CALCULATING GROUND CONCENTRATION AND DISTRIBUTION OF SO_2 BASED ON WARRANTY TESTS AFTER DESULFURISATION OF CHIMNEY IN THE THERMAL POWER PLANT KOSTOLAC B

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Abstract: *The most widespread and harmful pollutant for human health is sulfur dioxide. Coal power plants are the main pollutants of sulfur dioxide. In 2018, the coal power plants which were included into relevant National Emission Reduction Plan (NERP) in Western Balkans countries emitted a total of 603.988 tons of SO_2, which is six times more than permissible limit values for this region of 98.696 tons. According to the Climate Action Network (CAN), the Kostolac B thermal power plant, emits more SO_2 than permissible limit emission values of all countries in the West Balkans. For these reasons, the Electric Power Company of Serbia completed the flue gas desulfurisation plant in TPP Kostolac B in 2017, which reduces sulfur dioxide emissions. Today, there are no data about this plant. However, in this paper we analysed the impact of different scenarios (before and after the construction of the delufurisation plant) of sulfur-dioxide emissions from the TPP Kostolac B, applying the Gaussian dispersion model. Ground concentrations of SO_2 at different distances from the thermal power plant were calculated. Based on that, the paper presents the obtained results of flue gas dispersion and the distribution of ground SO_2 concentrations.*

Key words: *Kostolac thermal power plant, sulfur dioxide, Gaussian model;*

INTRODUCTION

Sulfur dioxide (SO_2), one of the most common air pollutants, results from the combustion of fossil fuels rich in sulfur content. Sulfur compounds, as pollutants, emit natural and anthropogenic processes into the atmosphere. Natural systems (atmosphere, biosphere, ocean systems) emit H_2S, COS, CS_2, DMS, SO_2, various sulfates and mercaptanes. About 1.5 million tons of sulfur dioxide is emitted into the atmosphere from natural sources annually, which makes only 1-2 percent of the total amount of SO_2 present. The remaining 98-99 percent come from human activity. In this way, an additional amount of about 140-150 million tons per year is emitted into the atmosphere. The most important anthropogenic sources of sulfur dioxide are fossil fuel combustion plants, especially coal plants. The sulfur content of fossil fuels varies from 1-5%. Most often, about 95% of the sulfur in the fuel is emitted as SO_2, 1-5% as SO_3, and 1-3% as sulfate particles. This is why thermal power plants are considered the largest SO_2 emitters. Sulfur dioxide and sulfur trioxide in combination with suspended particles and humidity have the most harmful effect on humans, living organisms and material goods. These wind-borne droplets fall to the ground, which leads to a gradual decrease in its pH value. Increasing the acidity of the soil slows down the growth of forests and other plants, and increasing the acidity of water has a detrimental effect on the growth of flora and fauna in the waters. As a consequence of the emission of sulfur dioxide, acid rains also occur, which results in a noticeable harmful effect on plants, and can also cause corrosion.

Sulfur dioxide in a mixture with solid particles and sulfuric acid (which has a higher irritant effect than sulfur dioxide) at an averange annual concentration of 0.04-0.09·10^{-5} and smoke concentration of 150-200 $\frac{\mu g}{m^3}$ leads to symptoms of heavy breathing and lung disease, and at an average daily concentration of 0.02-0.5·10^{-6} and smoke concentration of 500-700 $\frac{\mu g}{m^3}$ sudden increase in morbidity and mortality can occur. At a sulfur dioxide concentration of 0.3-0.5·10^{-6} within a few days, chronic disease can be found in both plant leaves (especially spinach, lettuce, cotton and lucerne) and conifer needles, which causes acid rain [1].
The highest concentration of SO_2, occurs in urban areas and large industrial centers. Average annual concentrations of SO_2, in urban areas of developing countries are 40-80 μg /m^3, in urban areas of North America and Europe 10-30μg/m^3, and in EU Member States 6- 35μg /m^3 [2].

SULFUR DIOXIDE EMISSIONS FROM KOSTOLAC THERMAL POWER PLANTS AND THEIR IMPACT ON THE ENVIRONMENT

The Kostolac thermal power plants are part of the PE "Electric Power Industry of Serbia", and they consist of two production units, the "Kostolac" mines and the "Kostolac A" and "Kostolac B" thermal power plants. The basic product is electricity that is delivered to the power system from four blocks of thermal power plants. The annual electricity production of all four blocks of Kostolac thermal power plants is close to 6 billion KWh per year.
By emitting pollutants, primarily flue gas and ash, Kostolac thermal power plants are the dominant source of atmospheric pollution. Flue gas is a product of fuel combustion, in this case coal, and is a mixture of gases (sulfur dioxide, nitrogen oxides, carbon monoxide, fluoride and chloride), the concentration of which depends on the characteristics of the fuel. In addition to gases, flue gas contains solid particles that are the product of incomplete combustion of fuel, such as soot, and the influence of mineral components in the fuel (ash). The most common element in the flue gas is sulfur dioxide with about 97%.
According to PE "Electric Power Industry of Serbia" data, concentrations of harmful substances, in four settlements around the Kostolac thermal power plant, in the period from June 2016 to February 2018, some of the values were higher than the allowed on average every fourth day. On the days when the measuring of sulfur dioxide (SO_2) was performed, values above the maximum allowed were determined 118 times. The measurements were performed by the *Public Health Institute in Pozarevac and the Mining Institute*. Measurements of pollutants at the source of pollution, in the thermal power plant, were done by experts from the *Vinca Institute of Nuclear Sciences.* Their reports stated excessive SO_2 emissions, which caused the presence of SO_2 in the atmosphere 10.5 to almost 15 times higher than allowed.
It is estimated that pollution caused by coal-fired power plants leads to almost 4,000 premature deaths in the non-EU Western Balkan countries, as well as more than 2,000 premature deaths in countries within the European Union. Six well-known European organizations and international coalitions dealing with ecology, climate change and energy (HEAL, Sandbag, CAN Europe, CEE Bankwatch, Europe Beyond Coal, Agora Energiewende) have presented a new report on the impact of coal-fired power plants in the Western Balkans. Air pollution from thermal power plants in the Western Balkans each year leads to 3,900 premature deaths, 8,500 cases of bronchitis in children, as well as to other chronic diseases. The cost of treatment is 11.53 billion euros. In the EU, this pollution causes health costs of € 3.1-5.8 billion, while the economies of the Western Balkans bear a burden of € 1.9-3.6 billion a year [3].
These facts are confirmed by the health status of the inhabitants of the Braničevo district, which includes Požarevac, Kostolac and seven smaller municipalities, and is unfavorable as shown by the results of the *Public Health Institute Požarevac*. In 2016, 29.4% of the inhabitants of the district were affected by diseases of the respiratory system, significantly higher than the national average of 16.8%, which was the biggest problem in the previous ten years. The circulatory system diseases are the leading cause of death, with 58.4% of the total deaths in 2016, compared to 51.7% at the state level. A quarter of children under the age of six in the Braničevo district had throat and tonsils inflammation, while in second place, with almost 19% of patients, was an infection of the upper respiratory system. As in previous years, school-age children (7-18 years) have the biggest problems with the respiratory system, and in 2016, every other child was treated for such problems [4].

RESULTS OF WARRANTY TESTING OF THE DESULFURIZATION SYSTEM IN TPP KOSTOLAC B

The application of laws in the field of environmental protection implies the compliance of our regulations with the regulations of the European Union. One of the priority tasks is the adoption of measures and procedures for air protection

by reducing the emission of hazardous substances at the source of pollution. Considering the limit values of SO_2 emissions in flue gas, the construction of a system for desulfurization of SO_2 in Kostolac thermal power plants with regard to the installed capacity, concentrations in the air should be reduced more than 10 times, at full load of blocks and at combustion of worst-quality coal. This is one of the most important criteria that influenced the selection of TPP Kostolac B for the first thermal power plant in EPS in which a desulfurization system has been built. The construction of a system for desulfurization of SO_2 concentrations in the air should be reduced to 200 mg / Nm^3 or less, as provided by the European Directive on Industrial Emissions, which has been applied since 2016. As the best solution, a wet flue gas desulfurization process was chosen, using limestone for SO_2 absorption, whereby commercial quality gypsum was obtained as the final by-product of the absorption process. This procedure is based on spraying a suspension of lime or limestone into the flue gas, with the aim of converting SO_2 into a potentially commercial final product [5].
The Chinese company CMEC has realized the project for constructing a plant for desulfurization of flue gases of blocks B1 and B2 of the Thermal Power Plant "Kostolac". The construction works of the desulfurization system lasted from 2014 to 2017. The guaranteed mass concentration of sulfur dioxide at the outlet of the desulfurization plant is ≤ 200 mg /m^3 (dry gas, 6% O_2).
After the construction and commissioning of the flue gas desulfurization plant, warranty measurements were performed in March and April 2017 at the outlet of the flue pipes of block B1 and block B2. The measurements were performed in accordance with the Decree on measurements of emissions of pollutants into the air from stationary pollution sources ("Official Gazette RS", No. 05/16). Warranty tests at TPP "Kostolac B" at the waste gas desulfurization plant on block B1, were performed by the Mining Institute Belgrade, in March 2017.

Table 1: projected SO_2 values at the inlet and outlet of the desulfurization system[1]

parameter	units	Values at the plant inlets		Values at the plant outlets	
		Kostolac B1	Kostolac B2	Kostolac B1	Kostolac B2
SO_2	mg/Nm^3	5619	7661	30	200

The results of measuring the trial operation of the desulfurization system in March 2017 were presented on the basis of the report of the Laboratory for Environmental Protection and Work Safety of the Mining Institute from Belgrade.

Table 2: Desulfurization rates on Blocks B1 and B2 after the warranty tests[2]

TPP Kostolac B	C_0, mg/Nm^3	C_1, mg/Nm^3	S, %
Block B1	5439,6	55,3	99
Block B2	5419,9	143	97,4
Mean value			98,2

The calculation determined that the desulfurization rate in block B1 is in the range of 98.4-98.8%, and in block B2 in the range of 97.3 - 97.5%, which is within the limits [6].

GAUSSIAN MODEL FOR CALCULATING THE EMISSION AND DISPERSION OF SULFUR DIOXIDE FROM THE KOSTOLAC B THERMAL POWER PLANT, BEFORE AND AFTER FLUE GAS DESULFURIZATION

Mathematical modelling is one of the scientifically accepted methods for calculating the propagation of gaseous substances from stationary sources. By applying a mathematical model based on data on a stationary source, topographic, climatic and urban characteristics of the investigated area, it is possible to see the dispersion of gaseous substances through different scenarios. The number of parameters and the model used to assess the dispersion effect of gaseous substances may vary depending on the modelling category used.
The paper analyzes the data on sulfur dioxide emissions from the chimney of TPP Kostolac B in December 2008 and the sulfur dioxide emissions of the same thermal power plant with the desulfurisation plant during the April 2017 trial period. The Gaussian statistical model was applied to estimate the ground dispersion of gaseous substances for certain values of gaseous emissions and parameters related to the external environment. The basis of this model is the assumption that the impurities emitted by a continuous point source form a smoke column in which a symmetrical distribution of the particle concentration in relation to the axis of the smoke column is noticed. In analyzing the impact of the Kostolac B thermal power plant on air quality (mean one-hour concentrations), the software of the US Environmental Protection Agency SCREENVIEW 4 was

[1] [6] Environmental impact assessment study in Kostolac B, 2018
[2] Mining Institute D.O.O. Belgrade, Laboratory for Environmental Protection and Work Safety

used, which allows estimation of the pollution concentration at the ground level originating from a single source. The following input quantities were used to calculate the sulfur oxide concentrations:
1. Chimney height
2. Inner diameter of chimney
3. Mass flow rate and flue gas velocity at chimney outlet
4. Flue gas temperature at the outlet
5. Terrain characteristics around thermal power plants (rural and urban areas, absolute height, topographic features)
6. Condition of the atmosphere in terms of stability and wind speed.

Table 3: Production parameters at TPP Kostolac B in December 2008. [3]

PARAMETER	KOSTOLAC B
Chimney height [m]	250
Inner diameter of chimney at the outlet [m]	9,8
Fuel gas temperature at the outlet [°C]	170
Fuel gas velocity at chimney outlet VS [m^{-1}]	25,7
Mass flow rate [g/s]	1410, 8

The data which were used in modelling for calculating the ground concentration of air pollution were gathered from multy-year measurements by the Republic Hydrometeorological Institute of Serbia at the Main Meteorological Station Veliko Gradiste, 25 km east of the thermal power plant, which is climatologically representative of the area . The average monthly temperature in December 2008 in the Kostolac basin was 3.300 C. In December of the same year, stability class D and stability class F predominated. The calculation indicated that the terrain is characterized by lowland and plain features. It was also adopted that TPP Kostolac B is located in a rural area. After the performed analyzes for the stated parameters, the results are as follows.

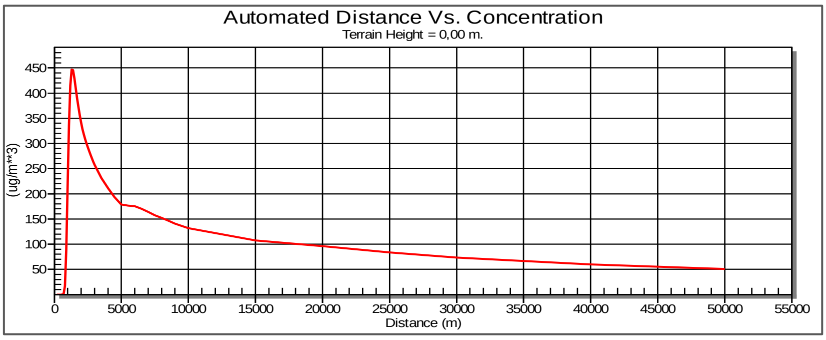

Figure 1: SO_2 concentration depending on source distance at Kostolac B thermal power plants in December 2008.

Figure 1 shows that at TPP Kostolac B the maximum concentration is 410.2 μg/m^3 at a distance of 1381 m. The SO_2 concentration is above the limit and is 150 μg/m^3 at a distance of 600 m to 7500 m. Based on the results of the test measurements of the trial run of desulfurisation system in April 2017 which were presented in the report of the Laboratory for Environmental Protection and Work Safety of the Belgrade Mining Institute we defined the production parameters.

Table 4: Production parameters of TPP Kostolac B in the trial run of the desulfurisation system[4] (April 2017)

PARAMETER	KOSTOLAC B (blocks B1 I B2)
Chimney height [m]	180
Inner diameter of chimney at the outlet [m]	13,4
Flue gas temperature at the outlet [^{0}C]	63,8
Flue gas velocity at chimney outlet vs [m/s]	21
Mass flow rate (g/s)	80,8

[3] Đorđević-Miloradović J. et al., 2012.
[4] Mining Institute D.O.O. Belgrade, Laboratory for Environmental Protection and Work Safety, 2018

When modelling to calculate the concentration and distribution of sulfur dioxide after using the desulfurisation system, we used meteorological data for April 2017, for the main meteorological station in Veliko Gradiste. The average monthly temperature in April was 11.7 ^{0}C. The maximum temperature in April of the same year was 18.3 ^{0}C and the minimum 3.2 ^{0}C. In this area, the prevailing wind direction is south - southeast and southeast, followed by winds from west and west-north-west direction. In April, stability class D and stability class F prevailed. Other parameters, such as the relief and urbanization of space, were identical to those of the previous modelling. The same input values were used to calculate the concentration of sulfur oxides in the trial run of the desulfurisation system. Modelling obtained the following results.

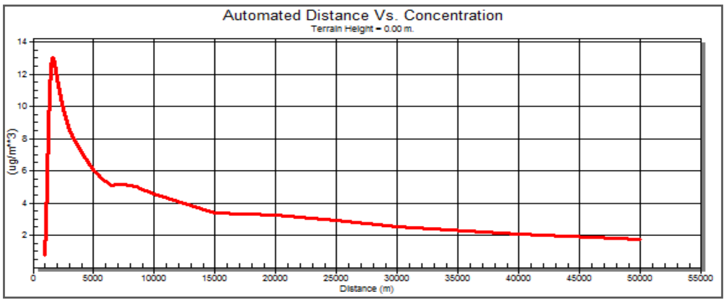

Figure 2: SO_2 concentration depending on source distance at Kostolac B thermal power plants in April 2017 (after chimney desulfurisation)

Figure 2 shows that the SO_2 concentration is more than 90% lower than in 2008. The maximum concentration is 13.06 μg/m^3 at a distance of 1642 m. The modelling results show that the flue gas desulfurisation system fully meets the requirements for the reduction of ground-level SO_2 concentrations in the area around the source of the pollution. Comparisons of the effects on air quality of certain atmospheric stability classes, for different wind speeds and different temperatures have also been made. In December 2008, atmospheric stability classes D and F prevailed with the highest percentage, therefore, comparisons were made for these stability classes.

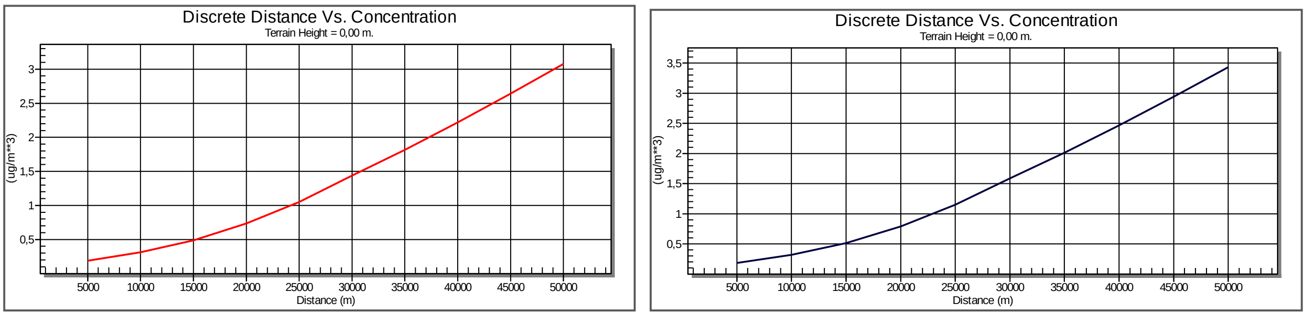

Figure 3: Distribution of SO_2 concentrations for stability class F at 3.3^{0} C and wind speed of 1m / s (left) and wind speed of 4m / s (right)

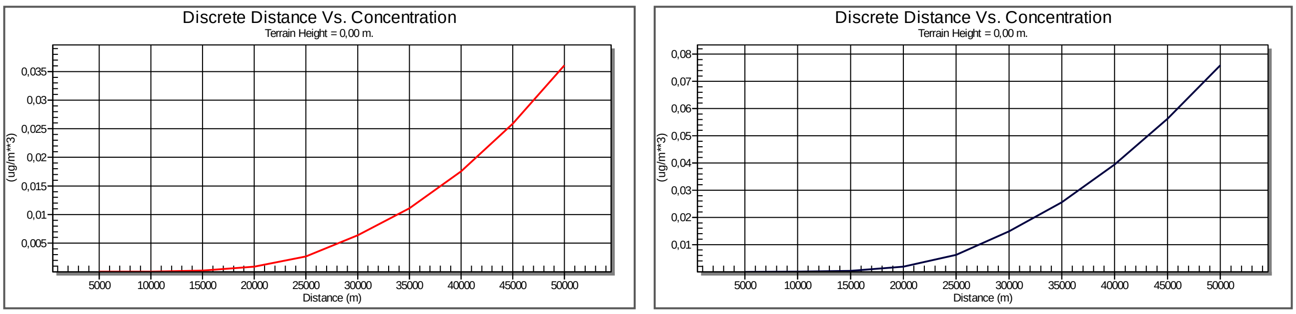

Figure 4: Distribution of SO_2 concentrations for stability class D at 3.3^{0}C (left) and 11.26 ^{0}C (right)

The graphs in Figures 3 and 4 show that an increase in ambient air temperature for the same atmospheric stability class leads to an increase in the concentration of pollutants compared to the lower temperature values. Although higher wind speeds should lead to lower concentrations, the modelling results indicate an increase in concentration with an increase in wind speed for the same atmosphere stability class. This indicates that other factors affecting the distribution are prevalent. In windy periods, aeolian erosion of the surface occurs, raising and carrying tiny ash fractions. Measurements have shown that the largest exceed of deposition of particulate matter are in the towns of Stari Kostolac and Kostolac, which are closest to the ash dump. In the case of the Kostolac B thermal power plant, the largest pollutants are surface sources of the discovery, open limestone landfills, ashes and gypsum landfill. Landfill particles generally pollute the immediate environment. The graphs in figures 3 and 4 show that, under the same conditions, the concentration of pollutants is higher for the atmospheric stability class D (neutral) than for the class F (very stable). In the comparative analysis of the results when the desulfurization system operates, examples are given for the two most common stability classes D and F, with an average air temperature in April of 11.7^{0}C and an average wind speed of 2.2 m / s.

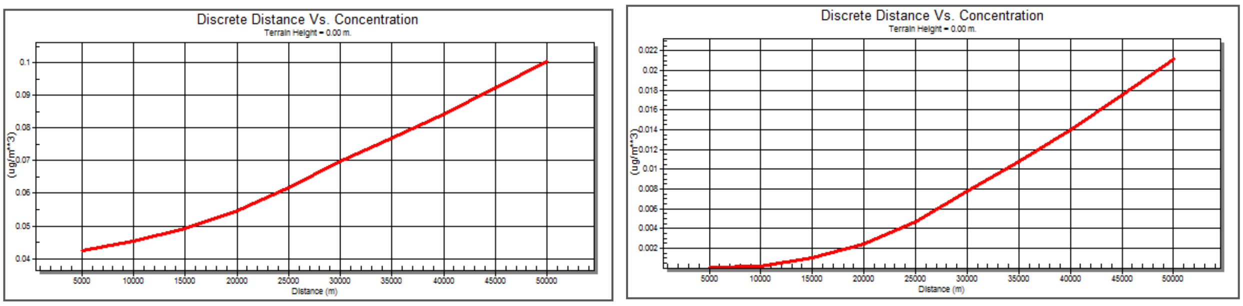

Figure 5: Distribution of SO_2 concentrations for stability class D at 11,7^0 C and wind speed of 2.2m / s (left) and stability class F for temperature of 11.7^0 C and wind speed of 2.2m / s (right)

The maximum SO_2 concentration for stability class D is 0.1006 μg/m^3 at a distance of 50000 m, and for stability class F 0.021 μg/m^3 also at a distance of 50000 meters. In both cases, it is observed that SO_2 concentrations are extremely low. Maximum concentrations occur at the same distances from the emission source. There is no pollution in Class D at ground level, which confirms the theory that in this class there is minimal soil pollution if the chimney is high enough and the terrain is flat. The increase in pollution is observed at a distance of 5 km away from the chimney. Stable atmospheric conditions, class F, cause more resistance to vertical movement and thus less dispersion in the vertical direction. The increase in SO_2 concentration is at a distance of about 8800m away from the chimney. We also analyzed the impact of wind on the spread of pollution for stability class F. We compared the maximum and minimum wind speeds in this area. According to meteorological data, the maximum wind speed for the ten-year period was 3.8 m/s and the minimum was 1.3 m/s. As a temperature value, we took the monthly average for April 2017, which was 11.7 ^{0}C.

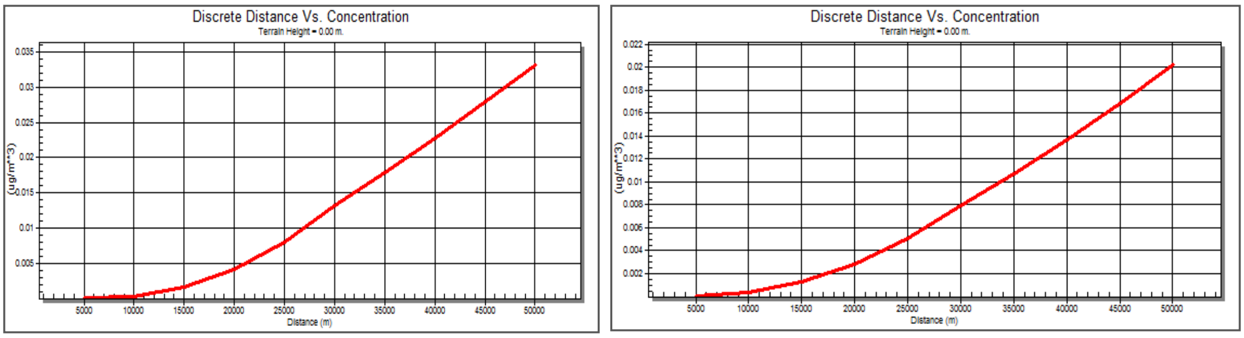

Figure 6: SO_2 concentration distribution for stability class F at wind speeds of 3.8 m/s (left) and 1.3 m/s (right)

As with sulfur dioxide spread analysis (Figure 1) in December 2008, the modelling results indicate an increase in the concentration of pollution with an increase in wind speed for the same atmosphere stability class. The maximum concentration of SO_2 when the wind speed is 3.8 m/s is 0.3313 μg/m^3 and at a wind speed of 1.3 m/s it is 0.2021 μg/m^3. There is also a difference in the distance of the increase in pollution from the source. With stronger winds, the increase in pollution starts at a distance of 10 kilometers, and with the lower wind speed at 7.5 kilometers.

The results obtained by modelling sulfur dioxide spread from TPP Kostolac B show that the desulfurisation system with a new wet chimney fully meets the requirements prescribed by law and that the ground concentrations around the emitters are minimal. Based on the results of SO_2 emissions in the trial run, the newly built desulfurisation system will significantly reduce the sulfur dioxide emissions from TPP Kostolac B. According to our calculation, the desulfurisation rate at TPP Kostolac B is 94.3%.

CONCLUSION

Applying the Model of the US Environmental Protection Agency SCREENVIEW 4, we compared the effects of one source under different operating conditions on air quality (mean hourly concentrations), operation of the Kostolac B thermal power plant without a desulfurisation plant and operation with a desulfurisation plant and a new wet chimney as an emitter. Examples of the results of the performed analysis for the two most common stability classes in the measurement period are given. Comparisons have been made for sulfur dioxide emissions which have the greatest impact on air pollution. The results obtained indicate that the construction of the desulfurisation system reduced the emissions of sulfur dioxide by 94.3%, on average for both units, thus fulfilling the condition that the minimum degree of desulfurisation according to the Regulation on limit values of pollutant emissions into the air from the combustion plant ("Official Gazette RS", No. 6/16), for existing large combustion plants with a thermal power exceeding 500 MWth, is 94%. This was reflected in the mean one-hour concentrations obtained by modelling. They are well below 200 mg/Nm^3 or less, as foreseen by the European Industrial Emissions Directive, which has started to apply since 2016. The distributions shown represent the usual results of modelled dispersion. For example, an increase in air temperature leads, as expected, to an increase in the concentration of sulfur dioxide compared to values at a lower temperature. It is also confirmed that higher wind speeds do not contribute to lower concentrations. Maximum ground concentrations occur at distances of about 50 km from the source (chimney), at appropriate wind speeds. The release of the flue gas desulfurisation system at TPP Kostolac B will significantly reduce the sulfur dioxide emissions of the Kostolac energy complex, thereby, significantly improving the quality of the environment, which is greatly endangered.

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