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Evaluation of the Processing of Multi-Crop Plants into Pelletized Biofuel and Its Use for Energy Conversion

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Abstract: Multi-crop plants (fibrous hemp, maize, and faba bean) can potentially be an alternative to wood biomass pellets, but there is no detailed knowledge to support the suitability of this biomass for solid biofuel production. The aim of this study is to analyze and justify the suitability of multi-crop plant biomass for the production of biofuel pellets and to assess the environmental impact of burning them. This paper presents studies of physical-mechanical, thermal, and chemical characteristics of biofuel pellets from multi-crop plants and emissions during their combustion under laboratory conditions. The main parameters of the produced pellets were determined according to international standards, which are detailed in the methodology part. The length of the produced pellets ranged from 17.6 to 26.6 mm, and the diameter was about 6 mm. The density of wet pellets varied from 1077.67 to 1249.78 kg m^{-3}. The amount of ash in the pellets varied from 5.75% to 8.02%. Determined lower calorific value of all pellets was close to 17.1 MJ kg^{-1}. The lowest CO and C_xH_y emissions were determined when burning MIX2-1 pellets (biomass of the binary crop); their values were 572 and 29 ppm, respectively. The lowest content of CO_2 was determined when burning S-Mz pellets (mono crop biomass), and it was 3.5%. The lowest NO_x emissions were also determined when burning the pellets of this sample, with a value of 124 ppm. Research results show that multi-crop plants are a suitable raw material for the production of solid biofuel, the burning of which does not cause negative consequences for the environment.

Keywords: fibrous hemp; field bean; harmful emissions; intercropping; maize; pellets utilization; solid fuel

Citation: Petlickaitė, R.; Jasinskas, A.; Domeika, R.; Pedišius, N.; Lemanas, E.; Praspaliauskas, M.; Kukharets, S. Evaluation of the Processing of Multi-Crop Plants into Pelletized Biofuel and Its Use for Energy Conversion. *Processes* **2023**, *11*, 421. https://doi.org/10.3390/pr11020421

Academic Editor: David Herak

Received: 23 December 2022
Revised: 26 January 2023
Accepted: 28 January 2023
Published: 31 January 2023

1. Introduction

The global annual energy demand continues to grow every year; for example, as recently as 2018, about 70% of this energy demand was met by using fossil fuels. The consequence of this is an increase in CO_2 emissions of up to 33 Gt [1].

The increasing global demand for energy, rising energy prices, and concern for less negative environmental impact are leading to greater use of biomass beyond forests and agriculture. More and more attention is drawn to the possibility of using non-wood biomass for the production of biofuel pellets. With a large variety of granulation raw materials, the quality of such biofuels needs to be evaluated, as this is one of the main aspects of sustainable biofuel use [2].

When choosing biofuel raw materials, it is important to consider the aspect of sustainability because burning biofuel can generate greenhouse gas emissions (GHG). Priority should be given to raw materials that reduce GHG in the short term, such as wood residues and agricultural waste, thus contributing to the development of sustainable energy systems [3].

Using biofuels instead of fossil fuels can significantly reduce GHG and air pollution. However, the environmental impact of biofuel production varies greatly depending on biomass type, land resources, and management practices. With good management of energy crop ecosystems, the risk of loss of biodiversity can be reduced [4]. Emissions from burning biomass should be reduced as much as possible. This is particularly important in the case of nitrogen oxides and particulate matters, as these pollutants are harmful to both human health and the environment [5].

High humidity, low calorific value, hydrophilic nature, and difficulties in the storage and transportation of raw biomass are the main factors limiting its use [6]. By compacting biomass, it is possible to obtain high-quality solid biofuel that can replace coal. Such pellets can be used both in households and industrial boilers. The use of pellets is very wide: from burning in residential furnaces to industrial boilers. The quality of pellets is also very important to ensure their storage and transportation [7,8]. The quality of produced biofuel pellets is determined by such parameters as diameter and length, moisture, bulk density, ash content, calorific value, elemental composition, and mechanical durability [9].

When evaluating biomass fuel, the most important thing is its energetic value, which is expressed as net calorific value and determines the thermal power of the fuel-burning device, as well as gas emissions. Taking into account the most important properties of the pressed biofuel from herbaceous plants, such as elemental composition, and thermal and chemical properties, the possibilities of the final use of this biofuel in various fields are decided [10]. Although it is stated that, with the growing demand for wood pellets, it is necessary to look for alternative sources of biomass for the production of solid biofuel, there is still a lack of detailed scientific knowledge about other types of biomass that could become a sustainable raw material for the production of biomass pellets.

Interest in the use of multi-crop biomass for biofuels may be increasing for several reasons. First of all, the cultivation of multiple crops is one of the means of sustainable farming. Crop areas in which several different crops are grown at the same time may expand significantly in response to rising energy and fertilizer prices, as well as the need to reduce the amount of synthetic fertilizers and pesticides. Research data show that intercropping helps fight weeds when growing annual plants [11]. Intercropping is an example of a sustainable farming system, as it creates a balance with the environment and reduces damage caused by diseases and pests. Intercropping improves soil erosion control and makes use of plant-growing resources (such as sunlight, water, and nutrients) more efficiently [12]. Growing cereal and leguminous crops in the same field ensures yield stability and food security, thus contributing to sustainable or ecological global food production [13].

Secondly, the use of multi-plant biomass can potentially improve the quality characteristics of the produced pellets. The conducted studies reveal that the quality parameters of biomass pellets can be improved by using mixtures of different biomass feedstock. For example, Gutierrez-Antonio et al. [14] research data showed that the pellets made from rice husk biomass were of insufficient quality, but after mixing this biomass with bean straw biomass, the quality parameters of the produced pellets got better.

After preliminary investigations, it was determined that fiber hemp, maize, and faba bean are suitable plants for intercropping. Fiber hemp is a competitive energy crop that can be used for bioenergy production as an alternative to fossil fuels and has great potential to reduce GHG [15]. Fiber hemp is classified as one of the best plants that can be used for energy purposes, both technically and chemically [16]. In terms of its properties, hemp biofuel is close to wood fuel and is better compared to biofuel made from cereal straw, reed, or Miscanthus biomass [17].

Maize produces large amounts of biomass and is widely used for energy purposes. In the European Union, it is the second most popular crop after wheat, of which 85% is used for bioenergy production [18]. Faba bean has the ability to fix atmospheric nitrogen and produces large amounts of waste biomass after harvest [19]. Romaneckas et al. [20]'s study shows that growing fibrous hemp, maize, and faba bean in the same field may not only increase biomass yield but also stabilize gas concentrations and emissions from the soil and reduce the fraction of microstructures in the topsoil.

The aim of this work is to justify the suitability of multi-crop plants biomass, such as hemp, maize, and faba bean, for the production of pressed solid biofuel.

The novelty of this research work is based on the fact that the biomass of multi-crop plants (maize, fibrous hemp, and faba bean), which was grown in one plantation and not fertilized with any fertilizers, can be used for the production of high-quality solid biofuel by pressing chopped and milled plants into pellets with a diameter of 6 mm.

2. Materials and Methods

Studies were conducted with 7 types of pellets produced using the biomass of plants grown in monocultures and multi-crop. Three types of pellets are produced using plants grown in monocultures: fibrous hemp (*Cannabis sativa* L.), sort "Austa SK", maize (*Zea mays* L.), sort "Pioneer" and faba bean (*Viciafaba* L.), sort "Vertigo". Another four types of pellets were produced using plants of the same species and variety grown in multi-crop, i.e., in binary and trinomial crops (Table 1).

Table 1. Plant growing in monocultures and multi-crop.

Experimental Plots	Plants in Experimental Plots	Code of the Biomass
1	Maize (mono)	S-Mz
2	Fibrous hemp (mono)	S-FH
3	Faba bean (mono)	S-FB
4	Maize and fibrous hemp (binary)	MIX2-1
5	Maize and faba bean (binary)	MIX2-2
6	Fibrous hemp and faba bean (binary)	MIX2-3
7	Maize, fibrous hemp, and faba bean (trinomial)	MIX3-1

Biomass from a total of 7 fields was used for the research: field 1—maize (monocrop; in the article, the pellets made from the biomass of this field are called S-Mz), field 2—fiber hemp (mono crop, S-FH), field 3—faba bean (monocrop, S-FB), field 4—maize and fibrous hemp (binary crop, MIX2-1), field 5—maize and field bean (binary crop, MIX2-2), field 6—fibrous hemp and faba bean (binary crop, MIX2-3), and field 7—maize, field bean, and fiber hemp (trinomial crop, MIX3-1).

All plants are grown in the test fields of Vytautas Magnus University Agriculture Academy (54°52′ N, 23°49′ E). In the spring, before sowing the plants, the land was cultivated with a cultivator and fertilized with mineral fertilizers NPK 15:15:15 (300 kg ha^{-1}). The experiment of growing these plants is planned to be carried out for 3 years. The article presents research data on the main properties of biomass pellets produced from plants grown in the second year of the experiment.

Pellets for research were made from plants naturally dried to 12–16% humidity. The plants were naturally dried in a storage room with an ambient temperature of 20–25 °C and relative humidity (RH) of 30–50%. Sufficiently dry plants were ground by a Retsch SM 200 hummer mill (Germany) before being crushed by a drum shredder of a forage harvester MARAL-125 (Germany). A 7.5 kW granulator with a 6 mm matrix (ZLSP200B, Poland) was used for the production of pellets.

The diameter and length of the pellets were determined by measuring them with a calliper (accurate to 0.05 mm). Ten units of each type of pellet were used for the research.

Pellet density was calculated by dividing the mass of the pellet by its volume. The requirements of the ISO 18847:2016 standard were followed [21].

The moisture content of the pellets was determined according to the requirements of the standard LST EN ISO 18134-1:2016 [22]. A sample of pellets (about 300 g) is distributed on the weighed tray that, on a 1 cm^2 surface area, fits in 1 g of the sample. They dried (about 16 h) at 105 °C temperature to constant weight. Constancy in mass is defined as a change not exceeding 0.2% of the total loss in mass during a further period of heating over a period of 60 min.

The ash content of the pellets was determined according to the standard LST EN ISO 18122:2016 [23]. Samples of ground pellets (about 1 g.) were combusted in the air atmosphere for 60 min at 250 °C and after 120 min at 550 °C—to constant weight. Calculate ash content according to weight losses of sample before and after combustion.

Elemental analysis of the pellets was performed according to several standards. Analysis of sulfur and chlorine was carried out according to standard LST EN ISO 16994:2016 [24]. Samples were grounded to a homogenous powder (1 mm sieve) by using an IKA MF 10.2 cutting mill. Approximately 1 g of milled powder for each combustion was weighed into a compressed tablet with pressure of 10 t in square centimeter. Samples were put into a quartz combustion crucible and closed in calorimetric bomb. After that, the bomb was pressurized with oxygen to 35 atm. The bomb was immersed in a water bucket and ignited via an electrical discharge. After the ignition and cooling step, released gases were dissolved after passing through an Erlenmeyer flask filled with deionized water. All of the parts of the interior of the bomb were rinsed with deionized water, and all of the washings and solutions were collected in 50 mL volumetric flasks. All samples were analyzed by the Ion chromatography system Dionex ICS 5000.

The total content of carbon, hydrogen, and nitrogen was determined according to the requirements of standard LST EN ISO 16948:2015 [25]. Analysis of the elements C, H, and N in pellets was performed using a Flash2000 analyzer. Samples were combusted at 950 °C under inert conditions. After the combustion or pyrolysis process, gas mixtures CO_2, H_2O, and NO_2 go to the gas chromatography column in which they are separated.

During the elemental analysis of the pellets, the samples were mineralized according to ISO 16967:2015 [26] and LST EN ISO 16968:2015 [27]. Analysis of major elements was performed according to standard LST EN ISO 16967:2015 [26], and analysis of minor elements—according to standard LST EN ISO 16968:2015 [27]. A Paar Multiwave 3000 microwave was used for the mineralization process with a power of 800 W, 6 MPa pressure, and a pressure rate of 50 kPa S^{-1}. The analysis of the chemical elements Al, Ca, Cd, Cu, Fe, K, Mg, Na, P, Pb, Si, and Zn was performed using an ICP-OES device (Perkin Elmer). Tests were performed with three replicates. Analysis of chlorine and sulfur was carried out using a calorimetric bomb and an Erlenmeyer flask. An ion chromatography system, Dionex ICS 5000, was used for sample analysis.

The calorific value was determined according to the requirements of the standard LST EN ISO 18125:2017 [28]. An automatic bomb calorimeter IKA C6000 was used. In preparation for this study, the samples were ground to a homogeneous powder, and pellets of about 13 mm in diameter and 1 g weight were produced by pressing by a hydraulic press with a force of 10 t.

Ash melting behavior was analyzed according to the requirements of the standard ISO 21404:2022 [29]. To keep the particle size as small as possible, the resulting ash was crushed with a pestle. The ash was moistened with ethanol (purity > 95%) to yield a paste-like consistency. The resulting mass was compressed by hand into 5 mm high cylindrical samples with a diameter of 5 mm. The compressed samples were positioned vertically to dry (for approx. 24 h). The analysis was performed in a reducing atmosphere using a mixture of carbon monoxide and carbon dioxide at a volume ratio of 60% and 40%. First, the furnace temperature was raised to 550 °C. The temperature was then raised steadily at a rate of 2 °C/min, and photographs were taken. After the ash melted, the temperatures at which the phases of the samples changed were visually recorded according to the melting phase [30].

Harmful emissions during the burning of pellets in the special burning implement (Figure 1) were determined according to the requirements of the standards LST EN 14785:2006 [31] and LST CEN/TS 15883:2009 [32]. The pellets were burned in a 5 kW residential space heating appliance for 8–10 min. The heating appliance maintains the smoke temperature at 200 ± 5 °C, and the flue draught in the measuring section is maintained at 13 ± 1 Pa. CO, CO2, NOx, and OGC emissions were determined using three different analyzers: IR source analyzer Datatest 400 CEM for CO and CO2 determination, chemiluminescent analyzer Topaze 32M for NOx, and flame ionization analyzer VE7 for OGC.

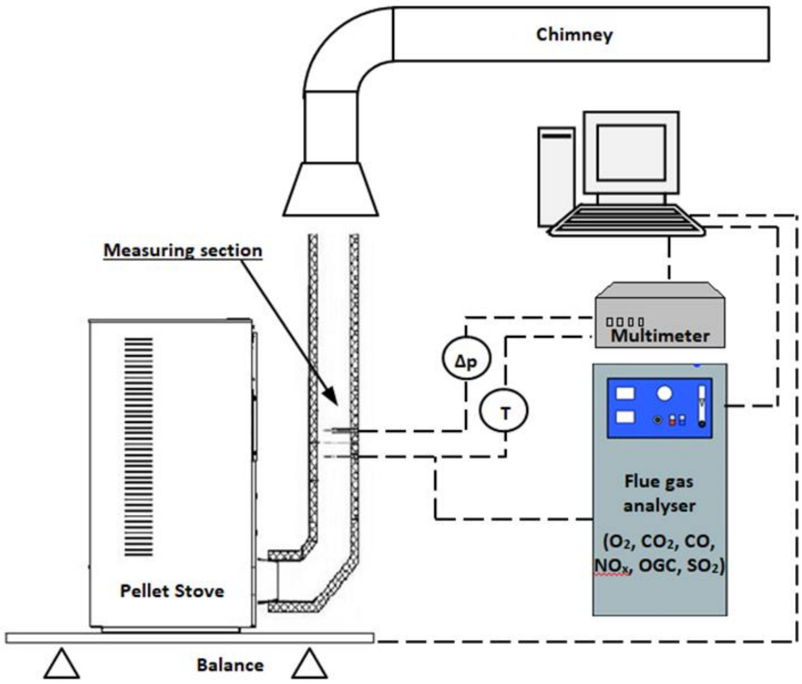


Figure 1. The schematic view of pellet combustion process.

In experimental studies analyzing and evaluating the investigated and determined properties of pressed biofuel, the research data were statistically evaluated by performing an analysis of variance, correlation, and regression. A STAT_ENG (vers. 1.55) program of SELEKCIJA software was used [33].

The mean values of the obtained results and their confidence intervals were calculated at the probability level P of 0.95.

3. Results and Discussion

3.1. Qualitative Parameters of Produced Pellets

All pellets from different biomass species were produced specifically for research purposes. All 7 pellet samples can be seen in Figure 2. The qualitative characteristics of the manufactured pellets can be seen in Table 1.

The diameter of the produced pellets was about 6 mm, and the length varied from 17.6 to 26.6 mm (Table 1). The density of produced pellets varied from 1077.67 kg m^{-3} (S-Mz pellets) to 1249.78 kg m^{-3} (S-FH pellets). Faba bean pellet density determined by Trejo-Zamudio et al. [34] was 1197.9 kg m^{-3} and the value of this parameter was very similar to that of our S-FB pellet sample (1210.04 kg m^{-3}). Maj et al. [35] found out that corn cob pellet density was 1140 kg m^{-3}, and corn cobs and corn husks mixture pellets density was 1150 kg m^{-3}. Tulumuru et al. [36] study showed that the density of 8 mm diameter pellets made of corn stover was about 1133 kgm^{-3}. Our sample (S-Mz corn biomass) density is lower than that obtained by the authors of the last two studies. However, for the samples MIX2-1, MIX2-2, and MIX3-1, where maize biomass is used in the mixtures, this parameter is very similar or even higher.

Figure 2. Samples of produced pellets.

Some researchers did not study the density of single pellets, but they determined the density of pellet bulk mass called bulk density. Jach-Nocoń et al. [37] studies of several types of biomass pellets showed that miscanthus pellet bulk density was 610 kg m^{-3}, sunflower husk—580 kg m^{-3} and corn stover pellet—570 kg m^{-3}. According to the research of these authors, wood biomass pellets had a higher bulk density (670 kg m^{-3}) compared to the investigated non-wood biomass pellets. The moisture content of the produced multi-crop pellets was also determined. The lowest moisture content was found in a sample of S-FB granules; it reached 5.52%. The moisture content of the sample of S-FH pellets was determined to be 16%, and the pellets of this sample were the only ones that slightly exceeded the requirements of standard ISO 17225-6:2021 [38] for non-wood biofuel according to moisture parameters.

After conducting an overview of the use of various biomass raw materials for the production of pellets, Ungureanu et al. [39] indicate that the moisture content of the pellets is strongly related to their physical properties. It is important to determine the optimal moisture content of the granules for the granules to have high stability and durability, as well as to select suitable storage conditions. The authors conclude that 5% moisture pellets are of low strength and produce a lot of dust during transportation. Pellets with more than 15% moisture will spoil during storage. According to these authors, the optimal moisture content of the pellets is 10–15%. However, Greinert et al. [40] indicate that the optimal pellet moisture content for the combustion process should be 6–8%. These authors studied the characteristics of straw and willow wood (4:1) pellets. The data provided by the above-mentioned authors do not allow us to draw unequivocal conclusions about the optimal moisture content of the granules because, in one case, it was found that it was higher, 10–15% humidity ensures the stability and durability of the granules, and otherwise, it is noted that the minimum is 6–8% humidity ensures optimal combustion conditions.

After evaluation of our research results, it was determined that the lowest ash content was found in a sample S-Mz and a sample of MIX3-1 pellets (5.75 and 5.98%, respectively), and the highest was found in a sample of S-FH biofuel pellets—8%. In comparison to

Nath et al. [41] research results, we can see that the ash content of wheat straw pellets is 7.09%, and this parameter was very similar to our MIX2-3 pellets (7.12%).

According to our determined ash content parameter, the pellets of all seven samples met the requirements of the ISO 17225-6:2021 standard. According to this standard, the pellets of S-Mz and MIX3-1 can be classified as class A pellets and the pellets of the remaining samples as class B pellets. Therefore, although not all samples can be assigned to a higher class, A, according to this parameter, all meet the standards of quality solid biofuel. On the other hand, it is necessary to further investigate the possibilities of further sustainable use of the ash produced by burning the pellets of this biofuel, e.g., for plant fertilization.

The lower calorific value (LCV) of dry biofuel of all produced pellets was quite similar and was close to 17 MJ kg^{-1}. According to this parameter, all produced pellets can be considered high energy efficiency; according to the ISO 17225-6:2021 standard, all samples can be classified as class A pellets. Pellets made from some other types of biomass have a similar LCV. Hrdlička et al. [42] found out that the LCV of grain straw pellets was 17.60 MJ kg^{-1}. Greinert et al. [40] found out that the calorific value of pellets made from straw and willow wood in a ratio of 4:1 was 17.3–20.1 MJ kg^{-1}. Therefore, it is similar to our pellets and in some variants even higher. Ozturk et al. [43] in their study report only the higher calorific value (HCV) of pellets made from corn stalks (with an average length of 17.28 mm and a diameter of 6.26 mm), which was 18.11 MJ kg^{-1}. Therefore, the lower calorific value of these pellets was probably lower and similar to the pellets produced by us.

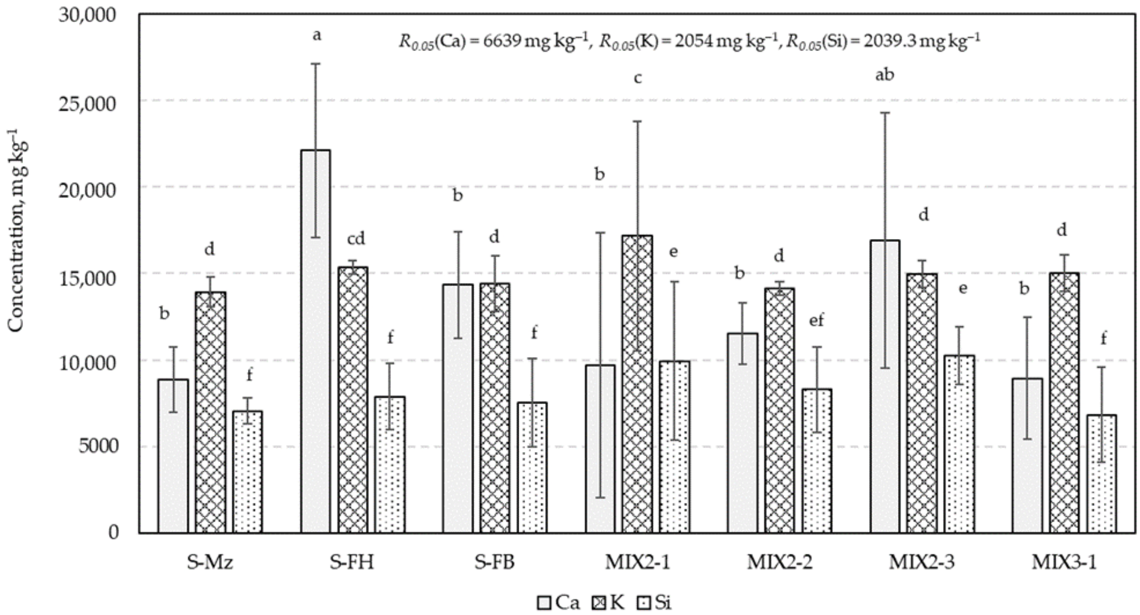
Rajput et al. [44] point out that the energy density of biomass is lower than that of fossil fuel, and in this respect, biofuel is less attractive to consumers. In these studies, some scientists are looking for ways to improve the properties of biomass fuel. For example, Whittaker and Shield indicate in their study that using additives such as fat or oil can increase the calorific value of fuel [45]. However, the authors also point out that the use of such additives has a negative aspect, as it can reduce the durability of the pellets. Therefore, the improvement of biofuel properties is likely to be an open question.

The requirements for non-wood biofuel are defined by the ISO 17225-6:2021 Solid biofuels—Fuel specifications and classes—Part 6: Graded non-woody pellets [38]. The standard defines the quality characteristics of non-wood biomass pellets and the permissible values of some chemical elements.

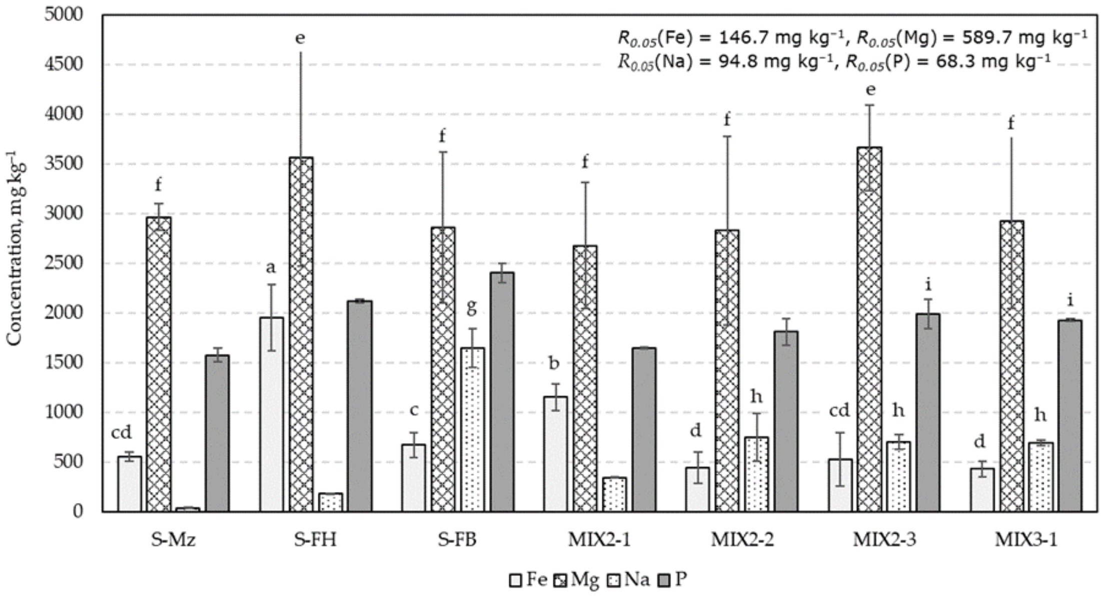
Only S-FB pellets N value (2.67%) exceeded the standard permissible N rate ($\leq$2.0%). All 7 types of pellets met the requirements of the standard for S content ($\leq$0.30%) and Cl content ($\leq$0.40%).

The chemical micro and macro elements of all 7 samples were analyzed, and the analysis results are presented in Figures 3 and 4 (graphs show mean value and standard deviation). During the elemental analysis of the pellets, it was determined that carbon, oxygen, and hydrogen elements accounted for 89.94–93.04%. The largest part of these elements was carbon, the amount of which varied from 45.18 to 46.63%. The amount of oxygen varied from 38.07 to 41.53%, and hydrogen content ranged from 4.88 to 5.68%.

For comparison with other biomass types, other information sources were analyzed. Barmina et al. [46] demonstrated that elements such as C, O, and H in wheat straw pellets were 47.40, 42.84, and 5.28%, respectively, and these elements accounted for 95.52% in total. Xu et al. [47] found that the C content in corn residue pellets was 38.72%, and in rice hull pellets—36.27%. The H content in these pellets was 5.56% and 5.08%, respectively. Kaczyński et al. [48] determined that the total amount of C, H, and O in sunflower pellets was 91.3%, and in spruce wood pellets—98.7%. These authors, having studied more types of wood and agrobiomass pellets, state that, regardless of the source of biomass, the composition of biomass in terms of carbon, hydrogen, and oxygen elements is very similar. The results of our analysis do not contradict this statement.



(**a**)



(**b**)

Figure 3. Results of macronutrient analysis of produced pellets: (**a**) Ca, K, and Si analysis; (**b**) Fe, Mg, Na, and P analysis. In the figures, any two samples with a common letter are not significantly different, as assessed using the least significant difference.

Although the elemental analysis of the pellets we presented is intended to support the compliance of the pellets with the non-wood biofuel standard, a more in-depth analysis is warranted in the future to determine how the elemental composition affects combustion processes, etc. In addition, Williams et al. [49] indicate that nutrients in biomass such as N, P, K, the main macronutrients Ca, Mg, Na, and Si, as well as the micronutrients Mn, Fe, Mo, Cu, and Zn and others, vary depending on the time of harvest and the growing conditions.

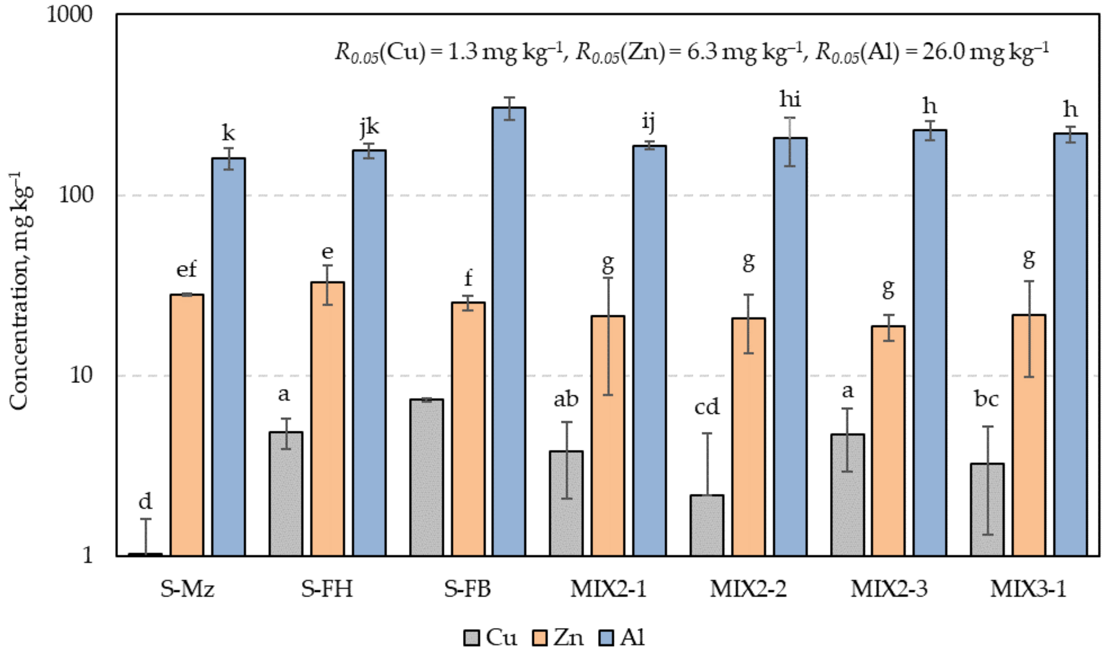


Figure 4. Results of Cu, Zn, and Al analysis. In the figure, any two samples with a common letter are not significantly different, as assessed using the least significant difference.

The content of Cu in the analyzed pellets varied from 1.03 to 7.31%; the content of Al in the analyzed pellets varied from 160.44 to 303.83%, and the content of Zn—from 18.66 to 32.67%. Cd and Pb in all 7 types of pellets were also determined. The cd value in all samples was less than 0.51%, and the Pb value was less than 1.20%. The results of the study show that the values of Cd, Cu, Pb, and Zn determined in the pellets do not exceed the limit values set for these elements in the ISO 17225-6 standard.

Standard ISO 17225-6:2021 does not specify requirements for ash melting temperatures; it only states that they must be specified. The ash melting temperatures of all sample pellets are given in Table 2.

Table 2. Main characteristics of produced multi-crop plant pellets.

Parameter	S-Mz	S-FH	S-FB	MIX2-1	MIX2-2	MIX2-3	MIX3-1
Length, mm	21.5 ± 3.15	23.4 ± 2.39	20.6 ± 1.69	17.6 ± 3.14	18.0 ± 5.30	23.3 ± 2.98	26.6 ± 1.42
Diameter, mm	6.3 ± 0.07	6.2 ± 0.04	6.0 ± 0.05	6.2 ± 0.06	6.0 ± 0.33	6.2 ± 0.06	6.1 ± 0.08
Density, kg m^{-3}	1077.67 ± 90.73	1249.78 ± 80.08	1210.04 ± 109.72	1164.78 ± 159.60	1160.21 ± 39.95	1211.54 ± 77.51	1238.20 ± 104.57
Humidity, %	12.18 ± 0.24	16.63 ± 0.03	5.52 ± 0.04	8.33 ± 0.03	6.66 ± 0.07	5.43 ± 0.06	6.28 ± 0.04
Ash content, %	5.75 ± 0.07	8.57 ± 0.14	8.02 ± 0.07	6.87 ± 0.10	6.03 ± 0.14	7.12 ± 0.18	5.98 ± 0.10
LCV, MJ kg^{-1}	16.99 ± 0.34	16.73 ± 1.11	16.72 ± 0.18	16.87 ± 0.58	16.95 ± 0.41	16.81 ± 0.04	16.79 ± 0.76
C, %	46.00 ± 0.14	46.55 ± 0.17	45.18 ± 0.34	46.63 ± 0.06	45.65 ± 0.15	45.73 ± 0.05	46.57 ± 0.06
O, %	41.53	38.51	38.07	39.25	40.82	39.34	39.90
H, %	5.51 ± 0.05	4.88 ± 0.01	5.67 ± 0.05	5.61 ± 0.05	5.65 ± 0.06	5.68 ± 0.04	5.68 ± 0.18
N, %	0.93 ± 0.01	1.17 ± 0.05	2.67 ± 0.25	1.34 ± 0.09	1.52 ± 0.02	1.93 ± 0.01	1.43 ± 0.12
S, %	0.08 ± 0.01	0.13 ± 0.01	0.12 ± 0.01	0.08 ± 0.01	0.10 ± 0.01	0.09 ± 0.01	0.10 ± 0.01
Cl, %	0.20 ± 0.01	0.19 ± 0.02	0.27 ± 0.03	0.22 ± 0.03	0.24 ± 0.01	0.11 ± 0.01	0.34 ± 0.04
SST, °C	948 ± 0.82	798 ± 1.42	810 ± 2.00	822 ± 0.34	923 ± 0.77	723 ± 0.20	1042 ± 0.14
DT, °C	1004 ± 0.28	1461 ± 0.29	>1550	1296 ± 0.65	1107 ± 0.13	1463 ± 0.01	1148 ± 0.74
HT, °C	1092 ± 0.19	1504 ± 0.38	>1550	1394 ± 0.20	1169 ± 0.36	1474 ± 0.26	1177 ± 0.36
FT, °C	1145 ± 0.62	>1550	>1550	1423 ± 0.10	1201 ± 0.12	1484 ± 0.19	1206 ± 0.70

Abbreviations: LCW—lower calorific value, SST—ash shrinkage starting temperature, DT—deformation temperature, HT—hemisphere temperature, FT—fusibility temperature.

3.2. Determination of Harmful Emissions

The usability of the pellets is also characterized by harmful emissions produced during burning. The determined harmful emissions during the burning of pellets of all 7 samples are presented in Table 3. The changes in CO and NOx concentrations during the burning of the pellets of the 7 samples are shown in Figures 5 and 6. The combustion process of sample of MIX3-1 pellets is presented in Figure 7.

Table 3. ISO 17225-6 requirements for non-woody pellets *.

Parameter	Class A	Class B
Length (L), mm and diameter, mm	$3.15 \leq L \leq 40$ (from D06 to D10), $3.15 \leq L \leq 50$ (from D12 to D25)	
Moisture, %	≤12	≤15
Ash content, %	≤6	≤10
Net calorific value (Q), MJ kg^{-1} or kWhkg^{-1}	Q14.5 ≥ 14.5 or Q4.0 ≥ 4.0	
Bulk density, MJ kg^{-1}	≥600	≥550
N, %	≤1.5	≤2.0
S, %	≤0.20	≤0.30
Cl, %	≤0.10	≤0.40
Cd, mg kg $^{-1}$	≤0.5	
Cu, mg kg $^{-1}$	≤20	
Pb, mg kg $^{-1}$	≤10	
Zn	≤100	

* Reproduced from International Standard ISO 17225-6:2021 Solid biofuels—Fuel specifications and classes—Part 6: Graded non-woody pellets after obtaining the permission of the Lithuanian Department of Standardization. Copyright is protected by the Lithuanian Department of Standardization.

The lowest CO_2 emission was determined when burning S-Mz pellets (3.5%), and the highest when burning MIX2-1 and MIX2-2 pellets (4.3%). However, CO_2 emissions when burning the pellets of all 7 samples were lower than when burning wood biomass pellets of the control variant (5.6%). When evaluating CO, C_xH_y, and NO_x emissions of all seven pellet samples, significant differences were observed. The lowest CO and C_xH_y emissions were determined when burning MIX2-1 pellets (572 and 29 ppm, respectively) and the highest when burning S-FB and S-FH pellets (1778 and 144 ppm). The lowest NO_x emissions were determined when burning S-Mz pellets (124 ppm) and the highest when burning S-FB pellets (270 ppm). However, CO, NO_x, and C_xH_y emissions when burning pellets of all seven samples were higher than when burning wood biomass pellets of the control variant (Table 4) because quality wood biofuel pellets were only used as a control option, they are used in combustion and emission studies using different types of plant biomass.

Table 4. Harmful emissions by burning pellets.

	CO_2	CO	NO_x	C_xH_y
Unit	**%**	**ppm**	**ppm**	**ppm**
S-Mz	3.5	786	124	42
S-FH	3.7	1609	160	144
S-FB	3.9	1778	270	111
MIX2-1	4.3	572	168	29
MIX2-2	4.3	1052	210	40
MIX2-3	4.1	1406	238	74
MIX3-1	4.1	815	194	34
Wood pellets	5.6	90	43	9

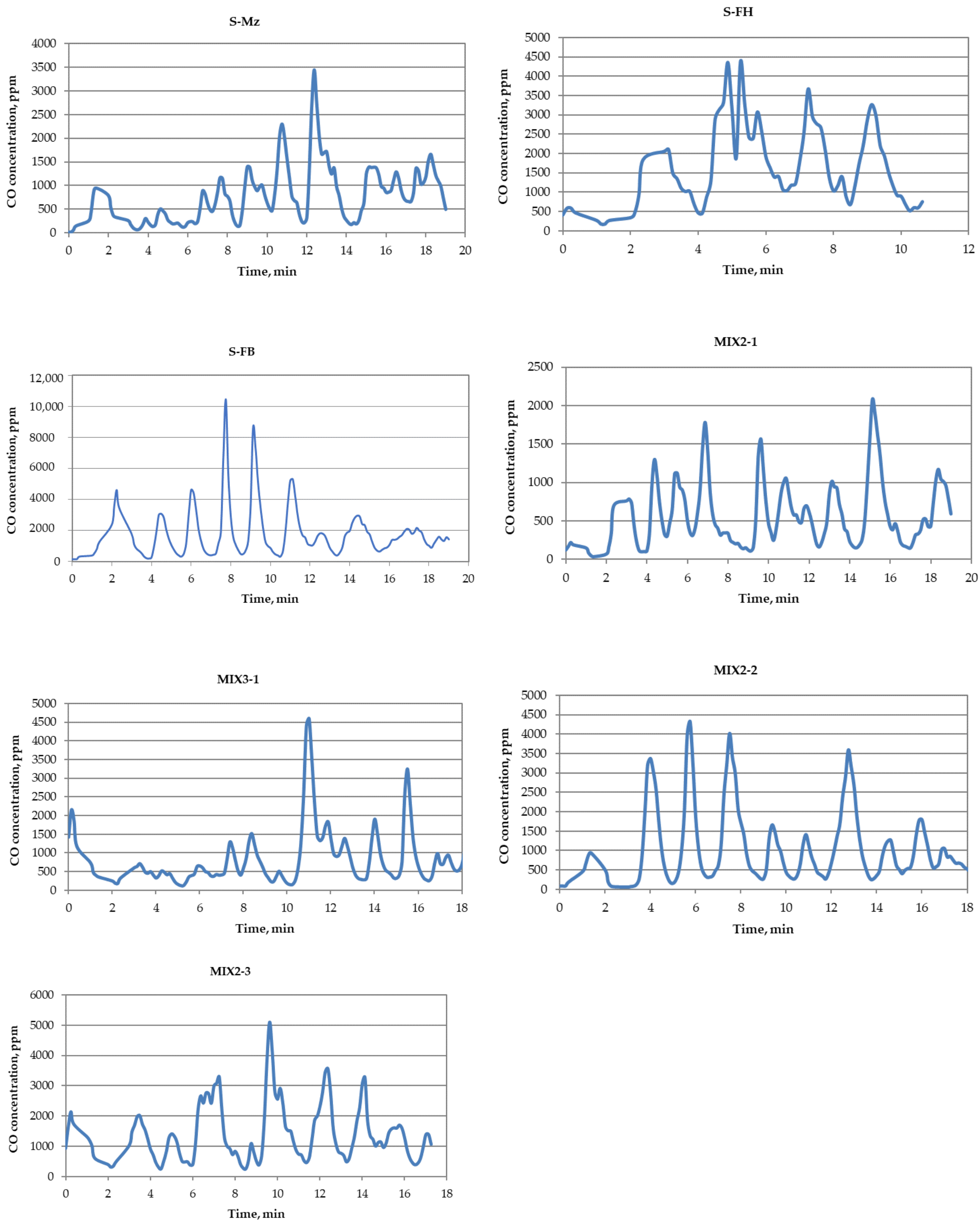


Figure 5. CO concentrations during combustion of plant biomass pellets.

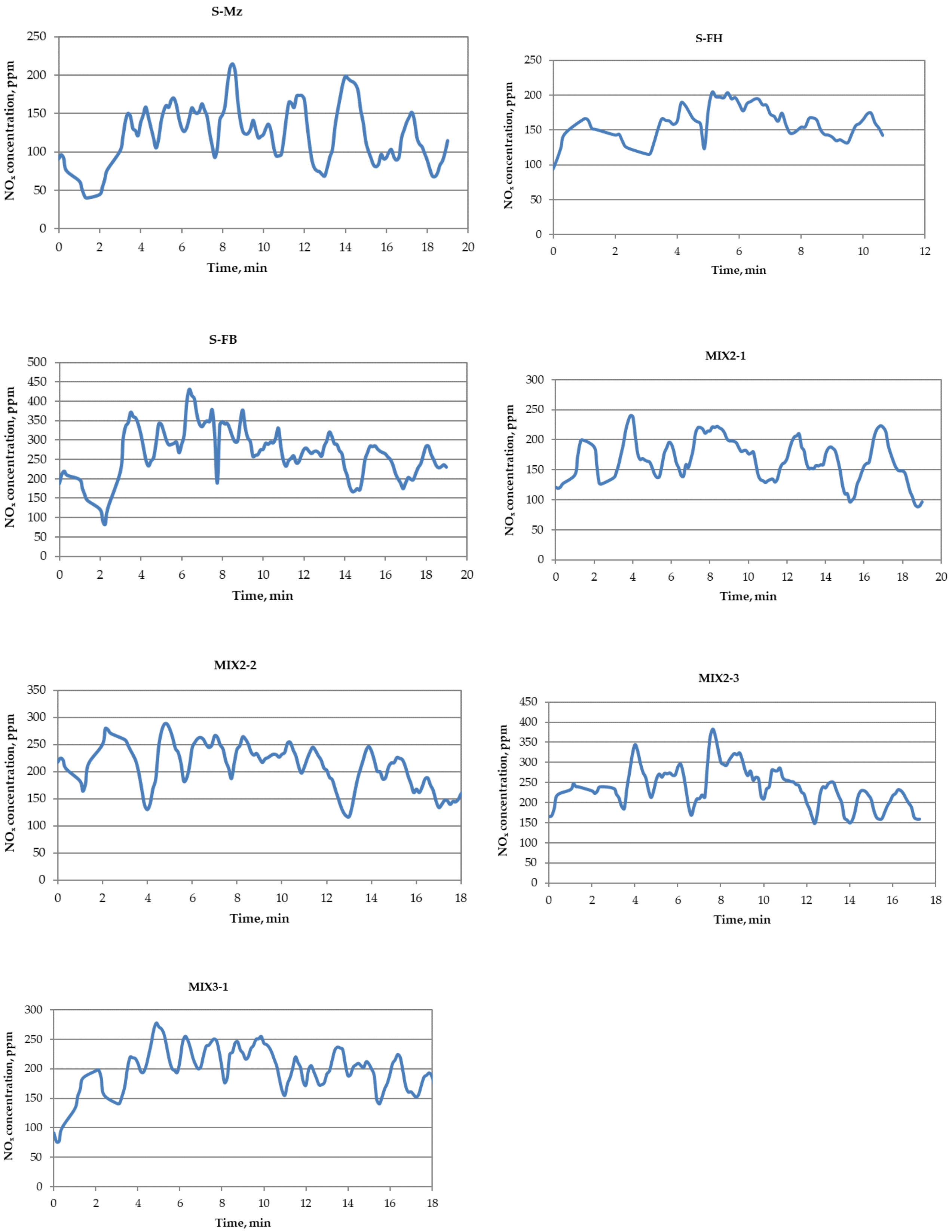


Figure 6. NO_x concentrations during combustion of pellets.

Figure 7. MIX3-1 pellets burning process.

The burning process of pellets MIX3-1 is presented in Figure 7. Pellets burned well all the time, and there were only several drops of flame. During the test period, pellet residue and slag accumulated on one-half of the burner.

Data from other authors' previous studies on emissions from burning biomass pellets are presented in Table 5.

Table 5. Harmful emissions from burning biomass under different conditions.

Type of Pellets	Boiler Type	Combustion Conditions	Emissions	Source of Literature
Sunflower husk pellets Pine pellets	10 kW domestic biomass boiler	The temperature distribution inside the combustion chamber (300–850 °C)	CO_2—9.82% CO—704 ppm NO—344 ppm NO_2—20.7 ppm CO_2—9.87% CO—665 ppm NO—640 ppm NO_2—30 ppm	[50]
Wheat straw pellets Rye straw pellets Birch sawdust pellets	10 kW nominal thermal power and 80% efficiency grate boiler	Air is supplied by a fan at a constant blowing speed of 1 m s^{-1}. The air supply channel was about m^3 h^{-1}	CO_2—0.76–7.71% CO_2—0.6–6.88% CO_2—2.81–7.66%.	[51]
Rape straw pellets	25 kW boiler. Fuel is automatically supplied to the furnace installed in the boiler	The fuel mass flow was 6.15 kg h^{-1} for wood pellets and 7.63 kg h^{-1} for rape straw pellets. The exhaust gas temperature was 138 °C and 134 °C, respectively.	CO_2—3.15% NO_x—119.2 ppm CH_4—579.6 ppm	[3]
Wheat straw pellets Spruce pellets	2 kW power experimental device	A propane flame flow (1.2 kJ s^{-1}) is supplied to the upper part of the biomass layer. Underneath the layer of biomass pellets, primary air is supplied with an average velocity of 0.57 g s^{-1}. Secondary swirl air with an average air supply speed of 0.6 g s^{-1} and a speed of S < 0.6 is supplied at the bottom of the burner	CO_2—11.94% CO—250 ppm NO_x—250.7 ppm CO_2—13.21% CO—121 ppm NO_x—67.2 ppm	[46]
Faba bean waste pellets	5 kW solid fuel boiler	Burning time of each sample—10–12 min. The oven maintains the smoke temperature at 200 ± 5 °C. The thrust in the measuring section is maintained at 13 ± 1 Pa.	CO_2—4.1–5.0% CO—1072–2785 ppm NO_x—133–266 ppm	[52]
Faba bean waste and potato peel pellets	5 kW solid fuel boiler	Burning time of each sample—10–12 min. The oven maintains the smoke temperature at 200 ± 5 °C. The thrust in the measuring section is maintained at 13 ± 1 Pa.	CO_2—3.41–4.00% CO—1103–3163 ppm NO_x—198–229 ppm C_xH_y—67–211 ppm	[53]

Bala-Litwiniak and Zajemska [50] found that CO_2 emissions from burning sunflower husk pellets were 9.82%, and CO emissions were 704 ppm. CO_2 emissions were slightly higher during the combustion of pine biomass pellets—9.87%, but CO emissions were lower—665 ppm. These authors indicated that NO_x emissions were lower during the combustion of sunflower husk pellets, and this is due to a higher amount of nitrogen in pine biomass pellets. The determined values of NO and NO_2 during the combustion of sunflower husk pellets were 344 and 20.7 ppm, respectively, while pine biomass pellets had the values 640 and 30 ppm, respectively. Jach-Nocoń et al. [37] stated that the amount of fuel-bound nitrogen has the greatest influence on NO_x emissions in low-power boilers. When burning corn stover and sunflower husk pellets made of intensively grown agricultural plants, the above-mentioned authors determined significantly higher emissions than when burning wood pellets. Forest wood emits the least NO_x because it is grown extensively and does not use nitrogen fertilizers [37]. However, Kraszkiewicz et al. [51] studies on the burning of biomass pellets of wood and herbaceous plants showed that the amount of nitrogen in the biomass is not the most important factor indicating the formation of NO emissions, and even the geometric shape of the fuel affects the amount of NO in the exhaust gases in the first phase of fuel combustion. He also found out that CO_2 emissions during the burning of wheat straw pellets ranged from 0.76 to 7.71% depending on the combustion phase, rye straw—from 0.6 to 6.88%, and birch sawdust—from 2.81 to 7.66%.

As indicated by Zajac et al. [52], it is not possible to draw generalized conclusions about the emissions obtained during the study since the combustion process is very sensitive to the conditions in which it takes place. Even a slight change in conditions can cause large changes in emissions. Therefore, the results obtained cannot be considered a very accurate source of information about emissions. Further emissions studies are needed considering the equipment used and different types of biomasses [52].

It should be noted that the pollutant level, when measured under laboratory conditions, may differ from emissions from, e.g., domestic boilers. The actual level of emissions depends on the fuel quality, boiler operation, and boiler maintenance. CO emissions are influenced not only by the type of biofuel but also by the type of device and the procedure for performing the test. The simpler and less controlled the device is, the higher the CO emission factor is. Meanwhile, different devices do not affect NO_x and SO_2 emissions. The determining factor for NOx emissions is the amount of nitrogen in the biofuel, and for SO_2 emissions, it is the amount of sulfur in the biofuel [42].

Wasilewski et al. [3] found that CO_2 emissions by burning rape straw pellets were 3.15%. NO_x emissions were 119.2 ppm, and CH_4 emissions were 579.6 ppm. Barmina et al. [46] determined that during the burning of wheat straw biomass pellets, CO emissions were 250 ppm, CO_2 emissions were 11.94%, and NO_x emissions were 250.7 ppm. Whereas for spruce biomass pellets, these data were 121 ppm, 13.21%, and 67.2 ppm, respectively. The authors explain that higher CO_2 emissions in spruce biomass pellets are due to the higher carbon content in these pellets and higher NO_x emissions in the combustion of wheat straw pellets are due to higher nitrogen content in these pellets.

Our data on emissions from burning biomass pellets can be objectively compared with Jansinskas et al. [53] and Manajeva et al. [54] research data because the combustion conditions were analogous, and the same burning implement was used. Our determined CO_2 emissions when burning S-FB (faba bean biomass) pellets were 0.2–1.1 percentage points lower compared to the data obtained by Jasinskas et al. [53] when burning faba bean waste pellets. Meanwhile, emissions of CO and NO_x were quite similar. Our determined CO emissions are similar to the research results obtained by Minajeva et al. [54] when burning faba bean waste and potato peel pellets. Our determined CO emissions are similar to the research results obtained by Minajeva et al. during the burning of faba bean waste and potato peel pellets. Meanwhile, the highest value of CO emissions determined by us when burning pellets of sample of S-FB was 1.8 times lower compared to the highest value of CO emissions determined by the author when burning pellets of the already mentioned mixture in different proportions.

4. Conclusions

For research purposes, seven types of pellets were produced using fibrous hemp, maize, and faba bean plants, which were grown as mono and multi-crop. The length and diameter, ash content, and calorific value of all pellets met the requirements of the ISO 17225-6:2021 standard. The moisture content of S-FH pellets exceeded the norm allowed by the standard by 1.63 percentage points. The amount of N exceeded the standard rate very slightly in the produced S-FB pellets. The concentrations of chemical elements S, Cl, Cd, Cu, Pb, and Zn determined in the granules of all samples did not exceed the values set in the standard. The highest NO_x emissions were found when burning S-FB pellets, which had the highest N content, and conversely, the lowest NO_x emissions were found in S-Mz pellets, which had the lowest N content. The lowest CO and C_xH_y emissions had MIX2-1 pellets, which were produced from binary crop biomass. In summary, it can be stated that multi-crop plants are a suitable raw material for the production of pressed solid biofuel, the burning of which does not cause negative consequences for the environment. In order to more accurately assess the effect of burning multi-crop plant biomass pellets on household boiler users, studies with higher-power industrial incineration implements should be conducted.

Author Contributions: Conceptualization, R.P., A.J. and N.P.; methodology, R.P., A.J., E.L., M.P. and S.K.; validation, R.P., A.J. and R.D.; formal analysis, R.P., A.J. and S.K.; investigation, R.P., A.J., E.L. and M.P.; data curation, R.P., E.L. and M.P.; writing—original draft preparation, R.P., A.J., E.L. and M.P.; writing—review and editing, A.J., R.D. and S.K.; supervision, R.P., A.J. and N.P.; funding acquisition, R.P., R.D. and N.P. All authors have read and agreed to the published version of the manuscript.

Funding: This research received no external funding.

Institutional Review Board Statement: Not applicable.

Informed Consent Statement: Not applicable.

Data Availability Statement: Not applicable.

Conflicts of Interest: The authors declare no conflict of interest.

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