Multi-criteria Analysis of Air Pollution with SO_2 and PM_{10} in Urban Area Around the Copper Smelter in Bor, Serbia

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Abstract This work presents the results of 4 years long monitoring of concentrations of SO_2 gas and PM_{10} in the urban area around the copper smelter in Bor. The contents of heavy metals Pb, Cd, Cu, Ni, and As in PM_{10} were determined and obtained values were compared to the limit values provided in EU Directives. Manifold excess concentrations of all the components in the atmosphere of the urban area of the townsite Bor were registered. Through application of a multi-criteria analysis by using PROMETHEE/GAIA method, the zones were ranked according to the level of pollution.

Keywords Heavy metals · SO_2 gas · PM_{10} · Pollution · Distribution · PROMETHEE/GAIA

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1 Introduction

At the beginning of the third millennium special attention is paid to the air quality in the urban zones in Europe, due to the increasing industrialization (Nikolaou 2003; Gotschi et al. 2005). The problem of air pollution in industrial zones is much bigger and particularly in the zones with developed production of nonferrous metals (Periera et al. 2007; Shanchez de la Campa et al. 2008). Special interest is directed towards SO_2 (Pires et al. 2008; Periera et al. 2007) and PM_{10} and $PM_{2,5}$ with the contents of heavy metals Cu, Zn, Pb, Ni, Cd, As... (Kozlov 2005; Parceval et al. 2006), with a special view on the contents of arsenic (Shanchez de la Campa et al. 2008; Daniel Sanchez-Rodas et al. 2007).

Sulfur dioxide (SO_2) is a traditional ambient air polluter so the monitoring of its concentration is of special interests for characterization of air quality (Periera et al. 2007), which is particularly emphasized by the World Health Organization (WHO 2000). SO_2 is one of the most important polluters of the environment and mostly originates from the oxidation of sulfur compounds. Anthropogenic emission of SO_2 is resulting from burning the fossil fuels (coal and heavy oils) or smelting of sulfidic ore concentrates (most frequently Cu, Pb, and Zn ores). In the last 20 years a lot of efforts have been made in view of reduction of emission of SO_2 into the air in industrially developed western countries (Nunnari et

al. 2004). Removal of SO_2 from the atmosphere is performed through acid precipitation. SO_2 is an irritating gas which causes breathing problems when people are exposed to high concentrations of it. Absorption of SO_2 in the nose due to its solubility in water burns mucous membrane and attacks upper breathing airways (WHO 2000). Although sulfur is useful for plants in small concentrations, pollution of the atmosphere with SO_2 gas due to its higher concentrations negatively affects plants and the size of the impact depends on its concentration. Due to a certain negative effect of SO_2 in the atmosphere European Union limits its mass contents: (1) limit per hour for protection of human health 350 μg m^{-3}, not to be exceeded more than 24 times per calendar year; (2) daily limit for protection of human health 125 μg m^{-3}, not to be exceeded more than three times per calendar year; and (3) annual limit for protection of ecosystems 20 μg m^{-3} (EC Directive 1999).

Suspended particles of PM_{10} and $PM_{2,5}$ are one of the most important ambient air polluters which harmfully affect human health (Koelemeijer et al. 2006). Prolonged exposure to PM_{10} and $PM_{2,5}$ particles often cause respiratory and cardiovascular diseases and increase mortality (Kappos et al. 2004). For the purpose of protection of human health the EU has introduced two limitations for PM_{10} and $PM_{2,5}$ which should be implemented in two periods: the first one at the beginning of 2005 and the second one in 2010. The limit values for 2005 and 2010 are as follows: (1) daily limit of 50 μg m^{-3} not to be exceeded more than 35 times per calendar year and (2) annual limit of 40 μg m^{-3}. The limits which will be implemented after the year 2010 are: (1) daily limit of 50 μg m^{-3} not to be exceeded more than seven times per calendar year and (2) annual limit of 20 μg m^{-3} (EC Directive 1999).

Arsenic is present in the earth's crust in the concentrations of 4.8±0.5 μg g^{-1} (Rudnick and Gao 2003). The sources of arsenic in the industrial area are natural and anthropogenic (Roy and Saha 2002) and it can be found in rocks, water, and atmospheric dust (Mandal and Suzuki 2002). One of the biggest anthropogenic sources of arsenic in PM_{10} are copper smelter plants which are considered the main environmental pollutants all over the world: Chile, USA, Sweden, Spain, Russia, Australia, and Serbia (Gidhagen et al. 2002; Hedberg et al. 2005; Mandal

and Suzuki 2002; Kozlov 2005; Martley et al. 2004; Shanchez de la Campa et al. 2008; Dimitrijević et al. 2008).

Arsenic is a toxic element and as such it is hazardous for human health considering that it shows carcinogenic qualities (Roy and Saha 2002). It has been established that arsenic attacks many human organs and weakens the immune system (Duker et al. 2005). The higher concentration of arsenic in the air in urban areas is always of anthropogenic origin which is usually the emission from technological plants. In 2001 the World Health Organization published the second edition of Air Quality Guidelines for Europe (WHO 2000) in which it was explained that value of arsenic in the air above 1.5×10^{-3} μg m^{-3} presents high risk for human life. Typical contents of arsenic in European regions are in the range from 0.2 to 1.5 ng m^{-3} in rural areas; 0.5 to 3 ng m^{-3} in urban areas and lower than 50 ng m^{-3} in industrial zones (Shanchez de la Campa et al. 2008). Also, DG Environment of the European Commission has developed a directive for air quality and determined the limit value for arsenic in PM_{10} of 6 ng m^{-3}—an average value on annual level (European Commission 1999, 2000, 2004).

Some toxicological studies indicate that toxicity of arsenic depends on its chemical form, oxidation state, physical state, (gaseous or liquid solution), chemical nature, rate of absorption in the cells, rate of elimination from the body etc. (Viraraghavan et al. 1992). Arsenic is present in various oxidation states: As(0) or in the form of ions, As(V) arsenate, As(III) arsenite, and As(III) arsine. There is a generally prevailing opinion that non-organic arsenates are more poisonous than organic ones and non-organic As(III) compounds are more poisonous than non-organic As(V) (Duker et al. 2005). Due to high arsenic toxicity, the EU air quality EU, 2004/170/CE defines the total threshold contents of arsenic regardless of its form (Oliviera et al. 2005). Arsenic is considered one of the most toxic elements for human health. Continual exposure to high concentrations of arsenic causes acute toxic effect which is easy to diagnose. However, low doses of arsenic do not cause acute toxic effect but they can cause cancer after a prolonged exposure (Roy and Saha 2002).

Other heavy metals such as Pb, Cd, Ni, and Hg are also harmful for human health. Although they do not show acute effect in exposure to contamination they

could get accumulated in the human body for up to 30 years and this way increasing mortality. This makes these elements extremely harmful for human health. The EU directives prescribe limiting value concentrations in PM_{10} at an average annual level of: Cd—5 ng m^{-3}; Ni—20 ng m^{-3}; Pb—5 µg m^{-3}; Hg—1 µg m^{-3} (1999/30/CE; 2004/107/CE).

One of the biggest copper smelters in Europe, from the aspect of quantity of environment pollution gasses emitted, has been operating in the town of Bor (Serbia) for more than 100 years. Since 2003 in the urban part of the townsite of Bor there has been continuous measurement of $S0_2$ emissions in the air in real time as well as measurement of contents of heavy metals in PM_{10} (Cu, Pb, Cd, As, Hg, Ni, and Mn). Obtained results show that concentrations of SO_2 in the air and As in PM_{10} as anthropogenic materials are above the prescribed values (Dimitrijevic et al. 2008). Also the increased contents of Pb and Cd were registered with sporadic registering of the contents of Ni and Hg while the content of Mn was not registered (Milosevic 2005; 2006; 2007; 2008; Dimitrijevic et al. 2008). The aim of this work was to reveal, through an analysis of the results in the period 2005–2008, the threats for human life from the environmental pollutants and thus to trigger better understanding of the effects of manifold excess concentrations of SO_2, As, Cd, and Pb in PM_{10} in urban area as well as future consequences of the EU air quality directive application.

The technology for copper production in this smelter plant is outdated (classic pyrometallurgy with melting in furnaces and utilization of SO_2 gas in production of H_2SO_4 with relatively small degree of utilization <50%) which leads to environmental pollution from higher concentrations of SO_2 and particles of floating dust PM_{10} as well as aero sediments $PM > PM_{10}$. The ore melted in this smelter plant is of chalcopyrite–pyrite type with increased contents of arsenic which is found in the form of FeAsS and Cu_3AsS_4. Through oxidation roasting and melting of such mineral forms leads to arise of the heavy metals oxides and SO_2 gas which in certain quantities contaminate the environment. When emitted from the smelter plant, the reach of SO_2 gas is up to 15 km and the pollution from particles is 2 to 3 km (Magaeva et al. 2000; Moldovanska et al. 2000; Zhukovsky 2000; Kishimoto et al. 2008). Concentrations of SO_2 gas and heavy metals in PM_{10} are much higher than limit value concentrations prescribed through EU Directives (Dimitrijevic et al. 2008; EU

Directives 1999/30/CE and 2004/107/CE). The main reason for such situation is a missed chance for introduction of new technology at the moment when life cycle of present technology required it (Živković & Živković, 2007). The real-time monitoring system for air pollution monitoring in the urban part of the town of Bor was installed in 2003 enabling continuous measurement of the contents of SO_2 in gasses and cumulative measuring of contents of heavy metals in floating particles at four measuring points. Also, there operates a mobile station which enables measurement of the contents of PM_{10} and aerial sediments at 15 locations.

The results obtained from this monitoring system serve as information for state bodies, local administration, and company management and so far they have been scarcely published in scientific literature (Dimitrijevic et al. 2008). The authorities do not pay enough attention to pollution of the environment from various polluters which is a consequence of company's operations, on account of which the operations of the smelter plant pose a risk for the region which justifiably leads to raising an ethical dilemma whether to produce at any cost (Halis et al. 2007).

One should also emphasize the fact (this is the data of RTB Bor Company) that as a consequence of operations of this company around 200,000 tons of SO_2 are emitted into the atmosphere every year which is around 3.5 tons per inhabitant. Per every ton of refined raw materials around 2.5 kg of dust are emitted into the atmosphere which leads to the situation that every year 5.3–19.6 kg of As, 4.86–7.99 kg of Zn, and 6.27–25.11 kg of Pb per inhabitant are emitted into the atmosphere which is many times higher compared to other industrial zones in Europe (LEAP 2003). These facts show that the word is about the most polluted region in Europe which, apart from harming human health in the region itself, poses a particular danger for wider area of southeastern Europe. Regardless of the size of this region the attitude of the company's management towards pollution must be based on global approach towards resolving this problem (Parnell 2006; Yorgun 2007).

This work, apart from the analysis of the state of pollution of the area around this smelter plant also aims at animating potential stakeholders in activities for prevention of further pollution of the environment from such operations of the copper smelter in Bor as well as at preventing a permanent soil degradation in the area of river basin of the Danube where more than 200,000 people live.

2 Study Area

The town of Bor is situated in the eastern part of Serbia at the junction of three countries: Serbian, Romanian and Bulgarian, at a distance of 100 km from Romania and 30 km from Bulgaria (Fig. 1). The Border with Romania is the river Danube while at the immediate vicinity of this area river Timok flows. In the direction towards Romania—Northeast there is a national park Djerdap. West and northwest there is an artificial dam called "Borsko jezero" (Lake Bor) and a mountain range Homoljske Mountains with preserved nature which, together with the national park Djerdap, represent significant tourist resources of the region (Fig. 1).

The source of air pollution with SO_2 gas, heavy metals in PM_{10} and aero sediments is the copper smelter plant within the RTB Bor Company (Mining and Copper Smelter Complex) which has been in operation for more than 100 years and by its capacity represents one of the biggest smelters in Europe. Location of the smelter is immediately beside the urban settlement of the town of Bor where more than 40,000 people live, while in the rural part in the immediate surroundings there are more than 20,000 inhabitants. In the wider area as shown in the Fig. 1, there are around 200,000 inhabitants whose health is imperiled by the emission of SO_2 gas and heavy metals of anthropogenic origin. The technology used in this smelter plant is outdated resulting with the utilization of sulfur lower than 50% while remaining contaminates the environment. This location could be represented as one of the riskiest areas in Europe (Dimitrijevic et al. 2008). Years long contamination of the soil with heavy metals of anthropogenic origin created a danger that those heavy metals may enter the food chains of animals and people which can lead to disastrous consequences (LEAP 2003). Previous

Fig. 1 The area of eastern Serbia with the location of copper smelter in Bor as the main air polluter
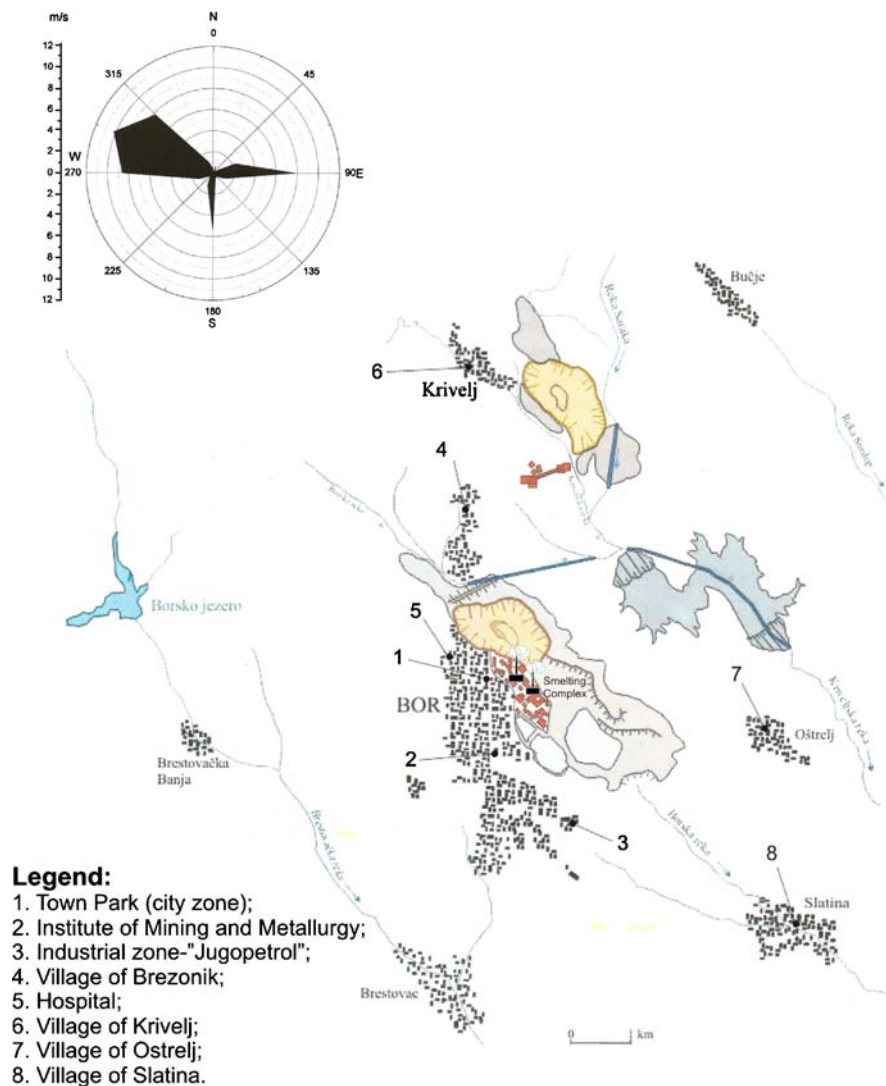
research (Dimitrijevic et al. 2008) unmistakably shows that this area is the most polluted area in southeastern Europe which forces the management of the company to take action aimed at global resolution of the problem.

The direction and the strength of wind in the period from 2005 to 2008 were mostly towards west–northwest and partially towards east and south which can be seen from the wind rose shown in Fig. 2. These facts show that the zone of pollution was directed by the wind rose to the location of the old urban center and measuring points Hospital and Town Park which increases contamination of the urban part of the town. During the year there are incidents of manifold pollution of the urban settlement by, above all, SO_2 gas and in those periods the smelter plant is

stopped. However, short-term contamination and attack on human health repeatedly happen during the year just in the area of the urban part of the old town center. In copper smelter plant in Bor there are two factory smokestacks, the height of one being 120 m ($D=3$ m) for smelter plant off-gasses with the contents 1–3% SO_2 and the other of 150 m ($D=3.5$ m) for gasses when the factory of sulfuric acid is not in operation (gasses resulting from roasting procedure in fluo-solid reactor mixed with converter gasses) with the contents of SO_2 in the gas of 5–6%. On the average the factory of sulfuric acid is out of operation for about 6 months in a year. Both smokestacks are situated in the immediate vicinity of the urban settlement at a distance lesser than 500 m from

Fig. 2 Locations of the places of origin of SO_2 gas and PM_{10} in the area of the townsite of Bor with its surroundings

Legend:
1. Town Park (city zone);
2. Institute of Mining and Metallurgy;
3. Industrial zone-"Jugopetrol";
4. Village of Brezonik;
5. Hospital;
6. Village of Krivelj;
7. Village of Ostrelj;
8. Village of Slatina.

the old urban center where numerous vital functions of the town are. Measuring stations for measurement of the contents of SO_2 and the contents of heavy metals in PM_{10} are installed in the urban part of the town as well as in the bordering part of rural area towards the urban area where the source of pollution of the environment is (Fig. 2).

3 Methodology

3.1 Sampling

In the Fig. 2 there are locations of measuring stations for taking samples of SO_2 gas and PM_{10} in the urban part of the town: measuring point 1 (Town Park); measuring point 2 (Institute of Mining and Metallurgy); measuring point 3 ("Jugopetrol"); measuring point 4 (Village of Brezonik); measuring point 5 (Hospital); measuring point 6 (Village of Krivelj); measuring point 7 (Village of Oštrelj), and measuring point 8 (Village of Slatina). At measuring points 1 and 3 there are fixed measuring stations from which, in real time, every 15 min a sample is taken automatically and the contents of SO_2 in the air is determined. In other places the samples of the PM_{10} are automatically taken cumulatively during 24 h, with mobile station periodically, depending on meteorological situation. The contents of heavy metals in PM_{10} are determined in the laboratory of the Institute for Mining and Metallurgy in Bor.

3.2 Chemical Analysis

At two measuring points (1 and 3), automatic measuring stations for real-time determination of the SO_2 contents in the air have been installed. Transfer of data from measuring stations is performed every 15 min to the control center in the Institute for Mining and Metallurgy in Bor. The content of SO_2 in the air is determined by UV-fluorescence after extraction to higher energy level and light emission measurement. This method enables automatic determination of the contents of SO_2 gas in ambient air in the range of concentrations from 0 to 10,000 μg m^{-3} according to the standard ISO 10498.

At measuring points 2 and 4 measurements were performed through classical acidimetric method. Hydrogen peroxide was used as an absorption solution while titration was performed by means of

sodium hydroxide. The results are comparable because parallel measurements had been performed at the beginning of work of thus defined combined monitoring (Milosevic et al. 2004).

4 Results and Discussion

4.1 Data Processing Methodology

For ranking of zones according to the level of ambient air pollution caused by SO_2 gas and PM_{10} by the quantity and contents of heavy metals in the studied area of the urban part of the town of Bor and its surroundings (Fig. 2), we decided to apply multi-criteria decision-making (MCDM) method (Rousis et al. 2008). Many authors use MCDM in analysis of the air and soil pollution problem (Lim et al. 2005, 2006; Al-Rashdan et al. 1999; Khalil et al. 2004). In this work the PROMETHEE method was used for ranking of locations at which sampling of air and PM_{10} was performed in accordance with determined contents of masses and contents of heavy metals in PM_{10} while geometrical analysis for interactive assistance (GAIA) plane as an option provides graphic interpretation of PROMETHEE method, namely it gives a clear picture of the decision-making problem in the way that it monitors PROMETHEE ranking (Visual Decision Inc 2007). The GAIA visual modeling method is providing the decision-maker with information about the conflicting character of the criteria and the impact of the weights of the criteria on the final results. The GAIA plane is defined by vectors resulting from covariance matrix obtained using principal components analysis (PCA). Using the PCA, it is possible to define a plane on which as few information as possible gets lost by projection (Brans and Mareschal 1994).

The reason for application of PROMETHEE/ GAIA method for processing of obtained results lies in certain advantages of this method compared to other MCDM methods, which are reflected in the way of problem structuring, in the amount of data which is possible to process, in the possibilities of quantifying the quality values, in good software support and in presentation of obtained results (Macharis et al. 2004; Visual Decision Inc 2004).

PROMETHEE represents an outranking method, for final set of alternatives (Vego et al. 2008). In the use of this method it is necessary to define a

corresponding function of preference and assign weight significance (weight coefficient) to each criterion. The preference function defines how a certain option is ranked in relation to another one and translates the deviation between two compared alternatives into a single parameter related to the preference level. The preference level represents an increasing function of deviation where, if the deviation is small, it relates to a weak preference while if the opposite is the case, i.e., if the deviation is large then it represents a strong preference of referent alternative. The PROMETHEE method uses six forms of preference function (Usual, U-shape, V-shape; Level, Linear, and Gaussian), whereby each form depends on two thresholds (Q and P). Indifference threshold (Q) represents the largest deviation considered irrelevant by the decision-maker while preference threshold (P) represents the smallest deviation which decision-maker considers decisive whereby P cannot be smaller than Q. Gauss' threshold (s) represents the average value of P and Q thresholds (Brans 1982; Brans et al. 1984; Brans and Vincke 1985; Herngren et al. 2006).

The PROMETHEE method is based on the determination of positive flow (Φ^+) and negative flow (Φ^-) for each alternative in relation to outranking relations and in accordance with obtained weight coefficient for each criterion attribute. Positive preference flow expresses how much a certain alternative dominates other alternatives, namely if the value is higher ($\Phi^+ \to 1$) the alternative is more significant. Negative preference flow expresses how much a certain alternative is preferred by other alternatives. The alternative is more significant if the value of outgoing flow is lower ($\Phi^- \to 0$). Complete ranking (PROMETHEE II) is based on the calculation of net flow (Φ), which represents the difference between the positive and the negative preference flow. The alternative with the highest value of net flow is ranked best etc. (Brans and Mareschal 1994; Albadvi et al. 2007; Anand and Kodali 2008).

4.2 Results of the Analysis

Average annual contents of SO_2 in urban ambient air in the town of Bor at measuring points 1, 2, 3, and 4 for the 2005–2008 periods are shown in the Table 1.

Obtained results of SO_2 gas (average values on annual level) in the air show high contents above

prescribed limits: for the protection of human health EC Directive 1999/30/CE prescribes 125 μg m^{-3} as daily limit not to be exceeded more than three times per calendar year and 20 μg/m^3 annually for protection of ecosystems (for winter period first October to 31st March). In the copper smelter in Bor gasses from melting containing an average of 1–3% of SO_2 are emitted into the atmosphere through a 120-m high smokestack while gasses from roasting and converter operation are used for production of H_2SO_4 with an average content of 5–6% of SO_2 are also emitted into the atmosphere through a 150-m high smokestack when the H_2SO_4 production factory does not operate. Both smokestacks are situated in the immediate vicinity of the old urban center where some vital functions of the town function (urban green market, a hospital, local self-government bodies, hotel, town hospital, technical faculty, one elementary school, one kindergarten...) at a distance lesser than 500 m. Every year an average of 200,000 t of SO_2 or 3.3 t per inhabitant, depending on the quality of the smelting furnace charge (LEAP 2003), is emitted into the atmosphere through the smokestacks. Modern copper smelter plants in the world, for example Harjavalta copper smelter (Finland) emitted 3,300 t of SO_2 in 2006 on account of annual production of anode copper of 160,000 t which is four times higher than the quantity of produced anode copper in Bor (Dimitrijevic et al. 2008). It should particularly be emphasized that during the analyzed period 2005–2008 concentration of SO_2 in the air reached an average 58–238 μg m^{-3} on annual level whereby concentrations at measuring points "Town Park" and "Jugopetrol" were extremely high which corresponds to the wind rose (Fig. 2). In addition to high concentrations of SO_2 gas one should also emphasize the fact that at these measuring points there is a large number of days with the values above the limit, namely 120 to 150 days a year on the average. On the location "Town Park" a large number of people are exposed to the effects of ambient air pollution particularly during the day as well as on the section from the industrial zone towards measuring station "Jugopetrol" where two villages are located: Ostrelj and Slatina. The values presented in Table 1 marked as Max. value represent average monthly values of the month when this value was the highest in a given year and these are: 1,567, 359, 2,002, and 351 for the year 2005; 2,441, 589, 1,288, and 469 for the year

Table 1 Average annual contents of SO_2 and PM_{10} particles in urban ambient air in the town of Bor for the 2005–2008 periods

Location		SO_2 [µg m^{-3}]				PM_{10} [µg m^{-3}]			
		2005	2006	2007	2008	2005	2006	2007	2008
Location 1	Min. value	<25	10	<25	<25	–	–	–	3
	Max. value	1,567	2,441	344	355	–	–	–	44
	Average value	169	238	175	105	–	–	–	16
	Days above limit	119	132	109	69	–	–	–	16
Location 2	Min. value	<25	<25	<25	<25	4	4	4	5
	Max. value	359	589	347	208	40	48	48	78
	Average value	66	86	82	61	7	8	8	17
	Days above limit	21	25	20	22	0	0	0	17
Location 3	Min. value	4	0	0	<25	–	–	–	5
	Max. value	2,002	1,288	957	561	–	–	–	20
	Average value	215	199	189	170	–	–	–	11
	Days above limit	155	144	150	126	–	–	–	2
Location 4	Min. value	<25	<25	<25	–	4	3	3	–
	Max. value	351	469	697	–	28	23	23	–
	Average value	58	104	91	–	7	5	5	–
	Days above limit	8	23	29	–	0	0	0	–

– no measurements

2006; 344, 347, 957, and 697 for the year 2007; and 335, 208, and 561 for the year 2008. These values were much larger then prescribed limit value of 125 µg m^{-3} for monthly averages. The contents of SO_2 in the air in incidental situations reach the values of 5,000–8,000 µg m^{-3} (Dimitrijevic et al. 2008) when the human health is seriously threatened because acute toxication takes place. The values of SO_2 contents in the air shown in the Table 1 are the highest values registered in relation to the reported values in 21 European cities (Nikolaou 2003; Gotschi et al. 2005), so this area can justifiably be considered as the most SO_2 gas-polluted area in Europe.

Along with gasses from the smelter plant, the PM_{10} particles, containing heavy metals and posing a grave danger for human health due to the fact that they are inhaled into the respiratory tract, are also emitted into the air. The average contents of PM_{10} in the air on annual level in the period of 2005–2008 were measured sporadically at four measuring points and the obtained values (average values on annual level) are shown in the Table 2.

Obtained values of concentrations of PM_{10} in the air are within the limits prescribed by EU Directives (1999/30/CE—50 µg m^{-3} averagely on annual level and maximally not to be exceeded more than 35 times

per calendar year or on annual level 40 µg m^{-3}). However, there are some phenomena of exceeding the limit for 15–20 days particularly in the year 2008 which points to the tendency of the increase of PM_{10} contents in the year 2008 in which maximal values were on average annual level of 44 and 78 µg m^{-3} at measuring points Town Park and the Institute. The reason for such increase of PM_{10} contents is deterioration of the filters resulting with larger quantity of PM_{10} that goes into the air along with the smoke gasses. The EU requirements (1999/30/CE) as of first January, 2010 are more stringent than the existing ones and they prescribe daily limit of the contents of PM_{10} up to 50 µg m^{-3}, not to be exceeded more than seven times per calendar year and 20 µg m^{-3} of PM_{10} on an annual average. The concentration of PM_{10} shows a trend of increase in the year 2008 compared to the year 2007 so that even current lower requirements are not met.

PM_{10} particles were analyzed on the contents of the following heavy metals: Cu, Pb, Cd, As, Ni, Hg, and Mn at eight measuring points the disposition of which in the urban part of the town and the suburban areas is schematically shown in the Fig. 2. The content of Mn was not registered in a single sample while the contents of mercury was registered in only a

Table 2 Contents of heavy metals in PM_{10} in the urban area of the townsite of Bor and its surroundings in the period 2005–2008

Location		Pb [µg m⁻³]				Cd [ng m⁻³]				Cu [µg m⁻³]				Ni [ng m⁻³]				As [µg m⁻³]			
		2005	2006	2007	2008	2005	2006	2007	2008	2005	2006	2007	2008	2005	2006	2007	2008	2005	2006	2007	2008
Location 1	Xmin[a]	0[d]	0.0[e]	0.0	0.1	0	0	0	1	0	0.1	0	0.8	0	0	0	0.025	0	0	1.6	3.3
	Xmax[b]	0.2	0.9	0.6	0.55	6	14	7	10	0.2	0.6	2.4	1.8	0.2	0	0.2	0.100	149	170	98.8	24
	Xavg.[c]	0.02	0.2	0.2	0.15	2	3	2	2	0.1	0.3	0.8	1.334	0.02	0	0.1	0.040	29.3	38.9	25.5	18
Location 2	Xmin	0	0.0	0	0.1	0	0	0	1	0	0	0.3	0.8	0	0	0	0.04	1.9	0	4.8	3.8
	Xmax	0.1	0.4	0.1	0.54	1	15	7	25	0.6	0.6	1.4	3.1	0	0	0.2	0.10	36.1	75.7	51.1	33
	Xavg.	0.01	0.01	0.01	0.18	1	3	2	7	0.2	0.2	0.5	1.6	0	0	0.1	0.04	12	15.5	21	15
Location 3	Xmin	0	0.0	0.1	0.05	0	0	0	2	0	0	0.4	0.7	0	0	0	0.02	0	0	7.8	26
	Xmax	0.1	0.5	0.3	1.5	8	18	5	33	0.4	1.2	2.7	2.6	27	0	0.5	0.10	94.2	148	71.3	179
	Xavg.	0.04	0.2	0.2	0.43	3	5	3	9	0.1	0.4	1.1	1.6	3	0	0.1	0.07	30.7	41.3	31.8	71
Location 4	Xmin	0.0	0	0	–	0	0	0	–	0	0	0	–	0	0	0	–	4.1	0	3.8	–
	Xmax	0.1	0.1	0.1	–	3	6	2	–	0.1	0.4	0.9	–	0	0	0.1	–	48.7	13	22.8	–
	Xavg.	0.1	0.1	0.1	–	1	2	1	–	0.1	0.1	0.4	–	0	0	0.1	–	19.5	4.7	10.3	–
Location 5	Xmin	0	0.0	0	–	0	0	1	–	0	0.1	0	–	0	0	0	–	0	5	0	–
	Xmax	0.1	0.5	0.2	–	8	12	6	–	0.3	0.7	0.8	–	0	0	0	–	84.7	193	41.7	–
	Xavg.	0.1	0.2	0.1	–	3	6	3	–	0.2	0.4	0.4	–	0	0	0	–	30.2	59.3	19.2	–
Location 6	Xmin	0	0	0.2	–	1	1	5	–	0	0.1	0.2	–	0	0	0	–	8	7	82.3	–
	Xmax	0	0	0.2	–	1	1	5	–	0	0.1	0.2	–	0	0	0	–	8	7	82.3	–
	Xavg.	0	0	0.2	–	1	1	5	–	0	0.1	0.2	–	0	0	0	–	8	7	82.3	–
Location 7	Xmin	0	0	0	–	0	0	0	–	0	0.1	0	–	0	0	0	–	0	9.9	0	–
	Xmax	0.1	0.1	0.1	–	0	6	3	–	0.2	0.4	0.8	–	0	0.9	0	–	9.3	43.8	14.8	–
	Xavg.	0.05	0.1	0.05	–	0	2	1	–	0.1	0.2	0.4	–	0	0.2	0	–	4.6	20.6	6.5	–
Location 8	Xmin	0	0.1	0.1	–	0	1	2	–	0	0	0.3	–	0	0	0	–	20	0	37.9	–
	Xmax	0.1	0.4	0.1	–	2	12	2	–	0	0.4	0.3	–	0	0	0	–	20.9	10.6	37.9	–
	Xavg.	0.05	0.2	0.1	–	1	6	2	–	0	0.2	0.3	–	0	0	0	–	20.4	5.3	37.9	–
Limit values according to the EU directive		5 µg m⁻³				5 ng m⁻³				–[f]				20 ng m⁻³				6 ng m⁻³			

– no measurements

[a] The minimal average value on month level

[b] The maximal average value on month level

[c] The average value on annual level

[d] Not registered concentration

[e] The concentration registered in trails

[f] Limit value of copper in the PM_{10} is not prescribed by the EU directives (1999/30/CE; 2004/107/CE)

few samples on account of which it will not be specially analyzed. The results obtained by the analysis of the composition of PM_{10} for the period of 2005–2008 were shown in the Table 2.

Obtained values of the contents of heavy metals in PM_{10} revealed extremely increased content of As due to its presence in the incoming raw material. The content of As was in all cases above the limit value of 6 ng m^{-3} and calculated on annual level is three to ten times as high at urban locations: town park, hospital, and "Jugopetrol". The values calculated on monthly average at times used to be 30 times above the limit (Milosevic 2005; 2006; 2007; 2008). The content of arsenic in the air measured in 21 European cities was the highest in Verona and it was 24.7 ng m^{-3} during winter (Gotschi et al. 2005). At the same time in Bor in December, the value of 193 ng m^{-3} was determined (Dimitrijevic et al. 2008). Bor occupies the first place in Europe according to the contents of arsenic emitted into the ambient air per inhabitant with 5.3–19.6 kg of As which has happened for the last few years with the tendency of increase (LEAP 2003). It should be emphasized that World Health Organization (WHO 2000) advises that the threshold of 1.5 ng m^{-3} is a risk limit for human health. Any additional comment on the status of air in the urban area of the town of Bor would be superfluous!

The elements Cd, Ni, Pb, and Hg are also toxic elements some of which (Pb and Cd) accumulate in the human body for the period of up to 30 years. EU Directives also prescribe their limit values in the air such as: Cd—5 ng m^{-3}, Ni—20 ng m^{-3}, Pb—5 μg m^{-3}, and Hg—1 μg m^{-3} in the form of PM_{10}. In PM_{10} (Table 3), Hg values were not registered except in a few cases, Ni only in a few cases while Pb and Cd were registered at almost all measuring points.

The maximal contents of cadmium in PM_{10} during the year 2006 at measuring points 5 and 8 was twice as high in relation to the limit value and at measuring point 3 the obtained maximal values were three times as high. During the year 2007 maximal contents of Cd in PM_{10} above limit values were measured at measuring points 1, 2, and 5. However, in the first part of the year 2008 higher contents of cadmium were registered at locations 2 and 3 with the values going even five to six times above the limit values when further measuring of the contents of Cd in PM_{10} was stopped!

The content of lead in all these cases was beneath limit values but it represents a potential danger due to

its propensity to be accumulated in the human body for a longer period of time. The contents of heavy metals in other areas surrounding the industrial zones are Tamilandu (India): Cu–0.2/0.7 μg m^{-3}, Ni—0.09/ 0.12 μg m^{-3}, Pb—0.1–0.5 μg m^{-3}; Cd was not registered (Vijayanand et al. 2008); industrial region of Korea: Pb—0.1/0.4 μg m^{-3}, Cd 0.01/0.03 ng m^{-3} (Nam and Lee 2006); and Spain: Cu 80/120 ng m^{-3}, Cd 0.8/0.9 ng m^{-3}, Pb 34/47 ng m^{-3}, As 7/9 ng m^{-3} (Daniel Sanchez-Rodas et al. 2007).

It is evident that as a result of the of copper smelter operations there are much higher contents of SO_2, As and Cd in the air in the studied region of the urban part of the town of Bor and its surroundings than prescribed by the EU directives.

Also, the increase in the content of PM_{10} contained in Cu, As, Pb, and Cd with the tendency of increase is evident which will inevitably lead to the increase in the content of already high contents of As and Cd. These facts clearly indicate (besides SO_2, As, and Cd which are evident) potential dangers of heavy metals intoxication in the air: Cu, Pb as well as Ni and Hg with the tendency of increase.

It is evident that the attention of the authorities to the problem of air pollution in 2008 and at the beginning of 2009 is reduced because the number of sampling and the number of components monitored in the air were reduced compared to 2007! With increase of the smelter plant capacity which is being announced, the concentration of SO_2, PM_{10}, and the contents of heavy metals will be increased which will significantly worsen air quality in the urban zone of the town of Bor and the region as a whole.

On the basis of available data, a multi-criteria analysis has been performed through the use of PROMETHEE/GAIA method for zone ranking in the urban part of the town of Bor with its surroundings according to the level of pollution: with the SO_2 gas, PM_{10}, and the contents of heavy metals: Cu, As, Pb, Cd, and Ni in PM_{10}. On the basis of available data obtained by measuring at all eight measuring points— locations (period 2005–2008), two scenarios for ranking of polluted zones have been developed:

- Scenario 1: ranking on the basis of concentrations of SO_2 gas, PM_{10}, and the contents of heavy metals Cu, As, Pb, Cd, and Ni in PM_{10} for locations: Town Park (1); the Institute (2); "Jugopetrol" (3), and Village of Brezonik (4).

Table 3 Weight coefficient setting on the basis of harmfulness of present metals

Criteria	Weights		Influence on human health
	Scenario 1	Scenario 2	
SO_2—sulfur dioxide	15	–	Harmful effect on respiratory organs
Number of days above SO_2 limit	10	–	
PM_{10}—Particulate matter	20	–	Heavy metals enter the body
Number of days above PM_{10} limit	5	–	
Pb—lead	15	30	II class, remains in the body and is carcinogenic
Cd—cadmium	15	30	I class, remains in the body and is carcinogenic
Cu—copper	5	10	Harmful in the body but the body gets rid of it
Ni—nickel	5	10	II class, carcinogenic substance
As—arsenic	10	20	II class biological halftime 3–5 days, carcinogenic
Σ	100	100	

- Scenario 2: ranking on the basis of contents of heavy metals in PM_{10}, Cu, Pb, Cd, As, and Ni for locations: Town Park (1); the Institute (2); "Jugopetrol" (3); and Village of Brezonik (4); Hospital (5); Village of Krivelj (6); Village of Ostrelj (7); and Village of Slatina (8).

For the needs of a model creation presented in this work, the required parameters for PROMETHEE/ GAIA method were assigned to each criterion. These values include the impact of the criteria, namely presence of harmful metals at certain measuring locations with tendency of their minimizing, so the model implies ranking of the best alternatives— locations with the least presence of harmful materials in the air in accordance with assigned set of preference functions and weights to each criterion (Table 3). Linear preference function was chosen as preference function for criteria which define the contents, the concentration of harmful metals with adopted thresholds of indifference and preference (Q and P) in the zones of 5% and 30%, respectively. For the remaining two criteria which define the average number of days with pollution above the prescribed limit, V-shape preference function with preference threshold (P) of 25% was assigned.

PROMETHEE performed a complete ranking from the best to the worst location from the aspect of presence of harmful metals in the air on those locations. By utilizing Decision Lab 2000 software package, with the PROMETHEE method, based on data in Tables 1, 2, and 3, values are acquired for positive (Φ^+) and negative flows (Φ^-) and thereby net flow (Φ) for both scenarios (Fig. 3).

The ranking results indicate that the best location in Scenario 1 is the measuring point, Village of Brezonik (location 4), while the measuring points in the Village of Ostrelj (location 7 and again location 4) are the best locations in Scenario 2. The most polluted location in both scenarios is Industrial zone "Jugopetrol" (location 3) while we should also mention the locations Hospital (Location 1) and Town Park (location 5) which are situated in the very center of the town.

Another advantage of the software package Decision Lab can be seen in the application of the option GAIA. Considering that the value Δ is satisfactory in both scenarios (Δ>75), we will discuss about the validity of use of this tool in further presentation of the results. Where, Δ presents the measure of the quantity of information being preserved by defined model. In the real world applications the value of Δ has always been larger than 60% and in most cases larger than 80% (Brans and Mareschal 1994).

The GAIA plane presents the projection of the set of n alternatives that can be represented as a cloud of n points in a k-dimensional space. Where n represents the number of alternatives and k is the number of criterions. The basis of the position of criteria in GAIA plane (squares), concord, or conflict between certain criteria can be determined. Also, the positions of alternatives (triangles) determine strength or weakness of the properties of actions in regard to criteria—the closer to the direction of the criterion vectors the better alternative itself according to that criterion. The coordinate axes, presented in Fig. 4, are dimensionless axes which are only used for segmentation of the space for the purpose of better presenting the strengths of the alternatives and criterions accord-
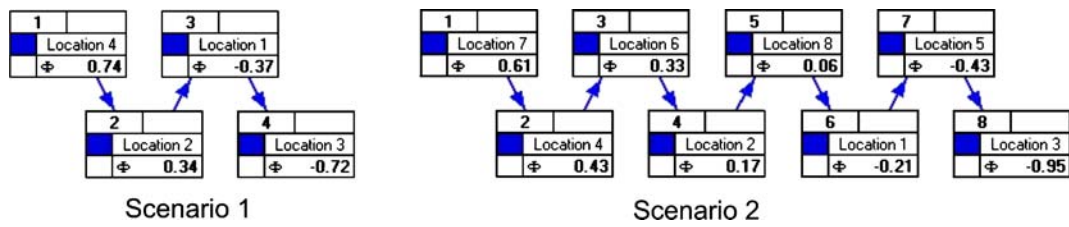
Fig. 3 PROMETHEE II ranking of locations for both scenarios (sampling places were ranked from right to left and from the best to the worst location for each scenario)

ing to their position in the GAIA plane. Especially, it is important to indicate their distance from the coordinate beginning, which only can be done using the coordinate axes. This way, within the set A and D of Fig. 4 (Cluster A and Cluster D), there are locations with the largest percent of harmful metals in the air (location 3, location 1, and location 5) which

evidently are not good according to any criterion and they are also directed in the opposite direction in regard to the decision stick *pi* which defines a compromising solution in accordance to the given weights of the criteria. Unlike them, location groups in Fig. 4 (Cluster B, Cluster C, and Cluster E) are good by a large number of criteria from which the

Fig. 4 GAIA analysis for defined scenarios
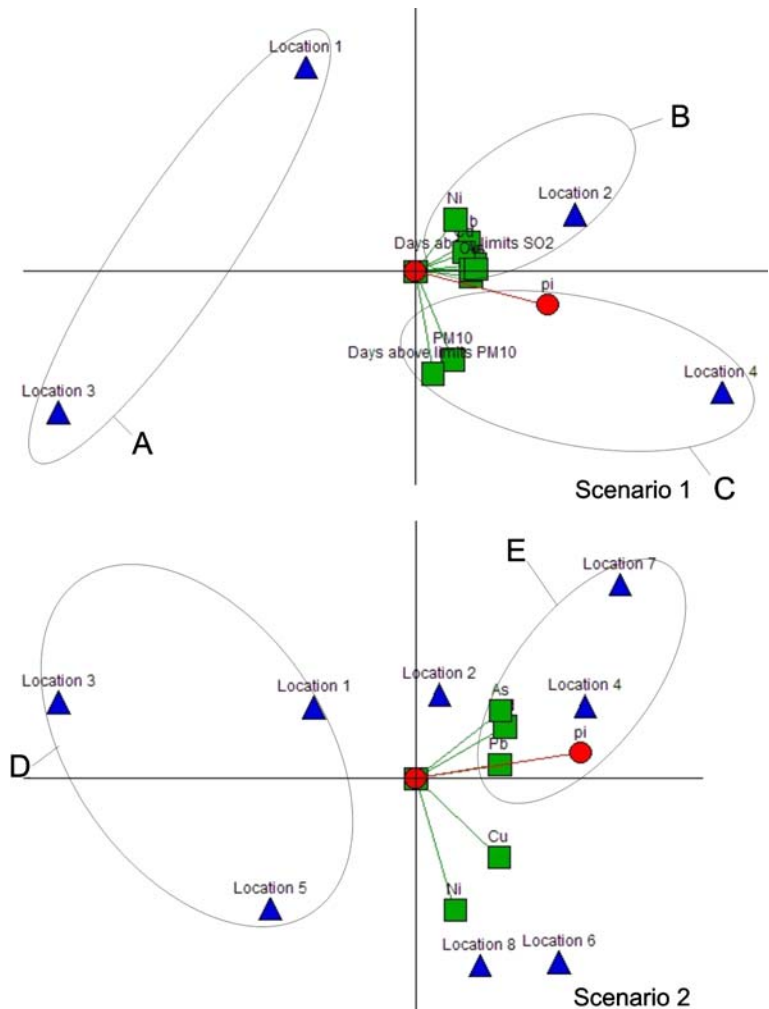
location the Village of Brezonik (location 4) stands out and which is by scenario 1 the closest to the decision stick and with the lowest concentration of PM_{10} particles in the air (cluster C), while the location Institute (location 2) is least exposed to concentration of SO_2 and contents of harmful heavy metals in PM_{10}. Also, according to scenario 2, locations Village of Ostrelj (location 7) and location Village of Brezonik (location 4) are least exposed to the contents of the most dangerous heavy metals Cd, As, and Pb (Cluster E) in PM_{10}. Apart from these, one should also point out the location Village of Krivelj (location 6) which is the best in relation to the contents of Ni and Cu in PM_{10}.

5 Conclusion

Obtained results show that in the studied area of the urban part of the townsite of Bor, situated in the immediate vicinity of one of the largest copper smelters in Europe, environmental pollution resulting from the SO_2 gas, PM_{10} particles, and the contents of As and Cd are several times above the limit values prescribed by EU Directives (1999/30/CE, 2000/105/CE) which seriously endangers human health in this part of Europe. Because of the location of the smelter plant there is also a risk of pollution on a wider scale even in other countries (Romania and Bulgaria).

PROMETHEE/GAIA method was used to rank the zones according to the level of total pollution through two scenarios: scenario 1, locations with simultaneous impact of SO_2, PM_{10}, and contents of heavy metals in PM_{10} and scenario 2, locations with the impact of contents of heavy metals in PM_{10}. Obtained clusters of the total pollution identify locations 1 and 3 in the first scenario and locations 1, 3, and 5 in the second as the most dangerous for human health. Location 1 (Town Park), 3 ("Jugopetrol"—in the vicinity of the new town center), and 5 (Hospital) are just those locations in which the largest number of people in the town of Bor are concentrated and whose health is exposed to the largest impact of harmful components from the atmosphere. Wind rose, in a long period, is directed just towards these locations.

An ethical issue, which the representatives of RTB Bor Company and government officials of Serbia who are in charge of environmental and human health

protection should mind, is the price of further copper production in the Bor smelter with technology which pollutes the environment with such huge quantities of substances harmful for human health!

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