UTICAJ NOVE TEHNOLOGIJE TOPLJENJA BAKRA NA SASTAV UKUPNIH TALOŽNIH MATERIJU U BORU, SRBIJA

THE IMPACT OF A NEW COPPER SMELTING TECHNOLOGY ON THE TOTAL ATMOSPHERIC DEPOSITION IN BOR, SERBIA

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U cilju ispitivanja uticaja nove tehnologije topljenja koncentrata Cu u topionici u Boru na količinu i hemijski sastav (As, Cd, Ni, Pb) ukupnih taložnih materija (UTM) izvršeno je poređenje rezultata merenja u periodu 2014-2015 (stara tehnologija topljenja Cu) sa rezultatima merenja iz perioda 2016-2017 (nova tehnologija topljenja Cu). Analiza je izvršena za tri merna mesta u urbanoj sredini grada Bora (Tehnički fakultet - TF, Gradski park - TP, i Institut za rudarstvo i metalurgiju - IN). Kao posledica promene tehnologije topljenja i boljeg tretmana otpadnih gasova, pre svega SO₂, na svim posmatranim lokacijama došlo je do promene pH vrednosti UTM, od kiselog (5.8 pH) ka baznom (7.8 pH). Takođe, u periodu rada nove topionice, na svim posmatranim lokacijama uočeno je povećanje sadržaja As u UTM kao i smanjenje sadržaja Cd. Sadržaj Ni i Pb u UTM je neznatno promenjen u posmatranom periodu. Na lokacijama TF i TP u periodu rada stare topionice prekoračena je maksimalno dozvoljena koncentracija UTM od 200 mg·m⁻²·dan⁻¹ za godišnji period usrednjavanja. Kao posledica boljeg tretmana otpadnih gasova iz topionice u 2017. godini zabeleženo je samo jedno prekoračenje maksimalno dozvoljene koncentracije UTM od 450 mg·m⁻²·dan⁻¹ u mesečnim uzorcima, i to na lokaciji TF. Jedino na ovoj lokaciji je u 2017. godini detektovano i prekoračenje maksimalno dozvoljene koncentracije UTM za godišnji period usrednjavanja. Pored toga, u periodu rada nove topionice, na svim posmatranim lokacijama uočeno je prisustvo veoma jake ($r > 0.8$) i jake ($0.8 > r > 0.6$) Pearson-ove korelacije između teških metala detektovanih u UTM, za razliku od perioda rada stare topionice kada su ove korelacije bile slabije.

Ključne reči: ukupne taložne materije; teški metali; topionica bakra

The influence of a new smelting technology in the Copper Smelter Bor on the content and chemical composition (As, Cd, Ni, Pb) of the total atmospheric deposition (TAD) in the Bor town urban areas was analyzed in this paper. The results of measurements the TAD in the period 2014-2015 (old smelting technology) were compared with the results for the period 2016-2017 (new smelting technology). The analysis was carried out for three locations in the Bor town (Technical Faculty - TF, Town Park - TP, and Mining and Metallurgy Institute Bor - IN). As a result of changes in the smelting technology and better treatment of waste gases, predominantly SO₂, the pH values of the TAD have changed at all sampling points from acid (5.8 pH) to base (7.8 pH). The increase in the As content as well as the reduction of Cd content in the TAD was also detected at all sampling points while the Ni and Pb contents in the TAD have been slightly changed in the observed period. At the sampling points of the TF and TP, the maximum permissible concentration of the TAD (200 mg·m⁻²·day⁻¹) for the annual averaging period is exceeded, during the old smelter operation. As a result of better treatment of the smelter waste gases in 2017, only one exceeding of the maximum allowed concentration of the TAD (450 mg·m⁻²·day⁻¹) in monthly samples was recorded (at the sampling point TF). Only at the TF, the maximum permissible concentration for the annual averaging period is exceeded in 2017. The presence of a very strong ($r > 0.8$) and strong ($0.8 > r > 0.6$) Pearson correlation between the heavy metals determined in the TAD was observed at all sampling points during the new

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smelter working period, as opposed to the old smelter working period when these correlations were weaker.

Key words: *total atmospheric deposition; heavy metals; copper smelter*

1 Introduction

One of the largest copper mines, as well as the Copper Smelter in Europe, is located in the Municipality of Bor, Serbia. Equipment in the Smelter and Sulfuric Acid Plants has long since become outdated, since it has been in use for almost 50 years. The existing technology, not providing the required utilization of sulfur and not meeting the environmental standards, has a high energy and maintenance costs [1]. Also, the obsolete technology and old equipment does not provide the designed technological copper recovery (95%). In addition to that, it was not possible to achieve more than 60% of sulfur utilization, which is put into the smelting process with the concentrate using the old smelting technology [1].

It is well known that the primary metal and metalloid constituents of the particulate matter (PM) emitted by the Copper Smelter are Cu and Fe oxides and other elements, such as As, Sb, Pb and Zn [2]. Fugitive emissions from the open pits, ore waste dump, flotation tailing dumps, and copper smelter, together with the emissions from the point sources in the Copper Smelter are the main sources of PM and total atmospheric deposition (TAD) in the Municipality of Bor [3]. In recent years, a bulk deposition has been widely studied with the aim to examine the elemental associations in the TAD with the potential sources of heavy metal contaminants using the Positive Matrix Factorization (PMF) model [4, 5].

Contamination with SO₂ and PM enriched with heavy metals, originated from the Copper Smelter in Bor, required the urgent measures to protect the human health and prevent further contamination of the working and living environment. Therefore, the Serbian Government has decided to invest the notable means in building the New Copper Smelter and Sulphuric Acid Plant in Bor. The new smelter facilities were built and started to operate in 2016. By modernization the Smelter, the emissions of SO₂, PM, and other pollutants in the waste smelter gas streams should be drastically reduced and brought under the prescribed limit values.

*Fig. 1. Sampling locations relative to the Copper Smelter Bor
(1. Technical Faculty in Bor -TF, 2. Town Park -TP, 3. Mining and Metallurgy Institute Bor- IN)*

The main goal of this research was to investigate the influence of the new smelting technology, applied in the Copper Smelter in Bor on the quantity and chemical composition (As, Cd, Ni, and Pb) of the total atmospheric deposition (TAD) in the Bor town urban areas. In that aim, the results of measurements in the period 2014-2015 (old smelting technology) were compared with the results from the period 2016-2017 (new smelting technology).

2 Materials and methods

Results presented in this paper are obtained from the sampling campaigns conducted from January 2014 to December 2017 at three locations in the Bor town urban area (Technical Faculty in Bor - TF, Town Park - TP, and Mining and Metallurgy Institute Bor - IN) as shown in Fig. 1. The TAD samples were collected continuously for the periods of 1 month. The total of 144 monthly samples were collected, 48 samples per each sampling point. Sampling campaigns were conducted by the Mining and Metallurgy Institute Bor, Department for Chemical and Technical Testing, in accordance with the monitoring program, adopted by the competent Ministry and local government.

2.1 Sampling locations

The sampling point, named TF, is located downwind of the S-SE prevailing winds from the Copper Smelter. This location is close to the Technical Faculty in Bor, about 1 km N-NW relative to the Copper Smelter. The sampling point, named TP, is located downwind of the E prevailing wind from the Copper Smelter. This location is close to the Town Park in Bor, about 1 km W relative to the Copper Smelter. The sampling point, named IN, is located downwind of N-NE prevailing wind from the Copper Smelter. This location is close to the Mining and Metallurgy Institute Bor, about 2 km S-SW from the Copper Smelter.

2.2 Sampling equipment and procedures

Analysis of the TAD samples has been done following the standard SRPS EN 15841, published by the Institute for Standardization of Serbia, in 2011 [6]. This standard is identical to EN 15841:2009 CEN/TC 264, and specifying the general requirement for sampling equipment and different sampling strategies relative to the site type. The bulk (bottle/funnel) and Bergerhoff samplers are recommended for collecting the TAD in the industrial and urban sites. A study that evaluated the sampler uncertainties of different collectors (wet only, bulk, Bergerhoff and bulk bottle/funnel) showed that at the industrial sites and possibly at very dry rural and urban sites, it is necessary to use the Bergerhoff or bulk bottle+funnel methods for the TAD collecting [7]. Therefore, the Bergerhoff collector was used in the present study. The collecting surfaces of all the collectors used in the present study were made of a high density polyethylene. The height of sampling orifice was about 1.5 m above the surface to avoid collection of the re-suspended dust from the surface.

3 Results and discussion

The total of 144 TAD monthly samples were collected from January 2014 to December 2017 in three sites in the urban area of Bor (sampling sites TF, TP and IN). The average daily and annual concentrations of the TAD, pH, and heavy metals (Pb, Cd, Ni, and As) were calculated and analyzed. The summary statistics of the daily average content of the TAD in Bor, for the annual averaging period, is presented in Table 1. The analysis showed that there are statistically significant differences (level 0.05) between the TAD concentrations at all sampling sites comparing the periods when different technologies were applied in the Smelter. The same stands also for the pH values of the TAD determined at all sampling sites, as well as for the Cd content in the TAD at the TF and As content in the TAD at the IN.

In accordance with data obtained in Table 1, it is obvious that the TAD concentrations at the TF and TP are almost 2 times higher in the period 2014-2015 compared to the period 2016-2017, as shown in Fig.2. The lower TAD concentrations are a direct consequence of changes in the smelting technology and better treatment of waste gases in the Copper Smelter. Also, the Cd concentrations in the TAD are higher in the period 2014-2015 compared to the period 2016-2017. Contrary to that, the As concentrations, determined in the TAD, and pH values of the TAD, determined at all sampling sites in Bor are higher in the period 2016-2017 compared to the period 2014-2015, as shown in Figs. 3 and 4. A higher As content in the TAD in the period 2016-2017 can be attributed to a

higher concentration of As in the copper concentrates, processed in the smelter in the observed period, as well as to a higher copper production during the operation period of the new smelter. The increase in the pH of the TAD in the operation period of the New Smelter is due to the better waste gas treatment from the Smelter (primarily SO₂). The Pb and Ni content in the TAD did not change significantly during the period 2014-2017. This is because that the waste gas emissions from the Smelter do not contribute to an increase in concentration of these elements to a great extent [3].

Table 1. Daily average TAD concentration in Bor ($\text{mg}\cdot\text{m}^{-2}\cdot\text{day}^{-1}$), together with Pb, Cd, Ni and As contents, determined in the TAD ($\mu\text{g}\cdot\text{m}^{-2}\cdot\text{day}^{-1}$) and average pH for the period 2014-2017

Parameter	Unit	Technology used in The Copper Smelter in Bor			
		Old		New	
	Year	2014	2015	2016	2017
TAD-TF	$\text{mg}\cdot\text{m}^{-2}\cdot\text{day}^{-1}$	498.9	366.2	256.4	211.8
TAD-TP	$\text{mg}\cdot\text{m}^{-2}\cdot\text{day}^{-1}$	473.7	312.7	202.4	181.8
TAD-IN	$\text{mg}\cdot\text{m}^{-2}\cdot\text{day}^{-1}$	179.0	110.7	91.7	97.7
Pb-TF	$\mu\text{g}\cdot\text{m}^{-2}\cdot\text{day}^{-1}$	136.9	34.6	69.3	52.4
Pb-TP	$\mu\text{g}\cdot\text{m}^{-2}\cdot\text{day}^{-1}$	113.8	21.8	51.7	48.1
Pb-IN	$\mu\text{g}\cdot\text{m}^{-2}\cdot\text{day}^{-1}$	28.1	11.6	18.7	15.4
Cd-TF	$\mu\text{g}\cdot\text{m}^{-2}\cdot\text{day}^{-1}$	6.1	3.8	2.4	1.3
Cd-TP	$\mu\text{g}\cdot\text{m}^{-2}\cdot\text{day}^{-1}$	4.5	1.7	2.0	0.9
Cd-IN	$\mu\text{g}\cdot\text{m}^{-2}\cdot\text{day}^{-1}$	0.9	0.3	0.6	0.4
Ni-TF	$\mu\text{g}\cdot\text{m}^{-2}\cdot\text{day}^{-1}$	5.2	7.0	7.8	5.1
Ni-TP	$\mu\text{g}\cdot\text{m}^{-2}\cdot\text{day}^{-1}$	7.1	4.5	8.1	8.0
Ni-IN	$\mu\text{g}\cdot\text{m}^{-2}\cdot\text{day}^{-1}$	1.3	1.4	3.1	2.4
As-TF	$\mu\text{g}\cdot\text{m}^{-2}\cdot\text{day}^{-1}$	42.7	4.5	62.5	49.2
As-TP	$\mu\text{g}\cdot\text{m}^{-2}\cdot\text{day}^{-1}$	37.1	3.2	44.2	38.9
As-IN	$\mu\text{g}\cdot\text{m}^{-2}\cdot\text{day}^{-1}$	2.7	0.1	9.1	10.1
pH-TF		4.6	6.8	7.5	8.1
pH-TP		4.9	6.2	7.3	8.0
pH-IN		5.5	6.6	7.3	7.9

The TAD concentrations on 49 sampling sites in the Republic of Serbia are presented in the "Annual Report on the Air Quality in the Republic of Serbia in 2016" [8]. The highest average concentration of the TAD in 2016 for the annual averaging period was recorded in Pirot ($421 \text{ mg}\cdot\text{m}^{-2}\cdot\text{day}^{-1}$), while the lowest concentration was recorded in Pancevo ($65 \text{ mg}\cdot\text{m}^{-2}\cdot\text{day}^{-1}$). If the Table with the TAD concentrations from reference [8] is sorted in a descending order, the TAD concentration at the TP in 2016 ($202 \text{ mg}\cdot\text{m}^{-2}\cdot\text{day}^{-1}$) occupies the 15th, and the TAD concentration at the IN ($92 \text{ mg}\cdot\text{m}^{-2}\cdot\text{day}^{-1}$) occupies the 47th position out of 49 positions. The TAD concentrations at the IN were below the maximum permissible concentration for the annual averaging period throughout the observation period. As a result of better treatment the smelter waste gases in 2017, only one exceeding of the maximum allowed concentration of the TAD ($450 \text{ mg}\cdot\text{m}^{-2}\cdot\text{day}^{-1}$) in monthly samples was recorded at the TF. Only at the TF, the maximum permissible concentration for the annual averaging period is exceeded in 2017. It can be said that these results show that the environment state is improved in terms of the TAD amount in the urban areas in Bor after the new smelting technology application in the Smelter.

In order to compare the annual content of heavy metals (Pb, Cd, Ni and As), determined in the TAD in Bor, to the results obtained for other regions, the results are presented in Table 2 along with the results from some other studies. In accordance with data, obtained in Table 2, it is obvious that the annual atmospheric deposition fluxes of Pb and Cd, determined at all sampling points in Bor, are higher in the period 2014-2015, compared to the period 2016-2017. The annual atmospheric deposition fluxes of Pb recorded in Bor are comparable with those recorded in Belgrade [4], but they are higher compared to the results of other studies (with the exception of the sampling point IN). The annual atmospheric deposition fluxes of Cd, recorded in Bor at the TF and TP, are higher compared to the results of other studies (with the exception of the sampling point IN where Cd fluxes are of the same order of magnitude or lower than in the other studies). The annual atmospheric dep-

osition fluxes of Ni and As, determined at all sampling points in Bor, are higher in the period 2016-2017, compared to the period 2014-2015. The annual atmospheric deposition fluxes of Ni, recorded in Bor, are of the same order of magnitude or lower compared to the results of other studies, but higher compared to the results recorded in Belgrade [4]. The annual atmospheric depositions fluxes of As, recorded in Bor, are higher compared to the results of other studies.

Fig. 2. TAD concentrations in Bor in the period 2014-2017

Fig. 3. Average As content in the TAD in Bor in the period 2014-2017

Fig. 4. Average pH of the TAD in Bor in the period 2014-2017

Table 2. Annual atmospheric deposition fluxes of Pb, Cd, Ni and As ($\text{mg}\cdot\text{m}^{-2}\cdot\text{year}^{-1}$) in different regions

Location	Period	Pb	Cd	Ni	As	References
TF, Bor, Serbia	2014-2015	31.3	1.8	2.2	8.6	This study
TF, Bor, Serbia	2016-2017	22.2	0.7	2.4	20.4	This study
TP, Bor, Serbia	2014-2015	24.7	1.1	2.1	7.4	This study
TP, Bor, Serbia	2016-2017	18.2	0.5	2.9	15.2	This study
IN, Bor, Serbia	2014-2015	7.2	0.2	0.5	0.5	This study
IN, Bor, Serbia	2016-2017	6.2	0.2	1.0	3.5	This study
Belgrade, Serbia	2002-2006	21.7	0.2	11.3		[4]
Tokyo Bay, Japan	2004-2005	9.9	0.4	6.8	2.9	[9]
Seine Estuary, France	2001-2002	18.0	0.4	4.1		[10]
Paris, France	2001-2002	4.2	0.2	0.6		[10]
Pearl River delta, China	2001-2002	13.0	0.1	8.4		[11]
Lake Superior, USA	1993-1994	1.5	0.5	0.8	0.2	[12]
North Sea	1992-1994	3.7		1.2	0.3	[13]

It can be said that the results presented in Table 2 show the improved environment state in the urban areas in Bor after the new smelting technology application in the Smelter, concerning the annual atmospheric deposition fluxes of Pb and Cd. Contrary to that, the annual atmospheric deposition fluxes of As are higher after the new smelting technology application in the Smelter.

Table 3 shows the Pearson coefficients of correlation between the heavy metals at the sampling points in Bor, before and after the new technology application in the Smelter. The presence of the very strong ($r > 0.8$) and strong ($0.8 > r > 0.6$) Pearson correlation between the heavy metals determined in the TAD was observed at all sampling points during the New Smelter operation period, as opposed to the Old Smelter operation period when these correlations were weaker.

Table 3. Pearson correlation matrix for the deposition of Pb, Cd, Ni and As in Bor before and after the application of the new smelting technology in the smelter

Technology used in The Copper Smelter in Bor									
Old					New				
	Pb-TF	Cd-TF	Ni-TF	As-TF		Pb-TF	Cd-TF	Ni-TF	As-TF
Pb-TF	1				Pb-TF	1			
Cd-TF	.791**	1			Cd-TF	.817**	1		
Ni-TF	-.178	.266	1		Ni-TF	.865**	.683**	1	
As-TF	.316	.412*	.170	1	As-TF	.885**	.775**	.707**	1
Old					New				
	Pb-TP	Cd-TP	Ni-TP	As-TP		Pb-TP	Cd-TP	Ni-TP	As-TP
Pb-TP	1				Pb-TP	1			
Cd-TP	.435*	1			Cd-TP	.781**	1		
Ni-TP	-.161	.268	1		Ni-TP	.753**	.761**	1	
As-TP	.703**	.338	.231	1	As-TP	.732**	.824**	.884**	1
Old					New				
	Pb-IN	Cd-IN	Ni-IN	As-IN		Pb-IN	Cd-IN	Ni-IN	As-IN
Pb-IN	1				Pb-IN	1			
Cd-IN	.604**	1			Cd-IN	.924**	1		
Ni-IN	-.007	.290	1		Ni-IN	.863**	.811**	1	
As-IN	.549**	.196	-.106	1	As-IN	.937**	.918**	.909**	1

** Correlation is significant at the 0.01 level (2-tailed).

* Correlation is significant at the 0.05 level (2-tailed).

4 Conclusion

The influence of the new smelting technology, applied in the Copper Smelter in Bor, on the quantity and chemical composition (As, Cd, Ni, and Pb) of the total atmospheric deposition in the Bor town urban areas was analyzed in this paper. In that aim, the results of measurements in the period 2014-2015 (old smelting technology) were compared to the results from the period 2016-2017 (new smelting technology).

It can be said that these results show that the environment state is improved in terms of the TAD amount in the urban areas in Bor after the new smelting technology application in the Smelter. The increase in the pH of the TAD in the period of operation of the New Smelter is due to the better processing of smelter waste gases (primarily SO₂). The increase in As content in the TAD in the operation period of the New Smelter can be attributed to a higher concentration of As in the copper concentrates processed in the Smelter, as well as to a higher production of copper. Decrease in the Cd content in the operation period of the new smelter can be attributed to a lower concentration of Cd in the copper concentrates processed in the Smelter. The content of Pb and Ni in the TAD did not significantly change in the operation period of the new smelter, but they are lower compared to the period when the Old Smelter was in operation. In general, the annual atmospheric deposition fluxes of As, Cd, and Pb, recorded in Bor, are higher compared to the results of other available studies, while the annual atmospheric deposition fluxes of Ni, recorded in Bor, are of the same order of magnitude or lower compared to the results of other studies.

The presence of the very strong ($r > 0.8$) and strong ($0.8 > r > 0.6$) Pearson correlation between the heavy metals, determined in the TAD, was observed at all sampling points during the New Smelter operation period, as opposed to the Old Smelter operation period when these correlations were weaker. This unequivocally indicates that they come from the same source - the Copper Smelter.

From all above mentioned facts, it can be concluded that in the period of operation of the New Smelter, a decrease in concentrations of the TAD and heavy metals in the TAD was observed, with the exception of As. Further research should be continued regarding to the identification the sources of As emissions within the Smelter in order to reduce the As content in the TAD.

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