Isoprene and monoterpene emissions from alder, aspen and spruce short rotation forest plantations in the UK

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Abstract

An expansion of bioenergy has been proposed to help reduce fossil-fuel greenhouse gas emissions, and short-rotation forestry (SRF) can contribute to this expansion. However, SRF plantations could also be sources of biogenic volatile organic compound (BVOC) emissions, which can impact on atmospheric air quality. In this study, emissions of isoprene and 11 monoterpenes from the branches and forest floor of hybrid aspen, Italian alder and Sitka spruce stands in an SRF field trial in central Scotland were measured during two years (2018–2019) and used to derive emission potentials for different seasons. Sitka spruce was included as a comparison as it is the most extensive plantation species in the UK. Winter and spring emissions of isoprene and monoterpenes were small compared to those in summer. Sitka spruce had a standardised mean emission rate of 15 μg C g^{-1} h^{-1} for isoprene in the dry and warm summer of 2018, more than double the emissions in 2019. However, standardised mean isoprene emissions from hybrid aspen were similar across both years, approximately 23 μg C g^{-1} h^{-1} and standardised mean isoprene emissions from Italian alder were very low. Mean standardised total monoterpene emissions for these species followed a similar pattern of higher standardised emissions in the warmer year: Sitka spruce emitting 4.5 μg C g^{-1} h^{-1} and 2.3 μg C

g^{-1} h^{-1} for 2018 and 2019, aspen emitting 0.3 μg C g^{-1} h^{-1} and 0.09 μg C g^{-1} h^{-1} and Italian alder emitting, 1.5 μg C g^{-1} h^{-1} and 0.2 μg C g^{-1} h^{-1}, respectively. In contrast to these foliage emissions, the forest floor was only a small source of monoterpenes, typically one or two orders of magnitude lower than foliage emissions on a unit ground area basis. Estimates of total annual emissions from each plantation type per hectare were derived using the MEGAN 2.1 model. The modelled total BVOC (isoprene and monoterpenes) emissions of SRF hybrid aspen plantations were approximately half those of Sitka spruce for plantations of the same age. Italian alder SRF emissions were 20 times smaller than from Sitka spruce. The expansion of bioenergy plantations to 0.7 Mha has been suggested for the UK to help achieve "net-zero" greenhouse gas emissions by 2050. The model estimates show that with such an expansion total UK BVOC emissions would increase between <1% and 35%, depending on the tree species planted. Where increases might be small on a national scale, regional increases might have a larger impact on local air quality.

1. Introduction

The UK has committed to reducing its carbon dioxide (CO_2) emissions to meet net-zero greenhouse gas emissions targets by 2050, and increasing bioenergy use is seen as a substantial pathway to this. Bioenergy was the largest contributor to renewable energy within the UK in 2018, accounting for 7% of the primary energy supply (Renewable Energy Association, 2019) and it has been suggested that this could grow to 15% by 2050 (Committee on Climate Change, 2019). Solid biomass, in the form of wood pellets, chips, and agricultural and forestry residues, is the primary type of biomass used to generate heat and electricity, accounting for 60% of bioenergy in 2016 (IEA Bioenergy, 2018). However, the majority of the 7.2 million tonnes of wood pellets burned in the UK in 2018 came from imports from North America (Renewable Energy Association, 2019). However, importing biomass contributes higher carbon emissions than biomass grown in the UK (Ricardo, 2020) so a larger contribution from domestic supply of bioenergy in the UK is required if the UK is to achieve net-zero.

Currently the most common bioenergy crops in the UK are coppiced willow (*Salix spp*.) and *Miscanthus*, a perennial grass. Only 1.6% of arable land has been used in recent years for biomass in the UK (DEFRA, 2019) but this needs to increase (Committee on Climate Change, 2019). Short rotation coppice (SRC), in which woody plants such as willow is grown on a 3–4 year cycle, provides high-volume short-term biomass yields but typically produces biomass of lower calorific value compared to short rotation forest (SRF). In SRF, single stemmed trees are grown over 10–20 years for either biomass or timber. This produces a better timber to bark ratio for higher biomass yields, is easily harvested and offers increased flexibility to growers in times of uncertain biomass markets (Keith et al., 2015; Leslie et al., 2012; McKay, 2011). The recent Committee on Climate Change report (2020) suggested that 0.7 million hectares of energy crops (*Miscanthus*, SRC or SRF) should be grown in the UK by 2050 as a 'Further Ambition' scenario in order to achieve net zero emissions and increase the domestic supply of biomass.

In 2010, Forest Research established SRF trials across the UK to determine biomass yields and assess the environmental impact of SRF (Harrison, 2010). The trials included a number of broadleaf tree species (hybrid aspen (*Populus tremula L.* x *tremuloides* Michx.), red alder (*Alnus rubra* Bong.), common alder (*Alnus glutinosa (L.) Gaertn*), Italian alder (*Alnus cordata* Desf.), sycamore (*Acer pseudoplatanus*), Sweet chestnut (*Castanea sativa* Mill.), eucalyptus spp. (*Eucalyptus gunnii*, *Eucalyptus nitens* (Vic*. nitens* (NSW), *E. glaucescens*) and the two conifer species Sitka spruce (*Picea sitchensis* Bong. Carr) and hybrid larch *(Larix x marschlinsii Coaz*) (Harrison, 2010). Sitka spruce is the most widely grown conifer tree species in the UK and a key plantation species. SRF plantations have previously been assessed for their environmental impact in the UK and Ireland (Keith et al., 2015; McKay, 2011; Tobin et al., 2016), but not for their potential future impacts on air quality in the UK, which is the focus of this work.

Trees are known sinks for CO_2 but can also be sources of other trace gases such as volatile organic compounds (VOCs) (Monson and Fall, 1989; Went, 1960). VOCs are emitted by tree foliage as a means of communication, plant defence against herbivory and during environmental stress such as heat or drought. Other sources of VOCs within a forest may include wood, litter, soils, fruits, flowers and roots (Dudareva et al., 2006). Emitted VOCs include, in particular, isoprene and monoterpenes, and their aliphatic, aromatic and oxygenated derivatives. These compounds are highly reactive in the atmosphere and contribute to the formation of tropospheric ozone in the presence of nitric oxide (NO) (Atkinson and Arey, 2003). Terpene composition has been found to be an important factor in the magnitude of ozone production (Bonn et al., 2017). Ground-level ozone is a concern for agriculture and natural ecosystems as it causes leaf damage, reduced plant growth (Emberson, 2020; Fares et al., 2013; Felzer et al., 2007) and is also a pollutant with impacts on human-health and as a greenhouse gas (UNEP/WMO, 2011). In addition, intermediates of VOC oxidation may act as condensation nuclei for the formation of secondary organic particles (Carlton et al., 2009), another atmospheric pollutant with detrimental effects on human health (Fuzzi et al., 2015).

The emissions of VOCs from plants are dependent upon a range of factors (which vary with emitting source and type of VOC) including species, plant age and environmental conditions such as light and temperature (Guenther et al., 1991; Monson and Fall, 1989) and, in the case of forest floor emissions, soil moisture, ambient temperature, soil type and the activity of the soil microbiome (Peñuelas et al., 2014). If the area of bioenergy crops expands, determining their VOC emissions becomes necessary for the wider assessment of air quality for a given region. Willow, a current UK bioenergy crop grown as SRC is a known emitter of VOCs (Morrison et al., 2016), but there is a lack of literature data generally for VOC emissions from trees in SRF plantations and from the forest floor.

In this study we focus on determining the contribution of the BVOC emissions from the two species with the largest growth in SRF trials in the UK: hybrid aspen and Italian alder (McEvoy, 2016; McKay, 2011; Parratt, 2018). In addition, we measured the BVOC emissions for young Sitka spruce plantations, also grown at the same location, as a comparison. Measurements were made in a plantation species-trial in central Scotland. Using dynamic enclosure sampling of BVOCs onto absorbent cartridges, the contribution of both foliage and forest floor emissions were measured simultaneously on occasions to form a plantation-scale assessment of BVOC emissions. The data were then used with the MEGAN 2.1 model (Guenther et al., 2012) to derive an estimate of the potential total annual contribution of expanded SRF to UK BVOC emissions.

2. Methods

2.1 Field site description

2.1.1 Tree species and planting

Measurements were made at East Grange, Fife, Scotland (Lat/Lon (WGS84) 56° 05' 21" N, 003° 37' 52" W), elevation 45–60 m, one of the 16 SRF trial locations established by Forest Research (Harrison, 2010; Stokes, 2015). Soil type and texture at the site is surface-water gley and sandy silty loam respectively, containing 4.9% clay, 53.0% silt and 42% sand (Drewer et al., 2017; Keith et al., 2015). In 2010, the ex-agricultural site was planted with a single block of 40 randomised tree species plots and 8 control plots. Each plot (20 m x 20 m) consisted of a single species containing 200 trees with a 2 m x 1 m spacing arrangement (Harrison, 2010). Ten species were planted, and the two broadleaved species with the best survival and growth rates across the trials in the first six years, hybrid aspen (*Populus tremula L. x tremuloides* Michx.) and Italian alder (*Alnus cordata* Desf.), were selected for the measurements here, along with Sitka spruce (*Picea sitchensis* Bong. Carr, produced by vegetative propagation) (McEvoy, 2016; Parratt, 2018). After initial establishment of the young

saplings, the site remained unmanaged. Branch and forest floor sampling chambers were installed in single south facing plots of each species.

2.1.2 Meteorological data

Meteorological data were collected from an unplanted plot in the middle of the site between May 2018 and July 2019. Minimum and maximum soil temperature (T107, Campbell Scientific, Shepshed, Leics, UK), air temperature and relative humidity (HMP45C, Campbell Scientific) were monitored hourly. In addition, photosynthetic active radiation (PAR, SKP 215 Quantum Sensor, Skye instruments, Llandrindod Wells, UK) was measured at the same site every 5 minutes. Monthly means and ranges are provided in Supplementary Information S1. Occasional power failure at the site led to some missing data. For the modelling of BVOC emissions using Pocket MEGAN 2.1 excel beta 3 calculator (Guenther et al., 2012)the missing PAR and mean temperature data were replaced by measurements from the Easter Bush site of the UK Centre for Ecology & Hydrology lying 45 km to the south east (Lat/Lon (WGS84) 55° 51′ 44″ N, 003° 12′ 20″ W). A summary of the combined East Grange and Easter Bush data used in the model can be found in Supplementary Information S2.

The climate in east Scotland is colder, with fewer sunshine hours than in the south of England. To encompass these climate differences, meteorological data from Alice Holt forest (51°09′13″N , 000°51′30″W), Hampshire, in southern England recorded during 2018 and 2019 was also used for the modelling and scaling up of the measured BVOC emission potentials from this study. A summary of the PAR and air temperature data for this field site is given in Supplementary Information S3.

2.2 Sampling enclosures

Branch sampling was conducted on the spruce, aspen and alder plantation plots on a total of 16, 11 and 13 days respectively between March 2018 and July 2019. The plantation floor sampling was conducted on a total of 18 (spruce and alder) and 20 days (aspen) for the same plots during the same period.

2.2.1 Forest floor enclosures

Forest floor in this context includes soil, leaf litter, fallen small twigs/branches and flowers, understorey vegetation, microorganisms and underground biomass that may all be sources of BVOC from the ground of the plantation. A static chamber method was used for the plantation floor enclosures. Polyvinylchloride plastic soil collars (with a flange), 40 cm diameter x 18 cm high, were installed per tree species plot prior to sampling (Asensio et al., 2007c, 2007b; Greenberg et al., 2012; Janson, 1993) and remained in the ground for the duration of the experiment. One or two collars were installed in 2017 and used during 2018. Additional collars were installed during 2018 resulting in a total of three soil collars per plot for the 2019 sampling. The collars were placed towards the centre of each plot to reduce the likelihood of plant debris from other plots contaminating them. Leaf litter and understorey vegetation were not removed from the collars prior to sampling to reflect actual changes in BVOC emissions with changes in the forest floor composition through the seasons.

A clear acrylic lid (with a foam lined flange), 40 cm diameter x 22.5 cm high, was placed over the soil collar during sampling periods only, enclosing a total chamber volume of 51 L. The lid was sealed using clamps around the rim. A small 12 V axial fan (RS components Ltd, Colby, UK), 4 cm x 4 cm x 1 cm, was attached to the chamber lid to mix the air inside the chamber (Janson, 1993). Samples of BVOC in the enclosed air were collected through PTFE tubing onto a 6 mm OD stainless steel automated thermal desorption (ATD) cartridge (PerkinElmer, Waltham, MA, USA) packed with 200 mg Tenax TA 60/80 (11982 SUPELCO, Sigma-Aldrich, St Louis, MO, USA) and 100 mg Carbotrap 20/40

(20273 SUPELCO, Sigma-Aldrich) at a flow rate of 0.2 L min^{-1} using a handheld pump (210-1003MTX, SKC ltd, Blandford Forum, UK). Samples were collected for 30 min after closure, equating to a total sample volume of 6 L. Pressure compensation was maintained through a small hole in the side of the chamber to prevent negative pressure inside the chamber and potential degassing of air from the soil pores. Ambient air samples were collected concurrently with the chamber sample in order to quantify BVOC emissions from the forest floor by difference. This is discussed further in Section 2.5.2. No ozone filter was used during sampling so amounts of some monoterpenes may have been reduced by reaction with ozone (Ortega et al., 2008). However, it has also been suggested that ozone may be lost by dry deposition onto the chamber walls in the first minute (Janson et al., 1999). Chamber air temperature (Electronic Temperature Instruments Ltd, Worthing, UK) and humidity (Fisherbrand™ Traceable™ Humidity Meter, Fisher Scientific, Loughborough, UK) were measured at the end of the 30 min sample collection period.

Volumetric soil moisture (ML3 ThetaProbe Soil Moisture, Delta T, Cambridge, UK) was measured at three locations around each chamber and soil temperature was measured at a single location at 7 cm depth close to, but outside the soil collar to avoid disturbance of the forest floor. Both measurements were performed after sample collection to prevent perturbation of the ambient air sample.

2.2.2 Branch enclosure

A dynamic chamber method was used for branch enclosures. Three sample points were established per tree species plot and used to mount a removable flow-through acrylic chamber (Potosnak et al., 2013), 53 L in volume. The chambers were set up during each sampling visit and used to enclose a single branch, alternating between three similar branches per tree species. The branches were selected to be of similar size and in a similar position on the tree. All branches were approximately 1.5 m from the ground and in a south-facing position. Ambient air flow was delivered from an oil-

free double-ended diaphragm pump (Capex V2, Charles Austen pumps Ltd, Surrey, UK) through PTFE tubing (Morrison et al., 2016; Purser et al., 2020) at a flow rate of 10 L min^{-1} to obtain the desirable air exchange rate of 4-5 minutes (Ortega and Helmig, 2008). In addition, the chamber contained a small 12 V axial fan (RS components Ltd, Colby, UK), 8 cm x 8 cm x 2.5 cm, to ensure sufficient mixing of air inside the chamber.

After set-up, the branch enclosure was left for a period of 30 min to attain a steady state. Both inside and outside of the enclosure were then sampled concurrently for 30 min at a flow rate of 0.2 L min^{-1} (total sample volume of 6 L) using a handheld pump (210-1003MTX, SKC Ltd, Blandford Forum, UK). In cases of low light levels, low temperatures or smaller volumes of foliage, the sampling time was sometimes extended (up to 60 minutes) to ensure sufficient sample was collected on the sample cartridge. Multiple sequential samples were taken over a given day. All enclosure sample tubes were stored in a fridge at 4 °C until analysis.

After BVOC sample collection, the leaves inside the chamber were counted and a representative subsample of approximately 10% of the total number of leaves on the measured branch removed from a nearby branch. The leaves were dried at 70 °C until constant mass, typically after 48 h. In the case of the Sitka spruce subsidiary branches were used. Measurements of chamber temperature and relative humidity (CS215, Campbell Scientific, Shepshed, UK) were made each minute during sampling. In addition, PAR (SKP 215 PAR Quantum Sensor, Skye instruments, Llandrindod Wells, UK) was measured outside but next to the branch chamber with measurements made every minute. The chambers had 85% transparency to PAR (400–700 nm), so the measured PAR values were correspondingly adjusted to represent the illumination conditions inside the chamber.

2.3 BVOC analysis

The BVOC samples collected on the sorbent were analysed using gas chromatography-mass spectrometry (GC-MS) with a two-stage automatic thermal desorption unit (ATD 400, Perkin-Elmer, Wellesley, MA, USA) using the method described in Purser et al. (2020). Calibration was carried out using standards (from Sigma-Aldrich, Gillingham, UK) of the monoterpenes α-pinene, β-pinene, d-limonene, α-phellandrene, β-phellandrene, 3-carene, camphene, γ-terpinene and β-myrcene, and the monoterpenoids (monoterpene-based compounds with, for example, additional oxygen or missing a methyl group) eucalyptol and linalool prepared as a mixed stock solution of 3 ng μL^{-1} in methanol. Aliquots of 1, 2, 3 and 4 µL of the mixed monoterpene stock solution were pipetted directly onto sample tubes under a flow of helium to produce a range of mixed monoterpene standards of 3, 6, 9 and 12 ng. Isoprene standards were prepared by direct sampling onto a sorbent tube from a certified 700 ppbv gas standard (BOC, UK) for 10, 30, 45 and 60 s using a sample pump (210-1003MTX, SKC ltd, Blandford Forum, UK) producing standards of 65, 198, 296 and 395 ng. Note that mass loadings of isoprene and monoterpene calibration standards were calculated to greater precision than quoted above but are shown here as nominal values for ease of discussion.

Unknown peaks in sample chromatograms were identified by comparison to the internal library of the GC-MS (National Institute of Standards and Technology) and by comparison with the retention time of the standard. The limit of detection (LOD) of the calculated measured emissions ranged from 0.12-0.35 µg C g_{dw}^{-1} h^{-1} for the branch chambers and 0.47-1.4 µg C m^{-2} h^{-1} for the forest floor chambers. Uncertainties on an individual calculated emission rates were 16% for isoprene and 17% for monoterpenes, which were derived via error propagation methods described in Purser et al. (Purser et al., 2020).

2.4 Calculation of standardised emissions

2.4.1 Forest floor BVOC emissions

As no substantial isoprene emissions were observed during an initial assessment, only monoterpenes were quantified from the forest floor. Monoterpene emissions from the forest floor (F_{floor}) were calculated as μg carbon for a given compound per ground surface area (μg C m^{-2} h^{-1}) using Eq. (1), where C_{sample} is the concentration of a monoterpene inside the chamber (μg C L^{-1}), $C_{ambient}$ is the concentration of a monoterpene in the ambient air outside the chamber (μg C L^{-1}), A is the area of forest floor inside the chamber (m^2), V is the volume inside the chamber, and , t is the sampling duration (mins).

$$F_{floor} = \frac{\left[C_{sample} - C_{ambient}\right] \times V \times 60}{A \times t} \quad (1)$$

In some cases, the concentration in ambient air was larger than inside resulting in a negative emission value, i.e. a net uptake.

2.4.2 Branch scale BVOC emissions

The isoprene or monoterpene emission (F_{branch}) from an enclosed branch was calculated as μg carbon (C) for a given compound per leaf dry mass basis, μg C $g(dw)^{-1}$ h^{-1}, using Eq. (2), where f is the flow rate through the chamber (L min^{-1}) and m is the dry mass (g) of foliage inside the chamber.

$$F_{branch} = \frac{\left[C_{sample} - C_{ambient}\right] \times f}{m} \quad (2)$$

Isoprene emissions have previously been shown to be controlled by both light and temperature and were standardised to 30 ^{o}C and 1000 μmol m^{-2} s^{-1}, respectively (Guenther et al., 1993b). Mean chamber air temperature and PAR for each period of sample collection were therefore used to standardise the measured F_{branch} emissions for isoprene (Eq. (3), (4) and (5)) and monoterpenes (Eq.

6) to facilitate comparison between this study and previous literature. The algorithms developed in Guenther et al. (1993b) are subsequently referred to as G93.

The standardised isoprene emission rate $F_{isoprene}$ at 30 ^{o}C and 1000 µmol m^{-2} s^{-1} PAR is a function of the measured emission F_{branch}, a term C_L to correct for the effect of light and a term C_T to correct for the effect of temperature Eq. (3).

$$F_{isoprene} = \frac{F_{branch}}{C_L \times C_T} \quad (3)$$

The light-correction term C_L is calculated from Eq. (4) where α = 0.0027 and C_{L1} = 1.066 are empirical coefficients in G93 and L is the experimentally-measured mean PAR (µmol m^{-2} s^{-1}) during sampling.

$$C_L = \frac{\alpha C_{L1} L}{\sqrt{1 + \alpha^2 L^2}} \quad (4)$$

The temperature-correction term C_T is calculated using Eq. (5) in which the terms C_{T1} (95000 J mol^{-1}), C_{T2} (230000 J mol^{-1}) and T_M (314 K) are all empirically-derived coefficients from G93. R is the molar gas constant 8.314 J K^{-1} mol^{-1}, T is the mean air temperature (K) during sampling, and T_s is the standardised temperature of 303.15 K, equivalent to 30 ^{o}C.

$$C_T = \frac{exp\frac{C_{T1}(T-T_S)}{RT_ST}}{1+exp\frac{C_{T2}(T-T_M)}{RT_ST}} \quad (5)$$

Monoterpene emissions from branch chambers, F_{branch} were standardised to temperature based on the calculations from G93 using Eq. (6). T_s is the standard temperature (303 K) and T is the mean air temperature during sampling. $F_{monoterpene}$ is the standardised monoterpene emission rate (µg C g$_{(dw)}^{-1}$ h^{-1}) and F_{branch} is the measured monoterpene emission rate (µg C g$_{(dw)}^{-1}$ h^{-1}).

$$F_{\text{branch}} = F_{\text{monoterpene}} \exp(\beta(\mathrm{T} - T_s)) \quad (6)$$

Standardised isoprene and monoterpene emission rates from sequential samples calculated for a given day were then averaged to give a single standardised branch emission rate per tree species per measurement day. In addition, daily measurements were grouped into seasons to give a standardised emission potential per season, F_{b_season}.

2.5 LAI determination

A Leaf Area Index (LAI) meter (LAI-2000 plant canopy analyser, LI-COR, Inc., Lincoln, NE, USA) was used to provide data to estimate a density of foliage, $m^2_{leaf}\ m^{-2}_{ground}$, for each species during two separate days, two weeks apart in July 2018, assumed to be the time of maximum foliage density (Ogunbadewa, 2012). LAI determinations were made in three hybrid aspen, two Sitka spruce and one Italian alder plots. Two above-canopy and eight below-canopy points were measured per plot, with a mixture of within and between row measurements. Where more than one plot was measured for a species, the mean LAI is reported.

2.6 Scaling up from emission per mass of foliage to an emission per area of ground of plantation

The standardised emissions of isoprene and monoterpenes from the canopy ($\mu g\ C\ m^{-2}_{ground}\ h^{-1}$), $F_{foliage}$, was determined using Eq. (7), multiplying standardised summertime branch emission measurements (F_{b_summer}) calculated in Section 2.5.2 with literature values of the leaf mass per leaf area (LMA) for each tree species (Table 1) and the measured LAI. As there was limited LMA data for Italian alder under climate conditions relevant for the UK, additional values were taken from literature on common alder (*Alnus glutinosa*). The LMA multiplied by the LAI gives the mass of

315 foliage per unit area of ground, known as the foliar biomass density. The calculated foliar biomass
316 density values in Table 1 for hybrid aspen (329 g m^{-2}) and Italian alder (315 g m^{-2}) are very similar to
317 the 320 g m^{-2} (Karl et al., 2009) and 375 g m^{-2} (Geron et al., 2000) used in previous modelling studies
318 for these two tree species. For Sitka spruce the foliage biomass density used here (619 g m^{-2}) is
319 about half that for the same species in previous modelling studies, 1500 g m^{-2} (Geron et al., 2000;
320 Karl et al., 2009) and reflects the immature Sitka spruce stand not yet achieving a closed canopy.

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322 $$F_{\text{foliage}} = F_{\text{b\_summer}} \times LMA \times LAI \quad (7)$$

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324 For times when the plantation canopy consisted of flowers only (catkins) or early leaf emergence,
325 during the months February to April on deciduous species, a different approach had to be applied. In
326 these instances the LAI was either reduced to reflect the canopy during leaf emergence or the
327 following estimate for catkins was applied. We assumed that there were approximately 66 catkins
328 per m^{-2} per ground area of the plantation canopy based on similar catkin forming species
329 (Boulanger-Lapointe et al., 2016). This equates to a catkin biomass density, for converting from
330 branch-scale to canopy-scale purposes, of 8.98 g m^{-2}_{ground} based on the mean mass of an alder catkin
331 measured during our study.

332

333 In measurements of LAI by Ogunbadewa et al. (2012), taken across a year in a deciduous forest in
334 the UK, the LAI was at its maximum by July and during spring the LAI increased such that it was
335 around a quarter of the maximum by late April and around a half by mid-May. These seasonal
336 changes in LAI were therefore adopted for use in the MEGAN 2.1 model (Table 2) in the absence of
337 multiple seasonal LAI measurements taken at East Grange during our study.

338 **Table 1 – Leaf mass per area data for calculating foliage emission rates per plantation ground area.**

Tree species	LMA / g m^{-2}_{leaf}	Literature source	Country of origin of literature measurement	Forest type	Stand age / years	Measured LAI during this study	Foliar biomass density / g m^{-2}_{ground}
Hybrid aspen	98.0	(Tullus et al., 2012)	Estonia	Trial plantation	4		
	73.5	(Yu, 2001)	Finland	Clone trial	1.5		
	61.7	(Johansson, 2013)	Sweden	SRF Plantation	15-23		
Mean	77.7	-	-	-	-	4.24	329
RSD / %	24						
Sitka spruce	222	(Norman and Jarvis, 1974)	NS	Plantation	NS		
	160	(Meir et al., 2002)	Scotland	Plantation	13		
	200	(Foreman, 2019)	Ireland	Greenhouse trial	3		
Mean	194	-	-	-	-	3.19	619
RSD / %	16						
Italian alder	114**	(Leslie et al., 2017)	England	Trial Plantation	2		
	102*	(Foreman, 2019)	Ireland	Greenhouse trial	2		
	75.1**	(Johansson, 1999)	Sweden	Plantation	21-91		
Mean	97.0	-	-	-	-	3.25	315
RSD / %	21						

*Mean of sun and shade leaves. NS = Not specified, RSD = relative standard deviation.

**Measurements from common alder (*Alnus glutinosa*)

2.7 From canopy emission to total annual emissions per hectare and the influence of increasing biomass planting on total UK BVOC emissions

Standardised foliage emission rates, $F_{foliage}$, for summer 2018 and 2019 (Table 3) were input to the Pocket MEGAN 2.1 excel beta 3 calculator (Guenther et al., 2012) with hourly mean PAR and temperature data from East Grange (gap filled with UKCEH site data), LAI and the other variables given in Table 2. For a detailed description of the equations and algorithms used in MEGAN 2.1 see Guenther et al. (Guenther et al., 2006, 2012). The model adjusts the standardised emission rate input in accordance with air temperature and PAR from the meteorology inputs per hour to produce a likely emission rate for the plantation. Input LAI measurements for alder and aspen were scaled in spring and autumn by 25% and 50% to simulate leaf emergence and senescence (Table 2). The LAI of Sitka spruce was assumed to remain constant through the seasons although it is recognised there will be a small increase in the spring, and a later decline. No LAI measurements were made in 2019 therefore 2018 measurements were used. The function that accounts for the effect of both the

356 previous 24 hours and 240 hours of light on the calculated emissions was applied in the model. The
357 latitude was set to 56^{o} for Scotland and 51^{o} for England and the vegetation cover was set to 1. The
358 functions in MEGAN2.1 that allow for consideration of soil moisture and CO_2 concentrations were
359 not used due to a lack of continuous data available for the field sites. The monoterpenes in the
360 model were calculated using the single value for mean total monoterpene from East Grange and
361 using the category named “other monoterpenes”. Although some individual monoterpene
362 compounds may be produced in the leaves in response to light and temperature to varying degrees,
363 due to the use of the collective “total monoterpenes” as a model input the simplification was used
364 that monoterpene emissions were driven by temperature only and no light specific emission factor
365 was applied.(Guenther et al., 2006, 2012, 1993a). Any other model input parameters remained as
366 default.

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368 The model output of hourly isoprene and total monoterpene emissions were summed to give annual
369 emissions per m^2 of SRF plantation. The combined mean total annual emission rate encompassing
370 both years of emission potentials (2018 and 2019) and meteorology from two contrasting UK sites
371 (E. Scotland and S.E. England), for each SRF species, was then compared to literature values for the
372 estimated annual UK isoprene and monoterpene emissions and combined total BVOC emissions.

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Table 2 – Seasonal time course of leaf area index (LAI) for estimating annual VOC emissions for different species plots at East Grange, Fife, Scotland, using MEGAN 2.1 model.

Date	Day of year	Sitka LAI	Aspen LAI	Alder LAI
1st January	1	3.19	0	0
19th February	50	3.19	0	0
31st March	90	3.19	0	0
19th April	109	3.19	1.06	0.81
30th April	120	3.19	2.12	1.63
1st June	152	3.19	3.18	2.43
15th July	196	3.19	4.24	3.25
1st August	213	3.19	4.24	3.25
1st September	244	3.19	3.18	2.43
20th October	304	3.19	1.06	0.81
31st October	334	3.19	0	0
31st December	366	3.19	0	0

Table 3 – Input parameters for estimating annual BVOC emissions for different SRF species plots at East Grange, Fife, Scotland using the MEGAN 2.1 model.

	Spruce		Aspen		Alder	
Emission rate (per unit ground area)	2018	2019	2018	2019	2018	2019
Isoprene / mg m^{-2}_{ground} h^{-1}	9.31	4.23	7.74	7.30	0.01	0.01
Total monoterpene / mg m^{-2}_{ground} h^{-1}	2.81	1.45	0.09	0.03	0.22	0.07

3. Results and discussion

3.1 Field observations of seasonality

The measured BVOC emissions were assigned to seasons as follows: winter (21st December – 19th March), spring (20th March – 07th June), summer (08th June – 22nd September) and autumn (23rd September – 20th December). 2018 is classified here as a dry year, being 25% drier at the East Grange field site than the 30 year mean for the area (Met Office, 2020). In contrast, 2019 was 50% wetter than the 30 year UK mean. In 2019, catkins were fully developed on the hybrid aspen and

Italian alder branches by February, but bud burst and leaf emergence was not observed until mid-April (19th). This was two weeks later than in 2018. The first new growth on the Sitka spruce was observed at the end of April (29th). Based on these differences in phenology at the site, measurements taken on 7th June 2019 was still categorised as spring.

For the forest floor it was noted that the soil temperatures during summer 2018 were higher than in 2019. After several dry weeks in spring and summer in 2018, the first significant rainfall event since May was noted as 14th July, and some leaf fall in the Italian alder and hybrid aspen plots was observed by the end of July. By February 2019, no leaf litter from the previous autumn season was observed on the forest floor of the plots except for those of Sitka spruce. Rapid understorey growth identified as hogweed (*Heracleum sp)* quickly developed from late April (29th) and by early June (7th) completely covered the forest floor in the alder plots. The hybrid aspen and Sitka spruce plots during both 2018 and 2019 had minimal understorey vegetation by comparison.

3.2 Leaf area index

The LAI of 3.19 for our 8-y old Sitka spruce plantation (Table 1) is lower than the value of 4.33 predicted for a 10-y old plantation from allometric relationships (Tobin et al., 2007). However, our measured LAI reflects a canopy not yet fully closed and the differences in site conditions are likely to produce different growth rates.

A maximum LAI of 4 was reported for a 9-y old aspen (*Populus tremuloides Michx.*) plantation in Canada (Pinno et al., 2001), which compares well with the LAI of 4.24 measured here (Table 1).

A 4-y old SRF plantation of Italian alder established in Ireland that was also measured in July gave an LAI of 2.8 or 3.4 for a 2 x 2 m or a 1 x 1m plant spacing respectively (Foreman, 2019). Other alder species such as common (or black) alder (*Alnus glutinosa)* and grey alder (*Alnus incana)* in Sweden had LAI values of 2.85 and 3.04, respectively; all comparable to the Italian alder LAI of 3.25 measured

here (Table 1). A study of SRF planting density trials in Ireland found that above-ground biomass growth was similar for Italian alder compared to Sitka spruce (Foreman, 2019) which also aligns well with our observations.

3.3 BVOC emissions from tree branches

3.3.1 Italian alder

Italian alder (*Alnus cordata*) emitted very low amounts of isoprene, ranging between <0.0005 – 0.035 µg C g_{dw}^{-1} h^{-1} (standardised 0.017–0.037 µg C g_{dw}^{-1} h^{-1}) depending on season (Table 4), comparable with previous standardised emission rates reported as <0.1–3 µg g_{dw}^{-1} h^{-1} (0.09 – 2.64 µg C g_{dw}^{-1} h^{-1}) (Calfapietra et al., 2009). The equivalent median and interquartile ranges for the data collected during this study can be found in the Supplementary Information S4.

Mean measured emissions for total monoterpene ranged between 0.041–0.393 µg C g_{dw}^{-1} h^{-1} (standardised 0.073–1.5 µg C g_{dw}^{-1} h^{-1}) with higher emission rates during spring and summer 2018 than in 2019. The major monoterpenes emitted were d-limonene, α-pinene, β-myrcene and β-pinene, which were consistently emitted through the spring and summer (Figure 1). No previous data for total or speciated monoterpene emission rates from Italian alder could be found in the literature. However, other alder species have also been reported to be low emitters of monoterpenes, and to emit slightly more monoterpenes than isoprene. Studies that report similar low levels of total monoterpene standardised emissions from alder include 0.8 µg C g_{dw}^{-1} h^{-1} from grey alder (Hakola et al., 1999), 0.13 µg C g_{dw}^{-1} h^{-1} from black (or common) alder (Aydin et al., 2014) and 1–2 µg C g_{dw}^{-1} h^{-1} from green alder (*Alnus rugosa*) (Isebrands et al., 1999). For speciated emissions, 3-carene, β-phellandrene, β-ocimene, p-cymene, sabinene have also been reported to be emitted from *alder spp*. (Aydin et al., 2014; Copolovici et al., 2014; Hakola et al., 1999; Huber et al., 2000). Emissions of some monoterpenes such as β-myrcene are suggested to be induced by herbivory by aphids (Blande et al., 2010). However, since no data on the composition of monoterpenes under laboratory studies in the absence of herbivory is available for Italian alder it is

446 difficult to know which, if any, of the monoterpenes measured in our field study may have been

447 induced by previous herbivory.

448 **Table 4 – Mean seasonal BVOC emissions (µg C g^{-1} h^{-1}) from branches of Sitka spruce, hybrid aspen**
449 **and Italian alder in SRF plantations, East Grange, Fife, Scotland. Figures in parentheses are**
450 **standard deviations.**

	Spring 2018			Summer 2018			Autumn 2018			Winter 2019			Spring 2019			Summer 2019		
	Sitka spruce	Hybrid aspen	Italian alder	Sitka spruce	Hybrid aspen	Italian alder	Sitka spruce	Hybrid aspen	Italian alder	Sitka spruce	Hybrid aspen	Italian alder	Sitka spruce	Hybrid aspen	Italian alder	Sitka spruce	Hybrid aspen	Italian alder
Days	4	1	1	2	4	3	-	-	-	3	-	2	4	3	4	2	2	4
N	18	5	4	12	18	12		-	-	10	-	8	8	10	10	7	7	13
chamber T / °C	15.4 (7.3)	29.9 (1.4)	20.1 (3.1)	24.7 (8.9)	23.8 (5.6)	30.6 (3.0)	-	-	-	19.3 (5.2)	-	16.9 (2.0)	25.5 (7.1)	23.0 (3.1)	22.6 (3.7)	30.1 (6.1)	29.9 (4.7)	26.5 (7.4)
PAR / µmol m^{-2} s^{-1}	607 (464)	957 (214)	362 (166)	662 (530)	539 (380)	1018 (447)	-	-	-	394 (217)	-	298 (106)	934 (481)	882 (357)	1081 (331)	977 (609)	957 (368)	866 (397)
chamber RH / %	65 (16)	66 (2)	82 (4)	62 (13)	67 (17)	39 (9)	-	-	-	66 (4)	-	74 (4)	49 (10)	78 (17)	61 (17)	69 (17)	66 (6)	59 (20)
Isoprene	0.365 (0.864)	3.091 (0.961)	0.010 (0.008)	5.904 (3.221)	21.115 (17.304)	0.035 (0.080)	-	-	-	0.031 (0.048)	-	0.011 (0.000)	1.526 (1.887)	0.053 (0.038)	0.017 (0.020)	3.639 (1.872)	14.547 (18.616)	0.000 (0.014)
Standardised Isoprene	0.688 (1.384)	3.163 (0.620)	0.060 (0.051)	15.046 (8.307)	23.487 (11.057)	0.037 (0.071)	-	-	-	0.139 (0.183)	-	0.000 (0.000)	1.830 (1.725)	0.186 (0.130)	0.048 (0.064)	6.833 (7.013)	22.149 (18.159)	0.017 (0.043)
Total MT	0.325 (1.045)	0.082 (0.042)	0.268 (0.114)	2.609 (2.888)	0.201 (0.251)	0.393 (0.340)	-	-	-	0.428 (0.902)	-	0.039 (0.029)	1.458 (1.317)	0.040 (0.069)	0.041 (0.039)	2.314 (1.517)	0.062 (0.077)	0.095 (0.366)
Standardised Total MT	1.949 (7.145)	0.090 (0.046)	0.711 (0.434)	4.534 (4.817)	0.259 (0.361)	1.503 (2.823)	-	-	-	0.665 (1.257)	-	0.478 (0.406)	1.913 (2.220)	0.082 (0.103)	0.075 (0.073)	2.344 (1.652)	0.087 (0.069)	0.212 (0.720)
α-pinene	0.035 (0.101)	0.000 (0.010)	0.049 (0.029)	0.158 (0.105)	0.034 (0.037)	0.063 (0.052)	-	-	-	0.012 (0.020)	-	0.019 (0.011)	0.026 (0.022)	0.009 (0.017)	0.013 (0.012)	0.189 (0.304)	0.006 (0.009)	0.047 (0.191)
Standardised α-pinene	0.202 (0.600)	0.004 (0.008)	0.126 (0.094)	0.280 (0.148)	0.044 (0.038)	0.236 (0.506)	-	-	-	0.026 (0.035)	-	0.070 (0.076)	0.036 (0.015)	0.024 (0.025)	0.024 (0.025)	0.221 (0.069)	0.011 (0.011)	0.106 (0.375)
β-pinene	0.006 (0.018)	0.003 (0.002)	0.000 (0.001)	0.025 (0.017)	0.005 (0.006)	0.004 (0.007)	-	-	-	0.005 (0.008)	-	0.003 (0.002)	0.013 (0.011)	0.001 (0.001)	0.001 (0.001)	0.070 (0.102)	0.002 (0.002)	0.001 (0.005)
Standardised β-pinene	0.036 (0.0124)	0.003 (0.002)	0.000 (0.000)	0.044 (0.025)	0.007 (0.006)	0.005 (0.004)	-	-	-	0.008 (0.012)	-	0.028 (0.029)	0.018 (0.022)	0.002 (0.002)	0.002 (0.002)	0.077 (1.06)	0.002 (0.002)	0.003 (0.009)
camphene	0.030 (0.088)	0.002 (0.001)	0.001 (0.007)	0.133 (0.099)	0.005 (0.009)	0.046 (0.061)	-	-	-	0.006 (0.012)	-	0.001 (0.001)	0.010 (0.007)	0.000 (0.000)	0.000 (0.000)	0.040 (0.055)	0.000 (0.001)	0.001 (0.003)
Standardised camphene	0.175 (0.599)	0.002 (0.001)	0.006 (0.008)	0.237 (0.148)	0.008 (0.009)	0.058 (0.060)	-	-	-	0.019 (0.035)	-	0.001 (0.003)	0.014 (0.015)	0.000 (0.001)	0.000 (0.000)	0.056 (0.068)	0.000 (0.001)	0.002 (0.006)
β-myrcene	0.174 (0.592)	0.025 (0.017)	0.02 (0.008)	1.772 (2.329)	0.010 (0.011)	0.149 (0.162)	-	-	-	0.264 (0.599)	-	0.001 (0.001)	0.850 (0.806)	0.000 (0.001)	0.001 (0.001)	0.884 (0.425)	0.001 (0.002)	0.001 (0.003)
Standardised β-myrcene	1.070 (4.052)	0.025 (0.0018)	0.051 (0.014)	3.055 (3.741)	0.013 (0.0012)	0.177 (0.132)	-	-	-	0.392 (0.839)	-	0.009 (0.003)	1.097 (1.256)	0.001 (0.002)	0.002 (0.003)	0.807 (0.279)	0.002 (0.002)	0.002 (0.006)

451 Values shown as 0.000 = <0.0005, - = Not measured, MT = Monoterpene

452 **Table 4 continued.**

	Spring 2018			Summer 2018			Autumn 2018			Winter 2019			Spring 2019			Summer 2019		
	Sitka spruce	Hybrid aspen	Italian alder	Sitka spruce	Hybrid aspen	Italian alder	Sitka spruce	Hybrid aspen	Italian alder	Sitka spruce	Hybrid aspen	Italian alder	Sitka spruce	Hybrid aspen	Italian alder	Sitka spruce	Hybrid aspen	Italian alder
α-phellandrene	0.000 (0.000)	0.000 (0.000)	0.001 (0.001)	0.015 (0.012)	0.000 (0.000)	0.000 (0)	-	-	-	0.001 (0.002)	-	0.000 0.000	0.003 (0.003)	0.000 (0.000)	0.000 (0.000)	0.013 (0.006)	0.000 (0.001)	0.000 (0.001)
Standardised α-phellandrene	0.000 (0.000)	0.000 (0.000)	0.001 (0.002)	0.028 (0022)	0.000 (0.000)	0.002 (0.006)	-	-	-	0.001 (0.003)	-	0.003 (0.004)	0.003 (0.003)	0.000 (0.000)	0.000 (0.000)	0.013 (0.006)	0.000 (0.001)	0.001 (0.002)
β-phellandrene	0.000 (0.000	0.000 (0.000	0.000 (0.000)	0.020 (0.011)	0.009 (0.011)	0.000 (0.00)	-	-	-	0.003 (0.006)	-	0.001 (0.000)	0.007 (0.006)	0.008 (0.018)	0.000 (0.000)	0.017 (0.009)	0.007 (0.010)	0.000 (0.004)
Standardised β-phellandrene	0.000 (0.000)	0.000 (0.000	0.000 (0.000)	0.035 (0.021)	0.008 (0.009)	0.000 (0.000)	-	-	-	0.004 (0.008)	-	0.000 (0)	0.010 (0.014)	0.012 (0.025)	0.000 (0.000)	0.016 (0.007)	0.008 (0.011)	0.001 (0.002)
d-limonene	0.078 (0.243)	0.047 (0.015)	0.160 (0.102)	0.426 (0.270)	0.108 (0.229)	0.092 (0.140)	-	-	-	0.120 (0.239)	-	0.015 (0.011)	0.398 (0.351)	0.004 (0.009)	0.014 (0.015)	0.958 (0.886)	0.014 (0.017)	0.022 (0.062)
Standardised d-limonene	0.460 (1.662)	0.048 (0.019)	0.426 (0.338)	0.748 (0.427)	0.143 (0.339)	0.876 (1.964)	-	-	-	0.185 (0.329)	-	0.285 (0.255)	0.588 (0.837)	0.010 (0.020)	0.024 (0.024)	1.039 (0.987)	0.023 (0.015)	0.040 (0.123)
eucalyptol	0.001 (0.003)	0.007 (0.003)	0.004 (0.002)	0.053 (0.110)	0.012 (0.013)	0.016 (0.016)	-	-	-	0.014 (0.024)	-	0.000 (0.020)	0.145 (0.384)	0.010 (0.023)	0.000 (0.001)	0.114 (0.088)	0.003 (0.04)	0.000 (0.001)
Standardised eucalyptol	0.006 (0.002)	0.007 (0.003)	0.010 (0.006)	0.094 (0.056)	0.015 (0.015)	0.030 (0.042)	-	-	-	0.023 (0.037)	-	0.010 (0.007)	0.139 (0.033)	0.016 (0.033)	0.000 (0.001)	0.092 (0.062)	0.005 (0.008)	0.001 (0.001)
3-carene	0.000 (0.000)	0.000 (0.004)	0.035 (0.008)	0.008 (0.009)	0.017 (0.013)	0.023 (0.039)	-	-	-	0.003 (0.006)	-	0.014 (0.003)	0.006 (0.006)	0.002 (0.003)	0.009 (0.013)	0.017 (0.015)	0.005 (0.007)	0.025 (0.101)
Standardised 3-carene	0.000 (0.000)	0.001 (0.03)	0.090 (0.042)	0.013 (0.007)	0.021 (0.013)	0.118 (0.247)	-	-	-	0.006 (0.008)	-	0.065 (0.062)	0.008 (0.008)	0.005 (0.003)	0.014 (0.017)	0.014 (0.009)	0.007 (0.006)	0.056 (0.198)
linalool	0.000 (0.000)	0.000 (0.000)	0.000 (0.000)	0.000 (0.000)	0.000 (0.000)	0.000 (0.000)	-	-	-	0.000 (0.001)	-	0.000 (0.000)	0.000 (0.001)	0.006 (0.010)	0.003 (0.005)	0.008 (0.006)	0.024 (0.030)	0.000 (0.000)
Standardised linalool	0.000 (0.000)	0.000 (0.000)	0.000 (0.000)	0.000 (0.000)	0.000 (0.000)	0.000 (0.000)	-	-	-	0.001 (0.001)	-	0.002 (0.002)	0.000 (0.001)	0.012 (0.024)	0.007 (0.013)	0.006 (0.004)	0.029 (0.003)	0.000 (0.001)
γ-terpinene	0.000 (0.000)	0.00 (0.00)0	0.000 (0.000)	0.000 (0.000)	0.000 (0.000)	0.000 (0.000)	-	-	-	0.000 (0.000)	-	0.000 (0.000)	0.000 (0.000)	0.000 (0.000)	0.000 (0.000)	0.004 (0.003)	0.000 (0.001)	0.000 (0.000)
Standardised γ-terpinene	0.000 (0.000)	0.000 (0.000)	0.000 (0.000)	0.000 (0.000)	0.000 (0.000)	0.000 (0.000)	-	-	-	0.000 (0.000)	-	0.003 (0.005)	0.000 (0.000)	0.000 (0.000)	0.000 (0.001)	0.003 (0.002)	0.000 (0.001)	0.000 (0.001)

453 Values 0.000 = <0.0005, - = Not measured, MT = Monoterpene

454

3.3.2 Hybrid aspen

Measured isoprene emissions from hybrid aspen ranged from 0.053 to 21 µg C g_{dw}^{-1} h^{-1} (standardised 0.19–23 µg C g_{dw}^{-1} h^{-1}) (Table 4). No measurements were made during autumn senescence or in winter on the bare branches. Measured emissions were lower in spring for the newly emerged leaves compared to summer (Figure 1). As noted in Section 3.1, the onset of spring at the field site was earlier in 2018 compared to 2019. European aspen (*Populus tremula*) measured in late spring (May) two weeks after bud burst has also previously been reported to have a lower emission rate than in summer (Hakola et al., 1998). Isoprene emission rates made on leaves (not branches) on aspen in spring in the boreal forest were also reported to be a third of the emission rate measured in the middle of summer (Fuentes et al., 1999). In our study, the hybrid aspen plantation showed signs of stress thought to be associated with lower rainfall and soil moisture locally during summer 2018 causing a yellowing of leaves and early leaf shedding in July. It is widely accepted that isoprene emissions increase with increases in temperature and PAR (Guenther et al., 1991; Monson and Fall, 1989) but that under stress during drought, isoprene can be emitted at much higher rates than usual, only to eventually decline as resources are depleted in the leaves (Brilli et al., 2007; Seco et al., 2015). However, standardised isoprene emissions measured during this study on green aspen leaves did not differ between the two years, 2018 (23 µg C g_{dw}^{-1} h^{-1}) and 2019 (22 µg C g_{dw}^{-1} h^{-1}) despite the signs of stress in 2018 noted above. The standardised isoprene emissions for hybrid aspen reported here were much lower than those previously reported for European aspen, 51 µg g_{dw}^{-1} h^{-1} (i.e. 45 µg C g_{dw}^{-1} h^{-1}) (Hakola et al., 1998).

Total monoterpene emissions measured for hybrid aspen ranged from 0.040 - 0.20 µg C g_{dw}^{-1} h^{-1} (standardised 0.082 - 0.259 µg C g_{dw}^{-1} h^{-1}) with substantially higher emissions occurring in summer 2018 (Table 4, Figure 1). Increased emissions for some monoterpenes have been shown to be predominately driven by increases in temperature (Guenther et al., 1991). In particular d-limonene,

480 the major monoterpene emitted here, was found to correlate with an increase in temperature,
481 comparable to elevated temperature experiments for European aspen (Hartikainen et al., 2009).
482 However, total monoterpene emission rates were an order of magnitude lower in summer during
483 our study, closer to the findings of Brilli et al. (2014) from a SRC plantation of poplar, and in contrast
484 to the 4.6 μg g_{dw}^{-1} h^{-1} (4.1 μg C g_{dw}^{-1} h^{-1}) reported for European aspen by Hakola et al. (1998). D-
485 limonene, α-pinene, carene and β-phellandrene collectively accounted for 50–95% of the total
486 measured monoterpene emissions, although the composition for different days was highly variable
487 (Figure 1). Emissions of α-phellandrene peaked at 27% of total monoterpenes measured in April
488 when catkins were present but were otherwise < 13% (except on 6 June 2018).

489

490 Previously studies on European aspen report monoterpene emissions of 3-carene, limonene, α-
491 pinene, trans-ocimene, eucalyptol, β-myrcene and sabinene (Aydin et al., 2014; Hakola et al., 1998;
492 Hartikainen et al., 2009) and on hybrid aspen (*Populus tremula* – *Populus tremuloides*) report α-
493 pinene, β-pinene and β-ocimene, (Blande et al., 2007), although differences between clones were
494 noted.

495

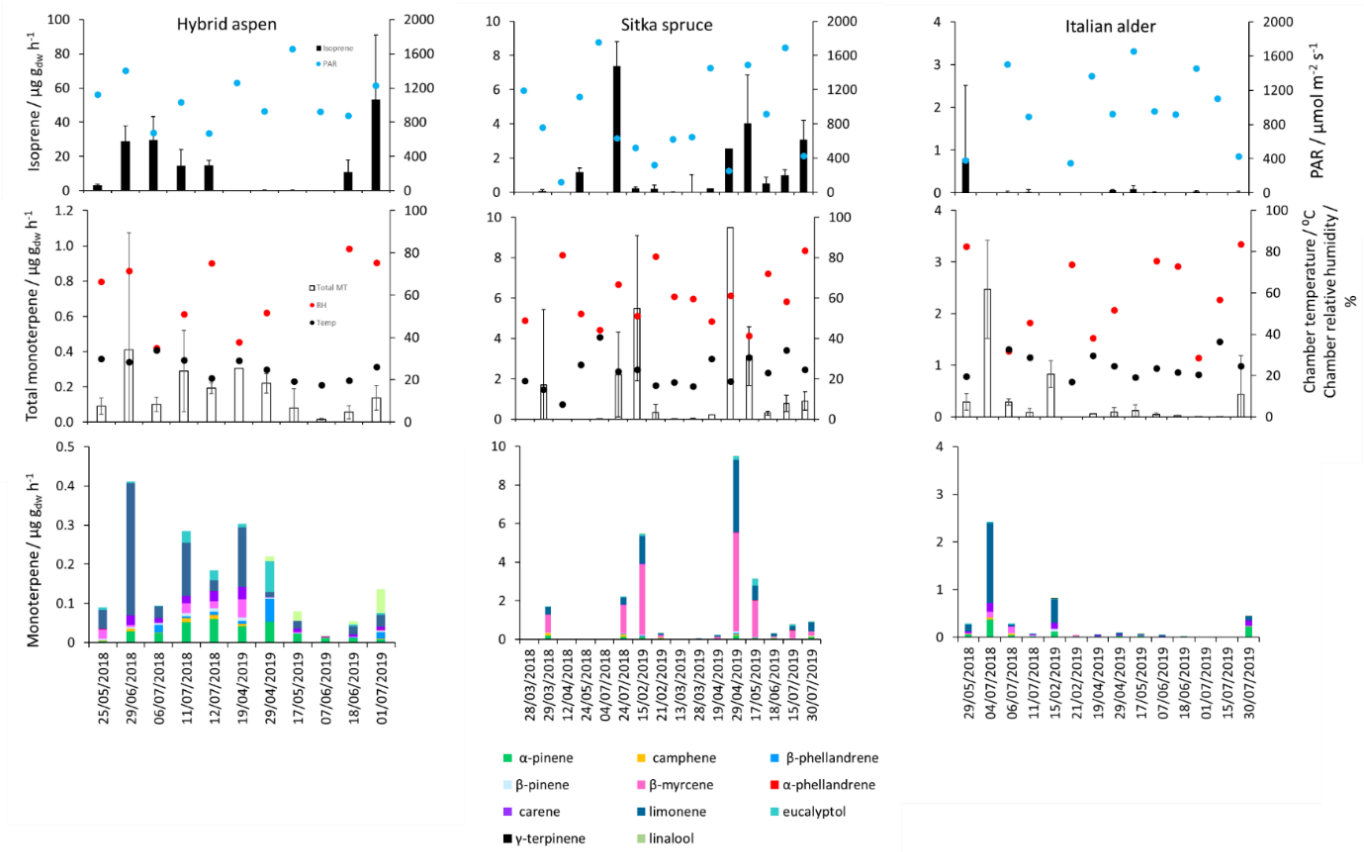


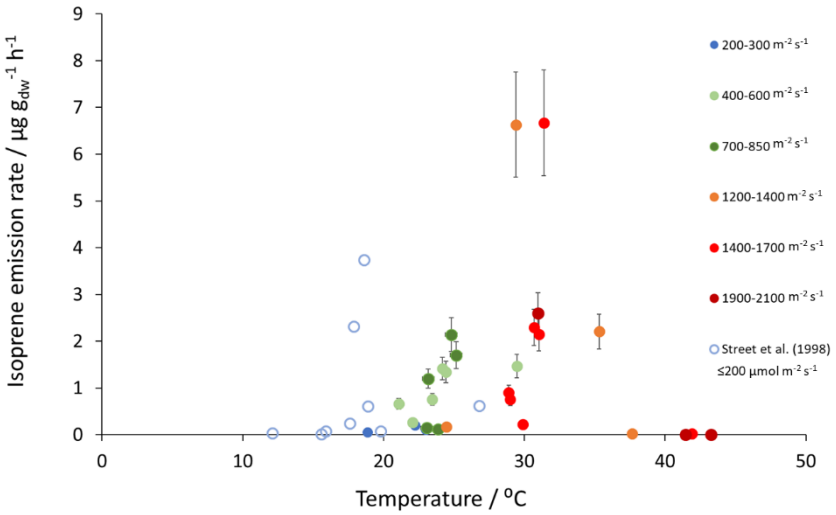
Figure 1 – Mean isoprene, total monoterpene and speciated standardised monoterpene emissions from branches of hybrid aspen, Italian alder and Sitka spruce trees in SRF plantations at the East Grange site, Fife, between March 2018 and July 2019. Error bars show standard deviation of all measurements made on a given day. Blue, red and black circles show mean PAR, chamber relative humidity and temperature, respectively. Note that emission scales differ between tree species

3.3.3 Sitka spruce

Mean measured isoprene emissions from Sitka spruce ranged from 0.031 µg C g_{dw}^{-1} h^{-1} (standardised 0.14 µg C g_{dw}^{-1} h^{-1}) in winter to 5.9 µg C g_{dw}^{-1} h^{-1} (standardised 15.0 µg C g_{dw}^{-1} h^{-1}) in summer (Table 4), which are comparable to the range of previously reported emissions from UK field measurements, 0.005-1.48 µg g_{dw}^{-1} h^{-1} (standardised 0.88–14.1 µg C g_{dw}^{-1} h^{-1}) (Street et al., 1996). Standardised isoprene emissions were lower in spring than summer during both years in our study (Figure 1). Standardised isoprene emissions in summer 2018 (15.0 µg C g_{dw}^{-1} h^{-1}) were more than twice those in summer 2019 (6.8 µg C g_{dw}^{-1} h^{-1}), likely reflective of the wetter and cooler conditions in 2019. However, laboratory measurements using trees acclimatised at a constant laboratory temperature of 20 °C and PAR of 1000 µmol m^{-2} s^{-1} for a week prior to sampling showed emission

512 rates similar to summer 2018 emission rates, 13.4 µg g_{dw}^{-1} h^{-1} (11.8 µg C g_{dw}^{-1} h^{-1}) (Hayward et al,
513 2004). The measured isoprene emissions in our study declined dramatically at higher chamber
514 temperatures, > 31 ^{o}C , despite the high PAR levels. An optimum of 33 ^{o}C for isoprene emissions
515 from Sitka spruce was noted by Street et al. (1996), although a higher optimum of 39 ^{o}C was
516 suggested by Hayward et al. (2004) based on a laboratory study. We therefore suggest that Sitka
517 spruce trees acclimatised under field conditions in Scotland with variable day and night
518 temperatures and light levels, may have a lower optimum temperature than observed under
519 laboratory conditions. The previous suggestion that Sitka spruce reaches maximum emissions of
520 isoprene at a low level of PAR of 300 µmol m^{-2} s^{-1} (Hayward et al., 2004) was difficult to confirm
521 under field conditions as high PAR values were correlated with high temperatures (Figure 2).
522 However, it is worth noting that the majority of field emissions collected by Street et al. (1996) align
523 well with the emissions measured at lower PAR and temperature in this study (Figure 2).

524



525

526 **Figure 2 –measured isoprene emissions as a function of PAR and temperature for Sitka spruce at**
527 **East Grange SRF site and from Street et al. (1996) at PAR ≤ 200 µmol m^{-2} s^{-1}.**

528

529 Total monoterpene emissions measured from Sitka spruce peaked on the 29th April 2019 (9.5 µg C
530 g_{dw}^{-1} h^{-1}) coinciding with the new shoot extension growth on the branches (Figure 1). Monoterpene

emissions have shown to be present in spring in advance of isoprene emissions for Norway spruce (*Picea abies*) (Hakola et al., 2003). Overall, monoterpene emissions were generally higher in summer than in spring (Table 4). Total monoterpene emissions were still higher in 2018 (standardised 4.5 µg C g_{dw}^{-1} h^{-1}) than in 2019 (2.3 µg C g_{dw}^{-1} h^{-1}) even once standardised to 30 ^{o}C, which could indicate an increased release of monoterpenes in response to the drier warmer conditions. The total monoterpene emissions in 2019 are comparable to the previously reported total monoterpene emission of 3.0 µg g_{dw}^{-1} h^{-1} (2.6 µg C g_{dw}^{-1} h^{-1}) from a laboratory study (Hayward et al., 2004). Monoterpene emissions from Sitka spruce comprised predominately of β-myrcene, d-limonene, α-pinene and eucalyptol, collectively accounting for 83–97% of total monoterpenes across all measurement days (Figure 1).

β-myrcene was the most abundant, consistent with the findings of Geron et al. (2000), and has been reported to be highest during spring in leaf oils, associated with new growth in this species, only to decline later in the growing season (Hrutfiord et al., 1974) but this was not evident during our study. d-limonene emission rates reported during our study are comparable in size to Hayward et al. (2004), although not the dominant monoterpene as previously reported. Furthermore, other studies have also reported limonene to be present in smaller quantities than α-pinene and β-myrcene (Beverland et al., 1996; Hrutfiord et al., 1974). Monoterpene composition was generally consistent between measurements throughout our study even though different branches and trees were measured. This may reflect that the trees grown via vegetative propagation could be from a genetically similar source. However, the variability between the previous literature discussed here may point towards the potential for different chemotypes within Sitka spruce, as suggested by (Forrest, 2011) and similar to that of Norway spruce (Hakola et al., 2017) and Scots pine (Bäck et al., 2012). Norway spruce has also been found to be significant emitters of sesquiterpenes (Hakola et al., 2017). Given the dominance of Sitka spruce plantations in the UK (and Ireland), the potential for

556 variation within this species, and the limited literature data on BVOC emissions, we suggest further
557 measurements are needed at the branch and canopy level to fully assess the terpenoid species
558 composition and their subsequent impact on air quality.

559

560 ## 3.4 BVOC emissions from forest floor

561 The forest floor has been reported as both a source of BVOCs (Asensio et al., 2007a, 2007b;
562 Bourtsoukidis et al., 2018; Greenberg et al., 2012; Hayward et al., 2001; Insam and Seewald, 2010;
563 Janson, 1993; Leff and Fierer, 2008; Mäki et al., 2019a; Peñuelas et al., 2014) and a sink, particularly
564 for isoprene (Cleveland and Yavitt, 1997, 1998; Owen et al., 2007; Trowbridge et al., 2020). Leaf litter
565 is a known source of forest floor BVOCs (Gray et al., 2010; Greenberg et al., 2012; Isidorov and
566 Jdanova, 2012). Data discussed here are the net flux of the opposing processes of source and sink.
567 Monoterpene emissions from the forest floor (Hayward et al., 2001) have previously been
568 standardised using G93 (Eq. (3)) on the assumption that air temperature is the main driver of
569 emissions of monoterpenes. However, these algorithms are based on empirical data and were not
570 designed to normalise negative emissions (uptake). In addition, what drives the sources and sinks of
571 the forest floor is often more complex; and although some models have been developed from
572 laboratory or field studies for litter, soils and the forest floor (Greenberg et al., 2012; Mäki et al.,
573 2017, 2019b) the models may be difficult to apply outside of the studies in which they were
574 developed. A process-based model applicable to a range of forest floor types is still lacking (Tang et
575 al., 2019). We therefore did not standardise the BVOC emissions from the forest floor and present
576 only measured fluxes in this section.

577

578 The total monoterpene emissions from the forest floor were highly variable between the three
579 chambers within the plots as demonstrated by a relative standard deviation range of 35% to 170%
580 for a given day, illustrating the highly heterogeneous soil and litter environment. All chamber

measurements made on the same day were averaged per species, and presented as a single flux value (Figure 3) and then grouped according to season and year (Table 5).

3.4.1 Italian alder

Negative fluxes for total monoterpenes were measured on two occasions, 4th July and 24th July. The highest total monoterpene emissions were observed on the 18th October 2018 (18 µg C m^{-2} h^{-1}) and 7th June 2019 (24 µg C m^{-2} h^{-1}) (Figure 3). Day to day variations were associated to some degree with changes in chamber temperature and soil moisture (Figure 3). Seasonal variations in mean emissions were also apparent (Table 5). The forest floor acted as a sink for monoterpenes during summer 2018 when there was bare soil inside the collars. During summer 2019 vegetation grew inside the soil collars and resulted in the forest floor being a more substantial source of monoterpenes (Figure 4). Monoterpene composition reflected the seasonal changes that occurred on the forest floor. The monoterpenes emitted in autumn (October 2018) were dominated by d-limonene, α-pinene and 3-carene and some β-myrcene, consistent with the composition of Italian alder foliage and attributed to the accumulation of leaf litter. However, the profile in June 2019 during the highest total monoterpene emissions showed significant emissions of γ-terpinene and α-phellandrene and likely reflects the changing understorey vegetation, hogweed sp., growing inside the chamber collars and which was only present in the alder plantations. The particular species at East Grange was not identified but *Heracleum mantegazzianum* (giant hogweed) has been determined to be a substantial γ-terpinene emitter (Matoušková et al., 2019). This highlights the importance of the specific understorey vegetation to the overall monoterpene flux composition.

3.4.2 Hybrid aspen

The highest measured total monoterpene emissions, 9.18 µg C m^{-2} h^{-1} and 5.83 µg C m^{-2} h^{-1}, occurred in July 2018 and were associated with the lowest soil moisture and warm temperatures. In contrast,

606 negative monoterpene emissions were also observed in July (24th) and seem to be associated with
607 an increase in soil moisture (Figure 3). Overall spring (0.30 µg C m^{-2} h^{-1}) and summer (0.06 µg C m^{-2} h^{-1}) total monoterpene emission rates in 2019 (Table 5) were smaller by an order of magnitude than
609 in spring (0.71 µg C m^{-2} h^{-1}) and summer (3.84 µg C m^{-2} h^{-1}) 2018. Higher rainfall during 2019
610 (Supplementary Information S1) resulted in increased soil moisture (Figure 3) which may have
611 suppressed some monoterpene emissions (Asensio et al., 2007b). In addition, during 2018, litterfall
612 started in July and peaked in October by which time the canopy had lost all its leaves.

613

614 The composition of the monoterpene emissions from the forest floor during 2018 was similar to
615 those measured from the branch chambers (Figure 1) and was consistent between days. The main
616 monoterpenes comprised α-pinene, β-pinene, camphene, d-limonene and 3-carene. The
617 contribution from the floor of an aspen plantation has not previously been investigated, although
618 soils taken from underneath aspen (*Populus tremula)* trees showed d-limonene as the predominant
619 monoterpene with a maximum emission of 15.9 µg C m^{-2} h^{-1} under laboratory conditions (Owen et
620 al., 2007). Quantifiable emissions of monoterpene from the leaf litter of American aspen (*Populus*
621 *tremuloides*) exist (Gray et al., 2010) although are not chemically speciated

622

623

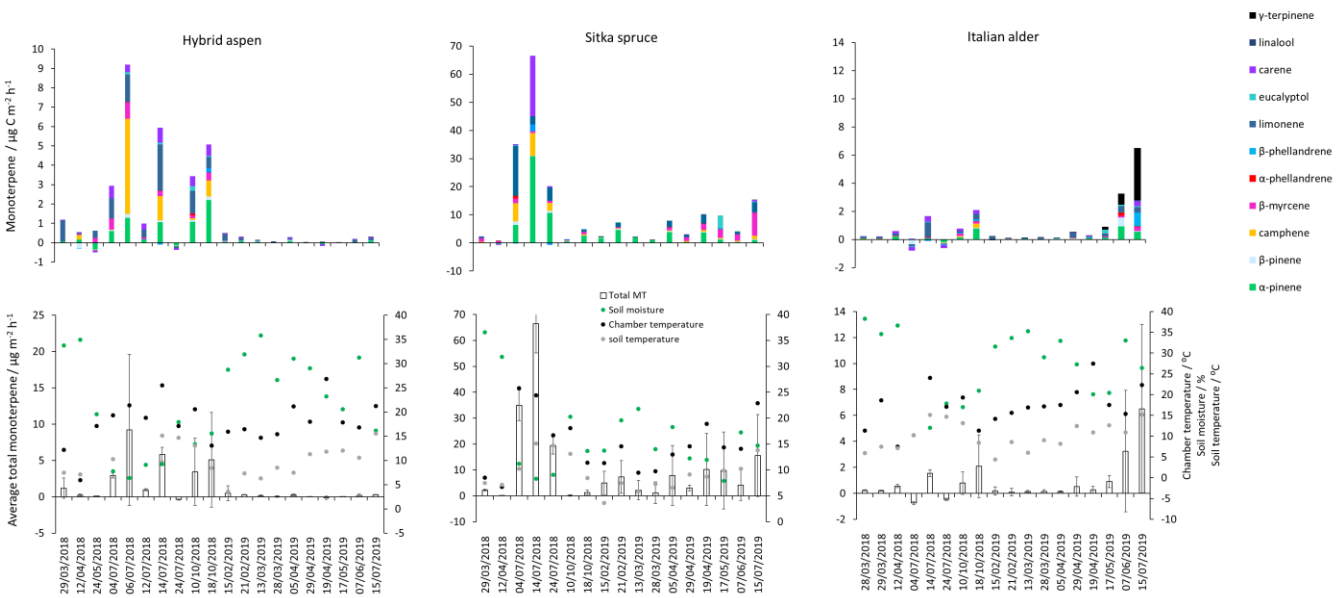


Figure 3 – Daily mean measured forest floor total monoterpene emissions from Sitka spruce, hybrid aspen and Italian alder SRF plots at East Grange, Fife during 2018-2019. Error bars represent the standard deviation of three forest floor chamber measurements. Green circles are volumetric soil moisture (%), black circles are chamber temperature (° C) and grey circles are soil temperature (° C). Note that emission scales differ between tree species plots.

3.4.3 Sitka spruce

Total monoterpene emissions measured from the Sitka spruce forest floor peaked during July 2018 (66.5 µg C m^{-2} h^{-1}) and coincided with the highest chamber temperatures and the lowest soil moisture readings (Figure 3). The lowest measured emissions (0.03 µg C m^{-2} h^{-1}) were observed on the 12th April 2018 when the temperature was lowest (7.5 °C, Figure 3) suggesting soil moisture and temperature are likely interacting controlling variables of monoterpene emissions. In addition, there were clear seasonal differences when measurement days were grouped. Mean measured summertime emissions of total monoterpenes from the forest floor in 2018 were larger than those measured in 2019 (Table 5). Temperatures measured in the chambers were 3 °C degrees higher on mean during 2018 compared to 2019 which could have contributed to the higher observed emissions although soil moisture at 7 cm depth was not significantly different. The young Sitka spruce plantation had litter present all year round unlike in the deciduous species plantations, but

643 the covering was sparse (Figure 4) compared to a mature plantation. Total monoterpene emissions
644 measured in summer 2018 (40.3 μg C m^{-2} h^{-1}) were slightly higher but similar in magnitude to the
645 33.6 μg m^{-2} h^{-1} (29.6 μg C m^{-2} h^{-1}) previously reported for the upper-most layers of the floor in a
646 mature Sitka spruce plantation (Hayward et al., 2001). Norway spruce plantation have also been
647 reported to have a slightly higher emission rate at 50 μg C m^{-2} h^{-1} (Janson et al., 1999).

648

649 The monoterpene composition profile in 2018 was comparable to 2019 and consistent with the
650 branch emissions recorded during our study, the major emitted monoterpenes being β-myrcene, α-
651 pinene, β-pinene, d-limonene and camphene. β-myrcene accounted for a larger percentage, 20–
652 50%, of emissions in summer 2019 compared to only 5–10% in summer 2018 (Table 5), although
653 there is no obvious explanation for this difference.

654

655

Figure 4 – Changes in the presence of leaf litter, herbaceous plants and grasses inside the forest floor chambers of (a) Italian alder (b) hybrid aspen and (c) Sitka spruce SRF plots at East Grange, Fife during 2019.

Table 5 – Seasonal variation in forest floor emissions (µg C m^{-2} h^{-1}) of monoterpenes from Sitka spruce, hybrid aspen and Italian alder SRF plots, at East Grange, Fife, Scotland, in 2018–19.

	Spring 2018			Summer 2018			Autumn 2018			Winter 2019			Spring 2019			Summer 2019		
Plantation type	Sitka spruce	Hybrid aspen	Italian alder	Sitka spruce	Hybrid aspen	Italian alder	Sitka spruce	Hybrid aspen	Italian alder	Sitka spruce	Hybrid aspen	Italian alder	Sitka spruce	Hybrid aspen	Italian alder	Sitka spruce	Hybrid aspen	Italian alder
Days	2	2	3	3	6	3	2	2	2	3	3	3	6	6	6	1	1	1
N	2	4	4	3	8	3	2	4	4	9	9	9	17	18	17	2	1	2
air T / °C	7.6 (1.3)	9.0 (3.6)	11.2 (5.2)	21.1 (4.5)	19.6 (4.1)	18.5 (4.2)	14.8 (4.7)	16.3 (4.3)	15.5 (3.7)	12.6 (1.1)	12.4 (1.5)	13.5 (0.5)	13.9 (2.0)	16.4 (2.4)	16.0 (3.8)	22.5 (0.0)	16.0	20.6 (0.0)
chamber T / °C	7.6 (1.3)	9.0 (3.6)	11.2 (5.2)	21.2 (4.2)	20.0 (4.2)	20.6 (4.9)	14.4 (4.2)	16.8 (4.4)	15.4 (4.6)	11.8 (2.3)	15.7 (1.5)	15.5 (1.3)	13.8 (2.8)	19.3 (4.0)	19.5 (4.2)	22.9 (0.7)	21.2	22.3 (0.0)
soil T / °C	5.3 (1.1)	6 (1)	6.9 (0.7)	14.3 (0.2)	14.3 (0.9)	13.4 (2.7)	9.8 (2.5)	10.6 (1.9)	10.8 (2.7)	6.2 (1.1)	5.7 (1.7)	6.4 (1.8)	8.5 (1.4)	10.3 (1.8)	10.7 (1.8)	13.8 (0.0)	15.6	15.2 (0.0)
chamber RH / %	-	-	-	-	-	-	-	-	-	88 (6)	81.4 (4.5)	77 (3)	74 (9)	73 (8)	88 (11)	70 (7)	78	79 (0)
soil moisture / %	34 (3)	36 (2)	37 (2)	20 (8.0)	12 (5)	13.4 (4.0)	14 (0)	14 (3)	19.0 (2.3)	21 (2)	32.2 (3.6)	34 (3)	14 (2)	27 (4)	27 (6)	15 (1)	31	26 (0)
α-pinene	-0.067 (0.372)	0.113 (0.075)	0.119 (0.111)	15.954 (13.059)	0.557 (0.736)	-0.050 (0.135)	1.627 (1.443)	1.634 (1.991)	0.454 (0.708)	2.661 (3.225)	0.230 (0.522)	0.020 (0.069)	2.167 (3.624)	0.005 (0.064)	0.156 (0.459)	1.067 (1.18)	0.112	0.557 (0.187)
β-pinene	0.052 (0.034)	-0.150 (0.176)	-0.019 (0.023)	0.724 (0.579)	0.076 (0.114)	-0.112 (0.165)	0.086 (0.010)	0.145 (0.166)	0.042 (0.038)	0.209 (0.271)	0.054 (0.111)	0.002 (0.007)	0.224 (0.387)	0.007 (0.023)	0.084 (0.305)	0.217 (0.191)	0.004	0.037 (0.003)
Camphene	0.130 (0.112)	0.126 (0.234)	0.013 (0.004)	5.775 (2.692)	1.386 (3.408)	-0.011 (0.038)	0.255 (0.174)	0.456 (0.784)	0.191 (0.275)	0.142 (0.235)	0.213 (0.634)	0.000 (0.008)	0.687 (1.578)	0.000 (0.004)	0.010 (0.022)	1.248 (1.453)	0.000	0.000 (0.000)
β-myrcene	0.930 (0.447)	0.014 (0.015)	0.009 (0.012)	1.046 (0.533)	0.426 (0.540)	0.024 (0.045)	0.521 (0.483)	0.272 (0.339)	0.172 (0.139)	0.115 (0.256)	1.255 (3.761)	0.011 (0.028)	4.839 (13.585)	0.005 (0.011)	0.034 (0.075)	8.145 (8.828)	0.002	0.270 (0.020)
α-phellandrene	0.006 (0.006)	0.004 (0.005)	0.000 (0.003)	0.355 (0.636)	0.009 (0.012)	0.002 (0.005)	0.000 (0.002)	0.064 (0.106)	0.002 (0.007)	0.011 (0.015)	0.025 (0.073)	0.000 (0.000)	0.055 (0.145)	0.000 (0.001)	0.027 (0.107)	0.118 (0.167)	0.000	0.075 (0.106)
β-phellandrene	0.000 (0.000)	-0.002 (0.003)	0.000 (0.000)	0.481 (1.669)	-0.020 (0.037)	-0.021 (0.058)	0.005 (0.006)	0.125 (0.226)	0.085 (0.120)	0.020 (0.035)	0.010 (0.028)	0.000 (0.000)	0.031 (0.092)	0.000 (0.000)	0.003 (0.013)	0.152 (0.112)	0.003	0.965 (1.290)
d-limonene	0.263 (0.391)	0.566 (1.014)	0.167 (0.078)	8.417 (8.037)	0.997 (0.888)	0.270 (0.679)	0.428 (0.373)	0.860 (0.933)	0.260 (0.199)	0.767 (0.983)	0.640 (1.450)	0.095 (0.210)	2.386 (5.456)	0.038 (0.053)	0.192 (0.298)	3.505 (3.375)	0.087	0.400 (0.021)
Eucalyptol	0.003 (0.002)	0.002 (0.002)	0.004 (0.011)	0.087 (0.160)	0.040 (0.088)	-0.025 (0.052)	0.133 (0.132)	0.150 (0.187)	-0.002 (0.007)	0.006 (0.011)	0.053 (0.144)	0.002 (0.004)	0.851 (2.980)	0.000 (0.003)	0.077 (0.152)	0.342 (0.346)	0.015	0.065 (0.007)
3-carene	-0.189 (0.276)	0.034 (0.032)	0.093 (0.125)	7.446 (12.140)	0.372 (0.496)	0.035 (0.335)	0.086 (0.006)	0.552 (0.621)	0.228 (0.233)	0.020 (0.029)	0.055 (0.063)	0.003 (0.054)	0.077 (0.147)	0.001 (0.066)	0.016 (0.047)	0.564 (0.077)	0.049	0.347 (0.066)
Linalool	0.000 (0.000)	0.000 (0.000)	0.000 (0.000)	0.000 (0.000)	0.000 (0.000)	0.000 (0.000)	0.000 (0.000)	0.000 (0.000)	0.000 (0.000)	0.001 (0.002)	0.005 (0.013)	0.000 (0.001)	-0.000 (0.002)	0.001 (0.002)	0.001 (0.004)	0.012 (0.003)	0.016	0.080 (0.007)
γ-terpinene	0.000 (0.000)	0.000 (0.000)	0.000 (0.000)	0.000 (0.000)	0.000 (0.000)	0.000 (0.000)	0.000 (0.000)	0.000 (0.000)	0.000 (0.000)	0.001 (0.002)	0.003 (0.003)	0.000 (0.001	0.011 (0.037)	0.000 (0.002)	0.128 (0.386)	0.157 (0.215)	0.007	3.709 (5.187)
Total MT	1.128 (1.559)	0.707 (0.977)	0.387 (0.210)	40.286 (23.999)	3.843 (5.490)	0.111 (1.254)	3.141 (2.615)	4.257 (4.706)	1.433 (1.664)	3.954 (4.970)	2.543 (6.737)	0.135 (0.225)	11.330 (24.084)	0.057 (0.174)	0.729 (1.567)	15.527 (15.797)	0.296	6.506 (6.488)

T = Temperature, N = Number of measurements, - = Not measured, RH = Relative humidity, 0.000 = values <0.0005, MT = Monoterpene

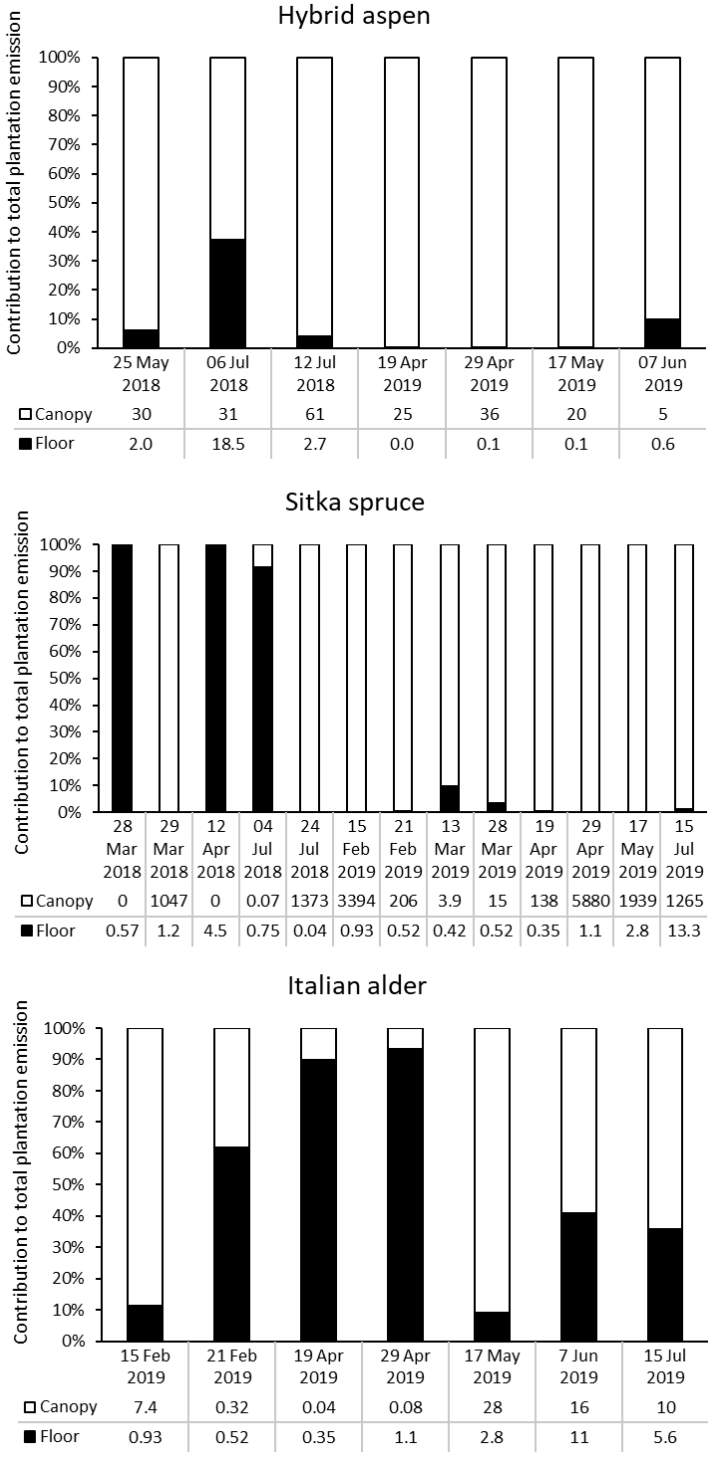
3.5 Plantation-scale isoprene and total monoterpene emissions

3.5.1 Relative contribution of forest floor and canopy emissions

Forest floor and branch emissions were sometimes measured on the same occasion enabling calculation of the contribution of each source to the total monoterpene emissions of the plantation per square metre of ground (based on non-standardised data) (Figure 5). In most cases, particularly in summer, emissions from the canopy dominated. For Sitka spruce, high monoterpene emissions from the plantation occurred when canopy emissions were high which supports previous summertime observations on conifer spp. that the forest floor contributes little to the overall forest monoterpene emissions (Hayward et al., 2001; Janson, 1993). We found that in some instances, more often in spring when canopy foliage was sparse (alder and aspen) or dormant due to cold

temperatures (spruce), the forest floor contributed the majority of the plantation monoterpene emissions. This trend was also reported for conifer sp. in the boreal forest (Mäki et al., 2019b).

For hybrid aspen the opposite was true with the forest floor contributing more in the summer, as a result of understorey vegetation or early litter fall, contributing up to 40% of the total monoterpene emissions of the plantation. In the Italian alder plantation the contribution was more mixed. Canopy emissions in late winter/ early spring were only from the alder flowers (catkins). The low observed emissions at this time of year from the forest floor were likely caused by colder temperatures and high soil moisture. However, later in spring (April) monoterpene emissions came largely from the forest floor (90%) as understorey vegetation began to grow and soil temperatures also increased. The canopy at this point was at the stage of leaf emergence when the foliage was sparse and so contributed little to the overall emissions. However, by summer just over half of the monoterpenes came from the canopy (now in full foliage) and the forest floor contributed around 40% of the monoterpenes, related to the presence of understorey vegetation.



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693

694 **Figure 5 – Percentage contribution of canopy (white bar) and forest floor (black bar) emissions to**
695 **the total monoterpene emissions from SRF plantations at East Grange, Fife, Scotland. Numbers**
696 **below the bars are the total monoterpene emissions in µg C m^{-2} h^{-1}.**

697

3.5.2 Modelled above-canopy fluxes

This section discusses modelled emissions of BVOC from the canopy per m^2 of ground. The "bottom up" approach of estimating BVOC emissions in this study using the chamber technique is useful for determining the contribution of different ecosystem components to BVOC emissions, but in this section emissions do not include modelled forest floor emissions. It is noted that forest floor processes are still being integrated into models in order to reliably capture the full complexity of the forest floor BVOC emissions for prediction purposes (Tang et al., 2019).

Mean standardised summertime emission factors for each tree species in section 3.3 (derived using the simplified G93 algorithms) (Table 3) were adjusted on an hourly basis by the Pocket MEGAN 2.1 excel beta 3 calculator to derive hourly BVOC emissions per unit ground area (Guenther et al., 2012). This allowed for a more advanced method of estimation of monthly and subsequent annual BVOC emissions from the canopy across two years (2018–2019) and two locations, East Grange (Scotland) and Alice Holt (England) for a given air temperature, PAR and the influence of these parameters over the previous 24 and 240 hours. In addition, changing LAI across the year (Table 2) had an influence on the biomass density of the canopy which influenced the emission rate of BVOCs per unit area of ground. Similar to previous modelling studies (Ashworth et al., 2015; Zenone et al., 2016) standardised mean summertime measurements were used as the basis for this calculation.

Given the above, modelled mean diurnal canopy emissions of isoprene for hybrid aspen were calculated to be approximately 2 mg C m^{-2}_{ground} h^{-1}, rising to a maximum of 7 mg C m^{-2}_{ground} h^{-1} in July, the warmest month, across both years (Figure 6A). These modelled emissions for the UK are broadly comparable to those reported from measured eddy covariance flux measurements above a hardwood forest, comprising primarily of aspen (*Populus tremuloides* and *Populus grandidentata,* LAI: 3.24-3.75) in Michigan USA and the boreal forest in Canada (predominantly *Populus tremuloides,*

723 LAI: 2.4) where the mean summertime emissions are reported to peak at 11 mg C m^{-2}_{ground} h^{-1} and
724 6.87 mg C m^{-2}_{ground} h^{-1} respectively (Fuentes et al., 1999; Pressley et al., 2006).

725

726 Mean total monoterpene emissions are two orders of magnitude smaller than isoprene (Figure 6B)
727 for hybrid aspen. Figure 6 (C and D)) highlights the difference in the relative magnitudes of emissions
728 between the three SRF species. Mean emissions from the canopy of Italian alder for isoprene (0.002
729 mg C m^{-2}_{ground} h^{-1}) and monoterpene (0.05 mg C m^{-2}_{ground} h^{-1}) were very small and no above-canopy
730 measurements could be found in the literature for comparison. For Sitka spruce mean canopy scale
731 emissions for July in Scotland were modelled to be 1.5 mg C m^{-2}_{ground} h^{-1} and 0.5 mg C m^{-2}_{ground} h^{-1} for
732 isoprene and total monoterpene respectively. There has only been one attempt in the UK to quantify
733 BVOC directly above a Sitka spruce plantation (Beverland et al., 1996) where a relaxed eddy
734 accumulation system was used and mean isoprene emissions were reported to be 0.146 mg C m^{-2}_{ground} h^{-1}
735 in a 24-h period in early July (temperature range 7-19 ^{o}C). These emissions are much lower
736 than our model estimates although it was reported that there were analytical difficulties with the
737 micrometeorological techniques and limited data which could account for this disparity.

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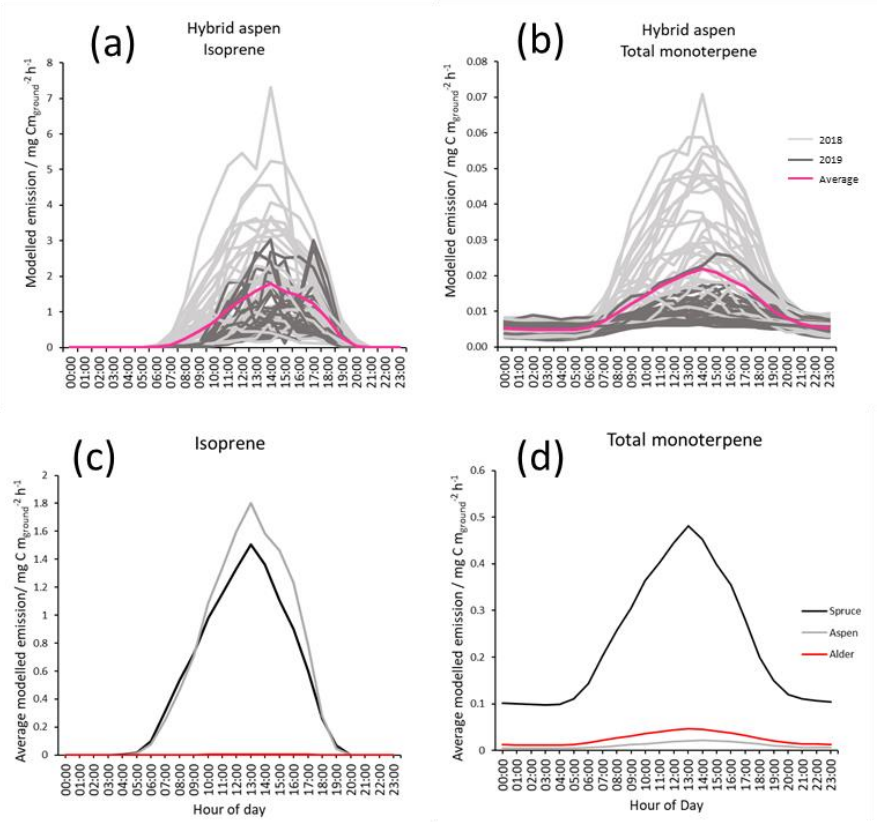

740

741 **Figure 6 – Modelled diurnal canopy emissions for July using MEGAN 2.1 of (a) isoprene from**
742 **hybrid aspen 2018 (light grey), 2019 (dark grey) and combined mean emission rate (pink), (b) total**
743 **monoterpene hybrid aspen (light grey), 2019 (dark grey) and combined mean emission rate (pink),**
744 **(c) mean modelled isoprene for three SRF species, spruce (Black), aspen (grey) and alder (red) for**
745 **July 2018–2109, (d) mean modelled total monoterpene for three SRF species, spruce (Black), aspen**
746 **(grey) and alder (red) for July 2018–2109. Results used measured PAR, temperature and the mean**
747 **summer branch emission potentials collected during this study (Table 3).**

748

749

750 3.5.3 Annual above-canopy fluxes per hectare for a UK planation

751 Table 6 shows the modelled annual BVOC emissions per hectare of plantation for each species for
752 the two meteorological years (2018-2019) at East Grange in Scotland, and for the contemporaneous
753 meteorology experienced in southern England (at Alice Holt). The modelled annual fluxes of
754 isoprene and total monoterpenes per hectare of Sitka spruce plantation averaged over the two
755 contrasting years were roughly similar, at 13.8 and 15.7 kg C ha^{-1} y^{-1}, respectively. Hybrid aspen was
756 modelled to emit only an average of 0.3 kg C ha^{-1} y^{-1} total monoterpene but much more isoprene

(15.5 kg C ha^{-1} y^{-1}), whereas the model estimated that Italian alder emitted minimal isoprene (0.02 kg C ha^{-1} y^{-1} on average) but larger monoterpene emissions of 0.81 kg C ha^{-1} y^{-1}.

It is worth noting that use of an mean summer flux could lead to a potential overestimation of emissions during other seasons and the subsequent total annual flux. Modelled isoprene emissions from Sitka spruce during 2018 for both East Grange and Alice Holt were higher than monoterpene emissions. In 2019, however, monoterpene emissions were more abundant than isoprene emissions using the East Grange meteorology data and of the same magnitude using the Alice Holt meteorology data. The lower PAR during 2019, which was more pronounced for East Grange than Alice Holt, limited the isoprene emissions. Monoterpenes were less affected as these were only temperature driven. The relative proportions of isoprene and monoterpenes in the atmosphere are important since they have differing effects on the formation and concentration of atmospheric pollutants such as ozone and secondary organic aerosol (SOA) (Bonn et al., 2017; Heinritzi et al., 2020). Long-term BVOC emissions measurement above Sitka spruce plantations is needed to confirm this model observation.

Table 6 – Modelled annual isoprene, total monoterpene and total BVOC emissions per hectare of SRF Sitka spruce, hybrid aspen and Italian alder plantations, using meteorology data from two locations, East Grange in east Scotland, and Alice Holt in south-east England.

			Total MT / kg C ha^{-1} y^{-1}	Isoprene / kg C ha^{-1} y^{-1}	Total BVOC / kg C ha^{-1} y^{-1}
Sitka spruce	2018	East Grange	12.3	18.0	30.3
	2019	East Grange	7.95	2.67	10.6
	2018	Alice Holt	21.2	30.3	51.5
	2019	Alice Holt	13.7	11.9	25.6
	Mean		13.8	15.7	29.5
Hybrid aspen	2018	East Grange	0.2	12.1	12.3
	2019	East Grange	0.3	13.0	13.3
	2018	Alice Holt	0.5	22.2	22.7
	2019	Alice Holt	0.2	14.8	15.0
	Mean		0.3	15.5	15.8
Italian alder	2018	East Grange	0.88	0.02	0.90
	2019	East Grange	0.33	0.01	0.34
	2018	Alice Holt	1.53	0.04	1.57
	2019	Alice Holt	0.52	0.02	0.54
	Mean		0.81	0.02	0.84

, MT = Monoterpene

3.6 Uncertainties in measured and modelled fluxes

There are several uncertainties and simplifications in our approach to scaling-up from periodic branch chamber emission measurements to annual canopy–scale predictions. We suggest that uncertainties in the quantification of individual measurements of BVOC emissions are likely to be 16-17% based on previous error propagation calculations (Purser et al., 2020). The nature of the chamber measurement technique is likely to have an impact upon the BVOC emissions due to the altered environmental conditions that may result. In addition, field-based measurements of emission rates, collected under natural conditions for the UK but far from standard conditions (PAR 1000 µmol m^{-2} s^{-1}, temperature 30 ^{o}C) introduce an uncertainty when standardised to form emission potentials.

Further uncertainty may then come from extrapolating these emission potentials in models for the prediction of fluxes using measured meteorology for a given field site. The modelling undertaken here does not include parameters such as soil moisture, humidity and wind speed as no continuous data for these parameters were available but it is noted these would further constrain the model estimate. In addition, there are uncertainties in collating data points to create seasonal means for each year, up to 25-50% based on the relative standard deviation in this case. Converting from emissions per leaf mass to per leaf area also adds uncertainty since leaf mass:area data is highly variable and dependent upon the tree species and sample location. However, we collected LMA data from a range of studies in areas close to the UK with a similar climate (Table 1), and the LMA uncertainty associated ranges from 16% to 24% RSD dependent upon tree species. The emissions predicted from the canopy are also lacking the influence of processes such as BVOC uptake by the forest floor, deposition to leaf surfaces and the influence of reactions with other atmospheric chemical species such as hydroxyl, ozone and nitrogen oxides.

Emissions in early spring measured in the chambers from flowers (catkins) were not included in this scale up exercise since only emission rates from foliage were used in the model. It is noted that these floral emissions may contribute significantly to spring time BVOC emissions across a two or three week time period (Baghi et al., 2012), but become less significant relative to the yearly contribution. It should be noted that BVOC emissions are predicted by the model in winter for Sitka spruce which maintains its canopy all year. However, this may be an over prediction of the emissions as, on some occasions, demonstrated by our chamber measurements, winter BVOC emission may be very low or absent from this species. Similarly, rain events have been shown to alter BVOC emissions and may have different effects in the short term (increasing) and the longer term (decreasing), which are also not accounted for in the model (Holzinger et al., 2006). These factors are likely to lead to an over estimation of emissions from all species but in particular Sitka spruce on a per annum basis.

Finally, algorithms used to scale up branch chamber emissions to canopy-level emissions have also been suggested to give variable results, with MEGAN 2.1 typically producing lower (but perhaps more realistic) flux estimates (Langford et al., 2017). This is an important consideration when comparing annual estimates to total UK BVOC emissions in section 3.7 where older, more simplified algorithms may have been applied.

3.7 Assessing potential impact of SRF plantation expansion on UK BVOC emissions

The annual mean BVOC emissions data from section 3.5.3 (Table 6) was used to explore the possible impact on total UK BVOC emissions arising from increased SRF planting under a suggested bioenergy expansion in the UK (see introduction). The following estimates assume all bioenergy expansion is SRF. However it is more likely that a combination of SRC, SRF and miscanthus could be used in the UK for biomass and as such these estimates should be treated as a single extreme case scenario. Meteorological data from Alice Holt and East Grange was used for model simulations as stated in section 3.5.2. Isoprene and monoterpene emissions are reported separately in Table 7 but also combined to give a "total BVOC" emission.

Table 7 – Modelled mean annual emissions from 0.7 Mha of SRF expansion.

0.7 Mha SRF expansion scenario	Total monoterpene / kt y^{-1}	Isoprene / kt y^{-1}	Total BVOC / kt y^{-1}
Sitka	9.7	11	20.7
Aspen	0.2	10.9	11.1
Alder	0.6	0	0.6

In the scenario of an expansion of 0.7 Mha of SRF, the total BVOC emissions from Sitka spruce SRF could equate to 20.7 kt y^{-1}. For Aspen it could potentially be 11.1 kt y^{-1}, whilst for Italian alder it is much smaller at 0.6 kt y^{-1}. These potential increases in BVOC emissions are compared in Table 8 to current predicted annual emissions of BVOCs from vegetation in the UK. Several air quality models

845 have been used to estimate the total isoprene and total monoterpene emissions from UK vegetation
846 (AQEG, 2020), with an earlier model (Simpson et al., 1999) determining isoprene to be the dominant
847 BVOC emission whilst later models suggest monoterpenes dominate (Hayman et al., 2017, 2010;
848 Stewart et al., 2003). The meteorological data used in some of these models are limited to a single
849 year, e.g. 1998, where the uncertainty in the model estimates could range by a factor of 4 (Stewart
850 et al., 2003), whilst others are the mean emissions across many years and so report a range (Hayman
851 et al., 2017). In addition, models of UK BVOC emissions are particularly reliant upon the emission
852 potential attributed to Sitka spruce as this accounts for nearly 21% of UK forest cover and, as
853 discussed in section 3.3.3, only a limited number of studies have been conducted on Sitka spruce
854 BVOC emissions. This simple impact assessment used a limited set of meteorological data to
855 represent two contrasting years (one warmer drier year and one cooler wetter year, relative to the
856 30 year mean) and for two 'ends' of the British climate range of temperature and PAR: north (East
857 Grange, Scotland) and south (Alice Holt, England).

858

859 However, given these uncertainties, simulations of the impact of potential future land–use changes
860 on atmospheric BVOC emissions are important first steps to gain a better understanding of any
861 potential future impacts on air quality.

862

863 It is worth noting that currently the UK has an estimated 3.2 Mha of woodland, of which 0.67 Mha is
864 covered by Sitka spruce (Forest Research, 2020) (similar in size to the future planting scenario used
865 here), a small area of alder (0.053 Mha, Forest Research, 2012) and even smaller area of aspen.
866 Comparing the total BVOC emissions for a 0.7 Mha SRF expansion scenario to the annual total BVOC
867 emissions for the UK suggests that the Sitka spruce and hybrid aspen scenarios could potentially
868 increase the total BVOC emissions in the ranges of 12–35% and 7–19% respectively, dependent upon
869 the original BVOC emission model used for this comparison (Table 8). For Italian alder this increase in

total BVOC is an order of magnitude smaller, ranging from 0.3–1%. It can therefore be suggested that future hybrid aspen SRF plantations for bioenergy will likely emit no more BVOC than equivalent expansion of young Sitka spruce plantations. Expansion of SRF with Italian alder may bring about no significant changes to the UK BVOC emissions at the national level.

Any future distribution of bioenergy crops including SRF in the UK will depend on several factors including available land, locations that are most suitable to obtain high biomass yields, locations that are close to energy-generation plants and locations close to opportunities for CO_2 storage, in the case of using BECCS to reach net-zero targets (Donnison et al., 2020). Further work is needed to better understand how these changes in BVOC emissions may impact air chemistry and potentially air quality (in particular ozone and SOA) at local to UK national scale.

Table 8 – Potential increase in isoprene, total monoterpene and total BVOC emissions from an additional 0.7 Mha of SRF plantations compared to previous modelled estimates of total UK BVOC emissions.

				Sitka spruce SRF			Hybrid aspen SRF			Italian alder SRF		
	Modelled UK total emissions / kt y^{-1}			% of modelled UK emissions			% of modelled UK emissions			% of modelled UK emissions		
Model Reference	MT	Isoprene	Total	MT	Isoprene	Total	MT	Isoprene	Total	MT	Isoprene	Total
Simpson et al. 1999	30	58	88	32	19	24	0.7	19	13	1.9	0.0	0.7
Stewart et al. 2000	83	8	91	12	138	23	0.3	136	12	0.7	0.2	0.6
Hayman et al. 2010 (forest only)	52	7	59	19	157	35	0.4	155	19	1.1	0.2	1.0
Hayman et al. 2017 (minimum)	110	33	143	9	33	14	0.2	33	8	0.5	0.0	0.4
Hayman et al. 2017 (maximum)	125	44	169	8	25	12	0.2	25	7	0.5	0.0	0.3

Values that are shown as 0.0 are < 0.05%; Hayman et al 2017 (minimum) and (maximum) values are the upper and lower estimates of BVOC emissions published that account for yearly changes in meteorology in the model scenarios.

4. Conclusions

Winter and spring emissions of isoprene and monoterpenes in the three potential short-rotation forestry (SRF) species of Sitka spruce, hybrid aspen and Italian alder were one or two orders of magnitude smaller than their respective emissions in summer. There were large differences in the BVOC emission rates and compounds between the three species, with d-limonene, α-pinene and β-myrcene being the major monoterpenes across all three species.

Sitka spruce emitted more isoprene and monoterpenes during the warmer, drier 2018 than in the cooler, wetter 2019. Isoprene emissions for hybrid aspen were similar in both years but monoterpene emissions were higher in 2018 compared to 2019. Italian alder did not often emit detectable amounts of isoprene in either year, and only a little monoterpene in 2018. The observed differences in emissions of the relative amounts of isoprene compared to monoterpenes in the case of Sitka spruce could lead to differences in SOA generation in warmer and cooler years.

Overall, forest floor emissions of monoterpenes were a factor 10 to 1000 times smaller than the canopy emissions. The forest floor emissions were more variable and acted as a source for most of the time with occasional instances (<4 measurement occasions out of 20) when the forest floor acted as a sink for monoterpenes. Further work is necessary under controlled conditions to fully understand the drivers and components of forest floor emissions.

Total annual emissions per unit ground area for each SRF species were derived using MEGAN 2.1 and scaled up to a 0.7 Mha future SRF expansion scenario for the UK. Under this scenario, total modelled UK BVOC emissions (the sum of isoprene and total monoterpene emissions) could increase by <1–35% depending on the species planted and the UK BVOC emissions model used. Future work to understand how any increase in forest cover and BVOC emissions may impact the atmospheric chemistry in NOx dominated regions is needed so that air quality impacts from pollutants such as ozone can be determined across the UK.

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Competing interests. The authors declare that they have no conflict of interest.

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