

GEOCHEMICAL BACKGROUND IN HEAVY METALS AND HUMAN HEALTH RISK ASSESSMENT AT AN ORE MINE SITE, GYÖNGYÖSOROSZI (NORTH HUNGARY)

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ABSTRACT The pseudototal concentrations of arsenic (As), cadmium (Cd), lead (Pb), mercury (Hg) and zinc (Zn) were determined in the topsoil samples collected at Gyöngyösoroszi (North Hungary) and outskirts of the town. The main aim of this study is to summarize the geochemical features of the area and to evaluate the human health risk for the residents in the town based on the pseudototal concentration values in soil. The results indicate variable pollution of the topsoil and determined two area types with the most hazardous heavy metal contamination. According to the high concentration values measured in the sediment of creeks, the lead and zinc contents are the most striking, whereas the real hazard is caused by the high arsenic concentration. Mercury values exceed the threshold level only at a few sites due to the geochemical features independent from the mining.

Keywords: arsenic, heavy metals, geochemical relationship, mining, soil pollution, human health risk assessment, Gyöngyösoroszi, Hungary

1. INTRODUCTION

The study is focused on the town of Gyöngyösoroszi with agricultural activities about 1000 m far from the tailing dump of the flotation plant. However the tailing dump is considered as a point source which may contaminate the surrounding area (Horváth & Gruiz, 1996), the most increasing threat is the high concentration of arsenic and heavy metals (e.g. cadmium and lead) in the banks and lens along the creeks. The Toka & Száraz creek cross the studied area, flood the floodplain twice a year and in consequence the vegetable gardens are contaminated with heavy metals.

These metals may accumulate to toxic concentration levels in the soil and in

plants (Andráš et al. 2007) which leads to the impairment of the quality of human life (Lee et al., 2005; Borošová, et al., 2008). The major object of this study is to estimate the health risk of arsenic, cadmium, mercury, lead and zinc in flooded and non-flooded vegetable gardens in the town considering the former mining activity and the natural geochemical relationship.

Elevated arsenic and heavy metal concentration values in home environment were also found at other former mining areas (Rieuwerts et al, 2006), where the arsenic represents the greatest threat to human health.

2. MATERIALS AND METHODS

2.1. Geographical setting and geology

The studied area is located at the SW part of Mátra Mountains, North Hungary along the left side of the Toka creek from the northern part of the mine (Fig. 1.). The Száraz creek run from Eastern to the Toka creek and it leak at first in to the Száraz brook from the drain pipe of the tailing dump, then to the Toka creek at the agricultural water reservoir. These creeks flow from North to South through the town Gyöngyösoroszi (47,826°N, 19,894°E) to a water reservoir at the western part of the city Gyöngyös and collect the runoff of the mountains and the abandoned tailing dump.

The territory is the southern part of the biggest collapse caldera in the Western Mátra. The mountains consist mostly of Miocene age amphibole andesite, its pyroclastics and pyroxene andesite on the top. The terrestrial stratovolcanic series contain dacite and rhyolite bodies around adventive craters (Vidacs, 1966; Nagy et al., 1986). The stratovolcanic andesite series is 950-1200 m thick around Gyöngyösoroszi (based on the structural prospective bore Gyo-2 in the valley Hidegkúti and Gyo-5 in the Western part of Károlytáró settlement) containing more than 1% Pb-Zn concentration in the 0.5-1 m thick dikes. The overlaying bed is formed by 1-21 m thick clay complex of Pleistocene and Holocene age which contains coarse-grained andesite detritus (Nagy et al., 1986).

According to the postvolcanic hydrothermal activities chalcophile elements build up the epithermal and mesothermal ore minerals (Nagy et al., 1986; Kun et al., 1988). In the Gyöngyösoroszi area the most frequent ore minerals are pyrite and marcasite (cubic and rhombic FeS₂), galena (PbS), sphalerite and wurtzite (ZnS) and chalcopyrite (CuFeS₂) in dikes (Vidacs, 1966; Vetőné Ákos, 1984, 1988) together with gangue minerals mainly quartz, (hydro)quartzite, calcite and gypsum, rarely several secondary sulfates (e. g. gypsum) and carbonates (e.g. siderite), bishop's stone (amethyst) and fluorite through potassic metasomatism (Vidacs, 1966; Nagy et al., 1986; Varga, 1992; Nagy, 2006). Around Gyöngyösoroszi cinnabarite (HgS) can be found in quartzite bodies locally only in boreholes and in Asztag-kő quarry 4.2 km far away from Gyöngyösoroszi to North-East. On the left bank of Toka creek the concentration values of mercury could be extremely high locally due to the hot springs (Kristó, 2004) (Fig. 1), former geysers and dikes in NE-SW direction from Asztag-kő (Nagy et al., 1986).

Three crystallization phases were separated based on the temperature of the mineral formation: at 260°C, 210-230°C and 150-175°C. Generally, lead and zinc are

contained in the dikes of the Badenian whereas copper is dominantly in the Karpatian coarsely crystalline stratovolcanic andesite series and arsenic occurs mainly in the dikes of the small altered porphyritic andesite or tuffs series (Nagy et al., 1986). From Middle Centuries the Pb-Zn-Cu mining activities were based on these revived and intersected dikes with several directions of dip.

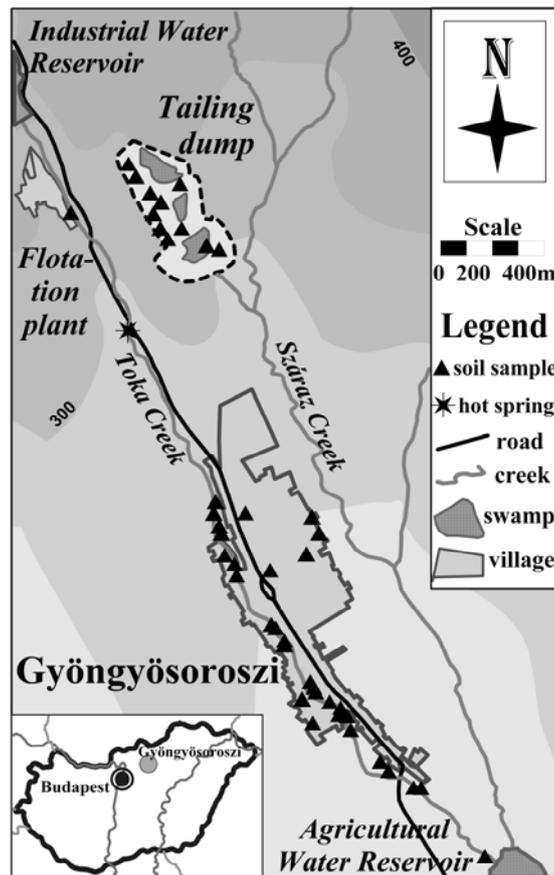


Figure 1. Topographic map of the studied area with the sampling sites (Sipter et al., 2008)

2.2. The origin of the soil pollution

Mining began in the Medieval times and it was expanding rapidly but intermittently throughout the 19th century until 1929. After two decades of interruption, mining continued more intensively and between 1954 and 1985 the total amount of 3 920 000 tons of ore were mined and transported to the flotation plant near Gyöngyösoroszi (Nagy et al., 1986). The water reservoir in Toka valley and the treatment of the water were necessary to meet the water demand of the flotation technology at the flotation plant and to lime the acidic mine drainage at the drain adit. The dams of the water treatment, the industrial (Gyöngyösoroszi) water reservoir and the tailing dump in Száraz valley periodically were built according to the mining from

the coarse fraction of waste material (Kun et al., 1966).

During the process of construction in the mine site several unfinished dams breakdowns follow each other and the pipe between the flotation plant and the tailing dump was washed often inattentive therefore the mine's waste was deposited into the creeks. The fine-grained fraction reached the agricultural fields (Kun, 1966) and its chemical components influenced the soil-forming process and accumulated in plants (Záray Gy. et al., 1991; Lonsták, 1992; Marth et al., 1994; Ódor et al., 1998; Fügedi, 2004, 2006; Fügedi et al., 2007). The over again resedimented medium- and coarse-grained fractions formed bars and lens along the creeks slowed down mainly southwards from Gyöngyösoroszi and became into the typical "yellow sand" strata generally in a depth of 20-50 cm (Fügedi, 2004; 2006).

Due to the mining, the sulphide minerals in the old drift gallery have been oxidized and sulfurous acid was formed by infiltration and by bacterial leaching (Andráš et al., 2008). The acid mine drainage was continuously limed and the lime-precipitate was settled and dumped in open disposals. Before the establishment of the neutralization plant in 1985, the mine outflow entered directly into the surface water system, where it was *in situ* neutralized. This lime precipitate has been part of the sediment of Toka creek and of water reservoirs. In 1996 a remarkable precipitation event (105 mm) occurred during one day, the water overflowed the dams and caused a huge flood in the village (Vánca, 1996).

More than 100 000 m³ of contaminated mud accumulated with high heavy metal content due to the liming at the drain adit in the industrial water reservoir and locally in the agricultural water reservoir. In 1996 the toxic sediment was spreading over the surrounding vegetable gardens and the huge flood resedimented again the former banks and lens along the creeks farther from the banks of creeks and further southwards than once earlier. The watering of agricultural fields from this reservoir and from the creeks contributed to the high heavy metal content in the soil. The continual changing but always high heavy metal contents in soil pose a long-term threat to the environment and the human health, too (Madarász, 2004).

2.3. Sampling and laboratory analyses

Two main territory types were selected because of different type of soil contamination and different land uses: a) soil forming the cover layer of the flotation-waste at the tailing dump and the soil used by inhabitants of the village as a vegetable gardens.

Sampling was carried out in July 2001 at both territory types together at 46 sampling sites (Fig. 1). At each sampling site duplicate samples were collected by random sampling method. Samples were taken from the upper 20 cm of soil and packed into polyethylene bags. Samples were dried at 25°C until constant weight and sieved through 2-mm nylon mesh. 0.1 g subsample was digested with 3 ml *aqua regia* (HNO₃:HCl = 1:3) according to Hungarian standard 21470-50 (Hungarian Standards Institution, 1998). The arsenic and metal contents were measured by inductively coupled-mass spectrometry (ICP-MS) according to US EPA Method 6020 (US EPA, 1994).

2.4. Risk assessment

Human health risk assessment to arsenic, cadmium, lead, mercury and zinc was evaluated based on soil contamination. Non-carcinogenic risk was characterized using a hazard quotient (HQ), what is the ratio of the average daily dose (ADD) to the reference dose (RfD). RfD is defined as a daily intake rate that is estimated to cause no appreciable risk to adverse health effects, even to sensitive populations, over a specific exposure duration. If HQ is larger than 1, so the ADD of particular metal exceeds the RfD, indicating that there is a potential risk associated with that metal. The hazard index (HI) is the sum of hazard quotient for each exposure routes (Paustenbach, 2002).

Two exposure pathways were selected due to the site-specific land uses and the feature of metals: ingestion of soil and ingestion of home-produced vegetables. The average daily dose was calculated with the following equations (US-EPA, 1997b):

$$\text{Ingestion of soil: } ADD = \frac{C_S \times IR_{SOIL} \times EF \times ED}{BW \times AT} \quad (1)$$

$$\text{Ingestion of vegetables: } ADD = \frac{C_S \times BCF \times F_{DRY} \times IR_{VEG} \times EF \times ED}{BW \times AT} \quad (2)$$

where C_S is concentration in soil (mg/kg), IR is ingestion rate (soil or vegetable) (kg/day), EF is exposure frequency (days/year), ED is exposure duration (years), BW is body weight (kg), AT is average time (days/year), BCF is bioconcentration factor for vegetables (-), F_{DRY} is fraction of dry weight (-).

3. RESULTS AND DISCUSSION

3.1. Geochemical features of the studied area

Median concentration and median absolute deviation (MAD) of As, Cd, Hg, Pb and Zn both in the soil of tailing dump and in the soil in the village are shown in table 1.

Table 1. Arsenic and heavy metal content of soil samples (mg/kg) compared to background concentration values (Fügedi, 2006) and to the B values (Hungarian Joint Decree No. 10/2000)

Sample type	Parameters [mg/kg]	As	Cd	Hg	Pb	Zn
Tailing dump	median	55.7	1.46	0.89	125.5	435.5
	MAD	30.0	0.40	0.28	49.5	113.5
Town	median	64.5	0.92	0.45	63.5	305.0
	MAD	20.0	0.43	0.45	33.0	137.0
Background	median	35.0	0.40	0.14	35.0	80.0
Pollution limit value (B)		15.0	1.00	0.50	100.0	200.0

The median concentration values of arsenic and heavy metals in the soil of tailing dump exceeded the Hungarian pollution limit values. In the soil of village the

median of arsenic and zinc exceeded this limit. The high median absolute deviation has shown the inhomogeneous distribution of soil contamination.

However, these data from the tailing dump and from the sediments of the water reservoirs are high, the concentration values of the samples near to the creek show greater lead risk values for the people growing vegetables in the flooded gardens as the non flooded gardens. The highest values of zinc and lead were measured around the flotation plant, the greatest value of cadmium occurred in the tailing dump. This is interpretable with the mining and flotation technology, which target was lead and zinc dressing. The correlation values show the relationship among the elements regarding their concentration (Tab. 2). The zero value means no relationship and a value near to 1 indicates close connection. The relatively high cadmium and low mercury concentration in the whole studied area and the high correlation values between lead, cadmium and zinc are characterized by their geochemical features.

Table 2. Correlation among the studied elements

Correlation	As	Cd	Hg	Pb	Zn
As	1				
Cd	0.452	1			
Hg	0.194	0.212	1		
Pb	0.440	0.934	0.156	1	
Zn	0.390	0.901	0.227	0.860	1

Zinc is in hydrothermal ore deposits most frequently accompanied by lead. Both these elements are the main elements of the two most important ore minerals galena and sphalerite at Gyöngyösoroszi. In the sphalerite (α -ZnS) crystal lattice some zinc atoms can be substituted by ferric (< 26 %), manganese, cadmium (< 1,7 %) and rarely also by mercury.

In the heteromorphic modification of sphalerite, wurtzite (β -ZnS) formed up to 1020°C is the most usual replacement cadmium (Koch et al., 1967). However, the wurtzite is not a frequent mineral due to the paramorphic alteration at 1020 °C, though first in the Gyöngyösoroszi area the third natural wurtzite modification (β' -ZnS) was found in dikes as microscopic crystals. This new modification was by Sándor Koch (in 1958) named “mátraít”. The frequent occurrence of heteromorphic modification of sphalerite interprets the incidence of cadmium in the Gyöngyösoroszi mining area.

Mercury occurs mainly in form of drops around the former geysers due to the latest hydrothermal phase at low temperature (at most at 100 °C). In postvolcanic activities the mercury drops can precipitate in the hydroquartzite and on the underlayers of the former thermal spring in the Toka valley and of the geysers mostly around Asztag-kő. Mercury generally has no accompanying elements, therefore its correlation values are low (Nagy et al., 1986).

The arsenic concentration is high both in the area of flotation plant and of the tailing dump, and also extremely high at some sampling sites in the village. On the northern part of the mine arsenopyrite was found together with sulphide minerals in the sediment of Toka creek (Nagy et al., 1986). Presence of arsenopyrite can probably explain the high arsenic concentration in the area. In galena lead can be substituted by

silver, copper and zinc, rarely also by arsenic in form of microscopic inclusions (Koch et al, 1967). The other accompanying ore minerals of sphalerite in the Gyöngyösoroszi area are pyrite (FeS₂) and chalcopyrite (CuFeS₂). Both these minerals contain zinc with copper, silver and gold as inclusions and also arsenic in the crystal lattice. This explanation is shown in correlation values of arsenic, cadmium, lead and zinc.

The data of mining and soil samples around Gyöngyösoroszi support that the As-Cd-Pb-Zn pollution are the most important and the mercury pollution due to the local mercury mineralization in the Toka valley and eastwards of Gyöngyösoroszi is quite small. The Hg concentration values are generally low and only in a single sample at the tailing dump reach 8 times the B value (Hungarian Joint Decree No. 10/2000). The mud of the thermal spring in the Toka valley has extremely high concentration of arsenic, cadmium, mercury and lead: 45, 13, 24 and 33 times higher as the B pollution limit values respectively (Kristó, 2004).

3.2. Human health risk assessment

The risk assessment was focused on the town with vegetable gardens. In the soil of tailing dump there is no agricultural activity, however the gardens in the town are mainly used to grow vegetables and fruits. These are used for local consumption of the families living in the village. The exposure factors were the default values of US EPA (US EPA, 1997a), the BCF values are from RIVM report (1992).

In general, the risk assessment procedure used 70 kg for bodyweight, 70 years for exposure duration and 350 days for exposure frequency. The reference doses were used from Integrated Risk Information System (US EPA, 2006), except for lead, where we used the formula $RfD = PTWI / 7$, where *PTWI* is provisional tolerable weekly intake (mg/kg/week) (JECFA, 1993).

The results of risk assessment in the town are shown in table 3. The summarized hazard index (ΣHI) is 1.16, indicating unacceptable risk. The most hazardous exposure pathway is ingestion of homegrown vegetable. The most hazardous element was arsenic, 86% of the summarized hazard index derived from arsenic contamination.

Table 3. Hazard indices (HI) in the village

ΣHI	Ingestion of soil	Ingestion of vegetable	HI
As	0.2524	0.7447	0.9971
Cd	0.0011	0.0561	0.0572
Hg	0.0018	0.0049	0.0067
Pb	0.0213	0.0389	0.0602
Zn	0.0012	0.0369	0.0381
HI	0.2778	0.8815	1.1593

4. CONCLUSIONS

The environmental research in the territory of Gyöngyösoroszi carried out has documented high concentration values of arsenic and heavy metals in soil. The source

of pollution was the tailing dump and the flotation plant during the mining however the geochemical background values are also higher than the Hungarian pollution limit values. The floods of Toka creek spread the contamination in the flood-plain between the village and the city Gyöngyös, whereas increasing the natural background concentration in soil.

The outcome of the risk assessment has indicated that soil contamination in the Gyöngyösoroszi mining area poses a potential health risk to the inhabitants. In order to reduce the health risk, decreasing or avoiding the consumption of homegrown vegetables is advisable. Determination of the risk assessment of the flooded and non-flooded areas in the village by several separated data is most desirable.

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