that new and mature foliage presents. This difference diminishes by the end of the growing season, as the potentials to emit are then similar for all needle age classes. Observations from SMEAR I and II were utilised due to data availability and in order to provide estimates across a latitudinal gradient.

2.3.4. Traditional approach: canopy emission potential with MEGAN algorithm

In our analysis, we compared the canopy emission potential resulting from Aalto et al. (2014) with a canopy emission potential that assumes that the emission potential of current year needles is enhanced in a similar manner as in Guenther et al. (2012). This "MEGAN style" canopy emission potential has been calculated as:

$$\varepsilon_{canopy,MEGAN\ style} = \varepsilon_{mature} \times F_{mature} + \varepsilon_{growing,MEGAN} \times F_{new} + \varepsilon_{new,MEGAN} \times F_{bud} \tag{3}$$

where $\varepsilon_{new,MEGAN}$ and F_{bud} are the emission potential and fraction of new foliage before needle elongation properly starts, respectively, while $\varepsilon_{growing,MEGAN}$ and F_{new} are the emission potential and fraction of new foliage during the period with a significant needle elongation rate, respectively. $\varepsilon_{mature,MEGAN}$ and F_{mature} are the emission potential of mature foliage and fraction of mature foliage, respectively. Using the coefficients from Guenther et al. (2012, Table 4) that describe the relative emission rates of buds, growing and mature foliage, Eq. (3) can be reformulated to:

$$\varepsilon_{canopy,MEGAN\ style} = \varepsilon_{mature} \times F_{mature} + 1.8 \times \varepsilon_{mature} \times F_{new} + 2 \times \varepsilon_{mature} \times F_{bud} \tag{4}$$

which can be shortened to:

$$\varepsilon_{canopy,MEGAN\ style} = \varepsilon_{mature} \times (1 + 0.8 \times F_{new} + F_{bud}) \tag{5}$$

since we did not consider periods with senescing needles. In our calculations, ε_{mature} is from Fig. 5c, while F_{new} and F_{bud} are from Fig. 2c. F_{bud} is the fraction of new foliage until ~13th of May in southern Finland (Fig. 2c and Aalto et al., 2014, Fig. 3b) and until ~27th of May in northern Finland (Fig. 2c). F_{new} is then the fraction of new foliage during 13/5–29/7 in southern Finland (Fig. 2c and Aalto et al., 2014, Fig. 3b) and during 27/5–26/7 in northern Finland (Fig. 2c).

2.4. Scots pine forest stand coverage in Finland

We utilised the coverage of Scots pine forests in Finland of different tree age classes (Fig. 6) from the Finnish Statistical Yearbook of Forestry (2014) (page 59, Table 1.13, Whole country, National Forest Inventory 11 (years 2009–2013), Pine dominated). The presented total area (12.931×10^6 ha) only includes Scots pine trees present on forest land,
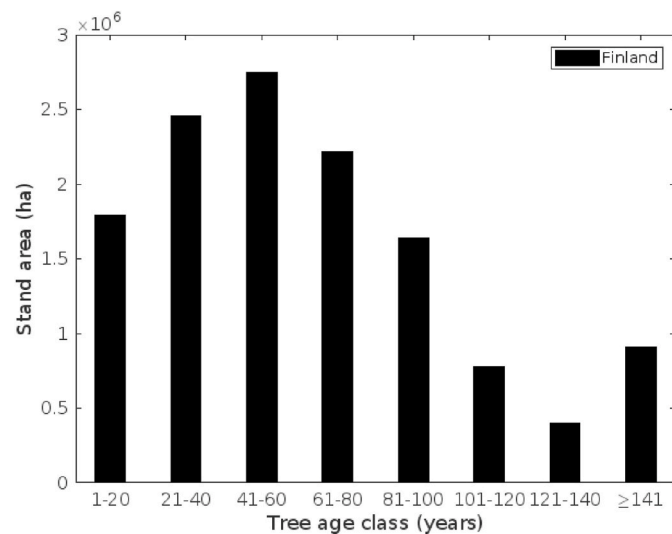

Fig. 6. Scots pine forest stand area in Finland expressed as a function of tree age. Data from Finnish Statistical Yearbook of Forestry (2014) (page 59, Table 1.13, Whole country, National Forest Inventory 11 (years 2009–2013), Pine dominated).

hence Scots pines growing on poorly productive forest land (~12% of forest land in Finland, Finnish Statistical Yearbook of Forestry 2014) are not accounted for, since no data is available. The coverage of Scots pine on forest land is 6.064×10^6 ha in southern Finland and 6.867×10^6 ha in northern Finland (Finnish Statistical Yearbook of Forestry, 2014). In our calculations, we assumed that there is an even distribution of trees of all ages within each tree age class (Fig. 6). Hence, within the first tree age class (1–20 years), we excluded 45% of the stand area, as it is assumed to be covered by trees aged 1–9 years.

3. Results and discussion

3.1. The emission potentials of new and mature Scots pine foliage as a function of season

Though the emission potential of new foliage is high, the corresponding biomass is low. Hence, in order to investigate the potential importance of new foliage to the whole tree's foliage emission potential, the products of the emission potentials of new (ε_{new}) and mature (ε_{mature}) foliage, respectively (Fig. 5), and the fractions that new (F_{new}) and mature (F_{mature}) foliage make of the total foliage, respectively (Fig. 2c), are compared ($\varepsilon_{new} \times F_{new}$ vs $\varepsilon_{mature} \times F_{mature}$) as a function of season, for trees of different ages and locations (Fig. 7). The high emission potential of new foliage counters the small mass of developing buds and needles in spring, and consequently new Scots pine foliage can be responsible for the majority of the whole tree's foliage emissions of monoterpenes during spring time, independently of tree age and location. In our estimations, new Scots pine foliage then generally accounts for ~80–90% of the total monoterpene emission potential of Scots pine trees of various ages growing in southern Finland, while the corresponding contribution is ~60–75% in northern Finland, though at times it could be even higher. Though the new foliage biomass increases as the season progresses, the very high new foliage emission potential collapses in the beginning of the summer (Fig. 5), and the importance of the emissions from new Scots pine foliage therefore decreases as a function of the season (Fig. 7). The contribution of new Scots pine foliage to the whole tree's emissions decreases with tree age (Fig. 7), because the proportion of new foliage of the total stand foliage mass decreases with an increase in tree age (Fig. 2c). Likewise, new foliage accounts for a larger fraction of the total Scots pine monoterpene emissions in southern than in northern Finland (Fig. 7), where needles are preserved for a longer time (Fig. 2c). We appreciate that the extrapolation of the emission potentials based on Aalto et al. (2014) to trees of different ages and for making estimates of trees growing in northern Finland can be very uncertain due to lack of observations.

3.2. The potential importance of new foliage to the whole Scots pine tree's foliage emission potential

The canopy emission potentials ($\varepsilon_{new} \times F_{new} + \varepsilon_{mature} \times F_{mature}$), as a function of season for trees of various ages and locations, are compared, in Fig. 8, to (1) the emission potentials of mature foliage (ε_{mature}, Fig. 5c), as several widely used models (e.g. LPJ-GUESS and ORCHIDEE) assume that the monoterpene emission potential is independent of needle age, and (2) canopy emission potentials that assume that the emission potentials of current year needles are enhanced in a similar manner as in Guenther et al. (2012) (see Sec. 2.3.4 for how this was calculated). We did not directly compare our canopy emission potentials to the potentials utilised in global BVOC models, as they do not use the same values, they do not utilise tree species specific, but instead plant functional type specific emission potentials, and often they assume some dependency on light. It is possible that models will greatly underpredict canopy emissions during the first ~2.5 months of the growing season in southern Finland if they assume that the monoterpene emission potential is independent of needle age or that the emission potential of new foliage is enhanced in a similar manner as in Guenther et al. (2012)
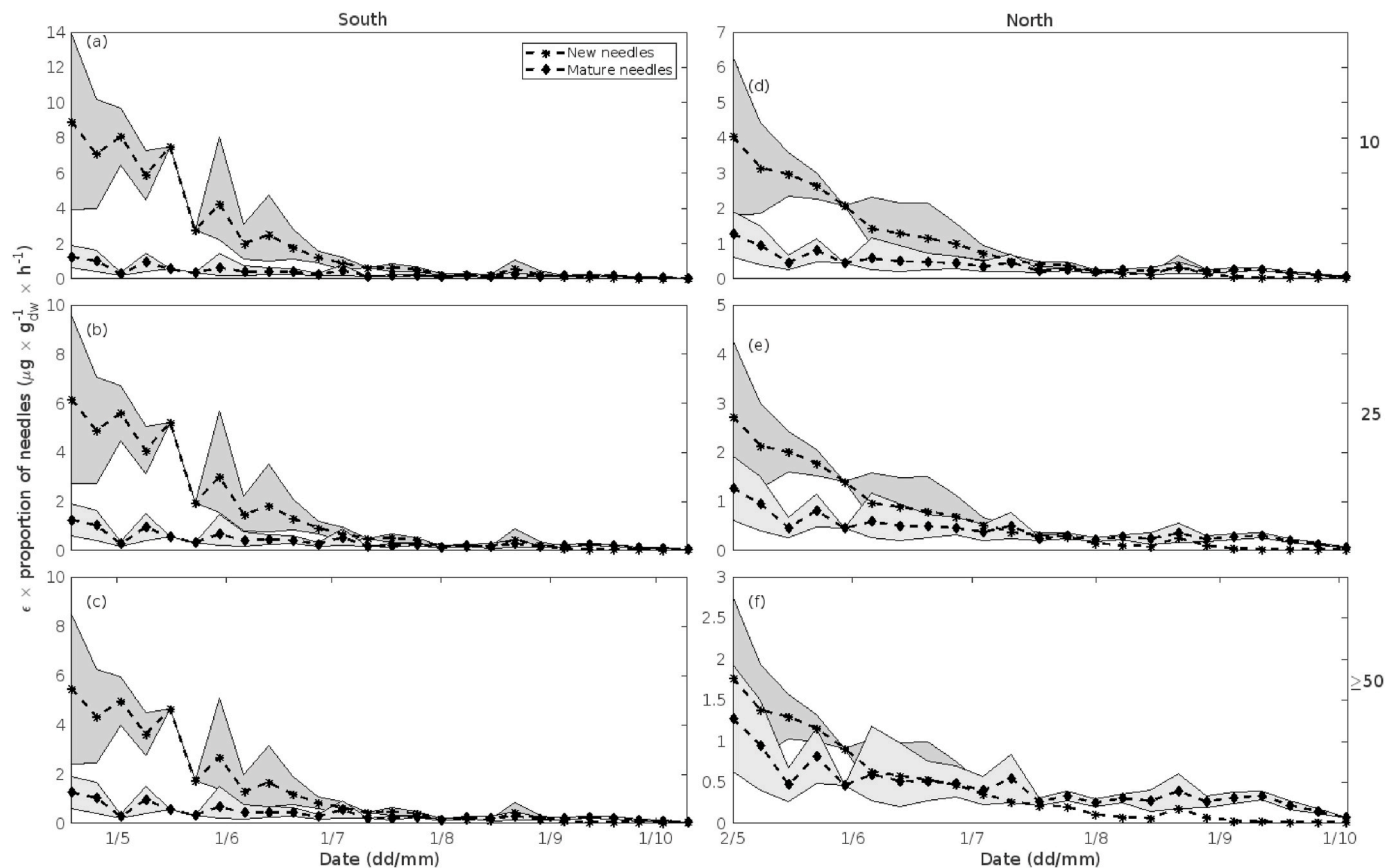
Fig. 7. The emission potentials of monoterpenes multiplied by the fraction of either new (black stars) or mature (black diamonds) needles for Scots pines of different ages (**a+d**: 10 years, **b + e**: 25 years, **c + f**: ≥50 years) and locations (**a-c**: southern Finland, **d-f**: northern Finland). The grey areas illustrate the ranges caused by interannual variations in the emission potentials (Fig. 5). Dark grey areas represent the range for new needles, while light grey areas indicate the range for mature needles. Be aware that the y-axis changes between the different subplots.

(Fig. 8). The estimated underestimation will be less severe for predictions of emissions from northern than from southern Finland (e.g. up to a factor of ~7 vs ~29 for 10 year old forest), and more severe for younger than older stands (e.g. up to a factor of ~29 vs ~19 for 10 vs ≥ 50 year old forest in southern Finland, Fig. 8). After ~1st of July, the estimated underestimation in the canopy emission potential of Scots pine growing in southern and northern Finland is less than a factor of 2.5 and 2, respectively. Assuming that the emission potential of new needles is enhanced as in Guenther et al. (2012) will only lead to a neglectable increase in the Scots pine canopy monoterpene emission potential (Fig. 8).

Canopy scale emission potentials by Taipale et al. (2011) and Rantala et al. (2015), derived from continuous micrometeorological flux measurements of a ~50 year old pine forest in SMEAR II, are included in Fig. 8c for comparison. We appreciate that the measured canopy, within an area with a radius of 200 m, is only covered by ~75% Scots pine (and ~25% other tree species). Thus our results cannot be directly compared to Taipale et al. (2011) and Rantala et al. (2015), but these two studies provide the most suitable observations for validation of our results. Be also aware that data from April from Rantala et al. (2015) represents the measured flux during the entire month, also before buds and elongating needles contribute to the emission. We refer to Table A2 in the Appendix for details on how these potentials (per ground area) have been converted (to per foliage mass). The reported canopy scale emission potentials agree very well with our suggested whole tree foliage emission potentials and the agreement is much better than that between Taipale et al. (2011) or Rantala et al. (2015) and assuming that the emission potential is independent of needle age or that the potential of new foliage is enhanced as in Guenther et al. (2012). Our enclosure-derived

canopy emission potential overestimates the canopy micrometeorological-derived potential by a factor of ~1.6 during May, and then slightly underestimates it during the summer. The overestimation can partly be due to interannual variations in emission rates and seasonal foliage mass development, and partly due to plant-to-plant variations (as rates by Aalto et al. (2014) were conducted on one tree). An underestimation during summertime is expected, since the emission potentials by Taipale et al. (2011) and Rantala et al. (2015) consider all sources of monoterpenes in the ecosystem, and not only Scots pine foliage. These additional sources include at least Scots pine stems, forest floor, understory vegetation, Norway spruce (15% of the stand) and deciduous species (~10%) (Bäck et al., 2010; Aaltonen et al., 2011, 2012; Vanhatalo et al., 2015; Mäki et al., 2019; Rissanen et al., 2020).

3.3. Effects of stand age and season on the estimated underestimation of the whole Scots pine tree's foliage emission potential

The estimated underestimation of the whole Scots pine tree's needle emission potential caused by not considering the enhanced potential of new foliage, is presented in Fig. 9 as a function of tree age, for southern and northern Finland separately. The ranges in the estimated underestimation are provided in Table A3. The estimated underestimation has been calculated individually for the spring and for the full season, since new particle formation events have been shown to occur more frequently during March–May in both southern and northern Finland (Vehkamäki et al., 2004; Dal Maso et al., 2005, 2007; Manninen et al., 2010; Nieminen et al., 2014; Vana et al., 2016). Hence, in our calculations, spring starts at the same time as emissions from new foliage is observed and lasts until the end of May, while the full season naturally
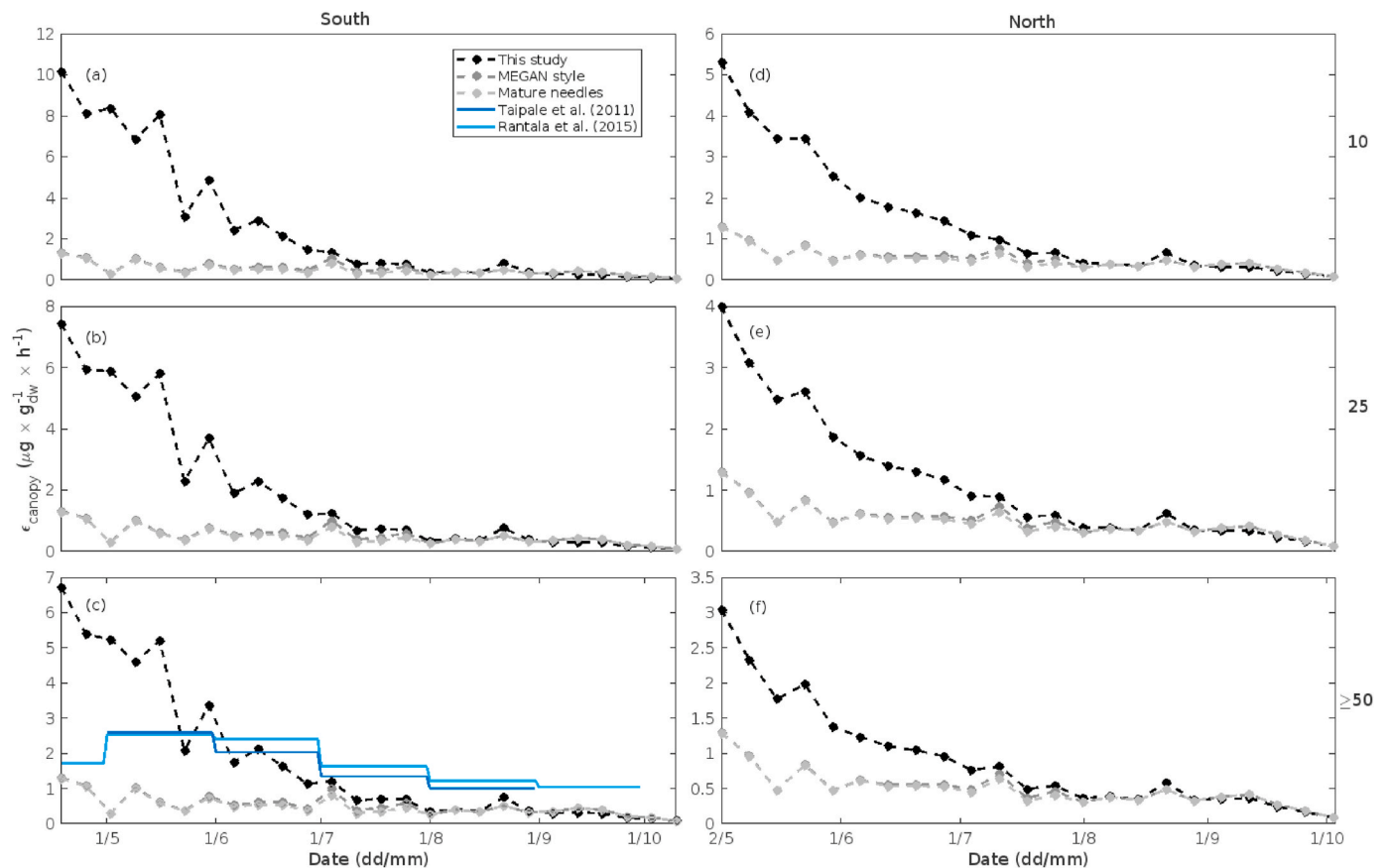
Fig. 8. (a–f) The monoterpene emission potential of Scots pine canopies of various ages and locations. The canopy emission potentials are illustrated for Scots pine stands aged 10 (a+d), 25 (b + e) and ≥50 (c + f) years old, growing in southern (a–c) or northern (d–f) Finland. "MEGAN style" assumes that the emission potentials of buds and growing needles are 2 and 1.8 times that of mature needles, respectively (see Sec. 2.3.4), while "Mature needles" presume that the emission potential is independent of needle age. Canopy emission potentials for a ~50 year old Scots pine forest derived from micrometeorological flux measurements by Taipale et al. (2011) and Rantala et al. (2015) are included for comparison in **c**. Ranges of the whole foliage emission potential are not included in this figure due to clarity, instead we refer the reader to Fig. 7 for an idea about the range. Please pay attention to changing scales on the axes.

includes the entire measurement period. Trees aged less than 10 years are excluded from our analysis, as it might not be reasonable to extrapolate conclusions extracted from emission rate measurements of a ~50 year old tree to very young trees. For example, Komenda and Koppmann (2002) showed that the emission potential of a 40 year old Scots pine tree was about five times higher than that of 3–4 year old seedlings. It should, however, be mentioned that measurements of seedlings were conducted in laboratory conditions, thus the difference in emission potential between seedlings and mature trees might be less.
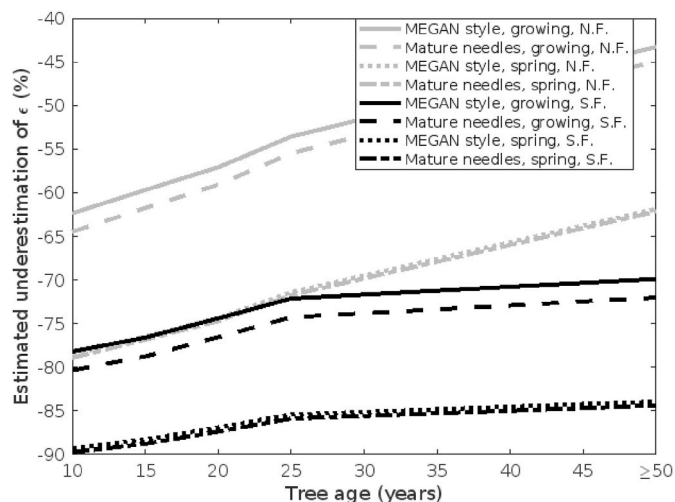
The estimated underestimation caused by not considering the enhanced emissions from new foliage during the entire growing season in southern Finland is similar to not accounting for the greater emissions from new needles during the spring in northern Finland, especially in the cases of younger Scots pine tree stands. An additional important conclusion from Fig. 9 is that it seems that neglecting the age of the stand only leads to a minor error if the longevity of needles is short (max ~8%), but to a larger error if more needle age classes prevail (max ~20%). This is because the relative proportion of new needles in stands that carry more needle age classes varies more between individual stands of different ages (Fig. 2c). Tree age is not usually considered specifically in BVOC models, instead only the biomass and/or leaf area index is/are included.

The spring time differences in emission potentials can lead to uncertainties in predictions of monoterpene emissions that are much greater than what has been estimated by Lamb et al. (1987) and Guenther et al. (2012). These investigators have estimated that the uncertainty on annual global emissions of monoterpenes into the

atmosphere could be around a factor of three in total, with about 15–25% of that uncertainty attributed to emission potentials (Lamb et al., 1987; Guenther et al., 2012). Guenther et al. (2012) emphasis that these uncertainties are estimated for *annual global* emissions, thus the uncertainty can be much greater for specific times and locations. Though the emissions from Scots pine species have been extensively measured, emissions during spring time have only relatively recently received more appropriate attention, thus it is reasonable to assume that model estimates of spring time Scots pine BVOC emissions are connected with a larger-than-average uncertainty.

3.4. Potential national level impacts caused by omitting the enhanced emissions from new Scots pine foliage

About 12.931 × 10⁶ ha in Finland, i.e. ~43% of the total land area in Finland, is covered by Scots pine forests (Finnish Statistical Yearbook of Forestry 2014). Hence, the estimated underestimation of not considering the emission potential of new Scots pine foliage (Fig. 9) is upscaled to Finland in Fig. 10. This has been estimated by (1) calculating the mean of the estimated underestimation shown in Fig. 9 within the respective tree age classes provided in Fig. 6 and (2) normalising the product of the mean foliage biomass (Fig. 1a) within each tree age class (Fig. 6) and the stand area within each tree age class (Fig. 6). For this calculation, we have assumed that there is an even distribution of trees of all ages within each tree age class, and we have excluded the fraction of trees younger than 10 years old. Hence, it is assumed that there is no underestimation connected with the emission potential of Scots pine

Fig. 9. The estimated underestimation of the whole Scots pine tree's needle emission potential caused by not considering the enhanced potential of new foliage, presented as a function of tree age. The estimated underestimation has been calculated as: (the integral of "other study" - the integral of "This study")/ the integral of "This study", where "other study" is either "MEGAN style" or "Mature needles" and the integrals are the areas under the curves presented in Fig. 8. The estimated underestimation has been calculated for the spring and for the growing season separately and for both southern (S.F.) and northern (N.F.) Finland. Ranges in the estimated underestimation are not indicated in the figure due to clarity, but they are provided in Table A3.
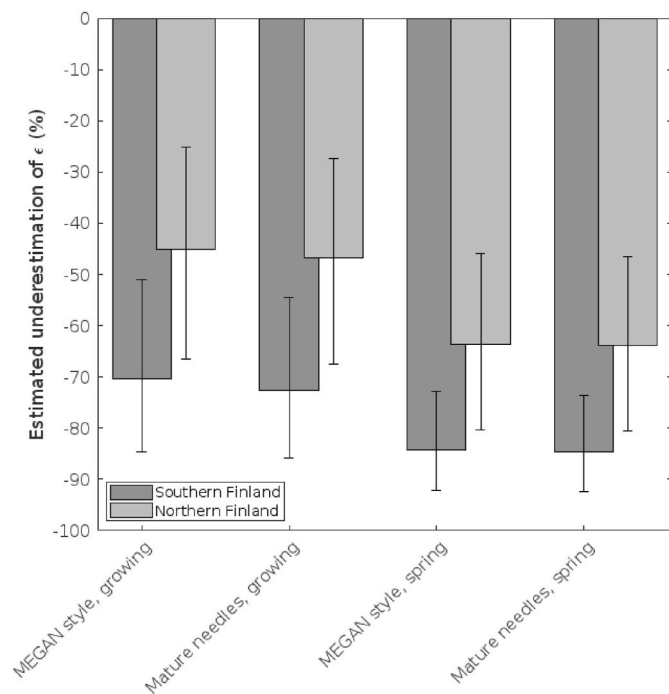
Fig. 10. The estimated underestimation of the whole Scots pine tree's needle emission potential caused by not considering the enhanced potential of new foliage, upscaled to Finland. The estimated underestimation has been calculated for the spring and full growing season separately, and for southern and northern Finland, separately. Errorbars are based on the interannual variations in the emission potentials (Figs. 5 and 7).

forest aged less than 10 years. The results presented in Fig. 10 only refer to potential underestimations in the emission potentials of Scots pine dominated areas and not to a general emission potential that would be representative for the entire Finland and hence also consider e.g.

Norway Spruce and various deciduous species. In our estimate, the national scale uncertainty is controlled by the uncertainty connected to trees aged $\geq$50 years, because the majority of trees in Finland are older than 50 years and their foliage mass is larger than that of younger trees. Thus, it seems largely unnecessary to include a tree age dependent emission potential for regional or global annual calculations of BVOC emissions. However, an exclusion will lead to an error of up to 20% in simulations of specific locations.

4. Implications

4.1. Emission potentials used in models

We emphasize that, in this study, we have not investigated how much MEGAN, LPJ-GUESS, ORCHIDEE or any other model could be underestimating the potential of Scots pine canopies to emit monoterpenes. This would, first of all, be inappropriate considering the fact that our estimations are based on a single measurement study. Secondly, it would largely be impossible, as it is not entirely transparent how models attain the emission potentials of their plant functional types. The sources of literature are provided in the model description, but often it is unclear if the plant functional type emission potentials are then an average of the considered literature or if there has been given consideration to tree species distributions. Such information is vital, since Scots pine was the sole focus of our study. Additionally, it is also unclear how literature values, which are most often standardised to either 25 or 30 °C, are re-standardised to also depend on light, when no information about light is provided in the literature sources. Instead, we have explored how such treatments of the emission potential, which are used in models, can lead to a potential underestimation. As ecosystem scale flux measurements become increasingly available, such data is progressively being incorporated into biogenic VOC emission models. This is fortunate, since such measurements capture the entire emissions from the ecosystem. Unfortunately, such measurements are most often conducted in summer. Thus, if the potentials they produce are not modulated by the seasons in models, a similar underestimation persists.

According to Guenther (2013), the emission potentials of Needleleaf Evergreen Boreal Trees in MEGAN v2.1 are based on enclosure and canopy micrometeorological measurements and landscape inverse modelling of various boreal forest species. However, almost all measurements of Scots pine utilised for compiling the monoterpene emission potential are enclosure measurements (Guenther, 2013). Results by Taipale et al. (2011) and Rantala et al. (2015) are not considered in MEGAN v2.1, at least in the latter case due to its (more) recent publication date. Micrometeorological measurements by Rinne et al. (2000, 2007) and Räisänen et al. (2009) are considered (Guenther, 2013), but these measurements were mainly conducted during summer time. The monoterpene emission potential of the boreal needleleaf evergreen tree type in ORCHIDEE is extracted from the corresponding emission potentials used in Guenther et al. (2006, 2012), and otherwise exclusively from literature on enclosure measurements when Scots pine is concerned (Messina et al., 2016). LPJ-GUESS by far mostly considers enclosure measurements for construction of their emission potentials, but as in the case of MEGAN, also ecosystem scale fluxes from Rinne et al. (2000) are used (Schurgers et al., 2009).

Monoterpenes are not the only atmospherically relevant VOCs that have been shown to be emitted in substantially greater quantities from new than mature Scots pine needles (Aalto et al., 2014). For example, Aalto et al. (2014) showed that the emissions of methanol, acetone and 2-methyl-3- buten-2-ol from developing needles can contribute with up to about 50, 35, and 75%, respectively, of the whole tree foliage emissions in case of a ~50 year old Scots pine stand. It is also possible that emerging foliage of other evergreen, conifers tree species would have a similarly much higher potential to emit VOCs than its corresponding mature foliage, as found in Aalto et al. (2014). If that is in fact the case, the impact of new needle emissions might be even larger than for Scots

pine forests, because many evergreen trees, especially in tropical regions, have several needle cohorts flushing annually. Since evergreen trees are dominating in many ecosystems around the world, more measurements are required in order to improve the representation of the emissions of atmospherically important VOCs from new evergreen conifers foliage in models.

4.2. Potential impacts on monoterpene emission predictions from Finland

The potential error of not accounting for new foliage monoterpene emissions in the canopy's emission potential translates directly into the predicted emission rates, as emission potentials are multiplied with various activity factors in models in order to produce the emission rates (e.g. Guenther et al., 2006, 2012). Thus, under the same environmental conditions and foliage mass or leaf area index, a change in the emission potential leads to a proportional change in the predicted emission rate (F):

$$\Delta F \propto \Delta \varepsilon \tag{6}$$

We investigated how many Gg of monoterpenes the emissions from Finland could be underestimated, if biogenic emission models only consider the emissions from mature foliage. For this analysis, we utilised Eq. (5) in Guenther et al. (1993) and considered the tree age (i) and time (j) dependant foliage mass per area (M, Fig. 2b) and the tree age dependant Scots pine stand area (A, Fig. 6):

$$\Delta F = \sum \left(\varepsilon_{new+mature,i,j} - \varepsilon_{mature,j} \right) \times exp\left(\beta \times \left(T_j - T_s \right) \right) \times M_{i,j} \times A_i \tag{7}$$

together with weekly averaged air temperature (T) during 2014–2018 at the SMEAR II (16.8 m, Aalto et al., 2019a) and SMEAR I (9 m, Aalto et al., 2019b) stations. In our calculations, it is assumed that the temperature of all needles equals the ambient temperature, which is a reasonable assumption for low density canopies (Pier and McDuffie, 1997; Martin et al., 1999; Zweifel et al., 2002; Leuzinger and Körner, 2007). T_s and β are the same as in Sec. 2.3.1. Eq. (7) considers our suggested canopy scale emission potentials (Fig. 8) and our emission potential of mature needles (Fig. 8). Our estimate suggests that about 27 Gg of monoterpenes could be additionally emitted from Finnish Scots pine forests yearly, if the enhanced emissions from new foliage are explicitly considered. The majority of these additional emissions, namely ∼23 Gg, originate from southern Finland. This is partly due to higher temperatures in the south (the difference in the weekly averaged temperature between SMEAR I and II was 3.1 °C during the investigated period), but mostly caused by a smaller production of new foliage in the north. The areas covered by Scots pine are almost identical in southern and northern Finland (Finnish Statistical Yearbook of Forestry 2014, Table 1.12).

The estimate of how many Gg of monoterpenes the emissions from Finland could be underestimated, is compared to several studies that have predicted the emissions of monoterpenes for Finland using different models and methods, in Table 1. Please be aware that these estimates consider emissions from all terrestrial land covers in Finland, and not only from Scots pine forests, except in the case of Kellomäki et al. (2001). Though Scots pine is the dominant forest species in Finland (∼65% coverage of forest land), Norway spruce and broadleaved species make up significant fractions of the forest land (∼25% and ∼10%, respectively, Finnish Statistical Yearbook of Forestry 2014).

Our estimate of emitted monoterpenes from new Scots pine foliage is comparable to Kellomäki et al. (2001)'s estimate of monoterpenes emitted from the complete Scots pine foliage in Finland. Other studies estimate that the emissions of monoterpenes from all forest types in Finland sum up to 105–230 Gg/yr, with all except one study ranging the emission to 105–160 Gg/yr. Though our estimate of additionally emitted monoterpenes is within the range covered in the literature, the addition is still very significant and in some cases corresponds to about 25% of the total monoterpene emission estimate from Finland.

Table 1

Other studies that have estimated the emissions of monoterpenes for Finland using different models and methods. Be aware that these values do not only cover the emissions from Scots pine, but all terrestrial land cover, unless otherwise specified.

Study	Monoterpene emission (Gg/yr)	Notes
Kellomäki et al. (2001)	30.3 (southern Finland: 15.9, northern Finland: 14.4)	These values are only for Scots pine and calculated using the total annual monoterpene emissions given in Kellomäki et al. (2001) Table 4 and multiplied by the Scots pine land cover in southern and northern Finland, respectively (Finnish Statistical Yearbook of Forestry 2014, Table 1.12).
Lindfors and Laurila (2000)	150	
Lindfors et al. (2000)	160	
Oderbolz et al. (2013)	105, 145, 230	The three different values listed correspond to three different vegetation inventories used for model simulations.
Simpson et al. (1999)	160	
Tarvainen et al. (2007)	110	

4.3. Potential impacts on predictions of new particle formation and growth

BVOCs, and especially monoterpenes, have been shown to participate in the formation (Kulmala et al., 1998, 2014; Donahue et al., 2013; Riccobono et al., 2014; Schobesberger et al., 2013) and growth (Ehn et al., 2014; Riipinen et al., 2012) processes of the climatically important secondary organic aerosol particles in the atmosphere. Though boreal forests are globally a small emitter of BVOCs when compared to e.g. the tropics (e.g. Guenther et al., 2012; Guenther, 2013), the ambient blend of BVOCs in boreal forests is, in contrast to e.g. the Amazonian rainforest, favorable for production of new particles (e.g. Lee et al., 2016). Previous studies from sites in the boreal forest indicate for example that 12–50% of aerosol mass and 50% of the climatically relevant cloud condensation nuclei originate from forests (Tunved et al., 2008; Sihto et al., 2011). In the specific case of Finland, it has been estimated that particle formation causes a local radiative perturbation of between −5 and −14 Wm^{-2} (global mean −0.03 to −1.1 Wm^{-2}) (Kurtén et al., 2003). As already stated earlier, the frequency of new particle formation events in boreal forests have been observed to be highest during spring time. We, therefore, extrapolate our results in order to assess the potential impact that an exclusion of the enhanced emissions of monoterpenes from new Scots pine foliage during spring time can have on predictions of formation and growth of small new particles in locations without measurements, or predictions of future climate.

As stated in Sec. 4.2, a change in the emission potential is proportional to a change in the (predicted) emissions under the same environmental conditions. Under the same boundary layer conditions, a change in the emissions of monoterpenes is largely proportional to a change in the atmospheric concentration of monoterpenes (MT), and hence in the concentration of oxidised organics (org), if the change in the concentrations is not extreme (see e.g. Smolander et al., 2014):

$$\Delta F \quad \propto \quad \sim \Delta [MT] \quad \propto \quad \sim \Delta [org] \tag{8}$$

The calculated canopy scale emissions of monoterpenes during spring time increase with 180% in northern Finland and by 560% in southern Finland, when the emission potentials of both new and mature foliage are considered, and compared to the situation when only the emission potential of mature needles is included. This has been

calculated as: (the integral of "This study" - the integral of "Mature needles")/the integral of "Mature needles", where the integrals are the areas under the curves presented in Fig. 8 during the spring time period. The values are therefore also different to Fig. 10, since those have been calculated as: (the integral of "Mature needles" - the integral of "This study")/the integral of "This study".

The formation of neutral 2 nm sized clusters, J_2, from sulfuric acid (H_2SO_4) and oxidised organic compounds can be expressed as follows (Paasonen et al., 2010):

$$J_2 = K_{s1} \times [H_2SO_4]^2 + K_{s2} \times [H_2SO_4] \times [org] + K_{s3} \times [org]^2 \qquad (9)$$

where K_{s1-3} are kinetic coefficients. The condensational growth rate, GR, of 2–3 nm particles can be calculated as follows (Nieminen et al., 2010):

$$GR = 0.5 \; nm \cdot h^{-1} \times CC \times 10^{-7} \; cm^3 \qquad (10)$$

where CC is the concentration of condensable vapours, which we assume to be the sum of sulfuric acid and organics. We assume that the molar mass of organics is four times higher than that of sulfuric acid (Ehn et al., 2014) and hence we can write:

$$GR = 0.5 \; nm \cdot h^{-1} \times \left([H_2SO_4] + [org] \times 4^{1/3} \right) \times 10^{-7} \; cm^3 \qquad (11)$$

Changes in the formation and growth rate depend on the absolute concentrations of sulfuric acid and oxidised organics. Hence, we have calculated the impact on formation and growth rates utilising sulfuric acid concentrations of $1 - 10 \cdot 10^6 \; cm^{-3}$ and concentrations of organic condensables of $1 - 5 \cdot 10^7 \; cm^{-3}$, which are reasonable ranges according to measurements of sulfuric acid and estimates based on observations of growth rates, respectively (Paasonen et al., 2010). The increase in the formation and growth rates are calculated in a similar manner as in the case of the emissions: (Y1–Y2)/Y2×100%, where Y1 = emission, formation or growth rate considering the emission potential of both new and mature needles, and Y2 = emission, formation or growth rate considering only the emission potential of mature needles. In our calculations, we assume that simulations including the emission potential of both new and mature Scots pine foliage would lead to concentrations of organic condensables in the range of $1 - 5 \cdot 10^7 \; cm^{-3}$. Thus, [org] is decreased by a factor of 2.8 (northern Finland) and 6.6 (southern Finland) in the calculations of the formation and growth rates using only the mature foliage emission potential. The resulting changes in the formation and growth rate are presented in Table 2 and illustrated in Fig. 11.

Models would predict significantly higher formation and growth rates of small new particles during spring time, if they considered the enhanced emissions from new Scots pine foliage. Since the increase in emissions of monoterpenes would be highest in southern Finland, also the induction in the simulated new particle formation and growth would be greatest there. The scale of the enlargement largely depends on the ratios of concentrations of sulfuric acid and organics originating from monoterpene oxidation. Hence, the increases in the predicted formation
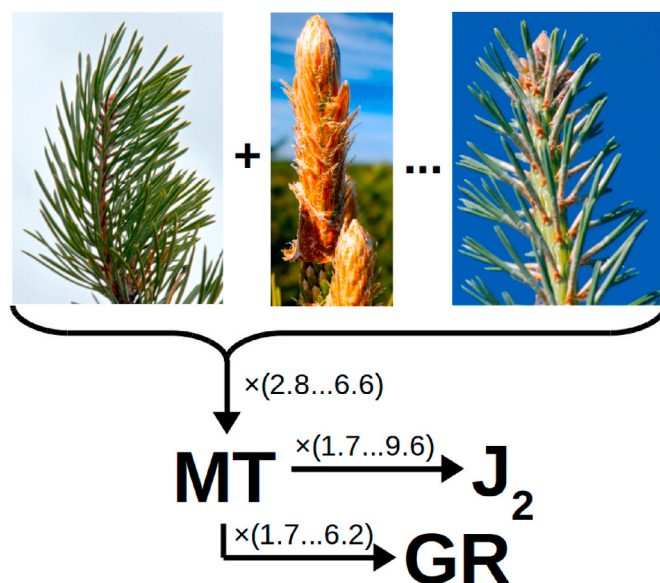

Fig. 11. The potential impact of considering the enhanced emission potential of new Scots pine foliage during spring. "MT" refers to both emissions and concentrations of monoterpenes. The factors are provided as a minimum-maximum range considering trees growing in northern and southern Finland and different concentrations of sulfuric acid and organics. The increases in the emission, formation (J_2) and growth (GR) rates are calculated as: (Y1–Y2)/Y2 × 100%, where Y1 = emission, formation or growth rate considering the emission potential of both new and mature needles, and Y2 = emission, formation or growth rate considering only the emission potential of mature needles.

and growth rates are modest at high $[H_2SO_4]/[org]$, but still greater than the uncertainty connected to the instrumentation used to obtain the rates (Manninen et al., 2016; Wagner et al., 2016; Kangasluoma and Kontkanen, 2017) and the uncertainty related to the calculation of these rates (Yli-Juuti et al., 2011). At low $[H_2SO_4]/[org]$ (e.g. ⅓ × 10^{-1} cm^{-3}), J_2 would be predicted to be ~10 times larger in southern Finland, when also considering the enhanced emissions from new foliage, while the corresponding growth rate would be ~6 times greater. Such increases in the predictions of new particle formation and growth would severely impact climate change predictions.

We emphasize that we are here not claiming that the discrepancy between field observations and model predictions of new particle formation can solemnly be explained by an exclusion of BVOC emissions from new foliage, but instead estimate how much aerosol processes would be predicted to increase if the enhanced emissions from new Scots pine foliage were included in models. Accounting for emissions from stems (Rissanen et al., 2020) and emission bursts from mature foliage earlier in the season (Aalto et al., 2015) would additionally work towards a closure of the gap. Pronounced early spring time emission bursts

Table 2

Observed ranges in the concentrations of sulfuric acid (H_2SO_4) and condensable organics (org) together with the differences in the formation rate of 2 nm clusters (J_2) and growth rate of 2–3 nm particles (GR) when the increased emission potential of new Scots pine foliage is considered in addition to the emission potential of mature foliage, and compared to situations where only the emission potential of mature foliage is included. All values are for spring time, while the resulting differences (ΔJ_2 and ΔGR) are provided for northern and southern Finland, individually. The concentrations of condensable organics (org) predicted for northern and southern Finland, using only monoterpene emissions from mature foliage, are assumed to be 2.8 times (northern Finland) and 6.6 times (southern Finland) less than the observed concentrations.

$[H_2SO_4]$ (cm^{-3})	[org] (cm^{-3})	[org] (cm^{-3}), northern Finland, only mature foliage is considered	[org] (cm^{-3}), southern Finland, only mature foliage is considered	ΔJ_2, northern Finland (%)	ΔJ_2, southern Finland (%)	ΔGR, northern Finland (%)	ΔGR, southern Finland (%)
10^6	10^7	$3.6 \cdot 10^6$	$1.5 \cdot 10^6$	180	470	150	400
10^7	10^7	$3.6 \cdot 10^6$	$1.5 \cdot 10^6$	73	130	65	110
10^7	$5 \cdot 10^7$	$1.8 \cdot 10^7$	$7.6 \cdot 10^6$	150	350	130	310
10^6	$5 \cdot 10^7$	$1.8 \cdot 10^7$	$7.6 \cdot 10^6$	280	860	170	520

from mature shoots are only partly included in our estimates (Fig. 5c), since such bursts have so far been shown to mainly take place before growth onset, and thus before the period that our study targets (Aalto et al., 2015). We accentuate that the influence of BVOCs on aerosol processes is far from simple and the formation of aerosol particles e.g. also depends on the absolute concentration of individual compounds and blend of VOCs (e.g. Lee et al., 2016; Faiola et al., 2018; 2019; McFiggans et al., 2019; Ylisirniö et al., 2020). The results presented in this section are connected with a large degree of uncertainty, since no other equations than those given in this section were used for these calculations and because our estimated underestimations of the emission potential in themselves are uncertain. One cause of this uncertainty is the assumption that emissions originate from storage pools only, and that a fixed value of β can be used to describe the emissions throughout the season. As also mentioned earlier, previous studies have, however, shown that a significant part of the emissions of monoterpenes from Scots pine can originate from *de novo* synthesis (Ghirardo et al., 2010; Taipale et al., 2011; Aalto et al., 2015; Rantala et al., 2015). Also, it is in reality known that β can vary during the season and can be different for individual monoterpene isomers (Hakola et al., 2006; Hellén et al., 2018), and hence can cause significant seasonal variations in the calculated emission potential which are not necessarily true (Hellén et al., 2018). Thus, a different handling of the emission potentials would impact the findings of this study, though it is unsure to which direction.

5. Conclusions

We have investigated the potential effects of considering the enhanced monoterpene emission potential of new Scots pine foliage on the whole tree's emission potential as a function of season, stand age and location. As methods, we used several years of continuous measurements of the emission rates of monoterpenes from new and mature Scots pine foliage, and growth models to predict the seasonal and yearly development of Scots pine needles. We found that the importance of the emissions from new Scots pine foliage decreases as a function of the season, tree age and latitude in Finland. During spring time, new Scots pine foliage could be responsible for the majority of the whole tree's foliage emissions of monoterpenes, independently of tree age and location. Our calculations suggest that neglecting the specific age (but not biomass or leaf area index) of the stand at most leads to an error of ~20% in simulations of specific locations. We demonstrate a good agreement between our whole tree foliage emission potentials, which account for the emissions from developing foliage, and monoterpene emission potentials derived from measured ecosystem scale fluxes of a Scots pine dominated forest. We also show that the ecosystem scale-derived emission potentials of monoterpenes are in better agreement with our whole tree foliage emission potentials than with the emission potential of mature Scots pine foliage or the whole tree potential when it is assumed that the emission from new foliage is enhanced in a similar manner as in MEGAN v2.1.

Our results suggest that the emissions of monoterpenes from Finland could be underestimated by ~27 Gg monoterpenes/year, which corresponds to a very significant fraction of the total monoterpene emissions predicted from Finnish forests. The estimated underestimation is especially severe during spring months where new particle formation is most frequent. Thus, the implications of our findings can lead to increases in the predictions of formation and growth rates of small particles during spring time in northern Finland by ~75–280% and ~65–180%, respectively, and in southern Finland by ~130–870% and ~110–520%, respectively.

We speculate that the emission trait observed by Aalto et al. (2014), on which our study is based, is probably not specific to Finnish Scots pine trees, nor is it sure that all Finnish Scots pines exhibit such trait, and thus our findings could be of importance in simulations of all places where Scots pines make up a major fraction of the land cover. Since the certainty of our conclusions are strongly limited by the availability of BVOC emission observations from new needles, we call for additional spring time measurements of new foliage. Such could either be conducted by separate enclosure measurements of new and mature foliage or by measuring the ecosystem scale flux with micrometeorological techniques. Since it is possible that other conifers species than Scots pine exhibit a similar emission behaviour, measurements of also other evergreen needle species are vital for improving emission algorithms.

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