

PLJEVLJA LIGNITE CARBON EMISSION CHARACTERISTICS

by

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The anthropogenic emission of GHG especially CO₂ has to be limited and reduced due to their impact on global warming and climate change. Combustion of fossil fuels in the energy sector has a dominant share in total GHG emissions. In order to reduce GHG emission, European Union established a scheme for GHG allowance trading within the community, and the implementation of the European Union emission trading scheme, which is a key to GHG reduction in a cost-effective way. An important part of emission trading scheme is prescribed methodology for monitoring, reporting, and verification of the emission of GHG including characterization of the local fuels combusted by the energy sector. This paper presents lignite characteristics from open-pit mine Borovica-Pljevlja, which has highest coal production in Montenegro (>1.2 Mt per year), including evaluation of its carbon emission factor based on the laboratory analysis of 72 coal samples. Testing of the samples included proximate and ultimate analysis, as well as, net calorific value determination. In accordance with the obtained results, linear correlations between net calorific value and combustible matter content, carbon content and combustible matter content, hydrogen content and combustible matter content, carbon content and net calorific value, were established. Finally, the non-linear analytical correlation between carbon emission factor and net calorific value for Pljevlja lignite was proposed, as a base for the precise calculation of CO₂ emission evaluation.

Key words: carbon emission factor, lignite characterization, laboratory analysis, GHG emission inventory.

Introduction

The anthropogenic emission of GHG, especially CO₂, together with tropical deforestation has to be limited and reduced due to their impact on global warming and climate change [1]. In 1992, United Nations Framework Convention on Climate Change (UNFCCC), International Environment Treaty, adopted in Rio de Janeiro, concluded that it is necessary to reduce the concentration of GHG in the atmosphere and thereby reduce their impact on the climate. As early as 1997, based on the Kyoto Protocol, commitments were made for developed countries in terms of reduction of GHG emissions for the period from 2008 to 2012. The UNFCCC conference held in Paris 2015, resulted in the consensus of all Parties, to give the best efforts to reduce GHG emission and limit temperature rise on the global level for no more than 1.5 °C. Factors that have a significant influence on GHG emissions, such as population, affluence,

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and technology, for the majority of world countries, have been presented [2]. The importance of each one depends on economic activity and technological development for a particular country. In addition to the listed factors, applied initiatives, policies and measures on the national and international level have a significant role. For 11 South-East European countries, review of implemented initiatives, policies and measures on the national level, in terms of GHG emissions and climate change, has been presented [3].

In EU series of legal documents are released in order to reduce emitted GHG, such as Directive 2003/87/EC on the establishing a scheme for GHG allowance trading within the community, and the implementation of the emission trading scheme (ETS), which is a key to GHG reduction in a cost-effective way. According to ETS, power plants and other plants, exceeding 20 MWth are covered by the scheme. The ETS scheme function under the cap and trade principle. Cap prescribes how much the total amount of GHG every energy plant within the ETS scheme can emit. Every plant has its own emission allowances, which can sell in case less emission than permitted, while a plant that exceeds the cap, must buy the emission allowances.

The initiatives, policies, and measures are pushing entities (countries) to adopt national strategies and plans. For Balkan countries, numerous publications and papers were published [4], on the topic on overall and sectoral mitigation analysis. Many authors pointed out *carbon leakage* within South-East Europe regional electricity market ETS scheme, due to the difference between ETS and non-ETS entities, which can delay the projected target 20-20-20, or even 40% GHG emissions reduction before the 2030 year [5].

The CO₂ is the most common GHG generated by human activities, accounting for about 60% of the increase in GHG emissions when compared to pre-industrial times. By far the largest source of CO₂ emissions represents carbon oxidation process occurring during fossil fuel consumption and accounting for 70-90% of total anthropogenic CO₂ emissions. During the fuel combustion process, most of the carbon contained in the fuel is emitted as CO₂. Some carbon is released as CO, CH₄ or non-methane hydrocarbons, which oxidize in the atmosphere to CO₂ over the period of few days up to 10-11 years.

In order to determine total amount of emitted CO₂ in the atmosphere by combustion of fossil fuels such as lignite, it is important to know total quantity of combusted fuel, efficiency of combustion process (oxidation factor) and characteristics of the fuel: net calorific value (NCV) and energy of burned fuel, or CEF value (ratio of carbon content to NCV of fuel in [t C/TJ]) which indicates how much carbon *i.e.* CO₂ is released into the atmosphere per unit of energy. Several different approaches were used for determination of CEF [6-9]. According to IPCC Guidelines for National GHG Inventories CO₂ emissions is calculated as the product of the fuel consumption, recommended value of NCV and the default value of CEF and oxidation factor for each fuel (coal) type. Such an approach is acceptable for most developing countries (if no other information is available) but characteristics of solid fuels for the same coal rank vary significantly between countries. For lignite NCV vary between 5.5-21.6 MJ/kg (default value 11.9 MJ/kg), and CEF between 24.8-31.3 t C/TJ (default value 27.6 t C/TJ) while for sub-bituminous coals NCV vary between 11.5-26 MJ/kg (default value 18.9 MJ/kg), and CEF value between 25.3-27.3 t C/TJ (default value 26.2 t C/TJ) [7]. This simplified assumption with the default fuel characteristics values is not suitable for precise determination of CO₂ emission. It is very important to point out that lignite CEF values from South Eastern Europe significantly differ from their default values defined by IPCC methodology [10-12]. The need for country-specific NCV and CEF values periodical determination and revision has been presented for Indian coals [13]. The example of China indicates that, based on data obtained from the energy sector, in the period 2002-2012, there was a decline in carbon content in used coals [14]. However, despite the carbon

content decline, emission in China is more than doubled for the same period, due to increased coal production. The stated fact from the practice, recommends the need for periodical coal testing on carbon content and consequently CEF determination. Most advanced methodology for the determination of fuel characteristics is according to Commission regulation 743/2014, which replaces annex VII of the regulation 601/2012: country-specific emission factors and other characteristics for the respective fuel or material should be experimentally determined based on default regular sampling and analysis. For example, for coal combustion thermal power plants like TPP Pljevlja 225 MWe, Montenegro, the minimum frequency for coal sampling and relevant analysis is at every 20000 t of used fuel or at least six times per year.

The laboratory experimental methodology that consists of ultimate and proximate coal analysis, net calorific value determination and calculations, described in this paper, authors implemented in their previously published paper [10], evaluating open-pit mine Kolubara, Serbia, lignite emission characteristics. For the first time, lignite from open-pit mine Borovica-Pljevlja, Montenegro, has been experimentally analyzed on such large scale, in order to determine its emission characteristics more accurately. At the same time, the validity of the methodology has been confirmed. The methodology can be applied later for emission characteristics determination of various coals. Certain improvements of the methodology can be implemented after, examining a larger scale of different types of coal samples and comparing the data with the previously published experiences *i.e.* [10-12].

In this paper results of laboratory analysis (including proximate, ultimate analysis and NCV determination) of 72 samples from open-pit mine Borovica-Pljevlja, Montenegro, are presented through obtained correlations: linear correlation between NCV and combustible matter content, linear correlation between carbon content and combustible matter content, linear correlation between hydrogen content and combustible matter content, linear correlation between carbon content and NCV, and non-linear correlation between CEF and NCV.

Laboratory analysis methodology

A sampling of Borovica-Pljevlja open-pit mine lignite samples for analysis has been done automatically for the purpose of the calibration of the instruments for on-line coal quality system, installed at TPP Pljevlja.

Samples were prepared according to ISO 5069, for the determination of moisture content and preparation of representative samples for proximate and ultimate analysis, while the determination of carbonate carbon content was done according to ISO 925.

Proximate and ultimate analyses were done in accordance with ASTM D7582-12, ASTM D5373-14, and ASTM D3176. For proximate analysis (determination of analytical moisture, ash and combustible matter content) thermogravimetric analyzer, LECO TGA 701 was used, while for ultimate analysis (the content of carbon, hydrogen, nitrogen and total sulfur) ultimate analyzer LECO CHNS 628 was used. Prior to laboratory measurement, all measuring equipment was checked and calibrated with certified referenced materials, supplied by LECO Company. A thermogravimetric analyzer was calibrated with a certified sample of coal with particulate smaller than 212 μm according to standards ISO 5068-2:2007 and ISO 1171:2010 while ultimate analyzer was calibrated with the referenced material sample, ethylene-diamine-tetra-acetic-acid (EDTA).

Determination of NCV was performed on calorimeter IKA 200, according to ISO 1928. Calorimeter calibration was done with certified benzoic acid pills with certified heating values. The calibration was performed prior to sample analysis. Achieved reproducibility was ≤ 64 kJ/kg between two consecutive measurements which is in good agreement with standard ISO 1928.

Achieved measurement repeatability values – r (the results of duplicate determinations, carried out at different times, in the same laboratory, by the same operator, with the same apparatus, on representative portions taken from the same test sample), were better than limits defined by used standards: for analytical moisture (in the range $W^a = 1.29$ -21.66%); $r < 0.21\%$ absolute, for ash (in the range $A^d = 2.93$ -16.73%); $r < 0.19\%$ absolute, for carbon (in the range $C^d = 54.9$ -84.7%); $r < 0.45\%$ absolute, for hydrogen (in the range $H^d = 3.25$ -5.10%); $r < 0.1\%$ absolute and for carbonate carbon content $r < 5\%$ relative.

Results and discussion

Characteristics of the lignite from open-pit mine Borovica-Pljevlje based on results of proximate and ultimate analysis are presented in tab. 1.

On the basis of the 72 analyzed samples from open pit-mine Borovica-Pljevlja, and obtained data values, five correlations have been established:

- between NCV at constant pressure and content of combustible matter,
- between carbon content and combustible matter content,
- between hydrogen content and combustible matter content,
- between carbon content and NCV at constant pressure, and
- between CEF and NCV at constant pressure.

Results of chemical analysis of tested samples from Borovica-Pljevlja open-pit mine have made it possible to carry out a linear correlation between NCV at constant pressure and combustible matter content presented in fig. 1. Since power plants furnaces operate at constant pressure, NCV of samples is calculated according to ISO 1928 for constant pressure. According to combustibles content in the samples, both raw and samples with analytical moisture were taken into account and presented in all figures. From fig. 1, it can be concluded that, in the investigated combustibles content range $>35\%$, there is an excellent agreement of experimental data with linear correlation, eq. (1), with high correlation coefficient value, $R^2 = 0.995$.

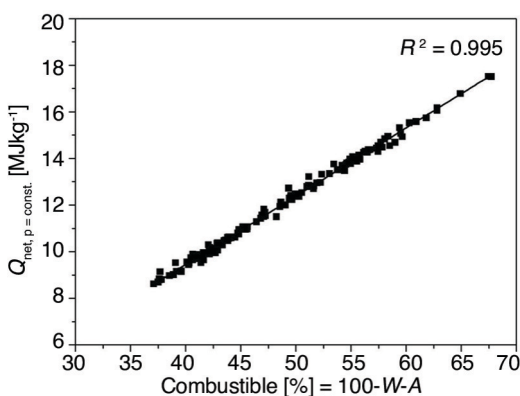


Figure 1. Linear correlation between the net calorific value and the combustible matter content of coal samples from Pljevlja open-pit mine

$$Q_{\text{net}, p = \text{const}} = 0.293 \cdot \text{Combustible} - 2.269 \quad (1)$$

Based on the tested samples from Pljevlja open-pit mine, experimental data of total carbon content vs. combustible matter content has been plotted in fig. 2. Experimental data are excellently correlated by, eq. (2), with correlation coefficient $R^2 = 0.991$:

$$C = 0.688 \cdot \text{Combustible} + 0.007 \quad (2)$$

Figure 3 presents the correlation between hydrogen content and combustible matter content in the samples of Borovica-Pljevlja

Table 1. Characteristics of the lignite from open-pit mine Borovica-Pljevlje

Parameter	Unit	As received	Dry basis
W	%	25.21-34.17	–
A	%	18.61-33.11	25.58-48.46
carbonate CO ₂	%	0.8-3.64	1.18-5.15
$Q_{\text{net}, p = \text{const}}$	MJ/kg	8.62-12.28	12.82-18.92
C_{tot}	%	25.36-34.66	34.82-49.81
H	%	1.99-2.71	2.85-3.96

lignite. Experimental results matching with linear correlation can be interpreted as well, but with a lower value of the correlation coefficient, $R^2 = 0.915$. The cause of higher deviation of the experimental results from correlated ones should be a matter of further analysis. Linearly correlated dependence of hydrogen content versus combustible matter content in Pljevlja lignite samples is presented by, eq. (3):

$$H = 0.054 \cdot \text{Combustible} + 0.049 \quad (3)$$

The experimental data and correlated linear dependency of total carbon content and NCV at constant pressure for 72 coal samples from Borovica-Pljevlja open-pit mine are presented in fig. 4.

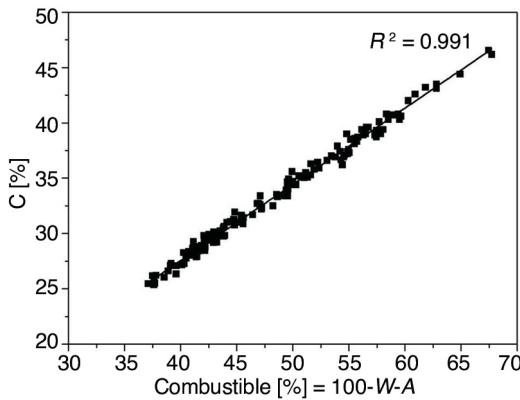


Figure 2. Linear correlation between the total carbon and the combustible matter content of coal samples from Pljevlja open-pit mine

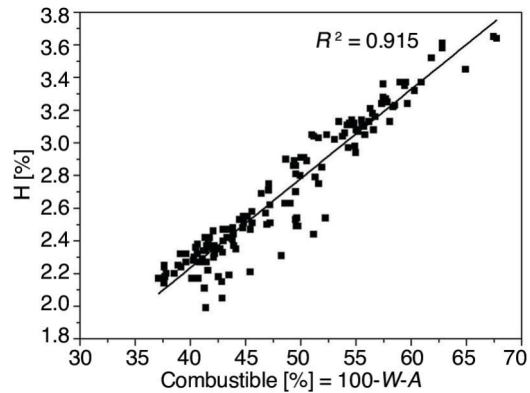


Figure 3. Linear correlation between the hydrogen and the combustible matter content of coal samples from Pljevlja open-pit mine

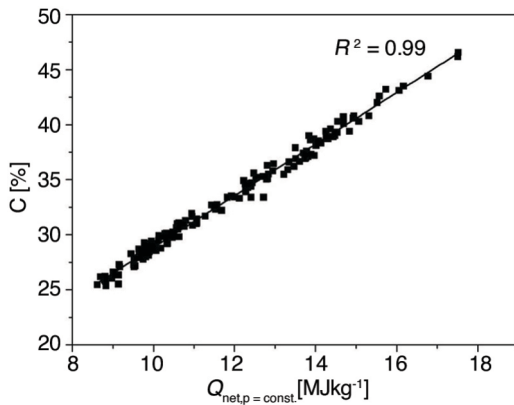


Figure 4. Linear correlation between the total carbon content and net calorific value of coal samples from Pljevlja open-pit mine

According to the diagram presented in fig. 4, it can be concluded that there is a good agreement between the experimental values and linear correlation eq. (4), with a high value of correlation coefficient, $R^2 = 0.99$.

$$C = 2.343 Q_{\text{net,p=const}} + 5.425 \quad (4)$$

Carbon emission factor as a ratio of the total carbon content to the NCV, is a characteristic of the fuel, which is used to calculate the amount of CO_2 released in the atmosphere per energy, by fuel combustion. According to the approach [6], it was proposed that the carbon emission factor for lignite has a default value of 27.6 t C/TJ. However, major variations in lignite quality, are followed by CEF variations too.

In order to determine CEF, the most appropriate correlation is the functional dependence of carbon content on NCV at constant pressure [10]. Analytical expression for CEF can be obtained, by dividing eq. (4), with NCV at constant pressure $Q_{\text{net,p=const}} [\text{MJkg}^{-1}]$, according to eq. (5):

$$\text{CEF} = 10 C / Q_{\text{net,p=const}} = 23.43 + 54.25 / Q_{\text{net,p=const}} \quad (5)$$

Comparative graphical presentation of CEF experimental values and calculated values according to eq. (5), is presented in fig. 5.

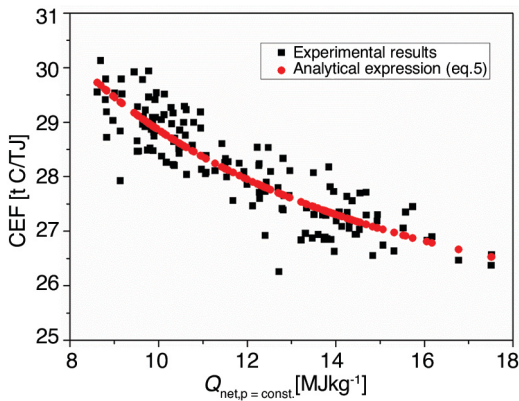


Figure 5. Comparative presentation of CEF values, for Borovica-Pljevlja lignite samples, obtained by experiments and analytical expression

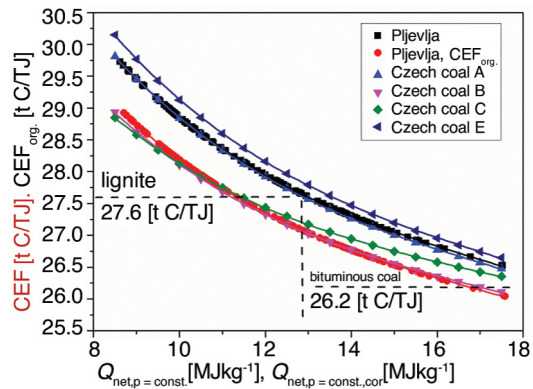


Figure 6. Correlation of the carbon emission factor and net calorific value for coal samples from Pljevlja open-pit mine and for Czech coal types [11, 12]

Figure 6 presents CEF correlation versus NCV for Borovica-Pljevlja lignite, according to eq. (5), correlations for three different sets of regional Czech coals and recommended correlation for European regional hard coals and brown coals [11, 12]:

- Set A - averaged coal data for bituminous coals with NCV 19.7-29.97 [MJkg⁻¹] and brown coal-lignite with NCV 9.52-17.72 [MJkg⁻¹] from the most important regional Czech coal districts (Moravia and Bohemia).

$$CEF = 23.33 + 55.11 / Q_{net,p=const} \quad (6)$$

- Set B - averaged coal data taken from an official Czech coal classification for bituminous coals with NCV 17.18-29.8 [MJkg⁻¹] and brown coal-lignite with NCV 9.36-23.86 [MJkg⁻¹].

$$CEF = 23.44 + 46.68 / Q_{net,p=const} \quad (7)$$

- Set C - experimental coal data based on proximate and ultimate analysis of coal samples from regional Czech district with 60 samples of bituminous coals and 17 samples of brown coal including lignite.

$$CEF = 24.0 + 41.23 / Q_{net,p=const} \quad (8)$$

- Set E - experimental coal data for European regional hard coals and brown coals-lignite.

$$CEF = 23.34 + 57.86 / Q_{net,p=const} \quad (8)$$

From fig. 6 one can conclude that non-linear correlation between CEF and NCV for Borovica-Pljevlja lignite, eq. (5), is equivalent to other cited/presented correlations and is especially close to correlation for set A coals. Still, there are differences in CEF due to: degree of coalification (coal age) and composition of the mineral matter in the coal.

Coals with a higher degree of coalification (hard coals and bituminous coals) have higher carbon content and lower oxygen and water content resulting in relatively higher NCV and lower CEF. On the opposite, younger coals (lignite) have lower carbon content but much higher oxygen and water content *i.e.* much lower NCV and as a result, higher CEF value. Sulfur content

in the coal has a positive influence on NCV resulting in lower CEF value compared to the same coal type but with lower sulfur content.

Chemical composition of mineral matter, for instance, the presence of carbonates could have an important influence on CEF value. Exposed to high temperature in the furnace, carbonates are decomposed, releasing CO₂. This calcination process is an endothermic reaction that requires heat to decompose chemical compounds (1786 kJ/kg CaCO₃) [15]. By combustion of a fuel with carbonates in the mineral matter, besides emission of CO₂ by organic carbon combustion, there is additional emission of CO₂ from carbonates (proportional to the content of carbonates in the coal) while real NCV is decreased (also proportional to the content of carbonates in the coal) due to endothermic calcination reaction. As a result fuels with increased carbonates content in the mineral matter has higher CEF compared with same fuel but without carbonates in the mineral matter. This is illustrated in fig. 7 where are plotted Borovica-Pljevlja lignite experimental data for C_{tot} vs. Q_{net,p=const} and calculated data for the same coals samples (with same ash content) but without carbonates: C_{org} [%] = C_{tot} [%] - 12 CO₂ [%]/44 vs. Q_{net,p=const,cor} corrected due to the heat of calcinations: Q_{net,p=const,cor} [MJkg⁻¹] = Q_{net,p=const} [MJkg⁻¹] + 4.059 [MJkg⁻¹] CO₂ [%]/100. In the calculated case of Pljevlja lignite samples without carbonates, the organic carbon emission factor would have lower values according to eq. (10), which would be much closer to eq. (7), for Set B representing averaged coal data taken from an official Czech coal classification. For the mean annual gross power generation (in period 2012-2014) at TPP Pljevlja of 1333 GWh/annum by combustion of 1655.33 kt/annum of Borovica-Pljevlja lignite with mean net heating value 9.15 MJ/kg and CEF = 29.359 t C/TJ, emission is 1.598 Mt CO₂/annum, while in the case of burning same lignite but without carbonates in the mineral matter, CEF would be 28.648 t C/TJ and emission would be 1.5591 Mt CO₂/annum, i.e. 2.4% less:

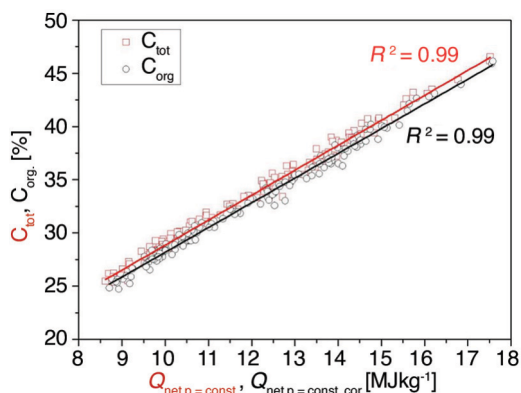


Figure 7. Correlation of the carbon content and the net calorific value for the same coal samples (same ash content) from Pljevlja open-pit mine, in the case with and without carbonates

$$CEF_{org} = 23.22 + 49.67/Q_{net,p=const,cor} \quad (10)$$

As a result of the aforementioned differences in coal samples characteristics, it could be concluded that there are differences in CEF values between coals from different regions/mines but also a difference in CEF values for the samples from the same region/mine, as seen in fig. 6. Due to that fact, determination of the GHG and CO₂ emission should be based on experimentally determined CEF correlation for each type of combusted coal, as is eq. (5) for open pit mine Borovica-Pljevlja lignite.

Conclusions

Based on proximate and ultimate laboratory analysis of 72 samples of open pit mine Borovica-Pljevlja lignite, the main coal emission characteristics for the improved determination of GHG-CO₂ emission are presented in the paper. Obtained linear correlation: between NCV and combustible matter content, between carbon content and combustible matter content, between hydrogen content and combustible matter content, between carbon content and NCV, and non-linear correlation between CEF and NCV at constant pressure are base for the precise calculation

of carbon *i.e.* CO₂ emission by Borovica-Pljevlja lignite combustion according to EU Regulation on Monitoring, Reporting and Verification [9], necessary for the ETS [8] implementation in Montenegro as a EU candidate country.

Obtained non-linear correlation of carbon emission factor and the net calorific value at constant pressure for open-pit mine Borovica-Pljevlja lignite is in excellent relation with analog correlations for regional Czech coal districts (Moravia and Bohemia) for bituminous and brown coal - lignite.

Analysis of the presented results of coal characteristics have shown that samples of open-pit mine Borovica-Pljevlja lignite has some content of carbonates in the mineral matter, which increases value of CEF (compared to same coal without carbonates) *i.e.* increases CO₂ emission during combustion of such coal due to endothermic calcination reaction of decomposition calcium-carbonate to CaO and CO₂ at elevated temperature in the boiler. Carbonate CO₂ content in the raw samples of open-pit mine Borovica-Pljevlja lignite is in the range CO₂^r = 0.8-3.64% (mean value 2.2%) which means that non-organic carbon in the raw coal is in the range C^r_{non-organic} = 0.218-0.99% (mean value 0.605%) while C^r_{tot} = 25.36-34.66%, (mean value 30.01%) corresponding to increased value of CEF in the range of 2.3% to 2.7% compared to CEF for the same coal without carbonates.

Based on presented results and analysis, determination of the GHG and CO₂ emission should be based on experimentally determined CEF correlation for each type of combusted coal, as is eq. (5) for open pit mine Borovica-Pljevlja lignite. These correlations should be periodically checked by frequent sampling followed by proximate and ultimate analysis, as it is prescribed by the EU Regulation on MRV [9].

Presented results indicate that exploitation of the lignite in open-pit mine Borovica-Pljevlja should be improved by selective excavation by which can be avoid inclusion of mineral matter from coal interlayers, resulting in decreased mineral matter and especially decreased carbonates content in the coal and increase its net heating value, *i.e.* giving a contribution to mitigate the impact on climate change (decrease in CO₂ emissions).

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Nomenclature

A	– ash content in the coal, [mass %]	$Q_{\text{net } p^{\text{const, cor.}}}$	– net calorific value at constant pressure corrected due to heat of calcination, [MJkg ⁻¹]
C	– carbon content in the coal, [mass %]	W	– moisture content in the coal, [mass %]
CEF	– carbon emission factor, [t C/TJ]	<i>Superscripts</i>	
CO ₂	– carbonate CO ₂ content in the coal, [mass %]	r	– as received (on raw coal bases)
Combustible	– combustible matter content in the coal, [mass %]	d	– dried (on dry coal basis)
H	– hydrogen content in the coal, [mass %]	<i>Subscripts</i>	
R^2	– coefficient of correlation, [-]	tot.	– total
$Q_{\text{net, } p^{\text{const}}}$	– net calorific value at constant pressure, [MJkg ⁻¹]	org	– organic

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