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Influence of tree provenance on biogenic VOC emissions of Scots pine (*Pinus sylvestris*) stumps

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HIGHLIGHTS

- Emissions of volatile organic compounds (VOC) from Scots pine stumps were measured.
- Emission rates relative to stump area were 27–1582 mg h^{-1} m^{-2} after cutting.
- Tree provenance affected the VOC composition.
- Emission rates were estimated for different aged pine stands.
- Fresh stump surface is a significant source of monoterpene emissions.

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ABSTRACT

Resin-storing plant species such as conifer trees can release substantial amounts of volatile organic compounds (VOCs) into the atmosphere under stress circumstances that cause resin flow. Wounding can be induced by animals, pathogens, wind or direct mechanical damage e.g. during harvesting. In atmospheric modelling of biogenic VOCs, actively growing vegetation has been mostly considered as the source of emissions. Root systems and stumps of resin-storing conifer trees could constitute a significant store of resin after tree cutting. Therefore, we assessed the VOC emission rates from the cut surface of Scots pine stumps and estimated the average emission rates for an area with a density of 2000 stumps per ha. The experiment was conducted with trees of one Estonian and three Finnish Scots pine provenances covering a 1200 km gradient at a common garden established in central Finland in 1991.

VOC emissions were dominated by monoterpenes and less than 0.1% of the total emission was sesquiterpenes. α-Pinene (7–92% of the total emissions) and 3-carene (0–76% of the total emissions) were the dominant monoterpenes. Proportions of α-pinene and camphene were significantly lower and proportions of 3-carene, sabinene, γ-terpinene and terpinolene higher in the southernmost Saaremaa provenance compared to the other provenances. Total terpene emission rates (standardised to +20 °C) from stumps varied from 27 to 1582 mg h^{-1} m^{-2} when measured within 2–3 h after tree cutting. Emission rates decreased rapidly to between 2 and 79 mg h^{-1} m^{-2} at 50 days after cutting. The estimated daily terpene emission rates on a hectare basis from freshly cut stumps at a cut tree density of 2000 per ha varied depending on provenance. Estimated emission ranges were 100–710 g ha^{-1} d^{-1} and 137–970 g ha^{-1} d^{-1} in 40 and in 60 year-old forest stands, respectively. Our result suggests that emission directly from stump surfaces could be a significant source of monoterpene emissions for a few weeks after logging in a Scots pine stand, but provenance properties strongly affect resin flow from root to stump surface.

1. Introduction

Boreal conifer forests form the largest terrestrial biome (Taiga) covering an approximately 15.1 million km^2 forested area (Taggart and Cross, 2009). This forested area includes at least 30% of global

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forest and has a significant impact on atmospheric quality through the emission of reactive volatile organic compounds which participate in secondary organic aerosol (SOA) and cloud formation (Spracklen et al., 2008). In the boreal area nearly 60% of forest loss is attributable to wild fires and the rest is caused by factors such as wind and forest clear-cuts (Potapov et al., 2008).

In Finland, forest covers 26.3 million hectares, which is 86% of the total land area of the country. The total growing stock in the last assessment was 2206 million m^3, half of which consists of Scots pine (*Pinus sylvestris* L.) (Peltola, 2009). In year 2010, ca. 7290 km^2 of forests were subjected to fellings, 73% of this was thinnings and 27% clear-cuts (Metla, 2011). This area corresponds to nearly 3% of the total forest land in Finland. Therefore, forest management and cutting are the dominant disturbances in the forests of Finland.

Foliage of Scots pine trees is known to be an important source of reactive mono- and sesquiterpenes (Rinne et al., 2000, 2007; Bäck et al., 2005, 2012; Ruuskanen et al., 2005; Räisänen et al., 2009), but monoterpenes are also emitted from the root system (Janson et al., 1999; Ketola et al., 2011). Above-ground VOC emission can also originate from trunk and branches, in which the xylem or phloem have higher concentrations of stored monoterpenes than needles (Sallas et al., 2003), and can thus, be significant sources of terpene emission. Monoterpene concentrations of pine trees in a 50-year old forest stand were 1.4–4.9 mg g^{-1} (FW) and 0.6–0.8 mg g^{-1} (FW) in stem heartwood and stem sapwood, respectively (Turtola et al., 2002). Fäldt et al. (2006) reported monoterpene concentrations of less than 10 mg g^{-1} in healthy phloem and more than 1000 mg g^{-1} in phloem within the fungi-infected reaction zone of Scots pine. VOC emission rates from air-dried pine xylem can be in the range of 900–1800 μg m^{-2} h^{-1} (Hyttinen et al., 2010) and monoterpenes form two thirds of the total emission (Manninen et al., 2002a). Ghirardo et al. (2010) determined the fraction of monoterpene emissions originating from *de novo* biosynthesis in *Pinus sylvestris* by applying $^{13}CO_2$ fumigation. They found that in actively growing pine seedlings as much as 58% of monoterpene emissions originated from light-dependent *de novo* synthesis in needles and the rest was from stored reserves (Ghirardo et al., 2010).

The analyses of forest management activities on emission of VOCs from resin-storing conifers to the atmosphere are limited. Räisänen et al. (2008) studied the impacts of thinning and clear-cut felling of a 40-year-old Scots pine stand on atmospheric monoterpene concentration. Stumps, needle debris and cut stems piled as 3 m long logs acted as sources of monoterpene emissions in the area. Three-fold increases in the atmospheric total monoterpene concentration (in the range of 5.3–15.5 μg m^{-3}) were detected in June, in the period after the clear cut felling. Thinning 60% and 30% of the stand density resulted in smaller increases in atmospheric monoterpene concentrations with no differences between thinning treatments at the end of the growing season in September (Räisänen et al., 2008). Schade and Goldstein (2003) measured monoterpene emissions from approximately 6 m above the average tree height of a ponderosa pine (*Pinus ponderosa* L.) stand with a Relaxed-Eddy-Accumulation (REA) GC-FID system. The daytime emissions of major monoterpenes, α-pinene, β-pinene, and Δ-3-carene, were enhanced by 40 fold in the thinned area when compared to the 34 day period before thinning. Schade and Goldstein (2003) attributed the majority of the rapid increase in monoterpene emissions to be from needle debris. Haapanala et al. (2012) estimated that annual monoterpene emissions from forestry operations could be 10% of the monoterpene emissions of intact forests in Finland.

Climate relevant and radiation-scattering SOA particles are rapidly formed in reaction chambers when monoterpene-dominated VOC emissions of Scots pine react with oxidants (Hao et al., 2009; Virtanen et al., 2010). Similar organic aerosol particles have also been detected in the atmosphere of forest sites (Virtanen et al., 2010). This suggests that changes in VOC emission potential of a forest stand could affect local atmospheric quality and may increase diffusion of solar radiation (Holopainen, 2011), which has been shown to improve canopy scale CO_2 uptake in conifer stands (Urban et al., 2012).

To better assess the impact of various environmental and forest management factors on Scots pine as a source of reactive VOCs we monitored emission dynamics of tree stumps after cutting. We wanted to know 1) what is the VOC emission rate from the surface of cut Scots pine stumps, 2) does tree provenance affect the composition and emission rate of VOCs from stumps, 3) how rapidly the stump emission cease after cutting of the tree and 4) what could be the consequences of clear cut and thinning on the forest stand scale emissions.

2. Materials and methods

2.1. Experimental site and tree provenances

Experiments were conducted in an experimental Scots pine (*P. sylvestris* L.) stand with nine provenances sown as a common garden experiment at the Suonenjoki Research Unit (latitude 62°37′) of the Finnish Forest Research Institute in 1991. Trees were grown from seeds originating from a 1200-km south–north transect from Estonia to Northern Finland. Trees of each provenance grew in five replicated 1000 m^2 blocks in fully replicated rows, and the area of the whole research field was 0.5 ha. More details of the site, sandy soil which was earlier prepared for production of nursery conifer seedlings, and growing conditions are described in Nerg et al. (1994) and Manninen et al. (2002b). The stand has been thinned several times since its establishment, with 30–40% of the original trees left at the time of the experiment. The provenances selected for the present study were Saaremaa (SAA, latitude 58°22′), Korpilahti (KOR, latitude 62°0′), Suomussalmi (SUO, latitude 65°10′) and Muonio (MUO, latitude 67°56′). The Saaremaa provenance originated in Estonia, the others were from Finland.

2.2. Sampling and analysis

To monitor VOC emissions from pine stumps after cutting, one tree of each provenance was randomly selected from each of four different blocks ($n = 4$). The trees were aged 19 years old, and were felled with a chain saw on 11 August 2010. The first VOC samples were collected within 2–3 h of cutting, and then from the same stumps 5, 13, 28 and 50 days later.

A pre-cleaned (120 °C for 1 h) polyethylene terephthalate (PET) cooking bag (size 25 × 55 cm) was tightened around stumps with rubber bands. The volume of the fastened bag was ca. 1 L. Crevices in the bark were blocked with aluminium foil to reduce air leakage. Bags were first flushed for 5 min with ozone-free air (Ozone Scrubber Cartridge, Environnement S.A., Poissy, France, to avoid degradation of VOCs in the adsorbent) purified with activated carbon (Wilkerson F03-C2-100, Monterrey, Mexico, to remove VOCs from background air entering the collection bags), which was channelled into the bags via Teflon tubing at a rate of 600 ml min^{-1}. When the bags had expanded and the air had been replaced, the flow rate was reduced to 300 ml min^{-1} and a purified stainless steel tube (ATD sample tubes, Perkin Elmer,, Norwalk, CT, USA) filled with approximately 150 mg of Tenax TA adsorbent (mesh 60/80, Supelco, Bellefonte, PA, USA) was inserted into a small hole cut in the upper corner of the collection bag and fastened with a plastic tag. The sample was pulled through the sample tube with a vacuum pump

(Thomas 5002 12V DC, Puchheim, Germany) at a rate of 200 ml min^{-1} for 5 min on 11 August. The sampling period was reduced to 2 min for the following three sampling dates (sampling time was shortened, because the quantities of a few monoterpenes were close to the upper detection limit of the GC–MS) and returned to 5 min for the last sampling date. Higher inlet than outlet flow was used to maintain a positive pressure and prevent VOCs from outside the bag contaminating the system. Air flows were calibrated with a mini-Buck calibrator (Model M-5, A.P. Buck, Inc., Orlando, FL, USA) before collections. Sample tubes were sealed with Teflon-coated brass caps immediately after collection and stored at 4 °C until analysis. Blank samples were also collected from empty collection bags. Temperature on the stump surfaces inside the bags was monitored by wireless temperature/humidity loggers (Hygrochron DS1923-F5 iButton, Maxim Integrated Products, Inc., CA, USA). Average temperatures on the stump surfaces inside the bags during collections were 21 °C (11 August), 22 °C (16 August, 5 days after cutting), 12 °C (24 August, 13 days after cutting), 16 °C (8 September, 28 days after cutting), 10 °C (30 September, 50 days after cutting). Solar radiation was diffused and low level on tree stump surface. Temperature outside the bag was max. 0.5 °C higher than inside the bag.

VOC samples were analysed by GC–MS (Hewlett Packard GC type 6890, Waldbronn Germany; MSD 5973, Beaconsfield, UK). Compounds trapped to the adsorbent were desorbed (Perkin Elmer ATD400 Automatic Thermal Desorption System, Wellesley, MA, USA) at 250 °C for 10 min, cryofocused in a cold trap at −30 °C and subsequently injected onto an HP-5 capillary column (50 m × 0.2 mm i.d. × 0.33 μm film thickness, J&W Scientific, Folsom, CA, USA). The temperature program was 40 °C for 1 min, followed by increases of 5 °C min^{-1} to 210 °C and 20 °C min^{-1} to 250 °C. The carrier gas was helium. Mono (C_{10}) and sesquiterpenes (C_{15}) were identified by comparing the mass spectra of compounds with those in the Wiley data library and pure standards. β-phellandrene was quantified using α-pinene as a reference substance. Emissions were calculated per hour and stump area, excluding dead bark (mg h^{-1} m^{-2}). Since VOC emissions from vegetation are temperature-dependent (Guenther et al., 1993) emissions were also calculated with temperature standardisation. The standardised measurements were calculated using algorithms by Guenther et al. (1993) relative to temperatures of 20 °C (Ekberg et al., 2011), which is typically used to represent boreal conditions, and 30 °C, which is a common reference temperature (Duhl et al., 2008) for biogenic VOC emission inventories. The temperature coefficient β was 0.09 for MTs (Guenther et al., 1993) and 0.16 for STs (average calculated from studies with several STs emitted from conifers, Table 1 in Duhl et al., 2008).

2.3. Emission estimates of forest cuttings

The estimated total terpene emission rates from tree stumps per hectare were calculated using the emission rate of terpenes per m^2 of stump surface standardised to 20 and 30 °C. We used the cut stand density of 2000 trees per hectare to calculate the stump area of cut trees. Stump area estimates per hectare for different aged forest stands were calculated according to Huuskonen and Miina (2007) using the modelled values of fresh (*Myrtillus* type, MT) growth sites. When grown at a density of 2000 Scots pine trees per hectare, the modelled stump areas of the cut trees were 13 m^2 ha^{-1} at the age of 20 years, 37 m^2 ha^{-1} at the age of 40 years, and 50.6 m^2 ha^{-1} at the age of 60 years, respectively. The estimates are based on the assumption that terpene content and consequent volatile emission is invariant with tree age.

2.4. Statistics

VOC-emission data was logarithm transformed to meet the assumptions of variance analysis. The general linear model for repeated measures, with provenance as a between-subject factor, and time as a within-subject factor, was used to investigate whether the sampling time during the 50 day period after tree cutting had any effect on VOC emissions, and if the time-response differed between the provenances. Helmert contrasts, which compare the mean of one sampling point with the means of later ones, were used to study how the emission changed during the study period. In addition, one-way ANOVA with Tukey HSD or Dunnett T3 tests was used to study differences between the provenances on two sampling occasions: 2–3 h and 50 days after cutting. Statistical analyses were done using SPSS 17.0. All the results are given standardised to +20 °C unless otherwise stated.

3. Results

3.1. Impact of time and provenance on VOC emission rates and composition

Total terpene emission rates from stumps varied from 27 to 1582 mg h^{-1} m^{-2} when measured within 2–3 h after tree cutting (data not shown). Total emission rates were highest in the southernmost provenance, Saaremaa (SAA), and lowest in Suomussalmi (SUO) (Fig. 1), but the difference was not significant. However, emissions of 3-carene were significantly higher from stumps of the SAA provenance than those of the SUO provenance, when measured soon after cutting (Fig. 2a, $p = 0.029$, Tukey's HSD). Emissions consisted mainly of the monoterpenes α-pinene (7–92% of the total emissions, average 57%) and 3-carene (0–76%, average 24%) (Fig. 3). Emissions of sesquiterpenes were low, on average 0.02% of the total emissions, and consisted mainly (90%) of longifolene (data not shown). Proportions of α-pinene, camphene, sabinene, 3-carene, γ-terpinene and terpinolene differed between the provenances (data shown for the major compounds, α-pinene and 3-carene in first and last sampling times, Fig. 3). Proportions of α-pinene and camphene were significantly lower and those of 3-carene, sabinene, γ-terpinene and terpinolene higher in the southernmost Saaremaa (SAA) provenance than the northern provenances, Korpilahti (KOR), Suomussalmi (SUO) and Muonio (MUO) ($p < 0.05$, Tukey's HSD).

Emission rates of most compounds followed a similar decreasing trend during the experimental period lasting until 50 days from cutting. Changes in the emission rates of 3-carene, β-pinene, α-pinene, limonene, β-phellandrene and terpinolene are given as examples (Fig. 2a–f). The drop in emission rates from the first measurement to five days after cutting was steep (Figs. 1 and 2). For example, the emission rate of total terpenes was typically 5% of that measured 2–3 h after cutting. Emissions of most compounds transiently increased after second sampling date and then dropped again between 28 and 50 days of cutting (Figs. 1b, 2, Table 1). Emission rates varied between 2 and 79 mg h^{-1} m^{-2} at 50 days after cutting. It should be noted that the emission rates of the provenances changed drastically within 50 days, and the pattern of change varied for the different provenances. The lowest emitter after cutting, SUO provenance, became the highest emitter and SAA provenance became the lowest emitter (Fig. 1). Decrease in 3-carene emissions from SAA provenance was steeper than in the other provenances, which resulted in significant provenance × time interaction (Fig. 2a, Table 1). The proportions of the terpenoids emitted did not significantly alter for any of the provenances for the period up

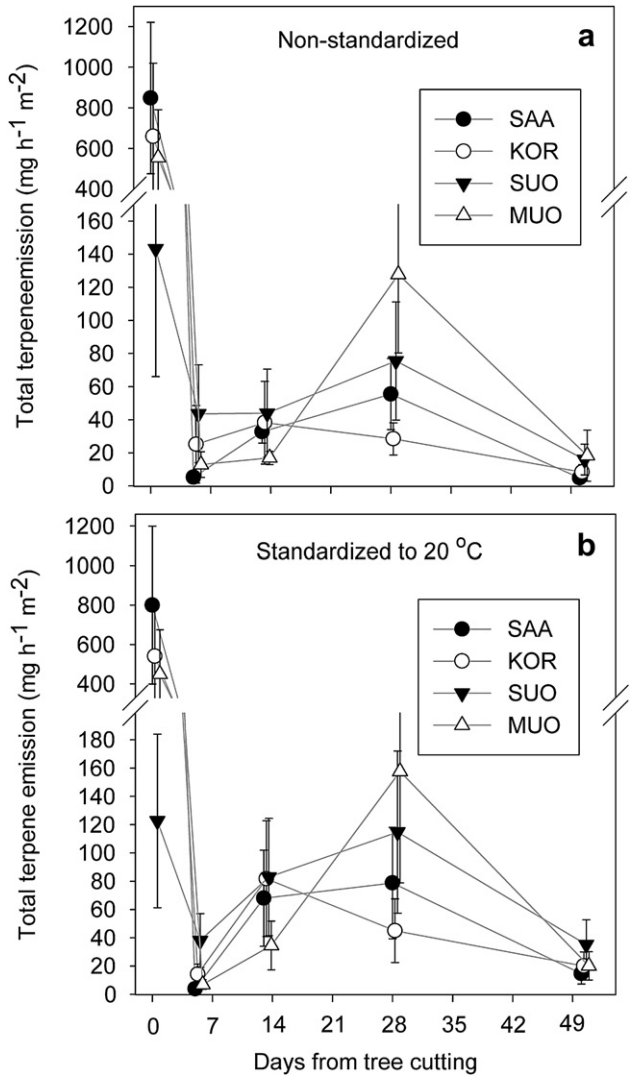

Fig. 1. Non-standardised (a) and standardised to 20 °C (b) emission rates of total terpenoids from stumps of four *Pinus sylvestris* provenances (SAA = Saaremaa, KOR = Korpilahti, SUO = Suomussalmi, MUO = Muonio) 2–3 h, 5, 13, 28 and 50 days after tree cutting on 11th August 2010. Note breaks in the *y*-axes, and that only one SE is visible in a few cases.

until 50 days after cutting (data shown for α-pinene and 3-carene in first and last sampling, Fig. 3, $p > 0.1$ for time and time × provenance of repeated measures ANOVA). Similar to the first sampling, the same significant differences in proportions of α-pinene, 3-carene, camphene, sabinene, γ-terpinene and terpinolene between the provenances were noted at 50 days after cutting (data shown for α-pinene and 3-carene Fig. 3, $p < 0.05$, Tukey's HSD).

3.2. Estimated stump emissions at a forest stand scale

The estimation of total terpene emission rates from tree stumps per hectare was based on the cut stand density of 2000 trees per hectare to calculate the basal stump area of cut trees. With this density of felled trees, emission rates standardised at +20 °C (Fig. 4a) on the cutting day varied between the provenances, ranging from 35 to 249 g ha^{-1} d^{-1}, from 100 to 710 g ha^{-1} d^{-1} and from 137 to 970 g ha^{-1} d^{-1} for 20, 40 and 60 year old forest stands, respectively. The emission rates standardised at +30 °C (Fig. A.1a) varied from 94 to 613 g ha^{-1} d^{-1}, from 268 to 1746 g ha^{-1} d^{-1} and from 366 to 2386 g ha^{-1} d^{-1} for 20, 40 and 60 year old forest stands, respectively. These values decreased rapidly and were substantially lower after 50 days (Fig. 4b and Fig. A1b) from cutting.

4. Discussion

4.1. Factors affecting quantity and quality of stump terpene emissions

Our results indicated that a few hours after cutting pine trees the upper surface of the stump is a very important source of VOC emission due to the resin flow through resin ducts to the cut surface from the root system. Monoterpene emission rates from the cut stump surfaces after harvesting indicated that they can reach 800 mg h^{-1} m^{-2} (direct measurements and at +20 °C standardised data) depending on the provenance under investigation. Rapid reduction of the emission rate within five days was typical for all provenances. Transient and slight increase in emission rates after five days, and decrease again at 50 days after tree cutting, cannot only be explained by the temperature differences, since temperature was higher five days after cutting than two and four weeks after cutting. Moreover, temperature standardisation was done to minimise the effect of temperature on emissions. Very little is known what happens in coniferous root systems soon after tree cutting when the connection between shoot and roots suddenly ends. Hernesmaa et al. (2005) reported increased enzyme activities in Scots pine rhizosphere two weeks of felling and increased microbial biomass until five weeks of felling, but 11 after felling soil chemistry and microbial community structure did not differ from bulk soil or living tree rhizosphere. Whether changes in rhizosphere and how resin storage of main roots affect resin flow to stump surface is not known.

Volatile monoterpene composition of the stumps over the study period remained the same. Eberhardt et al. (2009) observed the proportion of α-pinene and β-pinene to decline in the heartwood and β-pinene to decline in the sapwood of longleaf pine (*Pinus palustris* Mill.) stumps monitored in the field during a 1 year postharvest period. Flechtmann et al. (1999) reported increase of several oxygenated monoterpenes and decrease of α-pinene and many other non-oxygenated monoterpenes in billets cut from loblolly pine (*Pinus taeda*). Such changes in terpenoid composition were not noted in this study. Thus, the results suggest that up to two months after tree cutting the stump emissions probably originated from resin flow to the stump surface from stores of living roots, although decay of the roots may have started (Hernesmaa et al., 2005). In nature, monoterpene and ethanol emissions from conifer stumps are important in orientation of root-colonising weevils and constitute oviposition cues (Lindelöw et al., 1993), which promote the natural decomposition of the stumps.

Despite of overall decrease in monoterpene emission rate variation existed between the provenances. E.g. the most southernmost Saaremaa provenance (SAA) had the highest emission for the first sampling, but then had a faster reduction rate than the other provenances. The variation may be due to differences in monoterpene concentrations of oleoresin. Manninen et al. (2002b) found significantly higher monoterpene content in the wood of SAA provenance trees than in KOR, SUO or MUO provenances whereas resin acid content was more variable between three individuals than between provenances. High monoterpene concentration of resin in trees of the SAA provenance may partly affect faster resin outflow after cutting, but also soon lead to the

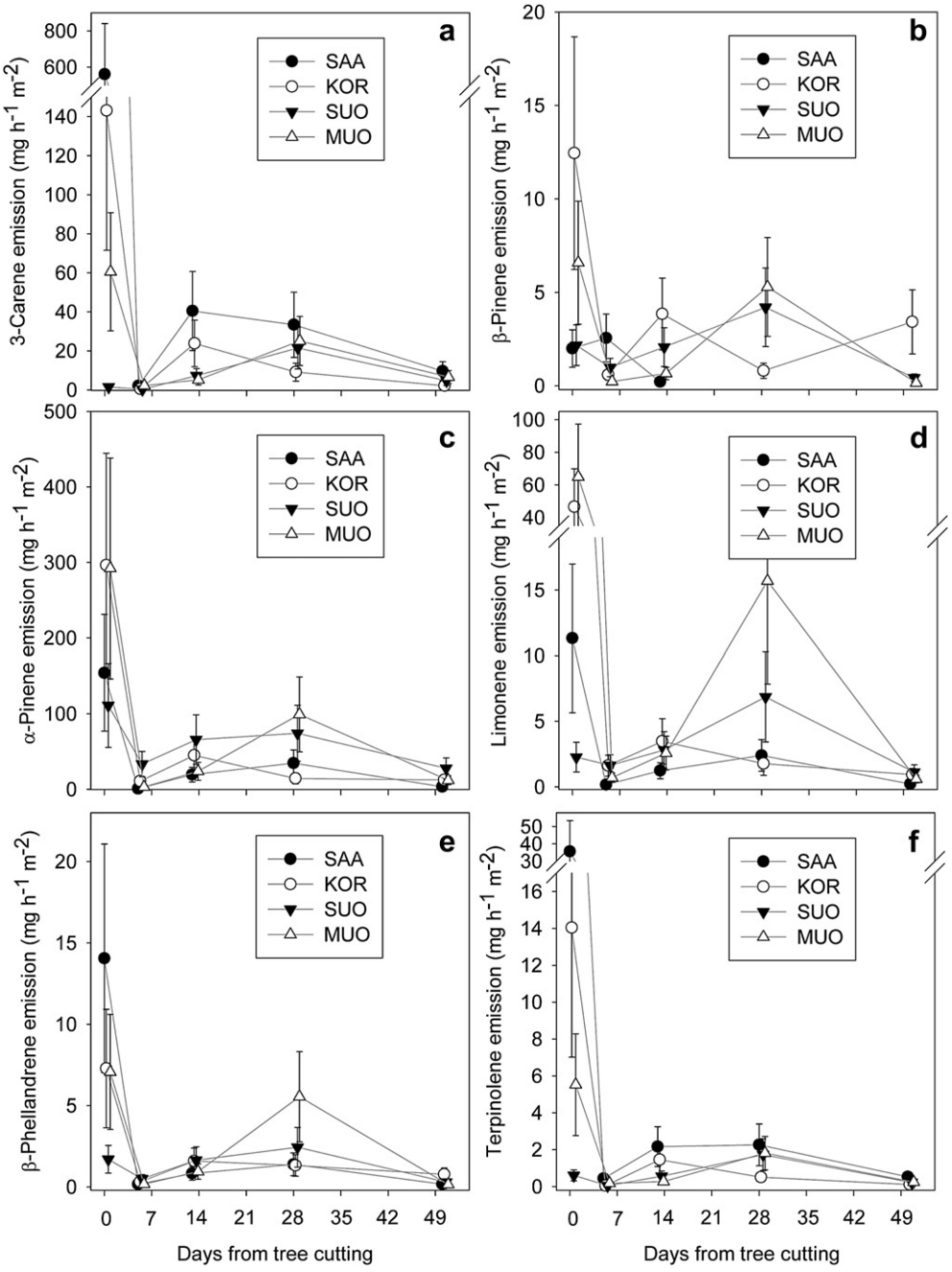

Fig. 2. Changes in emission rates of a few terpenoids (standardised to 20 °C) from stumps of four *Pinus sylvestris* provenances (SAA = Saaremaa, KOR = Korpilahti, SUO = Suomussalmi, MUO = Muonio) 2–3 h, 5, 13, 28 and 50 days after tree cutting on 11th August 2010. Note breaks in the *y*-axes, and that only one SE is visible in a few cases.

formation of thicker dry resin blockage on the stump surface (cf. Lombardero et al., 2000; see Fig. A.2). When the resin starts to dry, emissions will substantially decline within five days of cutting as a part of the natural healing process in damaged conifers (Lombardero et al., 2000). In healthy Scots pine trees the viscosity and flow of resin is relatively constant between individual trees, but induced production could increase resin flow by up to three-fold (Kytö et al., 1998). Thus, previous growth history of trees and the rate of induced stress may increase variability in resin production and flow between provenances and between individual tree stumps.

Monoterpene composition of needles and wood of Scots pine are under strong genetic control (e.g. Muona et al., 1986), which is also reflected in volatile emissions (Bäck et al., 2012). Genetic influence is also evident between the provenances of the present study. Provenances have been studied for terpene concentrations of the shoots of four and twelve-month-old seedlings (Nerg et al., 1994), as well as needles and wood of seven-year-old saplings (Manninen et al., 2002b) and needles of 12–15 years old trees (Heijari et al., 2008). For example, the proportion of α-pinene increased and 3-carene, sabinene and terpinolene were shown the decrease with latitude (Nerg et al., 1994; Manninen et al., 2002b). In this study, volatile monoterpene composition of the stumps after tree cutting was similar to the terpene concentrations of other tree parts in the earlier reports, and the differences between the provenances were the same.

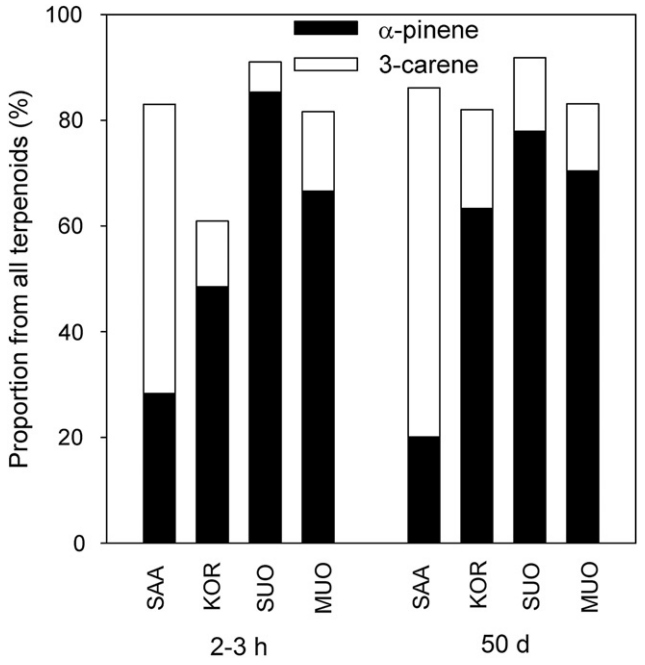

Fig. 3. Proportions of α-pinene and 3−carene of all volatile terpenoid emissions collected from stumps of four *Pinus sylvestris* provenances (SAA = Saaremaa, KOR = Korpilahti, SUO = Suomussalmi, MUO = Muonio) 2−3 h and 50 days after tree felling.

4.2. Impacts of forest cuttings on atmospheric biogenic VOC concentrations

Knowledge of the emission capacity of different components of a cut stand site is needed in order to understand the full impact of logging activities by modelling the atmospheric load of biogenic VOCs from resin storing tree species. This study was focused on VOC flux from cut surfaces of stumps. The basal area of trees per hectare increases during forest development leading to larger emitting surface areas per hectare in older forests. Our estimation of total terpene flux from tree stumps to the atmosphere after timber felling was based on the cut stand density of 2000 trees per hectare to calculate the basal stump area of cut trees. In young, 20−40 year old stands, this could represent a stand at the thinning stage, as some young stands could have tree densities higher than 4000 trees per ha (Huuskonen and Miina, 2007). In older, 60 year old forests, removal of 2000 trees ha^{-1} normally means a clear-cut harvesting.

Emission rates were substantially higher on cutting day than 50 days after cutting. Our results with data standardised to +20 °C indicated that maximum emission rates could be close to 1 kg ha^{-1} d^{-1} in a clear-cutting stage of 60 year old pine forest. If cutting is conducted on hot summer days (+30 °C) the emission rates could reach 2.4 kg ha^{-1} d^{-1}. It should be mentioned that temperature-standardised values should be treated with care, because it is not yet known how well the standardisation algorithm, originally developed for foliage, functions for other plant parts.

Haapanala et al. (2012) published results of monoterpene emission rates (standardised to +15 °C) from Scots pine annual logging residues in Finland. Their monitoring period of stump surface emissions lasted 130 d after cutting. The monoterpene releases over the six month active-growth period from the Scots pine stumps was 33 g m^{-2} per year (184 d was used to approximate the annual emitting days). They estimated this to correspond to an annual monoterpene emission rate of about 0.1 g m^{-2} in the stand scale which means 1 kg ha^{-1} y^{-1}. If we use our estimated emission rates of 20 y old trees per hectare after 50 d of cutting (Fig. 4b), in the range of 5−11 g ha^{-1} d^{-1}, depending on the provenance, and upscale it to an accumulated annual emission rate by multiplying it by 184 d [growing/emitting season May 1−October 31], we will have an annual emission rate estimation of from 0.92 to 2.02 kg ha^{-1} y^{-1}. Standardisation to +20 °C gives an approximately 36% higher value than standardisation to +15 °C (unpublished data). Thus the calculated annual stump monoterpene emission rate by Haapanala et al. (2012) standardised to +20 °C will be 1.36 kg ha^{-1} y^{-1}. This value is within our estimated annual range of monoterpene emission rates from stumps of cut Scots pine at the stand level.

For full ecosystem and stand level assessment of VOC emissions from conifer forest clearing and thinning activities, the emissions from needle and branch litter and fine roots should be measured. The current trend is to collect the uprooted stumps of conifer trees in piles after clear-cutting so that they can dry for use as a bioenergy fuel (Palander et al., 2009). This may increase the monoterpene emissions in the stump pile area and affect local atmospheric monoterpene concentration in the summer following clear cutting.

Compared to the intact forest, cutting of trees will reduce the net emissions since stump emissions are lower than tree emissions (Haapanala et al., 2012). However, effect of thinning will create more space in the stand and stimulate the growth and VOC emissions of the nearby trees. Compared to the undisturbed forest floor, young Scots pine stands after thinning and older stands after clear-felling are important VOC sources in the forest environment. Monoterpene emission rates from a natural conifer

Table 1
Significance values for the main effect of time and interaction of tree provenance and time, and contrasts for time, tested by repeated measures ANOVA, of changes in the emissions of a few volatile organic compounds (standardised to +20 °C) and total terpenes (non-standardised, standardised to +20 °C) emitted by stumps of *Pinus sylvestris* trees cut on 11 August. Non-significant main effects of the provenance are not shown.

	Contrasts for time					
	Time	Provenance × time	11 Aug vs later	16 Aug vs later	24 Aug vs later	8 Sept vs 30 Sept
α-Pinene	<0.001	0.134	<0.001	<0.001	0.062	0.022
Camphene	<0.001	0.872	0.005	0.027	0.771	0.014
β-Pinene	<0.001	0.111	0.001	0.008	0.164	0.018
Myrcene	<0.001	0.608	0.001	0.001	0.594	0.004
3-Carene	<0.001	0.034	0.006	<0.001	0.525	0.003
Limonene	<0.001	0.189	0.001	0.004	0.787	0.001
β-Phellandrene	<0.001	0.354	0.004	0.002	0.506	0.001
Terpinolene	0.010	0.831	<0.001	0.004	0.001	0.014
Total terpenes non-standardised	<0.001	0.489	<0.001	0.022	0.189	<0.001
Total terpenes standardised to 20 °C	<0.001	0.320	<0.001	<0.001	0.132	0.002

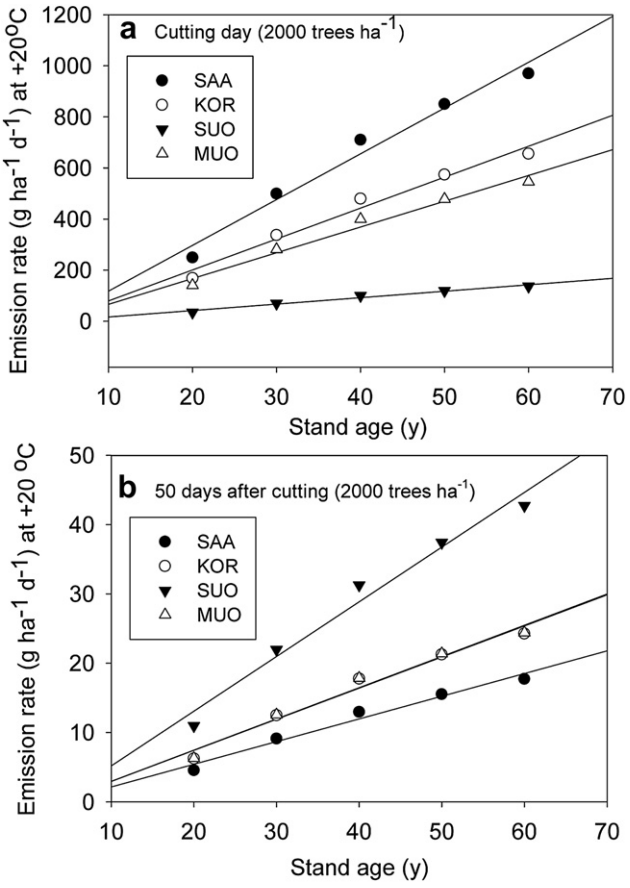

Fig. 4. Daily biogenic VOC emission rate estimates per hectare (standardised to 20 °C) from tree stumps after felling of different-aged *Pinus sylvestris* stands of four provenances (SAA = Saaremaa, KOR = Korpilahti, SUO = Suomussalmi, MUO = Muonio) grown at a density of 2000 trees per hectare at two time points; a) first 24 h after cutting and b) 50 days after cutting stand basal area estimates are calculated according to Huuskonen and Miina (2007).

forest floor with needle litter and moss vegetation are averaged at 5.76 g ha^{-1} d^{-1} (Janson et al., 1999), 1.3 g ha^{-1} d^{-1} (Aaltonen et al., 2011) or peaked at 89.5 g ha^{-1} d^{-1} (Hellén et al., 2006) in Scots pine dominated forest, and ranged between 6.8 and 9.2 g ha^{-1} d^{-1} in undisturbed Sitka spruce plantation forests (Hayward et al., 2001). Our measured monoterpene emissions from stumps of a nearly 20-year old stand on the cutting day (Fig. 4a, 35–249 g ha^{-1} d^{-1}) are higher than from an undisturbed conifer forest floor covered by needle litter. Fifty days later (Fig. 4b, 4.6–11 g ha^{-1} d^{-1}) they are at the same level. On the other hand, fresh natural needle litter in autumn may cause a peak in forest floor emissions of Scots pine forests. Janson (1993) reported that when standardised to 20 °C there was a peak monoterpene emission rate 139.2 g ha^{-1} d^{-1} in a Scots pine stand in October. Emissions from the forest soil of deciduous trees, which do not store resins, are probably lower than in emission from forest floor of coniferous forests.

5. Conclusions

Our result suggests that during the few weeks after logging and other forestry operations in a Scots pine stand, emissions directly from stump surfaces could be a significant source of monoterpene emissions. This should be considered together with the amount of needle and branch litter, when the terpenoid emission potential of deforested areas is estimated. Emission rates from individual trees and between tree provenances could be highly variable and this is probably mostly a result of accumulating resin on stump surfaces. Further studies are needed to determine the factors that influence the quantity of resin flow from the root system to the stump surface and to estimate emission algorithms for different plant parts.

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Appendix A. Supplementary data

Supplementary data related to this article can be found online at http://dx.doi.org/10.1016/j.atmosenv.2012.07.018.

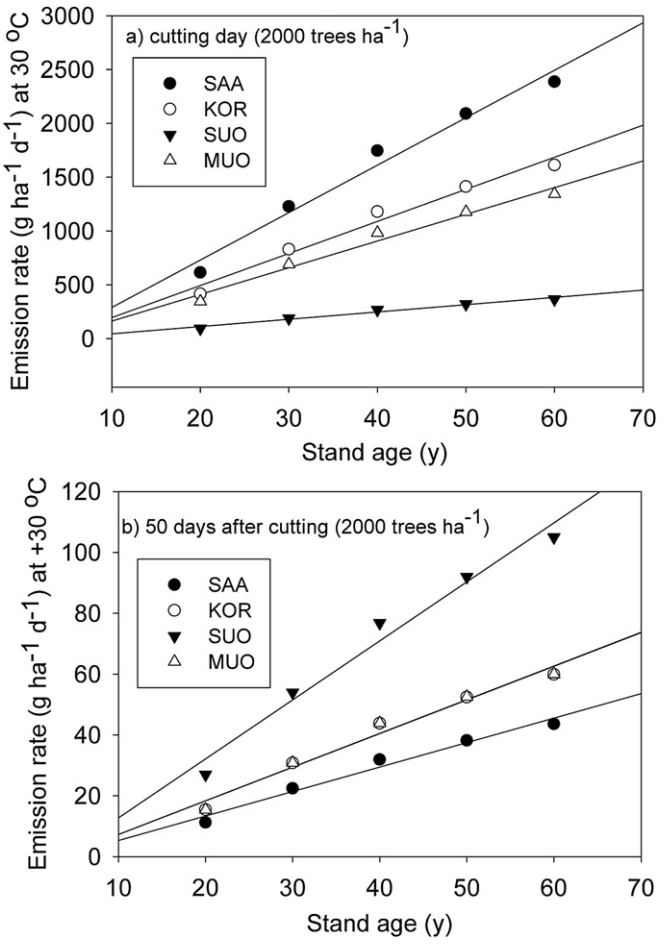

Fig. A.1. Daily biogenic VOC emission rate estimates per hectare (standardised to 30 °C) from tree stumps after felling of different-aged *P. sylvestris* stands of four provenances (SAA = Saaremaa, KOR = Korpilahti, SUO = Suomussalmi, MUO = Muonio) grown at a density of 2000 trees per hectare at two time points; a) first 24 h after cutting and b) 50 days after cutting. Stand basal area estimates are calculated according to Huuskonen and Miina (2007).

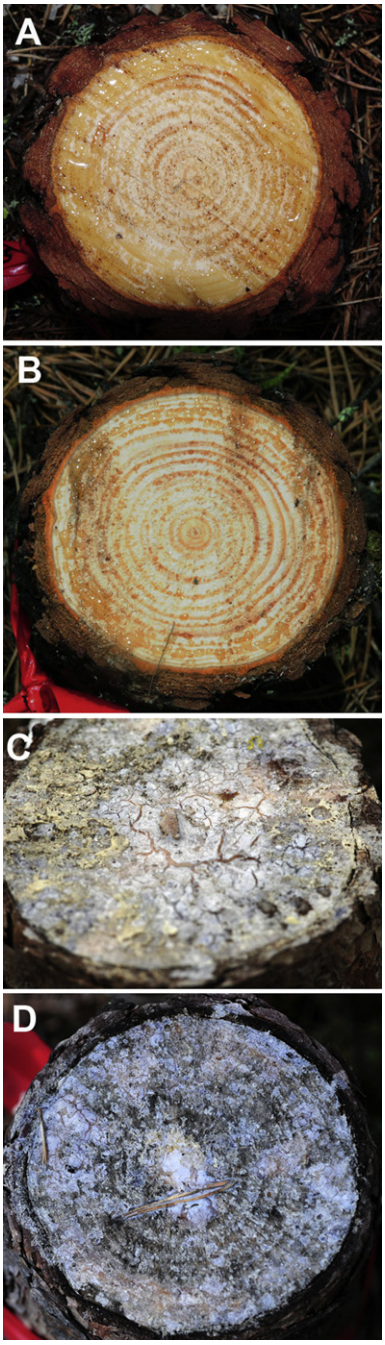

Fig A.2. Surfaces of pine stumps with fluid resin after 30 min of cutting A) high resinous tree (SAA3) and B) low resinous tree (MUO2). Surfaces of pine stumps covered by dry resin following spring after cutting C) high resinous tree (SUO4) and D low resinous tree (KOR1).

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