Low-cost agricultural measures to reduce heavy metal transfer into the food chain – a review

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ABSTRACT

Heavy metal contamination affects large areas of Europe and worldwide. Hot spots of pollution are located close to industrial sites, around large cities and in the vicinity of mining and smelting plants. Agriculture in these areas faces major problems due to heavy metal transfer into crops and subsequently into the food chain. This paper gives an overview on simple but effective countermeasures to reduce the transfer of heavy metals to edible parts of crops. Since crop species and varieties largely differ in their heavy metal uptake, choosing plants with low transfer factors (e.g., legumes, cereals) may reduce metal concentration in edible parts significantly. Cultivating crops with higher heavy metal uptake capacity, e.g., spinach or lettuce should be avoided. The application of soil amendments is another very effective measure to reduce the concentration of heavy metals in crops. Both organic (e.g., farmyard manure) and inorganic amendments (e.g., lime, zeolites, and iron oxides) were found to decrease the metal accumulation. Further effective methods to reduce metal transfer into food chain include crop rotation and cultivation of industrial or bio-energy crops. It is concluded that the methods presented here comprise several tools, which are easy to apply, and are effective to allow safe agriculture on moderately contaminated soils.

Keywords: heavy metals; soil contamination; agricultural soils; low metal uptake crops; soil amendments

As a consequence of industrialization during the last centuries, the heavy metal concentration of soils has increased worldwide (Adriano 2001). Hot spots of soil contamination are located in areas of large industrial activities, where surrounding agricultural areas are affected by atmospheric deposition of heavy metals. Also, agricultural practice, e.g. application of sewage sludge or phosphate fertilisers, has lead to increased metal concentration in soils.

The extent of the metal contamination in agricultural areas is demonstrated by the following examples: In Bulgaria, a survey has revealed that 19 360 ha are contaminated by heavy metals and 1913 ha are polluted by radionuclides. In Poland, 0.5% of the total country area is contaminated by heavy metals and/or other pollutants. In France, up to 800 000 sites are estimated to be potentially contaminated, whereas for the whole of Western Europe this figure may be up to 1 200 000 (NATO/CCMS 2002).

Metal contamination of soils may also be derived from geogenic sources. E.g., in Saxony (Germany) close to Chemnitz and Dresden, large areas are affected by elevated As, Cd and Pb concentrations (Sächsische Landesanstalt für Landwirtschaft 2003). In Cornwall and Devon (U.K.) up to 700 km^2 show As contamination (Mitchell and Barr 1995).

On sites with low or medium contamination levels, metal concentration in crops is mostly not as high to cause acute toxicity, but in the long run it may provoke chronic damage to health (Adriano 2001). Due to the heavy metal burden in human nutrition, there is a need for measures to reduce the metal transfer into agricultural plants. In areas, where conventional or other remediation technologies are not feasible or too expensive, other simple but effective approaches may help to reduce the accumulation of heavy metals in the edible part of crops. This review should highlight some of these measures and critically point out their effectiveness as well as possible drawbacks.

Table 1. Relative accumulation of heavy metals in edible plant parts of different crops (Kloke et al. 1984)

Relative accumulation level			
high	medium	low	very low
lettuce	red beet	maize	bean
spinach	radish	brussels sprouts	pea
mangold	cabbage	white cauliflower	melon
endive	potato	broccoli	tomato
cress	turnip	celery	pepper
carrot	kale	berries	drupe fruit

Selection of crops with low heavy metal uptake

Genotypic differences in plant uptake of mineral elements are well documented (Kloke et al. 1984). An overview on the general metal accumulation behaviour of the selected crop species is given in Table 1.

The transfer factor soil-plant, expressed as the ratio of plant concentration divided by the total concentration in soil, may be an indicator of the plant accumulation behaviour (Kabata-Pendias and Pendias 1992). Lübben (1993) showed large differences in the transfer of Cd, Zn, Ni, Cu, Pb and Cr from soil to different plant parts. The lowest transfer factors of Cd were found for grains of maize, peas, oats and wheat, whereas the highest values were reported for leaves of spinach and lettuce and the roots of various plants. Low transfer factors of Zn were reported for carrot and grains of maize and pea, whereas the highest were found for leaves of spinach and roots of radish and other plants. The transfer factors in the investigated plants were reported to be similar also for Cu, Cr and Pb.

The transfer factors in soil to plant of Cd, Cu, Ni, Pb and Zn for a range of plant species and plant parts are given in Table 2. This data indicates, that there is a large difference in the uptake of Cd, whereas differences in the uptake of Cu, Ni and Pb were less pronounced. Among the crops, spinach had generally very high transfer factors, indicating that cultivation of this species, along with other leaf vegetables like

Table 2. Transfer factors [total concentration in plant (mg/kg)/total concentration in soil (mg/kg)] of heavy metals from soil to different plant tissues (Machelett et al. 1993)

Plant species	Tissue	Cd	Cu	Ni	Pb	Zn
Fodder beet	leaves	5.55	0.25	0.52	0.07	6.04
Spinach	leaves	5.00	0.22	0.25	0.13	1.27
Celery	leaves	2.82	0.15	0.15	0.04	1.22
Celery	root	2.09	0.32	0.18	0.04	0.74
Lucerne	shoot	1.73	0.18	0.60	0.02	1.66
Maize	straw	1.09	0.10	0.06	0.09	1.53
Fodder beet	storage roots	0.84	0.23	0.28	0.02	1.18
Radish	tuber	0.50	0.10	0.15	0.02	0.72
Onion	tuber	0.47	0.06	0.09	0.01	0.54
Tomato	fruit	0.38	0.18	0.15	0.03	0.21
Potato	tuber	0.33	0.18	0.14	0.05	0.21
Maize	cob	0.30	0.11	0.15	0.01	0.68
Winter rye	grain	0.16	0.12	0.11	0.01	0.61
Bean	seeds	0.08	0.14	0.28	0.04	0.25

lettuce, should be avoided on contaminated soils. Heavy metal excluding crops like beans, potatoes or cereals should be preferred.

Due to the high toxicity and bioavailability of Cd, a number of studies focused on the accumulation of this particular metal. It has been shown that crop selection is effective to reduce the transfer into human food chain. The Cd accumulation by crop species decreases in the following order: leaf vegetables > root vegetables > grain crops (Bingham 1979, John 1973, Page et al. 1987). Compared to other plants, high levels of Cd have been found for spinach and lettuce (Kloke et al. 1984, Tlustoš et al. 1998) as well as for tobacco and celery (Isermann et al. 1983), whereas low Cd concentrations were observed in beans (Tlustoš et al. 1998). The large variation of Cd concentration in the edible part of crops is demonstrated by an experiment of Isermann et al. (1983), where several plant species where grown on a Cd-contaminated soil (Table 3).

Large differences in the Cd concentration of pasture species have been reported, where the highest concentrations were found in *Asteraceae* and *Brassicaceae* (Stoeppler 1991, Wagner 1993). Capeweed may accumulate 10 to 40 times the Cd concentrations of subterranean clover and ryegrass, respectively (Tiller et al. 1997). This indicates that changes of plant species available to grazing animals via farm management has great impact on Cd transfer to human food chain (Mench 1998).

Among cereals, wheat accumulates much higher amounts of Cd in grains than other species. Horak (1976) and Puschenreiter and Horak (2000) found that wheat takes up about twice as much Cd compared to rye, oat or barley. Cd concentration in durum wheat is generally higher than in summer wheat (Mench 1998). High concentrations of Cd were also reported for lineseed seeds, where 10-fold higher concentrations were observed compared to other seed crops (canola, lupin, Indian mustard, wheat) (Hocking and McLaughlin 2000).

Cd accumulation behaviour varies not only on the species level, but also between cultivars and individual plants. E.g., large genotypic variation of Cd accumulation was found for maize shoots (Kurz et al. 1999, Florijn 1993), where two main groups of inbreds were distinguished: a group with low shoot, but high root Cd contents (shoot Cd excluder) and a group with similar shoot and root Cd concentrations (non-shoot Cd excluder). Among Austrian wheat cultivars, a 2.5-fold variation of Cd concentration in grain was observed (Wenzel et al. 1996). Similarly, a 2.3-fold variation in seed Cd concentration was reported for 17 Australian lineseed lines (Hocking and McLaughlin 2000). These studies indicate that Cd transfer into edible parts of crops may be highly influenced by the selection of low-uptake cultivars.

Variation in heavy metal accumulation was also observed within the plant. Except for roots, the highest concentrations are found in leaves, whereas the lowest are typically observed in seeds (Machelett et al. 1993). Toxic metals like Cd seem to be restricted from transport into generative organs and seeds. In a field study by Puschenreiter and Horak (2000), the transfer factor of Cd from soil to plant was twice as high for straw compared to grain.

Crop rotation and growing of industrial crops

Rhizosphere effects of plants may affect the heavy metal bio-availability to the following cropping cycle. E.g., lupins are known to release citric acid, which may increase the Cd lability. Oliver et al. (1993) reported from two different locations that Cd concentration in wheat is highest when grown after lupins.

The cultivation of industrial plants has been considered as a valuable option for agricultural use of metal-polluted soil. Zheljazkov and Nielsen (1996b) observed, that lavender could be successfully grown in highly heavy metal polluted areas without any risk of essential oil contamination. Similar results were obtained for peppermint and cornmint, where no heavy metal contamination was found in the essential oils. Despite of the yield reduction (up to 14%) caused by heavy metal contamination, mint still remained a very profitable crop and it could be used as substitute for other crops (Zheljazkov and Nielsen 1996a). Other industrial crops suggested for cultivation on metal contaminated soils were fibre plants like flax, cotton, hemp (e.g., Yanchev et al. 2000, Angelova et al. 2004) and energy crops, such as *Salix* trees and reed canary grass (Börjesson 1999).

Table 3. Cd concentration in different crops grown on a highly Cd-contaminated soil (Isermann et al. 1983)

Crop species	Cd concentration (mg/kg)	
	mean	range
Lettuce	0.22 (FW)	0.19–0.25 (FW)
Spinach	1.71 (FW)	1.34–1.93 (FW)
Celery leaves	28.8 (DW)	21.0–39.3 (DW)
Celery tubers	20.1 (DW)	13.5–26.5 (DW)
Potato	1.76 (DW)	1.24–2.48 (DW)
Spring barley (grain)	0.64 (DW)	0.20–1.00 (DW)
Tobacco (leaves)	56.5 (DW)	46.8–69.4 (DW)

FW = fresh weight, DW = dry weight

Soil treatment measures

Fertilization

Several reports have shown, that Cd concentration in crops can be decreased by the application of Zn fertilizers (Abdel-Sabour et al. 1988, Choudhary et al. 1994, Oliver et al. 1994, 1997, Grant and Bailey 1998). However, some studies reported no interaction between Cd uptake and Zn application (MacLean 1976, Singh and Steinnes 1976, White and Chaney 1980). The effect of Zn fertilization seems to be largely dependent on the Zn status of soils and plants and most significant effects were found when soils were Zn deficient (Oliver et al. 1994). On a soil not Zn-deficient, Zn application of up to 100 kg/ha could reduce Cd levels in potato tubers by less than 20% (Tiller et al. 1997). Also, Cu competes with Cd for uptake by the plant (McLaughlin et al. 1998). Interactions between plant Cd uptake and other micronutrients than Zn and Cu (Mn, Fe) have been demonstrated in solution culture (Cataldo et al. 1983), but no glasshouse or field evidence is available.

Organic amendments

Organic amendments like compost, farmyard manure (FYM), bio-solids or bio-solid compost may effectively reduce the bio-availability of heavy metals in soils due to its high content of organic matter and high concentrations of P and Fe (Brown et al. 2003). The effect of manure on heavy metal availability is due to the introduction of organic matter to the soil, which may retain Cd in the soil against both leaching and crop uptake (Jones and Johnston 1989). Vácha et al. (2002) compared various organic amendments and concluded that the efficiency is strongly dependent on the quality of the organic matter. In their study, immobilisation was most effective on acid soils with low cation exchange capacity.

The effectiveness of FYM application is demonstrated by Narwal et al. (1992). Following the application of 2% farmyard manure, Ni concentration decreased by 39, 40, 45 and 35% in carrot, fenugreek, spinach, wheat grain, respectively. The long term effects of FYM application compared to NPK fertilization on Cd levels in crops are demonstrated by the Rothamsted Classical Experiments (Jones and Johnston 1989) and on the INRA experimental farm in Couhins (Mench 1998).

The effectiveness of the incorporation of straw into contaminated soil has been investigated by Tlustoš et al. (1995). They have observed that this treatment decreased the concentration of Cd and Zn in spinach by 40% and 25%, respectively.

Inorganic amendments

Inorganic amendments are very effective in decreasing the metal bio-availability due to the introduction of additional binding sites for heavy metals and due to pH effects. Many of these amendments are by-products of industrial activities and therefore inexpensive and available in large amounts (Mench et al. 1998). An overview on previously successfully applied amendments and their effectiveness for different metals is presented in Table 4. Some results on decreased heavy metal concentrations after application are presented in Table 5.

Lime has been used for centuries to increase pH and thus decrease metal uptake by crops (Knox et al. 2001). Repeated application (every 2–5 years; 2–10 t/ha) is necessary to maintain metal immobilization and therefore larger quantities are necessary compared to other inorganic amendments (Knox et al. 2001). Application of lime increases pH and thus decreases availability of metals, with the exception of Mo and the metalloid As. Christensen (1984) showed that the Cd adsorptive capacity of soils increased by a factor of three for one pH unit increase between pH 4.0 to 7.7. In contrast, Vlamis et al. (1985) have shown that the pH increase to a level of 6 is high enough to regulate metal uptake, whereas further enhancement seems not to be necessary.

The effectiveness of liming may be different for each metal. Singh and Narwal (1984) reported that lime application reduces the uptake of Zn and Ni more than that of Cd. Metal uptake in response to liming may also vary among plant species. Reduced Cd accumulation induced by liming was more pronounced for lettuce and carrot as for potatoes and peanuts (Chaney et al. 1987). Likewise, liming reduced Zn concentrations in soybean seeds to a greater extent than in corn grains or cotton seeds.

Successful reports on liming are available for pot and for field studies. In pot experiments, Brüne et al. (1984) and Lombi et al. (2002) found a significant reduction of Cd and Zn concentrations (Table 5). Tlustoš et al. (1995) observed a decrease of Cd and Zn concentration in spinach by 80% and 75%, respectively.

In the following, the effect of liming in field studies is presented. Brallier et al. (1996) reported a study about the effect of liming on the availability of heavy metals in a soil amended with sewage sludge 16 years previously. The soil-pH was adjusted from 4.6 to 5.8, 6.5 and 6.9. The most consistent trends in plant concentrations in this study were found for Cd, Ni and Zn. The concentrations of these elements in cabbage and tomato fruit were significantly reduced at liming rates of 15 and 22 t/ha (pH 6.5 and 6.9, respectively). Oliver

Table 4. Some examples of mineral amendments which were effectively applied for the reduction of heavy metal and metalloid bioavailability

Amendment	Effective for	Reference
Al-Montmorillonite	Cd, Ni, Zn	Badora et al. (1998)
		Lothenbach et al. (1999)
Clinoptilolite	Cd, Pb, Zn	Ghuman et al. (1999)
		Chlopecka and Adriano (1996)
		Taslidas et al. (1997)
Diammonium phosphate	Cd, Pb, Zn	McGowen and Basta (1999)
Ferrous sulfate	As, Cr	Brown (1997)
Hydroxyapatite	Cd, Cu, Ni, Zn	Boisson et al. (1999a, b)
		Geebelen et al. (1999)
Lime	Cd, Cu, Ni, Pb, Zn	Brüne et al. (1984)
		Lombi et al. (2002)
		Chlopecka and Adriano (1996)
		Cordovil et al. (1999)
		Derome and Saarsalmi (1999)
		McGowen and Basta (1999)
Manganese oxides	Cd, Pb	Chen et al. (2000)
Red mud	Cd, Pb, Zn	Müller and Pluquet (1999)
		Lombi et al. (2002)
		Friesl et al. (2003)
Synthetic zeolites	Cd, Cu, Ni, Pb, Zn	Gworek et al. (1992)
		Oste et al. (1999)
		Rebedea and Lepp (1994)
Water treatment sludge	Cd, Cu, Ni, Pb, Zn	Elliot and Singer (1998)
		Müller and Pluquet (1999)

et al. (1996) have observed, that Cd concentration in wheat and barley grain can be decreased when pH is increased from 4.0 to 5.0 by liming. Liming could reduce the Zn concentration in oat grains by 54% (Table 5) (Brüne et al. 1984). Mönicke et al. (1999) reported a reduction of Cd transfer by 50% caused by liming, however, the effect was lower on highly contaminated soils compared to soils with lower Cd levels (i.e. 1.7 mg Cd/kg). Rietz et al. (1983) reported that lime application of 3 t/ha reduced the uptake of Cd in winter wheat by –33% in straw and –36% in grain. In their study, only a slight reduction of Zn was found and levels of Pb remained unchanged, which is probably because Pb accumulation was mainly due to air pollution.

Adding to much lime to the soil can lead to the immobilization of essential nutrients and the mobilization of harmful anions (As, Mo) (Conyers 2002). Additionally, it should be noted that in some cases liming could not decrease the uptake of Cd due to the high buffer capacity of the soil (Tiller et al. 1997). Also in saline soils, the effect of the limestone application may be negligible (Mench 1998). Therefore, soil characteristics have to be evaluated to estimate the effects of the lime application before addition to the soil.

Zeolites are hydrated aluminosilicates that have selective capacities to adsorb metals. Therefore, zeolites are considered to be among the most effective mineral amendments reducing heavy metal transfer to plants; some examples are given in Tables 5 and 6. Beside natural occurring zeolites (clinoptilolite, philipsite etc.), also synthetic zeolites are promising. Chlopecka and Adriano (1997) found that zeolites could reduce the accumulation of Cd, Pb and Zn by up to 72,

Table 5. Selected literature reports on the effect of soil amendments (amount added to soil in brackets) on the reduction of metal uptake by crops

Amendment	Crop	Reduced metal concentration (%)	Reference
Farmyard manure (2%)	spinach leaves	18 (Zn)	Narwal et al. (1992)
		24 (Cd)	
	wheat grain	26 (Zn)	
		32 (Cd)	
Lime (1%)	oat grain	54 (Zn)	Brüne et al. (1984)
	oat straw	60 (Zn)	
Lime (0.25%)	oilseed rape leaves	12 (Cd)	Lombi et al. (2002)
		56 (Zn)	
	wheat grain	29 (Cd)	
		34 (Zn)	
Lime (0.1%)	winter wheat straw	33 (Cd)	Rietz et al. (1983)
	winter wheat grain	36 (Cd)	
Zeolite (0.25%)		0–52 (Cd)	Chlopecka and Adriano (1997)
Zeolite (0.50%)		0–72 (Cd)	
Zeolite (0.25%)	maize leaves	24–72 (Pb)	
Zeolite (0.50%)		24–81 (Pb)	
Zeolite (0.25%)		3–35 (Zn)	
Zeolite (0.50%)		15–41 (Zn)	
Apatite (0.25%)		51 (Cd)	
Apatite (0.50%)		59 (Cd)	
Apatite (0.25%)	maize leaves	41 (Pb)	
Apatite (0.50%)		44 (Pb)	
Apatite (0.25%)		50 (Zn)	
Apatite (0.50%)		51 (Zn)	
Hydroxyapatite (0.5%)	maize leaves	92 (Cd)	Boisson et al. (1999a, b)
		81 (Pb)	
		61 (Zn)	
Hydroxyapatite (1.0%)	maize leaves	95 (Cd)	
		54 (Pb)	
		85 (Zn)	
Hydroxyapatite (0.5%)	maize leaves	99 (Cd)	
		76 (Pb)	
		96 (Zn)	
Red mud (1.0%)	*Amaranthus hybridus* shoots	38–87 (Cd)	Friesl et al. (2003)
		50–81 (Zn)	
		66–87 (Ni)	

81 and 41%, respectively (Table 5). Other successful reports on zeolite application are summarized by Knox et al. (2001).

The use of phosphate compounds (apatite or hydroxyapatite) for the immobilization of metals was in the focus of many studies. Knox et al. (2001) found that hydroxyapatite from North Carolina (25 and 50 g P/kg) effectively immobilized Pb, Zn and Cd. On the other hand also nutrient concentration, such as P, Mn, and Fe decreased in the plant tissue, whereas Ca and Mg increased.

Other mineral amendments include red mud, Fe oxides, and beringite, among others. Friesl et al. (2003) have shown, that the application of red mud to four different heavy metal contaminated soils reduced plant uptake of Cd, Pb and Zn in *Festuca rubra* and *Amaranthus hybridus* by 38–87%, 50–81% and 66–87% on certain soils. Lombi et al. (2002) found that the application of red mud, lime and beringite was very effective in reducing heavy metal uptake of oilseed rape, pea, and wheat. Müller and Pluquet (1997) reported a reduction of Cd concentration in wheat straw by 75% and in wheat grain by 30–40% due to the amendment of Fe oxides. However, in the case of red mud application, precaution is required due to possible content of Cr and As (Friesl et al. 2004). Boisson et al. (1999a, b) reported, that Cd, Cu, Ni, Pb and Zn are immobilized very effectively by beringite, a modified clay mineral, combination of steel shots + beringite, and hydroxyapatite. Moreover, the effect of these additives is constant in different soils. However, it has to be considered, that also essential elements can be immobilized, leading to nutrient deficiency for plants (Knox et al. 2001).

Immediate effectiveness and the long-term sustainability are the main advantage of inorganic amendments. Since many of these materials are industrial by-products, availability and costs are reasonable. However, their use is physically limited to shallow contamination. Also, periodic application may be necessary to ensure continued effectiveness (Knox et al. 2001).

Conclusions

Several low-cost agricultural methods are available to reduce the transfer of heavy metals into human food chain. Choosing crop species or varieties with low metal transfer factors is one effective approach. Caution is required for the growth of leaf vegetables, e.g. spinach and lettuce, whereas cultivation of cereals, legumes, potatoes and other low-uptake vegetables is recommended for contaminated soils. However, also among cereals large differences where found and highest Cd concentrations were observed in wheat.

Growing low-uptake wheat cultivars or other cereals may effectively reduce the Cd transfer.

Another effective approach is to reduce the bioavailability of heavy metals in soils by the addition of various amendments. Organic amendments like farmyard manure or inorganic additives like lime, zeolites and Fe oxides where found to reduce the transfer of metals into crops. Most of these materials are available in large amounts and incorporation into soil is easy, if the contamination is restricted to topsoil. However, repeated application may be necessary and the effectiveness is largely dependent on soil conditions and has to be proved periodically.

Further effective methods to reduce metal transfer into food chain includes crop rotation and cultivation of industrial or bio-energy crops.

It is concluded the most effective method has to be chosen according to the specific site conditions. The combined implementation of several methods may be necessary to keep heavy metal concentrations below guideline values. However, on sites with moderate contamination levels in the soil the methods discussed here may help to establish safe agriculture at low cost.

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ABSTRAKT

Nízkonákladové možnosti omezující transfer těžkých kovů do potravního řetězce – studie

Kontaminace těžkými kovy ovlivňuje značná území jak v Evropě, tak po celém světě. Postižená místa jsou lokalizována blízko průmyslových center, kolem velkých měst a v blízkosti těžby a zpracování rud. Zemědělství v těchto oblastech čelí významným problémům kvůli transferu těžkých kovů do rostlin a následně do potravního řetězce. Tato studie dává stručný přehled jednoduchých, ale efektivních opatření, jak redukovat přenos těžkých kovů do poživatelných částí rostlin. Jelikož se rostlinné druhy a odrůdy značně liší v příjmu těžkých kovů, pro pěstování jsou vybírány rostliny s nízkými transfer-faktory (např. luskoviny, obilniny), které mohou podstatně redukovat obsahy kovů v konzumních částech rostlin. Pěstování rostlin s vyšší příjmovou kapacitou těžkých kovů (např. špenát či salát) by mělo být zcela eliminováno. Aplikace půdních meliorantů je další velmi efektivní krok k redukci koncentrace těžkých kovů v rostlinách. Jak organické hnojení (např. chlévská mrva), tak i přídavek anorganických meliorantů (např. vápno, zeolity nebo oxidy železa) vedl ke snížení obsahů kovů v rostlinách. Dalšími efektivními metodami

vedoucími k redukci transferu kovů do půdního řetězce jsou vhodná rotace plodin a pěstování průmyslových nebo energetických rostlin. Závěrem je možno konstatovat, že metody citované v tomto článku představují relativně snadný a efektivní způsob k dosažení bezpečného zemědělství na středně kontaminovaných půdách.

Klíčová slova: těžké kovy; kontaminace půd; zemědělské půdy; omezený příjem kovů rostlinami; půdní melioranty

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