



Utilization of black locust (*Robinia pseudoacacia*) sawdust as an alternative pelletization raw material

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Abstract

European pellet production will be a future challenge due to two effects: (1) the share of hardwood species in Europe will increase and (2) the pellet market will face raw material shortages. Therefore, we investigated the blending of conifer sawdust with black locust sawdust. Twenty-one physical and chemical pellet quality parameters were recorded, including combustion emissions. Our statistical evaluation showed a strong linear correlation ($p > 0.8$ or $p < -0.8$) of the share of black locust with nine quality parameters. Fifty-three percent of the overall variation in the data was explained by the major principal component, which included the share of black locust. The cause of the decreasing pellet quality with increasing share of black locust sawdust was attributed to the heat conductance in the dye, which was affected by the hydrophobicity and rigidity of the black locust saw dust. A share of 25% black locust in blends with conifer sawdust is proposed as the limit to meet the A2 standard criteria in the European DIN EN ISO 17255-2. This would allow a black locust sawdust consumption of app. 6 mio t per year in Europe, which is far above the estimated abundance and indicates a high potential for hardwood sawdust as an alternative feedstock for pellet production in general.

Keywords Combustion emissions · Waste biomass utilization · Feedstock analysis · Pellet production process · Alternative feedstock utilization

1 Introduction

The global wood pellet production increased continuously since 2012 with the USA as the largest producer [1]. In 2021, the European energy crisis induced by the war between Ukraine and Russia further increased the pellet production, especially in Germany [2]. Due to the predicted increase of the global energy demand, a long-term growth of wood pellet production is predicted [3].

This reality collides with the recent discussion of environmental policies in Germany and Europe that questions the sustainability of utilizing biomass as a fuel. In general, the impact of forests as a carbon sink has to be optimized by the utilization of wood. The utilization of wood for materials, e.g., in houses and furniture, should be favored over its energetic use due to the longer immobilization of CO₂ in these materials. The major source of wood for pellet production should be the waste of the wood processing industry [4].

However, the annual amount of harvested timber and the associated sawdust production is limited by the annual growth rate of forests, which is a fundamental concept of sustainable forest management since hundreds of years.

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Therefore, an increasing sawdust demand for pellet production can only be satisfied by upgrading sawdust that is not utilized for pellet production, yet, because it yields pellets with low quality.

Black locust (*Robinia pseudoacacia*) is a low-quality biomass that is not utilized for energetic use, yet. The black locust tree can be managed in a short rotation harvesting regime with 1–15 years rotation time and generates biomass yields up to $14 \text{ t} \times \text{ha}^{-1} \times \text{year}^{-1}$. With app. 2.5 million ha plantation area, black locust is the third most abundant hardwood plantation species worldwide [5]. In Hungary, 23% of the total forest area in 2011 [6] is covered by this tree species, while the USA, China, Japan, Germany, Slovakia, Italy, Greece, Bulgaria, and South Korea have large black locust plantations, as well.

The timber of black locust has an exceptional robustness against soft rot fungi and bacteria [7] and, therefore, is used for outdoor applications [5]. This robustness is caused by the high levels of antibacterial phenolic compounds [7, 8] in the hard wood. The flavonol robinetin and the flavanonol dihydrorobinetin are two frequently detected polyphenolic compounds present in the heartwood of black locust [7, 9–11]. The flavanonols fustin, fisetin, dihydromyricetin, and dihydroflavonol, the flavonones liquiritigenin, robtin, and butin, the chalcones butein and isoliquiritigenin, the stilbenol piceatannol, the phenylpropanoid hydroxycinnamic acid, the stilbenes resveratrol and piceatannol, and the polyphenol ellagic acid were also identified in the heartwood extract of black locust [9–11].

Due to these high phenolic compound levels that cause the exceptional rot resistance, there are several sawmills in Europe that are specialized in black locust stem processing. The residual sawdust is subject to waste management, because its energetic use is compromised by insufficient physical and chemical quality, e.g., of black locust pellets. These quality parameters are listed in the European wood pellet standard DIN EN ISO 17255-2 [12]. They should give an orientation for fuel characteristics that lead to low combustion emissions of fine dust, nitrogen oxides, sulfur oxides, and carbon monoxide. The limits of these combustion emissions are, e.g., fixed in the German emission law BImSchV (Bundesemissionsschutzverordnung) [13]. Most waste biomasses that are not utilized for pelletization, yet, e.g. leaves, bark, or hardwood sawdust, yield low-quality pellets according to DIN EN ISO 17255-2 [12] with combustion emissions above the allowed combustion emission limits [13].

Upgrading such unutilized waste biomasses, e.g., by water leaching [14–19] or low-temperature hydrothermal treatment (HTT) [20–22], requires expensive hardware and thermal energy. A promising approach is the mixing of low-quality biomass waste with conifer sawdust [23]. This reduces emission precursors like zinc, lead, chlorine, or potassium and,

thereby, can reduce the emissions below the respective federal emission limits, e.g., in the BImSchV [24].

Only few studies investigated the possibility for utilizing this waste stream for pellet [23, 25–27] or briquette [28] production. Kamperidou et al. calculated the heating values and ash contents of sawdust mixtures based on the values for the unmixed feedstocks [26]. The dye pressure and temperature during the pellet production process was modelled by Kocsis et al. [27]. Bucar et al. investigated unmixed black locust pellets and found that the bulk density was $680 \text{ kg} \times \text{m}^{-3}$, the mechanical durability was 80%, and the ash content was low with 0.29% [25]. Only Stachowicz and Stolarski investigated mixed pellets of black locust, pine, and birch and found that the mixtures did not meet the requirements of the A1 class in the standard DIN EN ISO 17255-2, except for the lower heating value [23].

This study investigates 21 chemical, physical, and combustion emission characteristics of mixed pellets produced with different shares of softwood and black locust sawdust on preindustrial scale. The results were evaluated according to the European standard DIN EN ISO 17255-2. This large data basis acquired for preindustrial scale pellet production allowed to calculate a maximum share of black locust sawdust for industrial pellet production that still meets the criteria of the European standard. Our study also firstly presents experimental data on combustion emissions from black locust pellets combusted in a pellet boiler used for one-family houses. Based on this large dataset, the cause of the low mixed pellet qualities is discussed in detail, addressing both the physical and chemical processes in the press channel and the thermal oxidation processes during combustion. This discussion leads to potential measures to improve hardwood sawdust pelletization in order to meet the long-term increasing pellet demand under increasing shares of deciduous forests in Europe in the future.

2 Material/methods

2.1 Raw material

Black locust saw dust was acquired from a local sawmill (Haisch GmbH & Co. KG, Neuweiler, Germany). The sawdust was a product of the stem processing with a wood milling head. The milling was applied in order to reduce sapwood for utilizing the rot-resistant heartwood for garden construction purposes. Conifer sawdust with residual bark (app. 80:15:5 *abies alba*:*picea abies*:*bark residue*) was acquired from the sawmill Vollmer Holz e.K. (Rottenburg, Germany). Both raw materials were dried to 12% water content and processed with a hammer mill (Münch SDHM 2, MÜNCH-Edelstahl GmbH, Hilden, Deutschland) equipped

with a 6-mm sieve. The resulting saw dusts were analyzed for particle size distribution and were pelletized.

Part of the raw materials were cut with a 1-mm sieve cutting mill (pulverisette 19, Fritsch GmbH, Markt Einer-sheim, Germany) and ground with 0.12-mm sieve laboratory grinder (ZM 200, Retsch Technology GmbH, Haan, Germany) for successive analysis.

2.2 Particle size distribution

The particle size distribution was analyzed with a vibrating screen (AS400, Retsch, Haan, Germany) according to DIN EN 15149-1:2010 and DIN EN 15149-2:2010.

2.3 Pelletization

Pellets were produced using a semi-industrial pellet mill (Münch RMR 250, Münch Edelstahl, Hilden, Germany) equipped with a rotating ring die and rollers. No pressurized steam was used. The ring die (RMR 250/250 wood, Münch Edelstahl, Hilden, Germany) had channels with 6 mm diameter and 30 mm length. The roller (RMR 250/250 wood, Münch Edelstahl, Hilden, Germany) was rippled on the surface and made from a 20mnCr5 alloy. The die was operated at 166 rpm and the material flow was adjusted according to the sawdust mixture (Table 1). Temperature and pressure in the ring die were not measured. The cutter was adjusted to 5 cm to control the pellet length. Four batches of pellets with different mixtures of conifer saw dust (CS) and black locust (BLS) were produced according to Table 1 and 1%_{DM} of wheat starch was added to each of the mixtures prior to pelletization.

2.4 Pellet density

The density of each produced pellet batch was determined by measuring length (edge to edge), diameter, and weight of 50 pellets from each batch with a caliper and a laboratory balance according to DIN EN ISO 17829:2015.

Table 1 The mixtures of conifer saw dust and black locust sawdust used in this study

No.	Batch	Share of conifer sawdust [%]	Share of black locust sawdust [%]
1	100 CS	100	0
2	75 CS / 25 BLS	75	25
3	50 CS / 50 BLS	50	50
4	25 CS / 75 BLS	25	75

2.5 Pellet diameter and length

The diameter and length of the pellets were measured according to ISO 17829:2015. App. Fifty pellets per batch were measured.

2.6 Pellet durability

The mechanical durability was determined according to DIN EN ISO 17831-1:2016 with a pellet durability tester (PDI 136, Serve Real Instruments, Shanghai, China). The water content of the pellets was tested three times before the analysis. Five hundred grams of the pellets was tumbled in a test chamber with 50 rpm for 10 min. The residual mass in the test chamber was calculated in % of the original mass.

2.7 Pellet bulk density

The average bulk density of the pellet batches was determined according to DIN EN ISO 17828:2015. The water content of the pellets was tested three times before the analysis.

2.8 Ash content

The ash content of the raw materials and the pellets was determined according to the standard DIN EN ISO 18122:2015 in an AAF 1100 muffle furnace (Carbolite Gero GmbH&Co.KG, Neuhausen, Germany).

2.9 Heating value

The higher heating value of the raw materials and the pellets was determined according to the standard DIN EN 14918 with a C6000 calorimeter (IKA-Werke GmbH & Co.KG, Stauffen, Germany). Prior to the experiment, 5 ml of water was added to the bomb for subsequent IC analysis. The lower heating value was calculated with the hydrogen content of the elemental analysis according to DIN 51900-1.

2.10 Elemental analysis (EA)

Twenty milligrams of the samples (raw materials and pellets) was analyzed with a varioMACRO cube elemental analyzer (elementar Analysesysteme GmbH, Langenselbold, Germany). For the analyses of the hydrochar samples, tungsten trioxide (WO₃) was added at a mixing ratio of 1:1. The molar amounts of C, N, and H were determined and the molar amount of oxygen was calculated as

the difference between the sum of ash, C, N, and H weights to the total sample weights to display the O/C and H/C ratio in the Van Krevelen diagram.

2.11 Ion chromatography (IC)

The residual solution of the calorimeter experiment was analyzed with an 833 Basic IC plus ion chromatograph (Methrom AG, Filderstadt, Germany) that was equipped with a Metroser A Supr 4 -250/4.0 column (Methrom GmbH&Co.KG, Filderstadt, Germany) to determine the chlorine content. Devolatilized $1.8 \text{ mmol} \times \text{l}^{-1} \text{ Na}_2\text{CO}_3$ / $1.7 \text{ mmol} \times \text{l}^{-1} \text{ NaHCO}_3$ solution was used as an eluent at a flow rate of $1 \text{ ml} \times \text{min}^{-1}$.

2.12 Induced coupled plasma–optical emission spectroscopy (ICP-OES)

A Spectro Blue-EOP-TI ICP-OES (Spectro Analytical Instruments GmbH, Kleve, Germany) was used to analyze trace elements in the saw dust and pellet samples according to EN ISO 1185. Four hundred milligrams of the samples was dissolved in 3 ml nitric acid and 9 ml hydrochloric acid using a microwave oven (Multiwave Go, Anton Paar GmbH, Graz, Austria). The samples were heated to 200°C with a ramp of $15.5^\circ\text{C} \times \text{min}^{-1}$ and a subsequent holding time of 30 min, cooled down to 180°C with a subsequent holding time of 5 min. The liquefied sample was sized to 50 ml with double distilled water (Carl Roth GmbH&Co.KG, Karlsruhe, Germany), filtrated, and analyzed. The injection flow rate was $20 \text{ ml} \times \text{min}^{-1}$ and the argon gas flow rate was $13 \text{ l} \times \text{min}^{-1}$. The elements Al, Ag, As, B, Ba, Be, Bi, Cd, Co, Cr, Cu, Fe, K, Li, Mg, Mn, Mo, Na, Ni, Pb, Sb, Se, Sr, Ti, Tl, V, Zn, Si, R, and S were quantified using element specific calibration curves.

2.13 Emission testing

The emission testing setup was built according to DIN EN 14785:2006. The water content of the pellets was determined with a quick test balance (MA 150, Sartorius AG, Göttingen, Germany) with five repetitions prior to each combustion experiment. The pellet boiler (Easyfire EF2 GS 12, KWB - Kraft und Wärme aus Biomasse GmbH, Mertingen, Germany) had 12 kW nominal heat output and was modified with an inlay of firebrick on the floor of the combustion chamber. This firebrick was installed to replace the layer of ash in order to facilitate the ignition process. The boiler was placed on a balance (CAISL 2, Sartorius AG, Göttingen, Germany) for online measuring of the consumed fuel. The flue gas was dragged with $12 \text{ Pa} \pm 2 \text{ Pa}$. K-type thermocouples were used to determine the temperature of the flue gas. The tests were performed

at nominal load and steady state conditions, while the flue gas was sampled non-isokinetic. Before each emission test, the boiler was run for at least one hour until the flue gas had a constant temperature. The temperature during testing was constant, with the allowed $\pm 2.5^\circ\text{C} \times 10 \text{ min}^{-1}$ deviation.

Carbon monoxide (CO) and nitrogen oxides (NO_x) were measured with nondispersive infrared spectroscopy (URAS 26, ABB Automation GmbH, Frankfurt a. M., Germany). Oxygen (O_2) was determined by a paramagnetic oxygen analyzer (MAGNOS 206, ABB Automation GmbH, Frankfurt a. M., Germany). Temperature, CO, NO_x , and O_2 values were measured every second.

The particulate matter measurements were based on DIN EN 13284-1:2017. A two-stage filter was used that had a sleeve filter filled with 2.4 to 2.5 g of glass wool (Assistent Glaswolle extrafein, Glaswarenfabrik Karl Hecht GmbH & Co. KG, Sondheim/Rhön, Germany) and a planar filter (Munktell MG 8 160 with $1.2 \mu\text{m}$ pore diameter, Ahlstrom-Munksjö, Helsinki, Finland). The filters were heated for 1 h to 200°C before the test and to 180°C after the test. Afterwards, they were cooled down in a desiccator that contained silica gel. For the weighting of the empty and loaded filters, an analytic balance (CPA 1245, Sartorius AG, Göttingen, Germany) was used. The tube containing the filters was heated to 180°C during testing. All three batches and the control were tested five times and each testing lasted 45 min.

2.14 Statistical analysis

Boxplots were used for the visualization of selected datasets, which comprised the mean, percentiles, maximum, and minimum values. The Pearson coefficient was calculated for all combinations of the 21 datasets to identify correlations between them. The results were visualized in a correlation plot and a PCA plot. Coefficients with values >0.8 and <-0.8 were defined as high correlation coefficient values. In addition, the sum of the absolute correlation values was calculated for each dataset to get a measure for the general impact intensity of the dataset on all other datasets. The software XACT (XactPro Version 8.05a, Sky Lab, Vélizy-Villacoublay, France), Excel (Microsoft® Excel® LTSC Professional Plus 2021, 64-Bit), and RStudio (Vs. 2022.07.2, R Foundation for Statistical Computing, Vienna, Austria) with the packages corrr, factoextra, and FactoMineR were used for these visualizations and calculations.

3 Results and discussion

3.1 Physical feedstock characteristics and their effect on pellet quality

3.1.1 Particle size distribution and hardwood fiber

The particle size distribution of the two feedstocks used in this study differed due to the higher share of fine particles in the conifer saw dust and the higher share of large particles (> 1 mm) in the black locust sawdust (Fig. 1).

The high share of fines explains the low durability of the 100% conifer sawdust pellets (Fig. 2).

An even particle size distribution is advantageous for pellet quality compared to a large fine dust fraction (<0.5 mm) [29–31]. The produced blends of conifer and black locust sawdust were affected by the high fine dust share of the conifer sawdust (Fig. 1), which had a negative effect on the pellet durability. Larger softwood particles are elastic, orient in the feedstock flow direction at the funnel shaped die entrance, entangle in the press channel, and yield durable pellets [32–34], but they were almost absent in this study (Fig. 1, CS, fractions >2.0 mm). Instead, the high rigidity of the larger hardwood particles [35, 36] (Fig. 1, BLS, >2.0 mm) led to a low flow orientation and a low entanglement during pressing, which caused higher friction in the die [35, 37]. Black locust has an average density comparable to oakwood ($0.74 \text{ g} \times \text{cm}^3$), which decreases the orientation and deformation in the press channel [38]. This reduces the contact area between the hardwood fibers in the press channel, which leads to two effects: (1) the hollow spaces between fibers increase, which causes reduced heat transport and, therefore, reduced lignin melting, and (2) the lignin that is melting has a lower contact area for interlinking the particles. Both effects cause a lower durability of the pellet. Kaliyan and Morey stated that rigid fibers can act as predetermined breaking points in pellets

Fig. 1 The particle size distribution of the conifer (CS) and black locust (BLS) sawdust after drying to 12% water content and processing with a hammer mill (sieve size 6 mm)

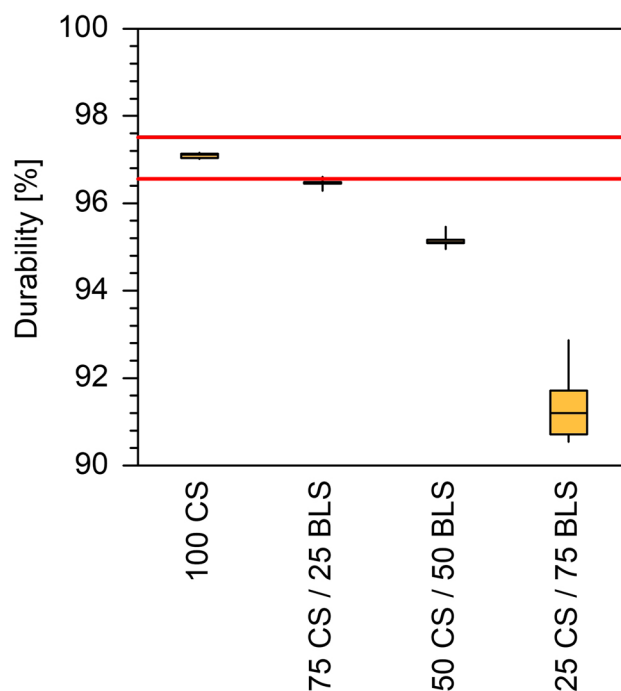
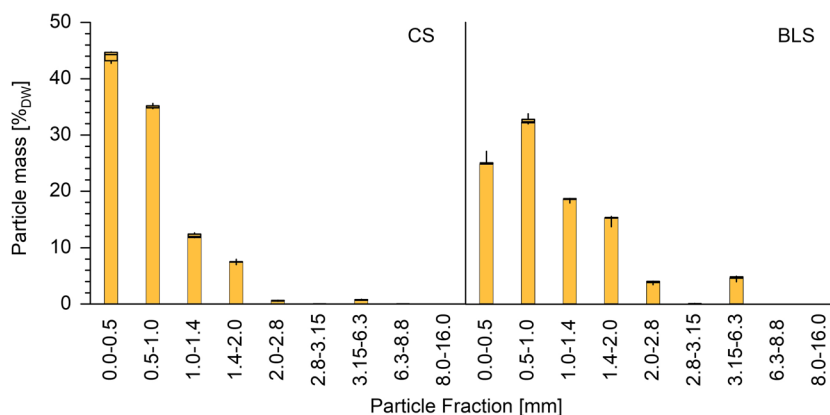


Fig. 2 The durability of the four pellet batches. The upper red line shows the quality class limit for A1 and A2 pellets (97.5%) in the European pellet standard DIN EN ISO 17255-2 and the lower red line shows the quality class limit for B pellets (96.5%)

[39]. The durability of the pellets in this study could have been increased by using larger sieve hole diameters in the hammer mill for the conifer sawdust to reduce the fine dust and smaller sieve hole diameters for the black locust sawdust to reduce the rigid hardwood fibers.

The share of the conifer and black locust sawdust feedstock (“batch” parameter, Fig. 3) had a high effect ($r > 0.8$ or $r < -0.8$) on nine of the 20 variables.

The batch parameter also had the highest sum of the absolute correlation values (Table S1) and correlated with physical, chemical and emission parameters.

Fig. 3 The Pearson correlation matrix with all physical, chemical, and emission variables quantified in this study. Concise values are given in Table S1 in the appendix. For all “batch” correlated variables, a positive *p*-value indicates an increasing variable value with increasing share of conifer sawdust and a negative *p*-value indicates a decreasing variable value with increasing share of conifer sawdust

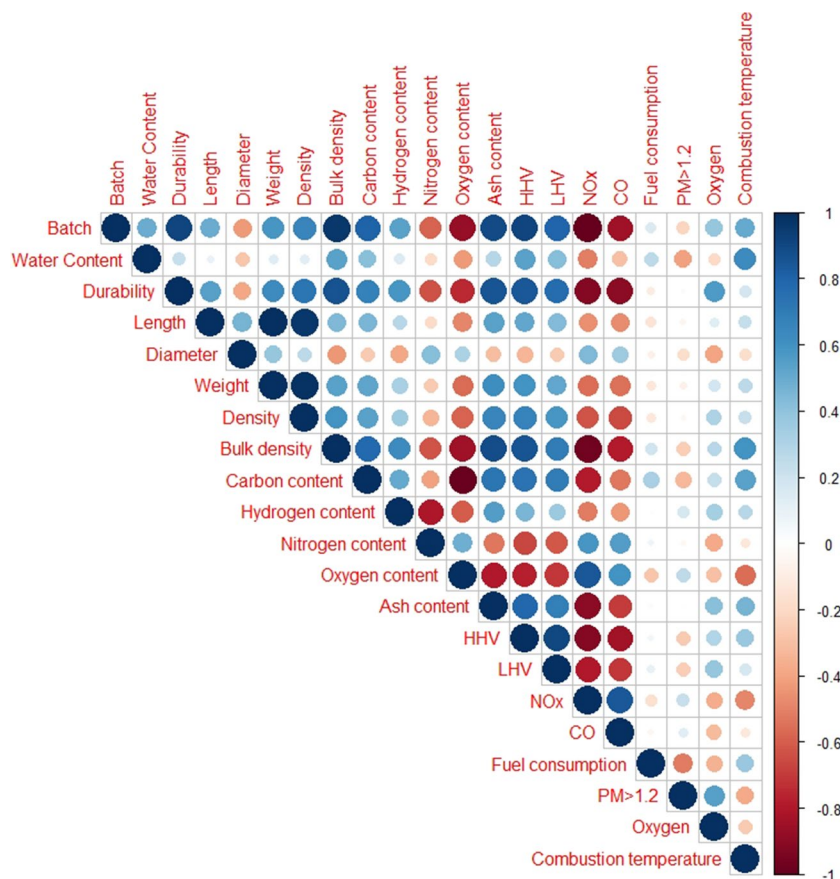


Table 2 Water and ash content as well as the heating values and the corresponding elemental compositions of the raw sawdusts before conditioning (CS: conifer sawdust; BLS: black locust sawdust) and the four mixed pellet types produced with different shares of conifer sawdust (CS) and black locust sawdust (BLS)

Batch	Water content [% _{DM}]	Ash content [% _{DM}]	Higher heating value [J×g ⁻¹]	Lower heating value [J×g ⁻¹]	C [% _{DM}]	H [% _{DM}]	O [% _{DM}]
CS	47.94 ±0.21 (0.44)	0.70 ±0.04 (5.06)	18,269 ±26 (0.14)	18,608 ±59 (0.32)	50.26 ±0.22 (0.00)	6.57 ±0.05 (0.01)	42.32 ±0.25 (0.01)
BLS	36.48 ±0.38 (1.05)	0.40 ±0.03 (7.24)	19,535 ±26 (0.13)	18,269 ±26 (0.14)	49.24 ±0.15 (0.00)	6.14 ±0.04 (0.01)	44.30 ±0.14 (0.00)
100 CS	9.22 ±0.67 (7.27)	0.82 ±0.03 (3.26)	20,225 ±82 (0.41)	18,858 ±99 (0.52)	51.92 ±0.26 (0.01)	6.46 ±0.04 (0.01)	40.62 ±0.26 (0.01)
75 CS / 25 BLS	8.39 ±0.09 (1.09)	0.67 ±0.04 (5.96)	20,177 ±30 (0.15)	18,866 ±53 (0.28)	51.56 ±0.19 (0.00)	6.39 ±0.07 (0.01)	41.16 ±0.22 (0.01)
50 CS / 50 BLS	8.00 ±0.05 (0.66)	0.67 ±0.04 (5.39)	20,045 ±36 (0.18)	18,718 ±32 (0.17)	51.10 ±0.03 (0.00)	6.43 ±0.03 (0.01)	41.61 ±0.05 (0.00)
25 CS / 75 BLS	8.45 ±0.83 (9.85)	0.52 ±0.06 (11.97)	19,955 ±23 (0.12)	18,637 ±27 (0.15)	51.04 ±0.35 (0.01)	6.34 ±0.07 (0.01)	41.85 ±0.39 (0.01)

3.1.2 Feedstock water content

The water content of conifer sawdust feedstock before drying was 23% higher in comparison to black locust sawdust and the water content of the pellets decreased with increasing share of black locust sawdust (Table 2, $r=0.5$).

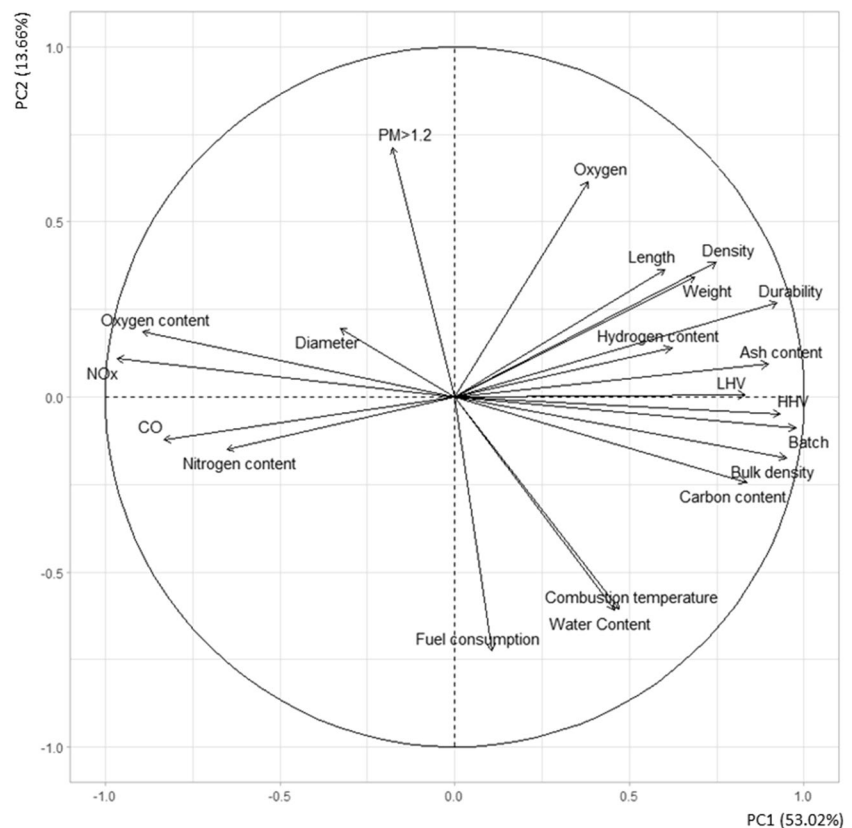
The higher hydrophobicity of the phenol rich black locust sawdust can save drying costs. The water content of the feedstock is very important for the heat transfer from the hot metal dye surface to the core of the forming pellet, which has a high impact on the pellet durability. At low water contents (app. $<10\%_{DM}$), the heat transport during the pressing process is too low to induce an even glass transition in the lignin. A high water content (app. $>14\%_{DM}$) causes high pressure in the dye due to steam formation. This leads to the Christmas tree effect, where the steam pressure is released directly at the dye exit and disintegrates the pellets [38–40]. On the other hand, large fibers become more flexible at high water content and entangle with each other easier, which increases the durability of the pellets. The effect of the water content on the durability, length, and density of barley straw pellets was very high in a study of Serrano et al. [41]. A comparable effect was shown by blending moist large fibers with dry fine dust [42]. The water content correlated to the pellet

durability ($r=0.81$), diameter ($r=0.99$), length ($r=-0.93$), bulk density ($r=-0.96$), and heating value ($r=-1$). Water also affects the dynamic mechanical properties of wood and the softening behavior of lignin [43]. Kelley et al. found an increase of the $\tan\delta$ of spruce wood with increasing water content, which proved the plasticizing effect of water on wood. In the pellet press, water has an additional indirect effect on the softening behavior of lignin due to its app. ten times higher thermal conductivity ($\lambda=0.56 \text{ W}\times(\text{m}\times\text{k})^{-1}$) in comparison to wood fibers ($\lambda=0.04\text{--}0.06 \text{ W}\times(\text{m}\times\text{k})^{-1}$) and air ($\lambda=0.02 \text{ W}\times(\text{m}\times\text{k})^{-1}$). This shows the negative effect of water-free spaces in the press channel induced by rigid hardwood fibers, which reduce the heat conductance to the core of the forming pellet.

In the presented study, the water content of the pellets had a low impact on the quality parameters of the pellets (Fig. 3) with r between -0.50 (NO_x emissions) to 0.63 (combustion temperature) (Table S1). There was a deviation from the linear water content decrease with increasing share of black locust for the 50% black locust share (Table 2), which caused the low Pearson correlations. It also caused a large distance to both axis of the PCA plot (Fig. 4).

The water content showed no high correlations and a low sum of correlation values (Table S1, sum of $|r|=6.59$)

Fig. 4 The principal component analysis (PCA), which grouped datasets with equal linear correlation functions close to each other and positioned datasets with different linear correlation functions in distance to each other. The length of the arrows indicates the internal sum of their squared deviations. The closeness to neighbor variables or the first or second principal component (x- or y-axis, respectively) indicates an equal slope



3.2 Chemical feedstock characteristics and their effect on pellet quality

3.2.1 Hardwood lignin

Lignin is a natural adhesive that enhances pellet durability. A lower lignin content in hardwood in comparison to softwood [38] decreases the durability of hardwood-blended pellets. Hardwood lignin contains both coniferyl alcohol monolignol units with one methoxy group and sinapyl alcohol monolignol units with two methoxy groups, while softwood lignin consists only of coniferyl alcohol monolignol units [44]. Therefore, hardwood lignin has more methoxy groups that form flexible β -O-4 bonds with propyl residues of other phenolic monomers [44]. The high β -O-4 bond flexibility contributes to the lower glass transition point in hardwood lignin [45]. This effect improves the durability of hardwood-blended pellets, but it could not compensate the low share of lignin in hardwood and the rigidity of hardwood fibers, which led to the low durability of the black locust blends in this study (Fig. 2). A higher pressure in the dye can improve the durability of hardwood pellets. It can be generated by the dye geometry and a combination of higher feedstock water content and higher dye temperature. This will decrease drying costs and increase the energy costs of the pellet press.

3.2.2 Ash content

The ash content of conifer sawdust (0.7%) was 43% higher in comparison to black locust sawdust (0.4%). This corresponds to the findings of Bucar Gornic et al., who found a low ash content (0.29%) in black locust, as well [25]. The pellets showed an ash content decrease with increasing share of black locust sawdust ($r=0.9$). The correlation to the batch variable (Table 2) caused non-explanatory high r values of NO_x content ($r=-0.90$), oxygen content ($r=-0.80$), density ($r=-0.88$), and durability ($r=0.86$) (Table S1). In contrast, the expected high correlation to the $\text{PM}_{>1.2}$ content in the flue gas was not found (Figs. 3 and 7, Table S1).

3.2.3 Elemental composition and heating value

The carbon content was 2.1% lower and the oxygen content was 4.5% higher in the black locust sawdust in comparison to the conifer sawdust. This was caused by the higher cellulose and hemicellulose content in hardwood species [38]. Cellulose and hemicellulose have a higher oxygen share and a lower carbon share in comparison to lignin [24]. The reduced oxygen content and the increased carbon content of all pellets in comparison to the feedstocks showed that the high dye temperature caused a mild pyrolysis (Table 2). The nitrogen content was neither correlated to the durability of the pellets nor to the heating value. The nitrogen content and the quantified trace elements did not exceed the limits in the standard DIN EN ISO 17225-2 (Table 3).

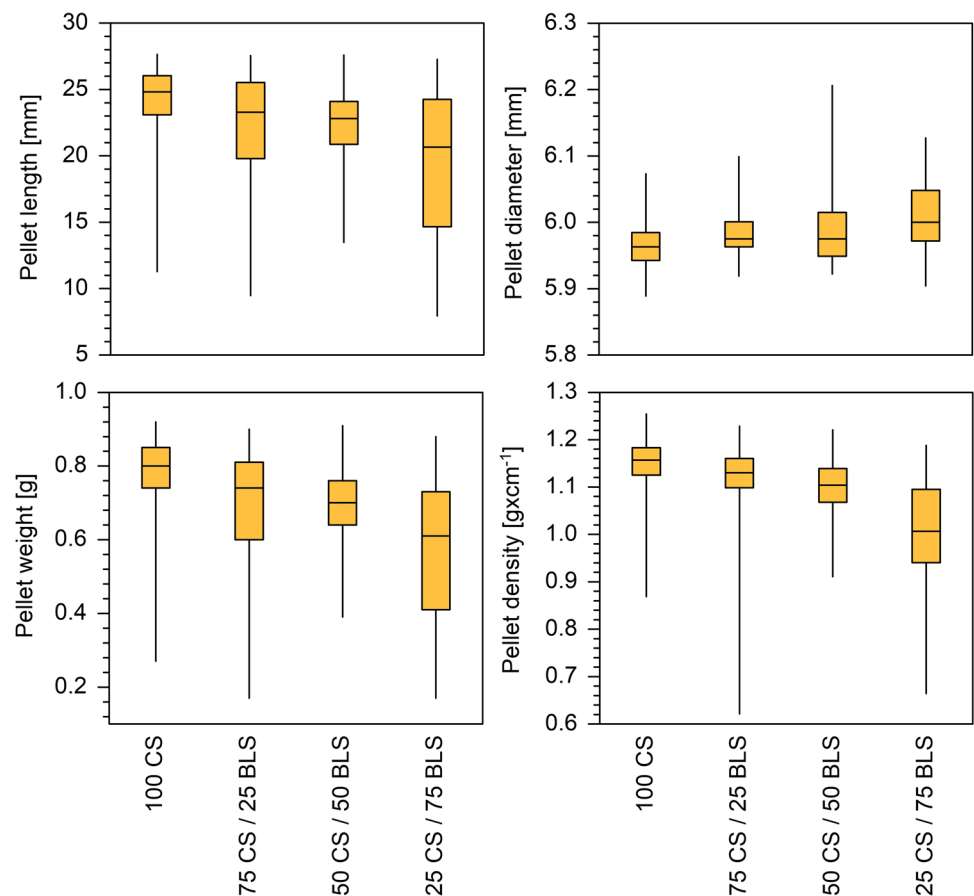
The higher heating value (HHV) had six high correlations (Table S1), which was the highest number among the chemical parameters. All six high correlations overlapped with the NO_x correlation, while both datasets were strongly negatively correlated ($r=-0.91$). This was caused by the endothermal nature of the NO_x formation (e.g., $\text{NO}=181$ kJ), while, e.g., the CO_2 formation is exothermic (-393 kJ). Therefore, the formation of NO_x in the calorimeter will consume heat energy, which will reduce the heating value of the fuel.

The PCA diagram showed that the HHV and the LHV contributed to the variance described by the first principal component PC1 (Fig. 4). The oxygen content and the ash content both had five high correlation values, while they were negatively correlated to each other. The correlations of the oxygen content overlapped with the batch and, except for the carbon content, also with the NO_x correlations, the durability and the bulk density (Fig. 3). Carbon content, hydrogen content, nitrogen content, and the lower heating value (LHV) had only two (batch and oxygen content), one (nitrogen content), one (hydrogen content), and one (HHV) high correlation values, respectively. While the HHV and the LHV positively correlated to the carbon content and negatively correlated to the oxygen content, they also positively

Table 3 The elemental composition of both raw materials ($N=5$) and the pellets ($N=1$) according to the DIN EN ISO 17225-2

Batch	N [% _{DM}]	S	Cl	As	Cd	Cr	Cu	Pb	Ni	Zn
	[mg×kg ⁻¹]									
CS	0.16 ±0.03 (0.18)	0.014 ±0.001 (0.07)	0.008 ±0.00 (0.04)	n.d.	n.d.	0.45 ±0.09 (0.19)	3.22 ±0.17 (0.05)	0.74 ±0.05 (0.07)	0.24 ±0.08 (0.33)	10.80 ±1.72 (0.16)
BLS	0.32 ±0.06 (0.20)	0.021 ±0.00 (0.01)	0.003 ±0.00 (0.10)	n.d.	n.d.	0.87 ±0.13 (0.15)	3.31 ±0.08 (0.02)	0.23 ±0.12 (0.50)	0.57 ±0.21 (0.38)	9.75 ±1.58 (0.16)
75 CS / 25 BLS	0.208	0.01	0.00	n.d.	n.d.	0.56	3.25	0.61	0.85	14.12
50 CS / 50 BLS	0.212	0.01	0.01	n.d.	n.d.	0.66	3.27	0.49	1.06	14.23
25 CS / 75 BLS	0.248	0.01	0.01	n.d.	n.d.	0.77	3.29	0.36	1.28	14.34

Fig. 5 Length, diameter, weight, and density of the four different pellet batches investigated in this study (CS = conifer sawdust; BLS = black locust sawdust). The limits for length and diameter for all three quality classes in the European pellet standard DIN EN ISO 17255-2 are $3.15 < L \leq 40$ mm and 6 ± 1 mm, respectively



correlated to the ash content (HHV: $r=0.79$; LHV: $r=0.69$, respectively).

3.3 Interdependency of physical pellet parameters

Pellet length, pellet weight, and pellet density were decreasing with increasing black locust sawdust share by 19.5% ($r=0.49$), 27.5% ($r=0.59$), and 13% ($r=0.66$), respectively, while the pellet diameter was increasing by 0.7% (Fig. 5).

The durability of the pellets decreased with increasing black locust share (Fig. 2, $r=0.91$) and only the conifer sawdust pellets and the pellets with 25% black locust share showed durability values acceptable according to the A2 quality of the pellet standard DIN EN ISO 79255-2 (Fig. 2).

The correlations between these physical pellet parameters resulted in two groups with high correlations, which were (1) share of black locust sawdust and durability ($r=0.91$ [35, 36]) and (2) length, weight, and pellet density. Group two correlated with $r=0.99$ (length to weight), $r=0.96$ (length to density), and $r=0.98$ (weight to density) (Table S1). The correlations between parameters of both groups were lower with values between $r=0.49$ and $r=0.73$.

Pellet length, weight, and density showed only two high correlations each, which were intercorrelations. In the PCA

diagram, the three variables were close to each other, but in distance to x - and y -axis. This showed that their contribution to both principal components was low. The pellet diameter did not show any high correlations to another parameter and was among the datasets with the lowest sums of the correlation values (Table S1, appendix). This dataset had the shortest arrow length and its angle to the x - and y -axis was close to 45° .

Durability and bulk density each had six high correlations (Table S1). They also had the third and fifth highest absolute sum of correlation values (Table S1). Both datasets shared most of their high correlations with chemical variables and the NO_x emission. This is confirmed in the PCA analysis (Fig. 4), as these variables were close to the x -axis.

3.3.1 Pellet durability

The decreasing durability was not caused by varying particle size distributions due to the increasing share of black locust sawdust (Fig. 1). A dilution of the large conifer fine fraction ($<0.5\text{mm}$) would have increased the pellet durability [29–31]. The durability could have been increased by adding 2% of starch instead of only 1% in this study. According to the limits in the DIN EN ISO 27255-2 [12]

for additives that are listed in the 1.BImSchV [13], a maximum of 2% is possible for pellets used in residential heaters. The decreasing durability in this study was caused by the rigid hardwood fibers that did not deform in the press channel and caused a larger air volume in the pellets that formed an insulation barrier. This resulted in a heat accumulation on the pellet surface, which is a major drawback in hardwood feedstock pelletization and causes a decrease of both pellet density and durability. In this study, a trend to lower durability (Fig. 2) with increasing diameter (Fig. 5) at increasing shares of black locust sawdust was visible. The low correlation between diameter and durability ($r=-0.39$) indicated that both variables were affected in a different way by the lack of heat transport to the pellet core. The diameter increase was caused by the accumulating heat on the press channel (surface effect) and the durability was affected by lignin melting in the whole pellet (volume effect).

3.3.2 Pellet length

The pellet length, with a linearly decreasing median from 23.9 mm (100% softwood) to 19.3 mm (75% black locust) (Fig. 5, $r=0.49$) was also within the range defined by the European pellet standard (3.15–40 mm). The Pearson correlation coefficient was low due to the high variation of the length values, which is common in pellet production [46]. This is reflected in the biplot of the PCA analysis with a large distance between batch and length dataset and the shorter arrow of the length variable (Fig. 4). The decreasing length was caused by the decreasing durability of the pellets (Fig. 2), as the cutter knife was always adjusted to 5 cm behind the dye exit. Therefore, a difference in length could only be caused by the impact of the pellets on the collector plate. The correlation of durability and length was $r=0.82$ in a study of Labbé et al. on softwood pellets [32], while in this study the Pearson correlation coefficient was $r=0.54$. The lower coefficient in this study can be explained with a high variation and a strong overlap of the pellet batch length values (Fig. 5). In the PCA plot (Fig. 4), the group of length, density, and weight was close to the durability data set. This showed that a higher density caused a higher durability, which caused less breaking on the collector plate and, therefore, yielded longer and heavier pellets.

3.3.3 Pellet density

The density of the pellets, which is not addressed in the European pellet standard, was decreasing with decreasing share of conifer sawdust ($r=0.66$), which further confirms that hardwood particles are less compacted in the press channel. Due to the reduced heat conduction in the forming pellet and the resulting reduced lignin melting, pellet density and durability correlated with $r=0.73$.

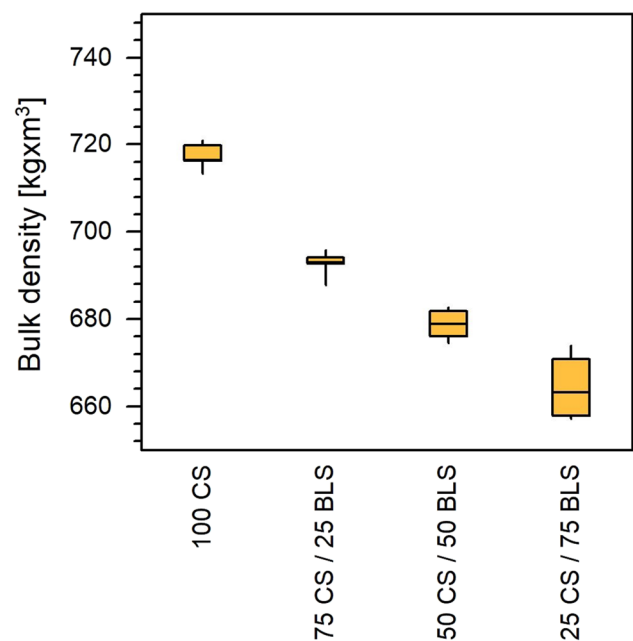


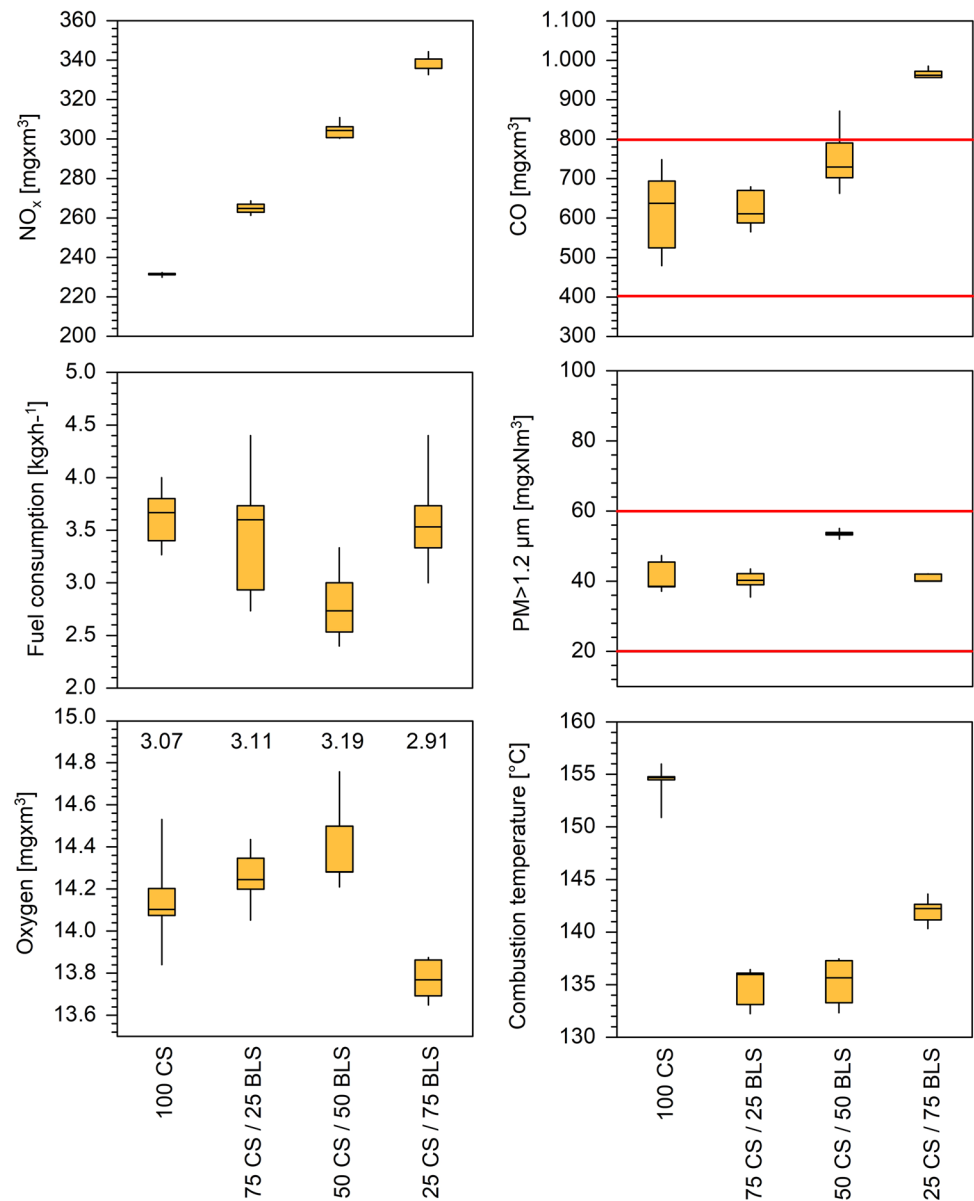
Fig. 6 The bulk density of the four pellet batches. The limit in the European pellet standard DIN EN ISO 79255-2 for all three quality classes is $\geq 600 \text{ kgxm}^3$

3.3.4 Pellet bulk density

The bulk density of all pellets varied within the standard limits in the DIN EN ISO 79255-2 (Fig. 6). The mean values linearly decreased by 7.5% from 716 kgxm^3 for conifer sawdust to 662 kgxm^3 for a 75% share of black locust ($r=0.97$), which resulted in a correlation to the durability of $r=0.86$.

This did not complement to a study of Mack et al. [47], where durability and bulk density correlated with $r=0.60$ for different commercial pellets. In our study, the durability varied between 97 and 90.5%, while the durability of the commercial pellets in the study of Mack et al. was above 98% for most investigated batches. The lower durability in this study had an impact on the pellet length ($r=0.54$) and, therefore, on the bulk density, while the length values in the study of Mack et al. were more homogeneous. In a study of Wöhler et al. [46], a high variation of the pellet length (three batches with a length range of 4–41 mm) affected the bulk density to a high extend ($632\text{--}711 \text{ kgxm}^3$) with decreasing bulk density at increasing pellet length. The report study published by Mack et al. on commercial pellets found a correlation between pellet length and bulk density of $r=0.54$ and a stronger correlation of bulk density and single pellet density ($r=0.66$) [47]. In our study, the correlation between pellet density and bulk density was $r=0.60$, which complements to the study of Mack et al. [47]. Apparently, the bulk density is affected by the pellet length, but only if the length

Fig. 7 The combustion parameters of the four pellet batches. The calculated lambda values were assigned to the oxygen values. The upper and lower red lines in the CO and PM > 1.2 µm figures show the emission limits according to the German 1.BImSchV, stage 1 (<500 kW, installation between 22.03.2010 and 30.12.2014) and stage 2 (<500 kW, installation since 31.12.2014), respectively



does highly vary. At low length variation it is more affected by the single pellet density.

3.4 Pellet combustion

3.4.1 Carbon monoxide and nitrogen oxides

The six parameters measured during the pellet combustion can be grouped in three categories. Nitrogen oxides (NO_x) and carbon monoxide (CO) emissions both showed a linear increase with increasing share of black locust sawdust (Fig. 7) with $r=-1$ and $r=-0.84$, respectively.

This corresponds to the linear decrease of the physical parameters durability and bulk density (Figs. 2 and 4), which correlated to the NO_x emissions ($r=-0.91$,

$r=-0.97$, respectively) and the CO emissions ($r=-0.89$, $r=-0.79$, respectively). CO, NO_x, durability, and bulk density were explained by the first principal component (Fig. 4). A decreasing pellet quality caused an increase in CO emissions, because the breaking of a pellet was correlated to its density. The breaking of pellets in a drop shaft stove, which was used in this study, yields fine particles. They are dragged by the airflow to low-temperature zones in the upper combustion chamber and the flue gas pipe. This causes the incomplete combustion of these particles, which increases the CO emissions. It also explains why the CO emissions do not correlate with the fuel consumption ($r=-0.05$), because the effect of the pellet durability superimposed the effect of the combusted wood quantity. A study of Wöhler et al. showed a correlation of pellet CO

emissions and pellet length. The CO emissions increased with increasing length in this study, because long pellets break with a higher probability than short pellets. In our study, the pellet length was varying between 25.8 (100% conifer sawdust) and 20.7 mm (75% black locust sawdust share). The low and inverse correlation of pellet length and CO emissions was caused by the lower length variation compared to the study of Wöhler et al. [46]. Therefore, longer pellets were more durable in this study ($r=0.54$), because pellets with low durability already broke on the collector plate of the press. CO emissions showed four high correlations, which all overlapped with the high correlations of batch, HHV, bulk density, and NO_x . The CO emissions also had the tenth highest absolute sum of correlation values and correlated negatively with the chemical and physical parameters (Fig. 3).

The emission parameter NO_x showed eight high correlations and also had the second highest sum of absolute correlation values (Table S1). This corresponded to the results of the PCA, where the NO_x dataset showed a contribution to the first principal component PC1. Like the CO parameter, the NO_x parameter was mostly negatively correlated to the physical and chemical parameters (Fig. 3). It correlated with the same parameters as the batch parameter, except for the carbon content ($r=-0.79$).

Nitrogen oxide emissions depend on the concentration of nitrogen in the fuel, with thermal and prompt NO_x formation as the only exception [24]. However, thermal and prompt NO_x form at combustion temperatures above 1300 °C, which is much higher than the combustion temperatures in this study (Fig. 7). The black locust sawdust contained twice the share of nitrogen in comparison to the conifer sawdust (Table 3). This nitrogen is acquired by nitrogen fixing microorganisms who live in a symbiosis with the black locust root system [5]. The nitrogen is bound in extractive compounds; therefore, extractive free black locust wood shows the same nitrogen levels as conifer wood ([48], Table 3). While robinetin and dihydro-robinetin are the most prominent extractives from black locust wood [8], there are several sources that report other non-nitrogen containing flavonoids. Most flavonoids are antibacterial, which explains the high resistance of the black locust wood in soil contact. Another source of nitrogen are extractable toxic proteins, e.g., robin or phasin, which are present in all black locust compartments [49]. These proteins can inhibit the protein synthesis of other organisms, which allows black locust to suppress competing plants [50]. In this study, the share of conifer sawdust and the nitrogen content were negatively correlated ($r=-0.58$), while the nitrogen content in the pellets and the NO_x emissions were positively correlated ($r=0.58$).

3.4.2 Fuel consumption and $\text{PM}_{>2.5}$

The second group of variables in Fig. 7 comprised the fuel consumption and the particulate matter emission, which correlated with $r=-0.52$. Both variables showed the same non-linear trend (Fig. 7). Figure 4 shows that both variables are explained by the second principal component and are oriented in opposite directions. This indicates that a partial load of the stove increased the particulate matter emissions. Particles can be formed by salt agglomerations around a heavy metal core in low-temperature zones above the fire bed. Heavy metals, e.g., zinc, mercury, or cadmium (Table 3), have a high vapor pressure ($\text{Zn}=0.401$ Pa; $\text{Cd}=12.6$ Pa; $\text{Hg}=50,800$ Pa; at 300 °C and atmospheric pressure). This leads to their vaporization in the fire bed and their condensation in the low-temperature flue gas zones [24]. They initiate the condensation of salts, e.g., KCl or K_2SO_4 , which form particulate matter (PM). Volatile organic compounds yielded by incomplete combustion of organic matter can condense on the salt particles, as well, or form organic particulate matter, e.g., tar or soot [24]. Both mechanisms require low temperatures above the fire bed, which are favored at partial load.

The black locust sawdust share did not correlate with the fuel consumption ($r=0.15$) or the particulate matter emission ($r=-0.22$) and neither did the ash content ($r=0.03$, $r=0.02$, respectively). Therefore, the $\text{PM}_{>2.5}$ emissions were only affected by the load of the stove, independent of the physical and chemical pellet property variation in this study.

3.4.3 Combustion temperature and oxygen concentration

The combustion temperature and the oxygen content in the flue gas constituted the third group of emission variables. Both were the only datasets that did not correlate to other emission datasets in this study (Fig. 4). All 100% conifer sawdust combustion temperature values were between 4.8 and 12.4% higher than the black locust share values (Fig. 7). The second highest temperatures were measured for the 75% black locust sawdust pellets. The oxygen content in the flue gas increased linearly from 100% conifer sawdust to 50% black locust sawdust share and then decreased to a lower value than the 100% conifer sawdust ($14.1 \text{ mg} \times \text{m}^3_{100\text{CS}}$; $13.8 \text{ mg} \times \text{m}^3_{25\text{CS}/75\text{BLS}}$; Fig. 7). The datasets combustion temperature and pellet water content were correlated ($r=0.63$), but did not show relations to other datasets or a contribution to PC1 and PC2 (Fig. 4). Water directly reduces the heating value and, thereby, the combustion temperature. The HHV was sampled on a water-free basis; therefore, the water content and the HHV did not correlate.

The thermal oxidation intensity is a function of the combustion temperature and the oxygen content in the flue gas, but both variables were not correlated to each other

($r=-0.25$). The high variation of the data (Fig. 7) and the lack of correlation to other datasets indicated that there are other factors than the ones investigated in this study that influence the combustion temperature and the oxygen content in the flue gas.

3.5 The Pearson correlation and PCA

The PCA (principal component analysis) showed one principal component that explained 53.02% of the variance of the datasets (Fig. 4). Two subsequent components explained 13.66% (Fig. 4) and 12.44% (not shown) of the variance of the datasets. The other components explained only < 5% (not shown) of the variance and were excluded in the data analysis, because they only explained the variance of one variable. As the principal component analysis is based on the Pearson correlation, the intercorrelation of all datasets (Table S1) is summarized in Fig. 4. The grouping of datasets in the diagram area of PC1 0.5 to 1.0 and PC2 -0.25 to 0.5 indicates that these eleven datasets were positively linearly dependent on each other. A second group with four datasets in the diagram section PC1 -0.5 to -1.0 and PC2 -0.25 to 0.25 also showed positive linear interdependencies. Both groups were described by the first principal component PC1 and the variables within the groups were negatively correlated to the variables in the respective other group (Figs. 3 and 7).

4 Conclusion

The effect of hardwood on pellet quality is high and it had negative effects on the pellet quality in this study. The potential advantage of a wider particle size distribution range of the blended sawdust batches was superimposed by the unfeasible physical and chemical properties of the black locust sawdust, e.g., fiber rigidity and lower lignin share. Physical, chemical, and emission properties of the blended pellets were decreased. Most variables were linearly correlated to increased shares of black locust and were explained by the first principal component of the PCA analysis. The most important variable was the share of black locust, which decreased durability, bulk density, carbon content, ash content, and the heating value, while it increased NO_x emissions, CO emission, and oxygen content.

The major challenge when pelletizing hardwood sawdust is its rigidity. Using a higher water content of the feedstock, grinding large hardwood fibers or using higher additive concentration can improve the pellet quality, but is limited due to the European pellet standard and emission regulations, e.g., the 1. BImSchV.

In our study, the maximum share of black locust sawdust that still would meet the requirements of class B for

residential pellets is 25%. Projecting this amount on the total production in Europe in 2022 (app. 24.3 mio t), a black locust sawdust mass of app. 6 mio t can be used. This indicates a high potential for hardwood sawdust blending in pellet production in general.

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Author contribution S.Pa.: study design, supervision of experimental section, data curation, draft writing; C.S.: study design, data curation; A.A.: data curation, draft writing; D.J.: supervision, funding; S.Pe.: study design, supervision of experimental section, funding.

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Data availability Data will be made available on request, please contact the corresponding author of this study.

Declarations

Ethical approval Not applicable.

Competing interests Not applicable.

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