Comparison of emissions from smokeless coal combustion in a household heating boiler used in Central Europe

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Abstract

The objective of this work was to evaluate the relationship between the fuel quality and the gaseous and particulate pollutant emissions generated from a hot-water boiler during the combustion of different types of innovative processed fuels: smokeless coal, smokeless briquettes 1–3, smokeless pellets and unprocessed hard coal. The aim of our research was to prove the presumption that smokeless processed coals produce less gaseous and particulate emissions. By using modern fuels in already used and manufactured older boilers, there is a possibility to significantly reduce emissions of organic gaseous compounds (OGC) and polycyclic aromatic hydrocarbons (PAHs). The emission situation in the heating season can be significantly improved even without costly production, and thus consumption of natural resources and energy, and installation of modern boilers. Physical and chemical characterization of solid-fuel samples, including determination of moisture content, ash, volatile organic content, calorific value and elemental composition analysis, were performed. Fuels were burned in one type of hot-water boiler class 1 according to EN 303-5 to determine the impact of applied fuel types on pollutant emissions. The pollutant emissions were characterized by the contents of gaseous components: nitrogen oxides NO_x , sulfur dioxide SO_2 , carbon monoxide CO , carbon dioxide CO_2 , organic gaseous compounds OGC and particle components: total suspended particles TSP, particulate matter less than $2.5\ \mu\text{m}$ and $10\ \mu\text{m}$ ($\text{PM}_{2.5}$ and PM_{10} , respectively) and polycyclic aromatic hydrocarbons PAHs in both phases. The emission factors from six types of fuel were compared with applicable European standards. The lowest NO_x content was observed for smokeless briquette 1, while the lowest SO_2 content was observed for smokeless pellets. The emission of CO was at a similarly low level of $200\ \text{g/kg}$ for smokeless briquette 1, smokeless briquette 2 and hard coal. Gaseous and pollutant emissions described by $\text{PM}_{2.5}$ and TSP were observed to be the lowest for smokeless coal, smokeless briquette 1 and smokeless briquette 2.

Keywords Boiler · Coal · Smokeless · Combustion · Emission · Particulate · Polycyclic aromatic hydrocarbons

Introduction

The last census in the Czech Republic in 2011 showed that from a total of 3.6 million households, 9.2% were heated by coal, and its combustion decreased by 41% within

2001–2011 (Horak et al. 2017). The next census in the Czech Republic is in 2021. In Poland, 36.5% of the total of 14.4 million households in 2018 were heated by coal, and its combustion decreased by 14.5% within 2009–2018 (GUS 2020). The energy sector in Poland and the Czech Republic has been changing by acts and directives of the European Union. As reported by Manowska and Rybak (2018), the energy demand in Poland from hard and brown coals should decrease by 15% and 80%, respectively, by 2050 in comparison with 2010. This energy policy includes steps such as improving energy effectiveness, increasing energy security, developing renewable energy sources, developing competitive fuels and energy markets and reducing the impact on the environment. To decrease emissions from solid-fuel combustion in households and commercial buildings, many countries in Europe were actively encouraged to implement

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residential heating with wood and other types of biomass. Biomass is a renewable fuel of utmost promise. However, the process of biomass combustion is quite complicated and consists of devolatilization, gas-phase combustion of volatiles and char combustion (Chen et al. 2010), which cause pollutant emissions. Research efforts should be focused on the development of high-quality, innovative fuels to reduce the concentrations of atmospheric aerosols. Particularly during winter, pollutant emissions due to solid-fuel combustion for residential heating are much higher in smog episodes (Mikuška et al. 2015). These new types of low emission fuel can be safely used for energy purposes for the environment and public health in accordance with the best available technology (BAT) practices (Kubica et al. 2007). For this reason, data must be collected, and knowledge must be developed about emissions from standard combustion installations, in which innovative types of fuel are tested.

As presented in the WHO document (WHO 2015. ISBN 978 92 890 50760), the latest WHO indoor air quality guidelines strongly advise against the residential use of unprocessed or raw coal. This coal requires higher ignition and combustion temperatures and is a source of SO₂ and NO_x emissions due to its higher content of sulfur and nitrogen than biomass. Coals may contain toxic elements (such as fluorine, arsenic, selenium, mercury and lead). A research focus on the content and exposure of these pollutants and toxic contaminants from the use of clean or smokeless processed coal is recommended by the WHO.

Emissions from the combustion of new solid smokeless coal fuels in residential heating units have rarely been published previously. Ściążko et al. (1993) showed that smokeless coal fuel exceeded the combustion efficiency by 20–30%, e.g., benzo[a]pyrene resulted in 90-fold smaller emission. Matuszek et al. (2016) found that emissions of CO, dust, organic pollutants, benzo[a]pyrene and PAHs in the flue gas from the combustion of smokeless fuel were much lower than those from the combustion of coal (nut assortment) and other wood waste materials. Smokeless coal has already been described by Lasek et al. (2019) and Matuszek et al. (2016) when a significant decrease in gaseous and particulate emissions was obtained even in outdated construction boilers. The measured average pollutant emissions from combustion of smokeless coal at 10% O₂ were as follows: (1) CO 4.64×10^3 mg/m³, (2) NO_x 180 mg/m³, (3) SO_x 220 mg/m³, (4) OGC 94 mg/m³, (5) PM 86 mg/m³, (6) benzo[a]pyrene 0.02 mg/m³ and (7) 16 PAHs in total 0.45 mg/m³ (Lasek et al. 2019) and (1) CO 4.78×10^3 mg/m³, (2) NO_x 89 mg/m³, (3) OGC 64 mg/m³, (4) PM 62 mg/m³, (5) benzo[a]pyrene 0 mg/m³ and (6) 16 PAHs in total 0.59 mg/m³ (Matuszek et al. 2016). Compared to hard coal, the emission factors from the combustion of smokeless coal were on average sixfold lower for OGC, 8.5-fold lower for TSP, 23-fold lower for 16 PAHs and 25-fold lower

for benzo[a]pyrene (Lasek et al. 2019). The advantages of smokeless coal are described online as well (Smokeless 2020). This type of low-carbon fuel has a high calorific value, less ash and lower gaseous and particulate emissions which are the advantages of smokeless coal combustion. The disadvantage could be the price of smokeless coal which is about 20–30% higher than of conventional hard coal.

Currently, in Central and Eastern European countries, an extensive change of old combustion devices for the improvement of air pollutant emissions is underway. The replacement of old boilers by new boilers leads to air quality changes; however, the process of boiler replacement could take a long time. The use of smokeless fuels could lead to a rapid improvement in the local air quality during the first winter heating period after the fuel change. Recent studies (Křůmal et al. 2019, 2021) combusted the same type of boiler (overfire boiler) fuel, which is regularly used in practice for heating. Knowledge is lacking about gaseous and particle emissions for the combustion of fuels for which an overfire boiler is certified (such as coke). Our research aimed to present the results showing a decrease in pollutant emissions (CO, OGC, PAHs, TSP) from the combustion of fuels similar to coke (such as smokeless coal). Research efforts should focus on high quality, innovative fuels due to demands for pollutant emissions decrease. Application of new smokeless fuels in household boilers could increase the local air quality after the fuel change. It is important to show the data about pollutant emissions from innovative fuels.

This paper compares the chemical composition of the emissions from combustion in a household boiler of five types of new smokeless coal fuel prepared by Polish producers. Smokeless solid fuels were obtained in the process of thermal hard coal devolatilization conducted in a dedicated furnace. The fine fractions of the smokeless coal were used to form briquettes and pellets. For comparison, the test was also performed for unprocessed hard coal, which was used as a reference in this study. The different shapes and forms of these types of fuels have not yet been compared and described. There is a potential to find the most cost effective and the most environmentally friendly coal fuel which could be processed and used in practice. The type of reactor is not novel as we applied the old-type boiler which is, however, widely used in households and is suitable for tested coal fuels similar to coke in their parameters. The emissions were characterized for the content of gaseous pollutants: NO_x, SO₂, CO, and OGC, as well as CO₂, TSP, PM₁₀, PM_{2.5} and PAHs. The obtained values were discussed and compared with data in European standards to verify the potential use of these fuels for heating.

The research was conducted in the Department of Chemical Engineering and Processes, West Pomeranian University of Technology in Szczecin, Poland, and the Energy Research

Centre, VSB-Technical University of Ostrava, Czech Republic, within the period of February–October 2019.

Materials and methods

Experimental setup for the combustion tests

The same design of a hot-water overfire boiler was used for all combustion tests. An overfire boiler was used because smokeless coal fuels cannot be used in any other boiler type (automatic, gasification, downdraft boiler). The reason is that the tested fuels have very low contents of volatile matter (except for hard coal, which was used for the comparison), and the characteristics of smokeless coal fuels are similar to those of coke. A schematic diagram and photograph of the boiler are shown in Figure S1 in the supplementary file. The boiler consists of cast iron cells connected to form a combustion chamber, an ashpan and a double-pass exchanger heater. The boiler was connected to the water-cooling system. The fuel was placed in one large batch onto a solid grate. The boiler was equipped with primary and secondary combustion air supplies. The primary combustion air, F_{A1} , was supplied through a water-cooled grate to the fuel layer and was automatically regulated by a thermostatic valve, while the secondary combustion air, F_{A2} , was supplied in front of the first exchanger draft.

The burning fuel, F_G , was in direct contact with a heat exchanger, which formed the boiler walls. Cooling water was supplied to the heat exchanger, F_{W1} , from which hot water, F_{W2} , was discharged into the water-cooling system.

According to the boiler manufacturer's characteristics, the boiler was designed for coke, hard coal and wood burning in the conventional burn-through process. The boiler was produced in output from 27 to 32 kW based on fuel and with 5 cells. The technical parameters of the boiler declared by the manufacturer are presented in Table S1 in the supplementary file.

The scheme of the measuring apparatus is shown in Fig. 1. The boiler was connected to the insulated chimney in accordance with EN 303-5:2012 (CEN 2012), which was used for testing the fuels. Flue gas removal was ensured by the dilution tunnel, and the dilution air was sucked from the laboratory. The temperatures of the input and output heating water were measured using a PT100 resistance sensor, while the flue gas temperatures behind the boiler and in the dilution tunnel were measured using a K-type thermocouple. The water flow was measured using a Krohne electromagnetic flowmeter.

Characteristics of the fuels

Six types of solid fuels (Tables 1, 2) consisting of standard and innovative smokeless fuels were examined in this study. Smokeless solid fuels were obtained in the process of high-temperature pyrolysis of noncaking coal in a grate furnace.

Fig. 1 Scheme of measuring apparatus

For the production of smokeless fuel, an installation for the production of carbon reductants is used. During the production process, smokeless fuel or carbon reductant is the main product, and heat is also generated, which is recovered in a heating boiler and sold as superheated steam to the neighboring chemical plant. The detailed method of coal modification cannot be introduced because of the manufacturer's know-how protection. To compare the properties of fuels, briquetting and pelleting were used to prepare samples made of smokeless char fines. Both technologies of agglomeration are applicable for Polish manufacturers.

The samples were named smokeless coal (SC), smokeless briquette 1 (SB1), smokeless briquette 2 (SB2), smokeless briquette 3 (SB3), smokeless pellets (SP) and unprocessed hard coal (HC). Smokeless briquettes 1, 2 and 3 differed among themselves in the content of one key component. Smokeless briquette 1 was modified by polymer—phenolic resin, smokeless briquette 2 was modified by starch and smokeless briquette 3 was modified by carboxymethylcellulose (CMC). The smokeless pellets contained starch.

All types of fuel were air-dried on the metal plate in the testing laboratory prior to the measurement and analysis. Fuel analyses were performed in accordance with the following standards: (ISO 2006, 2009, 2010a,b,c, 2011, 2013). Different shapes and sizes of solid fuels can be observed in the pictures presented in Figure S2 in the supplementary file. The smallest fuel particle diameters start from 7 mm for smokeless coal, while the largest diameters start from 90 mm for hard coal. Smokeless briquettes had the most regular shape, while the fractions of smokeless coal and hard coal had the least regular shape. Smokeless coal had smaller grains and the additional grate had to be inserted for no falling down of smokeless coal to ash tray. The exception was smokeless coal briquettes.

Gaseous and particulate matter emission measurements

Concentrations of the following components, CO, CO₂, NO (the catalytic convertor changed NO₂ to NO), O₂, OGC, PAHs, PM₁₀ and PM_{2.5}, were measured during tests. A

Table 1 Results of physicochemical analysis of six tested fuels (raw state)

	Unit	SC	SB1	SB2	SB3	SP	HC
Carbon	%	87.6	79.3	78.5	76.9	79.9	68.0
Hydrogen	%	1.06	1.67	1.90	1.63	2.14	4.33
Nitrogen	%	1.45	1.36	1.41	1.31	1.43	1.69
Sulfur	%	0.33	0.54	0.51	0.83	0.42	0.34
Oxygen	%	1.60	3.61	5.90	4.75	6.18	10.1
Ash	%	4.94	8.26	7.55	9.99	5.15	6.60
Total moisture	%	2.99	5.29	4.19	4.56	4.78	9.03
Analytical moisture	%	2.55	4.70	3.80	4.06	4.75	8.41
Volatile matter	%	4.65	9.92	13.3	11.2	12.9	32.8
Fixed carbon	%	87.42	76.53	74.96	74.25	77.17	51.57
Net calorific value	MJ/kg	30.7	28.3	28.3	27.6	28.9	26.3
Gross calorific value	MJ/kg	31.0	28.8	28.8	28.1	29.5	27.5

Table 2 Results of physicochemical analysis of six tested fuels (dry basis)

	Unit	SC	SB1	SB2	SB3	SP	HC
Carbon	%	90.3	83.7	82.0	80.6	83.9	74.7
Hydrogen	%	1.09	1.76	1.98	1.71	2.25	4.76
Nitrogen	%	1.49	1.44	1.47	1.37	1.50	1.86
Sulfur	%	0.34	0.57	0.53	0.87	0.44	0.37
Oxygen	%	1.65	3.81	6.16	4.98	6.49	11.1
Ash	%	5.09	8.72	7.88	10.5	5.41	7.26
Volatile matter	%	4.79	10.5	13.8	11.8	13.6	36.1
Fixed carbon	%	90.1	80.8	78.3	77.8	81.0	56.7
Net calorific value	MJ/kg	31.8	30.0	29.7	29.1	30.5	29.2
Gross calorific value	MJ/kg	32.0	30.4	30.1	29.4	31.0	30.2

nondispersive infrared (NDIR) analyzer was used to determine the concentrations of CO, CO₂ and NO. A paramagnetic analyzer was applied to estimate the concentration of O₂. The concentration of the organic gaseous compounds was measured by a flame ionization detector (FID). The gas sampling was continuous with a time interval of 1 s. All of the analyzers used were calibrated prior to taking measurements using calibration gases. To estimate the PM (TSP), PM₁₀ and PM_{2.5} distributions, flue gas samples were taken in the dilution tunnel at sampling point 2, as shown in Fig. 1. Multistage impactors with ranges below PM_{2.5}, PM_{2.5}–PM₁₀ and above PM₁₀ were used for sampling. The impactor is shown in Figure S3 in the supplementary file.

Flue gas samples were taken in the dilution tunnel by isokinetic sampling in the middle of the flue gas stream. Measurements of PAH concentrations were taken in an accredited laboratory by the GC-HRMS method in accordance with applicable standards and regulations (EPA 1997, 1999, 2012; ISO 2003) and with the help of the sampling system (filtration-condensation apparatus) shown in Figure S4 in the supplementary file. The average standard deviations of emission factors of gaseous pollutants were for CO₂ (1.8%), CO (20.9%), NO_x (33.3%), SO₂ (29.8%), OGC (33.5%), particulate pollutants PM (31.2%) and organic compounds (39.0%) (Křůmal et al. 2019).

Test conditions

Combustion tests were performed in a hot-water overfire boiler. There was no reason to conduct the tests with different types of boilers because tested fuels (coke-type fuels) are intended only for overfire boilers. The boiler was already presented in “[Experimental setup for the combustion tests](#)” section. The boiler was operated according to the operating manual and the requirements of the standard EN 303-5:2012 (CEN 2012). A sufficient amount of the fuel containing beech wood and the chosen type of briquette was burned to create a suitable basic layer of glowing ash and to heat the boiler to its operating temperature of approximately 50 °C. Boiler controls were set to achieve a nominal heat output. The combustion air intake was controlled automatically by a simple thermoregulator, which reacts to the water temperature in the boiler. After the preheating period, a fuel charge sufficient for the maximum filling height of the boiler was added into the combustion chamber. The differences in fuel loads were caused by different bulk densities and granulometries of the tested fuels. The combustion period was finished each time when the fuel layer reached the level of the basic layer of glowing ash before the combustion period. The

test duration and heat output depended on the fuel masses added into the combustion chamber and the pressure loss of the fuels.

Results and discussion

Coal fuels composition

The results of analyses performed in the laboratory for six solid-fuel samples are summarized in Table 1 (raw state) and Table 2 (dry basis). The lowest carbon content was found in the case of hard coal (68.0%), for which the hydrogen content of 4.33% was the highest. The nitrogen content in all solid fuels was at a comparable level. Significant differences in oxygen content were noted for the tested fuels. The lowest amount of oxygen was found for smokeless coal (1.60%), while the highest was found for hard coal (10.1%). Quite significant differences in the amount of sulfur were also found, and a particularly high level (0.83%) was noted for smokeless briquette 3, which also had the highest ash content (9.99%). As expected, the highest number of volatiles was measured for the hard coal. The content of volatiles in the case of hard coal was threefold greater than that in the other tested fuels. The moisture content was 2.99% for smokeless coal, approximately 4–5% for smokeless briquettes 1–3 and 9.03% for hard coal. In general, for comparison, wooden fuels are considered dry with a moisture content of 15%, while higher moisture contents represent wet fuel. The highest net calorific value was 30.7 MJ/kg obtained for smokeless coal, while the lowest value was 26.3 MJ/kg obtained for hard coal.

The coal fuel parameters were compared with those of other similar fuels published previously—hard coal (Křůmal et al. 2021) and smokeless coals (Šćicčžko et al. 1993; Lasek et al. 2019) in Table 3 (dry basis). The hard coal parameters of this study and Křůmal et al. (2021) show quite good matches. Smokeless coal analysis from the literature (Šćicčžko et al. 1993; Lasek et al. 2019) is limited because elemental analysis is missing. The calorific values of our smokeless coal (SC) are similar to those reported by (Lasek et al. 2019).

Gaseous emissions

Conducted combustion tests of six solid fuels in the overfire boiler provided extensive measurement data (also in Table S2 in the supplementary file). The general characteristics of the operational conditions of the combustion process are summarized in Table 4. The collected measurement data allowed us to assess the boiler efficiency

Table 3 Results of physicochemical analysis of published coal fuels (dry basis)

	Ref	Křůmal et al. (2021)	Lasek et al. (2019)				Ściażko et al. (1993)
	Unit	HC	HC1	HC2	BC1	BC2	SB
Carbon	%	75.19	–	–	–	–	–
Hydrogen	%	5.14	–	–	–	–	–
Nitrogen	%	1.46	–	–	–	–	–
Sulfur	%	0.62	–	–	–	–	–
Oxygen	%	9.59	–	–	–	–	–
Ash	%	8.00	4.2	2.9	4.0–6.4	4.1–5.1	13.7
Volatile matter	%	37.36	32.7	32.2	3.94–10.54	3.32–8.53	8.3
Fixed carbon	%	54.64	63.1	64.9	–	–	78
Net calorific value	MJ/kg	29.79	30.80	31.85	30.69–31.64	31.26–32.02	27.84
Gross calorific value	MJ/kg	30.89	31.84	32.89	31.03–31.96	31.59–32.32	28.86

HC hard coal, BC blue coal (smokeless coal), SB smokeless briquettes

Table 4 Operational conditions of the combustion process for six tested fuels

	Unit	SC	SB1	SB 2	SB 3	SP	HC
Combustion period	h	4.4	4.2	4.1	3.8	3.7	5.3
Atmospheric pressure	mbar	985	988	991	990	979	989
Ambient air temperature	°C	30.5	27.9	29.3	26.8	24.4	27.9
Ambient air relative humidity	%	43.5	26.6	38.1	50.0	55.9	54.3
Heat output	kW	20.2	22.5	21.2	19.8	17.0	15.2
Fuel consumption	kg/h	3.87	4.24	4.69	4.03	3.84	2.69
Water flow temperature	°C	58.8	60.4	60.4	58.0	54.9	52.1
Water return temperature	°C	47.1	47.5	48.1	46.6	45.1	43.4
Flue gases temperature	°C	244	313	295	282	222	230
O ₂ concentration in dry flue gases	%	8.1	8.5	7.8	7.5	9.2	9.1
Air excess	–	1.62	1.69	1.59	1.56	1.78	1.77

Fig. 2 Comparison of boiler thermal efficiency for six tested fuels. The loss through radiation, convection and conduction has been estimated in the laboratory based on their experience

Table 5 Emissions of CO, NO_x, SO₂, OGC, CO₂, PM_{2.5}, PM₁₀ and TSP for the six tested fuels

	Unit	SC	SB1	SB2	SB3	SP	HC
CO	g/kg	623	238	206	553	670	200
NO _x	g/kg	1.69	1.20	1.25	1.21	1.46	2.82
SO ₂	g/kg	3.80	4.33	4.54	5.95	2.48	4.69
OGC	g/kg	1.05	5.15	3.45	1.72	12.4	72.6
CO ₂	g/kg	2.67×10^3	2.63×10^3	2.58×10^3	2.19×10^3	2.34×10^3	2.31×10^3
PM _{2.5}	g/kg	0.275	1.75	0.776	9.18	4.74	30.0
PM ₁₀	g/kg	0.351	1.88	1.07	9.71	5.42	31.1
TSP	g/kg	0.427	2.18	1.48	10.0	6.19	31.6

NO_x is expressed as NO₂

depending on the type of fuel used and to determine the characteristic concentrations of pollutants: CO₂, CO, NO_x, SO₂, OGC and PAHs. The obtained results are presented in Fig. 2 and Tables 4 and 5. The longest combustion time was found when using hard coal (5.3 h), while the shortest time was found when using smokeless pellets (3.7 h). Notably, Table 4 shows that the lowest fuel consumption and heat output were recognized for the hard coal, equal to 2.69 kg/h and 15.2 kW, respectively. The operational conditions of the combustion process for the six tested fuels are shown in Table 4.

The highest boiler efficiency of 77% was found for hard coal, which resulted from the lowest total losses (23%) achieved with this standard type of fuel. The highest total losses (45%) were recorded when smokeless pellets were used.

The loss through sensible heat of the products of combustion is directly related to the amount of flue gas produced (oxygen excess) and the temperature of flue gases (the difference between the temperature of flue gases and the laboratory temperature). The higher the excess O₂ and the temperature are, the higher the resulting loss will be.

The loss through incomplete combustion depends on the amount of flue gas and the concentration of CO in the flue gas.

The loss through radiation, convection and conduction depend on the size of the surface of the individual boiler walls and the difference between the boiler temperature surfaces and the laboratory temperature. The temperature of the boiler surfaces is given by the temperature of the water in the boiler and the quality of insulation used.

The loss through unburned fuel in ash is an analysis of the mass of solid residues after combustion (ash) and the C content in these residues (combustible matter). The procedures referenced to the test standard EN 303-5:2012 were applied.

The emissions of CO, CO₂, OGC, NO_x, SO₂, TSP, PM₁₀ and PM_{2.5} for all types of fuels are presented in Table 5.

The highest CO emission was found in the case of smokeless pellet combustion. Moreover, equally high emissions of CO were also noted for smokeless briquette 3 and smokeless coal. The lowest value of CO emission among smokeless fuels was recognized for smokeless briquette 2.

Major factors influencing the complete combustion of carbon include the fuel reactivity, fuel fineness and particle size, fuel/air mixing efficiency, excess air available for complete combustion, residence time and temperature profile inside the boiler (Tawil 2021). No obvious correlation holds between flue gas temperatures and CO emission factors; however, by excluding HC from the data set, CO decreased with increasing flue gas temperature for smokeless coals (Figure S5 in the supplementary file). As reported in studies of biomass combustion (Vakkilainen 2017), combustion at a high furnace temperature and a long residence time decrease CO emissions. There is no direct correlation between fuel characteristics such as volatile matter and CO emissions factors (Figure S6 in the supplementary file).

From the comparison of the emissions of NO_x, SO₂ and OGC, the worst result was obtained for unprocessed fuel—hard coal. The amount of NO_x emission for smokeless briquettes 1–3, smokeless coal and smokeless pellets was twice as low as the emission from hard coal on average. The emission of SO₂ was on average 25% lower for smokeless fuels except smokeless briquette 3 in comparison with that from hard coal. In general, the lower sulfur content in fuels tested during the combustion process also resulted in a lower SO₂ emission and a lower SO₂ concentration in exhaust gases. Similar conclusions were drawn by Křůmal et al. (2019), who showed that combusted coal also contained a much higher amount of sulfur, ash and nitrogen than, for example, biomass (wood or wood pellets) and caused higher emissions of sulfur dioxide for a given boiler type. Yang et al. and Zhao et al. (2016) proved that the SO₂ and NO_x emissions were closely related to

the sulfur and nitrogen contents of the fuel. Both of these components remain in the boiler as ash or are released by the chimney as emission.

The largest differences were noted in OGC emissions. The emission of OGC was 69-fold lower during smokeless coal combustion compared to hard coal combustion. Moreover, the OGC emissions were 42-, 21-, 14- and sixfold lower during combustion of smokeless briquette 3, smokeless briquette 2, smokeless briquette 1 and smokeless pellets, respectively, compared to hard coal combustion.

PM emissions

The dominant source of $PM_{2.5}$ emissions in the EU-27 from small sources (the domestic sector) is the combustion of solid biomass, which contributed nearly three quarters of the total emissions, while the share of hard coal was approximately 25%. Sixty-nine percent of $PM_{2.5}$ emissions came from residential stoves, followed by single-family house boilers (16%) and fireplaces (11%) (Cofala and Klimont 2012). Therefore, the determination of PM_{10} and $PM_{2.5}$ contents was important for the six solid fuels chosen in this study.

The values of PM_{10} and $PM_{2.5}$ were over 100-fold lower for smokeless coal, 35-fold lower for smokeless briquette 2, 18-fold lower for smokeless briquette 1 and sixfold lower for smokeless pellets than for hard coal combustion. Only threefold lower values of PM_{10} and $PM_{2.5}$ were obtained during smokeless briquette 3 combustion. Considering the current legislation in the EU, $PM_{2.5}$ emissions from small sources must decrease by more than 20% until 2020, by 40% by 2030 and by 50% by 2050. Smokeless briquettes 1 and 2, as well as smokeless coal and smokeless pellets, meet these conditions.

Dust in flue gases can be removed by electrostatic precipitators from small-scale combustion devices. Filters and catalysts are not suitable for the adsorption of dust as underburned products, as they would be clogged after a short time. Electrostatic precipitators are now being used in small-scale combustion devices, especially in automatic boilers, but not in manually batched boilers. For our tested boiler (overfire), the price of the electrostatic precipitator would include a notable part of the boiler price.

Gaseous and particle emissions were studied previously, e.g., by Křůmal et al. (2019). Herein, wood biomass (dry and wet) combusted in the overfire boiler had lower values than coal in the studied parameters (such as SO_2 , CO, OGC and PM). However, OGC and TSP emissions are higher from combustion of biomass than from combustion of our smokeless coal samples. Sixteen EPA PAHs were described in the study of Křůmal et al., but benzo[a]pyrene emissions were also higher from biomass combustion than from smokeless coal combustion. The gaseous and particle emissions of hard

Table 6 Comparison of the emissions of NO_x , SO_2 , CO, CO_2 , OGC, TSP and BaP for six tested fuels with different solid fuels (Křůmal et al. 2019, 2021)

g/kg	Křůmal et al. (2019)				Křůmal et al. (2021)				Smokeless coal (overfire boiler)					
	Reduced output (40–60%)				Reduced output (average = 60%)				Reduced output (average = 67%)					
	B1-WW	B1-DW	B1-BC	B1-BCB	B1-HC	B1-BC	B1-HC	B1-HC	SC	SBI	SB2	SB3	SP	HC
NO_x	0.405	1.16	1.2	1.06	2.36	2.02	2.43	2.43	1.69	1.2	1.25	1.21	1.46	2.82
SO_2	nd	nd	9.97	3.7	10.2	1.7	9.24	9.24	3.8	4.33	4.54	5.95	2.48	4.69
CO	74.7	112	419	239	260	105	253	253	623	238	206	553	670	200
CO_2	1.01×10^3	1.53×10^3	1.53×10^3	1.79×10^3	2.52×10^3	1.79×10^3	2.46×10^3	2.46×10^3	2.67×10^3	2.63×10^3	2.58×10^3	2.19×10^3	2.34×10^3	2.31×10^3
OGC	27.6	33.9	85.5	48.8	149	20.2	57.7	57.7	1.05	5.15	3.45	1.72	12.4	72.6
TSP	15.4	18.7	81.6	28.7	89.4	3.13	17	17	0.427	2.18	1.48	10	6.19	31.6
BaP	1.04	2.75	5.2	3.27	8.55	0.83	6.37	6.37	0.017	0.439	0.229	0.037	1.29	5.35

B1 overfire boiler, WW wet wood logs, DW dry wood logs, BCB brown coal, BCB brown coal briquettes, HC hard coal, BaP benzo[a]pyrene

coal in an overfire boiler with similar heat output, as presented in Table 5, are compared with Table 6 (Křůmal et al. 2019, 2021). The emissions from the combustion of hard coal in the overfire boiler are comparable except for SO_2 , while our tested coal contained less sulfur in fuel.

CO and OGC are products of incomplete combustion. They are quite well correlated with TSP (Figure S7 in the supplementary file), except CO emissions from HC. This result proves that the quality of combustion is dependent not only on the type of combustion unit but also on the type of fuel.

The tested coal fuels SB1–SB3 have the same size. The smallest coal fuel is SC, followed by SP. The largest coal fuel is HC; however, it is a mixture of large and small pieces, which makes the fuel inhomogeneous. This causes the use of large coal fuels (HC, SB1–SB3) in the

overfire boiler to not increase the pressure drop so much. The airflow is not limited significantly, which leads to lower CO concentrations. This corresponds to the results in Table 5, except SB3, which corresponds to higher TSP concentrations.

The volatile matter in coal fuels (Tables 1, 2) influences the OGC concentrations, which was confirmed for HC (the highest volatile matter with the highest OGC) and SC (the lowest volatile matter with the lowest OGC). Volatile matter is also related to PAH concentrations.

PAHs content

For the overall quality assessment of smokeless fuels, the emission of PAHs is important. In Tables S3–S4 in the supplemental file, all emission factors of PAHs for all tested

Fig. 3 Emission of benzo[a]pyrene, 4 PAHs_{LRTAP} and 16 PAHs for six tested fuels

fuels are shown. PAHs were sampled according to ISO 11338-2 (ISO 2003). The 16 analyzed EPA PAH compounds were naphthalene, acenaphthylene, acenaphthene, fluorene, phenanthrene, anthracene, fluoranthene, pyrene, benzo[a]anthracene, chrysene, benzo[b]fluoranthene, benzo[k]fluoranthene, benzo[a]pyrene, indeno[1,2,3-cd]pyrene, dibenzo[a,h]anthracene and benzo[g,h,i]perylene.

The highest $\sum 4$ PAHs_{LRTAP} emissions (benzo[b]fluoranthene, benzo[k]fluoranthene, benzo[a]pyrene and indeno[1,2,3-cd]pyrene) were from hard coal combustion.

Table 7 Emission factors of PAHs for the smokeless briquette 1

		PAHs in PM	PAHs in the gaseous phase
		$\mu\text{g}/\text{kg}_{\text{fuel}}$	$\mu\text{g}/\text{kg}_{\text{fuel}}$
Naphthalene	NaP	1.9×10^1	2.2×10^2
Acenaphthylene	Acy	1.0×10^1	2.7×10^3
Acenaphthene	Ace	0.90	3.7×10^2
Fluorene	Fl	1.8×10^1	7.2×10^3
Phenanthrene	Phe	1.9×10^2	8.6×10^3
Anthracene	Ant	3.8×10^1	3.3×10^3
Fluoranthene	Flut	1.3×10^3	1.5×10^3
Pyrene	Py	1.2×10^3	1.0×10^3
Benzo[a]anthracene	BaA	1.1×10^3	4.2×10^1
Chrysene	Chr	6.0×10^2	3.0×10^1
Benzo[b]fluoranthene	BbF	5.8×10^2	2.8
Benzo[k]fluoranthene	BkF	1.9×10^2	1.3
Benzo[a]pyrene	BaP	4.4×10^2	2.3
Indeno[1,2,3-cd]pyrene	IP	2.1×10^2	1.2
Dibenzo[a,h]anthracene	DBA	7.4×10^1	0.40
Benzo[g,h,i]perylene	BghiP	3.3×10^2	1.5
$\sum 4$ PAHs (LRTAP)		1.4×10^3	7.6
$\sum 16$ PAHs		6.3×10^3	2.5×10^4

These emissions were sixfold higher than those for smokeless pellets, 13-fold higher than those for smokeless briquette 1, 29-fold higher than those for smokeless briquette 2, 90-fold higher than those for smokeless briquette 3 and 218-fold higher than those for smokeless coal. The $\sum 4$ PAHs_{LRTAP} emissions from the tested fuels are presented in Fig. 3 (LRTAP—long-range transboundary air pollution).

Notably, Fig. 3 shows that the highest emissions of $\sum 16$ PAHs were recorded for the hard coal. The lowest $\sum 16$ PAH content was obtained for smokeless coal.

The highest emission of benzo[a]pyrene also came from hard coal (Fig. 3). The measured benzo[a]pyrene emission was fourfold lower during smokeless pellet combustion and 12-fold lower during smokeless briquette 1 combustion. The lowest benzo[a]pyrene emission was measured during smokeless coal combustion. It was 315-fold lower than that of hard coal combustion.

The sample results of the emission factors of PAHs for smokeless briquette 1 are presented in Table 7, and the percent contributions of gaseous and particulate PAHs to the total PAHs are presented in Fig. 4. The largest PAH emissions in PM were fluoranthene, pyrene, benzo[a]anthracene, chrysene and benzo[b]fluoranthene. The largest PAH emissions in the gaseous phase were fluorene and phenanthrene. As reported in the literature (Holoubek 1996), phenanthrene, fluoranthene and pyrene are the predominant PAHs emitted from the combustion of fossil fuels.

All results of emission factors and the contribution of gaseous and particulate PAHs to the total PAHs for all tested fuels are presented in Table S5 in the supplementary file.

There is no uniform linear correlation between 16 PAHs (in total) and the CO of smokeless coal fuels (Figure S8 in the supplementary file). SC, SB3 and SP have similar CO emissions (in the range of 553×10^3 – 670×10^3 mg/kg), while SB1, SB2 and HC have approximately fourfold lower CO emissions than SC, SB3 and SP (206×10^3 – 238×10^3 mg/

Fig. 4 Percent contribution of gaseous and particulate PAHs in total PAHs for smokeless briquette 1

kg). A good linear correlation holds between 16 PAHs (in total) and OGC for smokeless coal fuels (Figure S9 in the supplementary file). A good linear correlation holds between 16 PAHs (in total) and TSP for smokeless coal fuels (Figure S10 in the supplementary file), except for SB3.

The type of emitted toxic organic substances depends on the fuel characteristics, boiler type and operating parameters. Operating parameters include temperature, residence time, fuel particle size and fuel/air mixing rate. Concentrations of VOCs quickly decrease with increasing temperature, efficient fuel mixing and adequate residence time (Arslan and Özdalyan 2020). From our results of OGC concentration, volatile matter is directly proportional to OGC (or 16 PAHs) emissions, which means that these parameters are dependent on fuel characteristics (Figure S11 in the supplementary file). Considering flue gas temperature, there is no obvious correlation with OGC (or 16 PAHs) emissions (Figure S12 in the supplementary file). In our case (overfire boiler), there is a small and cold combustion chamber and short heat exchanger, which means a short residence time and little space for the sufficient oxidation of products of incomplete combustion.

Evidently, the combustion of hard coal produced a larger proportion of 16 PAHs. By increasing the maturity of coal, the number of aliphatic linkages (alkanes) decreases, and the degree of aromaticity (PAHs) increases (Oros and Simoneit 2000; Křůmal et al. 2013).

The obtained PAH data were compared with those of other studies.

Comparison with data presented by the European Monitoring and Evaluation Programme/European Environmental Agency (EMEP/EEA) and other studies

The total emissions of 4 PAHs (BbF, BkF, BaP and IP) for the tested fuels were compared with the data published by EMEP/EEA (2019). For all tested fuels, the EFs of each of the 4 PAHs are below the reference values presented by EMEP/EEA for hard and brown coals (Table 8). The EF results of 4 PAHs measured for unprocessed hard coal combustion were very close to those presented by EMEP/EEA, except for that of benzo[k]fluoranthene.

The results of EF BaP (mg/GJ) from other studies are similar to the values from smokeless coal combustion presented in Table S4 in the supplementary file. Another study (Knigawka et al. 2020) describing smokeless coal fuels presents the results of PAH emissions.

Gaseous and particle emissions in overfire boiler, which was used in our study, were investigated previously, e.g., by Křůmal et al. (2019). Herein, wood biomass (dry and wet) combusted in the overfire boiler had lower values in studied parameters than hard and brown coal (such as SO₂, CO, OGC and PM). However, OGC and PM emissions are higher from biomass combustion than our smokeless coal samples. Not 16 EPA PAHs were described in these studies but benzo(a)pyrene emissions was as well higher from biomass combustion than from smokeless coal combustion, see Table S6. Therefore, the application of smokeless coal combustion may lead to decrease in gaseous and particulate emissions in small-scale combustion units and to increase the local air quality.

Toxicity equivalent (TEQ)

The toxicity equivalent, TEQ, for the emission of a total of 16 PAHs and a total of 12 PAHs was calculated according to a previous publication (Horak et al. 2017). The TEQs for 16 PAHs and 12 PAHs followed the order of smokeless coal < smokeless briquette 3 < smokeless briquette 2 < smokeless briquette 1 < smokeless pellets < hard coal (Table S3 in the supplementary file). All new types of smokeless fuel had a TEQ lower than the previously determined TEQ from the combustion of biomass (dry and wet spruce wood) and lignite, tested in the same combustion unit and a similar heat output (Horak et al. 2017).

Bottom ash comparison

The briquettes stayed quite compact during the whole combustion period. The briquettes did not crumble much; however, some unburnt fuel was found in the ashpan, as shown in Figure S13 in the supplementary file.

Table 8 EFs (mg/GJ) of 4 PAHs compared with the reference presented in the EMEP/EEA Guidebook (EEA 2019) and other literature (Ścicżko et al. 1993; Lasek et al. 2019)

	[mg/GJ _{fuel}]	BbF	BkF	BaP	IP
EMEP/EEA Guidebook (EEA 2019)	Solid fuel except for biomass	250	100	270	90
	Hard coal and brown coal	330	130	230	110
Lasek et al. (2019)	BW from WK1			9–18	
	BW from WK2			3–12	
Ścicżko (1993)	Smokeless coal			7.33	
The present paper	Smokeless coal	1.5	0.32	0.56	0.37
	Smokeless briquette 1	20	6.6	16	7.6
	Smokeless briquette 2	8.1	2.5	8.1	4.1
	Smokeless briquette 3	3.8	0.76	1.4	1.6
	Smokeless pellets	39	13	44	19
	Hard coal	320	86	200	100

BW blue coal (smokeless coal), WK hard coal

Fig. 5 Ashpan after the combustion tests

For all tested fuels, a specific amount of ash fell through the grate of the boiler into the ashpan. Accumulated ash from subsequent samples is shown in Fig. 5. Ash generated during combustion differs in grain size. Ash from smokeless coal was the most uniform in size, while ash from smokeless briquette 3 was the most diverse.

Conclusion

Research efforts should focus on the development of high-quality, innovative fuels because of demands for a decrease in pollutant emissions. The use of new smokeless fuels in household boilers could improve the local air quality. Data must be collected, and knowledge must be developed about pollutant emissions from innovative fuels. Therefore, the emission factors of gaseous and particulate pollutants from the combustion of new types of smokeless coal fuels in one type of hot-water boiler were analyzed. Substitution of unprocessed hard coal with smokeless fuels, such as smokeless coal, reduced NO_x , SO_2 and OGC emissions by 1.7-fold, 1.2-fold and 69-fold, respectively. The OGC emission factor

(1.05 g/kg) was the lowest for smokeless coal. Smokeless coal had the smallest fuel particle diameters. The influence of different grain sizes of coal was not the aim of this study. The obtained results confirm that the application of smokeless briquettes 1 and 2, as well as smokeless coal, reduced the $\text{PM}_{2.5}$ emissions at least 18-fold in comparison with hard coal, thus improving the environmental performance. $\text{PM}_{2.5}$ emissions were the lowest for smokeless coal (0.28 g/kg), followed by smokeless briquette 2 (0.78 g/kg) and smokeless briquette 1 (1.8 g/kg). Therefore, these fuels meet the expectations of EU legislation for decreasing $\text{PM}_{2.5}$ emissions from small combustion sources by more than 20% until 2020, by 40% by 2030 and by 50% by 2050.

An improvement in $\sum 4$ PAH and $\sum 16$ PAH emissions was also observed when replacing hard coal with smokeless solid fuels. $\sum 4$ PAHs (0.086 mg/kg) and $\sum 16$ PAHs (3.54 mg/kg) were the lowest for smokeless coal and the highest for smokeless pellets from the smokeless coal fuel group. Smokeless solid fuels named smokeless briquettes 1–3 and smokeless coal seem to maintain their low emissions with the fuel quality at the same time. Composition analyses and emission tests proved that the tested processed

types of fuels meet the requirements established in Polish regulations. Thus, it would be efficacious to replace the commonly used unprocessed hard coal with smokeless solid fuels for environmental and public health benefits.

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Declarations

Conflict of interest The authors declare that there are no conflicts of interest regarding the publication of this article.

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