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Original Research

Assessing the Emissions and Suitability of Agro-Based Pellets for Small-Scale Residential Heating Appliances in Serbia

Vasilije Matijašević[1], Viktor Tekáč[2], Zdeněk Beňo[2], Van Minh Duong[2]*

[1]Faculty of Technology and Metallurgy, University of Belgrade, Karnegijeva 4,
11000 Belgrade, Serbia
[2]Faculty of Environmental Technology, University of Chemistry and Technology Prague,
Technická 5, 166 28 Prague, Czech Republic

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Abstract

Emissions from small-scale heating appliances significantly contribute to exacerbating Serbia's severe air quality challenges. This study investigates two types of pellets, produced from wheat crop residue and a mixture of wheat and rapeseed, as potential alternatives to wood pellets. The analysis covers key solid biofuel characteristics, including heating value, moisture, volatile matter, elemental composition, ash content, and melting temperature, in accordance with normative specifications. Results reveal that agropellets from wheat and the mixture fail to meet established requirements, disqualifying them from any *ENplus* classification. Emission characteristic tests, conducted in an automatic residential pellet stove, measure O_2, CO_2, CO, NO_x, SO_2, and TOC. Comparative analysis with wood pellets quantitatively indicates higher gaseous pollutant emissions from agropellets. Notably, the combustion of agropellets proves unsuccessful, leading to the extinguishing of the fire in the appliance, rendering them unsuitable for household use. The findings underscore the importance of ensuring high-quality pellets are available on the market and in domestic appliances, presenting implications for end-users and policymakers. Addressing these issues is crucial for mitigating air quality concerns and further study associated with heating in households.

Keywords: small-scale heating, biomass, agropellets, air quality, emissions

Introduction

The surge in climate change, driven by fossil fuel emissions, escalating energy demands, and fossil fuel depletion, has propelled rapid advancements in renewable energy [1-3]. Bioenergy assumes a pivotal role in the transition to complete renewable energy, offering abundant biomass and minimal carbon emissions [4, 5]. Solid biofuels, derived from plant biomass, animal waste, and municipal waste, are integral for energy production in various forms (firewood, crop residue) or processed states (pellets, briquettes, wood chips) [6, 7].

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*e-mail: Van.Minh.Duong@vscht.cz
Tel.: +420 220 444 229

Presently, solid biofuels contribute 10-14% to the global energy supply, standing as the second most utilized energy source after fossil fuels [8].

Pellets, derived from woody biomass, offer superior fuel properties compared to traditional alternatives like firewood, boasting homogeneity, high packing density, and condensed energy content [9, 10]. Primarily sourced from forest and plantation wood residues, pellets undergo a production process involving drying, balling, cutting, pulverizing, pre-treating, and compression, resulting in pellets of 6 mm-8 mm diameter and lengths ranging from 5 mm-30 mm [11, 12]. Agricultural feedstock stands out as a significant alternative for pellet production, given its abundance, affordability, and the opportunity to repurpose agricultural waste [13, 14]. In Serbia, pellets are predominantly crafted from slabs, long-length roundwood, and firewood [15]. Between 2012 and 2015, pellet consumption witnessed a remarkable 20.4% average annual growth, reaching 89,000 tons in 2015, notably in households and commercial facilities. However, approximately 40% of total production is exported, representing a missed opportunity for domestic use in replacing fossil fuels, which could substantially alleviate air pollution and reduce CO_2 emissions [15].

Pellets are converted into energy in the process of direct combustion, a process in which biomaterials are burned in the presence of air at temperatures of 800–1000ºC, or co-combustion (or co-firing), a technique of burning biomass alongside conventional fossil fuels like coal [16-18]. Wood pellets are used in small-scale combustion appliances, namely pellet stoves, pellet boilers, and pellet ovens, for the production of heat and cooking in households [19]. In Serbian households, firewood is the most widely used heating means, with usage of nearly 43.2%, while pellets are in fifth place after district heating, electricity, and natural gas, with usage of only 5.1%. Firewood-burning stoves are the predominant heating appliance (45.9%), while pellet stoves amount to 0.9% and pellet boilers amount to 6% of the total share of household heating appliances. One-third of all Serbian individual heating systems are older than 15 years [20].

Combustion of solid biofuels causes the emission of various pollutants, including particulate matter (PM_{10}, $PM_{2.5}$), black carbon (BC), heavy metals, gaseous pollutants – CO, SO_x, NO_x, NMVOC, PAHs, hexachlorobenzene (HCB), and polychlorinated dibenzo-dioxins and furans (PCDD/F). In small appliances, emissions are significantly higher than in larger plants due to incomplete combustion, linked to factors such as inadequate fuel-air mixing, low oxygen concentrations, low temperature, or short residence times [19]. Emissions from woody biomass combustion are also shown to be influenced by fuel properties, appliance design and age, and operating conditions [21]. Biomass composition notably affects emissions of PCDD/Fs, NO_x, SO_x, and heavy metals [22]. Appliances with active control systems for combustion air supply, such as pellet stoves, demonstrate higher combustion efficiency and lower pollutant emissions compared to traditional biomass stoves and fireplaces [23]. Extensive research has established the link between air pollution and respiratory and cardiovascular diseases [24-27], with broad health effects ranging from short-term impacts to premature death, affecting respiratory, nervous, and cardiovascular systems, as well as reproductive capacities and child health [28].

Serbia faces severe air quality challenges, ranking as the 33rd most polluted country globally and the 5th in Europe in 2021, with an average US air quality index (AQI) of 79 [29]. $PM_{2.5}$ mass concentrations were notably problematic, exceeding the WHO air quality guideline value by 5.1 times. Primary contributors to air pollution in Serbia include household burning of solid fossil fuels and biofuels (mainly wood and coal), along with lignite and coal-fueled power stations for electricity production [30]. Key emitters of particulate matter include heating plants with less than 50 MW capacity and households [29]. The World Health Organization estimated that air pollution in Serbia caused 6,592 premature deaths and a loss of 131,183 years of life in 2016 [30]. The Health and Environmental Alliance (HEAL) reported over 1,000 cases of chronic bronchitis and 600 hospitalizations due to cardiovascular and respiratory issues linked to air quality. HEAL estimates from 2010 indicate 10,000 premature deaths in Serbia due to exposure to ozone and particulate matter, marking the second-highest air pollution-related death rate in Europe [28].

Given Serbia's pressing air quality concerns, a shift from firewood and lignite to less harmful alternatives like pellets is imperative. However, it is crucial to assess the emissions associated with combustion before the widespread adoption of solid biofuels. While prior studies have scrutinized emissions from pellets made of endemic wood species like oak, beech, and pine [31-34], limited research has explored agropellets produced from crop residue, particularly wheat and rapeseed straw, to understand the environmental impacts in Serbia.

Material and Methods

In this experimental work, two different types of pellets from Serbia were analyzed: wheat (*Triticum*) straw pellets and a blend of wheat (*Triticum*) and rapeseed (*Brassica napus*) straw pellets, complying with ISO 16559:2014 standards [35]. The pellets, with a commercial diameter of 6 mm, were free of additives or binders. Notably, these pellets were not yet available on the Serbian market during the study, which was in the testing phase. Wheat and rapeseed straws were explored as potential alternative feedstocks for heating in Serbia. Pellets were sampled from 15 kg bags using a scoop, meeting ISO 21945:2020 requirements [36]. Sample preparation followed ISO 14780:2017 guidelines [37] and underwent comprehensive analysis to acquire both fuel characteristics (proximate analysis, specific energy testing, ultimate analysis, and determination of ash melting temperature) and pollutant emission data.

For proximate analysis, pellets were comminuted using a pulverisette 15 to achieve a top size of 2 mm. Cleaning procedures prevented cross-contamination. The proximate analysis consists of three measurements of four parameters: moisture content (on a wet basis), volatile matter, ash content, and fixed carbon (on a dry basis), and the result was averaged. Moisture content analysis was performed in accordance with ISO 18134-3:2015 specifications [38]. The volatile matter analysis was performed following ISO 18123:2015 guidelines [39]. The analysis of ash content was conducted in compliance with ISO 18122:2015 standards [40]. The fixed carbon content (its mass fraction) was determined as the difference between 1 and the sum of the mass fractions of ash and volatile matter.

Specific energy analysis, adhering to ISO 18125:2017 specifications [41], utilized a bomb calorimeter (IKA C 200) to measure both the gross calorific value (HHV) and the net calorific value (LHV). The compressed sample was placed inside a combustion crucible, and the calorimeter, filled with water, was sealed. The weight of the sample was entered, and the measurement started, with the HHV obtained after 14 minutes, repeated twice for each sample type. Benzoic acid served as the weight standard, and the LHV [MJ/kg] was calculated from the HHV.

Elemental analysis, complying with ISO 16948:2015 [42] and ISO 16994:2016 [43] guidelines, investigated the mass fractions of carbon, hydrogen, nitrogen, chlorine, and sulfur. The Silfradent hydraulic press 660 was employed to shape two samples of each pellet type into cylindrical forms (12 mm diameter, 4 mm height), enclosed in metal cups. Thermo Scientific Flash EA 1112 CHNS analyzer conducted the analysis. Approximately 1 mg of each sample was weighed, placed in a tin container, and subjected to analysis. The system displayed mass fractions of carbon, hydrogen, nitrogen, and sulfur after a 10-minute process. Chlorine determination adhering to ISO 16995:2015 [44] utilized the titration method, based on the presence of water-soluble chlorides in solid biofuels. Following a digestion process, titration employed a 0.01 M silver nitrate ($AgNO_3$) solution with 5 ml of 5% potassium chromate (K_2CrO_4) as an indicator. The chlorine mass fraction was calculated using the volume of $AgNO_3$ solution consumed, following the principle of volumetric titration. The oxygen mass fraction was determined as the difference between 1 and the sum of the mass fractions of C, H, N, S, Cl, and ash, employing the principle of elemental mass balance.

Ash melting temperature determination, following ISO 21404:2020 requirements [45]. Approximately 1 g of ash from each sample underwent a controlled temperature increase to 250ºC (5ºC per minute) for 180 minutes, followed by a rise to 550ºC (10ºC per minute) for 120 minutes. The cooled ash was ground, moistened with ethanol, and pressed into a cylinder mold using a spring press. Test pieces were heated incrementally at room temperature, and temperatures were recorded at specific changes in ash shape: deformation temperature (DT), shrinkage starting temperature (SST), hemisphere temperature (HT), and flow temperature (FT). Recordings were made at intervals of 5ºC from 550ºC onward, ensuring the accuracy and reliability of the data readings.

The on-site test-bench apparatus, assembled in the Emission Measurement Laboratory at UCT Prague, adheres to the European standard EN 16510-1:2018 [46]. Utilizing a Toledo IV 32 residential space heater from Haas + Sohn, the setup incorporates integral components such as a pellet storage hopper, rotating auger conveyor, firebox with a burner pot, electric spark ignition system, fire door with a window, exhaust gas air heat exchanger, and control panel. Wood pellets were automatically fed into the burner pot through a rotating auger conveyor, with primary combustion air introduced through openings on the bottom and side. The residue falls into the ash pan below through the bottom grate. The combustion chamber, accessible through a firebox door with a viewing window, undergoes automated burn tests every 30 minutes, increasing air volume to cleanse the burner of ash and residue. The technical specifications and performance details of the test appliance are outlined in Table 1.

Table 1. Technical specifications of Toledo IV 32 residential space heater.

Parameter	Value	Unit
Nominal heat output	8	[kW]
Heat output range	2.4–8.3	[kW]
Fuel type	Wood pellets	
Ignition procedure	Electric	
Pellet hopper capacity	32	[kg]
Combustion time with one hopper (min./max.)	20/60	[h]
Thermal efficiency	90/95	[%]
Flue gas temperature	206	[ºC]
Chimney draught at nominal heat output	11	[Pa]
Chimney draught at minimal heat output	5	[Pa]
Mass flow of flue gases	5.6	[g/s]
Dimensions		
Height	1,201	[mm]
Width	544	[mm]
Depth	499	[mm]
Diameter of the flue gas outlet	80	[mm]
Weight	110	[kg]

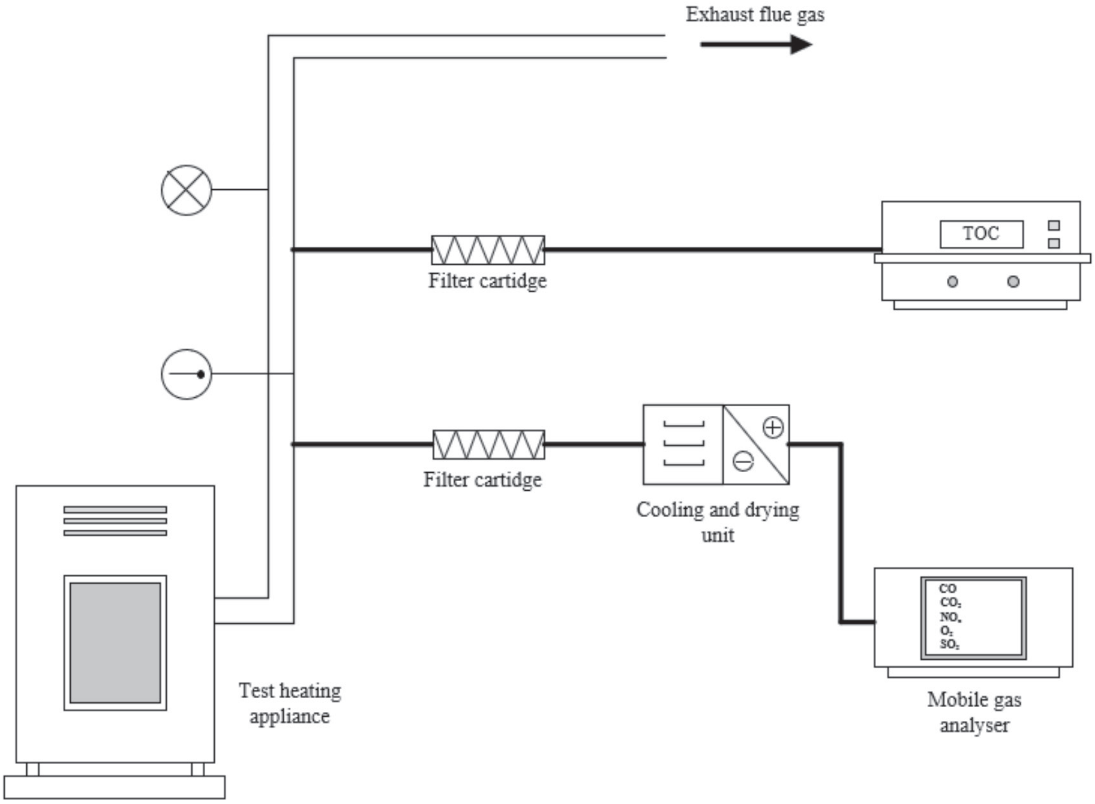

Fig. 1. Test-bench equipment arrangement layout scheme.

The test-bench configuration, construction layout, and measurement devices are illustrated in Fig. 1. An uninsulated flue connector (Ø 80 mm, 2 m long, 1 mm thick) linked the test appliance flue socket to the measuring section, connected by an uninsulated flue adapter. The downstream measuring section, featuring temperature and pressure measurement as well as analysis of gaseous inorganic pollutants and total organic carbon (TOC), had a Ø 80 mm diameter to match the flue socket. Temperature was recorded with GMH 3200 series thermoelements, and the chimney draft was measured by a GMH 3100 series differential manometer, with the optimal pressure specified by the Greisinger manufacturer as 5-15 Pa.

For quantitative analysis, two exhaust gas samples were drawn from the exit flow. PSP 4000-H gas sample probes (M&C Tech Group) with heated sintered ceramic filters (200ºC) were used to eliminate solid particles. Heated pipes at 200ºC transported the conditioned gas sample. Without further treatment, a THERMO FID analyzer (SK-Elektronik GmbH) directly on the heated line measured gaseous organic compounds as TOC. The upper measurement limit was established using a propane-nitrogen gas mixture with a certified propane (C3H8) mass fraction of 99.1 ml/m^3. A PSS 5 cooling and drying unit (M&C Tech Group) removed moisture from the gas sample to prevent interference with pollutant measurement.

Emission measurements during combustion in the small-scale heating appliance included oxygen, carbon dioxide, carbon monoxide, sulfur dioxide, nitrogen oxides, and organic gaseous pollutants. Post-gas conditioning, a PG 350 analyzer (HORIBA) was employed for the determination of O_2, CO, CO_2, SO_2, NO, and NO_2 in the exhaust gas. The analysis methods comprised paramagnetic analysis for O_2, chemiluminescence for NO and NO_2, and non-dispersive infrared spectroscopy for CO, CO_2, and SO_2. The measuring range was established using air (zero gas for CO_2 and gaseous inorganic pollutants) and certified SIAD gas mixtures. Analyte values were recorded at 30-second intervals in the internal memory of the analyzer and later manually transferred to a computer via USB after completion of the measurement. Pollutant volume fractions in wet flue gas were measured, recalculated into dry flue gas, and used for the calculations of the pollutant mass flow, the emission factor referring to fuel mass, and the emission factor referring to fuel energy content.

The test run comprised a pre-test lasting 30 minutes and main test periods for stable pellet burning, with three batch tests conducted for each type of analyzed agropellets. Each test run lasted either 25 minutes (wheat pellets) or 30 minutes (wheat and rapeseed mix pellets). Initial pellet loading was 5 kg, and the remaining pellets in the hopper were measured post-test for fuel mass calculation. The test parameters of 3 batch test runs and their average values for the wheat pellets and the wheat and rapeseed mix pellets are summarized in Table 2. For wheat pellets, 0.85 kg was burned on average, yielding

Table 2. Parameters measured during the three test runs of the analysed pellets.

Parameter	Value								Unit
	Wheat				Wheat and rapeseed mix				
	Test 1	*Test 2*	*Test 3*	*Average*	*Test 1*	*Test 2*	*Test 3*	*Average*	
Start time	07:40	09:15	10:10		09:50	11:05	12:00		[hh:mm]
End time	08:05	09:40	10:35		10:20	11:35	12:30		[hh:mm]
Actual measurement duration	0.42	0.42	0.42	0.42	0.5	0.5	0.5	0.5	[h]
Solid biofuel mass	0.87	0.85	0.84	0.85	1.10	0.72	0.74	0.85	[kg]
Solid biofuel mass flow	2.07	2.02	2.00	2.03	2.20	1.44	1.48	1.71	[kg/h]
Atmospheric temperature	12.3	12.3	12.3	12.3	5.2	5.2	5.2	5.2	[ºC]
Atmospheric pressure	1.021	1.021	1.021	1.021	1.023	1.023	1.023	1.023	[hPa]
Relative humidity	69	69	69	69	85	85	85	85	[%]
Average ambient room temperature	22.8	25.4	25.3	24.5	22.7	24.0	23.5	23.4	[ºC]
Average exhaust gas temperature	208.5	199.8	201.4	203.2	211.7	209.5	205.7	209.0	[ºC]
Average negative pressure in the exhaust gas draught	26.3	24.9	27.6	26.3	30.6	30.3	30.9	30.6	[Pa]
Average exhaust gas flow rate	20.80	20.30	20.10	20.40	20.13	13.18	13.54	15.62	[m3/h]
Power intake	34.01	33.18	32.86	33.35	34.53	22.60	23.23	26.79	[MJ/h]

a consumption rate of 2.07-2.00 kg/h in three tests. The average room temperature was 24.5ºC, and exhaust gas temperatures ranged from 203.2ºC to 208.5ºC. The draft pressure was 26.3 Pa, and the highest flow rate was 20.80 m^3/h to the average of 22.40 m^3/h. Wheat and rapeseed mix pellet tests burned 0.85 kg per run on average, with a consumption rate fluctuating from 2.20 kg/h (first test) to an average of 1.71 kg/h. Room temperature averaged 23.4ºC, and exhaust gas temperatures ranged from 205.7ºC to 211.7ºC. The draft pressure averaged 30.6 Pa, with flow rates ranging from 13.18 to 20.13 m^3/h across tests.

Results and Discussion

Fuel Characterization

Table 3 summarizes the results of fuel characterization analyses for wheat pellets and wheat and rapeseed mix pellets. On a dry basis, wheat pellets exhibit a carbon mass fraction of 44.93%, hydrogen at 5.92%, and a gross calorific value of 18.6 MJ/kg. The moisture content of 7.57% aligns with the European standard EN 14961 [47]. The high volatile matter content (77.2%) indicates significant vaporization before combustion. Nitrogen (0.73%) and sulfur (0.03%) meet EN*Plus* B requirements, specified by the ISO 17225-2 specifications [48]. However, ash (6.01%) and chlorine (0.41%) levels surpass specifications for the B quality class. In comparison to beech pellets, wheat pellets display lower ash melting temperatures: deformation at 980ºC, shrinkage starting at 1000ºC, hemisphere temperature at 1200ºC, and flow temperature at 1210ºC.

Mixed pellets, based on ultimate analysis, exhibit carbon and hydrogen values of 43.32% and 5.66% on a dry basis, resulting in a higher heating value of 17.0 MJ/kg. The moisture content, though high at 11.61%, falls within the acceptable range (≤15%) specified by European standard EN 14961 for fruit biomass or a mixture of wood or herbaceous biomass. Nitrogen content of 0.77% on a dry basis meets the required ≤1% for the EN*Plus* B quality type, but sulfur (0.12%) and chlorine (0.24%) significantly exceed the specified ≤0.05 level for sulfur and ≤0.03 level for chlorine according to the ISO 17225-2 standard [48]. Additionally, the ash content of 7.73% on a dry basis is much higher than the 2% limit for the B quality class. High ash content in wheat and rapeseed mix pellets contributes to a gross calorific value difference of 1.4 MJ/kg between dry without ash and dry basis. Ash melting temperature analysis reveals the lowest values compared to beech and wheat pellets, with temperatures of 960ºC, 980ºC, 1090ºC, and 1100ºC at which deformation, shrinkage, hemisphere shape, and thin layer spreading occur, respectively.

A comparison of wheat and wheat-rapeseed mixture pellets reveals distinct differences in fuel characteristics. The mix pellets exhibit lower carbon and hydrogen mass fractions and higher moisture and ash content, resulting in a lower calorific value. Additionally, the nitrogen and sulfur content of mix pellets is higher, indicating a potential for increased pollutant emissions. The mix pellets demonstrate inferior quality in terms of heating

Table 3. Characteristics of analysed pellets as the test solid biofuel.

Analysis	Value						Unit
	Wheat			Wheat and rapeseed mix			
	raw	dry	daf	raw	dry	daf	
Proximate, mass fraction							
Moisture content	7.57	0.00	0.00	11.61	0.00	0.00	[%]
Volatile matter	71.38	77.22	82.16	66.34	75.06	81.34	[%]
Ash content	5.55	6.01	0.00	6.83	7.73	0.00	[%]
Fixed carbon	15.50	16.77	17.84	15.22	17.22	18.66	[%]
Specific energy							
Gross calorific value (HHV)	17179	18586	19774	15009	16980	18402	[kJ/kg]
Net calorific value (LHV)	15852	17350	18460	13585	15690	17004	[kJ/kg]
Ultimate, mass fraction							
Carbon	41.53	44.93	47.80	38.29	43.32	46.94	[%]
Hydrogen	5.47	5.92	6.30	5.00	5.66	6.13	[%]
Nitrogen	0.68	0.73	0.78	0.68	0.77	0.83	[%]
Oxygen	38.79	41.97	44.65	37.27	42.17	45.70	[%]
Sulfur	0.03	0.03	0.04	0.10	0.12	0.13	[%]
Chlorine	0.38	0.41	0.44	0.21	0.24	0.26	[%]
Ash melting							
Deformation temperature (DT)	980			960			[ºC]
Shrinkage temperature (SST)	1000			980			[ºC]
Hemisphere temperature (HT)	1200			1090			[ºC]
Flow temperature (FT)	1210			1100			[ºC]

value, ash content, melting characteristics, and potential pollutant emissions.

A study by Carroll and Finnan measured the ash and moisture content, elemental composition, and gross calorific value of wheat and rapeseed pellets [49]. The recorded ash mass fraction for wheat (6.49%) and rapeseed (5.83%) is lower than that measured for the wheat and rapeseed mix pellets. Although the wheat moisture content (10.66%) is higher than measured for wheat pellets, both wheat and rapeseed pellets have a lower moisture content (11.39%) than that measured for mixed pellets. The gross calorific value of wheat pellets in the study (18.8 MJ/kg) closely aligns with the measured value, while rapeseed pellets' value (18.2 MJ/kg) surpasses the measured value of mixed pellets. Another study from 2020 examined wheat straw pellets with a recorded moisture content of 8.2%, volatile matter and ash content of 67.8% and 13.06%, respectively, and a gross calorific value of 18.0 MJ/kg on a raw basis [50]. In comparison to the measured values for wheat pellets from Serbia, the volatile matter content is lower, the ash content is significantly higher, and the high calorific value of wheat pellets from the study surpasses the measured 17.2 MJ/kg on the raw basis in this experiment. N. Dragutinovic et al. analyzed the characteristics of wheat and rapeseed agropellets [51]. The wheat agropellets exhibited a moisture content of 14.77% and an ash content of 6.65%, which are higher than the measured wheat pellet values. The rapeseed agropellets moisture and ash content from the study were 8.84% and 3.95%, respectively, lower compared to measured wheat and rapeseed. The gross calorific values were 19.6 MJ/kg and 18.5MJ/kg for wheat and rapeseed pellets, respectively, which are higher than the measured agropellets values.

A study from 2023 measured the ash melting temperatures of alfalfa, wheat straw, and hay agropellets [52]. Hay ash pellet ash melting temperatures were 916ºC (DT) – 1135ºC (FT), which is in line with recorded wheat and rapeseed pellet values, while alfalfa ash melting temperatures of 1017ºC (DT) – 1237ºC (FT) were similar to the measured temperatures of wheat pellet ash. The wheat straw pellets from the study exhibited the highest flow temperature value of 1308ºC. This study confirms that recorded values for ash melting temperatures are in line with those observed in agropellets.

Table 4. Gaseous emissions from analysed pellets run.

Air pollutant	Value								Unit
	Wheat				Wheat and rapeseed mix				
	Test 1	*Test 2*	*Test 3*	Average	Test 1	Test 2	Test 3	Average	
Measurement readings									
Oxygen	14.90	14.21	14.15	14.42	14.76	14.62	13.71	14.36	[%]
Carbon dioxide	5.76	6.43	6.48	6.22	5.86	6.00	6.88	6.24	[%]
Carbon monoxide	930	763	830	841	1164	1048	813	1009	[ml/m^3]
Nitrogen oxides as NO$_2$	183.2	202.3	201.4	195.6	223.7	236.2	247.2	235.7	[ml/m^3]
Sulfur dioxide	25.4	34.3	35.9	31.9	36.8	53.3	69.2	53.1	[ml/m^3]
Total organic carbon	14.2	14.5	11.6	13.4	45.1	21.1	19.5	28.6	[ml/m^3]
Mass concentration under standard conditions									
Carbon monoxide	1671	1372	1491	1511	2092	1883	1462	1812	[mg/m^3]
Nitrogen oxides as NO$_2$	540	596	594	576	659	696	729	695	[mg/m^3]
Sulfur dioxide	104	141	147	131	151	219	284	218	[mg/m^3]
Total organic carbon	33.17	33.68	27.00	31.28	105.20	49.11	45.37	66.56	[mg/m^3]
Average mass flow									
Carbon monoxide	34.75	27.85	29.97	30.86	42.11	24.82	19.80	28.91	[m^3/h]
Nitrogen oxides as NO$_2$	11.23	12.10	11.93	11.75	13.27	9.17	9.87	10.77	[m^3/h]
Sulfur dioxide	2.17	2.86	2.96	2.67	3.04	2.89	3.85	3.26	[m^3/h]
Total organic carbon	0.69	0.68	0.54	0.64	2.12	0.65	0.61	1.13	[m^3/h]
Emission factor referring to the fuel quantity									
Carbon monoxide	16.79	13.79	14.99	15.19	19.14	17.23	13.38	16.58	[g/kg]
Nitrogen oxides as NO$_2$	5.43	5.99	5.96	5.79	6.03	6.37	6.67	6.36	[g/kg]
Sulfur dioxide	1.05	1.42	1.48	1.32	1.38	2.00	2.60	2.00	[g/kg]
Total organic carbon	0.33	0.34	0.27	0.31	0.96	0.45	0.42	0.61	[g/kg]
Emission factor referring to the fuel energy content									
Carbon monoxide	977	803	872	884	1275	1148	891	1105	[mg/MJ]
Nitrogen oxides as NO$_2$	316	349	347	337	402	424	444	423	[mg/MJ]
Sulfur dioxide	61	83	86	77	92	134	173	133	[mg/MJ]
Total organic carbon	19	20	16	18	64	30	28	41	[mg/MJ]

Several studies evaluated the fuel characteristics of wood pellets from Serbia, including beech, pine, and oak [53-55]. Beech wood pellets and pine wood pellets had moisture contents of 8.4% and 8.8%, ash contents of 0.5% and 0.6%, and higher heating values of 18.4 MJ/kg and 19.8 MJ/kg, respectively [53]. Oak wood pellets measured values of ash contents and the higher heating values were 0.5% and 18.6 MJ/kg [54]. The ash melting temperatures of pine wood pellets were around 1200-1300ºC, while oak pellets' ash melting temperatures exceeded 1500ºC [55]. Compared to these wood pellet values, wheat and mixed agropellets exhibit inferior fuel characteristics, notably higher moisture, and ash content, as well as ash melting temperatures.

Emission Profiles

The combustion of wheat pellets and wheat-rapeseed mix pellets led to the expected emissions of oxygen, carbon dioxide, carbon monoxide, sulfur dioxide, nitrogen oxides (as NO$_2$), and total organic carbon. However, due to the high ash content, which posed challenges to proper combustion, the test period was divided into three shorter segments to accommodate technical limitations. Table 4 provides a summary of the measured values of volume fractions of criteria gases [%, ml/m^3] along with calculated values of mass concentration under standard conditions [mg/m^3], average mass flow [g/h], and average factors referring to the fuel amount [g/kg] and energy [g/MJ]. Fig. 2 illustrates the emission profiles of the main gaseous exhaust gas components (O_2, CO_2, CO, NO_2, SO_2, and TOC) during the tests conducted on wheat pellets, revealing the unstable nature of emissions caused by the non-homogeneous combustion mixture. This instability was evident in the fluctuating levels of pollutants and the eventual extinguishing of the fire due to the accumulation of combustion residue in the burner.

Across the three tests, the average volume fraction of oxygen in the exhaust gas was 14.42%, while carbon dioxide had an average volume fraction of 6.22%, as summarized in Table 4. Carbon monoxide (CO) exhibited the highest average volume fraction during the first test at 930 ml/m^3, with a three-test average of 841 ml/m^3. The average concentration of CO mass concentration was 1511 mg/m^3, resulting in an emission factor of 884 mg/MJ. This finding is notably higher than the CO emissions reported for straw pellets (728 mg/MJ and 311 mg/MJ) and wood pellets (94.8 mg/MJ and 455 mg/MJ) in an earlier study [31]. The incomplete combustion with elevated CO levels could potentially

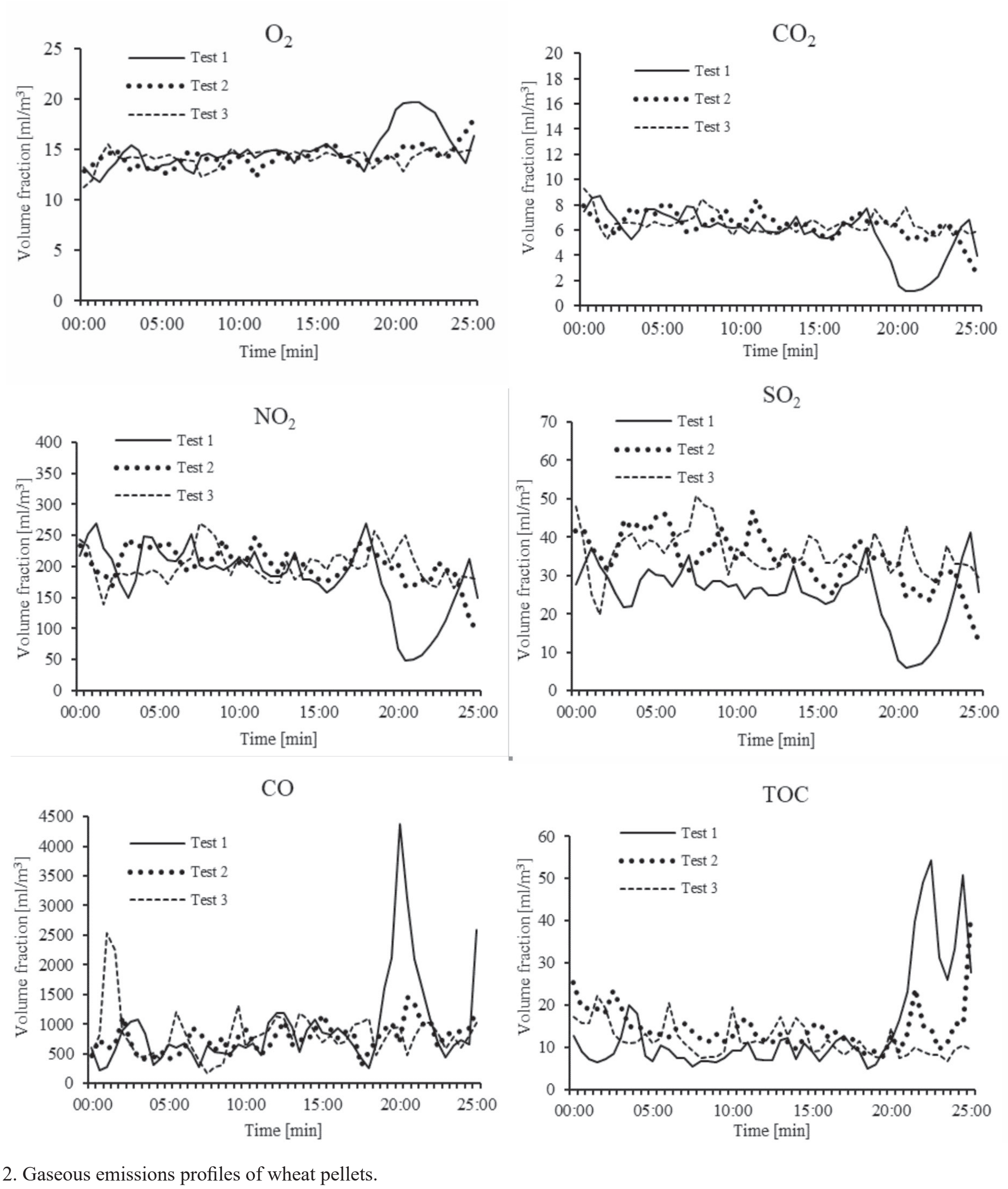

Fig. 2. Gaseous emissions profiles of wheat pellets.

be mitigated by optimizing combustion parameters, improving air distribution, and ensuring proper ventilation.

The high volume fraction of nitrogen oxides (NO_x), expressed as NO_2, ranged from 183.2 to 201.4 ml/m^3 across the three tests, averaging 195.6 ml/m^3. The NO_2 average emission factor referring to the fuel mass was 5.79 g/kg, comparatively lower than values reported for rapeseed bark pellets (7.15±0.60 g/kg) in a previous study [31]. However, it is significantly higher than NO_2 emissions from various wood pellets reported in other studies [32-34] (spruce stem pellets (95±28 mg/MJ), willow stem pellets (103±28 mg/MJ), pine stem pellets (82±16 mg/MJ), and oak pellets (147±30 mg/MJ), with an average energy-based emission factor of 337 mg/MJ. The elevated NO_x emissions resulted from the nitrogen content in the mixed pellets and suboptimal combustion conditions in small-scale heating appliances.

Sulfur dioxide (SO_2) was measured at an average volume fraction of 31.9 ml/m^3, with a mass concentration at a standard conditions value of 131 mg/m^3. The average SO_2 fuel mass-based emission factor across the three tests was 1.32 g/kg, considerably lower than the value of the rapeseed pellet (5.08±0.49 g/kg) in a prior study [31]. The SO_2 energy-based emission factor averaged 77 mg/MJ, which is lower than the value of 220 mg/MJ reported by L. Carvalho [56] for wheat pellets, while it is significantly higher than the values for wood pellets in the literature [31], which varied from 0.05 mg/MJ to 13.3 mg/MJ.

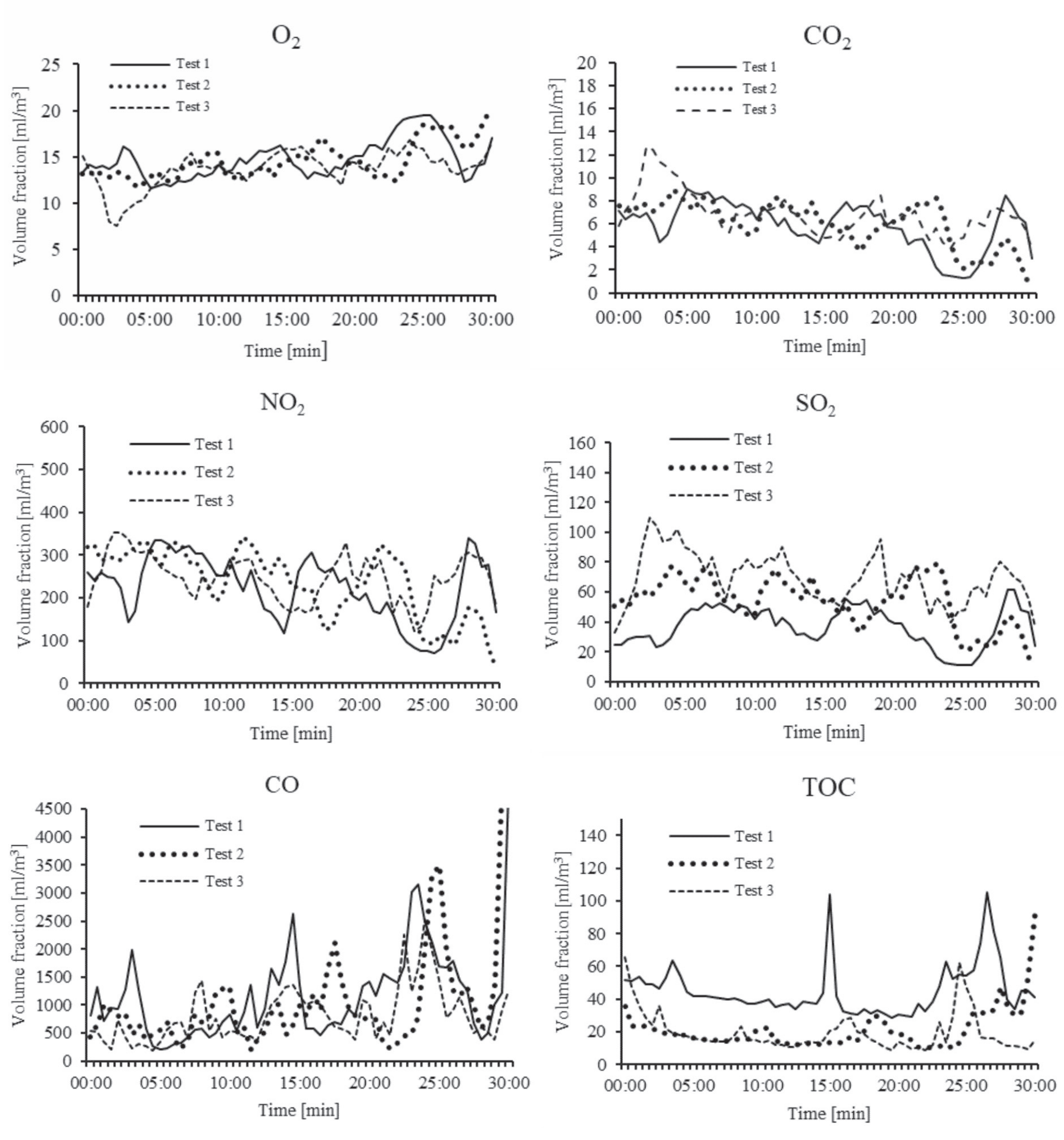

Fig. 3. Gaseous emissions profiles of wheat and rapeseed mix pellets.

Total organic carbon (TOC) had an average volume fraction of 13.4 ml/m^3, with an average mass concentration under standard conditions of 31.28 mg/m^3. The average energy-based emission factor for TOC was 18 mg/MJ, which is lower than the average value of agropellets of 28.7 mg/MJ [57], but is higher compared to wood pellets (2.9 mg/MJ to 38.2 mg/MJ), reported in earlier studies [58].

Fig. 3 depicts the emission profiles of six criteria gases in the exhaust gas during three 30-minute tests of wheat and rapeseed mix pellets. Emissions exhibited instability due to a non-homogeneous combustion mixture, leading to incomplete burning conditions, an unstable flame, and localized hotspots with elevated temperatures in the combustion chamber. Oxygen fluctuated, averaging 14.36% for the three tests, while carbon dioxide maintained an average volume fraction of 6.24% (see Table 4). Carbon monoxide, a byproduct of incomplete combustion, exhibited an average volume fraction of 1009 ml/m^3, an average mass concentration of 1812 mg/m^3, and an average emission factor of 1105 mg/MJ, significantly higher than wheat pellet emissions.

Nitrogen oxides (NO_x), expressed as NO_2, originating from the air nitrogen and fuel nitrogen content, averaged a volume fraction of 235.7 ml/m^3 with an average mass concentration of 695 mg/m^3. This value was 1.2 times higher than the NO_2 emissions from wheat pellets. Factors influencing this difference include the higher oxygen volume fraction during wheat pellet combustion, indicating a lower flame temperature, and the higher nitrogen content of wheat and rapeseed mix pellets (see Table 3), leading to increased NO_x production. The average NO_2 fuel mass-based emission factor of mix pellets was 6.36 g/kg, slightly lower than the value of rapeseed bark pellets (5.08 ± 0.49 g/kg) from an earlier study [31].

Sulfur dioxide (SO_2) exhibited volume fractions of 36.8, 53.3, and 69.2 ml/m^3 in tests 1-3, respectively, averaging 53.1 ml/m^3. The three test average mass concentration was 218 mg/m^3, 1.7 times higher than the average wheat value. Since SO_2 emissions originate from the sulfur content of the fuel, higher emissions align with the higher sulfur content of the mixed pellets (see Table 3). The SO_2 average emission factor of 133 mg/MJ was lower than the values of rapeseed bark pellet (224 mg/MJ) and wheat bran pellet (220 mg/MJ) reported in earlier studies [31, 56].

The total organic carbon (TOC) volume fraction varied from 42.6% (test 1) to 18.2% (test 3), with a three-test average mass concentration of 66.56 mg/m^3 and an average emission factor of 41 mg/MJ. This is higher than the values for agropellets from a previous study [57] and more than twice that of wheat pellets. Since TOC and CO emissions are associated with incomplete combustion, one of the factors that contribute to this phenomenon is the high moisture content of this biofuel (see Table 3). Furthermore, the higher chimney draft during the combustion of mix pellets, as evidenced by a higher oxygen volume fraction in the exhaust gas, may further contribute to these observed emissions differences.

Conclusions

The study provides key insights into the characteristics and applicability potential of wheat (*Triticum*) straw and a mixture of wheat and rapeseed (*Brassica napus*) straw blend biomass pellets for small-scale combustion. Agropellets derived from these crop residues in Serbia exhibit undesirable fuel composition and heating characteristics. High moisture and ash contents, along with high sulfur and chlorine contents, indicate non-compliance with the EN high-quality classification outlined in the normative reference ISO 17225–2:2021.

The combustion test revealed that both wheat pellets and wheat and rapeseed mix pellets were unsuitable for household appliances, with the fire being extinguished after approximately 30 minutes. The combustion residue showed non-homogeneous characteristics, suggesting incomplete combustion. Among the analyzed pellets, wheat and rapeseed mix pellets exhibited higher emissions than wheat pellets. Although NO_2, SO_2, TOC, and CO emissions aligned with average agropellets values from previous studies, pollutant emissions were notably higher compared to wood pellets. Incomplete combustion further contributed to elevated flue gas flow and related emissions. Future research should explore the combustion residue for potential contaminants, toxic elements, and ash-related issues.

The study underscores the limitations of automatic household pellet stoves, designed exclusively for high-quality wood pellets, rendering them incapable of burning the analyzed agropellets effectively. The findings indicate that agropellets from crop residues exhibit high pollutant emissions and are unsuitable for residential pellet appliances. Further investigations could explore the applicability of this solid biofuel in the industrial sector.

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Conflict of Interest

The authors declare no conflict of interest

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