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Influence of Sapwood/Heartwood and Drying Temperature on Off-Gassing of Scots Pine Wood Pellets

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Abstract

Wood pellets produced from fresh sawdust can form and release uncontrolled gases during bulk storage, a tendency referred to as off-gassing. This study investigated the off-gassing tendencies of Scots pine wood pellets made from separated sapwood and heartwood sawdust. The effects of drying temperature, raw material storage, as well as varying proportions of sapwood and heartwood were also investigated. There was a strong linear correlation between off-gassing and sapwood content, with correlation coefficient (R) values greater than 0.9 at $p < 0.001$ for all the off-gases. An increase in sapwood content of the feedstock led to a significant increase in off-gassing of CO_2, CO, and CH_4, and O_2 consumption. The drying temperature of the raw material had a significant effect on off-gassing of both sapwood ($F_{(8, 26)} = 51.32$, $p < 0.05$) and heartwood ($F_{(8, 26)} = 334.1$, $p < 0.05$) pellets. Increasing the drying temperature for heartwood resulted in increased off-gassing, while for sapwood, the off-gassing reduced. Storage of sapwood raw material before pelletization reduced the off-gassing of wood pellets, whereas for heartwood, it had no significant impact. Based on the results, it is suggested that a biological process, in combination with the chemical oxidation of fatty acids, lay behind the off-gassing of wood pellets.

Keywords Biofuel pellets · Bulk storage · Gas emissions · Carbon oxides · Methane · Feedstock composition

Introduction

Over the year, the use of fuel wood pellets, both for heating and electricity generation, has continued to increase due to their enhanced and standardized quality properties. Wood pellets are a preferred solid biomass fuel due to their uniform size, high energy density, and low moisture content, making them easy to transport and store [1]. These features make wood pellets suitable for use in small residential furnaces as well as in industrial combined heat and power plants, either as a substitute or co-fired with coal. The quality of wood pellets produced is influenced by several factors, including processing parameters, biomass feedstock, moisture content, and particle size [2–4].

In recent years, the global production of wood pellets surged from roughly 18 million tonnes in 2012 to around 42 million tonnes in 2020 [5]. The European Union remains the world's biggest market for wood pellets, with 75% of the total global consumption. In 2018, the EU's wood pellets consumption was approximately 27.35 million tonnes and is expected to increase by 30 – 40% between 2021 and 2026 [6]. However, this demand cannot be met by local supply within the EU; only 54% of the union's consumption share

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can be produced locally [7]. As a result, vast quantities of wood pellets are imported primarily from North America, leading to an increase in the amount of wood pellets in transportation and bulk storage to ensure a steady supply.

Bulk storage of wood pellets can pose a potential hazard because of the possibility of off-gassing and self-heating [8–10]. Off-gassing, particularly the release of carbon monoxide, carbon dioxide, and other volatile organic compounds, poses a risk to human health and the environment [11–14]. The underlying causes of self-heating and off-gassing in biomass, including wood pellets, are suggested to be a result of biological decomposition, physical transition reactions such as water absorption, chemical oxidation, or a combination of these processes [9, 15–17]. However, chemical oxidation is suggested to be the primary contributor to off-gassing of wood pellets. Several studies have indicated that off-gassing in wood pellets can be attributed to the oxidation of wood extractives [18–21], with the type rather than the amount of extractives having a significant impact [22]. Siwale et al. [23] specifically found that the off-gas emissions of carbon oxides were influenced by the availability of unsaturated fatty acids, while the resin acids and saturated fatty acids content had limited or less effect.

Fuel pellets are mainly produced from woody materials (i.e., secondary xylem), which include industrial residues, such as sawdust, shavings, and wood dust from primary and secondary wood processing [24]. Wood is divided into sapwood and heartwood. These are often easily distinguishable in a cross-section of a mature stem because of different colour; there are however exceptions such as spruce and birch. Sapwood, which is the outer layer of the xylem, is usually lighter in colour than heartwood, the inner darker wood [25, 26]. Sapwood contains physiologically active cells including some living cells and performs three primary functions of support, water conduction, and nutrition storage in a living tree. In softwoods, tracheids constitute the most abundant wood cells, accounting for approximately 95% of the total wood volume. These cells perform dual roles of support and transporting of water and minerals and are dead when thy carry out their biological functions, while parenchyma cells store nutrition in form of starch and fats, and work as a biological defence, excreting biological active matters such as antimicrobial toxins [27]. When a tree is felled and the wood dries, the starch and the fat are retained in the wood cells as a permanent deposit. Heartwood is the oldest part of the wood and lacks living cells. It has no conduction and storage functions, but only provides structural support. It is also impregnated with antimicrobial molecules [28].

The transition of sapwood into heartwood involves changes in the chemical composition of the extractives and primary chemical components of the wood cell walls [27, 29, 30]. In general, the sapwood of various pine species has been found to contain more cellulose than heartwood, while the differences in the composition of other non-cellulosic carbohydrates are small. The heartwood of pines has also been reported to contain slightly higher amounts of lignin than sapwood [31–33]. However, the most notable chemical difference between the two wood types is in the composition of extractives [34–37]. In a comprehensive study on wood extractives, Nisula [34] found that heartwood contains more extractives than sapwood. The amounts of extractives in different pine species were in the ranges of 2.3–8.9% for heartwood and 0.76–3.7% for sapwood. The wood extractives in heartwood contain more lipophilic and hydrophilic extractives than those in sapwood [26]. For instance, the extractives of Scots pine sapwood mainly contain triglycerides, while those of heartwood have more of pinosylvins, resin acids, and free fatty acids [38]. This reflects the different biological functions of the two types of wood; sapwood extractives are mainly for energy storage, while heartwood extractives prevent microbial decay.

To prevent the occurrence of off-gassing in wood pellets, Arshadi et al. [18] found that by adding low amounts (0.5%) of TBHQ (tert-butylhydroquinone) in sawdust, it was possible to block the autoxidation of fatty and resin acids and reduce off-gas emissions by 72 to 90% depending on the pellets temperature. Another study in which an antioxidant was used also reported that adding acetylsalicylic acid to sawdust reduced the emission of CO, CO_2, and VOCs during storage of wood pellets [19]. Attard et al. [39] further showed that using fat-free sawdust to produce the pellets can reduce the off-gassing by 80 – 90% compared to ordinary wood pellets. The wood pellet industry mitigates self-heating and off-gassing by using a larger proportion of stored sawdust in the biomass feedstock mix. The sawdust raw material is stored for 6 to 12 months before use [40], and this leads to reduction in the triglyceride extractive content, i.e., bounded fatty acids and changes in dry matter composition, thus making the stored sawdust lose the ability of fresh sawdust to form gasses [41, 42]. However, as demand for wood pellets increases, it is becoming less feasible to store the sawdust for long periods of time due to associated logistical costs such as capital binding, the need for a large storage area, and total wood weight loss caused by uncontrolled microorganism action [41]. Therefore, there is a need to explore ways of preventing self-heating and off-gassing in wood pellets produced from fresh sawdust.

Sawdust obtained from different parts or sections of a tree may have varying off-gassing and self-heating effects. Sorting sawdust and other residues to obtain the desired raw material for pelletization is technically feasible in sawmills and other wood product industries. However, accurate sourcing of desired raw material can only be achieved if sufficient information is available about their off-gassing and self-heating tendencies. Additionally, the diverse pretreatments, including drying systems, employed in wood

pellet production can also influence the extent of off-gassing and self-heating of the pellets. The purpose of this study is to generate knowledge that will assist in selecting raw materials and pre-treatment measures, with the goal of producing wood pellets that are not prone to self-heating and off-gassing.

The aim of this study was to investigate the off-gassing tendencies of Scots pine wood pellets made from separated sapwood and heartwood sawdust. Furthermore, the study also investigated the effects of drying temperature, raw material storage as well as varying proportions of sapwood and heartwood in the raw material on off-gassing abilities of the wood pellets. The extractive contents of the sawdust raw materials and selected quality properties of the pellets were also determined and are recorded here.

Materials and Methods

This section gives a detailed description of the materials and methods used to conduct this study. The particle size distribution of the sawdust raw material and quality properties of the wood pellets are also presented in this section and not under the results.

Raw Materials and Preparation

The raw material used in this study was Scots pine (*Pinus sylvestris*) sapwood and heartwood sawdust. The sawdust was prepared from freshly cut sawn timber sourced from Rinns Såg AB Sawmill located in Torsby, Sweden. Sapwood sideboards measuring $25 \times 100 \times 1500$ mm, cut from the outer part of the saw logs and heartwood pieces measuring $38 \times 125 \times 1500$ mm, cut from the inner part of the same saw logs, were used. To prepare the sawdust, the sawn timbers were initially ripped into wood strips using a bench power-driven ripping saw. These wood strips were further chipped into smaller particles using a garden chipper. The resulting wood chips were dried and milled through a 6-mm sieve to produce the sawdust. Sieve analysis was conducted to determine the particle size distribution of milled sapwood and heartwood sawdust. The procedure involved the screening of about 500 g milled material through horizontal vibrating standard sieves with sizes arranged in decreasing order. More than 99% of the particles for both sapwood and heartwood sawdust were less than 4 mm, with over 50% being smaller than 1.4 mm and about 28% smaller than 1 mm (Table 1).

Experimental Design

The undried wood chips obtained from both sapwood and heartwood materials were divided into two parts. One part of each material was further divided into three discrete batches, which were then dried separately in a bed drier using three different drying temperatures as follows: air drying at 40 °C, air drying at 80 °C, and steam drying at 180 °C, and thereafter milled. The remaining undried sapwood and heartwood chips were stored indoors in an open-air environment at room temperature for 7 months. After the storage period, the chips were air dried at 40 °C and milled into sawdust. The fresh sapwood and heartwood sawdust materials, which were air dried at 40 °C, were mixed in approximately 25 kg batches to achieve sapwood to heartwood mass percentage contents of 0:100%, 25:75%, 50:50%, 75:25%, and 100:0%. Furthermore, additional 25 kg batches were prepared from fresh sapwood and heartwood sawdust that were dried at 80 and 180 °C, as well as stored sapwood and heartwood sawdust air dried at 40 °C. This brought the total number of sawdust batches to eleven (Fig. 1). The target of 25 kg of sawdust per batch was aimed at obtaining sufficient raw material for the production of approximately 20 kg of wood pellets. This quantity was necessary for conducting off-gassing tests and pellet quality measurements. Each batch of sawdust was conditioned to around 11.4% moisture content by adding the required amount of water. To ensure uniform distribution and absorption of moisture, the materials were mixed separately using a SoRoTo 100 L Forced Action Mixer for approximately 10 min. After mixing, the materials were sealed in plastic bags and left for 48 h before they were pelletized.

Total Extractives and Fatty and Resin Acids

The total hydrophilic and lipophilic extractives were determined by separately conducting water and organic solvents extractions, in a Soxhlet apparatus. To determine the water-soluble extractives, 25 cycles of extraction were carried out for each sample, using water as the solvent, while for lipophilic extractives, a solvent mixture of petroleum ether and acetone, in a 90:10, v/v ratio was used, and the extraction was run for 12 cycles. For each material, about 3 g of

Table 1 Particle size distribution of the milled sapwood and heartwood sawdust

Particles passing through the sieve (%)						
Material	5.6 mm	4.0 mm	2.8 mm	2.0 mm	1.4 mm	1.0 mm
Sapwood	100	99.8	98.7	84.3	54.2	27.9
Heartwood	100	99.9	98.9	86.3	55.4	28.7

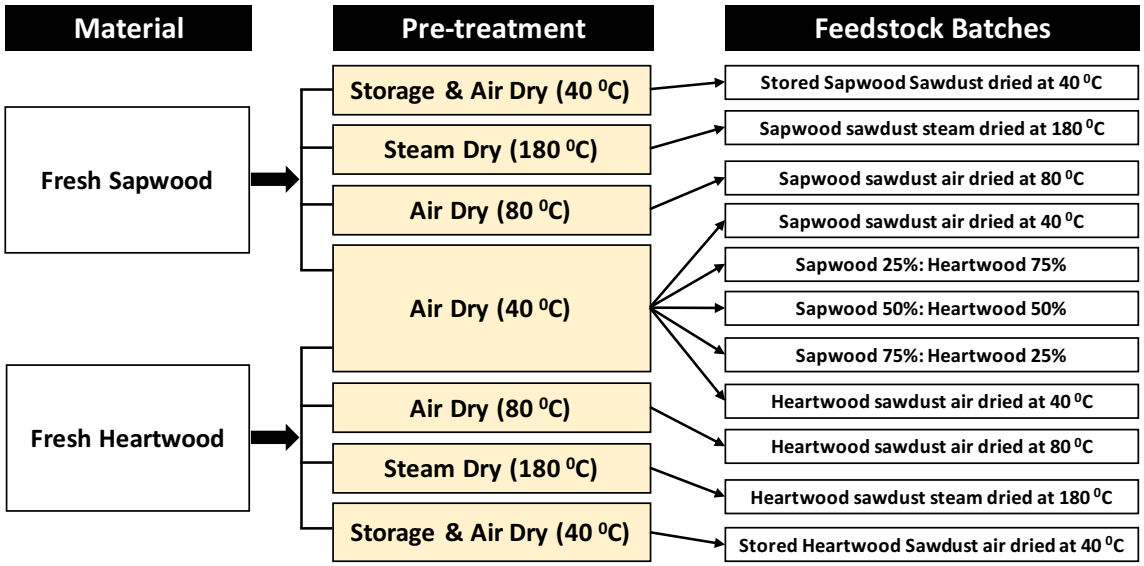

Fig. 1 Design of the study showing the raw material treatments and the feedstocks used to produce the wood pellets

sawdust samples was extracted in triplicate. The lipophilic extracts were further analysed for fatty and resin acids composition using a Hewlett Packard (HP6890-5973) GC/MS instrument. Full scan EI mass spectra (m/z 35–500) were recorded, and the NIST Mass Spectral Search Program (version 2.0) was utilized to identify the peaks. For quantitative analysis, an internal standard heptadecanoic acid with a known concentration was used. The fatty and resin acids determination method is described in detail in Arshadi and Gref [14].

Pelletizing

The production of pellets was carried out using an Amandus Kahl pellet press, Model 14–175. The fresh heartwood pellets were produced using a die with channel diameter of 6 mm and press channel length of 30 mm. The fresh sapwood, mixed, as well as stored sawdust were pelletized using a 24-mm press channel length die. Before production, the dies were pre-heated in an oven at 103 °C for 24 h and then run with trial raw materials until the die temperature reached and stabilized in the range of 70–100 °C, which is the required temperature for pelletization. For the heartwood 40 °C pellets, automated feeding was mostly used, while the rest of the materials had integrated automatic and manual feeding. the pellets were put into off-gassing containers on the same day of production, just after cooling.

Off-Gassing Measurement

The off-gases, CO, CO_2, and CH_4, and residual O_2 were measured using an ECOM J2KN Pro-IN gas analyser. This is a multi-instrument that detects and quantifies the gases using electrochemical and infrared (IR) sensors. The pellets (about 8–9 kg corresponded to about 70% of the container's volume capacity) were stored in 20-L airtight polyethylene containers with screw caps. Duplicate samples for each and every batch of pellets were stored and measured for off-gassing. At every measuring time, the gases were detected and measured in real time by inserting the gas analyser's probe into the headspace of the container, through the sampling port positioned at the top of the container. The probe was kept inside for 1 to 2 min so as to equilibrate before taking the reading. Measurements were taken every 24 h throughout the 13-day storage period, and the cumulative gas concentrations were calculated from these daily measurements. The detailed procedure of this method is described in Siwale et al. [22, 23].

Pellet Quality Properties

All pellet batches were subjected to analysis to determine their moisture content, bulk density, and durability. The pellet and die temperatures were also recorded for every batch of pellets during production. The pellet quality properties and production temperatures for all eleven batches of wood pellets that were investigated are listed in Table 2. The values are arithmetic means from replicate determinations, and the ± indicates the standard deviation. The moisture content was determined by drying at least 300 g of pellet samples in an oven at 105 ± 2 °C until a constant weight was obtained. The bulk density was measured using a standardized cylindrical container of 5 l volume. The tared weight of the container was first recorded and thereafter, the container was filled with pellets and subjected to two impact falls from a height of 15 cm. The weight of the

Table 2 Selected pellet quality properties and production temperatures

Material	Pellet MC (%)	Durability (%)	Bulk density (kg/m^3)	Die temp. (°C)	Pellet temp. (°C)
Fresh sapwood air dried (40 °C)	8.3 ± 0.04	96.8 ± 0.05	600 ± 1.7	89.6 ± 4.8	89.3 ± 2.5
Fresh sapwood air dried (80 °C)	8.0 ± 0.31	97.6 ± 0.09	602 ± 3.8	95.6 ± 2.5	95.1 ± 1.6
Fresh sapwood steam dried (180 °C)	8.3 ± 0.04	96.7 ± 0.01	611 ± 5.7	98.9 ± 1.5	94.6 ± 2.6
Fresh heartwood air dried (40 °C)	7.2 ± 0.05	98.4 ± 0.01	614 ± 2.1	90.9 ± 1.3	93.9 ± 3.1
Fresh heartwood air dried (80 °C)	7.3 ± 0.03	98.9 ± 0.04	635 ± 6.3	96.0 ± 1.6	92.0 ± 1.0
Fresh heartwood steam dried (180 °C)	5.2 ± 0.51	99.0 ± 0.01	733 ± 6.7	99.2 ± 3.5	103.5 ± 3.0
Fresh sap25%: heart75% air dried (40 °C)	7.9 ± 0.09	96.9 ± 0.03	574 ± 0.9	89.8 ± 2.9	93.2 ± 1.2
Fresh sap50%: heart50% air dried (40 °C)	8.0 ± 0.01	96.4 ± 0.16	587 ± 1.0	90.7 ± 0.9	92.2 ± 1.1
Fresh sap75%: heart25% air dried (40 °C)	8.0 ± 0.37	96.4 ± 0.28	594 ± 3.4	91.9 ± 1.7	92.4 ± 0.6
Stored sapwood air dried (40 °C)	6.8 ± 0.13	98.0 ± 0.12	635 ± 2.2	97.7 ± 3.7	95.6 ± 2.6
Stored heartwood air dried (40 °C)	7.1 ± 0.08	95.4 ± 0.05	544 ± 5.9	85.7 ± 3.7	83.6 ± 2.0

pellets in the 5 l container was then measured and bulk density calculated. The durability test was done by tumbling 500 ± 10 g of sieved pellets in a tumbling box device at 50 ± 2 rpm for 500 rotations. The abraded and fine broken particles were then sieved through a 3.15-mm sieve. These procedures are according to the standard methods, specified and detailed in respective European (EN) standard methods of determination for solid biofuels, namely EN 14774–1:2009 for moisture content, EN 15103:2010 for bulk density, and EN 15210–1:2009 for mechanical durability [43]. Most of the pellets met the quality guidelines for non-industrial use pellets.

Statistical Analyses

A number of statistical analyses were performed to test the effects of feedstock content, and raw material drying temperature and storage on off-gassing of sapwood and heartwood pellets. To test the effects of drying temperature and raw material storage, one-way multivariate analyses of variance (MANOVAs) were conducted, while Spearman's correlation analysis was used to test the effect of feedstock content. Follow-up tests to the MANOVA were performed using one-way ANOVA, then following significant effects from the MANOVA and ANOVA analyses, the Tukey HSD post hoc tests were conducted to determine which means were significantly different. The tests were performed using R version 4.2.3 software, and a significance level of $\alpha = 0.05$ was set for all the tests.

Results

This section presents the results obtained from the extractives analysis of the sawdust raw materials, off-gassing tests of the wood pellets, and statistical tests.

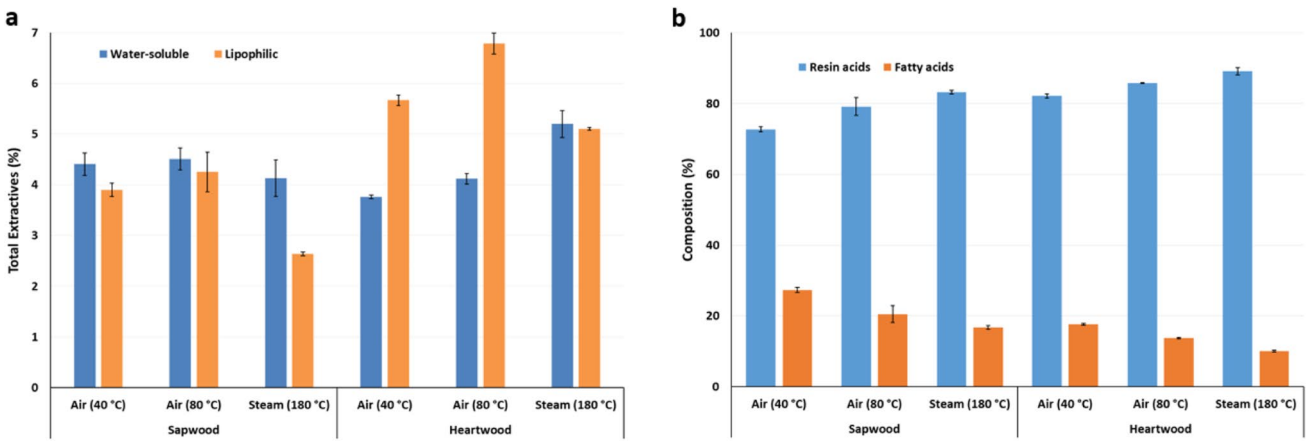

Fig. 2 Water-soluble and lipophilic extractives content of sapwood and heartwood sawdust dried at different temperatures and conditions (**a**), Percentage composition of fatty and resin acids (based on the total fatty and resin acids content for each material) in sapwood and heartwood sawdust dried at different temperatures and conditions (**b**). Error bars indicate the standard deviation of the mean

Table 3 Concentrations (mg/g) of fatty and resin acids in Scots pine sapwood and heartwood sawdust dried at different temperatures and conditions

IUPAC name	Common name	Air (40 °C)		Air (80 °C)		Steam (180 °C)	
		Sapwood	Heartwood	Sapwood	Heartwood	Sapwood	Heartwood
Nonanoic acid	Pelargonic acid	0.03 ± 0.00	n. d	0.04 ± 0.00	n. d	0.01 ± 0.00	n. d
Hexadecanoic acid	Palmitic acid	0.12 ± 0.02	0.08 ± 0.00	0.11 ± 0.00	0.08 ± 0.00	0.06 ± 0.01	0.06 ± 0.01
Heptadecanoic acid	Margaric acid	0.05 ± 0.01	0.03 ± 0.00	0.05 ± 0.00	0.03 ± 0.00	0.02 ± 0.00	0.02 ± 0.00
9-Octadecenoic acid	Oleic acid	3.68 ± 0.17	2.85 ± 0.09	2.94 ± 0.06	2.94 ± 0.10	1.43 ± 0.07	1.48 ± 0.14
9,12,15-Octadecatrienoic acid	Linolenic acid	0.23 ± 0.02	1.35 ± 0.01	0.14 ± 0.01	1.25 ± 0.04	0.04 ± 0.00	0.28 ± 0.03
9,12-Octadecadienoic acid	Linoleic acid	0.99 ± 0.04	4.29 ± 0.10	0.67 ± 0.02	4.04 ± 0.15	0.23 ± 0.02	1.07 ± 0.12
Pimaric acid	Resin acids	1.49 ± 0.02	4.54 ± 0.20	1.61 ± 0.30	6.09 ± 0.33	0.93 ± 0.06	3.20 ± 0.51
Pimaric acid, isomer		0.14 ± 0.01	0.64 ± 0.02	0.19 ± 0.04	0.86 ± 0.05	0.10 ± 0.01	0.47 ± 0.09
Isopimaric acid		0.82 ± 0.02	3.40 ± 0.19	0.99 ± 0.20	5.06 ± 0.23	0.55 ± 0.03	1.98 ± 0.29
Abietic acid		1.82 ± 0.12	7.28 ± 0.41	1.73 ± 0.83	7.60 ± 0.82	0.67 ± 0.02	1.89 ± 0.19
Dehydroabietic acid		5.86 ± 0.55	8.02 ± 0.24	5.68 ± 0.87	10.26 ± 0.33	4.09 ± 0.32	11.71 ± 1.42
Abietic acid, isomer		3.28 ± 0.20	16.25 ± 1.52	4.93 ± 0.54	21.98 ± 2.00	2.42 ± 0.25	6.22 ± 0.64
7-Oxo-dehydroabietic acid		0.21 ± 0.02	0.04 ± 0.00	0.30 ± 0.01	0.07 ± 0.01	0.19 ± 0.01	0.38 ± 0.13

n. d not detected

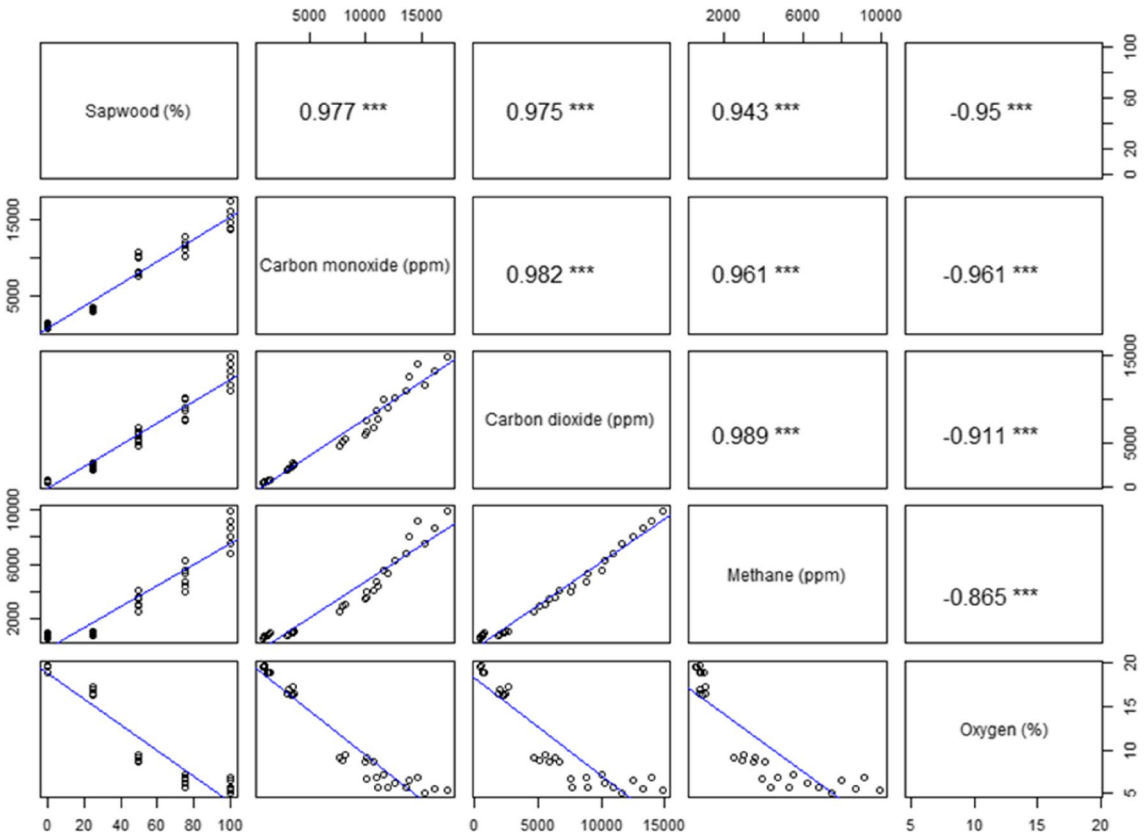

Fig. 3 Correlations between the percentage sapwood content of the feedstock, and concentrations of off-gases CO, CO_2, and CH_4 and residual O_2. The scatterplots with regression lines are shown in the boxes of the lower panel, and the corresponding correlation coefficient (R) values in the boxes of the upper panel. The three stars following each value indicate significance at $p < 0.001$

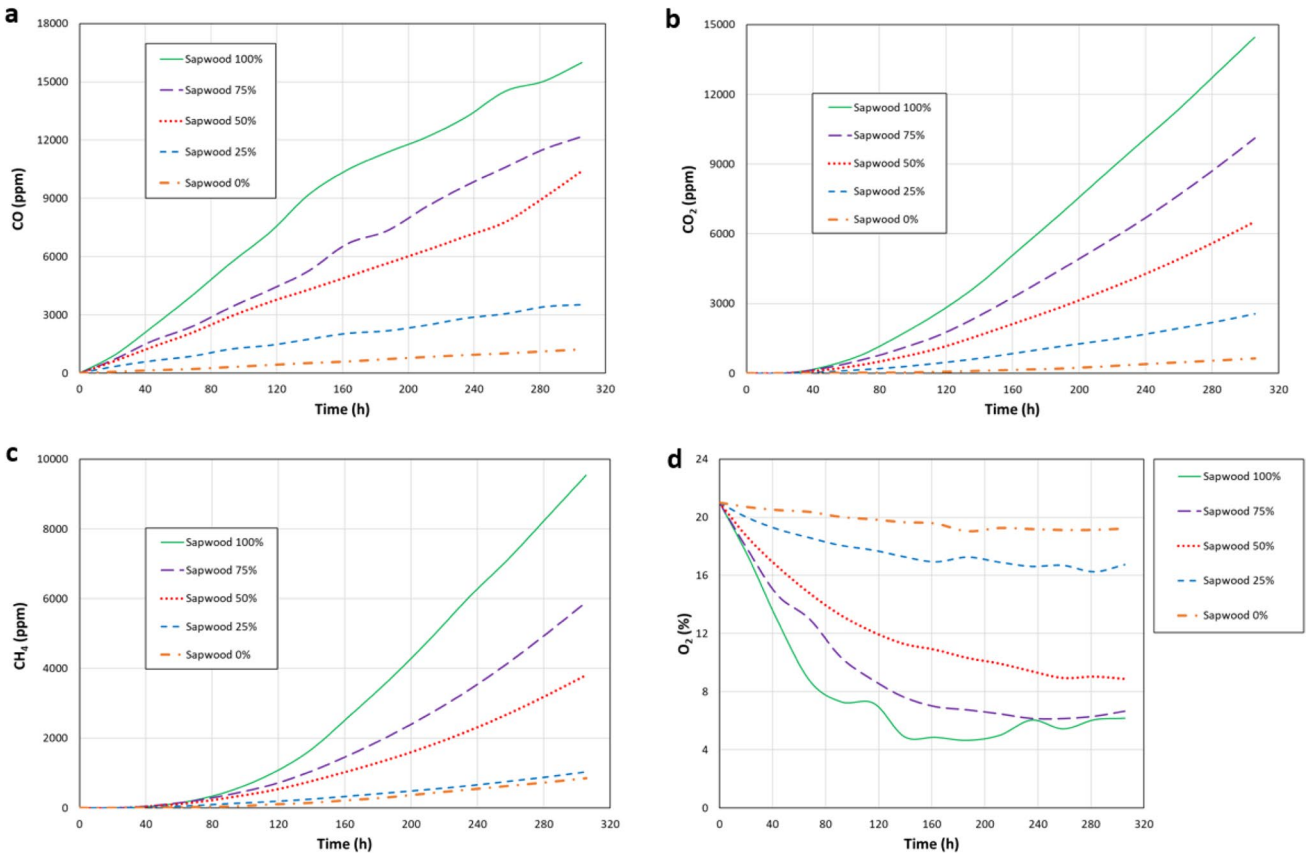

Fig. 4 Cumulative mean concentrations of the off-gases CO (**a**), CO_2 (**b**), and CH_4 (**c**), and residual O_2 (**d**) in the containers at each time point during the 13-day storage period for Scots pine wood pellets produced from sawdust with varying (0%, 25%, 50%, 75%, and 100%) mass percentage content of sapwood

Extractives

The total water-soluble and lipophilic extractives are presented in Fig. 2a. Thirteen fatty and resin acid compounds were detected and identified in sapwood, and twelve in heartwood. These are listed in Table 3, while Fig. 2b shows the total percentage of fatty and resin acids for each material. Generally, both sapwood and heartwood had higher concentrations of resin acids than fatty acids.

Effect of Feedstock Content on Off-Gassing

The relationship between off-gassing of wood pellet and the proportion of sapwood in the feedstock was investigated. Figure 3 shows the Spearman's correlation matrix of the relationships between the percentage content of sapwood and the concentrations of all the gases that were measured. The mean concentrations of the off-gases, CO, CO_2, and CH_4, increased with an increase in the percentage content of sapwood (Fig. 4a–c), while the level of residual oxygen decreased (Fig. 4d).

Effect of Drying Temperature on Off-Gassing

The results of the mean gas concentrations of sapwood and heartwood pellets produced from sawdust dried at different temperatures and conditions are presented in Fig. 5a–d. The one-way MANOVAs conducted to test the effect of drying temperature on off-gassing showed significant effects for both sapwood ($F_{(8, 26)} = 51.32$, $p < 0.05$) and heartwood ($F_{(8, 26)} = 334.1$, $p < 0.05$) pellets. To determine the effect sizes, univariate ANOVAs were carried out, and the results indicated significant effects, with eta-squared values falling between 0.61 and 0.98 for all gases in both sapwood and heartwood pellets. The Tukey HSD test was used in post hoc analyses to compare the differences in the mean off-gas concentrations for pairs of drying temperatures within each material. Significant differences were found between all pairs ($p < 0.05$), except for CO in sapwood ($p = 0.099$) between air (40 °C) and air (80 °C), CO_2 in sapwood ($p = 0.305$) between air (40 °C) and air (80 °C), and CH_4 in heartwood ($p = 0.636$) between air (80 °C) and steam (180 °C).

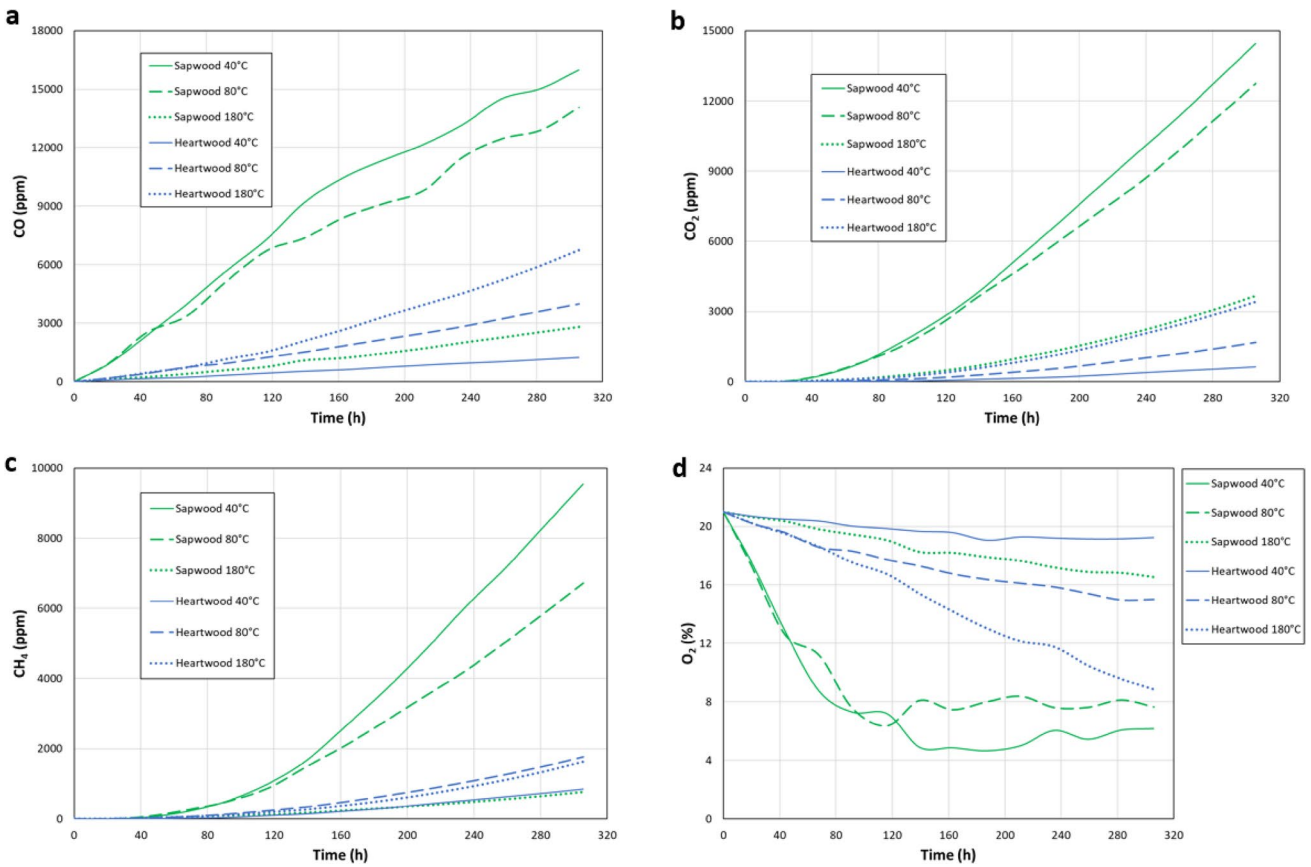

Fig. 5 Cumulative mean concentrations of the off-gases CO (**a**), CO_2 (**b**), and CH_4 (**c**), and residual O_2 (**d**) in the containers at each time point during the 13-day storage period for Scots pine wood pellets produced from sapwood and heartwood sawdust dried at different temperatures and conditions

Effect of Raw Material Storage on Off-Gassing

The results of the mean gas concentrations of the pellets produced from fresh and stored sapwood and heartwood sawdust are presented in Fig. 6a–d. A one-way MANOVA test was conducted to test the differences in off-gassing amongst fresh sapwood, fresh heartwood, stored sapwood, and stored heartwood pellets, and the results showed a significant difference, $F_{(12, 57)} = 22.97$, $p < 0.05$. The follow-up univariate one-way ANOVA results also indicated significant differences ($p < 0.05$) for all gases. The results of the Tukey HSD post hoc analysis showed that the mean concentrations of the off-gases, CO, CO_2, and CH_4, and residual O_2 were significantly different between fresh sapwood and fresh heartwood, as well as between fresh sapwood and stored sapwood pellets ($p < 0.05$). However, the differences were not significant between fresh heartwood and stored heartwood pellets ($p > 0.05$) for all the gases.

Discussion

The sawdust raw materials had varying extractive contents. Water-soluble extractives were relatively constant, around 4% in all sapwood samples, and increased with higher drying temperature from 4 to 5% in heartwood samples. Sapwood had higher content of water-soluble extractives than lipophilic extractives, while heartwood contained more lipophilic extractives than the water-soluble ones. The lipophilic extractives in sapwood and heartwood were found to be in the range of 2.6 – 4.2% and 5.1 – 6.8%, respectively (Fig. 2a). The higher content of lipophilic extractives in heartwood compared to sapwood was also observed by Willför et al. [26]. Although heartwood had higher concentrations of both fatty and resin acids (Table 3), based on the total amount of fatty and resin acids in each material, sapwood had a higher percentage compositions of fatty acids and lower percentage compositions of resin acids compared to heartwood (Fig. 2b). The amounts of fatty acids decreased

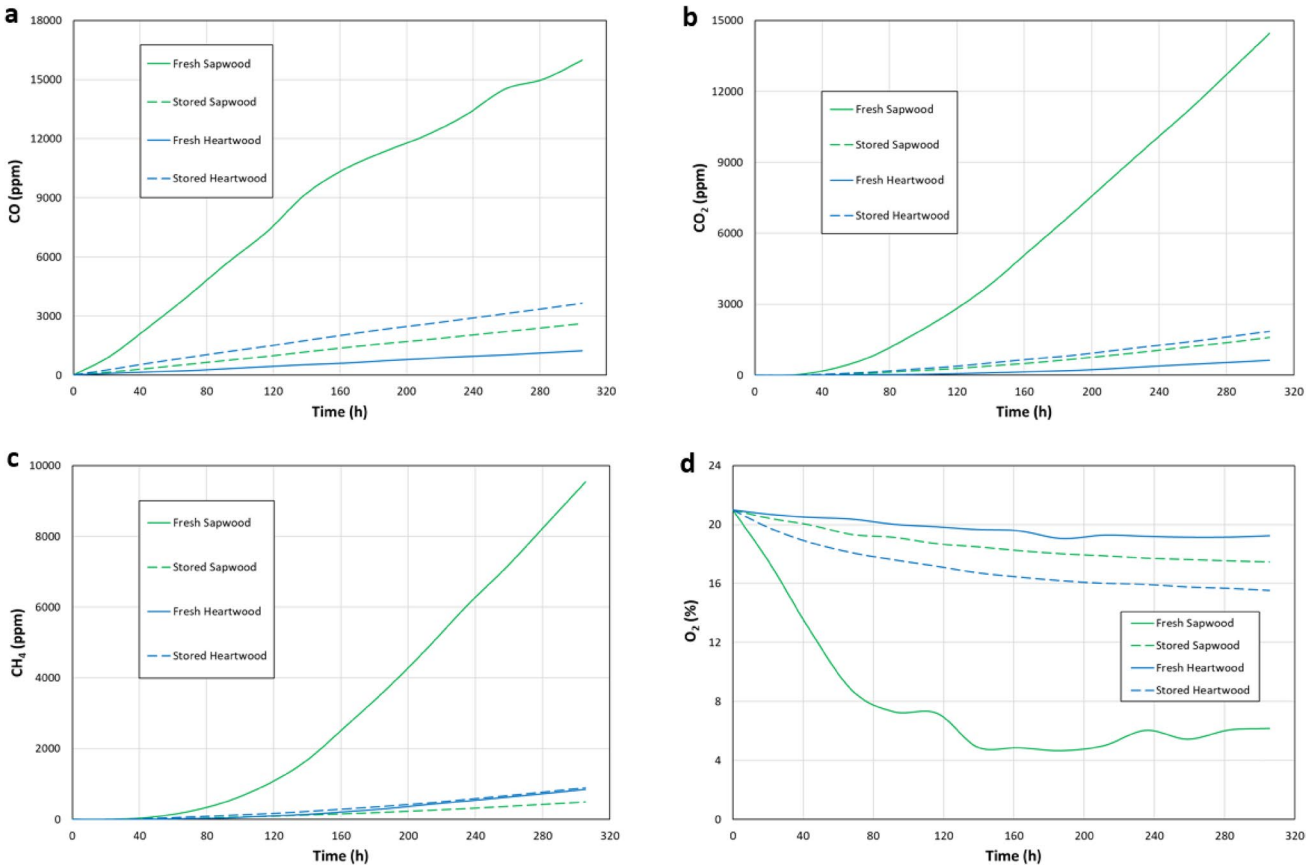

Fig. 6 Cumulative mean concentrations of the off-gases CO (**a**), CO_2 (**b**), and CH_4 (**c**), and residual O_2 (**d**) in the containers at each time point during the 13-day storage period for Scots pine wood pellets produced from fresh sapwood, fresh heartwood, stored sapwood, and stored heartwood sawdust

with an increase in drying temperature while the resin acids increased, in both sapwood and heartwood.

The off-gassing results in this work indicate that the gas emissions patterns were different for fresh sapwood and heartwood pellets. There was a strong linear correlation between off-gassing and sapwood composition, with correlation coefficient (R) values greater than 0.9 for all the three off-gases (Fig. 3). Specifically, increasing the sapwood content of the raw material increased off-gassing of the wood pellets (Fig. 4a–c). Fresh wood pellets produced from 100% sapwood sawdust produced the highest concentrations of CO, CO_2, and CH_4 under consumption of O_2, and the effects were diminishing with higher drying temperature. According to a number of different studies [18, 19, 23, 39], the oxidation of fatty acids is believed to be the primary cause of off-gassing in wood pellets. Therefore, the decrease in off-gassing of sapwood pellets due to increased drying temperature may be attributed to the reduction in the total fatty acids in the raw material (Fig. 2b). However, increasing the drying temperature for heartwood resulted in increased off-gassing of the wood pellets (Fig. 5a–c), even though the fatty acid content of the raw material had decreased. This indicates that other mechanisms than just the oxidation of fatty acids may contribute to off-gassing of wood pellets.

The emission of CO_2 and CH_4 and consumption of O_2 in sapwood pellets fits well with a biological explanation, where cells—either microbes, or maybe more probable surviving parenchyma cells—initially consume starch/fat (stored in the sapwood) by aerobic metabolism (producing CO_2) and thereafter anaerobically with methane fermentation [44]. This is apparently in contrast with earlier studies that suggested a connection with extractives and especially fatty acids, and an abiotic explanation for the gas formation. However, fats and fatty acids are excellent nutrition for living cells. That redox cycles of drying polyunsaturated fatty acids can produce gases is undisputed, but these gases are organic, and furthermore, despite containing lower amounts of both fatty and resin acids (Table 3), sapwood pellets produced much more gases than heartwood pellets. The formation of high concentrations CO is more puzzling; however, there are reports of biological processes forming CO in for instance cell signalling [45]. One possibility is that formation of this toxic gas is a stress reaction of the wood cells (with a purpose to harm

invading organisms). The formation of CO by stress has been reported for animals [46] and also for plants [47]. Another confusing observation is the reverted effect of increased drying temperature on off-gassing of heartwood pellets. There might be at least two different explanations to this, one is that another abiotic reaction is responsible for the gas formation, and that the high temperature pre-treatment in some way stimulates this—maybe by redistributing some extractives. Another explanation is that the high temperature in the pre-treatment inactivates antimicrobial chemicals in the heartwood; such effects are reported for antibiotics [48]. High temperature pre-treatment of sapwood results in the sterilization of its microbes or parenchyma cells, leading to a reduction in off-gassing emissions. Further studies are needed for understanding the exact nature of the off-gassing phenomena and its connection to self-heating, but the connection to biological processes indicated in this work can hopefully lead to practical efforts that can be performed to minimize these problems.

The storage of sawdust before pellet production has generally proven to be an effective strategy for reducing self-heating and off-gassing of wood pellets [40]. With specific to off-gassing of Scots pine wood pellets, the concentrations of CO, CO_2, and CH_4 significantly reduced in pellets produced from sawdust which was stored for 6 months compared to the ones produced from fresh sawdust [22]. In the present study, raw material storage significantly reduced off-gassing in sapwood pellets. The final cumulative concentrations of CO, CO_2, and CH_4 respectively decreased from 15,988 ppm, 14,453 ppm, and 9544 ppm in pellets made from fresh sapwood sawdust to 2620 ppm, 1598 ppm, and 498 ppm in pellets made from stored sapwood sawdust (Fig. 6a–c). Although there seemed to be an increase in the cumulative off-gas concentrations in heartwood pellets made from stored sawdust compared to those produced from fresh sawdust, the difference was not statistically significant. Furthermore, there were no significant differences in the off-gas concentrations between the stored sapwood and fresh heartwood pellets. This indicates that the storage of raw material had reduced the off-gassing of sapwood pellets to the same level as that of heartwood pellets produced from fresh sawdust. The lack of statistical significance in the difference of off-gassing between heartwood pellets made from fresh and stored sawdust, and the significant difference observed for sapwood pellets, suggest that raw material storage may have varying effects depending on the composition of the biomass raw material. Further studies could explore these differences and investigate the optimal storage time for sawdust obtained from different parts or sections of a tree and wood species to achieve maximum reduction in off-gassing and storage time of sawdust raw material.

Conclusion

The results of this study provided strong evidence of a relationship between sapwood and heartwood content of the feedstock and off-gassing of Scots pine wood pellets. The study also confirmed with established theory that off-gassing of wood pellet is caused by the oxidation of fatty acids. However, the distinct difference in off-gassing between sapwood and heartwood pellets suggests that biological mechanisms also play a role. Raw material storage and drying temperature and conditions can play a crucial role in reducing the off-gassing of wood pellets, but their effectiveness largely depends on the composition of the feedstock.

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Data Availability Data will be made available on request.

Declarations

Consent for Publication This version of the paper has been reviewed and approved by all the authors, and they have given their consent for its submission to BioEnergy Research Journal.

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