

# Geochemical investigation of Sasa tailings dam material and its influence on the Lake Kalimanci surficial sediments (Republic of Macedonia) – preliminary study

## Geokemične raziskave jalovinskega materiala rudišča Sasa ter njegov vpliv na sedimente Kameniškega jezera (Republika Makedonija) – preliminarna študija

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### Abstract

This research is aimed at investigating the mineralogical characteristics of the tailings material and heavy metal contents of the tailings material deposited close to the Sasa Pb-Zn Mine in the Osogovo Mountains (eastern Macedonia) and on its possible impact on Lake Kalimanci. The mineral composition of Sasa Mine tailings material is dominated by quartz, pyrite, galena, sphalerite, magnetite and others. Geochemical analysis was performed in a certified commercial laboratory for the following elements: Mo, Cu, Pb, Zn, Ni, As, Cd, Sb, Bi, Ag, Al, Fe, Mn, S. Analysis revealed very high concentrations of toxic metals in the tailing material – with average values [mg kg<sup>-1</sup>]: Mo 2.9, Cu 279, Pb 3975, Zn 5320, Ni 30, As 69, Cd 84, Sb 4.2, Bi 9.4 and Ag 4.1. The multi-element contamination of Sasa Mine tailings material was assigned a pollution index greater of 15, indicating that the tailings material from Sasa Mine contains very high amounts of toxic metals and represents a high environmental risk for surrounding ecosystems. For this reason the influence of discharged tailings dam material into Lake Kalimanci which lies approximately 12 km lower than Sasa Mine, was also established. Calculated pollution index values for Lake Kalimanci sediments vary from 21 to 65 and for Sasa mine surficial tailings dam material from 15 to 60.

### Izvleček

Raziskava se osredotoča na mineraloško sestavo in vsebnost težkih kovin v jalovišču svinčevo-cinkovega rudišča Sasa, ki se nahaja v Osogovskih planinah (vzhodna Makedonija); kot tudi na vplive rudarjenja na bližnje Kameniško jezero. Mineralno združbo jalovinskega materiala rudišča Sasa v večini sestavljajo kremen, pirit, galenit, sfalerit, magnetit in drugi minerali. Povprečne koncentracije izbranih elementov v površinskem jalovinskem materialu so [mg kg<sup>-1</sup>] Mo 2.9, Cu 279, Pb 3975, Zn 5320, Ni 30, As 69, Cd 84, Sb 4.2, Bi 9.4 in Ag 4.1. Za območje jalovišča, kot tudi za 12 km oddaljene površinske sedimente Kameniškega jezera je bil izračunan indeks onesnaženja (pollution index – PI), njegov razpon v površinskem jalovinskem materialu je od 15 do 60 in v površinskih sedimentih Kameniškega jezera od 21 do 65. Visoke koncentracije težkih kovin ter izračunan indeks onesnaženja kažeta na močno onesnaženje obeh območij, kar predstavlja veliko tveganje za bližnje ekosisteme.

### Introduction

Mining is one of the most important global industrial activities, occurring in almost every part of the world. A significant concern facing any mining area is the disposal of wastes produced during the mining, milling and smelting of ore minerals (MOORE & LUOMA, 1990; LAMBORS, 1994), because of the possibility of toxic metals entering the environment. Through ore extraction and

manufacturing at almost every metal mine site large amounts of waste material are being produced. Tailing material from mining activities is stored in special tailings dams (ROGAN ŠMUC et al., 2009), of which at least 3500 currently exist worldwide (DAVIES & MARTIN, 2000), ranging from a few to thousands of hectares in size. Because of their scale and composition, they usually have a negative effect on the surrounding area. Public concern regarding the risk and potential impact

on the surrounding environment caused by tailings dams has been growing since the first known occurrence of such incidents.

Composed of finely-ground rock particles with no particular economic value and mixed with water, tailings dams are supposed to last forever, but since 1970 there have been more than 35 major failures reported around the world (DIEHL, 2001; GÖRANSSON et al. 2001; GRIMALT et al. 1999; UNEP/OCHA, 2000). In 2003 such a major environmental disaster took place at Sasa Mine tailings dam. Tailings material from end of dam No. 3 and beginning of No. 4 collapsed and caused an intensive flow through the Kamenica River valley into Lake Kalimanci, with around 70,000 to 100,000 m<sup>3</sup> of tailing material discharged into the surrounding ecosystem. Tailing material contains a large amount of toxic metals (DOLENEC et al, unpublished) and as such can seriously affect the surrounding environment.

Involving a detailed preliminary search and re-evaluation of known historical research, here we present a study of the mineralogical and geochemical signature of tailings dam material. Both the mineralogical composition and total toxic metal concentration (Ag, As, Cd, Cu, Mo, Pb, Sb, Bi, Zn, Ni, Al, Fe, Mn) of the surficial material from the Sasa tailings dam are defined, with an environmental assessment of the 2003 failure of Sasa tailings dam also provided.

## Materials and methods

### Study area

Sasa Mine is located at an altitude of around 2000 m in the Osogovo Mountains, near the small town of Makedonska Kamenica in eastern Macedonia (Fig. 1). Part of the Serbo-Macedonian Massif, the area is comprised of highly metamorphic rocks containing gneiss, micas, amphibolites and schist. These rocks are paragenetically related to subvolcanic intrusions which are a product of tertiary calc-alkaline magmas and as a result there are also subvolcanic equivalents present which in dykes form favourable conditions for ore deposition. Sasa Mine has established itself as one of the largest ore districts within the Besna Kobilja Osogovo Tassos metallogenetic zone, within which the volcanic rocks are predominantly dacites, quartzlatites and andesites. The Sasa-Toranica zone has experienced hydrothermal alteration processes which have resulted in the creation of new mineral assemblages: quartz, orthoclase, coloured minerals (epidote, chlorite, carbonates) and accessory minerals (apatite, zircon and sphene). The typical mineral assemblage in the Sasa Mine area also includes several ore minerals: pyrite, galena, sphalerite, as well as occasional chalcopyrite and traces of ceruzite, anglesite and malachite (TASEV et al., 2005).

The important Pb and Zn ore bodies are usually found in quartz-muscovite-graphitic schists but also in greenschists and marbles. With Sasa's

ore field occupying an area of about 80 km<sup>2</sup>, Sasa Mine was in production for over 45 years, annually yielding around 90,000 tonnes of Pb-Zn high-quality concentrate (SERAFIMOVSKI et al., 2005). After the first geological surveys of Sasa Mine began in 1954, by 1966 the mine had an annual production capacity of 300,000 tonnes of material using the underground method. From 1979 to 2003 it produced a collective concentrate, with annual production reaching about 625,000 tonnes of material. Total ore production is estimated at 20 Mt, with concentrate grading at 10 % Zn, Pb and 30-35 g/t Ag. The average annual production of approximately 450,000 tonnes continued uninterrupted until the mine closed in 2003.

The tailings material from Sasa Mine is located between the mine and Lake Kalimanci. Deposited near the Kamenica River, it therefore poses a severe threat in terms of metal pollution to the surrounding area, as well as to areas further downstream towards both the river and the lake. The tailings dam at Sasa Mine is divided into four parts, representing an approximate total volume of 15,000,000 m<sup>3</sup>. The first three tailings are abandoned, but tailing No. 4 is still in production, with half of its projected capacity of 2,000,000 m<sup>3</sup> currently met.

### Sampling and sample preparation

Six surficial samples from Sasa tailings dam (No. 3 (H-1, H-2, H-3) and beginning of No. 4 (H-4, H-5, H-6)) were taken in October 2003, with the chosen sampling locations forming a profile along the dam from N to SE (Fig. 2). Surficial tailings dam material is composed chiefly of grey brown grinded Pb-Zn ores with grinded background rocks. Sandy samples were collected in plastic bags and stored in the laboratory at 4 °C, before being dried at 50 °C for 48 hours. Samples were then sieved through a 0.315 mm polyethylene sieve and homogenised in a mechanical agate grinder to a fine powder for further analysis. Seven samples of surface sediments were collected from Lake Kalimanci in September 2008 in transversal profiles (marked with roman numerals on Fig. 2). Measured pH in lake sediments is ranged from 5.5 to 7.5. Sampling was performed during a dry period (no precipitation – summer season) when the lake was empty. Surficial lake samples are composed of sand, silt and organic matter, and were collected with a plastic spade, transferred to pre-cleaned plastic bags, and stored in the laboratory at 4 °C. After collection, the samples were oven dried before dry sieving at a temperature of 50 °C for 48 hours until a constant weight was attained. Samples were sieved through a 0.315 mm polyethylene sieve to remove plant debris homogenised by mechanical agate grinder to a fine powder for subsequent analysis.

### Analysis

Mineralogy of the surficial samples was determined at the Department of Geology, Ljubljana



of the most important minerals in the Sasa ore district, were not found in all samples as was expected. One explanation for this may have been because samples were collected from the upper oxidation section, where reduction minerals are absent. In addition, SERAFIMOVSKI et al. (2006) reported that in the Zletovo ore district, located near the Sasa mine, sphalerite is usually found only in a few generations and is sometimes interstitially replaced by quartz, galena and other minerals.

The Serbo-Macedonian Massif is mostly metamorphic, with typically highly metamorphic rocks such as gneiss, micas, amphibolites and schist. This explains why XRD analysis showed (Table 1) mainly those minerals present in metamorphic rocks (quartz, calcite, mica, cordierite, epidote). Evidence was also found for the presence of clinopyroxenes, which are monoclinic and contain Ca, Na, Al, Fe<sup>3+</sup> or Li. Because of local hydrothermal processes, rocks of the Serbo-Macedonian Massif contain large amounts of lead, zinc, aluminium, iron, copper, manganese and other elements which are bound into different minerals such as pyrite, sphalerite, galena and magnetite.

Table 1. Results of XRD analysis.

Tabela 1. Rezultati rentgenske analize.

Minerals	Samples					
	H-1	H-2	H-3	H-4	H-5	H-6
Quartz – SiO <sub>2</sub>	☉	☉	☉	☉	☉	☉
Calcite – CaCO <sub>3</sub>	☉	☉	☉	☉	☉	
Sphalerite – (Zn, Fe <sup>2+</sup> )S	☉	☉		☉		☉
Chlinochlore – (Mg, Fe <sup>2+</sup> ) <sub>5</sub> Al(Si <sub>3</sub> Al)O <sub>10</sub> (OH) <sub>8</sub>	☉	☉	☉		☉	☉
Mica – KAl <sub>2</sub> /K(Mg, Fe <sup>2+</sup> ) <sub>3</sub> AlSi <sub>3</sub> O <sub>10</sub> (OH,F) <sub>2</sub>	☉	☉	☉	☉	☉	☉
Cordierite – (Mg, Fe) <sub>2</sub> Al <sub>4</sub> Si <sub>5</sub> O <sub>18</sub>	☉	☉	☉	☉	☉	☉
Clinopyroxenes – X <sub>2</sub> Si <sub>2</sub> O <sub>6</sub> (x=Ca, Na, Al, Fe <sup>3+</sup> , Li)		☉			☉	☉
Epidote – Ca <sub>2</sub> (Al,Fe <sup>3+</sup> ) <sub>3</sub> (SiO <sub>4</sub> ) <sub>3</sub> (OH)	☉	☉	☉	☉		☉
Pyrite – FeS <sub>2</sub>			☉			
Magnetite – Fe <sup>3+</sup> Fe <sub>2</sub> <sup>+2</sup> O <sub>4</sub>				☉		
Galena – PbS				☉		
Hematite – Fe <sub>2</sub> O <sub>3</sub>				☉		
Chlorite – (Mg, Fe, Mn, Al) <sub>4-6</sub> (Si, Al) <sub>4</sub> O <sub>10</sub> (OH, O) <sub>8</sub>				☉		

Table 2. Concentrations of different elements and pollution index values in Sasa surficial tailings dam material.

Tabela 2. Vsebnosti izbranih elementov in vrednosti indeksa onesnaženja v površinskem jalovinskem materialu rudišča Sasa.

Samples	Mo mg kg <sup>-1</sup>	Cu mg kg <sup>-1</sup>	Pb mg kg <sup>-1</sup>	Zn mg kg <sup>-1</sup>	Ni mg kg <sup>-1</sup>	As mg kg <sup>-1</sup>	Cd mg kg <sup>-1</sup>	Sb mg kg <sup>-1</sup>	Bi mg kg <sup>-1</sup>	Ag mg kg <sup>-1</sup>	Al %	Fe %	Mn %	S %	PI /
H – 1	2.9	188	3752	6092	28	54	53	2.5	6.7	2.8	4.6	12	1.6	4.4	19.23
H – 2	3.4	164	4573	3709	29	64	32	7.9	5.9	2.9	4.7	112	1.6	4.2	15.83
H – 3	2.7	244	5657	9747	32	49	81	3.6	12	3.7	4.9	11	1.5	4.8	29.13
H – 4	3.0	641	>10000	>10000	35	72	234	5.0	15	8.2	4.6	14	1.8	6.8	59.53
H – 5	2.7	213	4775	5761	29	85	48	3.0	6.0	3.1	4.9	12	1.5	4.2	20.27
H – 6	2.9	226	4995	6512	30	91	58	3.0	10	3.7	4.8	12	1.6	4.7	22.59
Mean	2.9	279	3975	5320	30	69	84	4.2	9.4	4.1	4.8	12	1.6	4.9	27.76
*	10	60	85	200	50	20	1	/	/	/	/	/	/	/	/

\*Permissible level of heavy metal content – Ur. list RS 68/96.

## Geochemistry and statistics of Sasa tailings dam

Results of toxic metal (Ag, As, Cd, Cu, Mo, Pb, Sb, Bi, Zn, Ni, Al, Fe, Mn) and sulphur (S) content analysis are given in Table 2.

If the average concentrations of examined elements from Sasa tailings dam are compared with average concentrations (Ag 16 mg kg<sup>-1</sup>, As 7.1 mg kg<sup>-1</sup>, Cd 56 mg kg<sup>-1</sup>, Cu 2.5 mg kg<sup>-1</sup>, Mn 730 mg kg<sup>-1</sup>, Ni 29 mg kg<sup>-1</sup>, Pb 172 mg kg<sup>-1</sup>, Sb 62 mg kg<sup>-1</sup>, Zn 1.7 mg kg<sup>-1</sup> and Fe 8.7 %) from Barroca Grande tailings dam (ÁVILA et al., 2008), it can be seen that concentrations of almost all toxic metals are much higher in surficial material from Sasa tailings dam. However, in both cases, toxic metals concentrations exceed the permissible levels adopted by the National Environmental Protection Agency of Slovenia (UR. LIST RS 68/96).

To identify multi-element contamination of the investigated material that may increase overall metal toxicity (NISHIDA et al., 1982), the pollution index (PI) of samples was calculated. PI is computed by averaging the ratio of the total concentration of toxic metals to the assumed permissible level of metals (JUNG, 2001), in this study represented by equation (1):

$$PI = (\Sigma(\text{Total conc. of element} / \text{Tolerable level of element})) / \text{Number of elements} \quad (1)$$

Samples	As mg kg <sup>-1</sup>	Mo mg kg <sup>-1</sup>	Zn mg kg <sup>-1</sup>	Ni mg kg <sup>-1</sup>	Pb mg kg <sup>-1</sup>	Cd mg kg <sup>-1</sup>	Cu mg kg <sup>-1</sup>	PI /
<b>I-4</b>	69	2.9	10700	47	9357	74	672	36
<b>II-3</b>	77	4.6	20900	79	16300	136	1162	65
<b>III-3</b>	73	3.7	14000	51	10900	90	723	44
<b>V-7</b>	66	3.7	12600	54	9472	81	596	39
<b>VI-11</b>	59	2.5	11600	53	7557	78	546	34
<b>VII-12</b>	66	2.7	9326	56	5144	54	398	25
<b>VIII-8</b>	62	2.3	7056	49	4863	47	327	21
Mean	67	40705	12312	56	9085	80	632	38
*	20	10	200	50	85	1	60	/

\* Permissible level of heavy metal content – Ur. list RS 68/96.

Table 3.  
Concentrations of the heavy metals and pollution index values in Lake Kalimanci surficial sediments.

Tabela 3.  
Vsebnosti izbranih elementov in vrednosti indeksa onesnaženja površinskih sedimentov Kameniškega jezera.

A PI value of greater than 1, indicates that, on average, toxic metal concentrations are higher than permissible levels (KIM et al., 2001).

Seven toxic metals (Mo, Cu, Pb, Zn, Ni, As and Cd) contained within Sasa tailings dam material were chosen to calculate the PI, the results of which are presented in Table 2. PI was also calculated for the same toxic metals from Lake Kalimanci samples (Table 3).

The value of the pollution index across all samples ranged from 15 to 60. According to KIM et al. (2001), the calculated PI for South Korean tailing samples varies from 3.9 to 58.6, which indicates that all tailings, both in South Korea and Macedonia (Sasa), contain toxic metals at a level high enough to cause toxicity in the local environment and consequently in the biota, including humans.

From the results it is clearly evident that a large amount of toxic metals has remind as residual material after the extraction process and is currently lying abandoned within the tailings dams. The studied toxic metals in Sasa tailings dam represent some of the most important ore-forming elements which are paragenetically related to the Pb-Zn polymetallic mineralisation of Sasa's ore district. The presence of such high levels of toxic metals represents a huge environmental disaster risk. If these metals enter an aquatic environment, they are redistributed through the water column where they accumulate in sediments and/or are consumed by biota (LINNIK & ZUBENKO, 2000; Long et al., 1996; SINGH et al., 2005). ROGAN ŠMUC et al. (2009) reported that because of the failure of the Sasa tailings dam in 2003, extremely high levels of toxic metals caused huge environmental damage to Lake Kalimanci and the Kočani field. However, further study is needed, to reveal more about the mineral composition of the Sasa tailings dam as well as a more in-depth assessment of toxic metal pollution in the surrounding area. Thus this preliminary study also includes an initial assessment of the influence of the discharged toxic metals on Lake Kalimanci.

## Impact on the surrounding environment

### Geochemistry and statistics of Lake Kalimanci sediments

Sediment samples from Lake Kalimanci were analysed in terms of their toxic metal content (As, Mo, Zn, Ni, Pb, Cd and Cu; Table 3). Concentrations of almost all metals, except Mo, exceeded the permissible levels adopted by the National Environmental Protection Agency of Slovenia (UR. LIST RS 68/96).

From analysis of the box and whisker plots in Figure 3, it is obvious that because of the failure of the tailings dam in 2003, higher concentrations of toxic metals (Cu, Pb, Zn, Ni and Cd) are present in Lake Kalimanci sediments than Sasa tailings dam material – with the exception of Mo and As. In general Mo is co-precipitated with fine grained sulphides such as FeS, ZnS, PbS, CuS and CuFeS<sub>2</sub>, all which are found in Sasa ore district material. Minerals within which As is a major constituent include sulphides, sulphosalts, oxides and arsenates. In the Sasa ore district, As is mostly bound to silver minerals (ALEKSANDROV et al., 1999) and arsenopyrite, which is the most abundant ore mineral.

Toxic metals in Lake Kalimanci sediments generally decrease in concentration from north to south. Such a pattern can be explained as a consequence of the Sasa tailings dam failure, when large amounts of material were deposited in the northern part of the Lake, before being slowly deposited southwards along the lake in suspension. The highest amounts of all toxic metals found in Lake Kalimanci sediments were found at sampling location II-3, which lies in the northern part of the Lake Kalimanci. This is likely due to the strong influence of the Kamenica River which enters Lake Kalimanci in the north.

To compare their relative toxic metal contamination with respect to Sasa tailings dam material pollution index levels were also calculated for Lake Kalimanci sediments samples (Table 3). The value of PI for these sediments varied from 21 to 65, representing much higher levels than those observed in other lakes; for instance in Vemband Lake sediments values of PI were in the range of 0.5–3.7 (PRIJU & NARAYANA, 2007). The source of

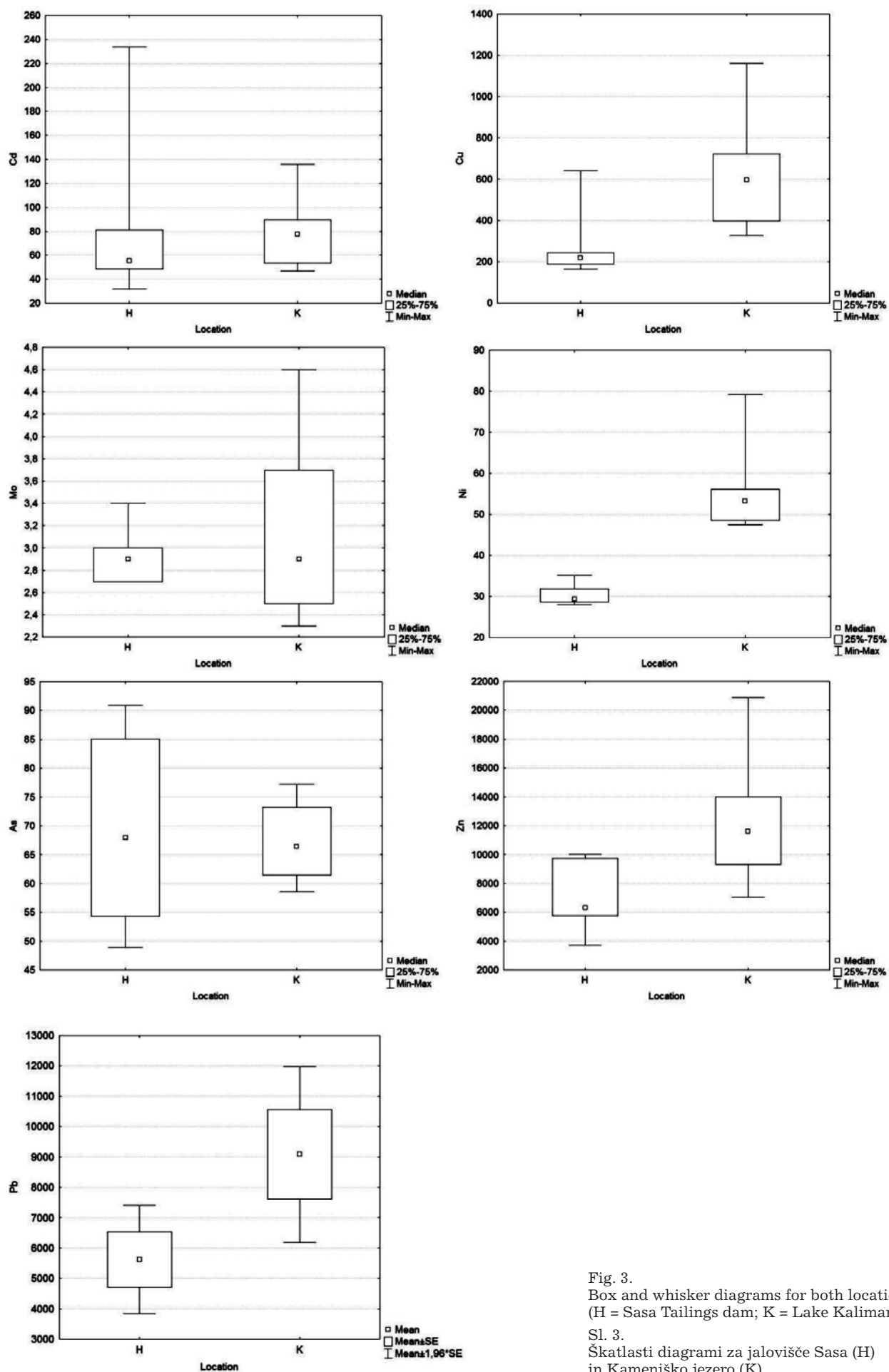


Fig. 3.  
Box and whisker diagrams for both locations  
(H = Sasa Tailings dam; K = Lake Kalimanci)  
Sl. 3.  
Škatlasti diagrami za jalovišče Sasa (H)  
in Kameniško jezero (K).

such high levels of toxic metals in Lake Kalimanci is most likely the tailing dam failure in August 2003, when large amounts of such material were discharged into the lake (ROGAN ŠMUC et al., 2009).

The PI values of sediments from Lake Kalimanci vary from 21 to 65, while those of tailings dam material are in the range 15–60. Normally the pollution index values of the latter should be higher than the former, but in this case, samples were collected from the upper oxic horizon (20 cm depth) of the tailings dam, where presumably the majority of toxic metals were washed into lower reductive horizons. In contrast, after the failure of the dam a large amount of mostly fine particles were discharged in suspension into Lake Kalimanci, with the bulk of that material presumably toxic metals originating from the tailings dam.

### Conclusion

Owing to environmental disaster in 2003, when Sasa tailings dam (part of No. 3 and No. 4) collapsed, we studied, in the present preliminary, the mineralogical and geochemical characteristics of surficial Sasa tailings dam material and geochemical characteristics of surficial sediments from Lake Kalimanci. The mineral composition of surficial tailings dam material generally reflects the surrounding geological background. However, calculated concentrations of toxic metals in Sasa tailings dam are much higher than permissible levels. According to the pollution index, surficial Sasa tailings dam material contains extremely high levels of toxic metals. All metals and industrial minerals produced in mining processes in the Sasa Mine could potentially find their way through Kamenica River to Lake Kalimanci as also into the environment and become pollutants. Evidence of mostly all fine grained pollutants (analysed toxic metals) found within the Sasa surficial tailings dam material has also been found in Lake Kalimanci sediments due to collapsed tailings dam, and thus the high concentrations of toxic metals in the former represent a significant environmental risk. However, after this preliminary study many questions remain unanswered therefore further investigations are required. The most important thing to do is sequential extraction procedure, of tailings dam material as also surficial lake sediments, to find out if toxic metals are available for plants, animals and humans. Because of high concentrations of metals in Sasa tailings dam it would be also necessary to find out a proper stabilization process for tailings dam and nevertheless to find a good mechanism (one of many remediation techniques) to clean out Lake Kalimanci sediments. But all this it is the matter of our future work.

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