

Assessment and Analysis of the Soot Load Emitted from Hard Coal Combustion in the Area of a Selected Settlement Unit

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ABSTRACT

Currently, the soot load from solid fuel combustion is not measured or counted, because it is included as part of particulate matter emitted from combustion sources. However, recent reports indicate that after carbon dioxide, soot is the most important contributor to current climate change. Therefore, an attempt was made to assess the soot load that is emitted during the winter season from individual heat sources where hard coal was burned or co-fired. Soot emission analysis and assessment were carried out in the selected settlement unit. Soot was collected monthly throughout the heating period at the chimney flue outlet, and analyzed for the PAH content. From these results and the information obtained from the users of individual heat sources, the soot load emitted from the installation in question during the entire heating period and the load of the sum of 16 PAHs and benzo(a)pyrene also during the entire heating period were calculated. It was found that the PAH content in soot largely depends on the type of boiler, the form of hard coal and wood addition burned, and the age of the boiler. The soot load, on the other hand, depends on the amount of hard coal burned, the type of boiler, how the combustion process is carried out, etc.

Keywords: soot, combustion, solid fuels, winter season, PAHs.

INTRODUCTION

Combustion of coal is inextricably linked to the formation of harmful components (by-products) (Kowalczyk 2021). However, the way the coal is burned, not just the fuel itself, has a major impact on the appropriate efficiency of energy transformations and the reduction of the negative impact on the environment. The composition of the fuel has a significant impact, but the conditions under which the combustion process proceeds is also important (Olszewski et al. 2012; Machnik et al. 2015). The effect of hard coal combustion is both carbon-graphite dust and soot. Both of these types of dust are responsible for air pollution (Szatyłowicz and Skoczko 2019).

The amount of gas or dust emissions into the air from the combustion of hard coal in boilers with a thermal capacity of up to 5 MW can be calculated

by determining the emissions of individual pollutants into the air, i.e. substances such as carbon dioxide, nitrogen oxides, carbon monoxide, sulfur dioxide, benzo(a)pyrene, or dust from fuel combustion. These values should also include the soot emission, which is emitted during the combustion of coal in boilers (Kubica et al. 2016).

During coal combustion in boilers with manual fuel feed and operating on natural draught, each fuel feed leads to a temporary cooling of the furnace and burning of the fuel to amorphous carbon, i.e. soot. An important factor here is the properly selected excess air factor, which according to the literature (King 2014) should be 1.8; unfortunately, practice shows that it is often around 3. This is related to the fact that the higher the excess air coefficient, the greater the blow-through of the furnace with the excess cold air. This leads to faster cooling of the furnace, which results in more soot emissions.

Soot is the finest fraction that is solid, rather than gaseous. According to the definition of dust, including TSP (total suspended dust), PM10, PM2.5, and PM0.1, dust is an air pollutant that consists of a mixture of solid and liquid particles, suspended in air, that are a mixture of organic and inorganic substances (Skotak et al. 2016; Kubica et al. 2016; Zhan et al. 2019). Therefore, the soot from the combustion of solid fuels is also included in the total dust. Some of it is emitted into the atmosphere, but some of it is retained in the flue gas ducts. According to the National Centre for Balancing and Emission Management, soot emissions from combustion processes outside the industry, i.e. combustion from individual heat sources, represent 22.3% of the total soot emissions to the atmosphere (Figure 1).

Furthermore, due to its physical and chemical properties, soot is seen as a significant pollutant that causes climate change. Not only does soot contribute to higher ground-level air temperatures, but it also disrupts the hydrologic cycle of the Earth by indirectly influencing the melting of the glaciers. Soot particles absorb solar energy, which is then released as heat (Skotak et al. 2016; Lohmann et al. 2020; Kurfeng et al. 2022). Given the above, it was considered appropriate to assess and analyze load emissions from individual heat sources burning or co-firing hard coal in various forms in the selected settlement unit, as soot is an important source of PAHs, including the most dangerous for health benzo(a)pyrene; it seems appropriate to determine them. For this purpose, 5

sampling points in Choroszcz were selected, and soot samples were taken for analysis throughout the heating period. On the basis of the tests and calculations, the amount of soot emitted per kilogram of fuel burned and the amount of PAH compounds emitted from individual heat sources during the heating season were determined.

MATERIAL AND METHODS

Soot samples were collected between October 2017 and March 2018 at selected measurement points, at 5 local heat sources in the Choroszcz municipality, where hard coal in various forms was burned. At each point, soot deposited in the chimney at the exhaust outlet was collected for laboratory testing. Table 1 shows the characteristics of the soot sampling points. Polyethylene plastic containers (total volume approx. 135 ml) were used for soot sampling, which did not further contaminate the sampled materials. Soot was collected from the inner walls of the chimney flue. A blunt tool was used for sampling to facilitate the removal of the soot from the wall surfaces. The containers filled with the test material were tightly closed and signed according to the point of collection, and transported to the laboratory. Table 1 presents the information obtained from users of heat sources regarding the combustion installation, fuel, type of boiler, etc.

The soot samples of 3 g were then extracted twice with a solvent mixture of hexane-acetone at

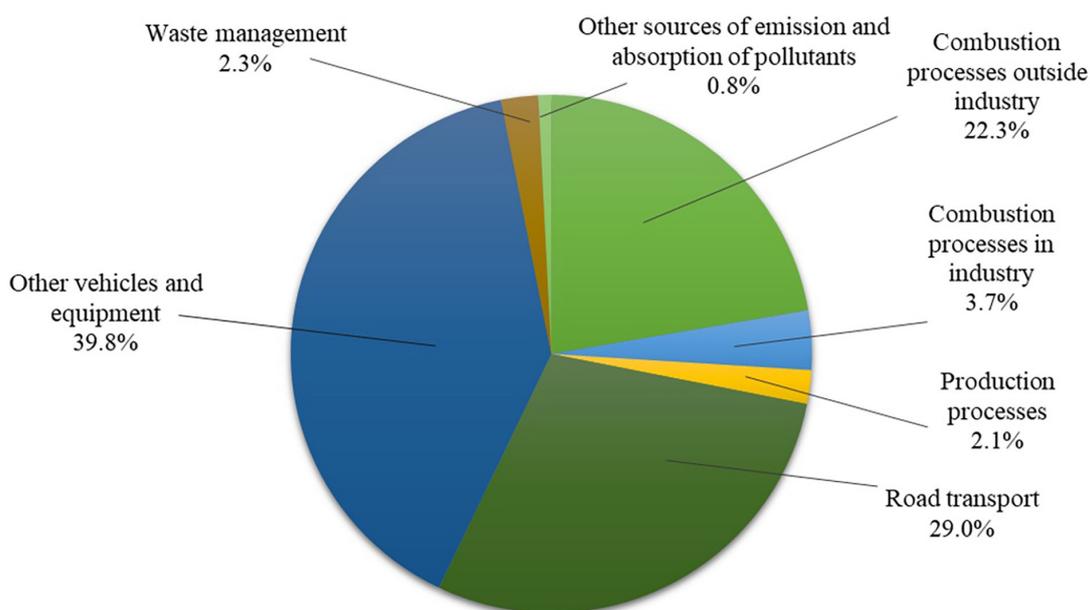


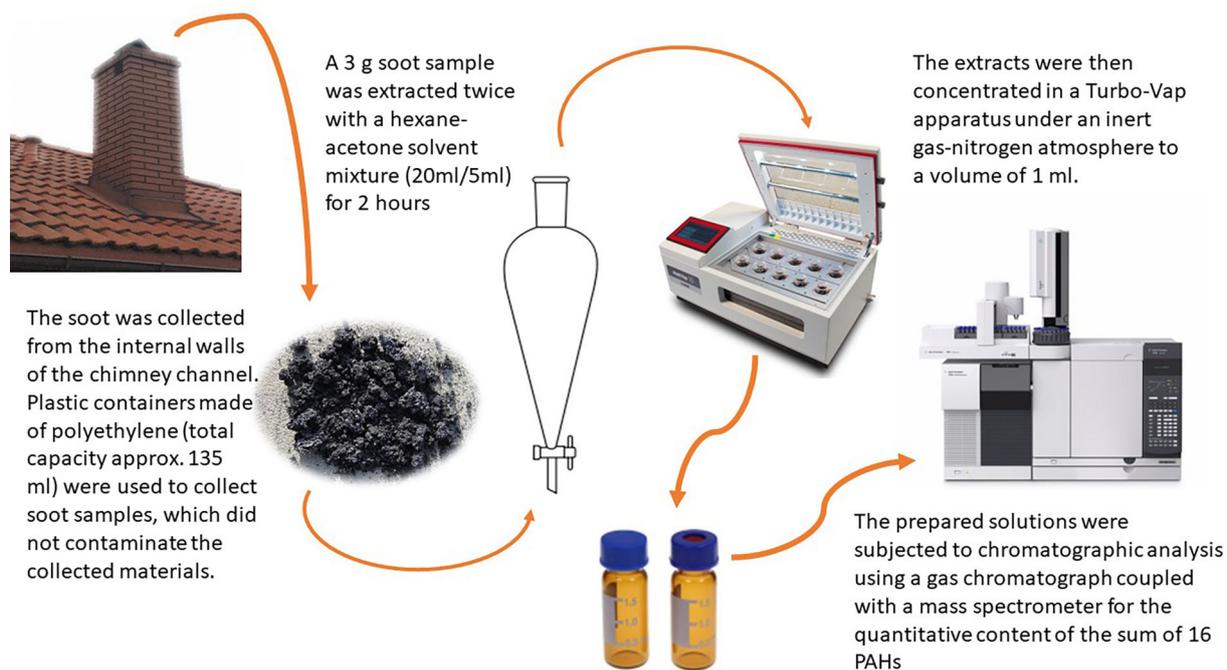
Figure 1. Share of significant sectors in soot emissions (KOBIZE 2019)

Table 1. Characteristics of soot sampling points

Number	1	2	3	4	5
Fuel type	Wood, hard coal	Hard coal	Eco-pea coal	Wood, hard coal	Eco-pea coal
Boiler type	Traditional boiler with an open combustion chamber	Boiler with a lower combustion chamber - charging	Automatic charging boiler	Boiler with a lower combustion chamber - charging	Boiler with a lower combustion chamber - charging
Start of boiler operation	1997	2000	2010	2011	2014
Boiler power [kW]	17,5	20,5	17	21	19
Chimney type	Internal	Internal	Internal	Internal	Internal
Chimney height [m]	8.5	12	11	9	5
Fuel consumption [t, m ³]	2,5 t hard coal, 14 m ³ wood	7 t hard coal	3 t	3 t hard coal, 6 m ³ wood	5 t

a ratio of 20 ml/5 ml for 2 hours. Acetone and the polar compounds extracted with it were removed by washing twice with 10 ml of deionized water. The remaining organic phase was dried over anhydrous sodium sulfate. The extracts were then concentrated in a Turbo-Vap apparatus under an inert gas-nitrogen atmosphere to a volume of 1 ml. The solutions thus prepared were subjected to chromatographic analysis using a coupled gas chromatograph (Agilent GC/MS Triple Quad 7000 C) equipped with a split/splitless manifold and a 30 m × 0.25 mm HP-5MS capillary column with a 0.25 µm thick layer with a mass spectrometer for the quantitative content of the sum of 16 PAHs: naphthalene, acenaphthylene, acenaphthene, fluorene, phenanthrene, anthracene,

fluoranthene, pyrene, benzo(a)anthracene, chrysene, benzo(b)fluoranthene, benzo(k)fluoranthene, benzo(a)pyrene, dibenzo(a,h)anthracene, benzo(g, h, i)perylene and indeno(1,2,3-c, d)pyrene. The American Environmental Protection Agency (US-EPA) indicates that the 16 above-mentioned PAHs as particularly dangerous (Szatyłowicz and Skoczko 2019). The scheme of sample collection and analysis is shown in Figure 2. During the extraction, no internal standard was added, while the method was controlled by simultaneously carrying out the entire analytical procedure for a sample of certified reference material – Clean soil Reference Material EDF-5183 (CERILLIANT Analytical Reference Standards). In the tests, external calibration was

**Figure 2.** Scheme for soot sampling and preparation

employed, performed by using calibration curves for individual compounds. The calibration solutions were prepared from a standard mixture of 16 PAHs (AccuStandard, Z-014G) (Szatylowicz and Skoczko 2019).

The soot emissions from the combustion of fuels without waste gas scrubbing were then calculated from equation 1:

$$E = B * w \quad (1)$$

where: E – pollutant emissions [kg];
 w – lift factor [kg/unit of fuel];
 B – the amount of fuel burned [unit solid fuel: Mg, oil: m³, gas: 10⁶ m³].

To calculate the ratio for hard coal, formula 2 was used from the Information and instructional materials of the Ministry of Environment Protection, Natural Resources and Forestry series 1/96 Warsaw from April 1996.

$$w = 0.05 * A' \quad (2)$$

where: A' – ash content expressed as a percentage.

The average ash content in hard coal was assumed to be equal to 15%, which is the ash content given by producers and distributors of hard coal-based solid fuels. The calculations above were performed for the content of total PAHs for the content of benzo(a)pyrene in carbon black collected at 5 collection points in Choroszcz.

RESULTS AND DISCUSSION

Table 2 summarizes and presents the averaged results of the sum of PAHs and benzo(a)pyrene in the entire heating period against 5 consumption points. At all measurement and control points, hard coal was burnt or co-fired in various forms to a greater or lesser extent. The highest averaged content of benzo(a)pyrene and the sum of PAHs was

Table 2. Mean and standard deviation of benzo(a)pyrene and total PAHs in carbon black from the whole heating season (October–March)

Sampling point	Benzo(a)pyrene [mg/kg]	Total PAHs [mg/kg]
1	20±14.32	1437.91±584.68
2	23±20.21	1214.33±1152.44
3	2±1.55	498.17±424.72
4	72±49.04	2292.89±1292.45
5	14±12.45	1274.92±1185.97

found in the soot collected from point 4, where hard coal with a cube size of more than 60 mm was burnt.

Similarly, the highest emission of soot was found (Table 2) in point 4, the sum of which was 2,292.89±1,292.45 mg/kg. The lowest content of the sum of PAHs and benzo(a)pyrene was determined in the black carbon samples during the entire heating period at the third measurement and control point, which was 498.17±424.72 mg/kg. Throughout the research period, in terms of the highest PAH content, the 4th consumption point was characterized by the use of mixed fuel (coal and wood). At this point, the local heat source was a boiler with a lower combustion chamber – a charging boiler from 2011. This source emitted the greatest amount of PAH into the atmosphere through the soot.

By analyzing the obtained results of benzo(a)pyrene in soot (Table 2) in the individual tested solid fuel combustion installations, it can be concluded that the highest amount of it during the entire heating period was obtained at measuring point 4 and amounted to 72±49.04 mg/kg. The smallest amount of benzo(a)pyrene in the soot was released into the atmosphere during the entire heating period by heating installation no. 3. According to the Tissari research group (2007), among the studied PAH emission sources, such as baking ovens, sauna ovens, and conventional brick ovens, which were combustible with

Table 3. Mass of soot, the sum of 16 PAH, and benzo(a)pyrene emitted during the whole heating season (October–March)

Sampling point	Amount of soot emitted [kg soot/heating season]	Amount of PAHs emitted to the atmosphere from coal combustion [mg/kg coal]	Amount of benzo(a)pyrene emitted to the atmosphere [mg/kg coal]
1	1.875	2.70	37.50
2	5.25	6.40	120.75
3	2.25	1.12	4.50
4	2.25	5.16	162.00
5	3.75	4.80	52.50

various types of wood, the highest emission of the 15 PAHs (apart from naphthalene) was about 202 mg/kg d.m. of soot, the sauna heater was fired with aspen and alder firewood. Peng et al. (2016) also conducted research on PAH emissions from the co-incineration of municipal solid waste. They noticed that the emission of the sum of 16 PAHs was 179.57–3616.96 mg/kg d.m. in the case of burning coal alone, and in the case of co-incineration of coal with municipal waste, it was: 120.49–2402.17 mg/kg d.m. The results obtained in the research are consistent with the obtained data. Table 3 summarizes the total amount of soot that is emitted to the atmosphere from the burnt fuel mass during the entire heating period. Hard coal was burnt or co-fired in various forms at all soot collection points. In addition, the total amount of the sum of 16 PAHs and the amount of benzo(a)pyrene emitted to the atmosphere at individual collection points are also presented.

The highest amount of soot in the whole period (Table 3) was emitted into the atmosphere from the 2nd soot collection point, this amount was equal to 5.25 kg p.m. At this point, there was a boiler with a bottom combustion chamber – batch-charged from the year 2000. This boiler burned 7 tonnes of hard coal during the heating season. The smallest load of soot was released into the atmosphere at intake point number 1, which was a traditional open combustion chamber boiler from 1997. At this point, 2.5 tonnes of hard coal and 14 m³ of firewood were burnt. The amount of soot formed is directly proportional to the amount of hard coal burned. The highest load of PAHs was introduced from the atmosphere at point number 2 and amounted to 6.40 g/kg of hard coal. A slightly smaller load, but also significant – in comparison with the remaining points – was introduced to the atmosphere at sampling point no. 4, amounting to 5.16 g/kg of hard coal. On the other hand, the highest benzo(a)pyrene emissions were emitted to the atmosphere at sampling point no. 4 and amounted to 162 mg/kg of hard coal. According to the data collected and studies carried out, it was noted that the PAH and anaerobic pyrene loads are not proportional to the amount of hard coal burned. The content of the sum of 16 PAHs in carbon black and their emission via carbon black to the atmosphere depends on many factors. On the basis of the conducted research and data from the literature, a large variation in the emission of these compounds can be observed. This is influenced by: the material burned,

the type of burner, and the combustion conditions (temperature, residence time, moisture content, and oxygen supply). The largest sources of PAHs in the atmosphere are solid fuel combustion processes in individual furnaces, which are characterized by widespread use, intermittent operation, lack of air pollution control devices, and lack of condition monitoring (Dat and Chang 2017).

CONCLUSIONS

Soot is also discussed as a factor changing the Earth's climate. Initially, the contribution of soot was treated very marginally and all attention was focused on carbon dioxide as the main contributor to climate change. Recent studies by American and Chinese scientists show that soot is the next most important driver of current climate change after carbon dioxide (Myhre et al. 2013). An additional feature of BC (black carbon) is that it absorbs sunlight due to its very well-developed specific surface area (soot grain size is about 0.03 µm and the specific surface area determined by the BET method is 70–100 m²/g) (Skotak et al. 2016). The absorbed energy is released in the form of heat and thus contributes to warming the air, resulting in accelerated melting of ice and snow, according to scientists. Furthermore, soot affects the climate change through such processes as cloud formation, water evaporation, and the disappearance or formation of precipitation (Schmidt, 2011; Anenberg 2012; Kubica et al. 2016).

Soot is a source of PAHs, and among them, benzo(a)pyrene is the most threatening; they are the most commonly studied pollutants in environmental samples. This is primarily dictated by their negative health effects, including toxicity, carcinogenicity, and mutagenicity (Kim et al. 2013; Jyethi et al. 2014; Kaminska et al. 2016). It is estimated that in Poland the majority, over 80%, of benzo(a)pyrene in the air comes from households – from cookers and coal-fired boilers, especially those that are outdated and of a low standard (commonly referred to as *kopciuchy* or *junk*) (Pytliński 2014).

The monitoring of polycyclic aromatic hydrocarbons in combustion residues from solid fuels, such as soot, has not been analyzed in detail so far, and studies conducted so far have mainly considered the observation of changes in PAH compounds in flue gases. The results obtained indicate that the soot load and the content of

total PAHs and benzo(a)pyrene from individual furnaces is influenced by the amount and type of coal burned or co-fired, as well as the type and quality of the boiler in which the combustion is carried out. The conducted research can be the basis for further work related to the utilization of residues from the combustion of solid fuels with activators, where the content of polycyclic aromatic hydrocarbon groups or individual compounds from this group is increased. Soot can be formed during the combustion of any fuel containing carbon. Therefore, it is necessary to prevent or at least reduce this phenomenon.

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