Investigating the Impacts of Winds on SO₂ Concentrations in Bor, Serbia

Viša Tasić ^{*1}, *Renata Kovačević* ², *Novica Milošević* ² ¹Department of Industrial informatics Mining and Metallurgy Institute, Bor, Serbia e-mail: <u>visa.tasic@irmbor.co.rs</u>

> ²Department of Chemical and Technical Control Mining and Metallurgy Institute, Bor, Serbia

Cite as: Tasić, V., Kovačević, R., Milošević, N., Investigating the Impacts of Winds on SO₂ Concentrations in Bor, Serbia, J. sustain. dev. energy water environ. syst., 1(2), pp 141-151, 2013, DOI: TBA

ABSTRACT

Air pollution is one of the most important environmental problems in the town of Bor, situated in the eastern of Serbia. The main source of air pollution with sulphur dioxide, heavy metals and other toxic and carcinogenic elements present in particulate matter (PM) and aero sediments is the Copper Mining and Smelting Complex Bor. The distribution of air pollutants is mainly determined by the copper smelter operation mode, as well as by meteorological parameters such as wind speed and direction. While information on air pollution with heavy metals and carcinogenic elements in the form of PM has been monitored and reported for longer period, availability of data related to the relationship between air pollutants and meteorological parameters in the Republic of Serbia is still limited. In the present study, the relationships between daily mean concentrations of SO_2 and the speed and wind direction were analysed. The analysis includes data collected in 2011 from the four monitoring stations (Park, Jugopetrol, Institute, and Brezonik) situated in a wider town area. Pearson correlation coefficients between daily average SO₂ and daily average speed and direction of winds are calculated. High wind speed reduces SO₂ concentration due to the dilution effect. Hence, correlations between SO₂ and the speed and direction of winds at almost all monitoring stations were negative, with one exception (Jugopetrol). This could be explained by the fact that winds, blowing from the north-west (NW) direction, carry polluted air towards this station. Moderate negative correlation found between SO₂ and wind direction at monitoring station Park. Generally, the observed correlations between SO_2 and wind speed and direction are weak, due to frequent changes in SO₂ emissions, wind speed and direction during the day.

KEYWORDS

Air Pollution, Sulphur-dioxide, Meteorology, Wind Speed, Wind Direction, Monitoring

INTRODUCTION

Urban air pollution is an important issue with different socio-economic and climatic aspects in different parts of the World. In the past century, most of the World population has moved to cities. Today, more than 75% of all people in the developed countries live in cities, and urbanization is a rapidly spreading process in the developing world. The origin of urban air pollution is mainly in anthropogenic emission sources, which include vehicles, industries, and domestic fuel combustion. Increased combustion of fossil fuels in the last century is responsible for the progressive change in the atmospheric composition. High concentrations of air pollutants are sufficient to cause adverse health effects, including increased morbidity or mortality [1 - 5]. Air pollution has both acute and chronic effects on human health, affecting a number of different systems and organs.

The Municipality of Bor is placed in a mountainous and forest area in the south-eastern part of Serbia, east from mountain Crni Vrh, south-east of mountain Veliki Krš and south of mountains Stol and Deli Jovan. It is located less than 50 km far from Bulgarian and less than 100 km far from Romanian borders (as shown in Figure 1). It has a total population of 50,000 citizens. The area has been the major centre for mining and processing of copper and other precious metals for more than a century. Air pollution is perceived as the main environmental problem in the Municipality Bor. The main source of air pollution with SO₂, heavy metals in PM and aero sediments are the copper smelter. The smelter operates within the RTB Bor Company (Copper Mining and Smelting Complex) which producing copper for more than 100 years [6 - 9].



Figure 1. The area of eastern Serbia with location of the Municipality Bor [9]

The SO₂ is one of the most important environments polluter. It mostly originates from oxidation of sulphur compounds. Anthropogenic emission of SO₂ results from burning the fossil fuels (coal and heavy oils) or smelting of sulphide ore concentrates (most frequently Cu, Pb, and Zn ores). Volcanoes and oceans are its major natural sources. Still, SO₂ is an irritant gas that causes breathing problems when people are exposed to its high concentrations. SO₂ as well as all SO_x gases can react with other compounds in the atmosphere to form small particles. These particles penetrate deeply into sensitive parts of the lungs and can cause or worsen the respiratory disease, such as emphysema and bronchitis, and can aggravate existing heart disease, leading to increased hospital admissions and premature death. Because of certain negative effect of SO₂ in the atmosphere, European Union issued the limits of its mass contents. Limit values and threshold alerts are as follows [10, 11]:

- limit value per hour for protection of human health is 350 μ g/m³, not to be exceeded more than 24 times per calendar year;
- daily limit for protection of human health is $125 \ \mu g/m^3$, not to be exceeded more than three times per calendar year;
- annual limit for protection of ecosystems is $20 \ \mu g/m^3$.

• alert threshold is set to 500 μ g/m³, measured over three consecutive hours at locations representative of air quality over at least 100 km² or specified air quality management zones, whichever is the smaller.

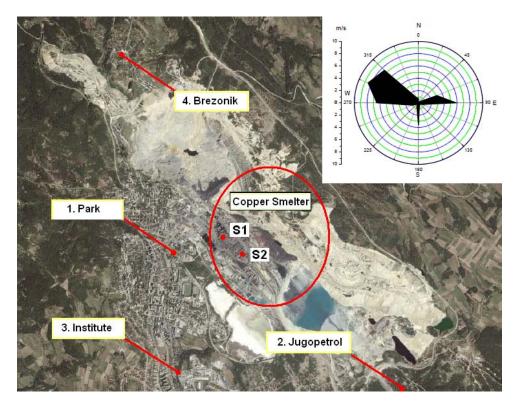


Figure 2. Map of the Municipality Bor area together with the locations of the copper smelter, monitoring stations (1. Park, 2. Jugopetrol, 3. Institute and 4. Brezonik) and wind rose diagram (2001-2011)

The technology of copper production in the copper smelter in Bor is outdated (classic pyrometallurgy with melting in furnaces and utilization of SO₂ gas in production of H_2SO_4 with a relatively small degree of utilization <60%) which leads to the environmental pollution of higher concentrations of SO₂ and particulate matter [8, 9] as well as aero sediments ($PM > PM_{10}$). The ore melted in the copper smelter plant in Bor is of chalcopyrite-pyrite type with the increased contents of arsenic, which is found in the form of FeAsS and Cu₃AsS₄. The oxidation, roasting and melting of such a mineral forms results in increased heavy metal's oxides and SO₂ gas which in certain quantities contaminate the environment. Reportedly, 170,000 to 250,000 tons of SO₂ are emitted to the atmosphere each year [6, 7 and 9]. There are two factory smokestacks in the Copper Smelter Plant in Bor. They are shown in Figure 2 as smokestacks S1 and S2. The height of smokestack S1 is 120 m (D = 3 m) for the Smelter Plant off-gasses with contents up to 1% SO₂. The other of 150 m (D = 3.5 m) is used for gasses when the Sulphuric Acid Plant is out of operation. In addition, gases resulting from the roasting procedure in fluo-solid reactor mixed with converter gases with SO₂ content of 5-6% in the gas. Both smokestacks are situated in the immediate vicinity of the urban settlement at a distance less than 500 m from the old urban centre with numerous vital functions of the town. In addition, airborne dust resulting from the open pits and surrounding waste heaps, which contains heavy metals, contributes to the air pollution of the area. Taking into account the location of the industrial complex and dominant wind directions, these pollutants are spread over the Bor town and its surrounding area. Therefore, the inhabitants in the town

of Bor are exposed to high levels of air pollution [9]. It is known that the distribution of air pollutants emitted from the copper smelter, is strongly influenced by the smelter operation mode and meteorological parameters such as wind speed and direction. There are numerous studies reported in the literature, which statistically determine the effects of meteorological parameters on SO_2 concentrations [12 - 16]. The influence of some meteorological parameters (wind speed and directions, relative humidity and temperature) on daily SO₂ concentrations at the four measuring sites in Bor town area during 4 years (2005–2008) was analysed in [16]. According to the values R^2 (cumulative), it can be said that meteorological factors (temperature, relative humidity and maximum wind gust) are not able to disperse and clean the pollutant concentrations completely from the atmosphere. The greatest influence of these three meteorological variables is at the measuring site Brezonik where R^2 (cumulative) is 10.3% [16]. Our analysis includes data collected in 2011 from the four monitoring stations (Park, Jugopetrol, Institute, and Brezonik) situated in Bor town area. Meteorological data (wind speed and directions) were used from 3 automatic measuring stations (Park, Brezonik and Institute) while in [16] meteorological data were used from one station, equipped with conventional measuring instruments, situated near Institute. Furthermore, in 2011 the copper production in the smelter was higher (3,000 tonnes of cathode copper per month on average), compared to the period from 2005 to 2008 observed in [16]. Correlation analyses were carried out to better quantify the relationship, if any, between daily average concentrations of sulphur dioxide and daily average winds speed and directions. In the cold season (October – March) the higher percentage of time without wind (65%) was detected, compared to the warm period (April – September) of the year (45%) in the time interval from 2005 to 2011. However, it was proved that diurnal changes in wind speed and wind direction fallowed the same pattern during cold and warm period of year [15]. It should be noted that the copper smelter running during night-time also. The emissions of SO_2 from the smelter are discontinuous and emissions have been changing hourly [14].

EXPERIMENTAL SETUP

Instrumentation

The equipment, used during the measuring period, consists of the four fixed stations for monitoring the ambient SO₂ concentrations and meteorological parameters. The Serbian Environmental Protection Agency (SEPA) has started measuring the air pollution with automatic monitors in 2006. Nowadays, SEPA operates with 37 automatic monitoring stations at the entire territory of the Republic of Serbia. Three of them are installed in the Bor town area during 2007-2010. The first was placed at Brezonik in the summer of 2007. It is equipped with HORIBA APSA-370 SO₂ analyser [17]. The second was placed at Park in spring of 2009. This station also contains HORIBA APSA-370 SO₂ analyser. The third station was placed at Institute in autumn of 2009. It contains EAS ENVIMET SO₂ analyser, Model 100E [18]. The forth monitoring station, placed at Jugopetrol in spring of 2009, operated by Municipality of Bor, is equipped with DKK TOA GFS-312E SO₂ analyser [19]. The reference method for sulphur dioxide measurement is described in [20]. Using the UV-fluorescence method, the analysers perform automatic measurements of SO₂ in the ambient air in a concentration range from zero to 10,000 μ g/m³ with linearity of ±1% and minimum detectable limit (2 σ) < 3 μ g/m³ [21]. Analysers are calibrated with the standard gas mixtures (200 ppb-500 ppb) from the certificated gas cylinders. All abovementioned SO₂ analysers have an auto calibration facility (zero and maximum calibration), definition of arbitrary interval sampling, averaging and saving the measured values. The data about SO₂ concentrations and meteorological parameters are available as 10-min, 15-min, 30-min or 1-hour average values. The analysers operate in accordance with the equipment manual. Maintenance schedules for replacement of consumable parts, diagnostic checks, and equipment fully follow the manufacturer recommendations. Routine and non-routine service visits are documented as well as the results of analyser tests or calibrations performed during monitoring programs. Data validation is performed in three-month intervals to ensure that they are reliable and consistent.

Measuring locations

The monitoring stations were installed close to the copper smelter (as shown in Figure 2) in positions downwind from prevailing wind directions. The prevailing winds are from west-northwest and therefore, tend to carry away the pollution from the main population centres (wind rose diagram in the time interval from 2001 to 2011 is also shown in Figure 2). During rainy periods, typical east or southeast winds are of more concerns. Low or zero wind conditions occur regularly (more than 50% of time). Light and variable winds are likely to cause very high localized concentrations of pollutants [7].

Monitoring station 1 Park – station is located within the Town Park, 650 m west from the copper smelter. A dense population (mainly high-rise) is directly downwind from the copper smelter during east winds. This station is also equipped with meteorological instrumentation.

Monitoring station 2 Jugopetrol – station is located 2 km south/southeast from the copper smelter. Close to the measuring point (1 km north-east) is the city dump.

Monitoring station 3 Institute – station is located about 2 km south/southwest from the copper smelter. This station is also equipped with meteorological instrumentation.

Monitoring station 4 Brezonik – station is located about 4 km north/northwest from the copper smelter. This station is also equipped with meteorological instrumentation. In the vicinity of the station is an open pit (1km south-east, as shown in Figure 2).

RESULTS AND DISCUSSION

Measurements of the SO_2 mass concentration levels were carried out in 2011, at the time intervals when the copper smelter was in operation, as well as at the time intervals when the Copper Smelter Complex Bor did not work continuously. Daily averages, used for the statistical considerations, are calculated from the 15-min and hourly averages. For calculation of daily averages, minimum 80% of the 15-min or 90% of hourly averages is required; otherwise, the value is considered as the missing one. Time series with daily mean concentrations of SO₂ are presented in the Figures 3-6 together with the wind rose diagram at each measuring point. Strong fluctuations of daily mean SO₂ mass concentrations that can be observed are mostly related to the changes in the weather conditions (wind speed and direction). This phenomenon is typical for meteorological conditions with the wind speed less than 2-3 m/s and such wind direction that causes the detectable air pollution at the monitoring stations [7]. The peaks over daily limit values of SO₂ usually occurred due to very high concentrations over a period of several hours during the day [7]. In addition to high concentration of SO_2 , at the measuring locations Park and Jugopetrol there are a large number of days with daily SO₂ concentration levels above the limit. According to the SEPA annual report, during the 2010 exceeding of the limit value of SO₂ concentrations have occurred at all measuring points in the Bor town, at some sites even over 150 days a year (Park) [22]. The same situation was also repeated in 2011, at measuring point Park, during 162 days, the SO₂ concentrations were above the limit value [23].

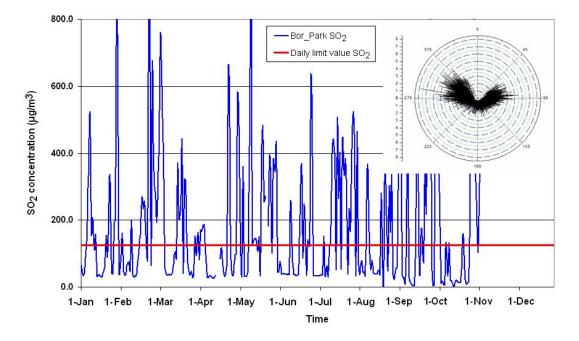


Figure 3. Time series of daily mean SO₂ levels at the measuring location Park together with the wind rose diagram in 2011

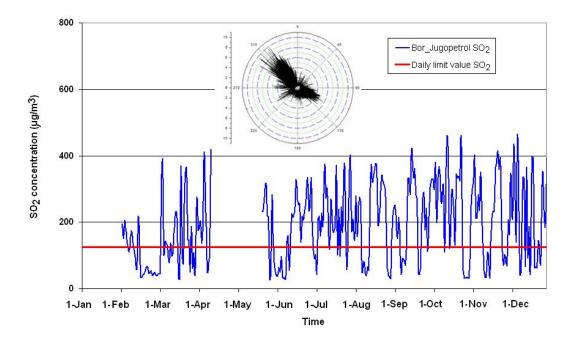


Figure 4. Time series of daily mean SO₂ levels at the measuring location Jugopetrol together with the wind rose diagram in 2011

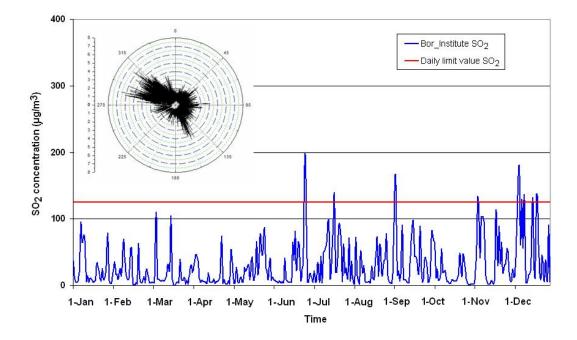


Figure 5. Time series of daily mean SO₂ levels at the measuring location Institute together with the wind rose diagram in 2011

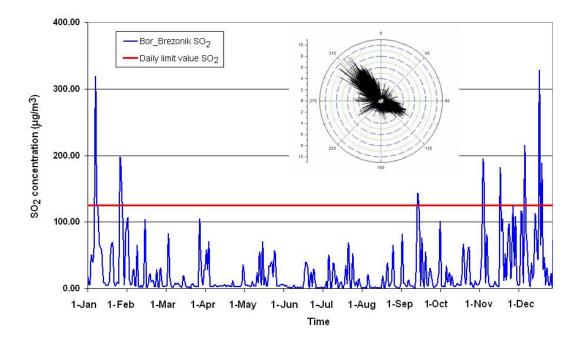


Figure 6. Time series of daily mean SO₂ levels at the measuring location Brezonik together with the wind rose diagram in 2011

A relationship between the SO_2 concentrations levels at the different measuring locations and the meteorological parameters of wind speed and direction was investigated by linear regression analysis. As seen in Table 1, the correlation of SO_2 concentrations with the wind speed and direction is not a similar for all measuring locations. The correlations between SO_2 concentrations at different measuring locations

are generally weak. The moderate correlation occurs only between the SO_2 concentrations at the measuring locations Brezonik and Institute (r = 0.514, n = 350, p < 0.001), and between the SO₂ concentrations at the measuring locations Park and Institute (r = 0.383, n = 330, p < 0.001). This could indicate a single source of SO₂ pollution dominant for the specified measuring locations in the time intervals with the slow winds, or without wind (calm weather).

 Table 1. Pearson correlation coefficients between daily mean SO2 concentration levels and meteorological parameters of wind speed and direction

	SO ₂ Park	SO ₂ Jugopetrol	SO ₂ Institute	SO ₂ Brezonik
SO ₂ Park	1			
SO ₂ Jugopetrol	0.009	1		
SO ₂ Institute	0.383	0.236	1	
SO ₂ Brezonik	0.168	0.150	0.514	1
Wind Speed	-0.107	0.092	-0.352	-0.414
Wind Direction	-0.551	0.307	-0.142	-0.077

Negative correlation between the SO₂ concentrations and wind speed is detected with exception at Jugopetrol (r = 0.092, n = 300, p < 0.001). This situation shows that when the wind speed is high, air pollution dilute by dispersion. The same conclusion applies in case of correlation between the SO₂ concentrations and wind speed at the measuring locations Institute and Brezonik. An exception that occurs at Jugopetrol can be explained by the fact that winds blowing from the northwest direction (winds that are with the highest frequency and maximum speeds as shown in Figure 4) carries air pollution in the direction of Jugopetrol.

Negative correlation between the SO₂ concentrations and wind direction is detected with exception at Jugopetrol (r = 0.307, n = 300, p < 0.001). An exception that occur at Jugopetrol can be explained by the fact that winds blowing from the north-west direction, that are with the highest frequency, carries air pollution in the direction of Jugopetrol. It was found that the moderate negative correlation occurs between SO₂ concentrations at Park and wind direction (r = -0.551, n = 330, p < 0.001). It could be explained by the fact that the most frequent wind direction that occurs at the measuring location Park improves the reduction of air pollution.

CONCLUSION

Monitoring of mass concentrations of SO_2 is very important from the aspect of risk assessment to human health, having in mind that SO_2 gas has negative effect to the completely human health especially to health of the children and older person. The aim of this study was to correlate data about daily average concentrations of sulphur dioxide and daily average winds speed and direction to better quantify the relationship between them. According to the shown results, it could be noted that the citizens of the Municipality Bor were exposed to the high concentrations of SO_2 and not so rare to the extremely high concentrations in 2011. The SO_2 concentration levels measured at the all four measuring locations in the Municipality Bor area exceeded the corresponding Serbian and European Union air quality standards. The lowest SO_2 levels were observed at Brezonik and Institute that is the least impacted by the Copper Smelter Complex Bor emissions of SO_2 . The annual average concentrations of SO_2 observed at Park and Jugopetrol are the greatest in the Republic of Serbia and in Europe as well. Therefore, Bor Municipality area is considered being one of the most polluted regions in Serbia and in Europe with SO₂. The Copper Smelter Complex Bor activities, coupled with calm weather are responsible for high SO₂ concentrations in the ambient air. It is proved that the calculated correlations between the average daily SO₂ concentrations and wind speed and direction are weak. The main reason for the poor correlation is frequent changes in speed and direction of wind, and the discontinuous emission of waste gases from the smelter. Even though, our findings talked about how wind speed and the dominant wind direction are important parameters for calculating the dispersion of pollutants. Negative correlations between the SO₂ concentrations at sites Park, Brezonik and Institute and wind direction are detected. This situation shows that when wind speed is high, pollutants dilute by dispersion. Positive correlation occurs between the SO₂ concentrations at Jugopetrol and wind direction. As stated above, the most frequent wind direction that occurs at Jugopetrol improves the air pollution. The most polluted part of the town is old town centre (near site Park), where more than 50% of people live and work. This situation of ambient air quality warrants the need to take serious steps to improve it.

ACKNOWLEDGMENT

This work is supported by a Grant from the Ministry of Education, Science and Technological Development of the Republic of Serbia, as a part of the Project III-42008: "Evaluation of Energy Performances and Indoor Environment Quality of Educational Buildings in Serbia with Impact to Health", within the framework of the Technological Development Program. We wish to thank the Serbian Environmental Protection Agency (SEPA) for assistance in technical issues and providing the useful pollutant and meteorological data.

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Paper submitted: 03.03.2013 Paper revised: 16.03.2013 Paper accepted: 16.03.2013