

Site-specific risk assessment in contaminated vegetable gardens

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Abstract

A field survey was carried on in Gyöngyösoroszi, Hungary, near to an abandoned lead/zinc mine to analyse the metal contamination of flooded and non-flooded vegetable gardens, and to evaluate the health risks to local population. Contamination levels of arsenic, cadmium, lead, mercury and zinc were measured in soil and homegrown vegetable samples and bioconcentration factors and hazard indices were calculated. The high metal contents of flooded vegetable gardens were caused by floods, the results indicated significant differences between flooded and non-flooded vegetable gardens. The most accumulating vegetable was sorrel, the most mobile elements were cadmium and lead. Arsenic was not available for vegetables. The health risk was calculated for two exposure routes: ingestion of soil and ingestion of vegetables. The site-specific exposure parameters were established after a population based survey and a special equation was created to calculate the health risk due to homegrown vegetable consumption. The highest risk was associated with ingestion of vegetables, the most hazardous element being lead. The hazard index did not exceed the threshold value of one in flooded or non-flooded gardens. The analyses of health risk indicated that despite the high metal concentrations of soil the contamination of vegetable gardens does not pose an unacceptable risk to the inhabitants of the village.

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1. Introduction

Mining and industrial processing are the main sources of heavy metal contamination in soil (Jung, 2001; Lee et al., 2001; Granero and Domingo, 2002; Kim et al., 2005; Lee et al., 2005). These metals may accumulate to a toxic concentration level which can lead to impairment in the quality of human life (Alam et al., 2003; Wang et al., 2005). Sites contaminated with arsenic and heavy metals have been ongoing problems in terms of exposure assessment. The aim of this procedure is to estimate the concentration to

which human populations may be exposed (Morra et al., 2006). As environmental measurements and modeling do not take into account differences in exposure routes, the human risk may be under- or overestimated (Nieuwenhuisen et al., 2006). The site-specific exposure assessment can be more realistic, and the exposure parameters obtained by questionnaire. Questionnaires typically provide qualitative information (WHO, 2000) and expand upon the data from site-specific risk assessments.

Consumption of vegetables is one of the most important pathways by which heavy metals enter the food chain (Cui et al., 2004; Wang et al., 2004). For some individuals, gardening is adopted as a way to provide fresh produce and save on food costs (Finster et al., 2004). Traditionally, the agricultural community has given little attention to the potential health effects of contaminated soil (Chaney et al., 1984).

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The area of our study is the region of an abandoned lead–zinc mine in Gyöngyösoroszi, a village in North-Eastern Hungary (47.826°N, 19.894°E). The Toka creek crosses the village and collects the runoff of the abandoned metal site. In the Gyöngyösoroszi area the most frequent ore minerals are galenite (PbS), sphalerite (ZnS), pyrite (cubic FeS₂), marcasite (rhombic FeS₂) and chalcopyrite (CuFeS₂) (Vető, 1988); the mining activities were based on these minerals. Mining was active between 1954 and 1985, but closure of the mine has not yet occurred. The ore dressing processes were carried on in the flotation plant next to the village and, in total, 3 millions tons of tailing material were transported to the tailing dump (Kovács et al., 2006). The sediment of the creek contains the *in situ* neutralized mine outflow and the sludge of the flotation technology containing elevated levels of heavy metals (Horvath and Gruiz, 1996). The creek floods the floodplain twice a year and contaminates the surrounding vegetable gardens (Ódor et al., 1998).

The main objects of this study are to analyse the metal content of soil and homegrown vegetables, to develop a more reasonable method for health risk assessment in metal contaminated sites, and to determine the differences in soil pollution and human health risk between the flooded and non-flooded vegetable gardens in the village.

2. Materials and methods

2.1. Sampling and laboratory analyses

Sampling of soils and vegetables was carried out at 44 sampling sites in July 2002 from flooded and non-flooded vegetable gardens in the village Gyöngyösoroszi. The flooded gardens are near to the bank of Toka creek; the non-flooded gardens are 100–400 m from the creek. The sampled locations are marked in Fig. 1. At each sampling site duplicate samples were collected by random sampling method. Vegetables and their rooted soil samples (at 0–20 cm in depth) were taken and packed into polyethylene bags.

Soil samples were dried at 30 °C until constant weight and sieved through 2-mm nylon mesh. Soil pH was determined in a soil:water suspension. Ten grams of dried soil and 25 ml of distilled water were mixed and taken in shaker for 30 min. Values of pH were measured using a glass electrode and pH analyser (Radelkis OP-300).

Vegetable samples were taken from flooded and non-flooded vegetable gardens in the village according to the type of harvest. Six vegetable species were selected for this study; these were representative of species consumed in the studied area: tomato (*Lycopersicon lycopersicum*), squash (*Cucurbita pepo*), bean (*Phaseolus vulgaris*), onion (*Allium cepa*), carrot (*Daucus carota* ssp. *sativus*) and sorrel (*Rumex rugosus*). Good quality vegetable samples were selected using a random sampling procedure, packed into polyethylene bags and transported to the laboratory. The sampling amount of vegetables were the fol-

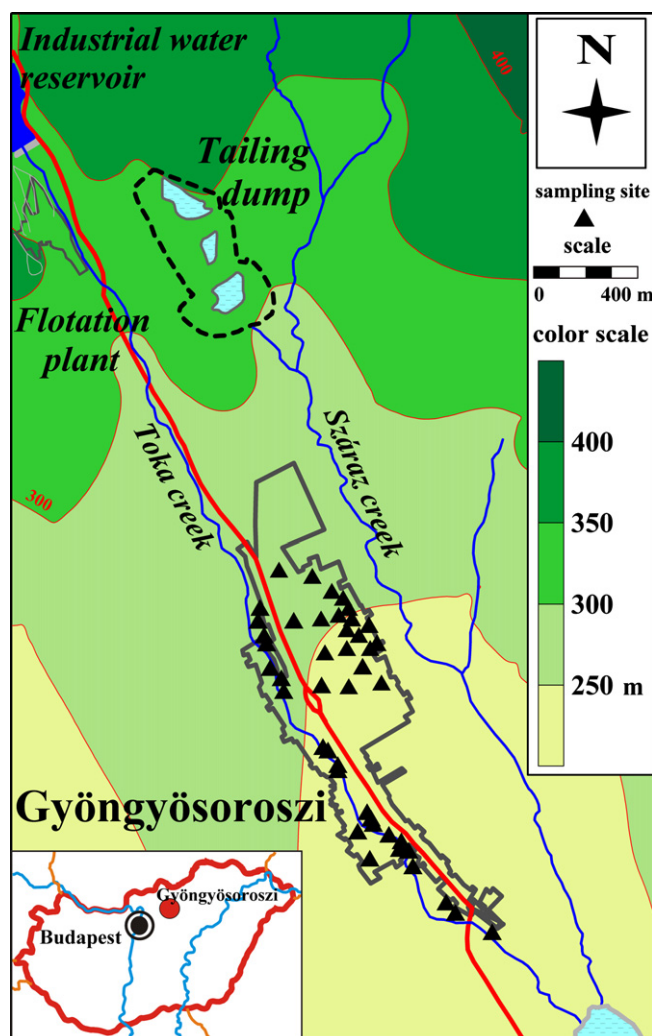


Fig. 1. Map of the studied area.

lowing 0.3–0.5 kg tomato, 1–2 kg squash, 0.2–0.3 kg bean, 0.4–0.6 kg onion, 0.3–0.5 kg carrot and 0.1–0.2 kg sorrel. The samples were washed with distilled water three times to eliminate the air-borne pollutants and soil particles and then blotted dry with tissue paper. Only the edible part of each vegetable was used for analytical purposes; the non-edible parts were removed using a plastic knife. The cook-ready vegetables were weighed, dried at 30 °C until constant weight, weighed again to determine water content and then ground using a ceramic-coated grinder.

One gram of dried soil and vegetable samples were treated with 4 ml of *aqua regia* (HNO₃:HCl = 1:3) in hermetic Teflon bombs and placed in a microwave digester (Milestone 1200 MEGA) under a power-controlled program. Solutions were filtered and made up to 25 ml with deionized water. The arsenic, cadmium, mercury, lead and zinc contents were measured by inductively coupled-mass spectrometry (ICP-MS) (Agilent HP4500 Plus). The limits of detection (LOD) for soils and vegetables were the following: 0.005 mg kg⁻¹ for Cd, Pb and Zn; 0.01 mg kg⁻¹ for

Hg; and 0.05 mg kg⁻¹ for As. The accuracy of the methodology was checked by determining the levels in duplicate samples as well as those of blanks. For quality control standard reference materials (GBW07404 and LGC6138) were used. The recovery rates ranged from 91% to 110%.

2.2. Questionnaires and statistical analyses

A standardized questionnaire was constructed to set the site-specific parameters for risk assessment. The door-to-door survey method was operated with open response and multiple choices. The age, gender, body weight, family members and the average yield of the home-produced vegetables were asked. The survey included 67 vegetable gardens in the village and 90 participants (43 males and 47 females).

The median concentrations and median absolute deviation were calculated and compared to their Hungarian pollution limit value. In analysing the differences among the two territory types the Mann–Whitney test was used for the statistical analysis taking $p < 0.05$ as significant. The statistical parameters were prepared using Statistica 6.0 (StatSoft, Tulsa, OK 2001).

2.3. Risk assessment

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 inhalation and dermal exposure routes were neglected in accordance with other studies (Granero and Domingo, 2002; Hough et al., 2004; Nadal et al., 2004; Grasmück and Sholz, 2005; Hellström et al., 2007).

The average daily dose was calculated using the following equations in the two selected exposure routes.

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

$$(2) \text{ Ingestion of vegetables: } ADD = \frac{\sum (C_{\text{veg}} \times Y_{\text{veg}}) \times ED}{BW \times AT}$$

C_S is concentration in soil (mg kg⁻¹), C_{veg} is concentration in vegetable (mg kg⁻¹ fresh weight), Y_{veg} is average yield of the selected vegetable in the garden (kg year⁻¹ garden⁻¹), n is average family size (–), IR_{soil} is ingestion rate of soil (kg day⁻¹), EF is exposure frequency (days year⁻¹), ED is exposure duration (years), BW is body weight (kg), AT is average time (days).

The average daily dose of soil ingestion was calculated separately for men, women and children, because of differences in bodyweight and ingestion rate soil. The average daily dose of vegetable ingestion for men and women was calculated separately, because of differences in bodyweight. All of the parameters applied in the calculations were site-specific and derived from a population survey, except ingestion rate soil, which was 16.7 mg day⁻¹ for children and 10 mg day⁻¹ for adults (Wcislo et al., 2002).

The non-carcinogenic risk was characterized using a hazard quotient (HQ), which is the ratio of the average daily dose (ADD) to the reference dose (RfD). The applied reference doses were the following: 0.0003 mg kg⁻¹ day⁻¹ for As; 0.001 mg kg⁻¹ day⁻¹ for Cd; 0.003 mg kg⁻¹ day⁻¹ for Hg; 0.035 mg kg⁻¹ day⁻¹ for Pb; and 0.3 mg kg⁻¹ day⁻¹ for Zn. Reference doses were obtained from Integrated Risk Information System (US EPA, 2006), with the exception of lead, in which we used the formula RfD = PTWI/7, where PTWI is provisional tolerable weekly intake (mg/kg/week) (JECFA, 1993). If HQ is bigger than 1, then 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 route and metal (Paustenbach, 2002).

3. Results and discussion

3.1. Arsenic and heavy metal content of soil and vegetable samples

The results obtained for the median, median absolute deviation (MAD), minimum and maximum for soil samples are presented in Table 1. The measured concentrations are the ‘pseudo-total metal contents’, which accounts for the *aqua regia* digestion not completely destroying silicates (Manta et al., 2002). This method is widely used in environmental geochemistry studies and recommended by the Hungarian Standards Institution. The median concentrations were compared to the Hungarian pollution limit value. The arsenic and metal content of flooded vegetable gardens exceeded these thresholds, with the exception of lead, whereas the contents of non-flooded vegetable gardens were, bar arsenic, under these thresholds. High arsenic concentrations (28 mg kg⁻¹) were also found in lower layers, and are thought to represent the natural background (Ódor et al., 1998).

The flooded and non-flooded vegetable gardens show highly significant differences ($p < 0.0005$ for As, $p < 0.00005$ for Cd, Hg and Zn, and $p < 0.00001$ for Pb). The median pH value of flooded vegetable gardens was 6.47, that of non-flooded vegetable gardens 6.11; the difference was not significant.

The differences in maximum concentrations are highest in the case of cadmium (19 times) and lead (17 times), and lowest in the case of arsenic (3.8 times). These could confirm that the origin of the high Cd and Pb content of flooded vegetable gardens is the floods of Toka creek.

The results of vegetable analyses are shown in Table 2. The arsenic content of vegetables was under the limit of detection in every case. The mercury content of vegetables grown in non-flooded vegetable gardens was also under the detection limit. The measured heavy metal levels were higher in vegetables grown in flooded vegetable gardens in every case, but it was significant ($p < 0.05$) in the Cd content of tomato, the Pb content of tomato, squash, onion and sorrel, and the Zn content of tomato, bean and squash.

Table 1
Arsenic and heavy metal content of soil samples (mg kg⁻¹)

	Flooded vegetable gardens					Non-flooded vegetable gardens				
	As	Cd	Hg	Pb	Zn	As	Cd	Hg	Pb	Zn
Median	46.6	1.31	0.81	85.2	366	31.4	0.43	0.30	27.8	142
MAD	9.9	0.50	0.26	37.8	98	3.0	0.12	0.06	6.4	28
Minimum	24.4	0.33	0.35	29.2	120	23.8	0.22	0.20	20.5	97
Maximum	142.0	13.60	5.40	694.0	2050	37.4	0.70	0.55	40.1	225
Limit value _{POLL}	15.0	1.00	0.50	100.0	200	15.0	1.00	0.50	100.0	200

MAD: median absolute deviation.

Limit value_{POLL}: Hungarian pollution limit value.

Table 2
Arsenic and heavy metal content of vegetable samples (mg kg⁻¹)

	Tomato		Squash		Bean		Onion		Carrot		Sorrel	
	NF	F	NF	F	NF	F	NF	F	NF	F	NF	F
As	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
Cd	0.008 ^a	0.060 ^a	0.005	0.033	0.010	0.020	0.056	0.070	0.068	0.130	0.101	0.115
Hg	<LOD	0.010	<LOD	0.037	<LOD	0.030	<LOD	0.020	<LOD	0.020	<LOD	0.060
Pb	0.083 ^a	0.480 ^a	0.079 ^a	0.673 ^a	0.141	0.260	0.130 ^a	1.060 ^a	0.278	0.810	^a 0.295	^a 0.990
Zn	3.16 ^a	18.00 ^a	1.41 ^a	30.90 ^a	7.70 ^a	30.50 ^a	17.20	42.20	11.80	27.60	29.30	60.50

NF: non-flooded vegetable gardens.

F: flooded vegetable gardens.

LOD: limit of detection.

^a $p < 0.05$.

We have not found any significant differences in the case of carrot. The highest concentration of Cd was found in carrot, the highest concentration of Hg and Zn found in sorrel, and the highest Pb content found in onion.

Despite of the arsenic concentration being high in the soil, in the vegetables it was under the limit of detection. In contrast with our results, other studies found elevated levels of arsenic in vegetables and crops cultivated in metal-contaminated soils (Queirolo et al., 2000; Lee and Chon, 2003; Rapant et al., 2006). The different mineralogy form and the presence of bioleaching microbes could explain the various phyto- and bioavailability of arsenic (Ruby et al., 1999; Mahimairaja et al., 2005).

In our study the most mobile metals were cadmium and zinc. Our results agree with Ullrich et al., who found that the potential phytoavailability of Pb, Zn and Cd in the top-soils between pH 5 and 7 declines in the order Cd, Zn and Pb (Ullrich et al., 1999).

The bioconcentration factor (BCF) was calculated from the ratio of the metal concentration of the vegetable (fresh weight) and the metal concentration of the soil. Fig. 2 illustrates the bioconcentration factors of different species in flooded and non-flooded vegetable gardens. The order of bioconcentration capacity of metals was Cd > Zn ≫ Pb and this strongly agrees with the literature data (McBride, 2003; Liu et al., 2005). Sorrel was the most contaminated vegetable while the lowest concentration was measured in bean. Other studies found spinach, which is very similar to sorrel, had the highest bioconcentration capacity among vegetables (Mattina et al., 2003) and bean had one of the

lowest (Liu et al., 2005). Generally, the BCFs are lower in flooded vegetable gardens, this is in agreement with BCF values decreasing as the metal concentration in soil increasing (Alam et al., 2003). In case of low accumulating vegetables (tomato, squash and bean) this effect was not predominating.

3.2. Human health risk assessment

Table 3 illustrates the site-specific exposure parameters, which were derived from the questionnaire survey method. In the village of Gyöngyösoroszi the migration of inhabitants was extremely low, thus the exposure duration was the same as a lifetime. Holidays amounted to only 1 week per year, therefore the exposure frequency was 358 days. The bodyweight of women was significantly higher than the average bodyweight of Hungarian women (67.21 kg), but the bodyweight of men was nearly the same (80.84 kg) (Melles, 2004). Location-specific human data were also necessary to calculate actual human exposure through ingestion of vegetables (Albering et al., 1999). Home gardening is a traditional family food production system in the village of Gyöngyösoroszi. Home-produced vegetables are for family consumption and not for commercial purposes, therefore in the calculation formula the yield of vegetables was divided by the average family size in the village. The yearly consumption of vegetables per capita was the following: 7.68 kg tomato, 6.21 kg squash, 2.74 kg bean, 2.44 kg onion, 2.15 kg carrot and 1.32 kg sorrel. These results are generally lower than Santos et al. found in

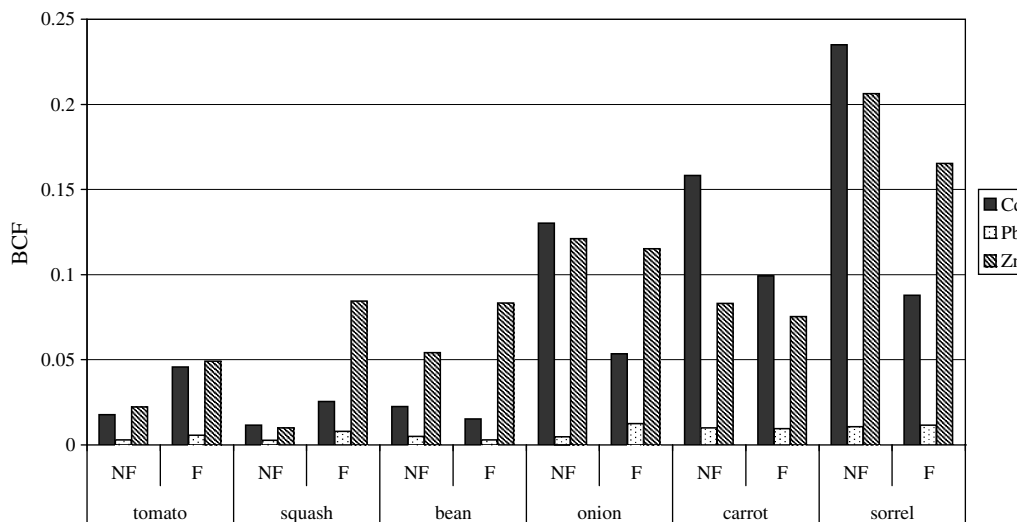


Fig. 2. Bioconcentration factors (BCF) of homegrown vegetables in flooded (F) and non-flooded (NF) vegetable gardens. $BCF = (c_{\text{metal vegetable}})/(c_{\text{metal soil}})$.

Table 3
Site-specific exposure parameters

Parameter	Value
Yield _{tomato} (kg year ⁻¹ garden ⁻¹)	21.56
Yield _{squash} (kg year ⁻¹ garden ⁻¹)	17.43
Yield _{bean} (kg year ⁻¹ garden ⁻¹)	7.68
Yield _{onion} (kg year ⁻¹ garden ⁻¹)	6.85
Yield _{carrot} (kg year ⁻¹ garden ⁻¹)	6.04
Yield _{sorrel} (kg year ⁻¹ garden ⁻¹)	3.70
Family size (garden ⁻¹)	2.8
Exposure frequency (days)	358
Bodyweight _{men} (kg)	80.07
Bodyweight _{women} (kg)	73.66

Rio de Janeiro (5.4 kg tomato, 4.1 kg onion and 3.7 kg carrot) (Santos et al., 2004) or that is given in the US EPA Exposure Factor Handbook (12.57 kg tomato, 5.1 kg bean, 4.42 kg carrot, 2.7 kg onion) (US EPA, 1997), because home gardening partly covers the vegetable requirements of families. The daily consumption of home-produced vegetables was 173.3 g day⁻¹, which is higher than found in Bangladesh (130 g day⁻¹) (Alam et al., 2003) or in China (105 g day⁻¹) (Liu et al., 2005).

The results of human health risk assessment are shown in Table 4. The summarized hazard index of flooded vegetable gardens is 0.394 and of non-flooded vegetable gardens is 0.092, which is an indicated acceptable risk. The largest contribution to hazard index was from Pb (about 35%) both in the flooded and non-flooded vegetable gardens. In all cases most hazard index was attributable to ingestion of home-produced vegetables (85% in the case of flooded vegetable gardens and 72% in the case of non-flooded vegetable gardens). The contribution to the hazard index from soil ingestion was largest for children in flooded vegetable gardens and 90% of the risk is derived from arsenic. The distribution of risk due to different exposure routes is similar to that found by Hough et al. in the United Kingdom (Hough et al., 2004). The elevated arsenic level of

Table 4
Hazard indices of flooded and non-flooded vegetable gardens

	Ingestion of soil	Ingestion of vegetable	Hazard index
<i>Flooded vegetable gardens</i>			
As	0.0514	–	0.0514
Cd	0.0002	0.0473	0.0475
Hg	0.0004	0.0664	0.0668
Pb	0.0053	0.1450	0.1503
Zn	0.0003	0.0780	0.0783
Hazard index	0.0576	0.3367	0.3943
<i>Non-flooded vegetable gardens</i>			
As	0.0235	–	0.0235
Cd	0.0001	0.0190	0.0191
Hg	0.0002	–	0.0002
Pb	0.0017	0.0287	0.0304
Zn	0.0001	0.0191	0.0192
Hazard index	0.0256	0.0668	0.0924

soil indicates the importance of soil ingestion pathway, especially for children (Rieuwertts et al., 2006). In order to reduce the uncertainty in estimating the risk associated with incidental ingestion of soil, further testing is necessary because the *in vivo* arsenic availability can vary widely (Juhász et al., 2007). Our results, as do other studies (Hellström et al., 2007; Huang et al., 2007), also emphasize that the consumption of locally grown vegetables is an important exposure pathway in metal contaminated sites. Despite the high concentration levels in soil, the summarized hazard index was low in flooded vegetable gardens. In our study the risk assessment was determined by special calculation of ingestion of vegetables and site-specific exposure parameters. Of special note is that the calculation using the concentration of vegetables could reproduce the real phytoavailability of metals in contaminated sites. The usage of yield of homegrown vegetables also refines the calculation because consumption of vegetables with higher BCF was much lower than with lower BCF.

4. Conclusion

The environmental research carried out in the village of Gyöngyösoroszi has documented increased concentrations of arsenic and heavy metals in the soil of vegetable gardens. The differences between the metal contents of flooded and non-flooded vegetable gardens were significant, which confirm that the floods of Toka creek spread the contamination over the flood-plain of the village. The source of pollution is the tailing dump and the flotation plant.

The metal contents of vegetables were low and the arsenic content was under the detection limit in every case. The sorrel cultivated in flooded gardens has the highest metal content. Significant differences were also found between the flooded and non-flooded vegetable gardens. The vegetable concentration results further support the view that soil metals are not always absorbed as well as soluble forms, therefore use of default bioconcentration factors in assessing human health risk may overestimate the hazard. In our study, all of the bioconcentration factors were under 0.25 and the mobile elements were cadmium and zinc. Generally, the bioconcentration factors of non-flooded vegetable gardens were higher; the sorrel was the most accumulating vegetable.

Site-specific exposure parameters and a newly created equation for ingestion of vegetables were applied in risk assessment process. The site-specific exposure factors were generated from the results of questionnaire survey in the village, while the equation was based on the cultivation habits of homegrown vegetables.

The outcome of risk assessment has indicated acceptable risk in the village of Gyöngyösoroszi, both in flooded and non-flooded vegetable gardens. In contrast with previous study in the area, home gardening does not increase the risk for inhabitants at present. The most relevant exposure route was ingestion of homegrown vegetables. It is possible to further reduce the risk of human exposure to soil metal contamination by selecting leafy vegetables such as sorrel in home gardening.

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