



29 h^{-1} for 2018 and 2019, aspen emitting $0.3 \mu\text{g C g}^{-1} \text{h}^{-1}$ and $0.09 \mu\text{g C g}^{-1} \text{h}^{-1}$ and Italian alder emitting,
30 $1.5 \mu\text{g C g}^{-1} \text{h}^{-1}$ and $0.2 \mu\text{g C g}^{-1} \text{h}^{-1}$, respectively. In contrast to these foliage emissions, the forest
31 floor was only a small source of monoterpenes, typically one or two orders of magnitude lower than
32 foliage emissions on a unit ground area basis. Estimates of total annual emissions from each
33 plantation type per hectare were derived using the MEGAN 2.1 model. The modelled total BVOC
34 (isoprene and monoterpenes) emissions of SRF hybrid aspen plantations were approximately half
35 those of Sitka spruce for plantations of the same age. Italian alder SRF emissions were 20 times
36 smaller than from Sitka spruce. The expansion of bioenergy plantations to 0.7 Mha has been
37 suggested for the UK to help achieve “net-zero” greenhouse gas emissions by 2050. The model
38 estimates show that with such an expansion total UK BVOC emissions would increase between <1%
39 and 35%, depending on the tree species planted. Where increases might be small on a national
40 scale, regional increases might have a larger impact on local air quality.

41

42 **1. Introduction**

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



53

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

66

67 In 2010, Forest Research established SRF trials across the UK to determine biomass yields and assess
68 the environmental impact of SRF (Harrison, 2010). The trials included a number of broadleaf tree
69 species (hybrid aspen, red alder, common alder, Italian alder, sycamore, horse chestnut, eucalyptus
70 spp.) and the two conifer species Sitka spruce and hybrid larch (Harrison, 2010). Sitka spruce is the
71 most widely grown conifer tree species in the UK and a key plantation species. SRF plantations have
72 previously been assessed for their environmental impact in the UK and Ireland (Keith et al., 2015;
73 McKay, 2011; Tobin et al., 2016), but not for their potential future impacts on air quality in the UK,
74 which is the focus of this work.

75



76 Trees are known sinks for CO₂ but can also be sources of other trace gases such as volatile organic
77 compounds (VOCs) (Monson and Fall, 1989; Went, 1960). VOCs are emitted by tree foliage as a
78 means of communication, plant defence against herbivory and during environmental stress such as
79 heat or drought. Other sources of VOCs within a forest may include wood, litter, soils, fruits, flowers
80 and roots (Dudareva et al., 2006). Emitted VOCs include, in particular, isoprene and monoterpenes,
81 and their aliphatic, aromatic and oxygenated derivatives. These compounds are highly reactive in the
82 atmosphere and contribute to the formation of tropospheric ozone in the presence of nitric oxide
83 (NO) (Atkinson and Arey, 2003). Terpene composition has been found to be an important factor in
84 the magnitude of ozone production (Bonn et al., 2017). Ground-level ozone is a concern for
85 agriculture and natural ecosystems as it causes leaf damage, reduced plant growth (Emberson, 2020;
86 Fares et al., 2013; Felzer et al., 2007) and is also a pollutant with impacts on human-health and as a
87 greenhouse gas (UNEP/WMO, 2011). In addition, intermediates of VOC oxidation may act as
88 condensation nuclei for the formation of secondary organic particles (Carlton et al., 2009), another
89 atmospheric pollutant with detrimental effects on human health (Fuzzi et al., 2015).

90

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



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

109

110 2. Methods

111 2.1 Field site description

112 2.1.1 Tree species and planting

113 Measurements were made at East Grange, Fife, Scotland (Lat/Lon (WGS84) 56° 05' 21" N,
114 003° 37' 52" W), elevation 45–60 m, one of the 16 SRF trial locations established by Forest Research
115 (Harrison, 2010; Stokes, 2015). Soil type and texture at the site is surface-water gley and sandy silty
116 loam respectively, containing 4.9% clay, 53.0% silt and 42% sand (Drewer et al., 2017; Keith et al.,
117 2015). In 2010, the ex-agricultural site was planted with a single block of 40 randomised tree species
118 plots and 8 control plots. Each plot (20 m x 20 m) consisted of a single species containing 200 trees
119 with a 2 m x 1 m spacing arrangement (Harrison, 2010). Ten species were planted, and the two
120 broadleaved species with the best survival and growth rates across the trials in the first six years,
121 hybrid aspen (*Populus tremula L. x tremuloides* Michx.) and Italian alder (*Alnus cordata* Desf.), were
122 selected for the measurements here, along with Sitka spruce (*Picea sitchensis* Bong. Carr, produced
123 by vegetative propagation) (McEvoy, 2016; Parratt, 2018). After initial establishment of the young



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

126

127 2.1.2 Meteorological data

128 Meteorological data were collected from an unplanted plot in the middle of the site between May
129 2018 and July 2019. Minimum and maximum soil temperature (T107, Campbell Scientific, Shepshed,
130 Leics, UK), air temperature and relative humidity (HMP45C, Campbell Scientific) were monitored
131 hourly. In addition, photosynthetic active radiation (PAR, SKP 215 Quantum Sensor, Skye
132 instruments, Llandrindod Wells, UK) was measured at the same site every 5 minutes. Monthly
133 averages and ranges are provided in Supplementary Information S1. Occasional power failure at the
134 site led to some missing data. For the modelling of BVOC emissions using Pocket MEGAN 2.1 excel
135 beta 3 calculator (Guenther 2012) the missing PAR and average temperature data were replaced by
136 measurements from the Easter Bush site of the UK Centre for Ecology & Hydrology lying 45 km to
137 the south east (Lat/Lon (WGS84) 55° 51' 44" N, 003° 12' 20" W). A summary of the combined East
138 Grange and Easter Bush data used in the model can be found in Supplementary Information S2.

139

140 The climate in east Scotland is colder, with fewer sunshine hours than in the south of England. To
141 encompass these climate differences, meteorological data from Alice Holt forest (51°09'13"N ,
142 000°51'30"W), Hampshire, in southern England recorded during 2018 and 2019 was also used for
143 the modelling and scaling up of the measured BVOC emission potentials from this study. A summary
144 of the PAR and air temperature data for this field site is given in Supplementary Information S3.

145



146 2.2 Sampling enclosures

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

151

152 2.2.1 Forest floor enclosures

153 Forest floor in this context includes soils, leaf litter, fallen small twigs/branches and flowers,
154 understorey vegetation, microorganisms and underground biomass that may all be sources of BVOC
155 from the ground of the plantation. A static chamber method was used for the plantation floor
156 enclosures. Polyvinylchloride plastic soil collars (with a flange), 40 cm diameter x 18 cm high, were
157 installed per tree species plot prior to sampling (Asensio et al., 2007c, 2007b; Greenberg et al., 2012;
158 Janson, 1993) and remained in the ground for the duration of the experiment. One or two collars
159 were installed in 2017 and used during 2018. Additional collars were installed during 2018 resulting
160 in a total of three soil collars per plot for the 2019 sampling. The collars were placed towards the
161 centre of each plot to reduce the likelihood of plant debris from other plots contaminating them.
162 Leaf litter and understorey vegetation were not removed from the collars prior to sampling to reflect
163 actual changes in BVOC emissions with changes in the forest floor composition through the seasons.
164 A clear acrylic lid (with a foam lined flange), 40 cm diameter x 22.5 cm high, was placed over the soil
165 collar during sampling periods only, enclosing a total chamber volume of 51 L. The lid was sealed
166 using clamps around the rim. A small 12 V axial fan (RS components Ltd, Colby, UK), 4 cm x 4 cm x 1
167 cm, was attached to the chamber lid to mix the air inside the chamber (Janson, 1993). Samples of
168 BVOC in the enclosed air were collected through PTFE tubing onto a 6 mm OD stainless steel
169 automated thermal desorption (ATD) cartridge (PerkinElmer, Waltham, MA, USA) packed with 200
170 mg Tenax TA 60/80 (11982 SUPELCO, Sigma-Aldrich, St Louis, MO, USA) and 100 mg Carbotrap 20/40



171 (20273 SUPELCO, Sigma-Aldrich) at a flow rate of 0.2 L min^{-1} using a handheld pump (210-1003MTX,
172 SKC Ltd, Blandford Forum, UK). Samples were collected for 30 min after closure, equating to a total
173 sample volume of 6 L. Pressure compensation was maintained through a small hole in the side of the
174 chamber to prevent negative pressure inside the chamber and potential degassing of air from the
175 soil pores. Ambient air samples were collected concurrently with the chamber sample in order to
176 quantify BVOC emissions from the forest floor by difference. This is discussed further in Section
177 2.5.2. No ozone filter was used during sampling so amounts of some monoterpenes may have been
178 reduced by reaction with ozone (Ortega et al., 2008). However, it has also been suggested that
179 ozone may be lost by dry deposition onto the chamber walls in the first minute (Janson et al., 1999).
180 Chamber air temperature (Electronic Temperature Instruments Ltd, Worthing, UK) and humidity
181 (Fisherbrand™ Traceable™ Humidity Meter, Fisher Scientific, Loughborough, UK) were measured at
182 the end of the 30 min sample collection period.

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

188

189 2.2.2 Branch enclosure

190 A dynamic chamber method was used for branch enclosures. Three sample points were established
191 per tree species plot and used to mount a removable flow-through acrylic chamber (Potosnak et al.,
192 2013), 53 L in volume. The chambers were set up during each sampling visit and used to enclose a
193 single branch, alternating between three similar branches per tree species. Ambient air flow was
194 delivered from an oil-free double-ended diaphragm pump (Capex V2, Charles Austen pumps Ltd,
195 Surrey, UK) through PTFE tubing (Morrison et al., 2016; Purser et al., 2020) at a flow rate of 10 L min^{-1}



196 ¹ to obtain the desirable air exchange rate of 4-5 minutes (Ortega and Helmig, 2008). In addition, the
197 chamber contained a small 12 V axial fan (RS components Ltd, Colby, UK), 8 cm x 8 cm x 2.5 cm, to
198 ensure mixing of air inside the chamber.

199

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

207

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

217



218 2.3 BVOC analysis

219 The BVOC samples collected on the sorbent were analysed using gas chromatography-mass
220 spectrometry (GC-MS) with a two-stage automatic thermal desorption unit (ATD 400, Perkin-Elmer,
221 Wellesley, MA, USA) using the method described in Purser et al. (2020). Calibration was carried out
222 using standards (from Sigma-Aldrich, Gillingham, UK) of the monoterpenes α -pinene, β -pinene, d-
223 limonene, α -phellandrene, β -phellandrene, 3-carene, camphene, γ -terpinene and β -myrcene, and
224 the monoterpenoids (monoterpene-based compounds with, for example, additional oxygen or
225 missing a methyl group) eucalyptol and linalool prepared as a mixed stock solution of $3 \text{ ng } \mu\text{L}^{-1}$ in
226 methanol. Aliquots of 1, 2, 3 and $4 \text{ } \mu\text{L}$ of the mixed monoterpene stock solution were pipetted
227 directly onto sample tubes under a flow of helium to produce a range of mixed monoterpene
228 standards of 3, 6, 9 and 12 ng . Isoprene standards were prepared by direct sampling onto a sorbent
229 tube from a certified 700 ppbv gas standard (BOC, UK) for 10, 30, 45 and 60 s using a sample pump
230 (210-1003MTX, SKC Ltd, Blandford Forum, UK) producing standards of 65, 198, 296 and 395 ng . Note
231 that mass loadings of isoprene and monoterpene calibration standards were calculated to greater
232 precision than quoted above but are shown here as nominal values for ease of discussion.

233

234 Unknown peaks in sample chromatograms were identified by comparison to the internal library of
235 the GC-MS (National Institute of Standards and Technology) and by comparison with the retention
236 time of the standard. The limit of detection (LOD) of the calculated emissions ranged from $0.12\text{-}0.35$
237 $\mu\text{g C g}_{\text{dw}}^{-1} \text{ h}^{-1}$ for the branch chambers and $0.47\text{-}1.4 \mu\text{g C m}^{-2} \text{ h}^{-1}$ for the forest floor chambers.
238 Uncertainties on an individual calculated emission rates were 16% for isoprene and 17% for
239 monoterpenes, which were derived via error propagation methods described in Purser et al. (Purser
240 et al., 2020).

241



242 2.4 Calculation of standardised emissions

243 2.4.1 Forest floor BVOC emissions

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

$$251 \quad F_{\text{floor}} = \frac{[C_{\text{sample}} - C_{\text{ambient}}] \times V \times 60}{A \times t} \quad (1)$$

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

254

255 2.4.2 Branch scale BVOC emissions

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

$$259 \quad F_{\text{branch}} = \frac{[C_{\text{sample}} - C_{\text{ambient}}] \times f}{m} \quad (2)$$

260 Isoprene emissions have previously been shown to be controlled by both light and temperature and
261 can be standardised to 30°C and $1000 \mu\text{mol m}^{-2} \text{s}^{-1}$, respectively (Guenther et al., 1993). Average
262 chamber air temperature and PAR for each period of sample collection were therefore used to
263 standardise the measured F_{branch} emissions for isoprene (Eq. (3), (4) and (5)) and monoterpenes (Eq.



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

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

$$269 \quad F_{\text{isoprene}} = \frac{F_{\text{branch}}}{C_L \times C_T} \quad (3)$$

270 The light-correction term C_L is calculated from Eq. (4) where $\alpha = 0.0027$ and $C_{L1} = 1.066$ are empirical
271 coefficients in G93 and L is the experimentally-measured average PAR ($\mu\text{mol m}^{-2} \text{s}^{-1}$) during sampling.

$$272 \quad C_L = \frac{\alpha C_{L1} L}{\sqrt{1 + \alpha^2 L^2}} \quad (4)$$

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

277

$$278 \quad C_T = \frac{\exp\left(\frac{C_{T1}(T-T_s)}{RT_s T}\right)}{1 + \exp\left(\frac{C_{T2}(T-T_M)}{RT_s T}\right)} \quad (5)$$

279

280 Monoterpene emissions from branch chambers, F_{branch} were standardised to temperature based on
281 the calculations from Guenther et al. (1993) using Eq. (6). T_s is the standard temperature (303 K) and
282 T is the average air temperature during sampling. $F_{\text{monoterpene}}$ is the standardised monoterpene
283 emission rate ($\mu\text{g C g}_{(\text{dw})}^{-1} \text{h}^{-1}$) and F_{branch} is the measured monoterpene emission rate ($\mu\text{g C g}_{(\text{dw})}^{-1} \text{h}^{-1}$).

284



285
$$F_{\text{branch}} = F_{\text{monoterpene}} \exp(\beta(T - T_s)) \quad (6)$$

286

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

291

292 2.5 LAI determination

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

300

301 2.6 Scaling up from emission per mass of foliage to an emission per area of ground of 302 plantation

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



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

315

$$316 \quad F_{\text{foliage}} = F_{\text{b_summer}} \times LMA \times LAI \quad (7)$$

317

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

326

327 Branch measurements made during April when leaves were young were assigned lower LAI values,
328 such as 1.06 for Hybrid aspen and 0.81 for Italian alder. This modification of LAI through the year
329 (Table 2) was based on multiple LAI measurements taken across the year in a deciduous forest stand
330 in the UK (Ogunbadewa, 2012) in which by late-April (day of year 120) a quarter of the maximum LAI
331 was reached and half the maximum LAI by mid-May (day of year 141). In that study the maximum
332 LAI was recorded in mid-July (day of year 210).



333 **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		
Average RSD / %	77.7 24	-	-	-	-	4.24	329
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		
Average RSD / %	194 16	-	-	-	-	3.19	619
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		
Average RSD / %	97.0 21	-	-	-	-	3.25	315

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

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

336

337

338 **2.7 From canopy emission to total annual emissions per hectare and the influence of**
 339 **increasing biomass planting on total UK BVOC emissions**

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



350 therefore 2018 measurements were used. The function that accounts for the effect of both the
351 previous 24 hours and 240 hours of light on the calculated emissions was applied in the model. The
352 latitude was set to 56° for Scotland and 51° for England and the vegetation cover was set to 1. The
353 functions in MEGAN2.1 that allow for consideration of soil moisture and CO₂ concentrations were
354 not used due to a lack of continuous data available for the field sites. The monoterpenes in the
355 model were calculated using the single value for average total monoterpene from East Grange and
356 using the category named “other monoterpenes”. An assumption was made that the emissions were
357 driven by temperature only and no light specific emission fraction was specified due to the different
358 behaviours of the collective “total monoterpenes”. Any other model input parameters remained as
359 default.

360

361 The model output of hourly isoprene and total monoterpene emissions were summed to give annual
362 emissions per m² of SRF plantation. The combined average total annual emission rate encompassing
363 both years of emission potentials (2018 and 2019) and meteorology from two contrasting UK sites
364 (E. Scotland and S.E. England), for each SRF species, was then compared to literature values for the
365 estimated annual UK isoprene and monoterpene emissions and combined total BVOC emissions.

366

367

368

369

370

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372



373

374 **Table 2 – Seasonal time course of leaf area index (LAI) for estimating annual VOC emissions for**
375 **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

376

377

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

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

380

381

382 **3. Results and discussion**

383

384 **3.1 Field observations of seasonality**

385 The measured BVOC emissions were assigned to seasons as follows: winter (21st December – 19th
386 March), spring (20th March – 07th June), summer (08th June – 22nd September) and autumn (23rd
387 September – 20th December). 2018 is classified here as a dry year, being 25% drier at the East
388 Grange field site than the 30 year average for the area (Met Office, 2020). In contrast, 2019 was 50%
389 wetter than the 30 year UK average. In 2019, catkins were fully developed on the hybrid aspen and
390 Italian alder branches by February, but bud burst and leaf emergence was not observed until mid-



391 April (19th). This was two weeks later than in 2018. The first new growth on the Sitka spruce was
392 observed at the end of April (29th). Based on these differences in phenology at the site,
393 measurements taken on 7th June 2019 was still categorised as spring.

394

395 For the forest floor it was noted that the soil temperatures during summer 2018 were higher than in
396 2019. After several dry weeks in spring and summer in 2018, the first significant rainfall event since
397 May was noted as 14th July, and some leaf fall in the Italian alder and hybrid aspen plots was
398 observed by the end of July. By February 2019, no leaf litter from the previous autumn season was
399 observed on the forest floor of the plots except for those of Sitka spruce. Rapid understorey growth
400 identified as hogweed (*Heracleum sp*) quickly developed from late April (29th) and by early June (7th)
401 completely covered the forest floor in the alder plots. The hybrid aspen and Sitka spruce plots during
402 both 2018 and 2019 had minimal understorey vegetation by comparison.

403 3.2 Leaf area index

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

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

410 A 4-y old SRF plantation of Italian alder established in Ireland that was also measured in July gave an
411 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
412 species such as common (or black) alder (*Alnus glutinosa*) and grey alder (*Alnus incana*) in Sweden
413 had LAI values of 2.85 and 3.04, respectively; all comparable to the Italian alder LAI of 3.25 measured
414 here (Table 1). A study of SRF planting density trials in Ireland found that above-ground biomass



415 growth was similar for Italian alder compared to Sitka spruce (Foreman, 2019) which also aligns well
416 with our observations.

417

418 3.3 BVOC emissions from tree branches

419 3.3.1 Italian alder

420 Italian alder (*Alnus cordata*) emitted very low amounts of isoprene, ranging between $<0.0005 -$
421 $0.035 \mu\text{g C g}_{\text{dw}}^{-1} \text{h}^{-1}$ (standardised $0.017-0.037 \mu\text{g C g}_{\text{dw}}^{-1} \text{h}^{-1}$) depending on season (Table 4),
422 comparable with previous standardised emission rates reported as $<0.1-3 \mu\text{g g}_{\text{dw}}^{-1} \text{h}^{-1}$ ($0.09 - 2.64$
423 $\mu\text{g C g}_{\text{dw}}^{-1} \text{h}^{-1}$) (Calfapietra et al., 2009).

424 Average measured emissions for total monoterpene ranged between $0.041-0.393 \mu\text{g C g}_{\text{dw}}^{-1} \text{h}^{-1}$
425 (standardised $0.073-1.5 \mu\text{g C g}_{\text{dw}}^{-1} \text{h}^{-1}$) with higher emission rates during spring and summer 2018
426 than in 2019. The major monoterpenes emitted were d-limonene, α -pinene, β -myrcene and β -
427 pinene, which were consistently emitted through the spring and summer (Figure 1). No previous
428 data for total or speciated monoterpene emission rates from Italian alder could be found in the
429 literature. However, other alder species have also been reported to be low emitters of
430 monoterpenes, and to emit slightly more monoterpenes than isoprene. Studies that report similar
431 low levels of total monoterpene emissions from alder include $0.8 \mu\text{g C g}_{\text{dw}}^{-1} \text{h}^{-1}$ from grey alder
432 (Hakola et al., 1999), $0.13 \mu\text{g C g}_{\text{dw}}^{-1} \text{h}^{-1}$ from black (or common) alder (Aydin et al., 2014) and $1-2 \mu\text{g}$
433 $\text{C g}_{\text{dw}}^{-1} \text{h}^{-1}$ from green alder (*Alnus rugosa*) (Isebrands et al., 1999). For speciated emissions, 3-carene,
434 β -phellandrene, β -ocimene, p-cymene, sabinene have also been reported to be emitted from *Alder*
435 *sp.* (Aydin et al., 2014; Copolovici et al., 2014; Hakola et al., 1999; Huber et al., 2000). Emissions of
436 some monoterpenes such as β -myrcene are suggested to be induced by herbivory by aphids (Blande
437 et al., 2010). However, since no data on the composition of monoterpenes under laboratory studies
438 in the absence of herbivory is available for Italian alder it is difficult to know which, if any, of the
439 monoterpenes measured in our field study may have been induced by previous herbivory.



440 **Table 4 – Average seasonal BVOC emissions ($\mu\text{g C g}^{-1} \text{h}^{-1}$) from branches of Sitka spruce, hybrid**
 441 **aspen and Italian alder in SRF plantations, East Grange, Fife, Scotland. Figures in parentheses are**
 442 **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 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 / $\mu\text{mol m}^{-2} \text{s}^{-1}$	607 (464)	957 (214)	362 (166)	662 (530)	539 (380)	1018 (447)	-	-	-	394 (217)	-	298 (106)	934 (481)	882 (357)	1081 (381)	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)	2.001 (0.251)	3.93 (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)	0.662 (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)	0.094 (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)	0.111 (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 (0.0134)	0.003 (0.002)	0.000 (0.000)	0.044 (0.025)	0.007 (0.006)	0.005 (0.004)	-	-	-	0.008 (0.012)	-	0.008 (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)	0.051 (0.008)	1.772 (3.229)	0.010 (0.011)	0.149 (0.162)	-	-	-	0.264 (0.599)	-	0.001 (0.001)	0.850 (0.806)	0.001 (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)	0.001 (0.003)	0.807 (0.279)	0.002 (0.002)	0.002 (0.006)

443 Values shown as 0.000 = <0.0005, - = Not measured

444

445 **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 (0.022)	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)	0.016 (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)	0.000 (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)	0.000 (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 (0.003)	0.000 (0.000)	0.000 (0.000)	0.000 (0.001)	0.003 (0.002)	0.000 (0.001)	0.000 (0.001)

446 Values 0.000 = <0.0005, - = Not measured

447



448 **3.3.2 Hybrid aspen**

449 Isoprene emissions from hybrid aspen ranged from 0.053 to 21 $\mu\text{g C g}_{\text{dw}}^{-1} \text{h}^{-1}$ (standardised 0.19–23
450 $\mu\text{g C g}_{\text{dw}}^{-1} \text{h}^{-1}$) (Table 4). No measurements were made during autumn senescence or in winter on the
451 bare branches. Emissions were lower in spring for the newly emerged leaves compared to summer
452 (Figure 1). As noted in Section 3.1, the onset of spring at the field site was earlier in 2018 compared
453 to 2019. European aspen (*Populus tremula*) measured in late spring (May) two weeks after bud burst
454 has also previously been reported to have a lower emission rate than in summer (Hakola et al.,
455 1998). Isoprene emission rates made on leaves (not branches) on aspen in spring in the boreal forest
456 were also reported to be a third of the emission rate measured in the middle of summer (Fuentes et
457 al., 1999). In our study, the hybrid aspen plantation showed signs of stress thought to be associated
458 with lower rainfall and soil moisture locally during summer 2018 causing a yellowing of leaves and
459 early leaf shedding in July. It is widely accepted that isoprene emissions increase with increases in
460 temperature and PAR (Guenther et al., 1991; Monson and Fall, 1989) but that under stress during
461 drought, isoprene can be emitted at much higher rates than usual, only to eventually decline as
462 resources are depleted in the leaves (Brilli et al., 2007; Seco et al., 2015). However, standardised
463 isoprene emissions measured during this study on green aspen leaves did not differ between the
464 two years, 2018 (23 $\mu\text{g C g}_{\text{dw}}^{-1} \text{h}^{-1}$) and 2019 (22 $\mu\text{g C g}_{\text{dw}}^{-1} \text{h}^{-1}$) despite the signs of stress in 2018
465 noted above. The standardised isoprene emissions for hybrid aspen reported here were much lower
466 than those previously reported for European aspen, 51 $\mu\text{g g}_{\text{dw}}^{-1} \text{h}^{-1}$ (i.e. 45 $\mu\text{g C g}_{\text{dw}}^{-1} \text{h}^{-1}$) (Hakola et
467 al., 1998).

468

469 Total monoterpene emissions for hybrid aspen ranged from 0.040 - 0.20 $\mu\text{g C g}_{\text{dw}}^{-1} \text{h}^{-1}$ (standardised
470 0.082 - 0.259 $\mu\text{g C g}_{\text{dw}}^{-1} \text{h}^{-1}$) with substantially higher emissions occurring in summer 2018 (Table 4,
471 Figure 1). Increased emissions for some monoterpenes have been shown to be predominately driven
472 by increases in temperature (Guenther et al., 1991). In particular d-limonene, the major

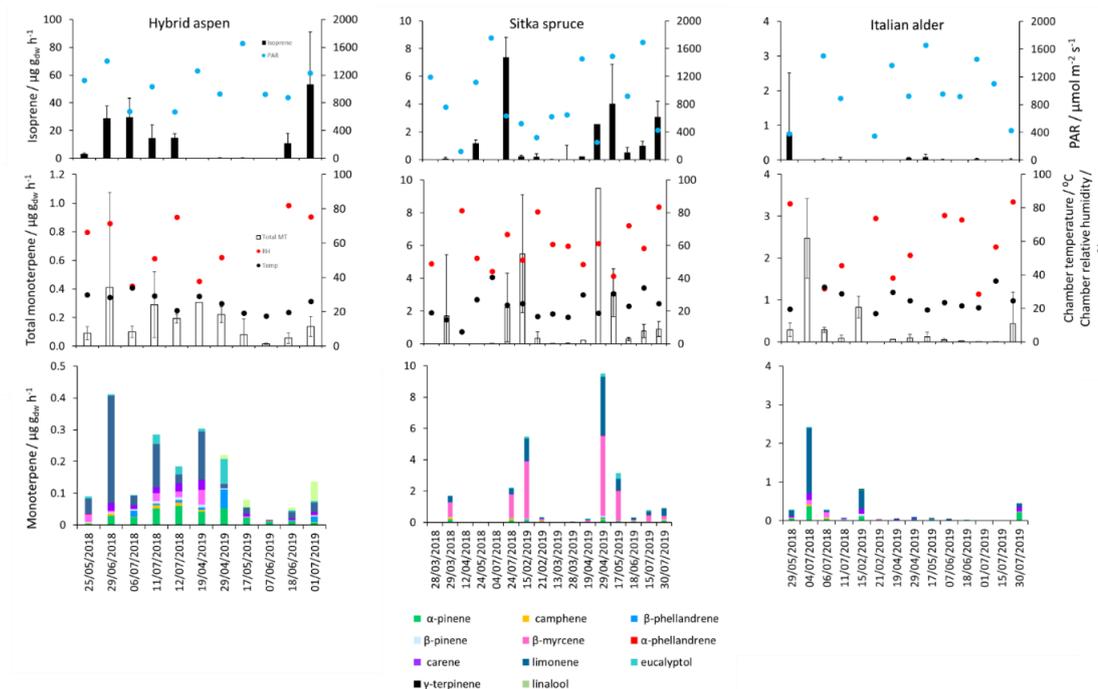


473 monoterpene emitted here, was found to correlate with an increase in temperature, comparable to
474 elevated temperature experiments for European aspen (Hartikainen et al., 2009). However, total
475 monoterpene emission rates were an order of magnitude lower in summer during our study, closer
476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

482

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

488



489

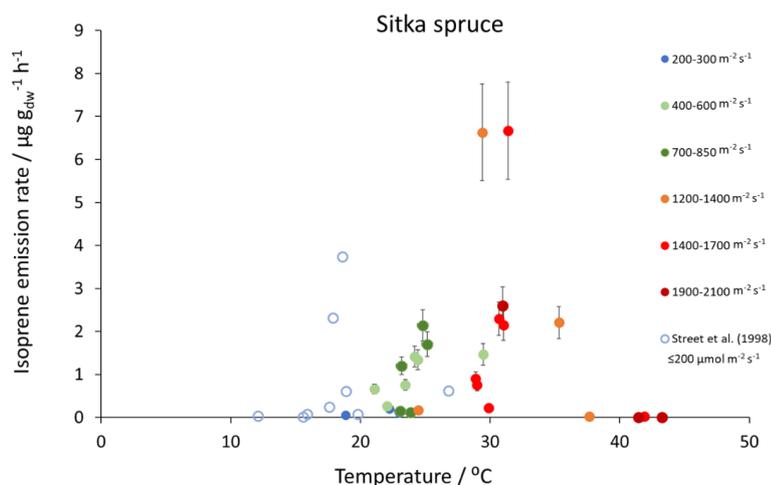
490 **Figure 1 – Average isoprene, total monoterpene and speciated monoterpene emissions from**
 491 **branches of hybrid aspen, Italian alder and Sitka spruce trees in SRF plantations at the East Grange**
 492 **site, Fife, between March 2018 and July 2019. Error bars show standard deviation of all**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

496 3.3.3 Sitka spruce

497 Average isoprene emissions from Sitka spruce ranged from $0.031 \mu\text{g C g}_{\text{dw}}^{-1} \text{h}^{-1}$ (standardised $0.14 \mu\text{g}$
 498 $\text{C g}_{\text{dw}}^{-1} \text{h}^{-1}$) in winter to $5.9 \mu\text{g C g}_{\text{dw}}^{-1} \text{h}^{-1}$ (standardised $15.0 \mu\text{g C g}_{\text{dw}}^{-1} \text{h}^{-1}$) in summer (Table 4), which
 499 are comparable to the range of previously reported emissions from UK field measurements, 0.005 -
 500 $1.48 \mu\text{g g}_{\text{dw}}^{-1} \text{h}^{-1}$ (standardised 0.88 – $14.1 \mu\text{g C g}_{\text{dw}}^{-1} \text{h}^{-1}$) (Street et al., 1996). Standardised isoprene
 501 emissions were lower in spring than summer during both years in our study (Figure 1). Standardised
 502 isoprene emissions in summer 2018 ($15.0 \mu\text{g C g}_{\text{dw}}^{-1} \text{h}^{-1}$) were more than twice those in summer 2019
 503 ($6.8 \mu\text{g C g}_{\text{dw}}^{-1} \text{h}^{-1}$), likely reflective of the wetter and cooler conditions in 2019. However, laboratory
 504 measurements using trees acclimatised at a constant laboratory temperature of $20 \text{ }^\circ\text{C}$ and PAR of



505 1000 $\mu\text{mol m}^{-2} \text{s}^{-1}$ for a week prior to sampling showed emission rates similar to summer 2018
506 emission rates, 13.4 $\mu\text{g g}_{\text{dw}}^{-1} \text{h}^{-1}$ (11.8 $\mu\text{g C g}_{\text{dw}}^{-1} \text{h}^{-1}$) (Hayward et al, 2004). The isoprene emissions in
507 our study declined dramatically at higher chamber temperatures, > 31 °C, despite the high PAR
508 levels. An optimum of 33 °C for isoprene emissions from Sitka spruce was noted by Street et al.
509 (1996), although a higher optimum of 39 °C was suggested by Hayward et al. (2004) based on a
510 laboratory study. We therefore suggest that Sitka spruce trees acclimatised under field conditions in
511 Scotland with variable day and night temperatures and light levels, may have a lower optimum
512 temperature than observed under laboratory conditions. The previous suggestion that Sitka spruce
513 reaches maximum emissions of isoprene at a low level of PAR of 300 $\mu\text{mol m}^{-2} \text{s}^{-1}$ (Hayward et al.,
514 2004) was difficult to confirm under field conditions as high PAR values were correlated with high
515 temperatures (Figure 2). However, it is worth noting that the majority of field emissions collected by
516 Street et al. (1996) align well with the emissions measured at lower PAR and temperature in this
517 study (Figure 2).



518
519 **Figure 2 – Isoprene emissions as a function of PAR and temperature for Sitka spruce at East Grange**
520 **SRF site and from Street et al. (1996) at PAR \leq 200 $\mu\text{mol m}^{-2} \text{s}^{-1}$.**

521



522 Total monoterpene emissions from Sitka spruce peaked on the 29th April 2019 ($9.5 \mu\text{g C g}_{\text{dw}}^{-1} \text{h}^{-1}$)
523 coinciding with the new shoot extension growth on the branches (Figure 1). Monoterpene emissions
524 have shown to be present in spring in advance of isoprene emissions for Norway spruce (*Picea abies*)
525 (Hakola et al., 2003). Overall, monoterpene emissions were generally higher in summer than in
526 spring (Table 4). Total monoterpene emissions were still higher in 2018 (standardised $4.5 \mu\text{g C g}_{\text{dw}}^{-1}$
527 h^{-1}) than in 2019 ($2.3 \mu\text{g C g}_{\text{dw}}^{-1} \text{h}^{-1}$) even once standardised to $30 \text{ }^{\circ}\text{C}$, which could indicate an
528 increased release of monoterpenes in response to the drier warmer conditions. The total
529 monoterpene emissions in 2019 are comparable to the previously reported total monoterpene
530 emission of $3.0 \mu\text{g g}_{\text{dw}}^{-1} \text{h}^{-1}$ ($2.6 \mu\text{g C g}_{\text{dw}}^{-1} \text{h}^{-1}$) from a laboratory study (Hayward et al., 2004).
531 Monoterpene emissions from Sitka spruce comprised predominately of β -myrcene, d-limonene, α -
532 pinene and eucalyptol, collectively accounting for 83–97% of total monoterpenes across all
533 measurement days (Figure 1).

534

535 β -myrcene was the most abundant, consistent with the findings of Geron et al. (2000), and has been
536 reported to be highest during spring in leaf oils, associated with new growth in this species, only to
537 decline later in the growing season (Hrutfiord et al., 1974) but this was not evident during our study.
538 d-limonene emission rates reported during our study are comparable in size to Hayward et al.
539 (2004), although not the dominant monoterpene as previously reported. Furthermore, other studies
540 have also reported limonene to be present in smaller quantities than α -pinene and β -myrcene
541 (Beverland et al., 1996; Hrutfiord et al., 1974). Monoterpene composition was generally consistent
542 between measurements throughout our study even though different branches and trees were
543 measured, which is perhaps a consequence of growing plantation trees propagated vegetatively
544 rather than by seed. However, the variability between the previous literature discussed here may
545 point towards the potential for different chemotypes within Sitka spruce, as suggested by (Forrest,
546 2011) and similar to that of Norway spruce (Hakola et al., 2017) and Scots pine (Bäck et al., 2012).



547 Given the dominance of Sitka spruce plantations in the UK (and Ireland), the potential for variation
548 within this species, and the limited literature data on BVOC emissions, we suggest further
549 measurements are needed at the branch and canopy level to fully assess the monoterpene
550 composition and subsequent impact on air quality.

551

552 3.4 BVOC emissions from forest floor

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

569

570 The total monoterpene emissions from the forest floor were highly variable between the three
571 chambers within the plots as demonstrated by a relative standard deviation range of 35% to 170%



572 for a given day, illustrating the highly heterogeneous soil and litter environment. All chamber
573 measurements made on the same day were averaged per species, and presented as a single flux
574 value (Figure 3) and then grouped according to season and year (Table 5).

575

576 3.4.1 Italian alder

577 Negative fluxes for total monoterpenes were measured on two occasions, 4th July and 24th July. The
578 highest total monoterpene emissions were observed on the 18th October 2018 ($18 \mu\text{g C m}^{-2} \text{h}^{-1}$) and
579 7th June 2019 ($24 \mu\text{g C m}^{-2} \text{h}^{-1}$) (Figure 3). Day to day variations were associated to some degree with
580 changes in chamber temperature and soil moisture (Figure 3). Seasonal variations in average
581 emissions were also apparent (Table 5). The forest floor acted as a sink for monoterpenes during
582 summer 2018 when there was bare soil inside the collars. During summer 2019 vegetation grew
583 inside the soil collars and resulted in the forest floor being a more substantial source of
584 monoterpenes (Figure 4). Monoterpene composition reflected the seasonal changes that occurred
585 on the forest floor. The monoterpenes emitted in autumn (October 2018) were dominated by d-
586 limonene, α -pinene and 3-carene and some β -myrcene, consistent with the composition of Italian
587 alder foliage and attributed to the accumulation of leaf litter. However, the profile in June 2019
588 during the highest total monoterpene emissions showed significant emissions of γ -terpinene and α -
589 phellandrene and likely reflects the changing understorey vegetation, hogweed sp., growing inside
590 the chamber collars and which was only present in the alder plantations. The particular species at
591 East Grange was not identified but *Heracleum mantegazzianum* (giant hogweed) has been
592 determined to be a substantial γ -terpinene emitter (Matoušková et al., 2019). This highlights the
593 importance of the specific understorey vegetation to the overall monoterpene flux composition.

594



595 **3.4.2 Hybrid aspen**

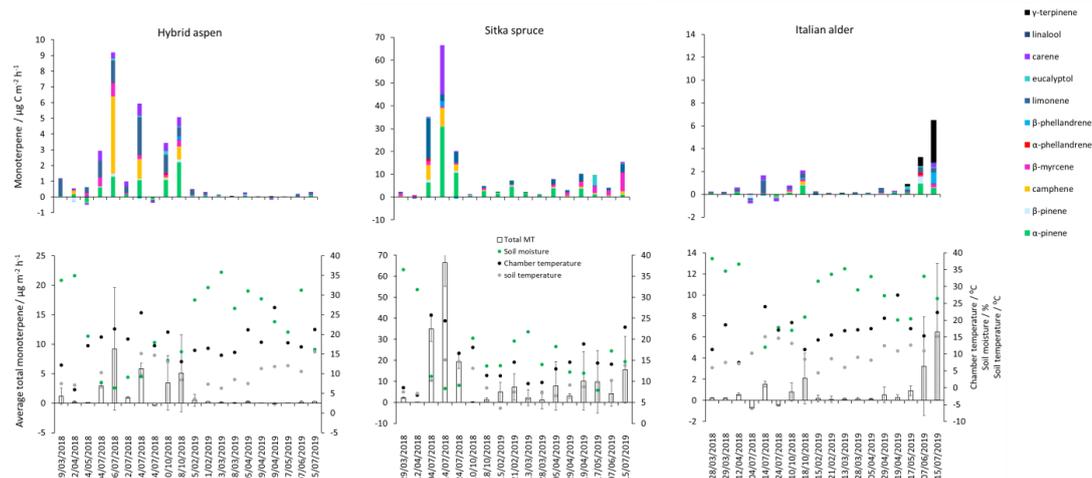
596 The highest total monoterpene emissions, $9.18 \mu\text{g C m}^{-2} \text{h}^{-1}$ and $5.83 \mu\text{g C m}^{-2} \text{h}^{-1}$, occurred in July
597 2018 and were associated with the lowest soil moisture and warm temperatures. In contrast,
598 negative monoterpene emissions were also observed in July (24th) and seem to be associated with
599 an increase in soil moisture (Figure 3). Overall spring ($0.30 \mu\text{g C m}^{-2} \text{h}^{-1}$) and summer ($0.06 \mu\text{g C m}^{-2} \text{h}^{-1}$)
600 total monoterpene emission rates in 2019 (Table 5) were smaller by an order of magnitude than
601 in spring ($0.71 \mu\text{g C m}^{-2} \text{h}^{-1}$) and summer ($3.84 \mu\text{g C m}^{-2} \text{h}^{-1}$) 2018. Higher rainfall during 2019
602 (Supplementary Information S1) resulted in increased soil moisture (Figure 3) which may have
603 suppressed some monoterpene emissions (Asensio et al., 2007b). In addition, during 2018, litterfall
604 started in July and peaked in October by which time the canopy had lost all its leaves.

605

606 The composition of the monoterpene emissions from the forest floor during 2018 was similar to
607 those measured from the branch chambers (Figure 1) and was consistent between days. The main
608 monoterpenes comprised α -pinene, β -pinene, camphene, d-limonene and 3-carene. The
609 contribution from the floor of an aspen plantation has not previously been investigated, although
610 soils taken from underneath *Populus tremula* trees showed d-limonene as the predominant
611 monoterpene with a maximum emission of $15.9 \mu\text{g C m}^{-2} \text{h}^{-1}$ under laboratory conditions (Owen et
612 al., 2007). Quantifiable emissions of monoterpene from the leaf litter of aspen (*Populus tremuloides*)
613 exist (Gray et al., 2010) although not chemically speciated

614

615



616

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

622 3.4.3 Sitka spruce

623

624 Total monoterpene emissions from the Sitka spruce forest floor peaked during July 2018 ($66.5 \mu\text{g C}$
625 $\text{m}^{-2} \text{h}^{-1}$) and coincided with the highest chamber temperatures and the lowest soil moisture readings
626 (Figure 3). The lowest emissions ($0.03 \mu\text{g C m}^{-2} \text{h}^{-1}$) were observed on the 12th April 2018 when the
627 temperature was lowest (7.5°C , Figure 3) suggesting soil moisture and temperature are likely
628 interacting controlling variables of monoterpene emissions. In addition, there were clear seasonal
629 differences when measurement days were grouped. Average summertime emissions of total
630 monoterpenes from the forest floor in 2018 were larger than those measured in 2019 (Table 5).
631 Temperatures measured in the chambers were 3°C degrees higher on average during 2018
632 compared to 2019 which could have contributed to the higher observed emissions although soil
633 moisture at 7 cm depth was not significantly different. The young Sitka spruce plantation had litter
634 present all year round unlike in the deciduous species plantations, but the covering was sparse



635 (Figure 4) compared to a mature plantation. Total monoterpene emissions in summer 2018 ($40.3 \mu\text{g}$
636 $\text{C m}^{-2} \text{h}^{-1}$) were slightly higher but similar in magnitude to the $33.6 \mu\text{g m}^{-2} \text{h}^{-1}$ ($29.6 \mu\text{g C m}^{-2} \text{h}^{-1}$)
637 previously reported for the upper-most layers of the floor in a mature Sitka spruce plantation
638 (Hayward et al., 2001). Norway spruce plantation have also been reported to have a slightly higher
639 emission rate at $50 \mu\text{g C m}^{-2} \text{h}^{-1}$ (Janson et al., 1999).

640

641 The monoterpene composition profile in 2018 was comparable to 2019 and consistent with the
642 branch emissions recorded during our study, the major emitted monoterpenes being β -myrcene, α -
643 pinene, β -pinene, d-limonene and camphene. β -myrcene accounted for a larger percentage, 20–
644 50%, of emissions in summer 2019 compared to only 5–10% in summer 2018 (Table 5), although
645 there is no obvious explanation for this difference.

646

647



648

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

652



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

Plantation type	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 N	2 2	2 4	3 4	3 3	6 8	3 3	2 2	2 4	2 2	3 9	3 9	3 9	6 17	6 18	6 17	1 2	1 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 (0.0)	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 (0.0)	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 (0.0)	15.2 (0.0)
chamber RH / %	-	-	-	-	-	-	-	-	-	88 (6)	81.4 (4.5)	77 (3)	74 (9)	73 (8)	88 (11)	70 (7)	78 (0)	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 (0)	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 (0.187)	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)	0.084 (0.003)	0.217 (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)	1.248 (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 (0.003)	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)	0.075 (0.106)	0.118 (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 (0.003)	0.152 (0.120)
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)	0.400 (0.021)	3.505 (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 (0.132)	0.133 (0.187)	0.150 (0.007)	-0.002 (0.011)	0.006 (0.144)	0.053 (0.004)	0.002 (2.980)	0.851 (0.003)	0.000 (0.003)	0.077 (0.152)	0.001 (0.346)	0.016 (0.346)	0.055 (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 (0.077)	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)	0.001 (0.002)	0.005 (0.013)	0.000 (0.001)	0.000 (0.001)	0.001 (0.002)	0.001 (0.002)	0.001 (0.004)	0.012 (0.003)	0.016 (0.003)	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)	0.001 (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 (0.215)	3.709 (5.187)	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 (6.488)	6.506 (6.488)

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

657

658 **3.5 Plantation-scale isoprene and total monoterpene emissions**

659

660 **3.5.1 Relative contribution of forest floor and canopy emissions**

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

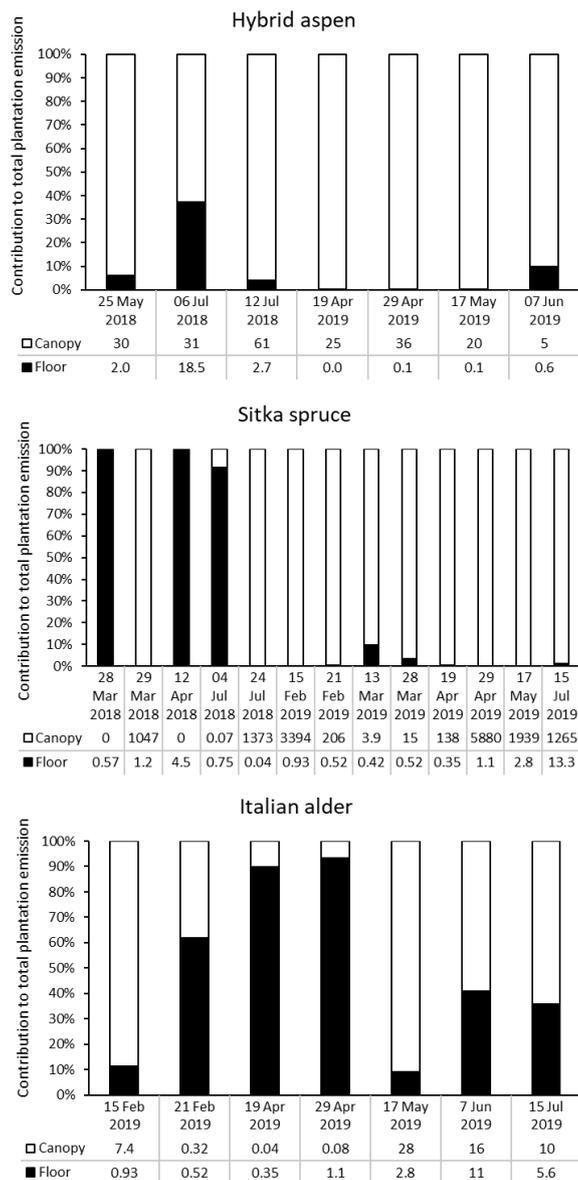


669 temperatures (spruce), the forest floor contributed the majority of the plantation monoterpene
670 emissions. This trend was also reported for conifer sp. in the boreal forest (Mäki et al., 2019b).

671

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

683



684

685

686 **Figure 5 – Percentage contribution of canopy (white bar) and forest floor (black bar) emissions to**
 687 **the total monoterpane emissions from SRF plantations at East Grange, Fife, Scotland. Numbers**
 688 **below the bars are the total monoterpane emissions in $\mu\text{g C m}^{-2} \text{h}^{-1}$.**

689



690 **3.5.2 Modelled above-canopy fluxes**

691 This section discusses modelled emissions of BVOC from the canopy per m² of ground. The “bottom
692 up” approach of estimating BVOC emissions in this study using the chamber technique is useful for
693 determining the contribution of different ecosystem components to BVOC emissions, but in this
694 section emissions do not include modelled forest floor emissions. It is noted that forest floor
695 processes are still being integrated into models in order to reliably capture the full complexity of the
696 forest floor BVOC emissions for prediction purposes (Tang et al., 2019).

697

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

709

710 Given the above, modelled average diurnal canopy emissions of isoprene for hybrid aspen were
711 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,
712 the warmest month, across both years (Figure 6A). These modelled emissions for the UK are broadly
713 comparable to those reported from measured eddy covariance flux measurements above a
714 hardwood forest, comprising primarily of aspen (*Populus tremuloides* and *Populus grandidentata*,



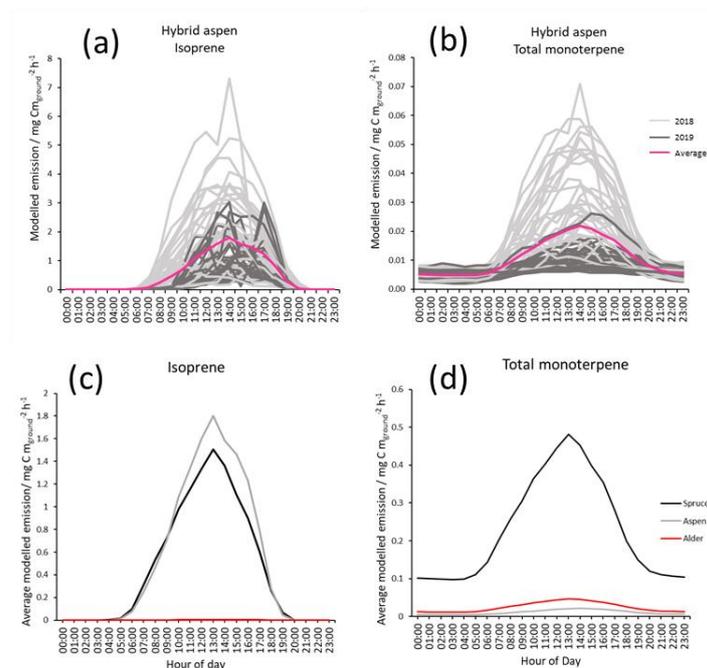
715 LAI: 3.24-3.75) in Michigan USA and the boreal forest in Canada (predominantly *Populus tremuloides*,
716 LAI: 2.4) where the average summertime emissions are reported to peak at $11 \text{ mg C m}^{-2}_{\text{ground}} \text{ h}^{-1}$ and
717 $6.87 \text{ mg C m}^{-2}_{\text{ground}} \text{ h}^{-1}$ respectively (Fuentes et al., 1999; Pressley et al., 2006).

718

719 Average total monoterpene emissions are two orders of magnitude smaller than isoprene (Figure
720 6B) for hybrid aspen. Figure 6 (C and D)) highlights the difference in the relative magnitudes of
721 emissions between the three SRF species. Average emissions from the canopy of Italian alder for
722 isoprene ($0.002 \text{ mg C m}^{-2}_{\text{ground}} \text{ h}^{-1}$) and monoterpene ($0.05 \text{ mg C m}^{-2}_{\text{ground}} \text{ h}^{-1}$) were very small and no
723 above-canopy measurements could be found in the literature for comparison. For Sitka spruce
724 average canopy scale emissions for July in Scotland were modelled to be $1.5 \text{ mg C m}^{-2}_{\text{ground}} \text{ h}^{-1}$ and 0.5
725 $\text{mg C m}^{-2}_{\text{ground}} \text{ h}^{-1}$ for isoprene and total monoterpene respectively. There has only been one attempt
726 in the UK to quantify BVOC directly above a Sitka spruce plantation (Beverland et al., 1996) where a
727 relaxed eddy accumulation system was used and average isoprene emissions were reported to be
728 $0.146 \text{ mg C m}^{-2}_{\text{ground}} \text{ h}^{-1}$ in a 24-h period in early July (temperature range 7-19 °C).

729

730



731

732 **Figure 6 – Modelled diurnal canopy emissions for July using MEGAN 2.1 of (a) isoprene from**
733 **hybrid aspen 2018 (light grey), 2019 (dark grey) and combined average emission rate (pink), (b)**
734 **total monoterpene hybrid aspen (light grey), 2019 (dark grey) and combined average emission rate**
735 **(pink), (c) average modelled isoprene for three SRF species, spruce (Black), aspen (grey) and alder**
736 **(red) for July 2018–2109, (d) average modelled total monoterpene for three SRF species, spruce**
737 **(Black), aspen (grey) and alder (red) for July 2018–2109. Results used measured PAR, temperature**
738 **and the average summer branch emission potentials collected during this study (Table 3).**

739

740

741 3.5.3 Annual above-canopy fluxes per hectare for a UK plantation

742 Table 6 shows the modelled annual BVOC emissions per hectare of plantation for each species for
743 the two meteorological years (2018-2019) at East Grange (EG) in Scotland, and for the
744 contemporaneous meteorology experienced in southern England (at Alice Holt (AH)). The modelled
745 annual fluxes of isoprene and total monoterpenes per hectare of Sitka spruce plantation averaged
746 over the two contrasting years were roughly similar, at 13.8 and 15.7 kg C ha⁻¹ y⁻¹, respectively.
747 Hybrid aspen was modelled to emit only an average of 0.3 kg C ha⁻¹ y⁻¹ total monoterpene but much



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

750

751 It is worth noting that use of an average summer flux could lead to a potential overestimation of
752 emissions during other seasons and the subsequent total annual flux. Modelled isoprene emissions
753 from Sitka spruce during 2018 for both EG and AH were higher than monoterpene emissions. In
754 2019, however, monoterpene emissions were more abundant than isoprene emissions using the EG
755 meteorology data and of the same magnitude using the AH meteorology data. The lower PAR during
756 2019, which was more pronounced for EG than AH, limited the isoprene emissions. Monoterpenes
757 were less affected as these were only temperature driven. The relative proportions of isoprene and
758 monoterpenes in the atmosphere are important since they have differing effects on the formation
759 and concentration of atmospheric pollutants such as ozone and secondary organic aerosol (SOA)
760 (Bonn et al., 2017; Heinritzi et al., 2020). Long-term BVOC emissions measurement above Sitka
761 spruce plantations is needed to confirm this model observation.

762 **Table 6 – Modelled annual isoprene, total monoterpene and total BVOC emissions per hectare of**
763 **SRF Sitka spruce, hybrid aspen and Italian alder plantations, using meteorology data from two**
764 **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
	Average		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
	Average		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
	Average		0.81	0.02	0.84

765



766 **3.6 Uncertainties in measured and modelled fluxes**

767 There are several uncertainties and simplifications in our approach to scaling-up from periodic
768 branch chamber emission measurements to annual canopy-scale predictions. We suggest that
769 uncertainties in the quantification of individual measurements of BVOC emissions are likely to be 16-
770 17% based on previous error propagation calculations (Purser et al., 2020). The nature of the
771 chamber measurement technique is likely to have an impact upon the BVOC emissions due to the
772 altered environmental conditions that may result. In addition, field-based measurements of emission
773 rates, collected under natural conditions for the UK but far from standard conditions (PAR 1000
774 $\mu\text{mol m}^{-2} \text{s}^{-1}$, temperature 30 °C) introduce an uncertainty when standardised to form emission
775 potentials.

776

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



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

802

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

808

809 **3.7 Assessing potential impact of SRF plantation expansion on UK BVOC emissions**

810 The annual average BVOC emissions data from section 3.5.3 (Table 6) was used to explore the
811 possible impact on total UK BVOC emissions arising from increased SRF planting under a suggested
812 bioenergy expansion in the UK (see introduction). The following estimates assume all bioenergy
813 expansion is SRF. However it is more likely that a combination of SRC, SRF and miscanthus could be
814 used in the UK for biomass and as such these estimates should be treated as a single extreme case



815 scenario. Meteorological data from AH and EG was used for model simulations as stated in section
816 3.5.2. Isoprene and monoterpene emissions are reported separately in Table 7 but also combined to
817 give a “total BVOC” emission.

818

819 **Table 7 – Modelled average 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

820

821 In the scenario of an expansion of 0.7 Mha of SRF, the total BVOC emissions from Sitka spruce SRF
822 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
823 much smaller at 0.6 kt y⁻¹. These potential increases in BVOC emissions are compared in Table 8 to
824 current predicted annual emissions of BVOCs from vegetation in the UK. Several air quality models
825 have been used to estimate the total isoprene and total monoterpene emissions from UK vegetation
826 (AQEG, 2020), with an earlier model (Simpson et al., 1999) determining isoprene to be the dominant
827 BVOC emission whilst later models suggest monoterpenes dominate (Hayman et al., 2017, 2010;
828 Stewart et al., 2003). The meteorological data used in some of these models are limited to a single
829 year, e.g. 1998, where the uncertainty in the model estimates could range by a factor of 4 (Stewart
830 et al., 2003), whilst others are the average emissions across many years and so report a range
831 (Hayman et al., 2017). In addition, models of UK BVOC emissions are particularly reliant upon the
832 emission potential attributed to Sitka spruce as this accounts for nearly 21% of UK forest cover and,
833 as discussed in section 3.3.3, only a limited number of studies have been conducted on Sitka spruce
834 BVOC emissions. This simple impact assessment used a limited set of meteorological data to
835 represent two contrasting years (one warmer drier year and one cooler wetter year, relative to the
836 30 year average) and for two ‘ends’ of the British climate range of temperature and PAR: north (East
837 Grange, Scotland) and south (Alice Holt, England).



838

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

842

843 It is worth noting that currently the UK has an estimated 3.2 Mha of woodland, of which 0.67 Mha is
844 covered by Sitka spruce (Forest Research, 2020) (similar in size to the future planting scenario used
845 here), a small area of alder (0.053 Mha, Forest Research, 2012) and even smaller area of aspen.
846 Comparing the total BVOC emissions for a 0.7 Mha SRF expansion scenario to the annual total BVOC
847 emissions for the UK suggests that the Sitka spruce and hybrid aspen scenarios could potentially
848 increase the total BVOC emissions in the ranges of 12–35% and 7–19% respectively, dependent upon
849 the original BVOC emission model used for this comparison (Table 8). For Italian alder this increase in
850 total BVOC is an order of magnitude smaller, ranging from 0.3–1%. It can therefore be suggested
851 that future hybrid aspen SRF plantations for bioenergy will likely emit no more BVOC than equivalent
852 expansion of young Sitka spruce plantations. Expansion of SRF with Italian alder may bring about no
853 significant changes to the UK BVOC emissions at the national level.

854

855 Any future distribution of bioenergy crops including SRF in the UK will depend on several factors
856 including available land, locations that are most suitable to obtain high biomass yields, locations that
857 are close to energy-generation plants and locations close to opportunities for CO₂ storage, in the
858 case of using BECCS to reach net-zero targets (Donnison et al., 2020). Further work is needed to
859 better understand how these changes in BVOC emissions may impact air chemistry and potentially
860 air quality (in particular ozone and SOA) at local to UK national scale.

861



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

865

Model Reference	Modelled UK total emissions / kt y ⁻¹			Sitka spruce SRF			Hybrid aspen SRF			Italian alder SRF		
	MT	Isoprene	Total	% of modelled UK emissions			% of modelled UK emissions			% of modelled UK emissions		
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

866

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

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

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

881 Overall, forest floor emissions of monoterpenes were a factor 10 to 1000 times smaller than the
 882 canopy emissions. The forest floor emissions were more variable and acted as a source for most of



883 the time with occasional instances (<4 measurement occasions out of 20) when the forest floor
884 acted as a sink for monoterpenes. Further work is necessary under controlled conditions to fully
885 understand the drivers and components of forest floor emissions.

886 Total annual emissions per unit ground area for each SRF species were derived using MEGAN 2.1 and
887 scaled up to a 0.7 Mha future SRF expansion scenario for the UK. Under this scenario, total modelled
888 UK BVOC emissions (the sum of isoprene and total monoterpene emissions) could increase by <1–
889 35% depending on the species planted and the UK BVOC emissions model used. Future work to
890 understand how any increase in forest cover and BVOC emissions may impact the atmospheric
891 chemistry in NO_x dominated regions is needed so that air quality impacts from pollutants such as
892 ozone can be determined across the UK.

893 *Author contributions.* JILM, JD and MRH conceptualized the study, acquired funds for the study,
894 supervised the study, and edited and reviewed the original draft. JILM gave permission for the use of
895 the field site at East Grange. JD provided laboratory equipment. GP contributed to the
896 conceptualization of the study, developed the methodology, collected field samples, conducted
897 measurements and analysis and wrote the original draft. RASS assisted in collection of field samples,
898 conducted measurements and analysis related to leaf area index at East Grange. LKD assisted with
899 collection of field samples and analysis.

900

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

902

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