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RESEARCH ARTICLE

SEASONAL VARIATIONS OF SULFUR DIOXIDE IN THE AIR IN ZENICA CITY DURING 11 YEARS PERIOD 2006 – 2016.

Halim Prcanovic¹, Sefket Goletic², Mirnes Durakovic¹ and Sanela Beganovic¹.

1. University of Zenica, Metallurgical Institute “Kemal Kapetanović”, Zenica, Bosnia and Herzegovina.
2. University of Zenica, Mechanical faculty, Zenica, Bosnia and Herzegovina.

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Abstract

This study presents the results of eleven years of measurement of a sulfur dioxide campaign, in three places in Zenica City between January 1 2006 and December 31 2016. This Study was performed to analyze the periods of activation/deactivation of domestic heating as a source of pollution. Sulfur dioxide was measured using the British standard number 1747 (24-hour samples). Analysis of temporal variations of SO₂ showed an uprising trend of pollution in the city of Zenica with a strong connection to activation/deactivation of the domestic heating period.

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Introduction:-

As one of the most polluted cities in Bosnia and Herzegovina due to mining and metallurgic activities, great number of boilers, small house stoves and traffic, Zenica has very bad air quality. The worst situation is in winter for two reasons: because of the beginning of the heating season and because of the temperature inversion which is common in the cold part of the year. This atmospheric condition favors the accumulation of pollutants in the valleys, in a phenomenon known as air mass stagnation [1, 2]. We can see how bad is occurrence of inversion layer in deep valley through air quality records data from December 2007 when a period with temperature inversion lasted for about 7 days, and the daily average concentration of SO₂ reach the value of 900 µg/m³ [3]. In these conditions, height of a chimney is of great importance. Study [4] made in 1987 showed that main sources of air pollution with sulphur dioxide, in the conditions of low dispersion (the winter period – with frequent inverse layers of atmosphere), due to their high chimneys, have no major impact on air quality in Zenica City [4]. Study [5] made after this “great discovery” named: Preliminary study of impact of pollution sources with low height chimneys in meteorological conditions characterised by high concentrations of sulphur dioxide in Zenica, made in 1989, gave just qualitative analysis such as: the ground level pollution in specific conditions named “trapped plumes” is a result of pollution emitting from low height chimneys. There was no quantitative analysis whatsoever. Two master thesis [6,7] made in 2013 and 2016, that used mathematical models (SelamaGis and AERMOD) to describe air pollution dispersion in Zenica valley gave almost the same results. According to these studies, major industrial sources of SO₂ have low influence on ambient air quality in Zenica city. Clearly, it is very difficult to determine the contribution of different pollution emission sources on ground level pollution, in specific meteorological conditions with inverse layer of atmosphere such as those in Zenica, using mathematical models. Perhaps, this is one of the reasons for not having quantitative data about impact of single sources on air quality in the situation of low dispersion.

Sulphur dioxide can affect both health and the environment. Short-term exposures to SO₂ can harm the human respiratory system and obstruct breathing. Children and elderly are particularly sensitive to effects of SO₂. Sulphur

Corresponding Author:- Halim Prcanovic.

Address:- University of Zenica, Metallurgical Institute “Kemal Kapetanović”, Zenica, Bosnia and Herzegovina.

dioxide emissions also lead to the formation of other sulfur oxides (SO_x), which can react with other compounds in the atmosphere to form small particles. These particles contribute to particulate matter (PM) pollution causing additional health problems. At high concentrations, gaseous SO_x can harm trees and plants by damaging foliage and decreasing growth. SO₂ can contribute to acid rain that can harm sensitive ecosystems [8].

The main source of sulfur dioxide in the air in Zenica City is metallurgic activity of the steel works Zenica that processes materials containing sulfur, and burn coal containing high amounts of sulfur (approximately 3,6%). In addition to the city heating plant (which is part of Steel Works Zenica), Zenica has a great number of boilers, small house stoves and traffic which are all important sources of sulfur dioxide.

Sulfur dioxide is also present in motor vehicle emissions. In the past, this emissions were an important, but not the primary source of sulfur dioxide in air. However, this is no longer the case, since the content of sulfur in the fuel has decreased significantly.

Emission of sulfur dioxide in Zenica from Steel Works is around 12.874 tons annually [9]. This estimate was made using material balance calculation. These emissions include emission of sulfur dioxide from the City heating plant, because the heating plant is part of Steel Works. Emissions from boilers and small house stoves are yet unknown.

The aim of this paper is systematic analysis of the period specific variability of sulfur dioxide, in Zenica City. The initial comparison with the recent ambient assessment thresholds set up by the domestic regulations for SO₂, was also performed. All data used in this study are the outcome of measurements of Metallurgical institute “Kemal Kapetanović” Zenica.

Methodology:-

The measurement campaign was conducted from January 1 2006 to December 31 2016 in three places “TETOVO”, “INSTITUT” and “CRKVICE” (Figure 1). Measurement site “TETOVO” is located near the Steel Works (Figure 1 a), 350 m from basic oxygen furnace (BOF). The second location “INSTITUT” is measurement site located in urban part of the city. This measurement site was chosen because it is located north of the steel works and detects pollution that the north wind, which blows approximately 11.2% of the time, carries from the Steel Works to the city. Therefore, when it comes to pollution from the Steel Works, this measuring site is always considered as a representative for the city [10]. The third measurement site “CRKVICE” is also an urban measurement site, located in east part of the city. Around of this measurement site there are numerous residential houses and a large boiler from the Cantonal hospital Zenica.

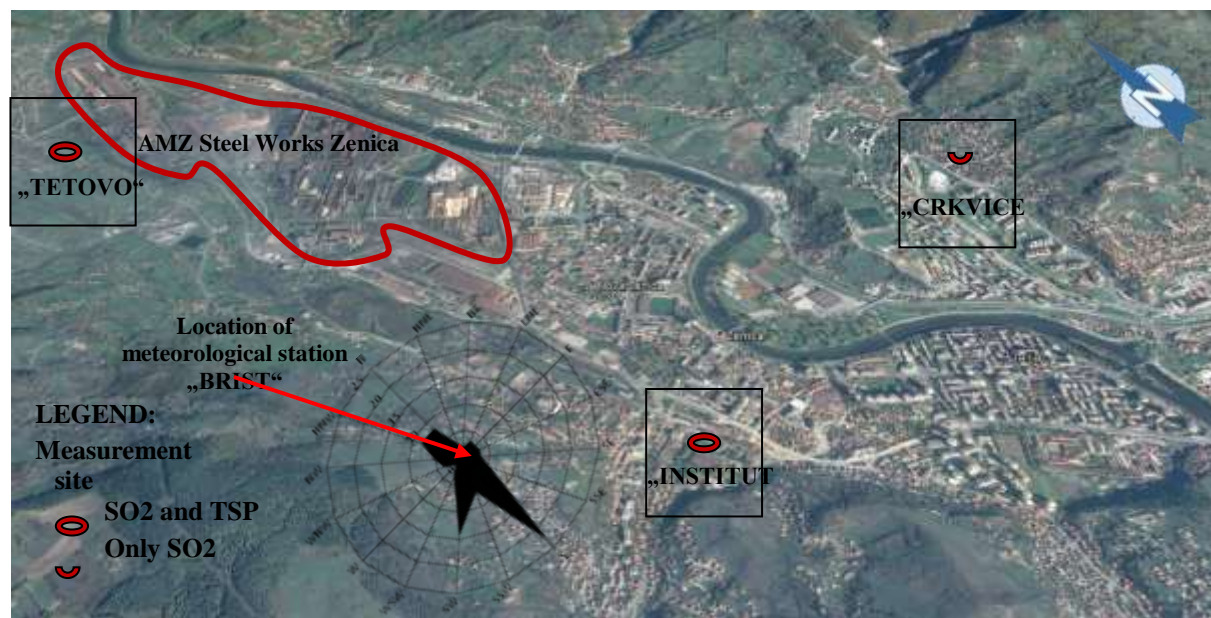


Figure 1:- Study area and spatial distribution of measurement sties

Results and discussion:-

Year average values of SO₂ for all measurement sites are shown in Figure 2. The highest year average of SO₂ in the examined period was found in “TETOVO” with total concentration of 175 µg/m³ and with a maximum value of 668 µg/m³. The maximum concentration of SO₂ in examination period was found at the measurement site “INSTITUT” in 2007 with a daily average value of 903 µg/m³.

Data clearly indicate that concentration of SO₂ is the highest on measurement site “TETOVO”, which is normal concerning the position of the measurement site. The uprising trend of SO₂ concentration at the all three of the measurement sites is evident. When concentration of SO₂ rises, it rises on all three sites. It is very hard to find part of the examined period in which the concentration of SO₂ on one site is stagnating or opposite to the concentrations on the other two sites. Analyzing the measurement data it is obvious that air becomes saturated with SO₂ soon after the formation of the inverse layer of atmosphere, which is frequent in the cold part of the year. This means that the capacity of Zenica valley to receive air pollution has been exhausted a long time ago, and authorities should consider whether to allow construction of any additional source of SO₂ in Zenica valley.

Since the production in Steel Works Zenica has not been changed in the last five years, this uprising trend of SO₂ concentration should be examined closely. The highest daily average concentrations of SO₂ were in the period 2007-2008, before the re-start of integral production in Steel Works Zenica. The origin of the SO₂ and constant rise of SO₂ concentrations should be examined closely.

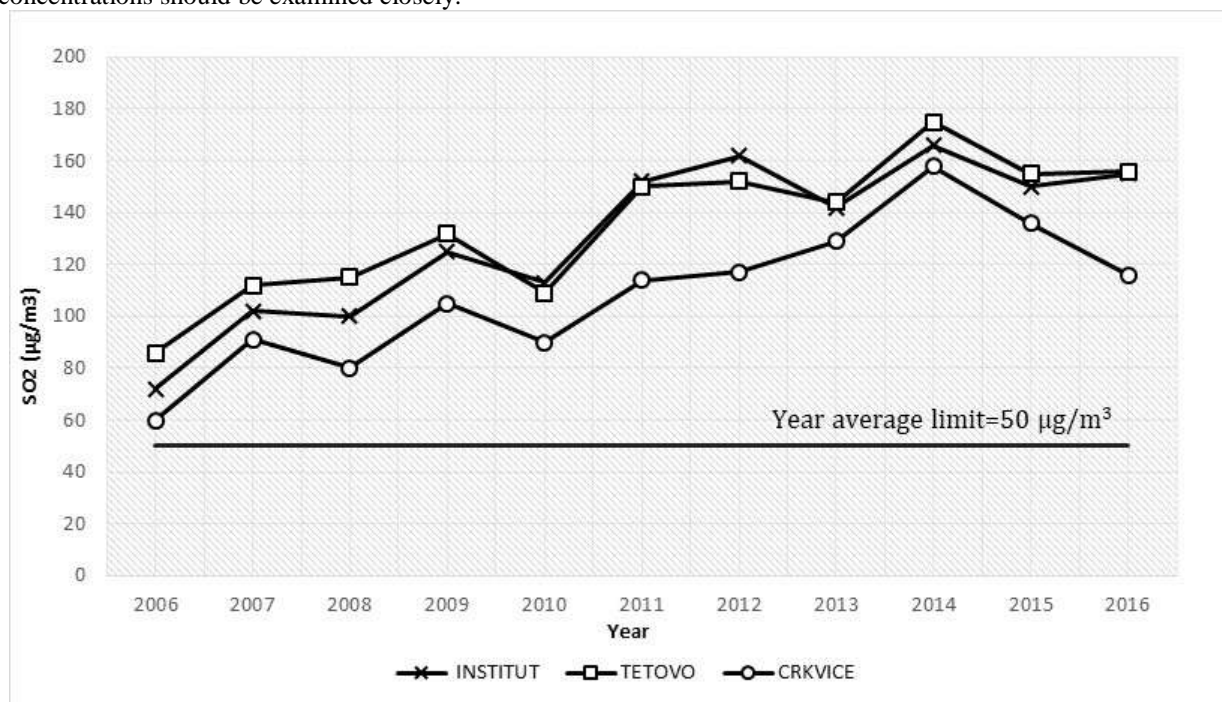


Figure 2:- Temporal variations of SO₂ year average in the examined period

Limit values proscribed by domestic legislation for SO₂ (year average 50 µg/m³ and daily average value of 125 µg/m³ SO₂, which should not be exceeded more than 3 days in a year, were violated in the entire examined period. The number of days of exceeding the daily average value of 125 µg/m³ in the observed period ranges from 66 days in 2006 to 226 days in 2014.

3.1 Meteorological conditions:-

Based on measurement data from meteorological station Zenica, frequencies and average wind speed for single directions for the period 2006-2016 are calculated. Data is obtained from Federal hydrometeorological institute BH. Wind rose and location of meteorological station is presented in Figure 1. Frequencies and average wind speed are given in Table 1. As it can be seen from Figure 1 and Table 2 prevailing wind was south wind with average speed of 1,7 m/s. North wind which carries pollution from Steel Works to the city had frequency of 8% with average wind speed of 2,1 m/s. Since the prevailing wind direction is south one measurement site should be placed in the north

part of the city to take measure pollution coming from industrial sources. Measurement sites position should be revised as well. All the sites are positioned at the base of the valley. At least one measurement site should be at a height above industrial emission sources height.

Table 1:- Frequencies and average wind speed for particular direction observed on meteorological station Zenica for the period 2006-2016.

Direction	N	NN E	N E	EN E	E	ES E	S E	SS E	S	SS W	S W	WS W	W	WN W	N W	NN W	C
Frequency (%)	8,0	2,7	1,9	3,1	2,8	3,1	3,1	5,2	22,1	6,8	13,9	3,7	1,6	3,6	5,1	8,3	5,0
Avg. speed m/s	2,1	1,1	1,3	0,8	0,9	0,9	1,1	1,1	1,7	1,3	1,2	1,0	1,0	1,2	1,5	1,9	0,0

The cold part of the year was characterized by a frequent formation of inversion layers. Figure 3 shows an average vertical temperature profile over the Zenica valley recorded in period 11-15 December 1980[4]. Unfortunately, this is the only recorded vertical profile of temperature over the Zenica valley. As shown in Figure 3a the inversion layer of atmosphere extends from 500 m to about 1000 m above sea level throughout day. First 250 m of the inversion layer is characterized by very unstable wind speed and direction [4]. This situation makes the modeling of dispersion very difficult. It is interesting to notice the temperature inversion in the first 20 meters above the ground throughout the day. This phenomenon is probably caused by the proximity of the water surface of the Bosna River, which causes a lower temperature of the air closest to the ground [4]. Ground level inversion increases local impact of pollution sources with low height chimneys and traffic. In these situations smoke from chimneys, after leaving the chimney goes towards the ground and pollutes the immediate environment.

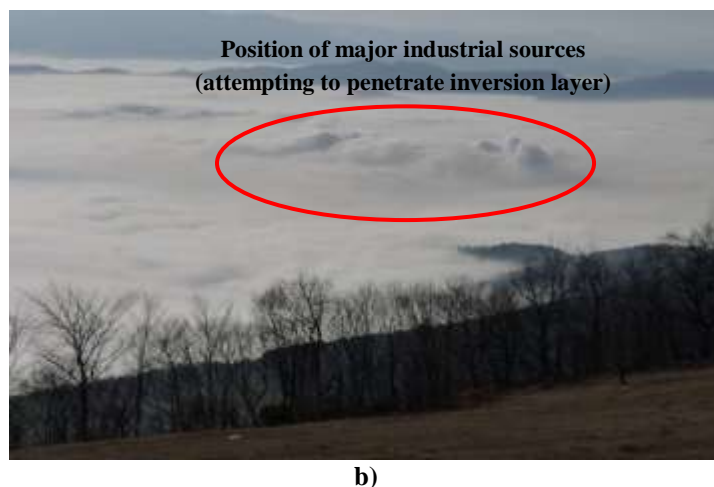
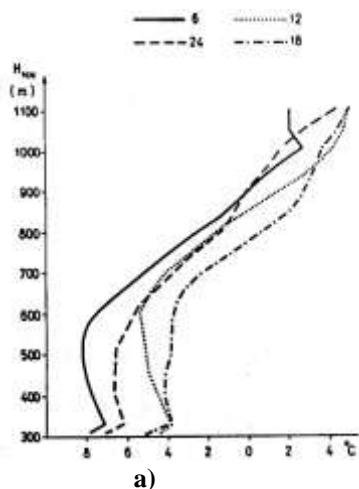


Figure 3:- Average vertical temperature profile (a) and fog formation in conditions with inversion layer (b)

Inversion layers are usually accompanied by fog since moisture in the air is trapped and builds up near the ground. Figure 3b shows a typical situation with an inversion layer and fog formation in Zenica. From the Figure 3 it can be seen how flue gases from mayor industrial emission sources are trying to penetrate the inversion layer.

Pollution in valleys is usually influenced by mountain and valley breeze that occur one after another in a daily cycle. Mountain breeze occurs in the evening when warm air rises in the middle of the valley and cold air is blowing down the hill. This wind is bringing the pollution in the valley and due to the inversion layer, which obstruct upward movement of the air in the middle of the valley, concentrations of pollutants rise gradually. When inversion is present emissions from industrial sources cannot disperse vertically. When it reaches the base of inversion layer the plume will begin to mix downward and contact with hillside at certain altitudes below the inversion layer base, as depicted in Figure 4b. Besides that, on the hill slopes there are a number of small house stoves. All that pollution is brought up to the valley with the mountain breeze. On the other hand, valley breezes blow during the day, blowing up the sides of the hill. Due to this wind and mixing of the air during the day, because of solar radiation

there is a decrease of pollutant concentrations. As it can be seen from Figure 4a typical daily diagram of SO₂ concentrations in Zenica has two distinctive parts. The first part, with small concentrations (during the day time period), and the second part with high concentrations (during the night time period). This situation can be connected with the valley and mountain breezes. As it can be seen from Figure 4a, the concentration of SO₂ starts to rise at 3 o'clock in the afternoon (sunset time for Zenica on 28 December 2015. at 16²²). The concentration of SO₂ reaches its maximum at ten o'clock in the evening.

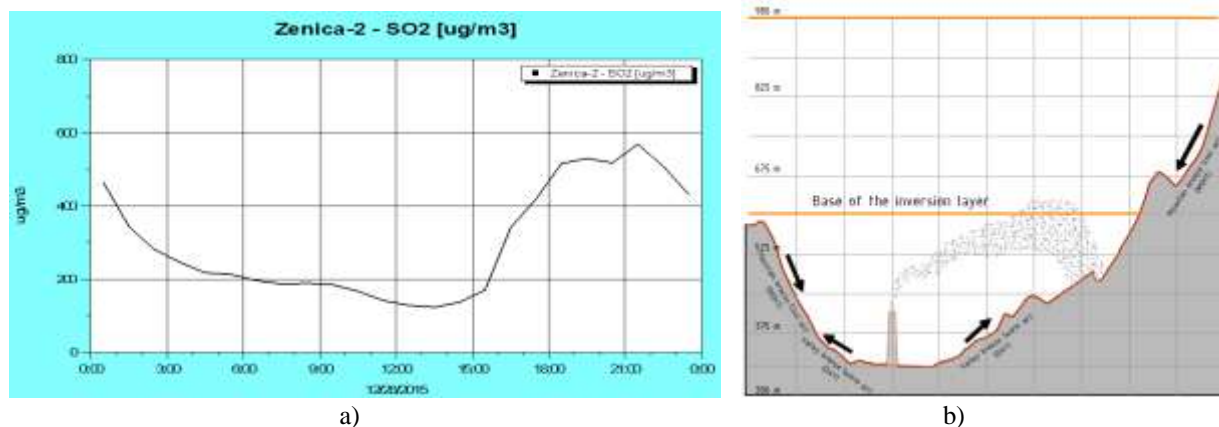


Figure 4:- Typical daily diagram of SO₂ concentrations in Zenica recorded in automatic measurement station Zenica-2 located near measurement site TETOVO on December the 28th 2015.(a), and (b) Zenica valley elevation profile in direction SW-NE, with an industrial stack average height and mountain and valley breeze illustration

3.2 Season specific variations of SO₂

Season specific analysis of SO₂ variations was conducted in order to investigate the impact of season specific emission sources on SO₂ concentrations in ambient air, like space heating (domestic and district heating which uses coal, containing 3.6 of sulfur, as fuel). The cold period is ranging between October the 15 of the current year and April 15 of the following year, to take into account the same winter conditions. This is the period when heating (district heating and domestic-small house stoves) becomes an effective source of SO₂. The warm period is ranging between April the 15 and October 15 of the current year. Descriptive statistics of measurement results are presented in Table 2. The hypothesis that concentrations are higher in cold part of the year is tested using the non-parametric Mann-Whitney U test. Significance at 95% level is also shown in Table 1. Data clearly show that concentration of SO₂ is higher during the cold part of the year (1,00 confidence level) at all three measurement sites for the entire examined period. It can also be seen that concentrations in cold part of the year are two to three times higher than those in warm part of the year. The number of violations of permissible daily average value of 125 µg/m³ was significantly higher in the cold part of the year. This could lead to a wrong assumption that domestic heating is the main source of SO₂ in the cold part of the year, and that an enormous amount of SO₂ (almost 13.000 tons per year) emitted from industrial sources have no significant impact on SO₂ concentration in ambient air in Zenica city. If domestic heating is the main source, why would measurement site "CRKVICE" have the smallest concentrations throughout the entire examination period? This measurement site is located in the east part of the city and with a prevailing south wind it is normal that this measurement site would have the smallest SO₂ concentrations during the examined period.

Furthermore, the height of the industrial stacks is such that the plume can't penetrate the inversion layer, and once the plume reaches the base of the inversion layer it will begin to mix downwards. In a deep valley like this one in Zenica, it will hit the lee side of the hill at a certain altitude below inversion layer base. Pollution doesn't go out from a valley and SO₂ concentration is rising day by day. The longer this situation lasts the higher the concentration will be (December 2007 SO₂ daily average concentration 903 µg/m³). In the December 2016 and January 2017 there were periods of 2 to 4 days without district heating. District heating is a considerable source of SO₂ in Zenica consuming 500 tons of coal per day. Even without district heating, in the condition with inversion layer, SO₂ concentrations were rising.

Table 2:- Period specific average of SO₂ (µg/m³) at all measurement sites

PERIOD*	“INSTITU”			“TETOVO”			“CRKVICE”		
	Mean ± SD		*Sig.	Mean ± SD		*Sig.	Mean ± SD		*Sig.
	Cold	Warm	-	Cold	Warm	-	Cold	Warm	-
2006-2007	128,57±72,59	26,66±8,58	0,0 0	97,73±55,56	52,23±20,77	0,0 0	150,76±76,67	51,34±16,13	0,0 0
2007-2008	211,0±170,5	43,21±16,02	0,0 0	39,86±109,3	54,48±23,05	0,0 0	214,0±144,8	61,09±22,78	0,0 0
2008-2009	140,77±87,65	77,74±19,58	0,0 0	107,60±58,32	92,26±28,82	0,0 0	141,95±65,70	102,19±34,89	0,0 0
2009-2010	166,92±89,65	64,14±31,74	0,0 0	115,81±50,22	67,32±28,28	0,0 0	154,83±74,36	69,51±27,52	0,0 0
2010-2011	192,55±114,37	77,54±29,63	0,0 0	117,68±57,60	82,15±39,68	0,0 0	166,69±92,51	100,07±41,13	0,0 0
2011-2012	256,65±127,15	93,68±50,85	0,0 0	140,67±84,81	109,51±62,39	0,0 0	221,46±110,31	116,69±72,09	0,0 0
2012-2013	168,59±93,39	88,76±43,64	0,0 0	116,75±51,70	119,37±80,40	0,0 0	238,75±63,03	122,68±68,87	0,0 0
2013-2014	215,15±116,68	124,74±62,07	0,0 0	169,91±89,26	144,53±81,99	0,0 0	200,09±90,51	153,14±89,39	0,0 0
2014-2015	214,62±114,04	85,63±63,34	0,0 0	151,99±85,37	123,42±110,93	0,0 0	180,49±98,46	129,27±111,88	0,0 0
2015-2016	194,67±107,86	76,83±80,70	0,0 0	137,63±107,78	93,46±96,55	0,0 0	170,54±105,97	107,18±110,75	0,0 0

* Significance level tested by the non-parametric Mann-Witney U-test.

** Cold part is considered from October 15 one year to April 15 of next year because of the heating season, and to take into account the same winter conditions.

Conclusion:-

This study was conducted to know the temporal and spatial variation of SO₂ during an eleven years period in the city of Zenica. The highest concentration of SO₂ was found in measurement site TETOVO. An incline trend of the SO₂ concentrations is recorded in all three sites. All limit values prescribed by the domestic legislations for SO₂ are violated multiple times. Season specific analysis of SO₂ variations showed that the concentration of SO₂ is higher during the cold part of the year (1,00 confidence level) at all three measurement sites during the entire examined period. Revision of the measurement sites should be made concerning the height of industrial sources of emission and the prevailing wind direction. Domestic heating is a considerable source of SO₂ emission but not the main source. Almost 13.000 tons SO₂ per year originate from industrial sources. The capacity of Zenica valley to receive air pollution has been exhausted a long time ago, and the air becomes saturated with SO₂ soon after the formation of the inverse layer of atmosphere, which is frequent in cold part of the year. Authorities should consider whether to allow any additional source of SO₂ to be constructed in Zenica city. Any attempt to apply existing mathematical models for dispersion of pollutants in Zenica city gave inconclusive results about the degree of impact of certain emission sources on ambient air quality. Existing models should be adapted to the specific meteorological conditions in Zenica city, complex terrain configuration and variety of pollutants. To gain best results a combination of existing models will probably be needed.

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