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Energy and emission properties of burley tobacco stalk briquettes and its combinations with other biomass as promising replacement for coal

Maja Malnar[1], Vesna Radojičić[1], Gordana Kulić[1], Zoran Dinić[2], and Olga Cvetković[3]

[1] University of Belgrade Faculty of Agriculture, Belgrade, Serbia
[2] Institute of Soil Science, Belgrade, Serbia
[3] University of Belgrade Institute of Chemistry, Technology, and Metallurgy, Belgrade, Serbia

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As a tobacco producer, Serbia has to deal with large amounts of leftover tobacco stalks after harvesting. One option for this type of biomass is to burn it, but burning is not encouraged in Serbia, since the levels of its combustion products have not been investigated yet. The aim of this study was therefore to determine the elemental composition, ash and nicotine content, heat values, and composition of gaseous combustion products of tobacco stalk briquettes and to see if their mixing with other types of biomass available in Serbia could improve their ecological profile. We made 11 different types of briquettes: six of pure raw materials, including burley tobacco stalks, sunflower head remains, wheat straw, corncob, soy straw, and beech sawdust and five by mixing tobacco stalks with these other raw materials in a 50:50 mass ratio. All briquettes meet the ecological criteria regarding the emission limits for nitrogen oxides (NO_x), sulphur dioxide, carbon monoxide, and carbon dioxide. Nicotine content in flue gases (<10 mg/kg) is far below the maximum level allowed by the European Union. Heat values of all biomass samples are acceptable, although lower than those specified for solid biofuels (≥16.0 MJ/kg), save for corncob and beech sawdust and their mixtures with tobacco stalks. Our findings therefore encourage the use of tobacco stalks as a viable biofuel.

KEY WORDS: combustion products; heat value; nicotine content; renewable energy

As the efforts to reduce the use of fossil fuel gain momentum, renewable sources have gained increasing economic and ecological importance (1). This particularly applies to countries that import fossil fuels, such as Serbia. One of the renewable options is to reduce pollution, the need for artificial fertilisers, and import of fossil fuel energy, and redirect development to rural areas is the use of agricultural biomass. Furthermore, agricultural biomass has lately been promoted to replace wood biomass by 2020, especially in central European countries (2).

One such potential biomass that presents significant disposal issues around the world are waste tobacco stalks (3). China, which produces over two million metric tonnes (t) of tobacco a year (4), has developed a wide range of efficient tobacco biomass uses, including the use of tobacco stalk to replace coal briquettes in tobacco drying (5) or bulk curing (6). Furthermore, studies of their chemical composition suggest that tobacco stalks can be used as biofuel (7–14), but there is still more left to learn about their use in combination with other types of biomass.

Serbia is at the top of European countries in the amount of available biomass, much of it coming from leftover agricultural plant mass (12.5 million metric tonnes). More than a half is corn, followed by wheat straw (>25 %) and harvest residues of sunflower, soybean, rapeseed, tobacco stalks, or pruning residues from orchards and vineyards (about 15 %) (15). In fact, of its total estimated biomass potential of 3.40 million toe (tonne of oil equivalent) per year, only about 30 % is currently being used (16), which roughly corresponds to the entire biomass potential originating from agriculture (about 1.05 toe). This volume of biomass can meet all the energy requirements in the agricultural sector (16). A large portion, more than 43000 t, comes from large-leaf burley or Virginia tobacco stalks, and only a small amount (25 %) is ploughed back into land, while the rest is disposed of as a waste or illegally burned at the field (17), which raises a variety of concerns, including waste of resources and environmental pollution (18, 19).

One of the solutions is to convert it to biofuel (20–22), considering that the toxic nicotine content is about 0.005 %, according to a Macedonian study (14) of briquettes made of oriental tobacco stalks.

The aim of our study was therefore to analyse elemental composition of tobacco stalk briquettes from biomass generated in Serbia, their heat values, and levels of their combustion products. All these parameters were also investigated for other crop biomasses and mixtures with tobacco stalks (50:50). Another aim was to determine the most favourable biomass

Corresponding author: Vesna Radojičić, University of Belgrade Faculty of Agriculture, Institute for Food Technology and Biochemistry, Nemanjina 6, 11080, Zemun-Belgrade, Serbia, E-mail: *mntabacco@agrif.bg.ac.rs*

combination in terms of thermal energy yielded and lowest pollution emission, as such combinations reduce the nicotine content in briquettes. We hoped that our findings would inform the industry how to repurpose biomass that would otherwise go to waste and to obtain quality biofuel that might be used to improve the self-sustainability of Serbian agriculture in the future.

MATERIALS AND METHODS

Materials

For this study we used 11 different biomass samples from Serbia in the form of fuel briquettes. Six were made of either burley tobacco stalks from Šabac tobacco fields (44° 45' N, 19° 42' E, Mačva district in Western Serbia), wheat straw, corn cob, both from Stara Pazova fields (44° 59' N, 20° 10' E, Srem district, Vojvodina province, Serbia), soybean straw and sunflower head remains, both from the Golubinci fields (44° 59' N, 20° 04' E, Srem District, Vojvodina province, Serbia), or beech sawdust from a wood processing company (origin unknown). Five samples were mixtures (in a 50:50 ratio) of tobacco stalks and one of the above mentioned biomass material. We opted for burley tobacco because stalks are mowed and dried together with leaves, which facilitates collection and transport and enables energy saving in drying. After drying, leaves are removed from and used for cigarette manufacture. Beech sawdust was selected as a material of the longest tradition in the production of energy briquettes in Serbia.

Sawdust and tobacco stalks were pre-dried as described elsewhere (23), whereas all other biomass was heaped and dried in a room for 30 days, during which time it was turned regularly to improve evaporation (24). The humidity of all biomass after drying was 9.66–10.85 % and complies with the European Standard of ≤12 % (25).

Dry biomass was then ground and homogenised in a mill, which is an integral part of the briquetting machine (Macinatore, MAC 500, CO.MA.FER, Collebeato, Italy), and subsequently by manual mixing to make the briquettes.

Briquettes (of 6 cm diameter, 5–11 cm long) were made with no binding materials using a briquetting machine (Macinatore, MAC 500) operating at a pressure of 1000 kPa and 50–150 kg/h output. They were then stored in large impregnated paper bags in an isolated room for 40 days until heat value and gaseous combustion product analysis, because Obernberger and Thek (24) found that four to six weeks of storing ensures pellet quality that is otherwise achieved only with biological additives. For combustion we only used briquettes of a uniform length of 8 cm.

Chemical analysis

The obtained briquettes were ground in a mill and sieved repeatedly to separate 0.5–1.0 mm particle fractions, which were taken for analysis. Samples were stored in paper bags in a cool and dry place for up to 15 days.

Moisture content was determined by oven-drying the samples (laboratory oven Digitheat 80 L, JP Selecta, Abrera, Spain) at 105 °C as described elsewhere (26).

Ash content obtained by combustion in the electric muffle furnace at 810 °C was determined following the EN 14775:2011 procedure (27).

Carbon (C), hydrogen (H), and nitrogen (N) content was determined as specified by the European Standard EN 15104:2011 (28). Sulphur (S) content was determined as specified by the European Standard EN 15289:2011 (29). Oxygen (O) content was calculated using the following equation:

O (%)=100-C (%)-H (%)-N (%)-S (%)-Ash (%) [1]

Nicotine content analysis

Nicotine (≥99 % purity) standard was purchased from Merck (KGaA, Darmstadt, Germany) and other solvents were HPLC-grade from Fisher Scientific (Pittsburgh, PA, USA).

Briquettes were ground in a mill (Wiley Mill, Model 4, Thomas Scientific, New Jersey, USA), passed through a 2-mm mesh, oven-dried to a constant dry weight at 60 °C for 24 h, distributed into 0.5 g lots, and extracted with 10 mL of 25 mmol/L sodium phosphate buffer (pH 7.8) at 30 °C for 24 h with constant agitation. The aqueous extract was filtered through a filter paper, diluted ten times with water, filtered again through a 0.45-μm pore mesh, and sealed in a screw-capped septum vial to permit automatic injection of a 20-μL aliquot. Samples were then eluted with an isocratic mobile phase containing 40 % (v/v) methanol and a 0.2 % (v/v) phosphoric acid buffer (pH 7.25) at a flow-rate of 0.5 mL/min. Nicotine was identified with a Waters high-performance liquid chromatograph (HPLC) (Waters Breeze, Binary Pump systems, Milford, MA, USA) at UV wavelengths between 210 and 400 nm and quantified at 254 nm as described elsewhere (30, 31).

Flue gas nicotine content was investigated in the smoke created by burning 1 kg of briquettes. At the top of the furnace flue we placed a Cambridge filter pad (92 mm in diameter; Bogwaldt, Germany) to collect smoke gases (32). The filter was then removed using laboratory tweezers, and nicotine particles extracted and quantified using the HPLC method described above (30, 31).

Determination of heat value

To determine heat values of each briquette type, samples were burned in an oxygen bomb calorimeter (IKA C400 Adiabatisch*)* according to the EN 14918 standard (33). The upper thermal power was calculated based on the amount of heat released minus the heat released by the formation of sulphuric and nitric acid during combustion in the calorimetric bomb. From the amount of available hydrogen and moisture we then calculated the lower thermal power as a realistic parameter for evaluating the heat value of the biomass.

Determination of gaseous combustion products

The combustion products (CO_2, CO, NO, NO_2, NO_x, and SO_2) were analysed in gas released from 3 kg of briquettes burnt in a 65 kW burning chamber at 1000 °C. The chamber had a fixed grid, and biomass was inserted manually.

Gases were determined following the protocol described elsewhere (34), with a probe of a flue gas analyser (VARIO plus industrial, MRU MessgerätefürRauchgase und Umweltschutz GmbH, Wiener Neustadt, Austria) inserted at the mouth of the furnace pipe exhaust, and read from the device's display. Three measurements were performed for each sample at minute 1, 5, and 9 of combustion that lasted ten minutes. Measurement characteristics (range and accuracy) of the MRU flue gas analyser are shown in Table 1.

All parameters met standard requirements for lab testing (35), measurements were done in triplicate, and the results are presented as means ± standard deviations (SD).

Calculation of the CO_2 emission factor

The CO_2 emission factor is the average amount of greenhouse gas emissions in relation to the emission source, assuming that all carbon from the biomass is completely oxidised (burnt) and converted into CO_2 (36). It is calculated based on the amount of carbon in the fuel at the lower heat value of the fuel, as follows:

$$EF = \frac{m(C)}{100} x \frac{3.664}{LHW} x 1000 \quad (tCO_2/TJ), [2]$$

where EF is the emission factor, m (C) – carbon mass in the fuel (with total moisture expressed in %), LHW the lower heat value of the fuel (with total moisture expressed in MJ/kg), and 3.664 the stoichiometric coefficient.

Statistical analysis

Data obtained from the experiments are expressed as means ± standard deviations (SD). We used one-way analysis of variance (ANOVA) to compare mean differences between samples. Correlations between samples were tested with Pearson's correlation coefficient. All analyses were run on the SPSS version 23.0 (37).

Table 1 Measurement range and accuracy of the MRU flue gas analyser

Component	Measurement range	Accuracy
CO	0–10000 mg/kg	±5 %
NO	0–3000 mg/kg	±5 %
NO_2	0–500 mg/kg	±5 %
SO_2	0–5000 mg/kg	±5 %
O_2	0–25 % vol.	±0.8 %
Temperatures	-40–1200 °C	±0.5 %
Speed	0–40 m/s	±0.4 %

RESULTS AND DISCUSSION

Table 2 shows mean percentages of ash content and elements, heat values, and CO_2 emission factors for all biomass samples.

Ash content is important in estimating biofuel quality. An increase in ash content by 1 % corresponds to a decrease in heat value of 200 kJ/kg (38), even though this relation is not strictly proportional in our results (Table 3). This may be owed to a certain level of heterogeneity of the samples, which is common and acceptable in briquette manufacture (35). Ash content in briquettes (≤7 %) turned out to meet standard criteria described elsewhere (39), save for sunflower head remains, which dropped to an acceptable level only in the mixture with tobacco. Burley tobacco stalk briquettes produced 5.13 % of ash, which is higher than reported in other countries (12, 40) but similar to the percentages reported for tobacco cultivated in Serbia (8, 9, 41). The composition of elements in the briquettes can also help to calculate their heat value using a simple and reliable procedure (9, 38, 42) based on the carbon content. Simply put, the higher its content, the higher its heat value.

In contrast, higher hydrogen content corresponds to lower heat values. However, it did not have much effect, as its content did not significantly differ between samples (5.12–5.91 %).

Nitrogen has a somewhat stronger heat lowering effect than hydrogen, and its content is the highest in tobacco stalk briquettes (3.70 %), where it mostly originates from nicotine. Due to the nicotine content, tobacco waste is generally classified as harmful (43). Some of the nitrogen content may originate from nitrogen/phosphorus/potassium fertilisers, because burley tobacco requires large amounts of nitrogen fertiliser, as reported elsewhere (44). However, judging by similar heat values between tobacco and soy straw or sunflower head remains, which had significantly lower nitrogen content, nitrogen did not significantly affect the heat value of burley tobacco briquettes.

In contrast, high ash mineral content, particularly the one in wheat straw (7.11 %) and sunflower head remains (12.17 %) seems to come with significantly lower heat values than expected from their high carbon content (Table 2). However, we should be careful with conclusions in this respect, as our samples were highly heterogeneous, which is common in briquette manufacture.

Speaking of heat value, only corncob and sawdust briquettes and their mixtures with tobacco stalks meet the standard of ≥16.0 MJ/kg (45), but considering that they are renewable energy sources and that their nitrogen and sulphur content is lower than that of coal (46–49), all biomass combinations are acceptable as biofuel. Another reason to replace coal is that the calculated CO_2 emission factor is about 100 tCO_2/TJ lower.

Table 3 shows significant dependencies between the chemical composition of various types of briquettes and their heat values (Pearson's correlation coefficient). Heat values show a strong negative correlation with the oxygen, nitrogen, and ash content and a strong positive correlation with the carbon and hydrogen content.

Table 2 Mean (±SD) ash content, heat value, and elemental composition of various types of briquetted biomass

Briquette	Ash (%)***	LHW (MJ/kg)**	Elemental composition (%)					EF (tCO_2/TJ)
			C***	H**	N**	S*	O**	
BTS	5.13±0.05	15.28±0.07	43.09±0.13	5.37±0.11	3.70±0.04	trace	47.84±0.20	103.33
CC	1.26±0.03	16.83±0.02	46.67±0.02	5.91±0.05	1.00±0.02	trace	46.42±0.06	101.6
BS	0.54±0.02	17.27±0.04	48.97±0.05	5.78±0.05	trace	trace	45.25±0.11	103.9
SHR	12.17±0.10	14.90±0.11	47.77±0.07	5.80±0.04	1.63±0.04	0.63±0.02	44.17±0.06	117.47
SS	3.15±0.06	15.61±0.04	44.44±0.03	5.66±0.06	1.60±0.02	trace	48.30±0.07	104.31
WS	7.11±0.07	15.16±0.06	43.25±0.05	5.12±0.04	0.72±0.02	trace	50.91±0.07	104.53
BTS/CC	3.66±0.04	16.47±0.04	44.69±0.05	5.81±0.07	2.66±0.03	trace	46.84±0.09	99.42
BTS/SB	2.31±0.04	17.28±0.04	44.90±0.05	4.57±0.05	2.52±0.02	trace	48.01±0.08	95.21
BTS/SHR	7.73±0.08	15.75±0.08	46.06±0.06	5.77±0.06	0.93±0.02	0.7±0.02	46.54±0.07	107.08
BTS/SS	4.13±0.05	15.96±0.04	44.23±0.03	5.42±0.06	2.41±0.04	trace	47.94±0.05	101.54
BTS/WS	6.20±0.07	15.49±0.07	47.63±0.05	5.74±0.02	1.55±0.03	trace	45.08±0.04	112.66

The mean difference is significant at the 0.05 level: *** between all samples; ** between more than 5 samples; * between 5 or fewer samples. BS – beech sawdust; BTS – burley tobacco stalk; CC – corncob; SHR – sunflower head remains; SS – soybean straw; WS – wheat straw

Table 4 shows the content of gaseous products of briquette combustion. Even though sunflower alone or combined with tobacco biomass contained sulphur (less than 1 %), no sulphur oxide was identified among combustion gases. This may be owed to different analytical methods used for biomass and gases (micro-elemental vs technical analysis). Furthermore, biomasses differ in oxygen content (Table 2) and burn differently. What is more important, however, is that all briquettes meet the requirements of the national Directive on emission limit values for air pollutants (50), since the CO content is lower than 4000 mg/m^3. The same is true with respect to NO_x content, which is below the limit value of 500 mg/m^3 (50). Combustion products of all briquettes are acceptable from the environmental protection point of view.

As for nicotine, its content in flue gases was lower than the detection limit of the instrument (Table 5) and meets both the national and the EU requirements (<500 mg/kg) (51) for all our briquettes to be considered environmentally acceptable.

Our comparison shows that in terms of environmental protection the optimal choice is the briquetted mixture of tobacco stalks and beech sawdust (NO: 203 mg/m^3, NO_x: 311.67 mg/m^3, CO: 1562 mg/m^3, CO_2: 7.44 %), even though the combination with sunflower head remains has even lower CO_2 emission.

Limitations of the applied experimental design

Our findings should be taken with some reserve, as the briquettes used in the experiments were not completely homogeneous, mixtures in particular. However, this relative heterogeneity in briquette composition reflects real-life situations and still has some informative value, especially in practical terms.

CONCLUSIONS

To our knowledge, this is the first research report profiling tobacco stalks as a renewable energy source in view of environmental and practical considerations. Our findings show that the chemical composition and heat value of tobacco stalks is quite similar to that of soy and wheat straw biomass. Its briquettes meet several standards in terms of ash content and CO and NO_x emission. With nicotine content of <10 mg/kg, it also meets the EU guidelines and has an acceptable heat of 15.28 MJ/kg, which can be improved to meet commercial use criteria (≥16.0 MJ/kg) by combining it with beech sawdust or corncob, either in separate briquettes or briquette mixtures.

Speaking of improvement, by mixing tobacco stalks (in a ratio of 50:50) with other forms of biomass, we also managed to reduce the amount of nicotine and nitrogen, which resulted in lower NO_x emission and increased heat value.

We therefore believe that tobacco stalks can be repurposed into useful, renewable biofuel, instead of remaining classified as hazardous waste.

Conflicts of interest

None to declare.

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Table 3 Pearson's coefficient of correlation for the lower heat value in relation to ash and element content in different types of biomass

LHW / Components	BTS	CC	SB	SHR	SS	WS	BTS/ CC	BTS/ SB	BTS/ SHR	BTS/ SS	BTS/ WS
BTS (A)	-	-	-	-	-	-	-	-	-	-	-1.000**
BTS (C)	0.998 *	-	-	-	-	-	-	-	-	-	-
BTS (N)	-1.000*	-	-	-	-	-	-	-	-	-	-
CC (C)	-	1.000**	-	-	-	-	0.997*	-	-	-	-
CC (H)	-	-	-	-	-	-	0.999*	-	-	-	-
SB (H)	-	-	0.999*	-	-	-	-	-	-	-	-
SB (O)	-	-	-0.997*	-	-	-	-	-	-	-	-
SHR (O)	-	-	-	-0.998*	-	-	-	-	-	-	-
SS (A)	-	-	-	-	-0.999*	-	-	-	-	-0.999*	-
SS (O)	-	-	-	-	-0.998*	-	-	-	-	-0.998*	-
WS (C)	-	-	-	-	-	0.999*	-	-	-	-	1.000**
WS (H)	-	-	-	-	-	1.000**	-	-	-	-	0.999*
WS (N)	-	-	-	-	-	-1.000**	-	-	-	-	-0.999*
WS (O)	-	-	-	-	-	-0.999*	-	-	-	-	-1.000**
BTS/CC (A)	-	-1.000**	-	-	-	-	-0.997*	-	-	-	-
BTS/CC (C)	-	0.999*	-	-	-	-	-	-	-	-	-
BTS/CC (H)	-	0.997*	-	-	-	-	1.000**	-	-	-	-
BTS/CC (O)	-	-0.998*	-	-	-	-	-1.000*	-	-	-	-
BTS/SB (C)	-	-	0.999*	-	-	-	-	-	-	-	-
BTS/SB (H)	-	-	-	-	-	-	-	0.999*	-	-	-
BTS/SB (O)	-	-	-0.998*	-	-	-	-	-	-	-	-
BTS/SHR (C)	-	-	-	-		-	-	-	0.999*	-	-
BTS/SHR (H)	-	-	-	-	-	-	-	-	0.999*	-	-
BTS/SHR (O)	-	-	-	-		-	-	-	-1.000**	-	-
BTS/SS (A)	-	-	-	-	-	-	-	-	-	-1.000*	-
BTS/WS (H)	-	-	-	-	-	-	-	-	-	-	0.999*
BTS/WS (N)	-	-	-	-	-	-	-	-	-	-	-0.999*

*p<0.05; **p <0.01. BS – beech sawdust; BTS – burley tobacco stalk; CC – corncob; SHR – sunflower head remains; SS – soybean straw; WS – wheat straw

Table 4 Mean percentage (±SD) of combustion products from briquettes

Briquette	O_2 (%) **	CO_2 (%) ***	CO (mg/m^3) ***	NO (mg/m^3) **	NO_x (mg/m^3) ***
BTS	12.88±0.04	8.10±0.04	1590.34±0.06	273.67±0.03	419.67±0.04
CC	12.53±0.06	7.90±0.04	2952.30±0.04	183.34±0.03	280.67±0.04
SB	12.52±0.06	9.44±0.04	2468.67±0.05	142.34±0.04	213.34±0.04
SHR	11.70±0.04	9.57±0.06	2105.34±0.04	273.67±0.07	419.34±0.05
SS	12.70±0.06	8.52±0.02	1593.00±0.02	247.34±0.03	366.34±0.04
WS	11.88±0.05	8.77±0.02	2815.00±0.04	173.34±0.04	301.34±0.03
BTS/CC	11.02±0.06	10.10±0.05	2592.67±0.05	244.67±0.06	374.67±0.04
BTS/SB	12.95±0.07	7.44±0.03	1562.00±0.06	203.00±0.06	311.67±0.05
BTS/SHR	14.31±0.06	6.40±0.06	2261.34±0.04	233.34±0.04	351.67±0.04
BTS/SS	10.98±0.06	9.74±0.04	2035.00±0.50	267.67±0.05	410.00±0.06
BTS/WS	11.05±0.06	9.97±0.05	3115.00±0.04	202.00±0.04	309.34±0.06

The mean difference is significant at the 0.05 level: *** between all samples; ** between more than 5 samples. BS – beech sawdust; BTS – burley tobacco stalk; CC – corncob; SHR – sunflower head remains; SS – soybean straw; WS – wheat straw

Table 5 Nicotine content in briquettes and flue gases produced by briquette combustion

Briquette	Nicotine content in briquettes (mg/kg)	Nicotine content in smoke (mg/kg)
BTS	715.6	<10.0
BTS/CC	532.7	<10.0
BTS/SB	486.2	<10.0
BTS/SHR	535.5	<10.0
BTS/SS	496.2	<10.0
BTS/WS	484.6	<10.0

BS – beech sawdust; BTS – burley tobacco stalk; CC – corncob; SHR – sunflower head remains; SS – soybean straw; WS – wheat straw

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Energetska i emisijska svojstva briketa od stabljika berlej duhana i njihovih kombinacija sa drugom biomasom kao obećavajuća zamjena za ugljen

Kao proizvođač duhana, Srbija raspolaže velikim količinama stabljika duhana koje ostaju nakon berbe listova. Jedna je od opcija za ovu vrstu biomase njezino izgaranje, ali se ono ne promiče u Srbiji jer još uvijek nisu istraženi njezini produkti izgaranja. Stoga je cilj ovog istraživanja bio utvrditi elementarni sastav, sadržaj pepela i nikotina, ogrjevnu vrijednost i sastav plinovitih produkata pri izgaranju briketa od duhanskih stabljika i ustanoviti može li njihovo miješanje s drugim vrstama biomase, koje su dostupne u Srbiji, poboljšati njihov ekološki profil. Proizveli smo 11 različitih vrsta briketa: šest od čistih sirovina, uključujući stabljike berlej duhana, ostatke glava suncokreta, pšeničnu slamu, kukuruzni oklasak, sojinu slamu i bukovu piljevinu, a pet od mješavina stabljike duhana s ostalim navedenim sirovinama u odnosu 50:50. Utvrđeno je da svi briketi ispunjavaju ekološke kriterije u pogledu ograničenja emisije dušikova oksida (NO_x), sumporova dioksida, ugljikova monoksida i ugljikova dioksida. Sadržaj nikotina u dimnim plinovima (<10 mg/kg) znatno je ispod maksimalne vrijednosti koju dopušta Europska unija. Prihvatljive su ogrjevne vrijednosti svih uzoraka biomase iako su ispod kriterija za specifikacije i klase čvrstih biogoriva (≥16,0 MJ/kg), osim za kukuruzni oklasak i bukovu piljevinu, kao i njihove mješavine sa stabljikama duhana. Naši nalazi stoga idu u prilog uporabi stabljika duhana kao održivoga biogoriva.

KLJUČNE RIJEČI: obnovljiva energija; produkti izgaranja; sadržaj nikotina; toplotna vrijednost