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Change in the Quality of Kolubara lignite Burned in the Thermal Power Plants "Nikola Tesla" Expressed in Terms of Emission Factor

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Abstract: The paper presents experimental investigation, calculations and analyzes of emission factors of low-quality domestic lignite from the Kolubara Basin, which is used for the operation of the Thermal Power Plant "Nikola Tesla". The studied lignite samples presented in this paper and their mutual comparison cover seven years of operation in the period 2012-2018. Standard analytical methods were used to determine the emission factor of lignite, including ultimate analysis, proximity analysis and determination of lower heating value. Based on the experimental results obtained, correlated dependencies of the emission factor of lignite were determined for the specified years and a comparison was made. From 2020, J.S.C. "Elektroprivreda Srbije", the operator of TPP "Nikola Tesla", imports coal of better quality, mixes it with Kolubara lignite and uses the mixture for firing the units of power plant B. A comparison was made between the lignite from Kolubara, which was offered for a long period of time, and the mixture with imported coal from last year. The change in the quality of domestic lignite, expressed as an emission factor, over time was presented together with the analysis of the change in the emission factor found for the coal blends used in recent years..

Keywords: Laboratory Analysis, Ultimate Analysis, Proximate Analysis, Carbon Emission Factor, Kolubara Lignite, Coal Blends.

1. Introduction

The methodology has been proposed at the international level (2006 IPCC Guidelines for National Greenhouse Gas Inventories. In order to harmonise the inventory of GHG emissions, some rafinements has been made – 2019 Refinement to the 2006 IPCC Guidelines for National Greenhouse Gas Inventories) [1-3]. Adopting for simplification, as already stated, predefined values for the net calorific value and the carbon emission factor (CEF) for fossil fuels, according to [2, 3], the proposed default value for the emission factor for lignite is 27.6 tC/TJ, and for the net calorific value for lignite is 11.9 TJ/Gg. However, the use of these default values leads to an error in the calculation of the GHG emissions inventory. For this reason, the international methodology [1-3] recommends the use of more accurate data. In the case of lignite from open-pit mines of the Republic of Serbia used for operation of utility boilers of TPP of the Joint Stock Company Electric Power Company of Serbia (EPS), the CEF and low temperature values deviate significantly from the recommended default values. The CEF values change over time, so it is necessary to determine them experimentally on a regular basis. Emission of Greenhouse Gasses in EPS for the Period from 1990 – 2008, has been presented in our internal publication [4], including complete data on emissions of pollutants from thermal energy plants within EPS for the specified period, as well as the methodology for calculating emissions and emission factors of used coals (mainly lignites). The calculation of the CEF for fuels for the production of electricity and thermal energy in the Republic of Serbia and the comparison with the IPCC standard values and other experimentally determined CEF values for lignite has been already presented in our previous work [5-13]. An alternative experimental methodology [4-6], has been described in detail, it was adopted and applied to determine the carbon emission characteristics of Serbian and Balkan lignites. The methodology has been established in the Laboratory for Thermal Engineering and Energy, VINČA Institute of Nuclear Sciences. The CEF is determined according to experimental measurements and calculations and the dependencies are established. It is found that the values of the emission factor are inversely proportional to the lower heating value, i.e. as the heating value increases, the value of the emission factor decreases and converges towards a certain value. Our group has a long time history investigating quality of the coals (lignites) used on TPPs in The Republic of Serbia and in the region (Bosnia and Herzegovina, Macedonia, Montenegro, Greece, Turkey etc.). The first publication was the internal one in which we presentented methodology in order to calaculate CEF factor based on experimental results and emission of greenhouse gasses from the utility boilers from Joint Stock Company Electric Power Company of Serbia, for the period 1990-2008, [4]. In 2011 and 2012 Evaluation of Kolubara Lignite Carbon Emission Characteristics has been presented, in Serbian [5] and in English [6]. The quality of Kostolac Lignite Carbon Emission Characteristics has been examined in 2017, [7]. Lignite from Montenegro (Pljevlja) and from Bosnia and Herzegovina (Lignite from Bogutovo selo) has been examined [9, 10] respectively. Carbon emission factor of Balkan lignites, including lignites from Serbia, Bosnia and Herzegovina, Montenegro, Turkey and Greece were investigated and mutually compared [11]. Variability of lignite from Kostolac basin for certain year period has been presented [12].

2. Laboratory methodology

In this work, samples mainly from the open-pit mine Kolubara for the the operation of Thermal Power Plant utility boilers "Nikola Tesla" were tested in the laboratory used. Kolubara lignite samples from 2015, 2017 and 2018 were tested in the laboratory, while the results of the 2012 test samples are presented and analyzed in our previous works [5, 6]. The laboratory testing of the samples from 2015 – 2018, were carried out, and also for samples with imported better quality coal from 2023, which represent a mixture of Kolubara lignite and imported coal. A total of 10 samples from 2015, 6 samples from 2017 and 7 samples from 2018 year, 4 samples of mixture from 2023 and 3 samples of imported better quality coal from 2023 were tested. As already stated in our previous works [5-7 and 9-12], sample preparation and laboratory analysis were performed at the Laboratory for Thermal Engineering and Energy of the"Vinca" Institute of Nuclear Sciences, National Institute of the Republic of Serbia, University of Belgrade, Department for fuel characterisation. Sample preparation was carried out prior to laboratory measurements in accordance with the standard ISO 5069-2: 1993. For the experimental laboratory tests, the samples were considered on an “as received”, “as determined” and “on dry” basis. Proximate analysis, ultimate analysis and determination of calorific value were performed in accordance with ASTM D7582, ASTM D5373 (Standard Test Methods for Determination of Carbon, Hydrogen and Nitrogen in Analysis Samples of Coal and Coke), ASTM D3176 (Standard Practise for Ultimate Analysis of Coal and Coke) and the standard ISO 1928: 2009, respectively. For experimental laboratory tests, LECO CHN628 ultimate analyzer, LECO CHN628 module S, for Sulfur content determination, Thermogravimetric analyzer LECO TGA 701 for proximate analysis and IKA C200 calorimeter has been used.

3. Experimental results

Experimental tests were performed for 10 Kolubara samples (year 2015), 6 samples (2017) and 7 samples (2018 year), 4 mixture samples (2023 year) and 3 samples of imported coal (2023 year), on an “as determined” basis, and the contents for the other two bases (“as received” and “dry”) were calculated from the first one. The contents of the Kolubara samples on an“as received” basis for years 2015, 2017 and 2018 are shown in the tables. 1., 2. and 3. respectively. Results of the experimental analysis for mixture samples are presented in table 4., and the results for the imported coal are presented in table 5. Results from previous test for Kolubara lignite for 2012 year were taken out from our previous work [5, 6]. Correlations were made on the basis of the results obtained: total Carbon content and combustible content, total hydrogen content and combustible matter content, total Carbon content and lower heating value, Lower heating value and combustible matter content and correlation between CEF and lower heating value. Based on correlations given, a comparison has been made for different years and different coal compositions and different type of coal.

As previously stated experimental investigation of the samples from 2015, 2017, 2018 and 2023 years were performed in laboratory environment. Experimental tests were performed on 27 samples, on an as determined basis. The other bases, as received and dry, were calculated from the first one. The contents of the samples on an “as received” basis,for the years 2015, 2017, 2018 and 2023 are presented in the Tables. 1., 2., 3. and 4, respectively.

Table 1. Table 1. The results of experimental test of lignite samples used for operation of TPP "Nikola Tesla". (Kolubara open-pit mine), "as received" basis, sampling year 2015

Sample Nº	Proximate analysis					Ultimate analysis				
	W	A	C_{fix}	Comb.	H_d	C	H	$S_{tot.}$	S_g	N + O
	[%]	[%]	[%]	[%]	[kJ/kg]	[%]	[%]	[%]	[%]	[%]
1	47.96	13.52	15.54	38.52	8447	25.09	2.16	-	-	11.27
2	47.86	13.89	15.24	38.25	8385	24.61	2.16	-	-	11.48
3	48.20	17.83	13.57	33.97	6920	21.89	2.29	0.35	0.10	9.68
4	47.13	17.91	13.30	34.96	7315	22.57	2.32	0.35	0.15	9.93
5	47.81	18.07	13.79	34.12	7135	22.18	2.27	0.28	0.14	9.54
6	47.49	18.42	13.48	34.09	7137	22.24	2.28	0.29	0.13	9.43
7	44.74	15.62	15.52	39.64	8866	26.43	2.19	0.34	0.14	10.87
8	50.97	18.26	7.72	30.77	6199	19.91	1.88	0.38	0.16	8.82
9	47.12	16.71	14.85	36.17	7842	23.85	2.08	0.41	0.15	10.08
10	45.69	22.56	12.50	31.75	6617	20.56	1.86	0.45	0.21	9.12

Table 3. The results of experimental test of lignite samples used for operation of TPP "Nikola Tesla". (Kolubara open-pit mine), "as received" basis, sampling year 2018

Sample Nº	Proximate analysis					Ultimate analysis				
	W	A	C_{fix}	Comb.	H_d	C	H	$S_{tot.}$	S_g	N + O
	[%]	[%]	[%]	[%]	[kJ/kg]	[%]	[%]	[%]	[%]	[%]
1	46.52	20.12	12.40	33.36	7177	21.63	1.98	-	-	9.76
2	47.70	17.92	13.60	34.38	7352	22.41	1.86	0.35	0.09	10.03
3	46.10	18.17	13.92	35.73	7532	23.50	1.99	0.35	0.12	10.11
4	45.70	21.90	12.69	32.40	7297	21.42	1.94	0.39	0.16	8.87
5	48.70	20.32	11.84	30.98	6382	20.33	1.95	0.49	0.22	8.48
6	48.10	18.97	12.79	32.93	7020	21.36	1.97	0.46	0.20	9.40
7	47.40	20.50	12.40	32.10	6706	20.99	1.97	0.43	0.19	8.96

Table 4. The results of experimental test of lignite samples used for operation of TPP "Nikola Tesla". (mixture of domestic lignite and imported better quality coal), "as received" basis, sampling year 2023

Sample Nº	Proximate analysis					Ultimate analysis				
	W	A	C_{fix}	Comb.	H_d	C	H	$S_{tot.}$	S_g	N + O
	[%]	[%]	[%]	[%]	[kJ/kg]	[%]	[%]	[%]	[%]	[%]
1	39.90	23.00	14.22	37.10	8131	24.53	2.12	0.49	0.42	10.04
2	39.84	19.44	15.72	40.72	9526	27.89	2.30	0.67	0.21	10.32
3	42.46	19.68	15.01	37.86	8312	24.24	2.15	0.36	0.09	11.37
4	50.33	15.84	12.95	33.83	7354	21.89	1.93	0.44	0.27	9.75

Table 5. The results of experimental test of samples of imported better quality coal, "as received" basis, sampling year 2023

Sample Nº	Proximate analysis					Ultimate analysis				
	W	A	C_{fix}	Comb.	H_d	C	H	$S_{tot.}$	S_g	N + O
	[%]	[%]	[%]	[%]	[kJ/kg]	[%]	[%]	[%]	[%]	[%]
1	40.74	4.24	24.39	55.02	13242	37.60	2.83	0.19	0.09	14.51
2	39.69	2.81	27.35	57.50	14789	40.24	2.88	0.37	0.09	14.29
3	44.87	5.10	22.71	50.03	12214	34.01	2.56	0.15	0.04	13.41

4. Calculations and comparison

According to the data presentented in the tables based on experimental tests and the calculations, linear dependences of the total Carbon content and combustible matter content has been calculated and presented, for each specified year:

2012: $$C = 0.7785 \cdot Combustible - 4.6405 \quad (1)$$

2015: $$C = 0.651 \cdot Combustible - 0.018 \quad (2)$$

2017: $$C = 0.651 \cdot Combustible - 0.089 \quad (3)$$

2018: $$C = 0.652 \cdot Combustible - 0.053 \quad (4)$$

2023: $$C = 0.660 \cdot Combustible - 0.087 \quad (5)$$

The experimental data of Carbon content on Combustible matter content are presented on diagram on Figure 1. Linear analytical correlations are presented by eq. (1-5), and graphically on diagram Figure 2., for the specified years and mutual comparison.

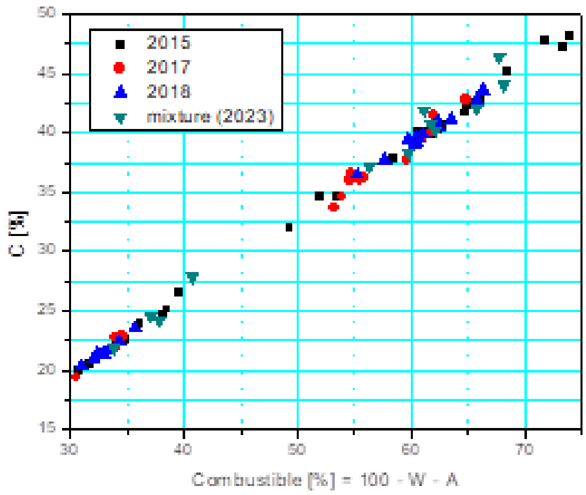

Figure 1. Total Carbon content as a function of combustibles content, for the samples of lignite, for 2015 – 2023 years, experimental data

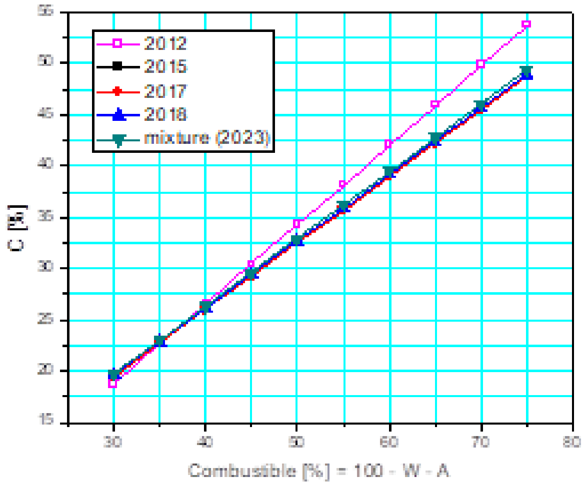

Figure 2. Total Carbon content as a function of combustibles content, for the samples of lignite, for 2012 – 2023 years, analytical data

According to the data presentented in the tables based on experimental tests and the calculations, linear dependences of the total Hydrogen content and combustible matter content has been calculated and presented, for each specified year:

2012: $$H = 0.0594 \cdot Combustible \quad (6)$$

2015: $$H = 0.059 \cdot Combustible + 0.067 \quad (7)$$

2017: $$H = 0.060 \cdot Combustible + 0.016 \quad (8)$$

2018: $$H = 0.057 \cdot Combustible + 0.069 \quad (9)$$

2023: $$H = 0.056 \cdot Combustible + 0.00004 \quad (10)$$

The experimental data of Hydrogen content on Combustible matter content are presented on diagram on Figure 3. Linear analytical correlations are presented by eq. (6-10), and graphically on diagram Figure 4., for the specified years.

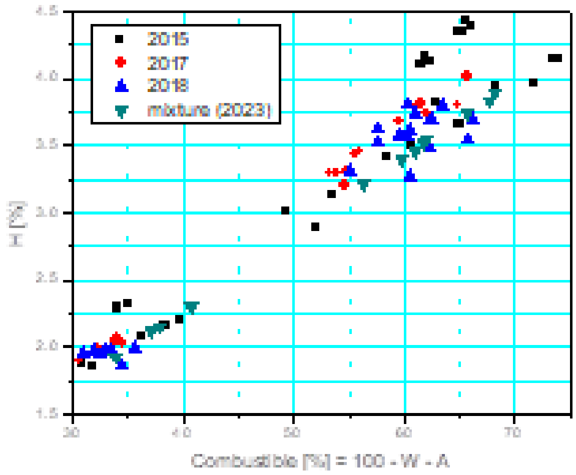

Figure 3. Total Hydrogen content as a function of combustibles content, for the samples of lignite, for 2015 – 2023 years, experimental data

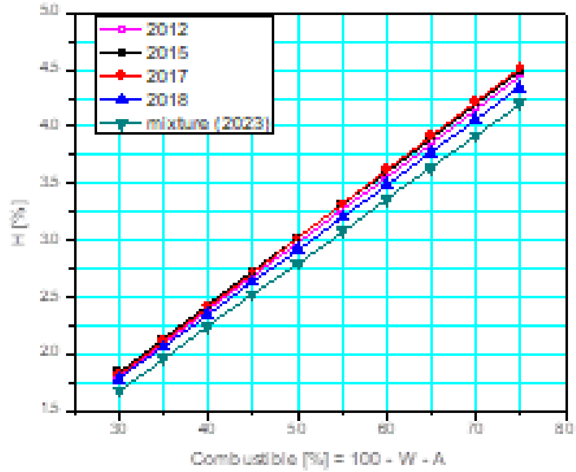

Figure 4. Total Hydrogen content as a function of combustibles content, for the samples of lignite, for 2012 – 2023 years, analytical data

The diagram in Figure 5 shows the experimental results of the total carbon content (%) as a function of the low heating value. The linear correlations for the years 2012 - 2023 are presented by dependencies, eq. (11-15), while the linear analytical correlations are presented together on the diagram on Figure 6. Based on the diagrams, a slight change in the dependency with the years is noticeable, which is especially clearly in the diagram in Figure 6, since the linear dependencies are almost superimposed over each other.

2012: $$C = 2.3718 \cdot H_d + 4.2637 \tag{11}$$

2015: $$C = 2.309 \cdot H_d + 5.639 \tag{12}$$

2017: $$C = 2.288 \cdot H_d + 5.571 \tag{13}$$

2018: $$C = 2.280 \cdot H_d + 5.535 \tag{14}$$

2023: $$C = 2.279 \cdot H_d + 5.653 \tag{15}$$

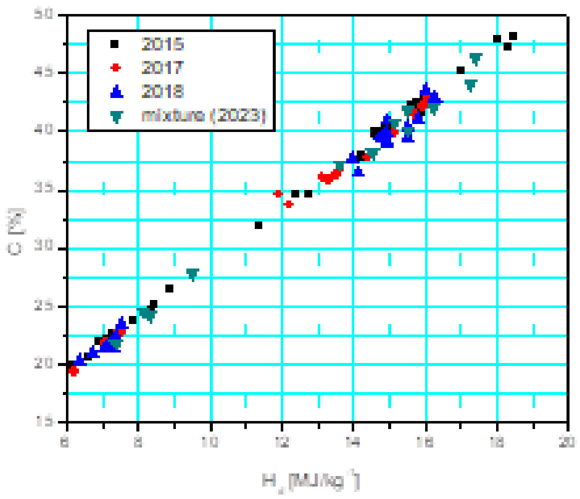

Figure 5. Total carbon content as a function of low heating value, for the samples of lignite, for 2015 – 2023 years, experimental data

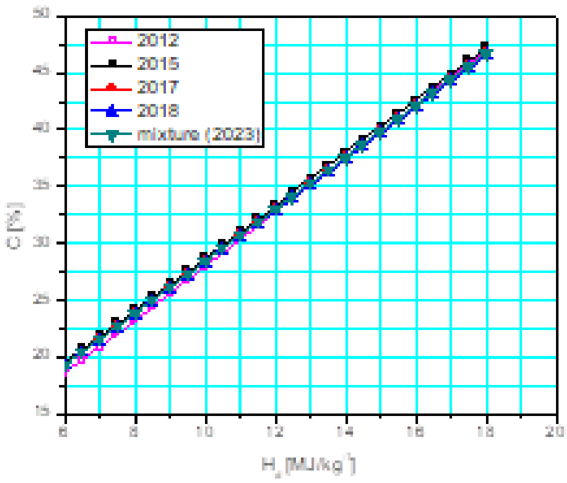

Figure 6. Total carbon content as a function of low heating value, for the samples of lignite, for 2012 – 2023 years, analytical data

The essential dependence is the dependence of the Carbon Emission Factor on the lower heatingl value, obtained based on the following formula, eq (16):

$$CEF = 10\frac{C}{H_d} \quad (16)$$

Including formulas eq.(11-15) in eq. (16), we get the following dependencies, eq. (17-21):

2012: $$\text{CEF}=23.718+\frac{42.637}{\text{H}_\text{d}} \quad (17)$$

2015: $$\text{CEF}=23.09+\frac{56.39}{\text{H}_\text{d}} \quad (18)$$

2017: $$\text{CEF}=22.88+\frac{55.71}{\text{H}_\text{d}} \quad (19)$$

2018: $$\text{CEF}=22.80+\frac{53.35}{\text{H}_\text{d}} \quad (20)$$

2023: $$\text{CEF}=22.79+\frac{56.53}{\text{H}_\text{d}} \quad (21)$$

Based on experimental measurements (Tables 1-4) and calculations eq. (16-21), the diagrams in Figures 7-9 show the dependence of *CEF* on the lower heating value. The diagram in Figure 7 presents the dependences of *CEF* on H_d for the years 2015-2023, obtained by experimental research. The diagrams in Figures 8 and 9 presents analytical dependencies for the years 2012-2023. Based on the analytical dependencies eq. (16-21) and from diagrams 8 and 9, one can notice and conclude:

- The deviations of the value of the emission factor by year are small, and for some years negligible,
- Based on the results of the emission factor taken over from our previous work [5, 6], for 2012 year and their comparison with other years (2015 - 2023), the *CEF* factor of lignite used for the operation of TPP "Nikola Tesla" had the lowest values, especially in the part up to 10 MJ/kg, which indicates that higher quality lignite was used then and less CO_2 was emitted compared to later years,
- Based on certain experimental measurements and calculations, the emission factor of lignite used in 2015 year has the highest value,
- Lignites from other years are found in terms of quality between these two cases.

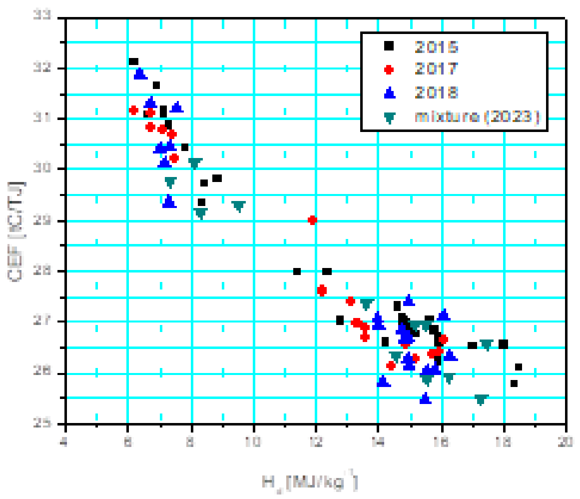

Figure 7. CEF as a function of low heating value, for the samples of lignite, for 2015 – 2023 years, experimental data

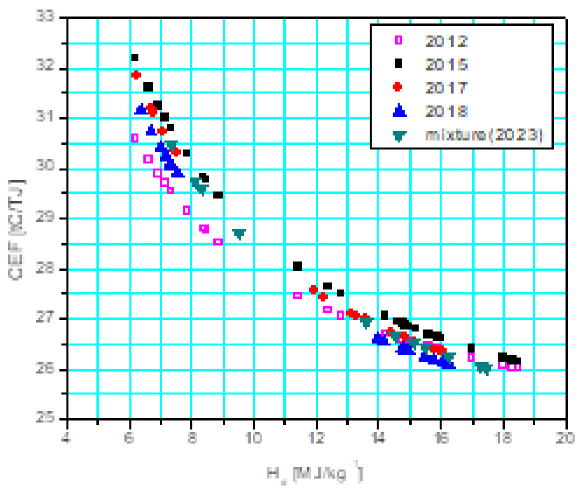

Figure 8. CEF as a function of low heating value, for the samples of lignite, for 2012 – 2023 years, analytical data

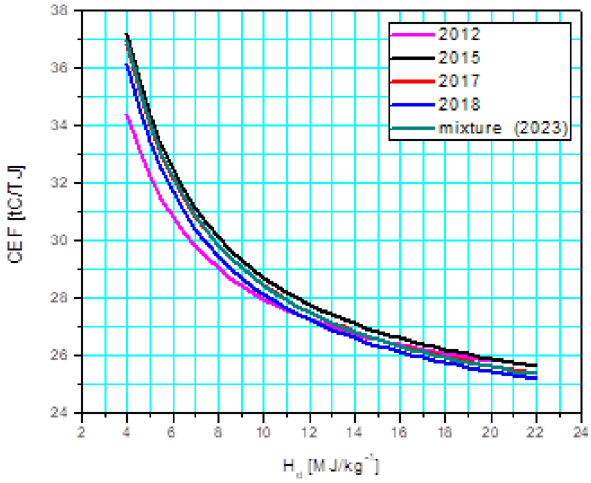

Figure 9. CEF as a function of low heating value, for the samples of lignite, for 2012 – 2023 years, analytical data

The deviation of the *CEF* value from the standard prescribed values recommended by the IPCC for lignite and for "as received" sample ($6 \leq H_d \leq 10$ MJ/kg) is of great importance. Therefore, it is necessary to carry out a detailed review of the stated deviations based on the diagram presented in Figure 9. Enlarged part of the diagram, Figure 10, shows the deviation of the *CEF* value for lower heating values of H_d =6 MJ/kg and H_d =8 MJ/kg, for the coal samples 2012 – 2023, with emphasis on values for 2012 and 2015 years (year with highest and lowest CEF values of the samples tested). These values are also presented in Table 6.

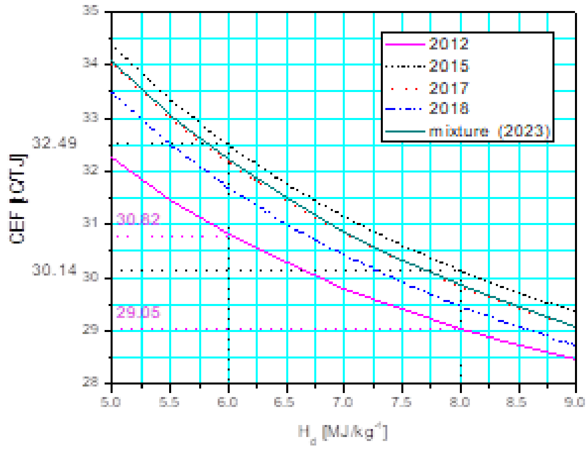

Figure 10. Deviation of CEF values as a function of lower heating values H_d = 6 MJ/kg and H_d = 8 MJ/kg, for the samples, for 2012 – 2023 years

Table 6. CEF values for the coal samples, years 2012 – 2023, for lower heating values, H_d = 6 MJ/kg and H_d = 8 MJ/kg.

Year	*CEF* tC/TJ (H_d =6 MJ/kg)	*CEF* tC/TJ (H_d =8 MJ/kg)
2012	30.82	29.05
2015	32.49	30.14
2017	32.17	29.84
2018	31.69	29.47
2023	32.31	29.86

The diagram in Figure 10 presents enlarged portion of diagram (Figure 9.) for the range of lower heating values $6 \leq H_d \leq 8$ MJ/kg, showing the differences in the CEF value of lignites and mixture for each of the years. For H_d = 6 MJ/kg and H_d = 8 MJ/kg the CEF values for each year are presented in Table 6 and also for the years 2012 and 2015 on diagram in Figure 10. For the coal samples from year 2012, for H_d = 8 MJ/kg, the CEF value 29.05 tC/TJ has been registered, while for H_d = 6 MJ/kg the CEF value is 30.82 tC/TJ. As in the previous case, for the coal samples from year 2015, for the value of H_d = 8 MJ/kg, the CEF value is 30.14 tC/TJ, while for

H_d = 6 MJ/kg the CEF value is 32.49 tC/TJ. It can be seen from the previous that the deviation of the CEF factor is higher than 2 tC/TJ for a particular lower heating value. The largest deviation was registered for the samples from 2023 and 2015, 2.35 tC/TJ, while the smallest deviation is 1.77 tC/TJ for the samples from 2012 year. It is evident that CEF values are certainly higher than the prescribed one (27.6 tC/TJ), recommended according to the IPCC, Table 6.

The diagram in Figure 11 shows the dependence of the lower heating value on the content of combustible matter in the samples for the years 2015 - 2023. The diagram shows slight deviations of the linear dependences by year. This is in agreement with the diagrams representing the dependence of CEF by years (diagrams in Figures 8 and 9).

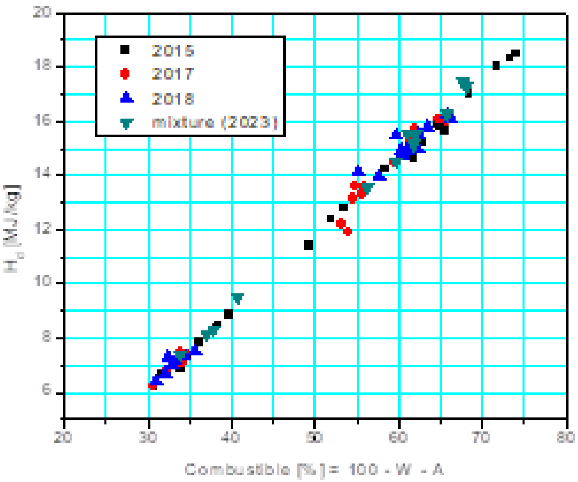

Figure 11. Lower heating value values as a function combustible matter content, for the samples, for 2015 – 2023 years

The diagram in Figure 12 shows the dependence of the CEF value on the lower heating value for imported better quality coal (Table 5). Considering that the content of Carbon (34 – 40 %) and Hydrogen (2.5 - 2.9 %) in the samples on the "as received" basis is significantly higher than that of Kolubara lignite. It is logical and evident that the Lower heating value (LHV) of that coal (over 10 MJ/kg, per "as received" basis) is also higher than Kolubara basin lignite, so this paper did not intend to compare the lignites used for operation of TPP "Nikola Tesla", nor with the mixture of these coals with the Kolubara lignites, but to indicate their differences by showing the emission characteristics. As an example, for the sample, "as received" basis, the lower heating value of better quality coal is from 10 MJ/kg, to over 26 MJ/kg, depending on the sample basis, in contrast to Kolubara lignite, where the lower heating value is in the range of about 7 - 19 MJ/kg.

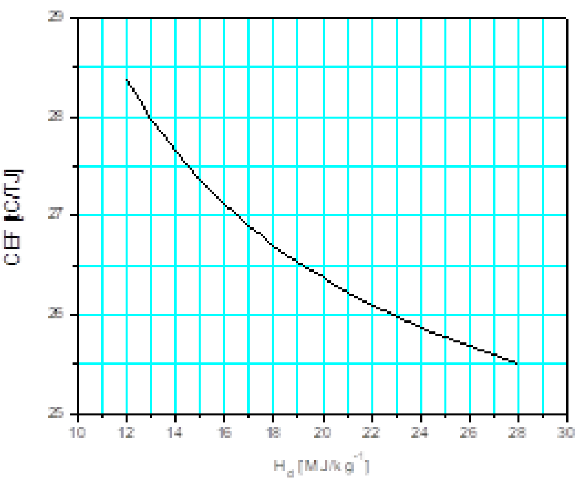

Figure 12. CEF as a function of low heating value, for the imported better quality coal, 2023 year, analytical data

5. Conclusion

This paper presents a methodology that represents a combination of experimental research and calculations in order to determine the emission characteristics of lignite and coal which is used for the operation of TPP within the Joint Stock Company Electric Power Company of Serbia. The results of experimental investigations of lignite used for the operation of TPP "Nikola Tesla", which are mainly lignite from the open-pit mine Kolubara, are given. Samples from 2015 (10 samples), 2017 (6 samples), 2018 (7 samples) and 2023 (4 samples) of a mixture of lignite from the open-pit mine of Kolubara and imported better quality coal from import were used for experimental research. Also, for the investigation, Kolubara lignite from 2012 (30 samples), were used in this paper, which were presented in our previous work. Based on the presented experimental research and calculations, the following conclusions can be drawn:

- The deviations of CEF factor values by years are not negligible, although at first glance they appear so. The fact leading to higher amounts of emitted CO_2, causing higher penalties for exceeded emissions,
- Deviations of the CEF factor for the lower heating values H_d = 6 MJ/kg and H_d = 8 MJ/kg, by years, are significant in absolute value,
- Based on the results of CEF taken over from our previous works [5, 6], for 2012 year and their comparison with other years (2015 - 2023), the emission factor of lignite used for the operation of TPP "Nikola Tesla " had the lowest values, indicating that year was used lignite of higher quality compared to later years,
- Based on certain experimental measurements and calculations, the emission factor of lignite used in 2015 year has the highest value,
- Lignites from other years are found in terms of quality between these two cases,
- The paper highlights the necessity for periodic sampling of coal, in order to determine the exact values of emission characteristics, instead of using pre-defined values by IPCC,
- Using the methodology described for experimental determination of CEF, it is possible to determine the emissions from particular energy sources and total CO_2 emissions on the contry level.

Nomenclature

A – ash content, [%]
C – carbon content, [%]
CEF – carbon emission factor, [tC/TJ]
Combustible – combustibles content, [%]
H – hydrogen content, [%]
H_d – lower heating value, net calorific value, at constant pressure, [kJ/kg] [MJ/kg]
N + O – nitrogen + oxygen content, [%]
S – sulfur content, [%]
W – moisture content, [%]

Subscripts
d – lower heating value (lower calorific value)
fix – fixed
g – combustible,
tot – total

Acronyms
TPP – thermal power plant,
IPCC – intergovermental panel on climate change

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