

## NO<sub>x</sub> AND SO<sub>2</sub> EMISSION FACTORS FOR SERBIAN LIGNITE KOLUBARA

by

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*Emission factors are widely accepted tool for estimation of various pollutants emissions in USA and EU. Validity of emission factors is strongly related to experimental data on which they are based. This paper is a result of an effort to establish reliable NO<sub>x</sub> and SO<sub>2</sub> emission factors for Serbian coals. The results of NO<sub>x</sub> and SO<sub>2</sub> emissions estimations based on USA and EU emission factors from thermal power plants Nikola Tesla Obrenovac A and B utilizing the Serbian lignite Kolubara are compared with experimental data obtained during almost one decade (2000-2008) of emissions measurements. Experimental data are provided from regular annual emissions measurement along with operational parameters of the boiler and coal (lignite Kolubara) ultimate and proximate analysis. Significant deviations of estimated from experimental data were observed for NO<sub>x</sub>, while the results for SO<sub>2</sub> were satisfactory. Afterwards, the estimated and experimental data were plotted and linear regression between them established. Single parameter optimization was performed targeting the ideal slope of the regression line. Results of this optimization provided original NO<sub>x</sub> and SO<sub>2</sub> emission factors for Kolubara lignite.*

Key words: NO<sub>x</sub>, SO<sub>2</sub>, emissions, emission factors, lignite, Kolubara

### Introduction

Nitrogen oxides (NO<sub>x</sub>) and sulphur oxides (SO<sub>x</sub>) are major combustion-generated pollutants from coal-fired power plants. Oxides of sulphur are a major contributor to acid rain. In the case of nitrogen oxides, NO<sub>x</sub> contributes to acid rain whereas N<sub>2</sub>O is a "greenhouse gas" which is also implicated indirectly in the depletion of the ozone layer. Those were the reasons for development of various estimation techniques for their emissions. Various estimation techniques of NO<sub>x</sub> and SO<sub>2</sub> emissions from boilers are described in literature [1-4].

Estimating NO<sub>x</sub> and SO<sub>2</sub> emissions from boilers can be important for a number of reasons, including [5]:

- developing, and assessing the effectiveness of emission control strategies and technologies,
- for regulatory purposes, including compliance with emission standards and reporting to pollutant inventories,
- determination of emission loads in association with economic instruments, such as "cap and trade" schemes and emission taxes,

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- validation of performance levels as part of plant commissioning tests,
- determination of emission rates as part of potential health and environmental assessments,
- developing appropriate mitigation strategies (as input data for design of emission reduction systems),
- determination of the effect of changes to fuel and or operational parameters., and
- providing input to air quality models and verification of model predictions.

The reasons for estimating emissions dictate the level of effort required, the data quality objectives and the resources required. The most important step in the emission estimation process is to define the end use and identify potential users of the data. For example emission estimation required to demonstrate compliance with regulatory standards may require more accurate and costly methods than those intended for the purpose of national inventory reporting.

Air pollutant emission factors are representative values that attempts to relate the quantity of a pollutant released to the ambient air with an activity associated with the release of that pollutant. These factors are usually expressed as the weight of pollutant divided by a unit weight, volume, distance, or duration of the activity emitting the pollutant (*e. g.*, kilograms of SO<sub>2</sub> emitted per kilogram of coal burned). Such factors facilitate estimation of emissions from various sources of air pollution. In most cases, these factors are simply averages of all available data of acceptable quality, and are generally assumed to be representative of long-term averages. Basic classification of emission factors is on: generic and site specific emission factors.

Generic emission factors are derived from actual emission tests conducted on representative sources within a particular industrial sector and are assumed to be long-term averages. Generic emission factors can be either simple or source specific. Source specific emission factors are developed using emission-testing data and source-activity information. Source specific emission factors for boilers are related to individual fuel characteristics (*e. g.* sulfur content of fuel), and to technology specific parameters (firing practice, ash removal, *etc.*). Emission factors that account for the influence of these parameters tend to yield more reliable estimates than those estimated using the generic factors.

Site specific emission factors are developed for a specific facility based on emission data generated from the site. They often require further validation and approval from regulatory authorities.

There are two principal sources of emission factors in the world, USA Environmental Agency USEPA (AP-42 document) and IPCC Guidelines for National Greenhouse Gas Inventories (on which the European Environment Agency – EEA emission factors are based) [6]. The approach to definition and development of emission factors of both agencies is mostly the same, but some factors are presented in different units. This is the case with NO<sub>x</sub> and SO<sub>2</sub> emission factors. Nevertheless, development of emission factors require extensive experimental research, both of fuels and plants used in combustion processes. Special attention is needed when dealing with low rank coals, more specific, Serbian lignites which are characterized by low heating value, high volatile, water and ash content, regarding the NO<sub>x</sub> and SO<sub>2</sub> emissions [7, 8].

Serbian energy reserves are mainly in lignite that is concentrated in three coal basins, Kolubara and Kostolac in Central Serbia and Kosovo in Kosovo and Metohia. First two basins provide all the coal used for electricity production in four great thermal power plants (TPP), TENT Obrenovac A and B and Kostolac A and B (65% of total electricity production in Serbia). The facts that there are no emission factors for these coals and that Public Enterprise “Electric Power Industry of Serbia” (JP EPS) has ongoing projects for reduction of NO<sub>x</sub> and SO<sub>2</sub> emissions for which the reliable estimations are needed, were the basis of the research that is presented in this paper.

## SO<sub>x</sub> and NO<sub>x</sub> emissions

The SO<sub>x</sub> emissions from lignite combustion are a function of the sulfur content of the lignite and the lignite composition (*i. e.*, sulfur content, heating value, and alkali concentration). The conversion of lignite sulfur to SO<sub>x</sub> is generally inversely proportional to the concentration of alkali constituents in the lignite. The alkali content is known to have a great effect on sulfur conversion and acts as a built-in sorbent for SO<sub>x</sub> removal.

The NO<sub>x</sub> emissions from lignite combustion are mainly a function of the boiler design, firing configuration, and excess air level. Tangential units, stoker boilers, and FBC typically produce lower NO<sub>x</sub> levels than wall-fired units and cyclones. New boilers constructed since implementation of the modern regulations for environmental protection have NO<sub>x</sub> controls integrated into the boiler design and have NO<sub>x</sub> emission levels that are comparable to emission levels from small stokers. In most boilers, regardless of firing configuration, lower excess combustion air results in lower NO<sub>x</sub> emissions. However, lowering the amount of excess air in a lignite-fired boiler can also affect the potential for ash fouling.

### Origins of SO<sub>x</sub> emissions

Considerable work on the properties that affect combustion originated sulphur emissions is made but despite of that, for the most part the extent of sulphur emissions in large-scale pulverised-coal combustion is straightforward – nearly all the sulphur in the coal is converted to SO<sub>2</sub>. The only coal properties that greatly affect the emission of SO<sub>2</sub> are the total sulphur content and the ash, and the amount captured by the ash is only a small part of the total. Most of the SO<sub>2</sub> is either emitted or captured by flue-gas desulphurisation. The sulphur content of coal ranges from less than 0.5% m/m to greater than 10 % m/m while those used for combustion are generally in the range of 0.5-3% m/m [9]. The sulphur is primarily associated with three phases in coal; sulphate minerals, sulphide minerals (predominantly pyrite, FeS<sub>2</sub>) and the organic matrix. The sulphate content is usually low except when the pyrite has been oxidised. Our knowledge of the organic sulphur in coal is a little less certain [10].

However, regardless of the form of sulphur in coal, combustion converts most of it to SO<sub>x</sub> (mainly to SO<sub>2</sub>, with some sulphur trioxide, SO<sub>3</sub>). The formation of SO<sub>3</sub> in a boiler is complex and is believed to occur through the oxidation of SO<sub>2</sub> by molecular oxygen, the oxidation of SO<sub>2</sub> in the flame by atomic oxygen, and the catalytic oxidation of SO<sub>2</sub>. Generally the ratio of SO<sub>2</sub> to SO<sub>3</sub> in combustion gas is in the range of 20:1 to 30:1 [11].

Despite the evidence for sulphur capture by ash, Okamoto [12] pointed out that the amount is so small that it can be neglected for practical purposes. Accordingly, he calculated the SO<sub>x</sub> emissions as follows:

$$V_{\text{SO}_2} = 1000 \frac{S}{100} 0.7 \quad (1)$$

where  $V_{\text{SO}_2}$  [m<sup>3</sup>t<sup>-1</sup>] is the volume amount of SO<sub>x</sub> per tonne of coal, 1000 kg – the mass of coal burned,  $S$  [%m/m] – the weight percent of total sulphur in coal, and 0.7 [m<sup>3</sup>kg<sup>-1</sup>] – the stoichiometric factor for sulphur combustion reaction.

The other calculation procedure was proposed by McInnes [13] in 1996 and lately adopted as a method by European environment agency (EEA):

$$C_{\text{SO}_{2\text{max}}} = 2C_{\text{S fuel}} \quad (2)$$

where  $C_{\text{SO}_2^{\text{max}}}$  [kgkg<sup>-1</sup>] is the maximum attainable amount of sulphur dioxide (in mass pollutant/mass fuel), and  $C_{\text{S fuel}}$  [kgkg<sup>-1</sup>] – the sulphur content of fuel (in mass element/mass fuel).

Afterwards, the value of maximum attainable amount of SO<sub>2</sub> is corrected by the sulphur retention in ash and reduction efficiency and availability of the secondary flue-gas desulphurisation system installed.

### Origins of NO<sub>x</sub> emissions

In contrary to the SO<sub>x</sub> emissions, NO<sub>x</sub> emissions are influenced by much more factors. NO<sub>x</sub> emissions from different fuels depend upon on the chemistry of combustion of the fuel. NO<sub>x</sub> emissions from lignite combustion are produced by two primary mechanisms:

- fuel NO<sub>x</sub> is related to the nitrogen content of the fuel and the firing mode used in combustion,
- thermal NO<sub>x</sub> is the chemical formation of NO from N<sub>2</sub> and O<sub>2</sub> in combustion air at temperatures exceeding 1400 °C.

The third mechanism of NO<sub>x</sub> formation – prompt NO<sub>x</sub> is negligible for temperatures occurring in the furnaces of coal fired steam boilers.

The production of NO<sub>x</sub> from any fuel cannot be simplified into a mathematical relationship, and NO<sub>x</sub> emission estimations for any fuel are therefore problematic. Furthermore, the proportion of fuel NO<sub>x</sub> and thermal NO<sub>x</sub> depends upon the type of boiler and the combustion conditions, much more than on the nitrogen content of the fuel [14].

A very brief list of influential factors on NO<sub>x</sub> emissions include: fuel nitrogen, proximate volatile matter and fuel ratio, volatile and char nitrogen, particle size, moisture and ash content, air-staging and reburning efficiency, *etc.*

### Emission factors

Emission factors are cost-effective means for development of emission inventories. One of the advantages when using emission factors is that emissions from many individual sources can be estimated by testing only a small fraction of those sources. Another advantage is that they can sometimes be used to generate default emission factors for non-measurable substances by applying specific knowledge of the process characteristics. Emission factors are often developed for “normal or typical” operating conditions so they do not reflect start-up, shut down or other modes of operation that could significantly contribute to air emissions.

Test data from individual sources are not always available and may not always reflect the variability of actual emissions over a prolonged period of time. Thus, assuming that they are used with sufficient knowledge, emission factors are appropriate method for estimating emissions in many cases, including emissions from the steam boilers in thermal power plants.

An emission factor is a tool that is used to estimate air pollutant emissions to the atmosphere. It relates the quantity of pollutants released from a source to some activity associated with those emissions. NO<sub>x</sub> or SO<sub>2</sub> emission factors are usually expressed as the weight of pollutant emitted per weight unit, (of coal burned) or per energy unit (of generated heat). Emission factors are used to estimate a source’s emissions by the general equation:

$$E = AEF \left[ 1 - \left( \frac{ER}{100} \right) \right] \quad (3)$$

where  $E$  is the emissions,  $A$  – the activity rate,  $EF$  – the uncontrolled emission factor, and  $ER$  [%] – the overall emission reduction efficiency.

ER is the product of the control device destruction or removal efficiency and the capture efficiency of the control system. When estimating emissions for a long time period (*e. g.*, 1 year), both the device and the capture efficiency terms should account for upset periods as well as routine operations.

Thermal power plants Obrenovac A and B, which were the objects of experiment do not utilize any emission control for SO<sub>2</sub> and NO<sub>x</sub>, so only the *EF* was analyzed.

Brief summary of preferred and alternative emission estimation methods for NO<sub>x</sub> and SO<sub>2</sub> from boilers is given in tab. 1 [15].

**Table 1. Summary of preferred and alternative emission estimation methods for NO<sub>x</sub> and SO<sub>2</sub> from boilers**

Parameter	Preferred emission estimation approach	Alternative emission estimation approach <sup>a</sup>
SO <sub>2</sub>	CEMS/PEM data	(1) Fuel analysis* (2) Stack sampling data (3) EPA/state published emission factors
NO <sub>x</sub>	CEMS/PEM data	(1) Stack sampling data (2) EPA/state published emission factors

\* May be used when no SO<sub>2</sub> control device is present.

### *USEPA emission factors*

USEPA AP-42 document provides emission factors for three main classifications of air pollutants: criteria pollutants and their precursors, hazardous air pollutants (HAP), and greenhouse gases. Besides these main groups, there are also specific pollutants like ammonia and stratospheric ozone depleters. The criteria pollutants are the most extensively covered, because they were the original focus of AP-42 and the Agency's regulatory efforts. The six criteria pollutants are sulphur dioxide, nitrogen oxides, carbon monoxide, lead, particulate matter less than 10 μm in diameter, and ozone. NO<sub>x</sub>, CO, and volatile organic compounds (VOC) are considered important because they are precursors of the pollutant ozone. Additionally, ammonia, SO<sub>2</sub>, NO<sub>x</sub>, and VOC are also considered precursors of PM.

Emission factors in AP-42 are appropriate to use in developing emission estimates for emission inventories. These inventories have many purposes including ambient dispersion modeling and analysis, control strategy development, and screening of sources for compliance determinations. However, because emission factors represent average emission rates for an entire source category, they are not recommended as emission limits or standards for any specific source. Actual test results from source specific tests or continuous emission monitoring systems (CEMS), when properly done, are more indicative of actual emissions for a specific source. When source-specific information is not available, use of emission factors may be necessary. Whenever AP-42 emission factors are used, one should be aware of their limitations in accurately representing the emissions from a particular facility, and the risks of using emission factors in such situations should be evaluated against the costs of further testing or analyses. Emission factors generally are developed to represent long-term average emissions, so testing is usually conducted at normal operating conditions. Lignite emission factors are principally developed for North Dakota and Texas lignites.

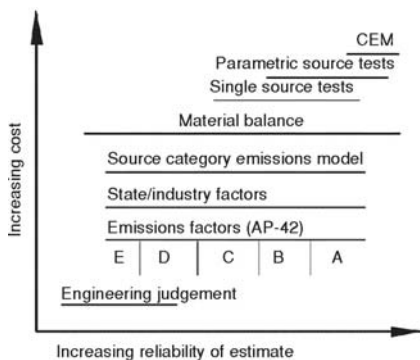


Figure 1. Approach to emission estimation [5]

Figure 1 depicts various emission estimation approaches that should be considered when analyzing the tradeoffs between the cost of obtaining the estimates and the quality of the resulting estimates. Data presented on fig. 1 are only indication of a typical relationship between cost and reliability while in practice there is a wide range of reliability possible for any one approach. Typically, using an emission factor to estimate emissions is cheaper than a source test, but the emission estimate may not be as reliable, although an “A-rated” emission factor may be as reliable as a CEMS. Selecting the protocol to be used to estimate source-specific emissions warrants a case-by-case analysis considering the costs and risks in the specific situation.

Letters from A to E under emissions factors (AP-42) represent the overall emission factor quality rating from the best to the worst. The overall emission factor quality ratings are described as follows:

*A – Excellent:* Developed only from A-rated test data taken from many randomly chosen facilities in the industry population. The source category is specific enough so that variability within the source category population may be minimized.

*B – Above average:* Developed only from A-rated test data from a reasonable number of facilities. Although no specific bias is evident, it is not clear if the facilities tested represent a random sample of the industries. As in the A-rating, the source category is specific enough so that variability within the source category population may be minimized.

*C – Average:* Developed only from A- and B-rated test data from a reasonable number of facilities. Although no specific bias is evident, it is not clear if the facilities tested represent a random sample of the industry. As in the A-rating, the source category is specific enough so that variability within the source category population may be minimized.

*D – Below average:* The emission factor was developed only from A- and B-rated test data from a small number of facilities, and there is reason to suspect that these facilities do not represent a random sample of the industry. There also may be evidence of variability within the source category population. Limitations on the use of the emission factor are noted in the emissions factor.

*E – Poor:* The emission factor was developed from C- and D-rated test data, and there is reason to suspect that the facilities tested do not represent a random sample of the industry. There also may be evidence of variability within the source category population. Limitations on the use of these factors are noted where applicable.

#### EEA emission factors

Emission factors according to EEA are classified in three tiers, from the lowest (Tier 1) to the highest level (Tier 3) of confidence. The basis of this approach is the 2006 IPCC (Intergovernmental Panel on Climate Change) Guidelines for National Greenhouse Gas Inventories (IPCC Guidelines) [16], while many emission factors are referenced to the USEPA AP-42 document [17].



Methodological choice for individual source categories is important in managing overall inventory quality and minimizing uncertainty. Generally, inventory uncertainty is lower when emissions are estimated using the most rigorous, higher tiered, methods. However, these methods generally require more extensive resources for data collection and calculation, so it may not be feasible to use most rigorous method for every category of emissions. It is, therefore, good practice to identify and prioritize the effort on those categories which make the greatest contribution to the overall inventory estimates (and where possible, the uncertainty). In this paper only one category is discussed – combustion in energy and transformation industries

The “Tier 1” method is a “simple” method using default emission factors only. To upgrade a Tier 1 to a Tier 2 method, the default emission factors should be replaced by country-specific or technology-specific emission factors. This might also require a further split of the activity data over a range of different technologies, implicitly aggregated in the Tier 1 method. A Tier 3 method could be regarded as a method that uses the latest scientific knowledge in more sophisticated approaches and models.

#### *Tier 1: Basic method*

A method using readily available statistical data on the intensity of processes (activity rates) and default emission factors. These emission factors assume a linear relation between the intensity of the process and the resulting emissions. The Tier 1 default emission factors also assume an average or typical process description. This method is the simplest method, has the highest level of uncertainty and should not be used to estimate emissions from key categories.

#### *Tier 2: More complex method*

Tier 2 is similar to Tier 1 but uses more specific emission factors developed on the basis of knowledge of the types of processes and specific process conditions that apply in the country for which the inventory is being developed. Tier 2 methods are more complex, will reduce the level of uncertainty, and are considered adequate for estimating emissions for key categories.

#### *Tier 3: Advanced method*

Tier 3 is defined as any methodology more detailed than Tier 2; hence there is a wide range of Tier 3 methodologies. At one end of the range there are methodologies similar to Tier 2 (*i. e.* activity data × emission factor) but with a greater disaggregation of activity data and emission factors. At the other end of the range are complex, dynamic models in which the processes leading to emissions are described in great detail.

The basic concept of the procedure to select the methods for estimating process emissions from combustion in energy and transformation industries is:

- if detailed information is available, use it,
- if the source category is a key source, a Tier 2 or better method must be applied and detailed input data must be collected. The decision tree directs the user in such cases to the Tier 2 method, since it is expected that it is easier to obtain the necessary input data for this approach than to collect facility-level data needed for a Tier 3 estimate. However, the inventory compiler should be aware that, because the number of sources may be comparatively small, in many instances the data required for a Tier 3 approach may be only a little more difficult to obtain than at Tier 2, and
- detailed process modeling is not explicitly included in this decision tree. However, detailed modeling will usually be done at facility level and results of such modeling could be seen as “facility data” (Tier 3) in the decision tree.

## Experiment

The experimental research covered on-site stack emissions measurement of air pollutants including SO<sub>2</sub> and NO<sub>x</sub> from boilers in all units of TPP Nikola Tesla Obrenovac A (6 units) and B (2 units) during period of nine years (2000-2008). All measurements were made as annual control measurements according to the acting rule in that period – Regulation on emission limit values (ELV), method and terms of measurement and data recording published in Official Gazette RS 30/97 (further on referred as Regulation [18]). Basic data about the units of TPP that were objects of measurements are given in tab. 2.

As it can be seen from tab. 2, the units vary in size (power) from the oldest (and smallest) unit A1 (210 MW<sub>e</sub>) to the newest and the biggest B1 and B2 (624 MW<sub>e</sub>), providing wide span of unit size. All units utilize the same combustion practice, tangential firing with dry bottom ash removal. There are no abatement techniques applied on all units, neither for SO<sub>2</sub> nor NO<sub>x</sub>. Besides, all the boilers are operating with relatively high excess air in furnace due to operating problems (coal mills failures, uncontrolled air penetration in coal-air mixture ducts) and suffer from slagging and ash fouling. All these facts have significant impact on NO<sub>x</sub> emissions.

**Table 2. Characteristics of the units of TPP Nikola Tesla A and B**

Unit	Boilers			Turbine	
	Manufacturer	Rated capacity [t <sup>h</sup> <sup>-1</sup> ]	Number of mills	Manufacturer	Rated power [MW]
A1, A2	SES, CSFR	650	6	LMZ (Russia)	210
A3	SES	920	6	CEM (BBC)	305
A4, A5, A6	SES, R-SULZER	920	6	CEM (BBC)	308.5
Rated power of TPP Nikola Tesla A: 1650.5 MW (on generator), 1502 MW (declared net capacity – DNC)					
B1, B2	RAFAKO-SULZER	1824	8	AA CEM (BBC)	624
Rated power of TPP Nikola Tesla B: 1240 MW (on generator), 1160 MW (DNC)					

The measurements were made strictly according to the requirements of the Regulation, on adequate measuring places, in the flue gas duct, after the electrostatic precipitator. The list of measured values include flue gas analysis (O<sub>2</sub> content in flue gas, concentrations of CO, NO<sub>x</sub>, SO<sub>2</sub> and particulate matter), flue gas temperature and flow rate, fuel (coal) consumption, coal ultimate and proximate analysis, and electric power of the unit. An example of measurement results is given in tab. 3.

Due to long period of measurement (nine years) flue gas analysis was made by extractive flue gas analyzers operating on different principles. At the beginning (year 2000), O<sub>2</sub> content was measured by the paramagnetic analyzer and afterwards by instruments with O<sub>2</sub> electrochemical cells. Flue gas NO concentration was measured principally by chemiluminiscense flue gas analyzer (THERMO ELECTRON model 12A) and in one case (year 2004) by electrochemical cells (Testo 360 flue gas analyzer). SO<sub>2</sub> concentration in flue gas was principally measured by non-dispersive infrared (NDIR) analyzer (MAIHAK UNOR 6R) and in one case (2004) by electrochemical cells (Testo 360 flue gas analyzer).



**Table 3. Emissions measurement results for Unit A3**

Year	Measurement number	Unit data		Coal data (as received)				Flue gas data (dry, at $p = 101,3 \text{ kPa}$ , $t = 0 \text{ °C}$ )			
								Total/average values after the electrostatic precipitator			
		$P_{el}$	$B$	$LHV$	$W_{tot}$	$A$	Stot	$V_{fgdryntot}$	$O_{2ave}$	$SO_2$	$NO_x$
[MW]	[ $\text{th}^{-1}$ ]	[ $\text{kJkg}^{-1}$ ]	[% m/m]	[% m/m]	[% m/m]	[ $\text{m}^3\text{h}^{-1}$ ]	[% v/v]	[ $\text{mgm}^{-3}$ ]	[ $\text{mgm}^{-3}$ ]		
2000	1	266	429	6935	54.00	15.00	0.48	1,390,000	9.2	2082	409
	2	297	386	8592	50.00	12.00	0.30	1,451,000	8.2	1286	375
	3	249	348	7993	52.00	13.00	0.53	1,393,000	9.0	2363	387
2001	4	233	329	7908	49.00	16.00	0.48	1,541,000	9.8	2111	260
	5	262	359	8160	49.00	15.00	0.31	1,472,000	8.9	1435	344
	6	211	269	8777	50.00	11.00	0.40	1,515,000	11.1	1639	336
2006	7	306	381	8989	50.00	11.00	0.44	1,098,000	7.9	1805	378
	8	305	417	8177	51.30	12.30	0.33	1,175,000	8.0	1350	396
2007	9	305	357	9562	51.40	7.72	0.61	1,316,000	8.8	2121	403
	10	305	369	9246	52.70	6.79	0.42	1,332,000	8.6	1052	433
	11	305	365	9353	52.30	6.41	0.50	1,231,000	8.2	1807	492
2008	12	288	476	6764	49.00	18.25	0.39	1,583,000	9.2	1978	375
	13	304	414	8214	51.25	11.36	0.50	1,641,000	9.1	1334	401
	14	301	380	8850	50.80	9.84	0.47	1,547,000	8.7	1325	422

Note: Concentrations of pollutants (SO<sub>2</sub>, NO<sub>x</sub>) are presented at reference O<sub>2</sub> of 6 %v/v.

Flue gas flow rate was measured according to the standard method of measurement the velocity field in appropriate cross-section of the flue gas duct by indirect method (Pitot probe for the pressure drop measurement and calculation of the velocity). Afterwards, the volume flow rate was calculated based on the average flue gas velocity and area of the cross-section. Flue gas temperature was simultaneously measured in the same cross-section to facilitate recalculation of the flow rate to normal conditions (pressure of 101.3 kPa and temperature of 273 K).

Water content of the flue gas was determined from the material balance of the combustion process, based on the difference between the wet and dry flue gas. This data was necessary for calculation of the dry flue gas flow rate as the concentrations of NO<sub>x</sub> and SO<sub>2</sub> were measured in dry flue gas (due to the measurement principle of the flue gas analyzers).

Due to the restrictions imposed by configuration of the flue gas ducts on the units of TPP (each unit has two electrostatic precipitators after which two ducts are joined into one), all

the measurements were made for each flue gas duct separately, and afterwards, based on the measured values of each measured parameter, the average value for complete unit was calculated. The only exception was the flue gas flow rate, for which two measured values were added and total flue gas flow rate of the unit calculated.

## Results and discussions

### Basic calculations

The reference method for comparison the results of emission estimation was the calculation of emission based on the emission measurement results as it is described in the Regulation [18]. Starting point for calculation is measured concentration of the pollutant in flue gas. Besides, the volume flow rate of dry flue gas is required. Both data are basic elements of the Report about emission measurement. Formula for calculation of reference emission is:

$$E_i = C_{in} \cdot 10^{-6} \dot{V}_{fgdryn} \quad (4)$$

where  $C_{in}$  is the concentration of pollutant "i" at normal conditions ( $p = 101.3$  kPa,  $T = 273$  K) and  $\dot{V}_{fgdryn}$  – the volume flow rate of dry flue gas at normal conditions.

Final result of the calculation is mass flow rate of the pollutant, based on which the annual emission of the boiler is calculated (by multiplication with annual number of operating hours).

Based on the USEPA emission factor ( $E_{iUSA}$ ) emission is calculated according to the [17]:

$$E_i = B \cdot EF_{iUSA} \quad (5)$$

where  $B$  is the fuel consumption by mass and  $EF_{iUSA}$  – the emission factor according to [17].

Based on the EEA emission factor ( $E_{iEU}$ ) emission is calculated according to the eq. [1]:

$$E_i = B \cdot LHV \cdot EF_{iEU} \quad (6)$$

where  $LHV$  is the coal low heating value, and  $EF_{iEU}$  – the emission factor according to [1].

An example of the calculation results is presented in tab. 4 for the same unit (A3) that the measurement results are given.

### Method of work

The basis of the calculations were experimental results of annual emissions measurement [as an example, refs. 19-23 are given]. The first step was to check out the possibility of utilization of USA and EU SO<sub>2</sub> and NO<sub>x</sub> emission factors on observed plants. The results of this were SO<sub>2</sub> and NO<sub>x</sub> hourly emissions marked with subscript USA for American, respectively EU for European emission factors.

Afterwards, those values were compared with the values measured and calculated according to the reference method (eq. 2) applying the regression analysis. Total set of 98 points for 8 units of TPP Obrenovac A and B was analyzed. Linear regression was performed using the fix intercept of regression line with axes at point (0,0). Linear correlations with coefficient of

**Table 4. Emissions calculation results for Unit A3**

Year	Measurement number	Unit data		SO <sub>2</sub> emissions					NO <sub>x</sub> emissions				
		$P_{el}$	$B$	$(E_{SO_2})_{meas}$	$(E_{SO_2})_{EU}$	$(E_{SO_2})_{EUopt}$	$(E_{SO_2})_{USA}$	$(E_{SO_2})_{USAopt}$	$(E_{NO_x})_{meas}$	$(E_{NO_x})_{EU}$	$(E_{NO_x})_{EUopt}$	$(E_{NO_x})_{USA}$	$(E_{NO_x})_{USAopt}$
		[MW]	[th <sup>-1</sup> ]	[kgh <sup>-1</sup> ]	[kgh <sup>-1</sup> ]	[kgh <sup>-1</sup> ]	[kgh <sup>-1</sup> ]	[kgh <sup>-1</sup> ]	[kgh <sup>-1</sup> ]	[kgh <sup>-1</sup> ]	[kgh <sup>-1</sup> ]	[kgh <sup>-1</sup> ]	[kgh <sup>-1</sup> ]
2000	1	266	429	2277	2439	2367	3088	3263	447	851	595	1522	732
	2	297	386	1599	2723	2643	1739	1838	466	950	664	1372	659
	3	249	348	2633	2283	2216	2769	2926	431	796	557	1236	594
2001	4	233	329	2440	2136	2074	2372	2506	300	745	521	1169	562
	5	262	359	1711	2402	2332	1669	1764	410	838	586	1274	612
	6	211	269	1631	1934	1878	1613	1704	334	675	472	954	459
2006	7	306	381	1737	2805	2723	2512	2654	364	978	684	1351	649
	8	305	417	1369	2796	2714	2064	2181	402	975	682	1480	711
2007	9	305	357	2280	2796	2714	3263	3448	433	975	682	1266	608
	10	305	369	1154	2796	2714	2323	2455	475	975	682	1309	629
	11	305	365	1898	2796	2714	2734	2889	517	975	682	1294	622
2008	12	288	476	2463	2640	2563	2785	2943	467	921	644	1690	812
	13	304	414	1744	2787	2705	3103	3279	524	972	680	1469	706
	14	301	380	1681	2759	2679	2681	2833	535	962	673	1350	649

determination higher than 0.90 (0.93-0.97) were established, but with the slopes that deviated more or less from 1, as it is presented on figs. 2 and 3 (for NO<sub>x</sub>) and 4 and 5 (for SO<sub>2</sub>). This was the indication of necessity to perform the optimization of the emission factors as to achieve the best correlation with measured data.

The slope of NO<sub>x</sub> emission linear regression line for original EU emission factor is 1.65 which mean that this approach, lead to overestimation of NO<sub>x</sub> emission for about 65%. At the same time, the slope of the same line for original USA emission factor is 2.40, meaning that American approach lead to even greater overestimation of NO<sub>x</sub> emission for about 140%.

Similar results, but less different were obtained for SO<sub>2</sub> emissions. The slope of SO<sub>2</sub> linear regression line for original EU emission factor is 1.17 which means that this approach, lead to moderate overestimation of SO<sub>2</sub> emission (about 17%). At the same time, the slope for original USA emission factor is 1.07, meaning that American approach lead to slight overestimation of SO<sub>2</sub> emission (about 7%).

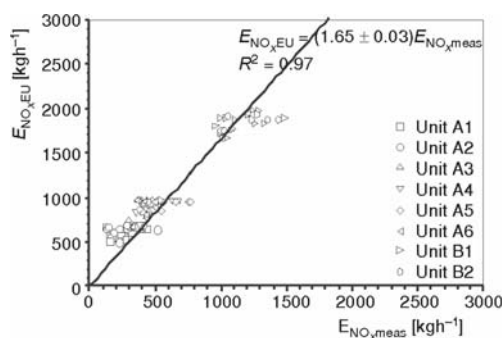


Figure 2. Calculated by EU emission factor vs. measured hourly NO<sub>x</sub> emissions

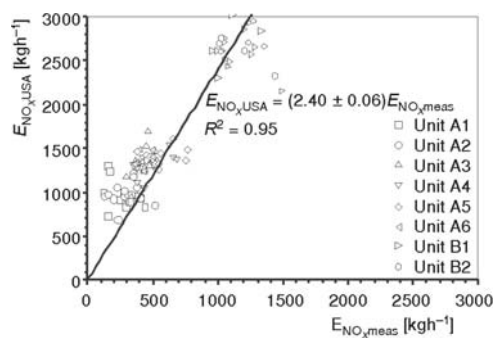


Figure 3. Calculated by USA emission factor vs. measured hourly NO<sub>x</sub> emissions

As it is defined in eqs. 5 and 6, emission of pollutant (NO<sub>x</sub> or SO<sub>2</sub>) is a function of only one parameter (emission factor) if all other parameters are held constant as it was assumed in this research. Regarding the target of investigations, the single parameter optimization (only per emission factor) of these correlations was performed aiming the goal to obtain the ideal slope of the regression line between the sets of calculated and measured values. The optimization was performed collectively for all units as they use the same coal, Serbian lignite Kolubara. The results of the optimization are given together with original emission factors in tab. 5. Calculated

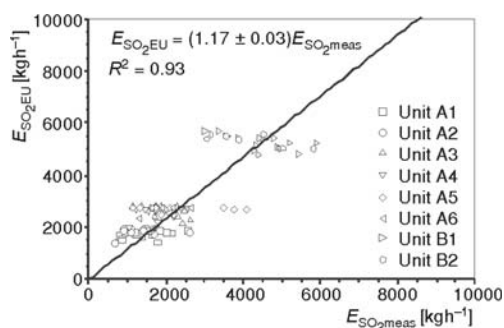


Figure 4. Calculated by EU emission factor vs. measured hourly SO<sub>2</sub> emissions

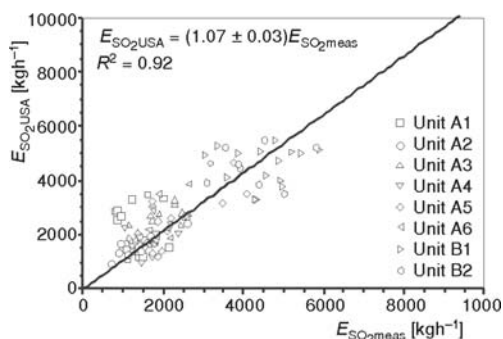


Figure 5. Calculated by USA emission factor vs. measured hourly SO<sub>2</sub> emissions

Table 5. Results of the single parameter optimization and original EU and USA emission factors

Emission factor	EU original	EU optimized	USA original	USA optimized
	[gGJ <sup>-1</sup> ]		[kg t <sup>-1</sup> ]	
NO <sub>x</sub>	286 <sup>a</sup>	173	3.55 <sup>b</sup>	1.48
SO <sub>2</sub>	820 <sup>a</sup>	700	15 <sup>c</sup>	13.99 S

a – Tier 2 emission factors for wet and dry bottom boilers using brown coal/lignite, no abatement techniques and coal S content of 1 %m/m; b – for lignite pulverized coal, dry bottom, tangential firing configuration with no abatement techniques, emission factor rating A; c – for lignite pulverized coal, dry bottom, tangential firing configuration with no abatement techniques, emission factor rating C (S is total sulphur content in coal, as received)

NO<sub>x</sub> emissions based on emission factor for lignite Kolubara (EU optimized) are presented in figs. 6, and 7 for USA optimized. Respectively the same data for SO<sub>2</sub> are presented in figs. 8 and 9.

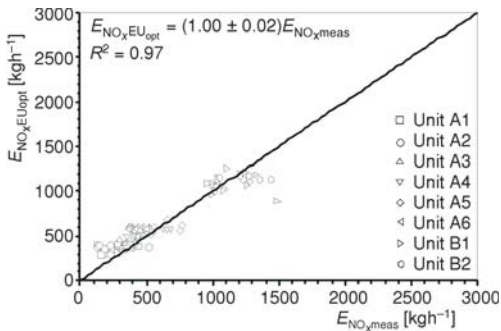


Figure 6. Calculated by optimized EU emission factor vs. measured hourly NO<sub>x</sub> emissions

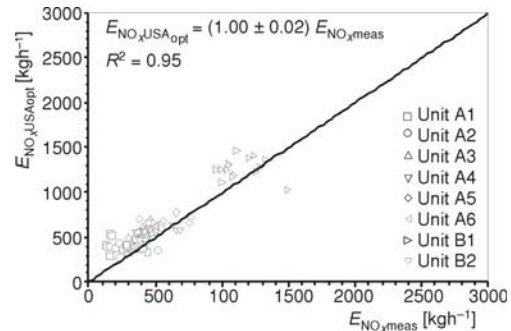


Figure 7. Calculated by optimized USA emission factor vs. measured hourly NO<sub>x</sub> emissions

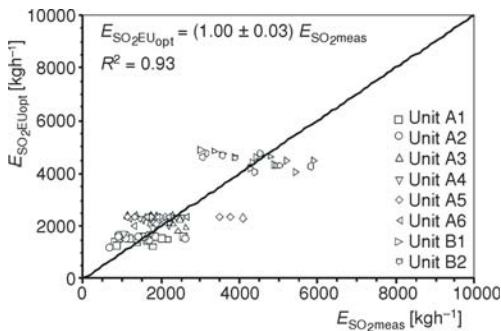


Figure 8. Calculated by optimized EU emission factor vs. measured hourly SO<sub>2</sub>

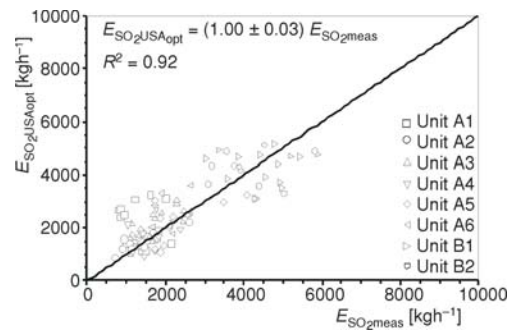


Figure 9. Calculated by optimized USA emissions emission factor vs. measured hourly SO<sub>2</sub> emissions

Optimized values of EU or USA emission factors are the new emission factors for Serbian lignite Kolubara. As it can be seen from tab. 5, the new emission factor for NO<sub>x</sub> is 40% lower than the original EU emission factor, while for the USA NO<sub>x</sub> emission factor the difference is even greater, the new emission factor is almost 58% lower than the original. At the same time, the improvements for SO<sub>2</sub> emission factors are much smaller. The new emission factor for SO<sub>2</sub> is 15% lower than the original EU emission factor, and for USA SO<sub>2</sub> emission factor the difference is even smaller, the new emission factor is about 7% lower than the original USA emission factor for SO<sub>2</sub>.

The results of investigations imply that original emission factors for NO<sub>x</sub>, both EU and USA should not be used for estimation of NO<sub>x</sub> emission in TPP Obrenovac and Serbian lignite Kolubara. Concurrently, original emission factors for SO<sub>2</sub>, both EU and USA could be used for estimation of SO<sub>2</sub> with minor corrections.

## Conclusions and future work

Emission factors for criteria pollutants like NO<sub>x</sub> and SO<sub>2</sub> are widely accepted tool for estimation emissions of these gases. Both USEPA and EEA have developed their own emission factors based on extensive research on various sources of emission including the TPP. Nevertheless, applicability of these factors is limited to the fuels and TPP that are recorded in their databases. It is strongly recommended to perform own measurements and tests on specific objects before utilizing those emission factors. In this respect, the authors of this paper have analyzed the results of regular annual emissions measurement from steam boilers on 8 units of TPP Nikola Tesla Obrenovac A and B. All boilers are dry bottom type and use the same firing practice (tangential) and the same coal, Serbian lignite Kolubara. There are no abatements techniques applied on any unit.

Original emission factors (USEPA and EEA) were used to calculate the emission based on fuel consumption (USEPA) and fuel consumption and lower heating value of the coal (EEA). The results of the calculation were compared to the experimental results and linear regression analysis performed. It was concluded that the NO<sub>x</sub> emissions are overestimated for both USA and EU emission factors. This is the result of differences in Serbian lignite Kolubara and American and European lignites for which the original emission factors were developed. The other influential factor are the operating parameters of the boilers. Basic design and firing practice are the same as listed in the USEPA and EEA documents, but the real operational status of the boiler systems affecting the NO<sub>x</sub> generation are different in the case of TPP Nikola Tesla boilers. Synergetic effect of both coal and plant characteristics resulted in lower NO<sub>x</sub> emissions than estimated according to the USA and EU emission factors.

SO<sub>2</sub> emission was 17% overestimated by EU emission factor and 7% if calculated by USA emission factor compared to measured emission. Deviations from the values obtained utilizing optimized emission factors are negligible, within the 5% margin of error.

Final results were the new NO<sub>x</sub> and SO<sub>2</sub> emission factors for lignite Kolubara which have provided closer estimation of emissions than the original USA and EU factors. The optimized EU NO<sub>x</sub> emission factor is 40% lower than the original and USA NO<sub>x</sub> emission factor is 58% lower than the original factor. For SO<sub>2</sub> the improvements are much smaller, the optimized EU SO<sub>2</sub> emission factor is 17% lower than the original and the optimized USA SO<sub>2</sub> emission factor is only 7% lower than the original factor. Deviations from the original factors for NO<sub>x</sub> are consequence of already mentioned specific characteristics of lignite Kolubara (low heating value, high volatile, water, and ash content) and operating conditions of the boilers systems. For SO<sub>2</sub> the deviations from original emission factors are smaller, and they are within the margins of error defined in respective documents [1, 17].

Future investigations will be focused on determination of NO<sub>x</sub> and SO<sub>2</sub> emission factors for second Serbian lignite Kostolac.

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## Nomenclature

$A$	– coal ash content, [%m/m]	$(E_{NO_x})_{EUopt}$	– calculated NO <sub>x</sub> emission based on optimized EU emission factor, [kgh <sup>-1</sup> ]
$B$	– coal consumption of the unit, [th <sup>-1</sup> ]	$(E_{NO_x})_{meas}$	– measured NO <sub>x</sub> emission, [kgh <sup>-1</sup> ]
$EF_{iEU}$	– emission factor (European), [kgkg <sup>-1</sup> ]	$(E_{NO_x})_{USA}$	– calculated NO <sub>x</sub> emission based on USA emission factor, [kgh <sup>-1</sup> ]
$EF_{iUSA}$	– emission factor (American), [kgkg <sup>-1</sup> ]	$(E_{NO_x})_{USAopt}$	– calculated NO <sub>x</sub> emission based on optimized USA emission factor, [kgh <sup>-1</sup> ]
$E_i$	– reference emission, [kgh <sup>-1</sup> ]	$LHV$	– coal low heating value, [kJkg <sup>-1</sup> ]
$(E_{SO_2})_{EU}$	– calculated SO <sub>2</sub> emission based on EU emission factor, [kgh <sup>-1</sup> ]	$O_{2ave}$	– average dry flue gas oxygen content, [%v/v]
$(E_{SO_2})_{EUopt}$	– calculated SO <sub>2</sub> emission based on optimized EU emission factor, [kgh <sup>-1</sup> ]	$P_{el}$	– unit power (electrical), [MW]
$(E_{SO_2})_{meas}$	– measured SO <sub>2</sub> emission, [kgh <sup>-1</sup> ]	$S_{tot}$	– coal total sulphur content, [%mm <sup>-1</sup> ]
$(E_{SO_2})_{USA}$	– calculated SO <sub>2</sub> emission based on USA emission factor, [kgh <sup>-1</sup> ]	$\dot{V}$	– volume flow rate, [m <sup>3</sup> h <sup>-1</sup> ]
$(E_{SO_2})_{USAopt}$	– calculated SO <sub>2</sub> emission based on optimized USA emission factor, [kgh <sup>-1</sup> ]	$V_{fgdryntot}$	– total dry flue gas flow rate at normal conditions, [m <sup>3</sup> h <sup>-1</sup> ]
$(E_{NO_x})_{EU}$	– calculated NO <sub>x</sub> emission based on EU emission factor, [kgh <sup>-1</sup> ]	$W_{tot}$	– coal total moisture content, [%]

## References

- [1] \*\*\*, EMEP/EEA Air Pollutant Emission Inventory Guidebook 2009, Technical Guidance to Prepare National Emission Inventories, European Environment Agency, Copenhagen, 2009, ISBN 978-92-9213-034-3
- [2] \*\*\*, Step by Step Guideline for Emission Calculation, Record Keeping and Reporting for Airborne Contaminant Discharge, Ontario Ministry of the Environment, Toronto, Ont., Canada 2004
- [3] Nelson, P., Coal Nitrogen & NO<sub>x</sub>, ACARP Report No. 01, Australian Coal Association Research Program, Australia, 2006
- [4] Mereb, B., Assessing the Effects of Coal Properties on NO<sub>x</sub> Emissions in Combustion Systems, in: EPRI-DOE-EPA Combined Air Pollution Control Symposium: the MEGA Symposium, Vol. 2: NO<sub>x</sub> and Multi-Pollutant Controls, Atlanta, Geo., USA, 1999, EPRI-TR-113187-V2, Palo Alto, Cal., USA, pp. 14.1-14.14, 1999
- [5] \*\*\*, Procedures for Preparing Emission Factor Documents, EPA-454/R-95-015, USEPA, 1997
- [6] \*\*\*, Initial National Communication of the Republic of Serbia under the United Nations Framework Convention on Climate Change (Eds. D. Bozanic, M. Gasperic), The Ministry of Environment and Spatial Planning, Republic of Serbia, Belgrade, 2010
- [7] Stojiljkovic, D., et al. Devolatilisation and Combustion of Yugoslav Lignites, *Journal of the Energy Institute*, 78 (2005), 4, pp. 1-6
- [8] Mladenovic, M., et al., Criteria Selection for the Assessment of Serbian Lignites Tendency to form Deposits on Power Boilers Heat Transfer Surfaces, *Thermal Science*, 13 (2009), 4, pp. 61-78
- [9] Laban, K., Atkin, B., The Direct Determination of the Forms of Sulphur in Coal Using Microwave Digestion and i.c.p.-a.e.s Analysis, *Fuel*, 79 (2000), 2, pp. 173-180
- [10] Davidson, R., Quantifying Organic Sulfur in Coal: A Review, *Fuel*, 73 (1994), 7, pp. 988-1005
- [11] Singer, G., Control of Power-Plant Stack Emissions, in: Combustion Fossil Power (Ed. J. G. Singer), Combustion Engineering Inc., Windsor, Conn., USA, pp. 15.1-15.76, 1991
- [12] Okamoto, A., Evaluation of Thermal Coal Quality (Japan) (III), Coal and Safety (1998), No. 12, pp. 18-39
- [13] \*\*\*, Joint EMEP/CORINAIR Atmospheric Emission Inventory Guidebook, 1<sup>st</sup> ed. (Ed. G. McInnes), Copenhagen, European Environment Agency, CD-ROM, Feb. 1996
- [14] Davidson, M., Nitrogen in Coal, IEA Coal Research, IEAPER/08, London, 1994

- [15] \*\*\*, Preferred and Alternative Methods for Estimating Air Emissions from Boilers, Eastern Research Group Inc., 2001
- [16] Eggleston, H. S., *et al.*, IPCC Guidelines for National Greenhouse Gas Inventories, IGES, Japan, 2006
- [17] \*\*\*, AP-42, Compilation of Air Pollutant Emission Factors, Volume I, Stationary Point and Area Sources, Fifth Edition, 1997
- [18] \*\*\*, Regulation on Emission Limit Values (ELV), Method and Terms of Measurement and Data Recording, Official Gazette RS 30/97, Belgrade, 1997
- [19] \*\*\*, Single Measurements of Hazardous and Toxic Matter Emissions in Air from Unit A4 of TPP Nikola Tesla A in 2000, Mining Institute – Belgrade, Department for HVAC and Environmental Protection, Belgrade, 2001
- [20] \*\*\*, Single Measurements of Hazardous and Toxic Matter Emissions in Air from Unit A1 of TPP Nikola Tesla A in 2002, Mining Institute – Belgrade, Department for HVAC and Environmental Protection, Belgrade, 2003
- [21] \*\*\*, Single Measurements of Hazardous and Toxic Matter Emissions in Air from Unit B1 of TPP Nikola Tesla B in 2003, Mining Institute – Belgrade, Department for HVAC and Environmental Protection, Belgrade, 2004
- [22] \*\*\*, Single Measurements of Hazardous and Toxic Matter Emissions in Air from Unit B2 of TPP Nikola Tesla B in 2003, Mining Institute – Belgrade, Department for HVAC and Environmental Protection, Belgrade, 2004
- [23] \*\*\*, Single Measurements of Hazardous and Toxic Matter Emissions in Air from Unit B2 of TPP Nikola Tesla B in 2008, Mining Institute – Belgrade, Laboratory for environmental protection, Belgrade, 2008