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Impact of metalliferous ore mining activity on the environment in Gyongyosoroszi, Hungary

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Abstract

In the area of Gyongyosoroszi mining activity was in progress between 1945 and 1985. A complex survey was carried out to assess the impacts of the mining activity. The survey covered the sampling, analysis and data evaluation of waste material, soils from several parts of the village, water samples from the brooks and lakes, sediments from the surface waters and plants growing in home gardens and in fields. Results of the environmental survey indicated that heavy metal contamination of the examined area is due to mining activity and this creates a high ecological risk; remediation of the contaminated area was designed.

Keywords: Mining; Hungary; Heavy metal contamination; Waste material; Tailing dump; Acid mine discharge

1. Introduction and general survey

Mining, metal industries and transport are the main sources of heavy metal contamination throughout the world. Heavy metal content of rivers, lakes and their sediments in industrial areas is increasing continuously (Baudo, 1989). The soil is also in danger; besides direct contamination by industries and transport, waste-water irrigation and the application of sludge to the land contribute to soil contamination (Fergusson, 1990).

A better understanding of heavy metal bonding and transport in soil (Merrington, 1993), sediment and living organisms is needed to clarify how There are several publications from Canada (Joshi et al., 1989), Australia (Craze, 1980), and the USA (Wai and Mok, 1989; Ragiani, 1977; Landrigan et al., 1975; Ma et al., 1995) as well as from Europe (Haakansson et al., 1989; Leenaers and Rang, 1989) about assessing, analysis and the spreading of such contaminants.

1.1. Short description of the area

The area of Gyongyosoroszi is rich in metalliferous ores such as galena, sphalerite, and to a lesser extent chalcopyrite and pyrite. Mining started in Gyongyosoroszi in 1945 and continued intermittently until 1985. During this period 3 920 089 M.T. (Metric ton) of ores have been ex-

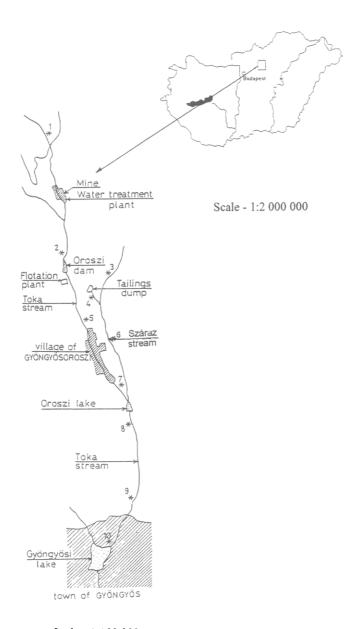
heavy metals enter the food chain (Beveridge, 1989).

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ploited. Although since 1985 the mining has been interrupted, so far the site has not been closed. Fig. 1 shows the area examined with surface waters: Toka, Szaraz.

Approximately 1500-2000 m³ of acid water leaves the mine every day. There are three dams containing the mine effluent. A dam burst in 1988

and 400 000 m³ of acid water flooded the area of Gyongyosoroszi. The mining company used in situ liming to neutralise the flooded water. This treatment resulted in hundreds of m³ of sludge containing large quantities of heavy metals to settle out onto the bed of rivers. The mine effluent is mainly treated in the neutralising plant. The



Scale - 1:100 000

Fig. 1. The study area.

precipitates which originate from the neutralising process are a dangerous contamination source as the storage lagoon is leaking.

The tailing dump is situated northeast of the village of Gyongyosoroszi. The area of the dump is $\sim 33~000~\text{m}^2$. The dump has been built up from the by-products of ore processing by flotation techniques. The waste material has a high water content which is collected by drainpipes and diverted into a collecting pond. The tailing dump has not been covered for several years. In 1988, 20-30 cm of soil was inefficiently dumped onto one side of the tailing dump. This covering is not satisfactory for the establishment of plants. Regarding its prominent colour (yellow, where it is covered, red and grey where it is not), it causes visual intrusion. The town of Gyongyos and the village of Gyongyosoroszi, two rivers (River Toka. River Szaraz) and several lakes are also in the examined area.

1.2. Investigation of the contaminated site Once the hazardous situation was discovered

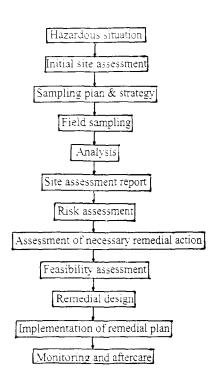


Fig. 2. Decision tree for contaminated site investigation and remediation.

the first step was to carry out an initial site assessment (Fig. 2), which is a vital step where immediate or acute health or environmental risk are suspected. The sampling was planned and carried out according to Hungarian sampling standards (MSZ 21472-85, MI 12739/1-78, MSZ 14475/18-80) (HSO, 1978; 1980; 1985).

Analysis was carried out jointly by the Technical University of Budapest and the Eotvos Lorand University of Science with contributions from the Pharmaceutical Research Institute and HUMIL Ltd. Hundreds of samples originating from the contaminated area were measured using multielemental analytical methods such as x-ray fluorescence spectrometry (XRF) and potentiometric stripping analysis (PSA). The next step was the completion of the site assessment report, which is the main concern of the present paper, to be followed by the risk assessment. We found that remedial action is required; recultivation of the tailing dump and measures for the remediation of surface waters were designed together with some alternative measures for the treatment of contaminated soil.

2. Materials and methods

2.1. Sampling water matrices, sample preparation, chemical analysis

Sampling was carried in mid June, 1991 and tailored for the specific situation in the examined area. Water samples were collected along the streams and at several parts of the lakes. The streams and the lakes were not deeper than 5 m, therefore stratification was not expected. Sampling was carried out at one depth in all cases. The pH of each sample was measured in situ using a site pH-meter (Radelkis OP-110). Samples were filtered through a $0.45-\mu m$ filter paper before preservation in HNO₃ (to pH 2) and then stored in plastic containers, leaving no head-space, until chemical analysis.

Chemical analysis was carried out by potentiometric stripping analysis according to Jagner (1979). The measuring cell was a 6.5-cm high, 3.5-cm wide plastic vessel with a magnetic stirrer. The three-electrode cell consisted of a saturated calomel reference electrode (OP-0830 P, Radelkis

type), a platinum wire counter electrode (OH-8583, Radelkis type) and a glassy carbon working electrode (F 3600, Radiometer type). The working electrode was first polished with 3 µm of diamond paste for 60 s, then cleaned carefully with acetone according to Jagner (1979). All three electrodes were immersed in 0.1 M HCl containing 25 mg Hg(II)/1. The plating potential was adjusted to -0.95 V (SCE) and the plating time to 1 min. This process was repeated 5 times. The precoated electrode was then used for the analysis of 15 samples. When not in use the electrode was stored in distilled water. For measurement, 10 ml of sample was added to 20 ml of blank solution (0.589 g NaCl, 1 ml 0.2 g Hg/l Hg(II), 3 ml cc HCl in 20 ml double distilled water). The blank was used for background correction. Zinc, Cd and Pb was plated at -1.25 V (SCE) according to Jagner (1976). The plating time for Zn was 1 min, while 5 min was applied in the case of Cd and Pb. The heavy metal content of the samples were determined using the standard addition method (Adam, 1983; Jagner, 1979). The detection limit was 10-20 ppb for Cd and Pb, and 100 ppm for Zn. Stock solutions of Hg(II) and solutions of the metals were prepared from analytical grade chemicals and dissolved in double distilled water. The HCl used was Merck Suprapur quality.

2.2. Sampling soil matrices, sample preparation, chemical analysis

One hundred and sixteen soil samples were collected along the streams (diffuse source) radially around the tailing dump (point source) and randomly in the previously flooded area. The banks of the streams were sampled vertically at several sites in order to assess the impact of irrigation and spring floods. The waste material of the tailing dump was sampled as shown in Fig. 3. Since there is a high background concentration of heavy metals in the examined area, reference soil samples were collected from uncontaminated sites (local control site). Stratification of the ground was investigated at three depths.

Sampling was carried out in the following manner: a 10×10 -m grid was identified for all sampling sites and three samples were taken along the diagonal line of the grid at equal distances, 0-0.4

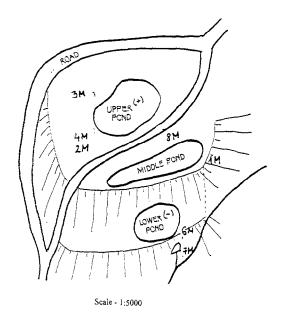


Fig. 3. The tailing dump.

m depth. The three samples were mixed and not less then 1.5 kg of soil was stored in plastic bags until the samples were shipped to the laboratory (Hungarian standard method, MSZ 21472-85) (HSO, 1985). The samples were refrigerated when not in use. For chemical analysis the samples were air-dried, milled and passed through a 2-mm sieve. Gravel, if present, was removed before milling, and quantified. This was taken into consideration when the heavy metal content of the sample was calculated (Hungarian standard method, MSZ 21470/2-81).

Chemical analysis was carried out by XRF spectrometry. One gram of the sample was pressed by 30 000 N and the disc produced (2 cm in diameter) was used for analysis. X-rays were produced by an 125 I isotope (half life, 59.4 days), which emits a 27.4-keV photon (Te K α line). The activity of the 125 I isotope was 410 MBq during analysis. The detector was a semi-conductor Si(Li) Canberra type (SL 30180-7500). Processing of the signals was carried out by a Canberra 2000 Nim Bin module which was fitted with a power supply, spectroscopic amplifier (2020 type) and ADC (8075 type). Data evaluation was carried out by Canberra MICRO AXIL software. To avoid matrix effects

we used calibration curves produced by the standard addition method. The measurement time was 1 h. The detection limit for the medium atomic number (Cu-Pb) was 1-5 ppm (Muia, 1991).

2.3. Sampling biological matrices, sample preparation

Plants were sampled at the site of soil sampling. Both natural (rye grass, great burdock) and manmade (garlic, onion, parsley, carrot, radish, string bean, sorrel, potato, rhubarb, kohlrabi, tomato, plum, red beet, currant, raspberry, gooseberry, melon, grapes, pumpkin, peach, lettuce, apple, pear, strawberry, maize) vegetation were of interest. Reference samples were collected from local control sites. The plants were pulled up by the roots and washed, then dried at 76°C, milled and used for chemical analysis (Hungarian standard method, MSZ 14474/18-80) (HSO, 1980). Chemical analysis was carried out by XRF spectrometry, in the same way as the soil analysis.

3. Results and discussion

3.1. Heavy metal content of the tailing dump

The Gyongyosoroszi tailing dump (Fig. 3) and its surroundings contain several metals. The most important ones are Zn, Pb, Cd, As, Cu, Mn and Co. Zinc is the main component, but because of the high toxicity of the other metals, even at low concentrations, they must be taken into account in the assessment of the contamination. The heavy metal concentration of seven representative samples, and the mean values (x) and standard deviation (S.D.) of heavy metal concentration of the waste material is summarised in Table 1. Accord-

ing to Table 1, the waste material has a very high heavy metal content. From the large deviation of the averages we conclude that the material of the tailing dump is heterogeneous (the larger the S.D./x value the more heterogeneous the sample).

3.2 Investigation of soil samples

Seventy soil samples were investigated. The tailing dump was considered as a point source which may contaminate the surroundings by the action of wind. Fig. 4 summarises our sampling strategy and the measured data around the tailing dump.

According to Fig. 4 the wind does not contribute to a great extent to the contamination of the surroundings of the tailing dump. Only one sample, which originated from a previously flooded area — marked with a cross on Fig. 4 — showed a high heavy metal content.

It is more likely that heavy metal contamination of the soil is the result of the spring floods as well as dam bursting. Therefore, the heavy metal concentration of the soil on moving away from the stream was also measured, and the results are given in Fig. 5 and Table 2.

All the highly contaminated soil samples were near the riverside, which may be explained by spring-floods and irrigation. According to Fig. 5 the concentration of the heavy metals in the soil decreases on moving away from the stream. In low lying areas — even 10 m away from the stream — very high heavy metal content ($c_{Zn} = 3056$ ppm, $c_{As} = 145$ ppm, $c_{Pb} = 950$ ppm) (Gruiz et al., 1993) was measured.

3.3. Contamination of the surface water

Thirty-five water samples were analysed. The

Table 1 Heavy metal content and mean values (x) of the samples from the tailing dump (mg/kg)

Metals	1 M	2 M	3M	4M	6M	7 M	8M	x	S.D.	S.D./x
Cr	0	0	1680	1450	0	270	90	499	738	1.48
Co	212	354	606	0	0	297	175	235	212	1
Cu	516	1070	5940	6140	0	500	1010	2168	2670	1.23
Zn	100	2400	9100	11 300	22 100	100	1900	6714	8090	1.20
As	363	0	0	0	84	0	0	64	136	2.13
Cd	0	0	6	0	66	5	46	18	27	1.54
Pb	889	3960	7041	21 120	53	657	2970	5241	7406	1.41

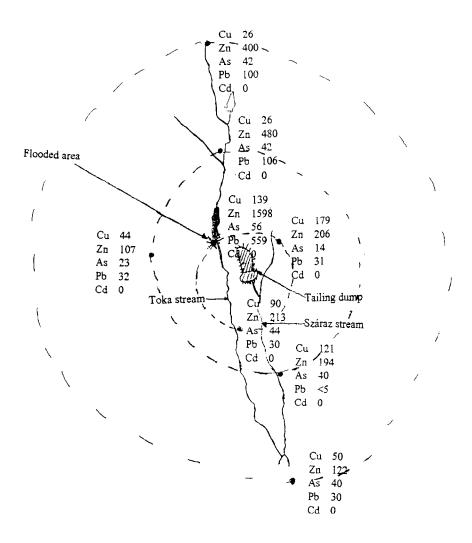


Fig. 4. Soil sampling around the tailing dump (concentrations in mg/kg dry wt.).

most representative results of the water investigation are summarised in Figs. 6 and 7.

The heavy metal concentration in the brooks and lakes were often above the limit set by 3/1984 (II.7.) OVH legislation. In general, the heavy metal content of surface water decreases suddenly after the neutralising plant — between sample 4b and the Oroszi dam (Fig. 1) — which is due to liming.

3.4. Plants

Plants growing in fields as well as in home gardens were investigated. In the case of natural vegetation a comparative study was carried out.

Plants from badly contaminated areas (e.g. on the tailing dump) were compared with those from an uncontaminated site. Two species, rye grass (Lolium perenne) and great burdock (Arctium lappa) were studied. Plants growing on the contaminated areas contained more types of heavy metals in higher concentrations than the control. The heavy metal content of great burdock plants growing on both the tailings and at an uncontaminated site are summarised in Table 3.

Usually, minor components of the tailings, such as La, Sm and Eu, occur in the plant as a finger-print of the contamination.

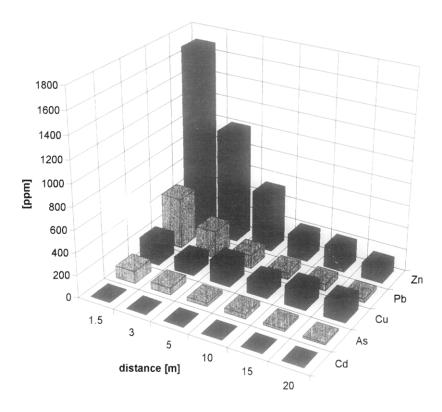


Fig. 5. Heavy metal content (mg/kg dry wt.) plotted against the distance from the stream.

In the case of cultivated plants, 300 samples were measured at the request of local people. Plants which are likely accumulate heavy metals and therefore dangerous for human consumption are given in Table 4.

The cultivation of plants and agricultural activity in the vicinity of Gyongyosoroszi needs careful consideration. Garlic, rhubarb, lettuce, horse radish, garden sorrel and parsnip accumulate heavy

metals to a large extent. On the other hand, grape, raspberry, currant, peach and apple as well as melon and pumpkin are recommended for growing.

4. Sources, fate and impacts of heavy metal contamination in the investigated area

On the basis of the analysis three contamination sources were distinguished, two of which are con-

Table 2
Heavy metal content of soil at various distances from the stream (mg/kg dry soil)

Metals	1.5 m	3 m	5 m	10 m	15 m	20 m	Uncontaminated control
Zn	1685	998	520	208	200	136	107
Pb	462	248	120	63	58	35	32
Cu	210	127	200	131	172	165	44
As	110	63	31	41	23	17	23
Cd	5.1	<1	<1	< 1	<1	<1	<1

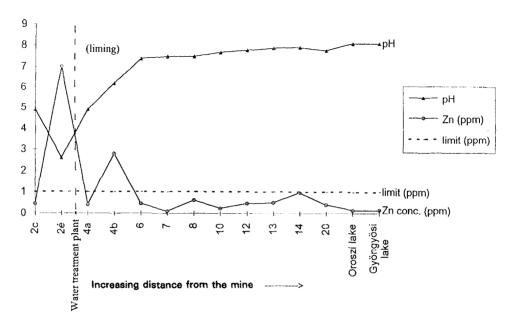


Fig. 6. pH and Zn content along the Toka stream.

sidered to be direct (tailing dump, acid mine discharge) and the third as indirect (contaminated sediment of surface waters) pollution sources.

4.1. Tailing dumps

Tailing dumps have mainly a local impact and

it is most likely that the activity of wind does not contribute significantly to contamination of the surroundings. The higher heavy metal content in close proximity to the tailings is likely due to leaching and surface runoff. The latter is a consequence of an inadequate deposition of the waste

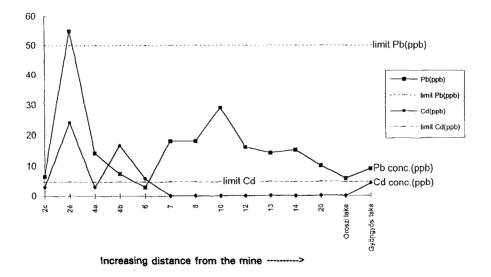


Fig. 7. Cd and Pb content along the Toka stream.

Table 3

Comparison of heavy metal content of great burdock (Arctium lappa) plants growing on the tailings and at an uncontaminated site

Sample	Metals (mg/kg)										
	Fe	Cr	Со	Zn	As	Sc	La	Eu			
Tailings	6100	11.3	4.1	400	4.6	2.5	6.5	0.24			
Uncontaminated soil	170	1	-	38	0.22		0.16				

material (steep slopes, absence of covering). Fig. 8 shows the fate and impact of the tailing dump.

The leachate of the tailing dump may contaminate the soil and groundwater. Surface runoff increases the heavy metal content of top soil, surface water, and indirectly, the tissues of plants. These above mentioned factors lead to the contamination of water courses, as well as an increase in sedimentation. The soil particles may adsorb considerable amounts of heavy metals, depending on their adsorption capacity. Heavy metals may be accumulated in plants growing in the examined area, and may enter the food-chain. This has an extreme impact since this area is used for agricultural activity.

Some soils and sediments have a large capacity to store and immobilise heavy metals, thus the direct effects may not be observed for a long period of time. This does not mean, however, that the chemicals will be stored there forever. Saturation or changing conditions influencing the storage capacity can lead to sudden and often unexpected mobilisation of heavy metals from soil and sediment. This effect is called the 'chemical time bomb', which is described by Stigliani (1988)

4.2. Acid mine discharge

Acid mine discharge has a complex influence on the environment (Fig. 9). Unfortunately, acid mine discharge is just partly neutralised and the rest of this acid water, with a high heavy metal content, directly enters the Toka stream. Due to the alkaline outflow of the neutralising plant, the heavy metals precipitate out, which increases the turbidity of the water and leads to sedimentation. In addition, large quantities of heavy metals are present in the sediment of the surface water as a result of past dam-bursting.

The sediment of surface waters therefore contains high concentrations of heavy metals which may dissolve at any time if the environmental conditions change (i.e. chemical time bomb).

Since pH plays such an essential role in the immobilisation of heavy metals from sediment, a trial was carried out to determine the amount of dissolved metals from the sediment of the lakes in relation to decreases in pH. The results for two metals in two seperate samples are summarised in Fig. 10.

Decreases in pH resulted in an increased concentration of dissolved heavy metals. This shows

Table 4
Accumulation of heavy metals in some cultivated plants (8/1985 (X.21) EuM Departmental Order, Hungary)

Metal	Plants (mg/kg)								
	Rhubarb	Onion	Parsley	Carrot	Garlic				
Zn	168	110	83	41	47				
Cu	101	53	80	54	48	10			
As	0	7	< 0.5	< 0.5	< 0.5	0.5			
Cd	0.3	4.9	10.3	< 0.03	0.2	0.03			

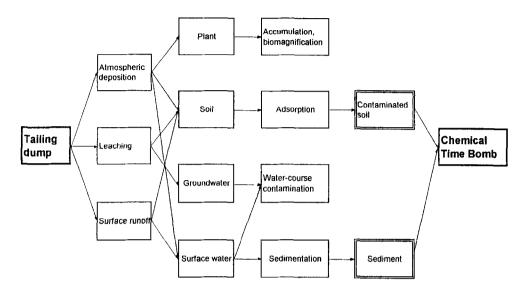


Fig. 8. Impact of the tailing dump.

the danger of the contaminated sediments, since they may leach continuously, especially if the pH decreases (e.g. ceasing the activity of the neutralising plant).

Fig. 9 also shows that besides the direct biota impact, an indirect effect may occur due to irrigation and spring floods which contaminate the soil along the streams.

5. Remediation of the contaminated area

The remediation plan for Gyongyosoroszi is tailored to the specific problems of each contamination source.

The waste material of the tailing dump has a very high heavy metal content and neither the stability nor the nutrient and water supply is ade-

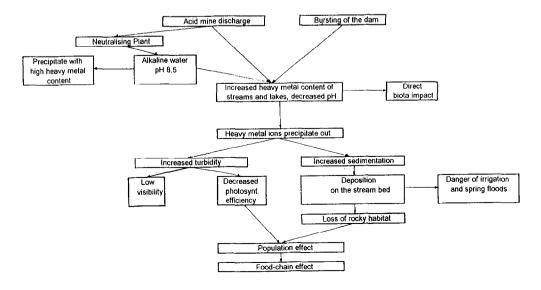


Fig. 9. Impact of the acid mine discharge.

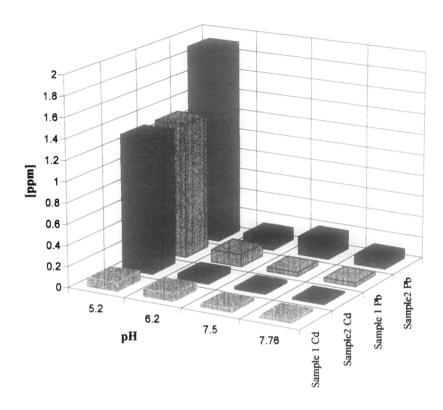


Fig. 10. Heavy metal leaching as a function of pH.

quate for plant establishment. Moreover, due to the problem of stability (very fine particles) erosion may occur. Part of the problem is due to the topography, since erosion is greatest on steep slopes. The tailing dump should be recultivated as follows. To avoid erosion, the dump has to be reshaped, and this has to involve the following steps: reduction in the height of the dump; construction of slopes with grades no steeper than 1 in 3 and no flatter than 1 in 20 (Ritcey, 1989) in order to minimise scouring, to facilitate vegetation and to provide surface and internal draining. This step is followed by chemical stabilisation of the dump by applying lime. The dump should then be isolated by a waterproof layer, such as clay, and covered by a drainage layer (sand and gravel) and finally by top soil to provide good conditions for re-vegetation.

In the case of acid mine discharge the short term solution is the continuous neutralisation of the water, which keeps the pH alkaline, and thus precipitates the heavy metal content of the mine effluent, and prevents immobilisation of heavy metals from the deposited sediment. Additional and continuous problems arise from the storage of the increasing amount of precipitate. This may be eliminated by the long-term solution, which is the complete closure of the abandoned mine. The water of the springs currently entering the mine should be diverted directly into the surface waters to prevent its contamination in the mine.

The surface water and its sediment is also hazardous for the environment. The short-term solution to maintain the present state requires continuous neutralisation. In this case, however, irrigation should be avoided, and floods should be controlled. The long-term solution is the dredging and safe disposal of the dredged sediment, followed by remediation of the bed. An alternative may be the use of geotextiles, which could prevent the immobilisation of heavy metals from the sediment.

In the area examined three alternatives were

considered in the case of contaminated soil. Firstly, if soils contain large quantities of heavy metals and this contamination occurs in relatively small areas, soil change is recommended. The removed soil should be deposited to hazardous waste disposal sites. If the contaminated area is large, it is impossible to remove the soil and therefore two alternatives are considered. In the case of high heavy metal content the area should be set aside. The area may be used for purposes other than agriculture. Forestation, however, should be avoided because of the significant risk in converting agricultural land to forest land, as it may result in a decrease of pH and an increase in the mobility of trace metals in the top soil (Kauppi, 1990).

If the heavy metal contamination is moderate, agricultural activity may be continued, but it is advocated to avoid plants which accumulate heavy metals. Liming should be considered.

The third alternative for soil treatment is to grow plants which are likely to accumulate heavy metals, and then to dispose of the harvested plants circumspectly.

6. Conclusion

The environmental survey, carried out in the vicinity of the Gyongyosoroszi tailing dump shows, that the contamination of surface waters, soils, sediments and plants is due to the mining and mine waste disposal activity.

On moving away from the tailing dump and acidic mine discharge the heavy metal content of the water and soil decreases. The sediment of surface waters contaminates the surroundings at spring floods and during irrigation, and results in a different spatial distribution of contaminants. High heavy metal content was observed in low lying areas, where spring floods frequently occur. There is also a significant impact from the lagoon of the neutralising plant, which stores large amounts of precipitates and is not adequately isolated. The agricultural activity in the vicinity of Gyongyosoroszi requires careful consideration. Recultivation of the tailings and the remediation of surface water and soil are recommended (Horvath et al., 1991).

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