CONTAMINATION OF SEDIMENTS BY SELECTED HEAVY METALS IN ARTIFICIAL WATER RESERVOIRS OF REGION BANSKÁ ŠTIAVNICA (THE SLOVAK REPUBLIC)

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Abstact: *Sediments are reliable indicators of environmental pollution. The territory of the Banská Štiavnica region belongs to the contaminated areas of the Slovak Republic. Among the present polluters is an aluminum manufacturing plant in the Žiar nad Hronom, whose emissions influence a wide area, while in the past it was an extensive mining activity, which had a significant adverse impact on the environment of the Banská Štiavnica and its environs. In this study, in the year 2010, the concentrations of selected heavy metals (Cd, Pb, As, Cu, Hg, Zn) have been studied in the sediments of the four artificial water reservoirs (the Počúvadlo, the Little Richňava, the Great Richňava, the Windsachta). In term of contamination importance is eminent of interaction vein and surface water of sediments with inappropriateness anthropogenic activity (past and present mining activities, irresponsible activity by gardening and agricultural), with diversity waste materials, which are standing in wild dumps form. Analyses samples of sediments we were carried by (certificate no. 01871/101/1/2001) flow electro chemistry methods (EcaFlow 150GLP). At present contaminants are relevant especially cadmium (concentration in the space of value from 0.996 to 5.016 mg.kg^{-1} Cd dry matter) and lead (18.13-57.08 mg.kg^{-1} dry matter). Other analyzed elements were arsenic (from 0.068 to 0.298 mg.kg^{-1} dry matter), copper (from 4.22 to 8.97 mg.kg^{-1} dry matter), mercury (from 0.086 to 0.398 mg.kg^{-1} dry matter), and zinc (from 25.9 to 78.64 mg.kg^{-1} dry matter), in the selected artificial water reservoirs. From statistical processing of data we had ascertained, that dependency of selected heavy metals were significantly correlated and strongly positive association, particularly Cd:Pb relation. Where is increase of cadmium concentration in the environment, there is spotted also high accumulation of lead contents, these connection is dependent. We had observed the expressive effect as a toxic elements to the potential increased incidence of individual affection for organisms of this monitored territory. Increasing concentration of chemical elements in the human environment can cause to origination of various disturbances and diseases. The uptake of cadmium, lead and other metals in food is one of the main exposition ways for organisms. Between the most important exposition sources for people belong roll and other root vegetables, green-stuff - vegetables products, cereals.*

Key words: *artificial water reservoirs, Banská Štiavnica ,environment, heavy metals, sediment*

INTRODUCTION

The territory Banská Štiavnica and its surroundings present area loaded with negative factors of environment. Pollution of the natural aquatic environment by metals is receiving still a considerable attention worldwide (LIU ET AL., 2009, HILLER ET AL., 2010). The contaminant sources are especially from anthropological activities. Contaminants defile single components of environment as deposit factor by past mining activities and present whirlwind atmospherically streaming from the Žiar basin territory and from surroundings of the towns the Žiar nad Hronom, the Hliník nad Hronom, the Žarnovica and the Nová Baňa. Also in area of the Banská Štiavnica, there are many factors which negative intervene and influence accumulated level in ground sediment of the artificial water reservoirs. This material contains organics impurities and heavy metals, which can threaten living organisms in the environment. Problems of the environment touches our all and it is inevitable activity and responsible standpoint of people to the environment protection. Industrial and agricultural waste macro- and micropolluting are so large that with their physical and chemical attributes endangers the life of water animals, soil edaphon and plant societies. The most serious social/economic consequence of the polluted environment is a possible injury to health of people and damage to agricultural production (CIBULKA ET AL. 1991, KHUN ET AL., 2000, KHUN ET AL., 2008). Among special questions are the hygienic-toxicological problems of use of dangerous chemical elements. Like presents HEGEDÜSOVÁ ET AL. (1997) in URMINSKÁ ET AL. (2004) and RAPANT ET AL. (2002) in the Slovak Republic occur a few specific areas in which to basic contaminants are associated also undegradable contaminants they are cumulating in several components of environment. These are toxic metals and their compounds.

One of the very exposed area is also the past mining region Banská Štiavnica and its artificial water reservoirs. Sediments, which act as the important repository by sedimentation of suspended particles from the overlying water column (HILLER ET AL, 2010). Dissolved metals adsorb to the suspended sediment depending on many factors such as pH, redox condition and mineralogical and chemical composition of suspended material (JAIN ET AL., 2005, HILLER ET AL., 2010).

The Windsachta reservoir, a water volume of 305.8 m^3, a water depth 14.2 m. The Počúvadlo reservoir, a water volume of 754.3 m^3, a water depth 10.8 m. The Litte Richňava reservoir, a water volume of 26.5 m^3, a water depth 14.2 m.

The Great Richňava reservoir, a water volume of 769.9 m^3, a water depth 21.1 m. These reservoirs are lies at the southwest of the Banská Bystrica in central the Slovak republic (LICHNER ET AL., 2005). These water reservoirs are contaminated by multiple sources of geogenic and anthropogenic origin, especially mining and industrially activities are located in various parts of catchment environment.

The aim of our research was to analyzed the sediments and determined concentrations of selected heavy metals [cadmium (Cd), lead (Pb), arsenic (As), copper (Cu), mercury (Hg), zinc (Zn)] in the surface sediments from four artificial water reservoirs the Počúvadlo, the Little Richňava, the Great Richňava, the Windsachta in the Slovak Republic. Determine of discover the statistical significance of selected heavy metals in the research area and to define the correlation relations.

MATERIAL AND METHODS

The environment´s evaluation quality was based on terrain observation of the region Banská Štiavnica, laboratory analyses of sediments of selected artificial water reservoirs in study area and consequently were the data statistic processed. The eight surface sediment samples from the Počúvadlo, the Little Richňava, the Great Richňava, the Windsachta were collected and using a stainless steel corer in September 2010.

Taking of samples were realized from 0 cm to 20 cm surface. Samples assigned to chemical elements statement were carefully stored in a PVC bags and kept frozen to processing and analysis. To determine the total content of contaminants, we used the sediment fraction with grain size below 0.125 mm. Samples were dried at 40 °C and milled to analytical fineness of 0.09 mm. The 25 g fine soil we added 125 cm^3 2 $mol.dm^{-3}$ HNO_3 and samples were extracted by shaking on a horizontal shaker for 120 minutes.

The extract was obtained by filtration through filter paper no. 390. The analyses of the sediment samples were carried out (certificate no. 01871/101/1/2001) by the flow electrochemistry method. EcaFlow 150GLP is based on flow-through electrochemistry-coulometry. The analysis including the calibration proceeds totally automatically and with much higher sample through put than in classical stripping voltammetry.

The presented systems is enables fully automatic determination of metal ions in the mg.dm^{-3}/kg^{-1} and lower concentration range in the calibration less mode (URMINSKÁ, ET AL., 2010). At present contaminants are relevant especially cadmium and lead. Other analyzed elements were arsenic, copper, mercury and zinc in the selected artificial water reservoirs. For statistical processing were used methods by Spearman's coefficient of correlation. The Spearman's coefficient of correlation ρ_s – is measure of dependency based on the measurement of dependencies between orders.

RESULTS AND DISCUSSIONS

The concentration of selected heavy metals in sediments of the four artificial water reservoirs are summarized in Table 1. At present contaminants are relevant especially cadmium (concentration in the space of value from 0.996 to 5.016 mg.kg^{-1} Cd dry matter) and lead (18.13-57.08 mg.kg^{-1} dry matter). Other analyzed elements were arsenic (from 0.068 to 0.298 mg.kg^{-1} dry matter), copper (from 4.22 to 8.97 mg.kg^{-1} dry matter), mercury (from 0.086 to 0.398 mg.kg^{-1} dry matter), and zinc (from 25.9 to 78.64 mg.kg^{-1} dry matter), in the selected artificial water reservoirs (the Počúvadlo, the Little Richňava, the Great Richňava, the Windsachta). The pH/KCl was in the range of 6.43 to 6.7. Limit values of the monitored elements in the sediments are summarized in Table 2. In the study of years are grading negative effects to the nature initiated by the tendency of land economic self-sufficiency and that above all in the safety the sufficient amount of the raw material and in the increase of industrial production. Actions in the nature don't sweep isolated but they are each another coupled. Therefore when we are creating a new and extending the existing residential and industrial centers, we are gradually changing the nature and globally human environment.

Table 1

Concentration of selected heavy metals in sediment of the monitored water reservoirs in the year 2010

Sampling localities of sediments	Cd mg.kg^{-1} dry matter	Pb mg.kg^{-1} dry matter	Hg mg.kg^{-1} dry matter	As mg.kg^{-1} dry matter	Zn mg.kg^{-1} dry matter	Cu mg.kg^{-1} dry matter
1. sampling place (barrage) the Počúvadlo	0,996	18,13	0,086	0,0689	25,9	6,14
2. sampling place (opposite of barrage) the Počúvadlo	1,128	22,04	0,094	0,0715	31,26	7,59
3. sampling place (barrage) the Little Richňava	2,14	42,09	0,287	0,0812	78,64	4,22
4. sampling place (opposite of barrage) Little Richňava	3,72	39,16	0,398	0,095	86,99	4,823
5. sampling place (barrage) the Great Richňava	1,37	15,43	0,21	0,096	28,13	6,63
6. sampling place (opposite of barrage) Great Richňava	1,12	18,3	0,34	0,11	33,5	8,59
7. sampling place (barrage) the Windsachta	3,127	45,3	0,24	0,23	48,7	7,19
8. sampling place (opposite of barrage) the Windsachta	5,016	57,08	0,319	0,298	59,16	8,97

From the aspect of possible effects of natural geochemical or anthropogenic environment to the health state of the population it is necessary to remember that most important factor is the form in it is given element present, i.e. bioavailability for the living organisms.According to „Methodical direction of Ministry of the Environment the Slovak Republic no. 549/98-2“ concentration of cadmium exceeded target value (0,8 $mg.kg^{-1}$) on sampling places from 1. to 8. from 0.996 $mg.kg^{-1}$ to 5.016 $mg.kg^{-1}$ increase about 24.5 % till 527 %.

Table 2

Limit values of the monitored elements in the sediments

Elements	Methodical direction of Ministry of the Environment SR no. 549/98-2 in $mg.kg^{-1}$ dry matter		Canadian *Sediment* Quality Guideline for the protection of Aquatic Life in $mg.kg^{-1}$ dry matter	
As	TV (1)	29.0	A (3)	6.0
	MPC (2)	55.0	B (4)	33.0
Cd	TV	0.8	A	0.6
	MPC	12.0	B	10.0
Cu	TV	36.0	A	16.0
	MPC	73,0	B	110.0
Hg	TV	0.3	A	0.2
	MPC	10.0	B	2.0
Pb	TV	85.0	A	31.0
	MPC	530.0	B	250.0
Zn	TV	140.0	A	120.0
	MPC	620.0	B	820.0

(1) TV – Target Value - fractional risk, (2) MPC - Maximum Permissable Concentration, (3) A - Dauntessness concentration, (4) B - Imperilling concentration

Concentration of mercury exceeded target value (0,3 $mg.kg^{-1}$) on sampling place no. 4 with value 0,398 $mg.kg^{-1}$ increase about 32.6 %, no. 6 value 0.34 $mg.kg^{-1}$ increase about 13.3 % and no. 8 concentration value 0.319 $mg.kg^{-1}$ increase about 6.3 %.

According to „Canadian Sediment Quality Guideline for the protection of Aquatic Life“ concentration of cadmium exceeded A value (0,6 $mg.kg^{-1}$) on sampling places from 1. to 8. from 0.996 $mg.kg^{-1}$ to 5.016 $mg.kg^{-1}$ increase about 66 % till 736 %.

Table 3

Correlation coefficient between elements in the surface sediments of the selected artificial water reservoirs

	Cd	Pb	Hg	As	Zn	Cu	pH/KCl
Cd	1						
Pb	**0,916448**	1					
Hg	0,607482	0,50985	1				
As	**0,793126**	**0,759603**	0,33328	1			
Zn	0,669087	**0,721798**	**0,715882**	0,167719	1		
Cu	0,10374	-0,00517	-0,06805	0,56422	-0,54696	1	
pH/KCl	0,399268	0,405711	0,577384	0,737891	-0,01355	0,618834	1

Bold type – correlation are significant at the 0.05 level

Concentration of lead exceeded A value (31.0 $mg.kg^{-1}$) on sampling places no. 3 with concentration value 42.09 $mg.kg^{-1}$ increase about 35.77 %, no. 4 with value 39.16 $mg.kg^{-1}$ increase about 26.32 %, no. 7 with value 45.3 $mg.kg^{-1}$ increase about 46.13 % and no. 8 concentration 57,08 $mg.kg^{-1}$ increase about 84,13 %.

Concentration of mercury exceeded A value (0.2 mg.kg^{-1}) on sampling places no. 3 with concentration value 0.287 mg.kg^{-1} increase about 43.5 %, no. 4 with value 0.398 mg.kg^{-1} increase about 99.0 %, no. 5 with value 0.21 mg.kg^{-1} increase about 5.0 %, no. 6 with value 0.34 mg.kg^{-1} increase about 70.0 %, no. 7 with value 0.24 mg.kg^{-1} increase about 20.0 % and no. 8 concentration 0.319 mg.kg^{-1} increase about 59.5 %. Correlation coefficient between monitored elements in the surface sediments of the selected artificial water reservoirs are summarized in Table 3.

For the relation Cd : Pb – Spearman's coefficient have the value 0.91645 and presents dependency between the cadmium and lead in the sediment. By the significance level 0.05 so with 95% reliability can be stated that Spearman's coefficient 0.91645 is high probative because the significance level 0.0014 is lower than 0.05. Dependency is higher positive. For the relation Hg : Zn – Spearman's coefficient have the value 0.71588 and presents dependency between the mercury and zinc in the sediment.

By the significance level 0.05 so with 95% reliability can be stated that Spearman's coefficient 0.71588 is probative because the significance level 0.0458 is lower than 0.05. Dependency is positive. For the relation Pb : Zn – Spearman's coefficient have the value 0.72180 and presents dependency between the lead and zinc in the sediment.

By the significance level 0.05 so with 95% reliability can be stated that Spearman's coefficient 0.72180 is probative because the significance level 0.0432 is lower than 0.05. Dependency is positive. For the relation As : Cd – Spearman's coefficient have the value 0.79313 and presents dependency between the arsenic and cadmium in the sediment.

By the significance level 0.05 so with 95% reliability can be stated that Spearman's coefficient 0.79313 is probative because the significance level 0.0188 is lower than 0.05. Dependency is positive. For the relation As : Pb – Spearman's coefficient have the value 0.75960 and presents dependency between the arsenic and lead in the sediment.

By the significance level 0.05 so with 95% reliability can be stated that Spearman's coefficient 0.75960 is probative because the significance level 0.0288 is lower than 0.05. Dependency is positive.

From statistical processing of data we had ascertained, that dependency of selected heavy metals were significantly correlated and strongly positive association, particularly Cd : Pb relation. Where is increase of cadmium concentration in the environment, there is spotted also high accumulation of lead contents, these connection is dependent. This problems of risk chemical elements action are the same as a many other countries (WILDI ET AL., 2004, KAMALA - KANNAN ET AL., 2008, ÖZTŰRK ET AL., 2008).

According to SALMINEN ET AL., (2005) high Cd values in sediment (>0.44 mg.kg^{-1}) occur in a north-west to south-east belt extending from Ireland through England, Germany, the Czech Republic, Slovakia and eastern Hungary. The spreading of sewage sludge and phosphate fertilizers, both of which may contain high levels of Cd, is prevalent in this whole area. In this region there are also known Pb-Zn ore deposits, which contain enhanced Cd levels (e.g. Rammelsberg and Freiberg in Germany, Kutná Hora in the Czech Republic, Banská Štiavnica in Slovakia and Recsk in Hungary).

Point anomalies in sediment occur in the Lavrion mining area in Greece (23.6 mg.kg^{-1}), in Slovakia (15.2 mg.kg^{-1}), which is possibly related to the Banská Štiavnica Pb-Zn mineralization, in the Vesdre basin in Belgium (13 mg.kg^{-1}), caused by metallurgical pollution, east-central Germany (12.0 mg.kg^{-1}) downstream of the mineralized Erzgebirge region, in north-east England (11.7 mg.kg^{-1}) with mining activities. In the Extremadura-Córdoba region in southern Spain, two high values correspond to catchment basins containing important Pb-Zb mining districts (Valle de Alcudia-Pedroches and Azuaga). Cadmium in sediment has a strong correlation with Zn, a good correlation with Pb, Cu and Hg (SALMINEN et al., 2005). High total

Pb values in sediment (>35 $mg.kg^{-1}$) occur throughout England, Wales and southern Scotland, related to extensive Pb-Zn mineralization, but also to anthropogenic contamination in industrial areas and leaded petrol.

Another extensive belt with high total Pb values in sediment stretches from eastern Belgium over most of central Germany to the Czech Republic and southern Poland, which includes many Pb-Zn mineralized areas and industrialized areas. Point total Pb anomalies in sediment occur in the French Pyrenees, north-western Poland, in southern and eastern Slovakia (Banská Štiavnica Pb-Zn, Rudnaňy Fe-Cu-Hg-Ba), and the Lavrion polymetallic mineralized area in Attika, Greece. Total Pb in sediment shows a strong correlation with Zn, a good correlation with Cd. This association points to mineralization and possibly contamination by mining (SALMINEN ET AL., 2005).

High values of selected heavy metals in sediment map mineralized areas, and the pollution caused by mining and metallurgical processing, but also the spreading of sewage sludge and phosphate fertilizers in agricultural areas (URMINSKÁ ET AL., 2004, SALMINEN ET AL, 2005, HRONEC ET AL., 2010).

CONCLUSIONS

In conclusion, the patterns of high values of selected heavy metals in sediment map mineralized areas and the pollution caused by mining and metallurgical processing, urban and other polluting anthropogenic activities. Our results are certificated, that toxic metals and their behavior in the environment dependence on their chemical-geochemical behavior. Specified region is but strongly loaded with others chemical elements (cadmium and lead) that significantly affect the state of disease damage for plants and animals in the mining region Banská Štiavnica.

Ascertained facts are expressly related to geochemical ground and implicitly to unsuitable mining, industrial and agricultural activity in the region and with migration and consequently with the increased concentration of insurants of various chemical-physical character in each individual factors of environment.

The risk heavy metals recently is this term being narrow connected with these elements, which are cause to the undesirable toxic effects and contaminating the environment. Consequently can get the heavy metals into environment from various anthropogenic sources or from natural geochemical processes.

They are being considered like elements: relative enough widened in earth crust; they are being mined and exploited in average quantity; they are a content of materials, which people coming in contact; they have got a toxic effect to organisms and they have been cause to undesirable effects in biogeochemical cycle (FERGUSSON, 1990, URMINSKÁ et al., 2008). Increasing concentration of chemical elements in the human environment can cause to origination of various disturbances and diseases.

The uptake of cadmium, lead and other metals in food is one of the main exposition ways for organisms. Between the most important exposition sources for people belong roll and other root vegetables, green-stuff - vegetables products, cereals (HEGEDŰSOVÁ - HEGEDŰS – MUSILOVÁ, 2006, HRONEC et al., 2002, HRONEC et al., 2010).

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