



# Impact of Former Mining Activity to Soil Contamination by Risk Elements

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# Abstract

The level of contamination of the environment in the past few decades is largely influenced by the anthropogenic activity. The tailing pond, where is stored landfilled flotation sludge with the high content of mercury (more than 46 mg/kg), is located about 500 metres from the main processing plant. In the present work, we have focused on the assessment of the level of contamination of the upper horizon (0-0.1 m) of agricultural and forest land in the cadastral territory of Markušovce, where is located the emission source and the pond. We determined 28 sampling points by GPS method on areas of our interest. All soil samples have been subjected to the analyses for the detection of active and exchange soil reaction, the content of humus (%), total concentration of Hg and the concentrations of Cd, Pb, Cu and Zn (mg/kg) in the solution of the aqua regia. The results confirmed an extremely high level of soil contamination by the studied elements. Mercury concentration was in the range of 0.69-90.7 mg/kg, while the median value exceeding the standard established by applicable legislation over 30 times. The content of other heavy metals closely correlated with contaminant concentration, which varied in wide intervals as evidenced by high relative standard deviations (Cd: 1.62 ± 34.2 mg/kg; Pb: 37.6 ± 60.7 mg/kg; Cu: 23.5 ± 124 mg/kg Zn: 108 ± 40.4 mg/kg). Even in the case of these two elements (Cd and Pb) we have in excess of the limit value at the level of the averages. The results observed that heavy metal concentrations indicate a high level of soil contamination in the study area, which will be reflected to the contamination of other components of environment and food chain.

## 1. Introduction

The level of environmental contamination with risk elements represents a significant risk in relation to the quality of the food chain. In the last decades a significant increase in the concentration of contaminating elements such as cadmium, mercury and arsenic in all components of the environment was reported (Granero & Domingo, 2002; Li et al., 2008). It is mainly due to continuous increase of industrialization of human society (Govil et al., 2008; Huang et al., 2007; Li et al., 2008). Increasing demands for raw materials result in adverse The level of environmental interventions in nature. contamination on the local scale is also amplified by metallurgical industry, which increases the level of contamination of topsoil in forest and agricultural ecosystems mainly through atmospheric deposition (Biester et al., 2002; Jiang et al., 2006). Heavy metals are ubiquitous part of the environment as a result of mutual natural and anthropogenic activities causing increased exposure of human populations to their effects through various channels (Póty et al. 2012). Increasing concentrations of certain risk elements, especially their mobile forms can cause serious environmental concern about contamination and accumulation in soil, vegetation, animals, respectively, surface and ground waters (Chopin & Alloway, 2007). The primary source of environmental contamination is mainly metals, whose main part is particularly lead, zinc or copper, in addition, antimony, arsenic, mercury, cadmium, thallium, gallium, and others. In addition to the production of metals is a very important source of environmental contamination with metals and burning of fossil fuels, especially coal. Fly ash from the incineration of atmospheric leakage through polluted soil (Steinnes et al., 2005). Increasingly important source of environmental pollution with heavy metals is becoming a burning municipal waste streams and pollution effluents containing elevated levels of toxic metals (Bencko et al., 1995). Soil contamination reveals itself as overreaching the limit value of at least one risky substance. In case of the risk elements we talk about their overall content. The available data regarding the total content (after the soil decomposition using potent mineral acids, especially Aqua regia used in the Slovak Republic since 2004) may give us the necessary information in accordance with the valid legislation (including the approved maximum content of risk elements and the level of contamination). Relationships between the risk element contents in the soil and their concentrations in plants have been observed only in case of overreaching their established total concentrations in soils. Analyses the transport of the risk elements to the plants, the interactions soil - plant are especially crucial and studied by a variety of authors (**Castaldi** *et al.*, **2009**). On the other hand, low concentrations of risk elements in soils, e.g. in acid soils, are related to their exceeded limits in plants (**Kobza** *et al.*, **2007**).

# 2. Material and methods

# 2.1 Study area

In the present work we have focused on determination the level of contamination of top soil in the cadastral territory of Markušovce, which is located about 10 kilometres south-east from the district of Spišská Nová Ves. In the southern part of the territory is an old environmental burden (tailing pond, areal of the former mercury smelter and heaps of pyrites). The history of mining and processing iron ores containing Hg industry dates to the 2<sup>nd</sup> half of the 19<sup>th</sup> century. About 52 small mercury smelters were worked and were processed mined cinnabar and tetraedrite till the year 1963. According to estimates several hundred tons of elemental Hg had been issued to the environment during this period. New plant on the surface was launched into operation in 1963. The major sources of mercury were the heat of operation, agglomerations, mercury smelter, tailing pond and heaps of roasted material. The main mercury smelter was launched into operation in 1969. The summary of Hg emissions from all sources was 142 tonnes for the years 1963 - 1993 (Svoboda et al., 2000). The mercury smelter emitted to the atmosphere 2.60-4.64 tons of mercury per year, during the period of 1980-1988 (Hančuľák et al., 2006; Hronec et al., 1992). The emitted mercury subsequently contaminated the area located in the direction of prevailing winds (north-south), which is strongly reflected in the level of pollution of all elements of the environment within 5 kilometres from the mercury smelter. The study area is of volcanic origin, characterized by frequent occurrence of geochemical anomalies, which penetrate up to surface. The substrate consists of naturally high concentration of mercury, copper, lead, zinc and cadmium, which sometimes occurs in the elemental form (Angelovičová & Fazekašová, 2014). The study area, indicating the emission source and sampling points is shown in Figure 1.



**Figure 1.** The study area, indicating the emission source (sampling point No. 1) and sampling points.

## 2.2 Sampling

Sampling and pre-analytical procedure. Soil samples (n = 28) of studied area were taken by randomly using pedological probe of depth 0.0 - 0.1 m in 2013. Sampling points were determined using GPS navigation device at regular intervals of 1 km from the emission source (sampling point 1). Each sample consisted of a mixed sample of soil from the sampling point (5 samples were taken from each sampling point and mixed together).

## 2.3 Pre-analytical procedure

The samples were dried at room temperature to constant weight and sieved through a sieve with a mesh diameter of 2 mm before all analyses. We carried out an analysis to determine the active (pH - H<sub>2</sub>O) and exchange (pH - CaCl<sub>2</sub>) soil reaction, humus content (%), total concentration of mercury and total concentration of monitored heavy metals (Cd, Cu, Pb, and Zn) in extract of Aqua regia (ISO 11466). High purity chemicals for all operations were used. Mineralization of soil samples was done by 10 cm<sup>3</sup> of Aqua regia (2.5 mL HNO<sub>3</sub> and 7.5 mL HCl, Merck, Germany) using microwave digestion unit Mars X-press 5 (CEM Corp., USA) in closed PTFE vessels. Digestion conditions for the applied microwave system were as follows: the heat was run up to 180°C for 15 minutes and kept constant for 15 minutes. A blank sample was carried out in the same way. The digests were subsequently filtered through a quantitative filter paper Filtrak 390 (Munktell, Germany) and filled up with deionized water to a volume of 100 mL (Árvay et al., 2013).

#### 2.4 Analytical procedure

The element determinations were performed in a Varian AA240Z (*Varian*, Australia) atomic absorption spectrometer with Zeeman background correction. The content of copper and zinc were determined by Atomic Absorption Spectrometry device with flame AA240FS Varian (Varian, Australia). The graphite furnace technique was used for the determination of Cd and Pb. The total mercury concentration was determined in the homogenized dried soil samples (0.005-0.01 g) using a cold-vapour AAS analyser AMA 254 (*Altec*, Czech Republic) with a detection limit of 0.5 ng/g. Mean difference between duplicates were up to 5% (Árvay et al., 2015; Svoboda et al., 2006).

#### 2.5 Statistical analysis and risk assessment

All statistical analyses were carried out using the statistical software Statistica 12.0 *(Statsoft,* USA). Descriptive data analysis included a minimum value, maximum value, median and standard deviation. The obtained data on the concentration of risk elements we have compared with the limit values that define the legislative norm Act No. 220/2004. Graphical outputs of spatial interpolation of monitored parameters are isoline maps that were created by the program ArcView 9.1 environment, which are depicted the concentration of monitored contaminants.

#### 3. Results

According to our findings relatively small study area of cadastral territory Markušovce (19.2 km<sup>2</sup>) is in all parameters significantly heterogeneous. Active but also exchange soil reaction as the basic chemical parameters, which have a significant influence on the behaviour of contaminants in the soil environment, varied in wide intervals. Active soil reaction varied in the range  $6.40\pm1.34$  (median $\pm$ st.dv). Exchange soil reaction in CaCl<sub>2</sub> solution with c=0.01 Mol/L was at the level of the median value in the range  $6.07\pm1.31$ . A wide range of both parameters is

probably due to the different morphology of the territory, the nature of the samples and the long-term effects of immission fallout, which correlates with the long-term prevailing winds (north-westerly and westerly).

Our findings show that  $3.51 \text{ km}^2$  of area (18.3%) is strongly acidified (pH<4.5), which can affect the which can affect the mobility and subsequent bioavailability of monitored contaminants. The content of humus (%) varied in a wide range (2.72±3.41), which is caused by the different nature of the samples (Table 1).

Statistically significant effect of soil parameters on the level of contamination of topsoil with heavy metals is also confirmed by

correlation coefficients (P<0.05, P<0.01) and these results are shown in Table 2.

**Table 1.** Basic characteristics of sampling points and monitoring parameters of soil samples

SP – sampling point; D-ES – distance from emission source; SC – sample characteristic; ES – emission source; AS – agricultural soil; F – forest; UA – urban area; **XXX** – exceed the limit value that is defined by law; \*Act No. 220/2004.

SP	D-ES (km)	SC	Soil parameters							
			pH H₂O	pH CaCl₂	Humus (%)	Hg <sub>tot.</sub> (mg/kg)	Cd (mg/kg)	Pb (mg/kg)	Cu (mg/kg)	Zn (mg/kg)
SP-01	0	ES	6.81	5.93	3.75	63.2	3.52	54.0	300	126
SP-02	1	AS	6.58	5.16	2.06	9.13	1.69	33.2	40.6	82.2
SP-03	1	AS	6.05	4.83	2.42	7.25	1.16	23.4	36.5	99.3
SP-04	1	AS	6.01	5.21	1.45	7.03	1.56	26.4	29.5	71.5
SP-05	1	AS	5.61	4.93	2.72	11.3	0.97	25.1	36.5	104
SP-06	1	F	7.06	6.48	11.2	10.4	3.73	125	136	253
SP-07	1	F	3.82	2.85	14.8	73.1	1.14	47.6	7.40	40.7
SP-08	5	F	4.30	3.26	5.63	6.74	1.29	35.8	33.7	38.8
SP-09	6	F	4.47	3.60	6.29	15.7	1.85	44.3	37.5	68.7
SP-10	1	AS	8.02	7.16	1.57	3.08	3.85	64.0	48.2	173
SP-11	2	F	4.91	4.01	3.15	6.65	1.74	43.6	38.1	57.2
SP-12	3	AS	6.17	5.41	4.05	1.26	3.00	56.2	26.9	151
SP-13	1	AS	5.94	7.04	1.45	1.35	1.02	24.9	52.5	78.7
SP-14	2	AS	5.08	5.91	1.82	1.05	1.34	27.9	36.6	84.5
SP-15	3	AS	6.67	7.35	3.81	1.81	5.15	43.7	39.1	88.6
SP-16	1	AS	8.13	7.32	1.33	2.74	3.01	45.3	38.2	94.3
SP-17	2	UA	7.94	7.19	2.72	0.70	3.17	47.7	105	387
SP-18	3	UA	7.94	7.27	1.57	0.72	1.68	25.6	24.0	86.2
SP-19	4	AS	8.13	7.16	3.27	0.69	1.80	25.9	18.9	79.3
SP-20	1	AS	7.22	7.34	4.30	90.7	1.94	45.8	331	189
SP-21	2	AS	5.40	6.33	1.88	1.64	1.29	27.1	47.9	112
SP-22	4	F	7.61	6.82	12.3	9.36	3.24	133	46.1	364
SP-23	5	AS	6.85	6.05	3.51	1.45	0.94	29.2	19.7	127
SP-24	6	F	6.38	5.34	1.82	2.27	1.07	27.4	20.3	282
SP-25	5	F	7.78	6.91	1.94	16.3	1.06	25.2	52.2	125
SP-26	5	AS	4.33	6.08	2.54	1.01	1.06	39.4	35.0	152
SP-27	6	AS	6.41	6.84	3.93	0.70	1.51	41.2	55.8	270
SP-28	7	AS	4.10	5.48	1.51	0.45	1.52	32.7	29.6	191
Limits value and descriptive statistics										
Limit value*			-	-	-	0.50	0.70	70.0	60.0	100
Minimum			3.82	2.85	1.33	0.45	0.96	23.4	7.40	38.8
Maximum			8.13	7.35	14.8	90.7	5.15	133	331	387
Median			6.40	6.07	2.72	2.91	1.62	37.6	37.8	108
Standard deviation			1.34	1.31	3.41	23.1	1.11	26.5	76.1	92.3

The highest levels of humus content were detected in the southern part of the territory. There is located forest, which is characterized by a higher content of organic matter in the top layer of soil compared with agricultural land. There is a significantly higher level of oxidation and the consequent degradation of organic matter occurs due to periodic aeration of topsoil. Data of the pH and humus in the study area are shown in Table 1 and their graphical interpolation on Figure 2.



**Figure 2.** Spatial interpolation of active, exchange soil reaction and humus content of soil samples from study area

The total mercury concentration in the study area varied in a wide range. The level of the median value was 2.91±23.1 mg/kg. Figure 3 shows strong local contamination of soil within a radius of 1 km from the emission source in the direction of the prevailing winds. From the hygienic point of view, it is necessary to emphasize that in an area of more than 15 km<sup>2</sup> (78.5%) was detected an excess of the mercury limit value (0.5 mg/kg). The results of mercury content in the soil closely correlate with the results of other authors, who pointed to the high level of contamination of all environmental elements (Árvay *et al.*, 2017; Bobro *et al.*, 2006; Demková *et al.*, 2017; Dombianová, 2005; Takáč *et al.*, 2008). It represents a significant risk of possible contamination of agricultural production and food and feed chains (Árvay *et al.*, 2014).

The cadmium content of study area varied in the range 1.62  $\pm$ 34.2 mg/kg. Exceeding a limit value of 0.70 mg/kg was recorded at the level of the mean more than two times. High level of topsoil contamination is shown by isoline map (Figure 3). We can state that study area is contaminated with risk elements. The highest concentration of the contaminant was ascertained at the sampling point SP-15 and in the north-eastern part of the study area. The limit values were exceeded for almost 96% of the study area. The lead content exceeds the limit value of 75 mg/kg at two sampling sites (6 and 22). These points were located at the northern part of the area. The median values of the lead content varied in the range 23.4 ± 60.7 mg/kg. The concentration of heavy metals exceeded the limit value by 10.3 % of total area. It does not pose any risk to contamination of the agricultural products, because it is a forest soil. Between the lead content of the topsoil and humus content is a high positive correlation (Table 2) and it demonstrates a high level of affinity of the monitored parameters, what is confirmed by the results of other authors Árvay et al. (2017).

The concentration of copper and zinc in the topsoil was compared to the level of mercury. It is not represent a significant risk. The level of contamination of study area is showed in Figure 3. Detected concentration of copper (37.8±76.1 mg/kg) and zinc (108±92.3 mg/kg) point to a high variability of monitored contaminants in the study area (Figure 3). In the case of all contaminants, we have seen local pollution in the vicinity of the emission source, which is confirmed by the findings of the authors **Angelovičová & Fazekašová (2014)**. Two sampling points (17 and 18) are an exception, where was measured higher concentration of zinc compared to the other sampling points. Our findings show that it is contaminated with copper 19.8% and zinc 69.9% of the study area. Obtained data with descriptive statistical evaluation are shown in Table 1.

**Table 2.** Correlation between risk elements and chemicalparameters

	Hg	Cd	Pb	Zn	Cu
рН – H2O	-0.08	0.46*	0.24	0.39*	0.25
pH – CaCl <sub>2</sub>	-0.17	0.41*	0.15	0.42*	0.27
Humus(%)	0.42*	0.20	0.70**	0.19	0.07
Hg		0.02	0.11	-0.09	0.72**
Cd			0.61**	0.31	0.30
Pb				0.55**	0.24
Zn					0.23

\*/\*\* correlations are significant at P $\leq$ 0.05, P $\leq$ 0.01 level, respectively



**Figure 3.** Spatial interpolation of total concentrations Hg, Cd, Cu, Pb, and Zn (mg/kg).

# 4. Conclusion

In the present paper, we aimed at detection of the level of burdened topsoil at the area of the former metallurgical company, which in 1993 was processing ore with a high content of Hg and other monitored elements. According to the results, it can be concluded that the area within 5 km from the emission source is heavily contaminated with monitored heavy metals, thus permitting passage of contaminants from the abiotic environment to the food chain, which in turn reflected the deterioration of health of the local population and thus to increased mortality.

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#### **Declaration of interest**

The authors report no conflicts of interest. The authors alone are responsible for the content and writing of the paper.

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