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Methane emissions from forested closed landfill sites: Variations between tree species and landfill management practices

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### Methane emissions from forested closed landfill sites: Variations between tree species and landfill management practices



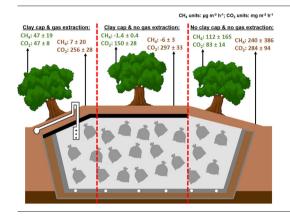
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#### HIGHLIGHTS

- Tree stem and soil GHG emissions varied between different landfill types.
- Trees on a landfill without modern management emitted the largest stem CH<sub>4</sub> fluxes.
- Stem GHG fluxes did not vary significantly between different tree species.
- Environmental conditions (waterlogging) and site age affected stem and soil fluxes.
- Including stem fluxes in total landfill GHG flux estimates would improve accuracy

#### GRAPHICAL ABSTRACT



#### ARTICLE INFO

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#### ABSTRACT

Trees in natural and managed environments can act as conduits for the transportation of methane (CH<sub>4</sub>) from below ground to the atmosphere, bypassing oxidation in aerobic surface soils. Tree stem emissions from landfill sites exhibit large temporal and spatial variability in temperate environments and can account for approximately 40% of the total surface CH<sub>4</sub> flux. Emission variability was further investigated in this study by measuring CH<sub>4</sub> and CO<sub>2</sub> fluxes from landfill sites with different management strategies and varying tree species over a 7-month period. Stem and soil measurements were obtained using flux chambers and an off-axis integrated cavity output spectroscopy analyser. Analysis showed average stem and soil CH<sub>4</sub> emissions varied significantly (p < 0.01) between landfills with different management practices. On average, tree stem CH<sub>4</sub> fluxes from sites with no clay cap but gas extraction, clay cap and gas extraction, and no clay cap and no gas extraction were  $1.4 \pm 0.4 \,\mu\text{g m}^{-2}\,h^{-1}$ ,  $47.2 \pm 19.0 \,\mu\text{g m}^{-2}\,h^{-1}$ , and  $111.9 \pm 165.1 \,\mu\text{g m}^{-2}\,h^{-1}$ , respectively. There was no difference in stem CH<sub>4</sub> fluxes between species at each site, suggesting environmental conditions (waterlogging) and site age had a greater influence on both stem and soil fluxes. These results highlight the importance of management practices, and the resultant environmental conditions, in determining CH<sub>4</sub> emissions from historic landfill sites.

#### 1. Introduction

Methane ( $CH_4$ ) is one of the most potent greenhouse gases (GHGs) with a 100-year global warming potential 28 times greater than carbon dioxide

 $(CO_2)$  (Myhre et al., 2013). Atmospheric  $CH_4$  concentrations have risen from 715 ppb in pre-industrial times to 1857 ppb in 2018 (Saunois et al., 2020). This increase in atmospheric concentration is largely a result of human activities, which includes agriculture, fossil fuel use and waste

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management practices (Ciais et al., 2013). Emissions of  $CH_4$  from the waste management sector account for approximately 12% of the global methane budget (Saunois et al., 2020). As  $CH_4$  has a relatively short atmospheric lifetime in comparison with other GHGs (approximately 8 years), it is more responsive to variations in the balance of sources and sinks (Saunois et al., 2020; Stevenson et al., 2020), and therefore, a focus of climate change mitigation strategies (Ravishankara et al., 2021).

The microbial metabolic process of CH<sub>4</sub> oxidation by methanotrophic bacteria in aerobic soils accounts for approximately 4% of the global CH<sub>4</sub> sink (Kirschke et al., 2013). Moreover, approximately 40% of the CH<sub>4</sub> produced from landfill sites is mitigated (oxidised) by the presence of aerobic surface soils (Spokas and Bogner, 2011). The rate of oxidation in aerobic surface soils is influenced by abiotic factors such as moisture content, soil texture and pH (Le Mer and Roger, 2001). Vegetation can also increase the CH<sub>4</sub> oxidation capacity in surface soils by releasing oxygen via roots into the rhizosphere (Bian et al., 2018). Moreover, vegetation modifies the chemical and physical properties of soils, such as density, water content and porosity, which alters CH<sub>4</sub> oxidation capacity (Reichenauer et al., 2011; Bian et al., 2018).

Trees growing in natural tropical and temperate ecosystems, provide a pathway for  $CH_4$  emissions from underground sources to the atmosphere via diffusion or within the transpiration stream (Terazawa et al., 2007; Gauci et al., 2010; Pangala et al., 2013; Maier et al., 2018; Barba et al., 2019;). This pathway allows  $CH_4$  to bypass oxidation in surface soils and accounts for between 27% and 87% of the total (surface and tree stem) ecosystem fluxes in temperate and tropical wetlands, respectively (Pangala et al., 2013; Pangala et al., 2015). Moreover, stem  $CH_4$  fluxes vary significantly between tree species in wetland (Pangala et al., 2015) and upland ecosystems (Warner et al., 2017; Pitz and Megonigal, 2017). In a landfill context, trees also provide a pathway for belowground  $CH_4$  to bypass oxidation, however, tree stem flux variations between species have not previously been detected (Fraser-McDonald et al., 2022).

Landfill site design and management has evolved over time to reduce both the public health and environmental risks associated with uncontrolled GHG emissions and leachates. Old-style landfills (approximately pre 1970 in the developed world) are generally located in old quarries or excavated holes with no pollution controls and a thin covering of native soil. Conversely, because of legislative drivers, a modern sanitary landfill is constructed to contain all materials and has active leachate and greenhouse gas management (Council of the European Union, 1999; Environment Agency, 2004; HMRC, 2018). Variations in landfill site design and management influence surface  $CH_4$  emissions (Environment Agency, 2004), as does the amount of organic waste deposited in the landfill (Jones and Tansey, 2015). Due to variations in gas production and consumption, and permeability rates, this results in a wide range of surface  $CH_4$  emissions that spans 0.0004 to over 4000 g m $^{-2}$  d $^{-1}$  (Bogner et al.,

1997). High temporal and spatial variability are also replicated from CH<sub>4</sub> emissions from trees stem surfaces on landfill sites (Fraser-McDonald et al., 2022). To provide further insight into this variability three hypotheses were tested: (1) GHG tree stem and soil fluxes vary between landfill sites with different management practices and ages; (2) Tree stem GHG fluxes differ between tree species; (3) Environmental controls (such as, temperature, soil moisture and pH) influence tree stem emissions.

#### 2. Methods

#### 2.1. Study sites

Field sites were selected based upon their management history and the range of tree species present. Detailed records of the quantity of organic waste deposited at each site were not available, but the management histories of the selected sites were the most comprehensive of the available field locations. Table 1 provides the details of the field sites used in this investigation.

#### 2.2. Sampling procedure

The dates of sampling visits and the stem and soil flux chambers sampled during each site visit can be seen in Table 2. Measurements from 15 trees and 5 soil locations were taken from site CC-GE. At sites CC-noGE and noCC-noGE, fluxes from 24 tree stems and 5 soil locations were sampled. The number of tree and soil sampling locations at each site was determined according to practical limitations and to allow appropriate statistical analysis. At sites noCC-noGE and CC-GE, tree species were well-mixed, so they were randomly sampled using the random walk method (Allaby, 2018). At site CC-noGE, coniferous species were growing in one area and deciduous in another. Therefore, the sampling area was split into two halves according to the type of tree present and the random walk method was used to sample trees in each half (stratified random sampling). Soil sampling locations were located on a transect across each site. A tree measurement height of 90 cm from ground level was selected based on the range of heights sampled in previous studies to aid comparisons (Pangala et al., 2013; Wang et al., 2016; Pangala et al., 2017; Maier et al., 2018). The number of each tree species sampled at site CC-GE reflected the woodland composition (Table 1). At sites CC-noGE and noCC-noGE, six trees from the four most dominant species were selected (Table 1). For each tree flux measured the GPS location, tree species and Diameter at Breast Height (DBH) was recorded. Air temperature and air pressure at each location were recorded with a Comet C4141 Thermo-hygro barometer (with an accuracy of  $\pm$  0.4 °C for temperature and  $\pm$  2 hPa at 23 °C for pressure). Tree stem surface temperature was measured using an infrared thermometer (RS Pro RS1327k; accuracy  $\pm 0.1\%$ ). At each soil and tree location, soil

Table 1

A summary of the site classifications, landfill management strategies, tree species and sampling frequency for each field site used to compare fluxes from landfill sites with varying ages and management strategies.

Site classification	Landfill management	Tree sampling	Tree species sampled		
1. Clay Cap and Gas Extraction (CC-GE)	<ul> <li>Clay cap (depth ≥ 1 m)</li> <li>Active gas extraction system</li> </ul>	15 trees     Sampled once a month	<ul> <li>10 × Betula pendula (Silver birch)</li> <li>1 × Fraxinus excelsior (European ash)</li> </ul>		
	Accepted inert, industrial, commercial and household waste from 1964 to 1998     Trees planted in 2004	Sampled once a month	4 × Prunus avium (Wild cherry)		
2. Clay Cap and no Gas Extraction (CC-noGE)	<ul> <li>Clay cap (1.8 m depth, installed in 1998)</li> <li>No gas extraction system</li> <li>Accepted household and industrial waste between the 1960s and 1990s</li> <li>Trees planted from 2000 onwards</li> </ul>		6 × Betula pendula (Silver birch)     6 × Fraxinus excelsior (European ash)     6 × Pinus nigra (Corsican pine)     6 × Prunus avium (Wild cherry)		
3. No Clay Cap or Gas Extraction (noCC-noGE)	<ul> <li>No clay cap or gas extraction</li> <li>Accepted household and industrial waste from 1971 to 1977</li> <li>Trees planted in 1998</li> </ul>	• 6 trees × 4 most dominant species • Sampled once every 4 months	<ul> <li>6 × Betula pendula (Silver birch)</li> <li>6 × Fraxinus excelsior (European ash)</li> <li>6 × Pinus nigra (Corsican pine)</li> <li>6 x Quercus rubra (Red oak)</li> </ul>		

Table 2
Sampling dates (at each stem measurement height and soil location) for all landfill sites during the sampling period. The site classified as CC-GE had a clay cap and gas extraction system. The CC-noGE site had a clay cap and no gas extraction system. The site labelled as noCC-noGE had no clay cap or gas extraction system.

		Month						
		Aug-19	Sep-19	Oct-19	Nov-19	Dec-19	Jan-20	Feb-20
Flux chambers sampled	Stem 90 cm	CC-GE CC-noGE	CC-GE	CC-GE noCC-noGE	CC-GE	CC-GE CC-noGE	CC-GE	CC-GE noCC-noGE
	Soil	CC-GE CC-noGE	CC-GE	CC-GE noCC-noGE	CC-GE	CC-GE CC-noGE	CC-GE	CC-GE noCC-noGE

temperature (Thermapen soil temperature probe; accuracy  $\pm 0.4\,^{\circ}$ C) and soil moisture (Delta-T Devices HH2 moisture meter with ThetaProbe type ML2x; accuracy  $\pm 1\%$ ) were measured at 10 cm and 6 cm depth, respectively. Soil cores were taken (within a 1 m area around tree stems) to determine bulk density and pH (Thermo Scientific Orion Versa Star Advanced Electrochemistry meter with Orion 8157 BNUMD ROSS Ultra pH ATC Triode; accuracy  $\pm 0.002$ ).

#### 2.3. Gas flux measurements

GHG fluxes from tree stems and soil surfaces were obtained using a recirculating closed loop system between gas flux chambers and a GHG analyser (ultraportable off-axis integrated cavity output spectroscopy analyser, Los Gatos Research). Semi-rigid tree flux chambers (30 imes 15 imes2 cm) were secured to tree stems (as per Welch et al., 2018), and rigid chambers (diameter 30 cm, height 20 cm) were inserted into the soil (as per Pangala et al., 2015). The GHG analyser had a measurement range of 0.01 to 100 ppm  $\pm$  2 ppb for CH<sub>4</sub>, and 1 to 20,000 ppm  $\pm$  300 ppb for  $CO_2$  measurements (Wilkinson et al., 2018). Gas concentrations in stem and soil chambers were measured over 10-min periods (approximately 600 measurements). Typically, the first 100 s of each flux measurement time series was discarded to account for setup disturbances. A linear regression line was plotted for each data set and the slope and R<sup>2</sup> values were calculated. Where R<sup>2</sup> values for fluxes were low, the regression graphs for both CH<sub>4</sub> and CO<sub>2</sub> were inspected further to inform whether the flux should be carried forward to statistical analysis. Fluxes with R2 values below 0.1 were converted to zero as no emission trend was detectable above the GHG analyser's detection capabilities. The proportion of fluxes converted to zero from sites CC-GE, CC-noGE and noCC-noGE were 16%, 20%, and 6%, respectively. Gas fluxes were determined using the ideal gas equation and standardised for temperature and pressure.

For upscaling calculations at each field site, the mean tree surface area was determined by considering the stem as a cylinder using an average diameter (of all measured trees) and a height of 3 m. The average tree surface area was multiplied by an estimated number of trees to determine the overall stem surface area at each site. An overall tree flux value was calculated from the product of the overall stem surface area and the average stem flux. The average soil surface flux and area for each site were multiplied to estimate the overall soil GHG fluxes. The magnitude of tree stem fluxes

across the site was compared with soil emissions, and the percentage contribution of tree fluxes to the overall surface flux was calculated for each site.

#### 2.4. Statistical tests

Graphs were produced using Origin (version 2020) and statistical tests were carried out in SPSS (24) and R (3.5.1). Where site and species flux data met the assumptions of normality (Shapiro-Wilk test) and equal variance (Levene's test), one-way ANOVA tests were carried out, followed by post-hoc Tukey's tests. Comparisons were made between non-normal data using Kruskal-Wallis tests, followed by Dunn-Bonferroni tests where appropriate. Full details of the statistical tests carried out are displayed in Supplementary Table 1. Stepwise multiple regression analysis was used to evaluate the relationships between tree stem fluxes and measured environmental variables. Air temperature was highly correlated with stem temperature and soil temperature (R $^2 > 0.9$ ); therefore, these variables were excluded from the stepwise multiple regression analysis. Results of the stepwise multiple regression analysis for site CC-GE were previously reported in Fraser-McDonald et al. (2022).

#### 3. Results

## 3.1. Variations in gas fluxes between landfill sites with different management practices

Stem  $CH_4$  fluxes were significantly different between landfill sites with different management techniques (p < 0.01). On average, tree stems at site CC-noGE consumed  $CH_4$ , whereas those at sites CC-GE and noCC-noGE emitted  $CH_4$ . The range of stem  $CH_4$  emissions was much greater at sites CC-GE and noCC-noGE than CC-noGE (Table 3).

Average soil  $\mathrm{CH_4}$  flux values showed uptake at site CC-noGE, relatively low emissions at site CC-GE and higher emissions at location noCC-noGE (Table 3). However, there was not a significant difference between the soil  $\mathrm{CH_4}$  fluxes from the landfill sites with varying management strategies (p > 0.05). This lack of significant difference may be due to the large range of  $\mathrm{CH_4}$  flux values, particularly from site noCC-noGE. The pattern observed between sites in stem  $\mathrm{CH_4}$  fluxes (the highest from site noCC-noGE and the lowest from site CC-noGE) was replicated in soil  $\mathrm{CH_4}$  emissions (Table 3). Based on the results shown in Table 3, excluding stem  $\mathrm{CH_4}$  emissions from flux estimates results in an underestimation of total surface

Table 3 Summary of  $CH_4$  and  $CO_2$  fluxes from a closed landfill sites with different management strategies. SE is standard error and n is the number of measurements. Sites classified as CC-GE had a clay cap and gas extraction system. CC-noGE sites had a clay cap and no gas extraction system. Sites labelled as noCC-noGE had no clay cap or gas extraction system.

	Measurement type	$CH_4 (\mu g m^{-2} h^{-1})$			$CO_2 (mg m^{-2} h^{-1})$				
		Average	SE	Range	n	Average	SE	Range	n
Site noCC-noGE	Tree stem 90 cm	111.9	165.1	4169.8	48	83.2	13.7	402.6	48
	Soil	239.6	386	3969	10	283.9	93.5	925.8	10
Site CC-noGE	Tree stem 90 cm	-1.4	0.4	10.5	48	150.3	28.1	792	48
	Soil	-6.3	3	31.3	10	297	33.4	314.6	10
Site CC-GE	Tree stem 90 cm	47.2 <sup>a</sup>	19	1406.3	105	46.7 <sup>a</sup>	7.8	453	105
	Soil	7.1 <sup>a</sup>	20.3	787.6	35	256.0 <sup>a</sup>	27.8	660.3	35

<sup>&</sup>lt;sup>a</sup> Previously reported in Fraser-McDonald et al. (2022).

emissions from forested areas of 18% and 71% for sites noCC-noGE and CC-GE, respectively. Conversely, excluding tree stem CH<sub>4</sub> fluxes from total surface flux estimates for site CC-noGE results in an underestimation of CH<sub>4</sub> uptake by 20%.

Stem  $\rm CO_2$  fluxes were significantly different between sites CC-GE and CC-noGE (p < 0.01) and sites CC-GE and noCC-noGE (p < 0.01). There was no significant difference in the stem  $\rm CO_2$  fluxes between sites CC-noGE and noCC-noGE (p > 0.05). On average, stem  $\rm CO_2$  fluxes from sites CC-noGE and noCC-noGE were higher than those from location CC-GE (Table 3). There was no significant difference between the soil  $\rm CO_2$  fluxes from the different landfill sites (p > 0.05). The averages and ranges of the measured environmental variables for the CC-noGE and noCC-noGE landfill sites are in Supplementary Table 2. Averages and ranges of ancillary variables measured at site CC-GE are reported in Fraser-McDonald et al. (2022). Full results of the stepwise regression analysis for sites CC-noGE and noCC-noGE are in Supplementary Table 3. At site noCC-noGE, none of the measured ancillary variables significantly accounted for the variance in CH<sub>4</sub> fluxes. At site CC-noGE, soil pH was correlated with CH4 fluxes, although this variable only accounted for around 10% of the variance in fluxes.

#### 3.2. Variations in gas fluxes between tree species

At site noCC-noGE, *Pinus nigra* trees emitted more CH<sub>4</sub> on average than other species, particularly *Fraxinus excelsior* (Supplementary Table 5).

However, the variation between the  $CH_4$  fluxes from different species at site noCC-noGE was not significant (p > 0.05). This is most likely due to the large range of flux values from *Pinus nigra* trees.

There was no significant difference in mean  $CO_2$  fluxes between the tree species at site noCC-noGE (p>0.05) and the range of fluxes did not vary substantially between species (Fig. 1B; Supplementary Table 5. However, in October 2019, *Quercus rubra* trees emitted significantly more  $CO_2$  than *Fraxinus excelsior* trees (p<0.05).  $CO_2$  fluxes from other species did not show a significant difference in October 2019 (p>0.05).

There was no significant difference in mean  $CH_4$  fluxes between different trees species at the 90 cm measurement height at site CC-noGE (p > 0.05). The ranges of  $CH_4$  fluxes from *Quercus robur* and *Betula pendula* were slightly larger than those for *Prunus avium* and *Pinus nigra*, but this did not significantly alter the variation between species (Fig. 1A; Supplementary Table 4).

Despite higher average fluxes from *Quercus robur*, there was no significant difference in  $CO_2$  fluxes between tree species at site CC-noGE (Fig. 1B). However, in August 2019, *Quercus robur* trees emitted significantly more  $CO_2$  than *Pinus nigra* (p < 0.01) and *Betula pendula* trees (p < 0.05). The  $CO_2$  fluxes from other species did not show a significant difference in August 2019 (p > 0.05).

Full results relating to the differences in GHG fluxes between trees species at site CC-GE have previously been published (Fraser-McDonald et al., 2022).

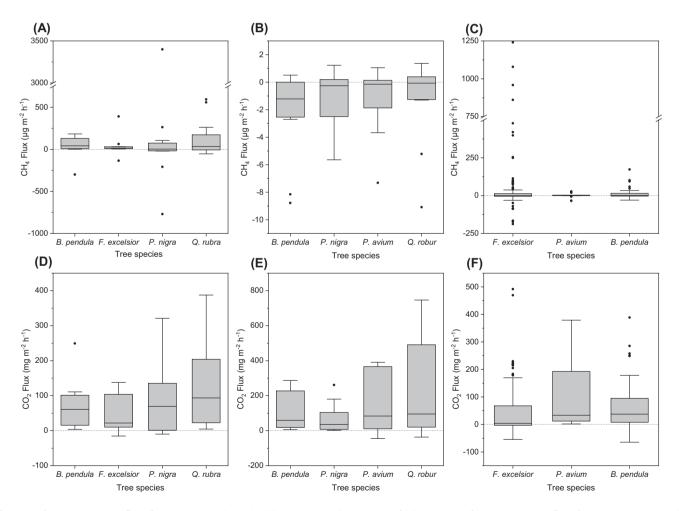


Fig. 1. Boxplots comparing  $CO_2$  fluxes between tree species at site (A) noCC-noGE, (B) CC-noGE, and (C) CC-GE. Boxplots comparing  $CO_2$  fluxes between tree species at site (D) noCC-noGE, (E) CC-noGE, and (F) CC-GE (outliers have been removed; data in full in Fraser-McDonald et al., 2022). The middle line indicates the median value, and the whiskers are determined by the 5th and 95th percentiles. The dots represent outliers (below  $Q_1$ –1.5 Interquartile range or above  $Q_3$  + 1.5 Interquartile range). Sites classified as CC-GE had a clay cap and gas extraction system. Sites labelled as noCC-noGE had no clay cap or gas extraction system.

#### 4. Discussion

# 4.1. Variations in gas fluxes between landfill sites with different management strategies

On average, sites CC-GE and noCC-noGE were a net source of CH<sub>4</sub> during the measurement period, whereas site CC-noGE was a sink. UK landfills with no engineered cap emit more CH<sub>4</sub> in the first 15 years after waste deposition than those with engineered caps (2.14  $\times$  10<sup>-2</sup> and 1.39  $\times$  10<sup>-5</sup> mg m<sup>-2</sup> s<sup>-1</sup>, respectively) (Environment Agency, 1999). However, peak CH<sub>4</sub> production occurs approximately 5 to 7 years after waste has been deposited, with most gas being produced within 20 years after deposition (ATSDR (Agency for Toxic Substances and Disease Registry), 2001). Therefore, it was expected that site noCC-noGE would have a low rate of CH<sub>4</sub> production and the lowest stem CH4 emissions. However, our results do not concur with this expectation as stem CH<sub>4</sub> emissions from site noCC-noGE were the highest. Soil CH<sub>4</sub> emissions from site noCC-noGE were also higher than those from the other sites, although this was not significant (likely due to the large range of soil fluxes from this site, as shown in Table 3). The soil fluxes from site noCC-noGE were not atypical of landfill surface fluxes, which can range between 0.0004 g  $m^{-2} d^{-1}$  and over 4000 g  $m^{-2} d^{-1}$ and are highly variable (Bogner et al., 1997).

Higher stem and soil CH<sub>4</sub> fluxes at the noCC-noGE landfill compared with other landfills are most likely explained by waterlogging at this site. Average soil moisture at site noCC-noGE (40.3  $\pm$  1.5%) was greater than at sites CC-GE (25.6  $\pm$  0.8%) and CC-noGE (24.0  $\pm$  1.3%), with visible waterlogging during some sampling visits. The formation of waterlogged regions in the soil would lead to localised anaerobic zones where CH<sub>4</sub> was produced (Le Mer and Roger, 2001). If tree roots grow in these areas, CH<sub>4</sub> can be transported via diffusion into the roots and stem, before being emitted to the atmosphere (Covey and Megonigal, 2019). If the management of closed landfills allows a surface structure to develop that is non-uniform and not free-draining (as at site noCC-noGE), these managed  $\,$ environments have the potential to emit higher levels of CH4 than expected from historic sites due to natural biochemical processes. Indeed, average stem CH<sub>4</sub> fluxes at site noCC-noGE were of a similar magnitude to fluxes recorded in temperate wetlands (Terazawa et al., 2007; Gauci et al., 2010; Pangala et al., 2015; Terazawa et al., 2015). The results from site noCC-noGE agree with experimental findings that trees grown in conditions where the water table is high emit significantly higher levels of CH<sub>4</sub> from the stem surface, when compared to trees grown under free draining aerobic soil conditions (Pangala et al., 2014).

Relatively high stem CH<sub>4</sub> emissions were expected from site CC-noGE. Landfill caps are designed to prevent the uncontrolled release of landfill gases from waste, but with no extraction system to remove this CH<sub>4</sub>, there would potentially be more transported to the atmosphere via the tree methane pathway (Dobson and Moffat, 1993). However, stem fluxes at site CC-noGE were significantly lower than those from the other landfill sites. The site stopped accepting waste in 1998 and as peak CH<sub>4</sub> emissions may have occurred between 5 and 15 years after the site closed, it is possible that CH<sub>4</sub> production was no longer sufficient to result in significant emissions (Environment Agency, 1999). Additionally, CH<sub>4</sub> produced in the waste may have been transported away from the source laterally, particularly due to increased subsurface pressure when capping took place and if any flaws existed in the side wall lining (Christensen et al., 1989; LGG (Landfill Guidance Group), 2018). Moreover, landfill caps are approximately 85% effective at preventing the release of GHGs from landfill sites (Jardine et al., 2006) and, on average, 40% of CH<sub>4</sub> emissions are offset via oxidation by methanotrophic bacteria in the overlying cover soil (Abushammala et al., 2014). CH<sub>4</sub> can also be oxidised by bark-dwelling methanotrophic bacteria, which would further reduce CH<sub>4</sub> emissions from tree stems (Jeffrey et al., 2021a; Jeffrey et al., 2021b). Consequently, the soils and tree stems at some landfills, including site CC-noGE, will exhibit negative CH<sub>4</sub> fluxes (Spokas and Bogner, 2011). Average stem and soil CO2 emissions at site CC-noGE were higher than those at sites CC-GE or noCC-noGE. Emissions at site CC-noGE, particularly from the soil, are similar to those from temperate upland environments which are net sinks of  $CH_4$  and sources of  $CO_2$  (Warner et al., 2017; Pitz and Megonigal, 2017; Maier et al., 2018).

Emissions from site CC-GE were expected to be lower than CC-noGE as the final soil cover and gas control system adhere to modern design requirements set by the EU directive of 1999 (Environment Agency, 2004). However, average stem CH<sub>4</sub> fluxes from site CC-GE were significantly higher than those from site CC-noGE. As gas extraction systems are not 100% effective at capturing all CH<sub>4</sub> (50 to 90% efficiency range), it is possible that the gas control system at site CC-GE is not removing all the GHGs produced in the waste (Abushammala et al., 2014). As site CC-GE was closed most recently of those investigated, it was likely to still have the greatest rate of CH<sub>4</sub> production from the waste and was therefore expected to have higher emissions than site noCC-noGE (the oldest landfill). However, this pattern was not observed. There were hotpots of emissions at site CC-GE, similar to noCC-noGE, despite there being no evidence of waterlogged soils. It is likely that rather than saturated soils causing anaerobic zones, the localised CH<sub>4</sub> fluxes from site CC-GE were caused by leaks in the landfill cap or gas extraction system. If landfill caps are subjected to cycles of wetting and drying or desiccation fissures can form, resulting in hotspots with significantly higher surface fluxes and high temporal variability (Rachor et al., 2013; Sinnathamby et al., 2014).

The results presented here have enabled a novel comparison of stem CH<sub>4</sub> fluxes at the 90 cm measurement height on different landfill sites. Results indicate that omitting tree stem fluxes from emissions estimates for forested landfill sites may result in an underestimation of the overall site flux. However, as CH4 emissions are not uniform across the surface of tree stems (Terazawa et al., 2015; Pangala et al., 2017), measuring fluxes at one stem height may not be representative of fluxes across an entire site. Nevertheless, the results do suggest that excluding tree stem fluxes from emission estimates for forested landfill sites would not provide an accurate representation of the overall site flux. This is particularly important in relation to modern sustainably managed landfill in which the oxidation of residual GHG emissions in cover soils plays an important role in minimising uncontrolled gas emissions (Grossule and Stegmann, 2020). Average soil surface flux values obtained from this study are expressed in different units in Supplementary Table 6 to allow for comparison with the target flux limit for sustainably managed landfill (0.5 l m<sup>-2</sup> h<sup>-1</sup>) (Cossu et al., 2020). Soil fluxes from CC-GE and CC-noGE sites are below the target, whereas the average flux from the noCC-noGE site was above this value. It should be noted that the landfill sites sampled during this investigation were not designed to be sustainable and the flux variations were likely due to environmental factors (such as the waterlogging at site noCCnoGE). Tree stems provide a conduit for GHG transport from belowground to the atmosphere, thus bypassing oxidation in cover soils, it is important that this emission pathway is considered in modern landfilling practices.

#### 4.2. Variations in gas fluxes between tree species

CH<sub>4</sub> and CO<sub>2</sub> emissions vary between some tree species in forested temperate wetland and upland environments (Pangala et al., 2015; Warner et al., 2017; Pitz and Megonigal, 2017; Pitz et al., 2018). Factors such as wood specific density, lenticel density, stem diameter, sap flow and transpiration rates contribute to the difference in stem GHG fluxes between species (Pangala et al., 2013; Pitz et al., 2018). However, no significant differences in CH<sub>4</sub> fluxes between different tree species were observed at sites CC-noGE and noCC-noGE; this concurs with results from a landfill site with a cap and gas extraction system (Fraser-McDonald et al., 2022). The lack of variation in CH<sub>4</sub> fluxes between different species at site CC-noGE may be due to the overall low flux values from all trees on this site, however, fluxes from site noCC-noGE were of a similar magnitude to those from natural temperate ecosystems (Pangala et al., 2015; Pitz et al., 2018). CH<sub>4</sub> fluxes from the tree species sampled in this research have not previously been measured before in natural temperate woodlands, suggesting stem surface emissions from the species listed in Table 1 are not different, or that ephemeral conditions do not produce the same stem emission profiles observed from trees

growing in permanent waterlogged conditions. The measurement of  $CH_4$  fluxes from a greater variety of tree species in natural and managed environments, may aid in determining how tree species influence the magnitude of stem  $CH_4$  emissions in temperate environments. The results presented here therefore suggest that the magnitude of  $CH_4$  emissions was more likely determined by landfill site conditions than tree characteristics.

#### 5. Conclusion

This study has revealed that trees growing on closed landfill sites with different management techniques and environmental conditions emit varying quantities of GHGs. On average, trees on the oldest site (noCC-noGE) and the most recently closed site (CC-GE) were a source of CH<sub>4</sub>, whereas trees on site CC-noGE were a CH<sub>4</sub> sink. Evidence suggests that the variation in average CH<sub>4</sub> fluxes between the different landfill areas was likely a result of the rate of CH<sub>4</sub> production in the waste (linked to the ages of the site), the susceptibility of the area to waterlogging, and landfill management techniques put in place upon closure. CH<sub>4</sub> emissions from site noCC-noGE indicated that the management (or lack thereof) of some closed landfill sites can result in surface drainage becoming impeded in places. Subsequently, soil and stem CH<sub>4</sub> emissions from this site were greater than expected from a relatively old landfill site and were similar in magnitude to a natural wetland ecosystem. These results indicate that management strategies used during and after closure, and resultant environmental conditions, can affect the magnitude of GHG emissions from former landfills. Findings show that excluding stem CH<sub>4</sub> emissions from flux estimates results in an underestimation of total surface emissions from forested areas of 18% and 71% for sites noCC-noGE and CC-GE, respectively. Conversely, excluding tree stem CH<sub>4</sub> fluxes from total surface flux estimates for site CC-noGE results in an underestimation of CH<sub>4</sub> uptake by 20%. This has implications when considering the contribution of legacy emissions from different closed landfill sites to carbon assessments and may inform landfill policy and practice.

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#### CRediT authorship contribution statement

A. Fraser-McDonald: Conceptualization, Methodology, Formal analysis, Investigation, Writing – original draft, Writing – review & editing, Project administration. C. Boardman: Conceptualization, Funding acquisition, Methodology, Writing – review & editing, Project administration, Supervision. T. Gladding: Funding acquisition, Methodology, Writing – review & editing, Project administration, Supervision. S. Burnley: Funding acquisition, Methodology, Writing – review & editing, Project administration. V. Gauci: Conceptualization, Funding acquisition, Methodology, Writing – review & editing, Project administration, Supervision.

#### Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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