

Designing Cropping Systems for Metal-Contaminated Sites: A Review^{*1}

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ABSTRACT

Considering that even contaminated soils are a potential resource for agricultural production, it is essential to develop a set of cropping systems to allow a safe and sustainable agriculture on contaminated lands while avoiding any transfer of toxic trace elements to the food chain. In this review, three main strategies, *i.e.*, phytoexclusion, phytostabilization, and phytoextraction, are proposed to establish cropping systems for production of edible and non-edible plants, and for extraction of elements for industrial use. For safe production of food crops, the selection of low-accumulating plants/cultivars and the application of soil amendments are of vital importance. Phytostabilization using non-food energy and fiber plants can provide additional renewable energy sources and economic benefit with minimum cost of agricultural measures. Phytoextracting trace elements (*e.g.*, As, Cd, Ni, and Zn) using hyperaccumulator species is more suitable for slightly and moderately polluted sites, and phytomining of Ni from serpentine soils has shown a great potential to extract Ni-containing bio-ores of economic interests. We conclude that appropriate combinations of soil types, plant species/cultivars, and agronomic practices can restrict trace metal transfer to the food chain and/or extract energy and metals of industrial use and allow safe agricultural activities.

Key Words: agronomic practices, food safety, hyperaccumulator, phytoremediation, trace element

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INTRODUCTION

The scarcity of soil resources as human population continuously increases will inevitably force farmers to cultivate on contaminated areas. In the long run, this issue will be of most importance for land contaminated with heavy metals (*e.g.*, Cd, Hg, Ni, Pb, and Zn) and other trace elements (*e.g.*, As and Se), as regard to their persistence in soils. Soil contamination is a widespread phenomenon, and soil-to-plant transfer and effects of metals have been well documented (*e.g.*, Morel, 1997). It is now established that metals can be phytotoxic, plants are unable to grow in some circumstances (*e.g.*, high metal concentration and low

pH), and the toxic metals can be transferred to edible plant parts at rates sufficient to cause a threat to human health. Such phenomena have led governments to ban food production in large surface areas of land considered too contaminated regarding the risks for population health unless decontamination procedures are previously conducted.

Hot spots of metal pollution can generally be managed and remediated by means of heavy technologies based on excavation, treatment, and subsequent reuse of treated materials for landscaping and soil construction (Séré *et al.*, 2008; Simonnot and Croze, 2008), but such technologies are neither economically nor environmentally feasible for large surface areas. In general,

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vast sites are abandoned and under strong surveillance regarding their possible negative impacts on human health and environmental targets, such as water bodies, air, and ecosystems. In principle, they are seldom used for economical purposes, and even less for food production.

However, in densely populated countries such as China, some contaminated land is already exploited for food production to compensate the loss of agricultural surface due to urbanization. It has been reported that 1/5 of agricultural land of the country is currently contaminated (Chen, 2007), making agriculture on metal-contaminated soils obligatory. Similarly, in other industrialized countries, such as Europe (*e.g.*, France), land has to be removed from traditional agricultural use (*i.e.*, food production) as a result of contamination with metals (*e.g.*, deposition from metal smelters and mine sites and wastewater spreading over long period of time). Moreover, with the increasing demand for safer food, which will undoubtedly reduce the quality standards, more care will be required for a large portion of land with soil-to-plant transfer risks of toxicants.

Therefore, there is no other alternative besides developing new agricultural scenarios suitable for such environments, to allow safe and economically valuable agriculture on areas that are currently considered inappropriate and toxic. The new strategies to be developed should be based on the designing of suitable agroecological and even forest systems combining biomass production and management of contaminants in order to minimize the risks for adjacent environments, crops, and human beings, and provide regular revenue to the farmers. There are two main options: i) removing pollutants according to economically and technically reasonable means, applicable to large surfaces, while using land for valuable plant production and ii) managing contaminants, *e.g.*, growing plants with specific methods to minimize transfer from soil to edible plant part and avoid dispersion of contaminants in the environment; in the latter case, designed cropping systems have to focus on production quality rather than on soil remediation. Of course, in both options, plants (*e.g.*, species, variety, and agricultural practices) are part of the challenge, and phytoremediation appears as a relevant remediation process. Phytoremediation, a concept which has emerged in the 1990s (Salt *et al.*, 1994), after having been suggested earlier (Chaney, 1983), covers a set of soil treatments based on processes induced by plant roots on pollutants (*i.e.*, stabilization, extraction, and degradation). Following the study of Baker *et al.* (1991), which opened the way

for phytoextraction of Zn using hyperaccumulators, much work has been conducted at the international level demonstrating the potential of accumulating and hyperaccumulating plants for the remediation of metal-contaminated soils. Removal of metals by plants significantly reduces the available pool (Chardot *et al.*, 2005), such as that for Ni (Echevarria *et al.*, 2006), Cd (Gérard *et al.*, 2001), and Zn (Sinaj *et al.*, 2004). Another application of phytoremediation is stabilization through the reduction of solubility and transport (soluble, colloidal) of pollutants as a result of rhizosphere interactions (*e.g.*, Morel *et al.*, 1986) and root activity. Phytostabilization is generally combined with amendment additions to increase metal immobilization (Rutens *et al.*, 2010), and allows growth of edible plants. However, phytoremediation is only part of the solution, and there is a great need for comprehensive agro-economic approaches to deal with contaminated soils. As a remediation process, phytoremediation can only be considered as an element to be included in sustainable production systems adapted to contaminated environments.

Considering that even contaminated soils are a potential resource for plant production, it is necessary to propose sets of agricultural scenarios and recommendations to permit plant growth and avoid dissemination of toxic elements, and to extract them for further economical use. In this perspective, this paper is a contribution to the future development of cropping systems which would allow a safe and regular agricultural activity on contaminated soils while avoiding any transfer of metals to the food chain. From the knowledge acquired in the management of contaminated land, we have examined three main situations where agriculture has to be conducted on metal contaminated soil: i) production of edible plants, ii) production of non-edible plants of industrial interests, and iii) extraction of elements for industrial use. Then, we have attempted to suggest appropriate cropping systems which would require more scientific and technologic attention in the near future.

CROPPING SYSTEMS FOR PRODUCTION OF EDIBLE PLANTS

General principles and goals

There is increasing concern that cultivation of edible plants on contaminated soil can lead to the uptake and accumulation of heavy metals in the edible parts with their concentrations exceeding statutory or advisory limits (*e.g.*, legal limit set by FAO/WHO) (Zhuang *et al.*, 2009; Nabulo *et al.*, 2010; Singh *et al.*,

2010; Luo *et al.*, 2011). It is therefore imperative to develop suitable strategies to restrict the metal accumulation in such crops and its further transfer to the food chain. The main principle is to use phytoexclusion strategy, which is involved in the selection and breeding of low-accumulating cultivars, reduction of bioavailable metals in the soil, and restriction of their potential uptake and translocation by plants.

Cereals

In the past decades, there have been reports that the uptake and translocation of heavy metal pollutants in plants varies greatly, not only among plant species but also among cultivars within the same species (John, 1973; Morishita *et al.*, 1987; Wu *et al.*, 1994, 1999; Cieslinski *et al.*, 1996; Yu *et al.*, 2006; Wu *et al.*, 2007a). Recently, there have been studies of significant differences in the uptake of heavy metals among cultivars of rice (*Oryza sativa* L.), maize (*Zea mays* L.), wheat (*Triticum aestivum* L.), and soybeans (*Glycine max* L.). Studies on the genotypic differences in heavy metal concentrations in cereal crops are summarized in Table I.

The selection of rice cultivars to minimize heavy metal accumulation by the plant has been extensively studied. Wu *et al.* (1999) evaluated 20 cultivars of rice in South China on a contaminated soil and their results showed that the grain Cd concentrations ranged from 0.48 to 1.17 mg kg⁻¹. The main reason causing the differences was that the low-accumulating cultivar exhibited a much lower Cd translocation rate to grains than the high accumulating cultivar. In recent years, most studies have been conducted on the molecular mechanism of the low-accumulating rice cultivar. A novel mapping population consisting of 39 chromosome segment substitution lines (CSSLs) was used to locate the putative quantitative trait loci (QTLs) for Cd concentration in brown rice (Ishikawa *et al.*, 2005a). Xue *et al.* (2009) found that 3 QTLs on chromosomes 6 and

7 controlled root and shoot Cd concentrations. Ueno *et al.* (2010) reported that the gene (OsHMA3) was responsible for low Cd accumulation in rice.

Heavy metal concentrations have also been shown to vary among wheat cultivars (Oliver *et al.*, 1995; Cieslinski *et al.*, 1996; Zhang *et al.*, 2002a; Gao *et al.*, 2011). Erdman and Moul (1982) evaluated 7 cultivars of durum wheat and reported that their grain Cd, Cu, and Zn concentrations ranged from 0.13 to 0.25 mg kg⁻¹ with an average of 0.22 mg kg⁻¹. Gao *et al.* (2011) also found that the wheat cultivars showed significant genotypic differences in grain concentrations of Cd and Zn. Greger and Lofstedt (2004) observed that differences among the cultivars in the ability to accumulate Cd in grain were related to variations in the translocation from root to shoot and within the shoot, rather than to differences in root uptake. Similar trends were observed by Harris and Taylor (2004). Clarke *et al.* (1997) found that grain Cd concentrations were largely controlled by a single gene. The low-Cd wheat cultivar Strongfield (Clarke *et al.*, 2005) was released in 2004 and is now sown on more than 25% of the durum area in Canada (Grant *et al.*, 2008).

Guo *et al.* (2010) observed that the grain Cd, Pb, Zn, and Cu concentrations in sweet corn cultivars were higher than those of normal corn cultivars. Analysis of 17 varieties of soybean showed that the soybean variety En-b0-1-2 was the low-Cd cultivar in both field and pot experiments (Arao *et al.*, 2003b). Low-accumulating maize and soybean cultivars appeared to retain more heavy metals in the root and translocate less to the grain than high-accumulating ones (Arao *et al.*, 2003b; Ishikawa *et al.*, 2005b; Guo *et al.*, 2010).

Vegetables

Similar to cereal crops, vegetables have a variety of capacities in accumulating heavy metals among different types of species. The leafy vegetables tend to accumulate higher amounts of heavy metals in edible

TABLE I

Studies on genotypic differences in heavy metal concentrations in cereal crops

Crop	References
Rice (<i>Oryza sativa</i> L.)	Morishita <i>et al.</i> , 1987; Wu <i>et al.</i> , 1994, 1999; Arao and Ae, 2003a; Liu <i>et al.</i> , 2003b, 2005a, 2007a; Li <i>et al.</i> , 2005b; Yu <i>et al.</i> , 2006, 2008; Fu <i>et al.</i> , 2008; Shi <i>et al.</i> , 2009
Wheat (<i>Triticum aestivum</i> L.)	Erdman and Moul, 1982; Oliver <i>et al.</i> , 1995; Cieslinski <i>et al.</i> , 1996; Clarke <i>et al.</i> , 1997; Zhang <i>et al.</i> , 2000, 2002a; Greger and Lofstedt, 2004; Harris and Taylor, 2004; Clarke <i>et al.</i> , 2005; Gao <i>et al.</i> , 2011
Maize (<i>Zea may</i> L.)	Florijn and Beusichem, 1993; Gao <i>et al.</i> , 1995; Kurz <i>et al.</i> , 1999; Samake <i>et al.</i> , 2003; Tudoreanu and Phillips, 2004; Guo <i>et al.</i> , 2010
Soybean (<i>Glycine max</i> L.)	Reddy and Dunn, 1986; Arao <i>et al.</i> , 2003b; Ishikawa <i>et al.</i> , 2005b

parts than fruit or even root vegetables (Zheng *et al.*, 2007; Wang *et al.*, 2009a; Zhuang *et al.*, 2009; Khan *et al.*, 2010), although this is not always the case. The high metal accumulation in leafy vegetables is regarded as a result of their high rates of transpiration and translocation of metals, particularly Cd (Khan *et al.*, 2010). In addition, leafy vegetables are fairly susceptible to physical contamination by soil dust and splash because of their high foliar surface area (Zhuang *et al.*, 2009). Accumulation of Pb and Cr in leafy vegetables resulting from aerial deposition is substantially greater than that of Cd, Zn, Ni, and Cu, of which the uptake occurs predominantly *via* the roots and subsequent transport to the edible shoot tissues. This highlights the importance of mulching vegetable gardens to reduce exogenous metal contaminants on leaves (Nabulo *et al.*, 2010).

Heavy metal accumulation may also differ greatly within cultivars of an individual species when grown on the same soil. Wide variation of metal accumulation within cultivars has been documented in potato (Dunbar *et al.*, 2003), carrot, pea (Alexander *et al.*, 2006), asparagus bean (Zhu *et al.*, 2007), and several leafy vegetables (Wang *et al.*, 2007b, 2009a). Alexander *et al.* (2006) tested five cultivars of six common vegetables and found that carrot and pea cultivars exhibited significant intra-species differences in Cd and Cu. Wang *et al.* (2009a) observed that there were large genetic variances in water spinach Cd concentration, ranging from 0.593 to 1.824 mg kg⁻¹ among 30 tested cultivars when grown on Cd-treated soil. Among them, six cultivars were identified as pollution-safe cultivars and the preferential sequestration of Cd in root and cell wall fragment was one possible mechanism leading to low Cd accumulation. Using suppression subtractive hybridization analysis, Huang *et al.* (2009) found tremendous differences in Cd-induced gene expression in two water spinach cultivars with contrasting Cd-accumulating capacity, suggesting that Cd-accumulating ability of this species is a genotype-dependent and inherited trait.

Orchards

One of the most common elevated trace metal contaminants in fruit orchard soils is Cu. The long-term use of Cu-based fungicides has resulted in elevated accumulation of Cu in soils of vineyard and apple orchard, typically ranging from 200 to 500 mg kg⁻¹ (Chopin *et al.*, 2008). The values are over ten times higher than the typical background Cu concentrations in soils (5–30 mg kg⁻¹) (Wightwick *et al.*, 2010).

There are increasing concerns of Cu availability in orchard soils and its subsequent accumulation in

grapevines (Mirlean *et al.*, 2005; Lai *et al.*, 2010). The adverse effects of Cu accumulation in apple orchard and vineyard soils on enzymatic activity, microbial community, and soil invertebrates have also been emphasized (Wang *et al.*, 2009b; Zhou *et al.*, 2011). In some vineyard sites of central Taiwan, high accumulation of Cu up to 86 mg kg⁻¹ in grape berries was reported, far exceeding the maximum residue level of Cu in wine grapes (20 mg kg⁻¹) proposed by the European Regulation (Lai *et al.*, 2010). In contrast, much lower Cu content (4.5 mg kg⁻¹) was observed in berries grown in a calcareous contaminated Champagne vineyard area, France (Chopin *et al.*, 2008). In fact, many studies have demonstrated that the availability of Cu is dominated by soil factors such as pH, cation exchange capacity (CEC), clay content, and organic matter content. Cu availability in soil is generally lowest in neutral soils and increases with increasing soil acidity and alkalinity (Wightwick *et al.*, 2010). This may have important implications in considering strategies (*e.g.*, soil pH management) to reduce Cd availability and toxicity in vineyards. On the other hand, several studies suggested that organic carbon is responsible for a substantial retention of Cu mainly by increasing CEC, and thus application of compost is another feasible option to reduce Cu mobility in vineyard soils (Lai *et al.*, 2010; Wightwick *et al.*, 2010). From the long run, it is wise to restrict or even stop the use of Cu-based fungicides, and develop environmentally safer alternatives to Cu-based fungicides, which have been banned in some European countries like the Netherlands (Lai *et al.*, 2010).

Chemical immobilization of heavy metals in soil

In-situ chemical immobilization is a cost-effective way to reduce metal concentrations in plants through the addition of chemical amendments in soil, which may provide a solution through the formation of low solubility and/or precipitation of minerals (Basta and McGowen, 2004). Kumpiene *et al.* (2008) reviewed the published papers during 2000–2005 on immobilization of As, Cr, Cu, Pb, and Zn in soils.

Research on chemical immobilization of heavy metals has included alkaline substances, phosphate- and silicon-based materials, adsorption agents, and organic matters. Alkaline substances, such as lime, have been used in traditional agricultural practice to raise soil pH levels for plant growth (Simon, 2005). The addition of alkaline amendments reduced heavy metal solubility in soil by increasing soil pH and formed insoluble compounds (Maddox and Soileau, 1991; Chen *et al.*, 2000; Khan and Jones, 2009). One of the disadvantages of liming is that the gradual reduction of the liming agent

over time due to dissolution and leaching, which can be accelerated by acid deposition, may have a potential risk that needed to be considered prior to long-term phytoexclusion practices (Ruttens *et al.*, 2010).

Silicon (Si) is not considered to be an essential mineral element for plants, but has been shown to ameliorate the deleterious effects of metals on plants grown on contaminated soils (Liang *et al.*, 2005, 2007; da Cunha and do Nascimento, 2009). Silicon applied in agriculture came mainly from by-products of fly-ash (Dwivedi *et al.*, 2007), steel sludge, furnace slag (Chen *et al.*, 2000), and Si chemicals (Shi *et al.*, 2005; Li *et al.*, 2009c; Gu *et al.*, 2011). Liu *et al.* (2009) reported that foliar application of silica sols significantly increased the dry weight of rice grains and shoots grown on Cd-contaminated soils, whereas the Cd concentration apparently decreased in the grains and shoots. The mechanism of Si foliar application to alleviate the toxicity and accumulation of Cd in rice grains may be related to the co-precipitation of Cd with Si in stems (Gu *et al.*, 2011), and probable Cd sequestration in the shoot cell walls (Liu *et al.*, 2009).

Phosphate-based materials added to contaminated soils reduced the Pb mobility by ionic exchange and precipitation of pyromorphite-type minerals, $Pb_5(PO_4)_3X$, where $X = F, Cl, \text{ or } OH$ (Kumpiene *et al.*, 2008). The phosphate-based materials include phosphate rock (Zwonitzer *et al.*, 2003; Cao *et al.*, 2004; Brown *et al.*, 2005), diammonium phosphate (Basta and McGowen, 2004; Chen *et al.*, 2007; Khan and Jones, 2009), phosphate-based salts (Ownby *et al.*, 2005; Simon, 2005; Li *et al.*, 2008), and phosphoric acid (Cao *et al.*, 2003).

Adsorption agents such as zeolites, iron and/or manganese oxides, and clay minerals have also been widely studied because of their high cation exchange capacity and high specific surface. In a pot experiment, natural zeolites decreased the available part of Pb and restricted Pb uptake by rape (Li *et al.*, 2009a). Among 15 metal ions investigated, addition of vermiculite significantly reduced the uptake of various metal pollutants by lettuce and spinach (Malandrino *et al.*, 2011). A similar reduction effect on Cd and Zn accumulation in radish and Japanese mustard spinach was reported when 5% lherzolite was applied in a contaminated soil (Kashem *et al.*, 2010).

Organic matters such as peat, animal excrement, and composts can also effectively be used in metal stabilization. Li *et al.* (2008) reported that pig manure and peat could increase grain yields and decrease Cu and Cd concentrations in grain.

CROPPING SYSTEMS FOR PRODUCTION OF NON-EDIBLE PLANTS

Energy crops

The technology development and utilization of phytoremediation using tolerant energy crops can provide additional economic benefits (Meers *et al.*, 2005, 2010). These crop cultivars are cultivated on metal-contaminated soils for energy production purposes, and the sustainable development of resilient energy plants has turned into a new trend in the developed and developing countries.

Industrial non-food crops have been used for both remediation of contaminated land and renewable energy sources (Meers *et al.*, 2007). Canola (*Brassica napus* var. Hyola) plants grown under adverse soil conditions in the west side of the San Joaquin Valley (SJV) accumulated Se in various plant organs including seeds. The Se-enriched economically viable phyto-products of canola such as seed oil have been used as biofuel additive and organic fertilizer (Bañuelos, 2006; Bañuelos *et al.*, 2010).

A number of studies have indicated that many energy plants (*e.g.*, sunflower, willow, and poplar) have high tolerance to heavy metals such as Cu, Cd, Cr, and Zn (Lin *et al.*, 2003; Lunáčková *et al.*, 2003; Rosselli *et al.*, 2003). In addition, soybean (*Glycine max*) oil, palm (*Trachycarpus fortunei*) oil, and coconut (*Cocos nucifera*) oil have been considered as possible substitutes for fossil diesel (Agarwal, 2007). Hence, metal-tolerant industrial and energy crops are suitable candidates for long-term field remediation and sustainable economic development. This process could be of particular importance in countries with increasingly less arable land, such as China and India, due to the rapid expansion of population and urbanization.

Jatropha curcas L. (Euphorbiaceae) is a perennial oil-yielding plant cultivated in the tropical and subtropical regions of the world. It has been widely used for energy plantations and wasteland reclamation due to the ability to tolerate extreme environmental conditions such as drought, non-fertile, and heavily polluted soils. Based on the accumulation of different trace elements (Cu, Cr, Mn, Ni, and Zn) and nutrients (Ca, K, Mg, Na, and P) in *J. curcas* grown in the vicinity of a mica belt, it could be used for phytostabilization of mining areas (Nagaraju and Karimnlla, 2002). Recent studies have mainly focused on the improvement of growth performance and metal tolerance of *J. curcas* grown on metal-contaminated calcareous soils with the amendments of organic wastes (biosludge and dairy

sludge) and a biofertilizer (*Azotobacter chroococcum*) (Juwarkar *et al.*, 2008; Kumar *et al.*, 2008; Yadav *et al.*, 2009, 2010).

J. curcas can also grow well on the acid mine drainage irrigated soils ($\text{pH} \geq 3.78$) with addition of $\leq 1\%$ limestone in extremely acidic soils (Li *et al.*, 2009d; Wu *et al.*, 2011). The growth performance of *J. curcas* was enhanced at 0.50% of lime addition due to the increase of soil pH and the decrease of phytoavailable Al (about 95%), Zn (about 75%), and Cu (about 65%) in soil. The high ability to tolerate Al was suggested to relate to cell wall binding and the organic acids secreted by the roots of *J. curcas* (Wu *et al.*, 2011). Based on the inherent tolerance ability of *J. curcas* in existing adverse environmental conditions, it could be used as a suitable candidate for phytostabilization in acid mine tailings. Effective stabilization of liming is an additional possible practice to improve the soil quality.

Fiber crops

Bast fiber crops are widely grown in China and throughout the world. The characteristics of fiber crops include fast growth, strong adaptability, large biomass, and high economic value. The bast fibers are widely applied in textiles, adsorbents, and building materials (Liu *et al.*, 2003a; Hasfalina *et al.*, 2010). These non-wood fibers were also proved to be suitable for papermaking (Saikia *et al.*, 1997; Ververis *et al.*, 2004).

Recent attention has been given to fiber crops as a potential plant for heavy metal-contaminated soils due to its tolerance (Angelova *et al.*, 2004; Ho *et al.*, 2008; Yang *et al.*, 2010). Hemp (*Cannabis sativa*) could grow under 17 mg kg^{-1} Cd in soil and 1 mmol L^{-1} Cu in culture solution without showing any phytotoxicity symptoms (Citterio *et al.*, 2003; Arru *et al.*, 2004). Through a large-scale field investigation and a hydroponic culture experiment, ramie (*Boehmeria nivea*) has been revealed to possess a certain degree of constitutional metal tolerance to As, Cd, Pb, and Zn (Yang *et al.*, 2010).

However, due to the presence of toxic metals, high soil acidity, and poor physical properties with deficiency of nutrients in the vicinity of mine tailings (Pichtel and Salt, 1998; Ernst, 2005), fiber crops are hard to grow in these contaminated areas. Therefore, revegetation process should combine with application of soil amendments, especially those that can neutralize soil pH and enhance nutrition. A 3-year field experiment on a stripmine land showed that the yield of kenaf (*Hibiscus cannabinus*) increased with sewage sludge application (Carlson *et al.*, 1982). Similar results were also found in a pot trial by applying chicken manure to a Pb-contaminated soil (Ho *et al.*, 2008).

In another field trial conducted in Shangba Village, Guangdong, China, where the farmland soil was highly polluted by acid mine drainage from upstream mine tailings, the ameliorative effects of lime, dolomite, fly ash, and compost were tested (Table II) (Huang, 2009). The contaminated soil was characterized by its low pH (3.36) and high concentrations of heavy metals (1500 , 10 , 660 , and 480 mg kg^{-1} of Pb, Cd, Cu, and Zn, respectively) (unpublished data). The soil pH was adjusted to 7–8 by amendment application. The yield of kenaf shoots could reach 20 t ha^{-1} or above (Fig. 1) (Zou, 2008), which is similar to those on normal soils, indicating a successful and sustainable mode of reclaiming the acid mine soils by using kenaf in combination with soil amelioration. Besides, low levels of heavy metals were detected in kenaf fibers. Meanwhile, it was suggested that heavy metals had no significant negative effects on fiber quantity and quality (Linger *et al.*, 2002; Wang *et al.*, 2008). A highly contaminated soil is always considered to be a wasteland. However, with a certain amendment applied into the soil, fiber crops could grow healthily and rapidly. Therefore, these crops are good candidates to cultivate on highly contaminated soils for revegetation as well as economic purposes.

Though the heavy metals in fibers may not easily enter the food chain and do harm to human beings di-

TABLE II

Effects of different amendment treatments on yields and agricultural and economic characteristics of kenaf (Huang, 2009)

Treatment	Agronomic trait			Biomass	
	Yield	Stem diameter	Stem height	Bast fiber	Stem core
	t ha^{-1}	cm		g plant^{-1}	
Lime (0.6%)	22.0ab ^{a)}	4.4a	308a	24.7a	63.0a
Dolomite (5%)	40.5b	6.2b	374a	47.9b	129.0b
Fly ash (5%)	30.7ab	5.9ab	358a	37.1ab	93.3ab
Lime (0.6%) + compost (5%)	35.0ab	5.4ab	356a	40.2ab	110.0ab
Lime (0.6%) + fly ash (5%)	20.1a	5.6ab	351a	36.9ab	94.4ab

^{a)} Means followed by the same letter(s) in a column are not significantly different at $P < 0.05$.

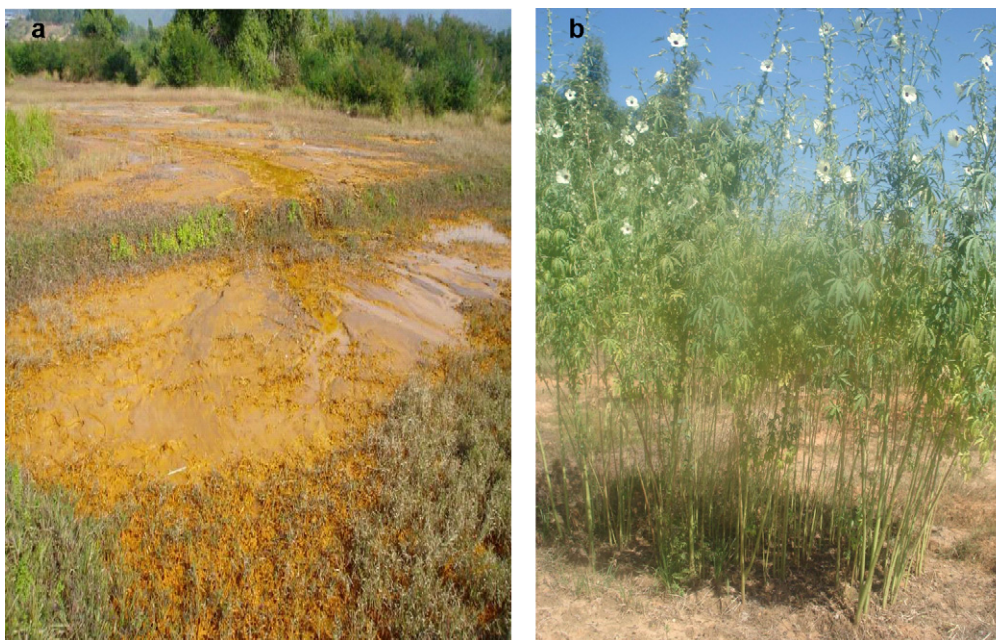


Fig. 1 A field trial of acid mine contaminated soil amendment in Shangba Village, Guangdong, China: (a) before growth of kenaf and (b) after growth of kenaf (Zou, 2008).

rectly, a minimum amount of metals is expected in these industrial raw materials. So, more attention should be paid to amendment selection and soil management practices to achieve the best crop yield and least potential hazard.

CROPPING SYSTEMS FOR EXTRACTION OF ELEMENTS FOR INDUSTRIAL USE

Phytoextraction of Cd, Ni and Zn: perspectives for phytomining and soil remediation

General principles and goals. Nickel is the most accumulated metal among hyperaccumulator plants. Approximately 390 plant species have been reported worldwide (Verbruggen *et al.*, 2009) to hyperaccumulate Ni in their shoot tissues (*i.e.*, Ni concentrations of above 1 000 mg kg⁻¹) and most of them easily reach 1% Ni in their leaf tissues when growing on soils with sufficient Ni availability (Massoura *et al.*, 2004; Chardot *et al.*, 2005). Hyperaccumulation of Cd and Zn occurs to be less than that of Ni (Peer *et al.*, 2006; Verbruggen *et al.*, 2009). Also, only a few plant species are able to take up both metals at high levels; these plants belong mainly to the family *Brassicaceae* (several species of the *Thlaspi*/*Noccaea* genus and *Arabidopsis hallerii*) as well as the species *Sedum alfredii* (Yang *et al.*, 2004). The co-hyperaccumulation is highly important because most of the Cd-contaminated sites also display high concentrations of Zn as these two metals are often present together in mineral ores and in industrial wastes and by-products. Metal-rich soils and mainly

ultramafic soils (*i.e.*, serpentine soils) are widespread over different areas of the world including the Alps, the Balkans, Turkey, New Caledonia, Cuba, Australia, and many other regions. These soils are a major problem for the development of agriculture due to extreme nutrient deficiency and toxicity of trace metals. The recovery of such metals from soils with hyperaccumulator plant crops as an alternative to traditional agriculture is a challenge for the rural development of these areas as well as for the local mining and metallurgical industry (Bani *et al.*, 2007). Very often, ultramafic soils are too poor in metal contents for viable traditional mining operations; phytomining, *i.e.*, recovery of metal hyperaccumulated in aboveground plant parts, proves to be profitable in these soils and becomes a true alternative to classical crops (Bani *et al.*, 2007; Chaney *et al.*, 2007). Also, the prices of some metals (*e.g.*, Ni, Co, and Ti) have dramatically increased for the past five years although a noticeable breakdown occurred during the 2009 crisis. During 2010, the prices of metals went up again due to increasing demand of Asian industries; at the time this text was written, the price of Ni at the London Metal Exchange was USD \$ 26 per kg. This alone justifies their mining from naturally rich soils or their recycling when remediating metal-contaminated soils. Industrial brownfield and mining sites are also often contaminated with trace metals (Cd, Co, Cr, Cu, Ni, and Zn), representing a major environmental concern (De Kimpe and Morel, 2000). In the case of Cd and Zn, the main objective of remediation is often removal of the metal fractions that present a risk in co-

ntaminated soils. Removal of total contents of metals in contaminated soils is often impossible thermodynamically speaking but has long been considered as an issue by stakeholders (Schwartz *et al.*, 2003; Keller and Hammer, 2004). Now, it is commonly accepted that the main goal of phytoextraction of Cd and Zn is to deal with keeping low the bioavailable fractions of these metals to prevent excessive uptake in following crops or leaching to groundwater. However, the recovery of metals in the produced biomass could be a way of reducing the overall costs of contaminated soil treatment, especially considering the expected increases in Cd and Zn market values in the forthcoming years.

Phytomining of Ni: agronomic perspectives. Several field trials and research programs have led to a better understanding of phytomining and proposed several sets of agronomic practices to end-users (Li *et al.*, 2003; Bani *et al.*, 2007; Chaney *et al.*, 2007). It is quite easy to grow Ni-hyperaccumulator plants in pot conditions on a wide array of soils and treatments (Robinson *et al.*, 1999) and significant amounts of Ni can be recovered after three months with Ni concentrations in shoots reaching more than 3% and depletion of total soil Ni reaching almost 5% (Kukier *et al.*, 2004; Massoura *et al.*, 2004; Chardot *et al.*, 2005). However, field-scale phytomining requires approaches that integrated species selection and breeding, phenology and cropping calendar, fertilization practices, and pest control (Chaney *et al.*, 2007). Two approaches have been developed so far: one intensively designed that includes all aspects of mechanized agriculture available in developed countries (Li *et al.*, 2003; Chaney *et al.*, 2007) and the other extensively designed that is adapted to ultramafic areas in the world where no modern cropping tools are available (Bani *et al.*, 2007; Bani, 2009). The latter needs the same scientific basic knowledge to optimize agronomic options. Some studies on the phenology of *Alyssum murale* showed that the curves of Ca, Mg, and Ni uptake during plant growth and development defined the mid-flowering stage as the best stage for harvesting with the highest Ni concentrations and the lowest Ca concentrations in shoots (Bani, 2009). Chaney's group led a breeding program with various species of *Alyssum* (Li *et al.*, 2003), among which *A. murale* from eastern Albania proved to be one of the best candidates (Bani *et al.*, 2009). The selected strains were able to accumulate 3% Ni in the shoots (including stems) as compared to 2% as a best value in the wild (Bani *et al.*, 2009), with reasonably increased yields (up to 20 t ha⁻¹ in field conditions) and a better retention of leaves that are the richest parts in the plant (Chaney *et al.*, 2007). Optimization

of soil fertility concerns mainly nitrogen (Bani *et al.*, 2007). *A. murale* is very efficient in taking up phosphorus (P) and soil P fertility is seldom a limiting factor (Shallari *et al.*, 2001; Bani *et al.*, 2007). However, N fertilization is seriously required and it was possible to limit N inputs to 100 kg N ha⁻¹ by fractioning its application (Chaney *et al.*, 2007; Bani, 2009). Since most Ni hyperaccumulators take up large amounts of Ca from Ca-deficient serpentine soils, it is necessary to maintain Ca fertility of such soils at a reasonable level (Kukier *et al.*, 2004; Chaney *et al.*, 2007).

Phytoextraction of Cd and Zn with hyperaccumulator plants at the field scale. Several teams in the past twenty years have carried out experiments on Cd and Zn phytoextraction by hyperaccumulators at the field scale and confirmed what had been studied at the lab scale although the adaptation of species such as *Nocca caerulea* to the field is not so easy (Schwartz *et al.*, 2003; Keller and Hammer, 2004; Chaney *et al.*, 2005, 2007). To date, most studies have shown that although *N. caerulea* can be used to clean up soils slightly or moderately contaminated by Cd and Zn (Ernst, 2005; McGrath *et al.*, 2006), the technology usually needs a long period (*e.g.*, 10 years) for cleanup due to the limited biomass production of hyperaccumulator species used (Koopmans *et al.*, 2008). Also, the phytoextraction duration can be underestimated since the bioavailable metal pools in soil decrease during phytoextraction, which leads to a decrease of metal uptake by plants and lower metal removal rates (Koopmans *et al.*, 2008). Thus, phytoextraction appears possible for Ni and As, but for Cd and Zn, the strategy is still far from large-scale practice (Van Nevel *et al.*, 2007). One of the keys for successful phytoextraction operations based on *N. caerulea* requires an increased biomass production.

Metal recovery in hyperaccumulator biomass. Ni is used in a wide variety of alloys and superalloys but the largest needs concern the production of stainless steel. For this reason, there is a wide variety of processes for Ni recovery that mainly depend on the type of and the concentrations of Ni in ores. Currently, about twenty processes are known, among which 6 are significantly used at the industrial scale, *e.g.*, pyrometallurgy (INCO and Falconbridge, Canada) and hydrometallurgy (Eramet-SLN, France). Some scientists have shown that pyrometallurgical processes can be used to produce pure Ni from hyperaccumulator plant residues (Boominathan *et al.*, 2004). This process is however difficult to promote in developed countries because of the strict environmental regulations. Other scientists promote co-incineration of hyperaccumula-

tor plants along with municipal wastes (Keller *et al.*, 2005). This also causes problems in terms of social acceptability and difficulty to recover metals that may escape with fly ashes. These ashes would then need a special treatment as dangerous wastes. One alternative to pyrometallurgy to valorize phytoextracted metals could be a hydrometallurgical process that better responds to clean technology standards (Barbaroux *et al.*, 2009, 2011, 2012). Hydrometallurgy consists in a set of techniques used for a long time (Habashi *et al.*, 2005). The yearly world production of Ni (over one million tons) is mainly based on pyrometallurgy but hydrometallurgy is also used because of the possibilities it offers for the elaboration of end products and for the recovery of metals in complex systems. Therefore, it is essential to continue development and adaptation of hydrometallurgical methods, in particular lixiviation methods, lixivate purification, and production routes of end products, in order to valorize metals present in various environmental materials. Biomass produced by phytomining is a substantial resource (*i.e.*, 2% Ni), for which a specific separation process should be developed.

Phytoextraction of As

Several plant species have been reported as As hyperaccumulators, primarily represented by terrestrial *Pteris* species (Ma *et al.*, 2001; Chen *et al.*, 2002b; Wang *et al.*, 2007a) and aquatic seaweeds (Nguyen *et al.*, 2011). Some angiosperms, *e.g.*, Brassicaceae *Isatis cappadocica* (Karimi *et al.*, 2009) and *Hesperis persica* (Karimi *et al.*, 2010), and ferns, *e.g.*, marsh fern (Anderson *et al.*, 2010), have also been reported as As hyperaccumulators. *Pteris vittata* has the merits of large biomass, high As concentration, and wide distribution; thus, it has been widely used in phytoextraction projects (Chen *et al.*, 2007; Ebbs *et al.*, 2009).

P. vittata has a wide distribution. Diverse habitats result in different genotypes. Although all ecotypes of *P. vittata* could hyperaccumulate As, the ability of hyperaccumulating As was very variable. The above-ground As concentration of *P. vittata* ranged from 643 to 3 009 mg kg⁻¹ (Cai *et al.*, 2007). It means that using different genotypes in phytoremediation projects could lead to several times of variations in the remediation efficiency. Therefore, differences in As accumulation by *P. vittata* genotypes were investigated and the 3–4 fold As accumulation differences were confirmed (Wang *et al.*, 2002; Zhao *et al.*, 2002; Wu *et al.*, 2009). It was found that some *P. vittata* ecotypes also had huge potentials in phytoextracting other toxic trace elements, *e.g.*, Zn, Cd, Cr, and Hg (Anet *et al.*, 2006; Su *et al.*,

2008b; Xiao *et al.*, 2008; Roccotiello *et al.*, 2010; Feng *et al.*, 2011; Kalve *et al.*, 2011). The reasons behind such ecotypic variations have not been identified yet, but these research results clearly showed the necessity to select appropriate genotypes according to the situation of contaminated fields.

Besides plant selection, complementary approaches to optimize plant growth and increase As concentration in *P. vittata* have been adopted. The uses of fertilizers (phosphate and nitrogen) and mycorrhizal fungi have received focused attention and both were able to increase the remediation efficiency (Chen *et al.*, 2002a; Liu *et al.*, 2005c; Wei and Chen, 2006; Huang *et al.*, 2007; Liao *et al.*, 2007; Leung *et al.*, 2010; Muthukumar and Bagyaraj, 2010; Xiong *et al.*, 2010). The effect of phosphate fertilizers is still unclear (Huang *et al.*, 2007; Santos *et al.*, 2010).

In addition, the optimization of environmental factors including soil properties, seedling age, and harvesting regimes has also been reported (Gonzaga *et al.*, 2007; Wan *et al.*, 2010; Xu *et al.*, 2010; Natarajan *et al.*, 2011). Younger seedlings have been found to extract As more quickly, but whether we should adopt young plants or apply more frequent harvest regimes still depends on the balance between larger biomass and higher concentrations.

A field experiment conducted showed that *P. vittata* can achieve maximum extraction efficiency by mowing 3 times a year, and the stubble height had a great impact on the *P. vittata* growth and extraction efficiency: a stubble height of 7.5 cm benefited the regeneration of *P. vittata* and the extraction efficiency of arsenic (Li *et al.*, 2005a).

In the plant tissues of arsenic hyperaccumulators (*Pteris* species), inorganic arsenic species arsenate (As(V)) and arsenite (As(III)) account for more than 95% of the total arsenic, while organic species have been detected as minor compounds (Ma *et al.*, 2001; Tu *et al.*, 2002, 2004; Zhang *et al.*, 2002b; Mathews *et al.*, 2010). Regardless of As(III) or As(V) supplied, As exists predominantly as As(V)–O in roots while mainly as As(III)–O in the rhizomes and fronds (Huang *et al.*, 2008; Mathews *et al.*, 2010). Arsenite was firstly oxidized in the growing medium and then further oxidized in the roots of *P. vittata* (Huang *et al.*, 2004; Mathews *et al.*, 2010). Arsenate was mainly reduced in the rhizomes (68%–71% as As(III)) and pinnae (> 90% as As(III)), and limitedly reduced in the roots (7%–8% as As(III)) (Huang *et al.*, 2004; Mathews *et al.*, 2010). Monomethylarsonic acid (MMA) can be methylated to dimethylarsinic acid (DMA), while DMA cannot be demethylated to MMA. Both MMA and DMA

can be oxidized to arsenate (As(V)) and arsenite (As(III)) (Chen *et al.*, 2004). Arsenic transformation mechanism has not yet been clearly explored and still leaves much to be resolved.

Phytoextraction faces the challenge of safe disposal of large quantities of contaminant-rich biomass after harvesting from contaminated sites (Cao *et al.*, 2010). Incineration was considered to be suitable for hyperaccumulators after harvest. During incineration of *P. vittata* containing high As content (1170 mg kg⁻¹), about 24% of total As was emitted at 800 °C, and 62.5% of the total emitted As was volatilized below 400 °C. Simultaneously, below 400 °C, As(V) reduced to As(III) (Yan *et al.*, 2008). The ash weight was about 4.4%–7.7% of the dried weight and the concentration of heavy metals presented regular variations in different stages of incineration. As concentration in the bottom ash was lower than that in the fly ash.

CO-CROPPING FOR REDUCING HEAVY METAL CONTENTS IN CROPS

Co-cropping of a hyperaccumulator (*N. caerulescens*) with a non-accumulator (*Thlaspi arvense*) increased the growth of the non-accumulator and reduced its Zn uptake, while in contrast, increased Zn uptake of the hyperaccumulator (Whiting *et al.*, 2001). Comparing with monoculture, intercropping decreased the Cd content in the aboveground parts of alfalfa (a normal crop) by 57.1%, while increased the Cd content in the aboveground parts of *Brassica juncea* (a high Cd accumulator) by 14.5% for the soils with Cd contents in the range of 0.37–5.37 mg kg⁻¹ (Li *et al.*, 2009e). The Cd contents in the aboveground parts of alfalfa under monoculture and intercropping were 0.21 and 0.09 mg kg⁻¹, respectively, neither of them being over the feed safety standard (Li *et al.*, 2009e). High Cd-accumulating varieties of oilseed rape could reduce the Cd uptake of co-cropped cabbage, but the Cd concentration in cabbage was still high (Liu *et al.*, 2007b; Su *et al.*, 2008a). Therefore, a low-metal crop is needed to produce safe agricultural products on contaminated agricultural lands in association with a hyperaccumulator. Growing a metal hyperaccumulator plant *S. alfredii* alongside a low metal-accumulating cultivar of maize was introduced to simultaneously phytoextract heavy metals from contaminated sewage sludge and to produce an agricultural crop (Liu *et al.*, 2005b). Results showed that this co-planting system effectively removed heavy metals from the sewage sludge by the hyperaccumulator while the harvested corn grains or *Alocasia* tubers were conformed to the Chinese standard for animal feeds (Liu *et al.*, 2005a; Wu *et al.*,

2007b).

However, the beneficial effect on the co-cropped normal crop was not always observed from the co-cropping with a hyperaccumulator. Combined cropping of *N. caerulescens* and *Hordeum vulgare*, though caused a decrease in Zn concentration in barley biomass, had a significant increase in Cd accumulation (Gove *et al.*, 2002). Similar results were also obtained by Jiang *et al.* (2010). The average Cd concentration in ryegrass shoots was increased by co-cropped *N. caerulescens*, but the Zn concentration and total uptake by ryegrass were decreased by co-cropping with *N. caerulescens* compared with mono-cropping or co-cropping with *T. arvense* (non-hyperaccumulator). These observations suggest that the hyperaccumulator competes for Zn against the co-planted crop, but not always true for Cd, probably because the hyperaccumulator does not need Cd for the physiological functioning of the plant and may mobilize too much Cd which may be absorbed by the co-planted crop.

Co-cropping of two normal crops may also alter the availability of metals. Wheat/rice intercropping could reduce Cd uptake by wheat and rice in shoots and reduce Cd concentration in the wheat grain; however, Cd concentration in rice grain was increased, compare to that in the monocultured plant (Wu *et al.*, 2003). In the intercropping system of *Cunninghamia lanceolata* in tea gardens, the concentrations of Pb, Mn, Cu, and Zn in the tea leaves were significantly lower than those in the mono-cropping one, indicating that intercropping *C. lanceolata* in tea gardens can improve the quality of the tea (Xue and Fei, 2006).

Continued agricultural production of safe animal feeds or energy crops on contaminated soils is necessary considering the limited arable land in China and time required for phytoremediating metal-contaminated soils. Co-cropping or intercropping may alter conditions in shared rhizosphere and thereby affect the availability of selected metals to neighboring plants. Therefore, it is possible that planting some low-metal crops in association with hyperaccumulators or other appropriate plants may allow agricultural production on heavy metal-contaminated soils.

CO-CROPPING FOR ENHANCING PHYTOEXTRACTION

Co-cropping *N. caerulescens* with *T. arvense* increased Zn uptake by the hyperaccumulator (Whiting *et al.*, 2001). Combined cropping of *N. caerulescens* and *Hordeum vulgare* caused a higher concentration of heavy metals (Cd, Pb, and Zn) in the biomass of

the hyperaccumulator *N. caerulea* (Gove *et al.*, 2002). It may indicate a potential approach to increase the remediation efficiency of metal-contaminated soils in a natural way.

A co-planting system, comprised of a Zn and Cd hyperaccumulator (*S. alfredii*) and a low-accumulating crop (*Z. mays*), was established in a rice field that was historically irrigated with water affected by Pb and Zn mining activities (Wu *et al.*, 2007c). The co-planting system enhanced not only the removal of heavy metals from the contaminated soils but could also produce safe corn for animal feed, allowing farmers to continue their agricultural activities. The co-cropping of a Cd hyperaccumulator *Brassica napus* with *B. parachinensis* or *Zea mays* increased the Cd concentration and accumulation in the shoot of the hyperaccumulator plant (Selvam and Wong, 2009). Co-cropping system could combine with chemical enhancement, including chelator-enhanced phytoextraction (Wu *et al.*, 2007c) and chemical washing (Huang *et al.*, 2010), to further increase the phytoextraction efficiency.

For the non-hyperaccumulators, Li *et al.* (2009b) studied the effect of 7 intercrops on Cd uptake by maize for naturally enhancing the phytoremediation with high biomass plants and the results showed that legumes substantially increased Cd uptake by maize. However, maize is not a hyperaccumulator so that its phytoextraction efficiency is low, and the disposal of the produced biomass with metal contents generally too low to be recovered should be considered.

In a pot experiment conducted to study the interaction mechanisms between the roots of two co-cropped plants with the corn roots separated from *S. alfredii* with a mesh barrier, co-cropping decreased pH of the sludge solution, increased dissolved organic carbon (DOC), and resulted in higher Zn/Cd concentration than the *S. alfredii* mono-cropping, which caused more Zn/Cd transported to the *S. alfredii* side and then enhanced the uptake of the heavy metals by the hyperaccumulator (Hei *et al.*, 2007). The concentrations of Cu, Pb, Zn, Cd, and Fe in the shoots of peas grown in a mixed culture with barley in pots were 1.5, 1.8, 1.4, 1.4, and 1.3 times higher than those of the monoculture (Luo *et al.*, 2008). The root exudates from barley in the mixed culture system played an important role in the process of solubilizing metals in soil and facilitating the uptake of metals by peas.

The enhancing effect on the phytoextraction depends on plant species in co-cropping. For example, intercropping of *Salix caprea* with the hyperaccumulator *Arabidopsis halleri* did not enhance total Zn and Cd

extraction compared to single plantings (Wieshammer *et al.*, 2007), which is potentially due to competition for water or nutrients and possibly metal sequestration into plants. *Athyrium yokoscens*, a fern that hyperaccumulates Zn and Cd, is often accompanied by another Zn and Cd-hyperaccumulating plant, *Arabis flagellosa*; a mixed planting of two plants of each species did not largely change the levels of Zn and Cd accumulated by these two plants (Chen *et al.*, 2009), which may be related to competition for metals, and indicated that it would be better to intercrop plants that hyperaccumulate different metals.

CONCLUSIONS

This review presents a broad summary and evaluation of cropping systems that are available for trace element-contaminated sites. Three major cropping systems are identified, each of which targets on one of the following crop production: edible plants, non-edible plants, and bio-ores for industrial use (Fig. 2). The primary strategies include phytoexclusion, phytostabilization, and phytoextraction/phytomining. Each strategy has benefits and limitations, depending on the remedial goals targeted and on-site and species-specific factors.

For safe production of edible food crops such as cereals and vegetables, further research is needed to select low-accumulating species/cultivars by means of agronomic and genetic breeding. Chemical amendments (*e.g.*, alkaline agents, adsorption agents, and fertilizers) should be optimized according to their efficiency of metal immobilization and the economic feasibility. In this case, the designed cropping system needs to be evaluated mainly by production quality rather than soil remediation.

At contaminated sites with harsh soil conditions (*e.g.*, acidity, low fertility, and multiple metals), cropping industrial non-food energy and fiber plants will likely be preferred. However, more attention should be paid to amendment selection and soil management practices to achieve a best crop yield and least potential hazard. On the other hand, for large-scale sites with low to moderate levels of trace elements (*e.g.*, As, Cd, Ni, and Zn), a plant-based phytoextraction will be a possible option for decontamination of metals and/or extraction of metals with high price (*e.g.*, Ni). However, the technology can in some cases be time-consuming and it is anyway limited to the soil depth prospected by roots of hyperaccumulators. Thus, high-biomass hyperaccumulating species should be selected, and further research is required for better un-

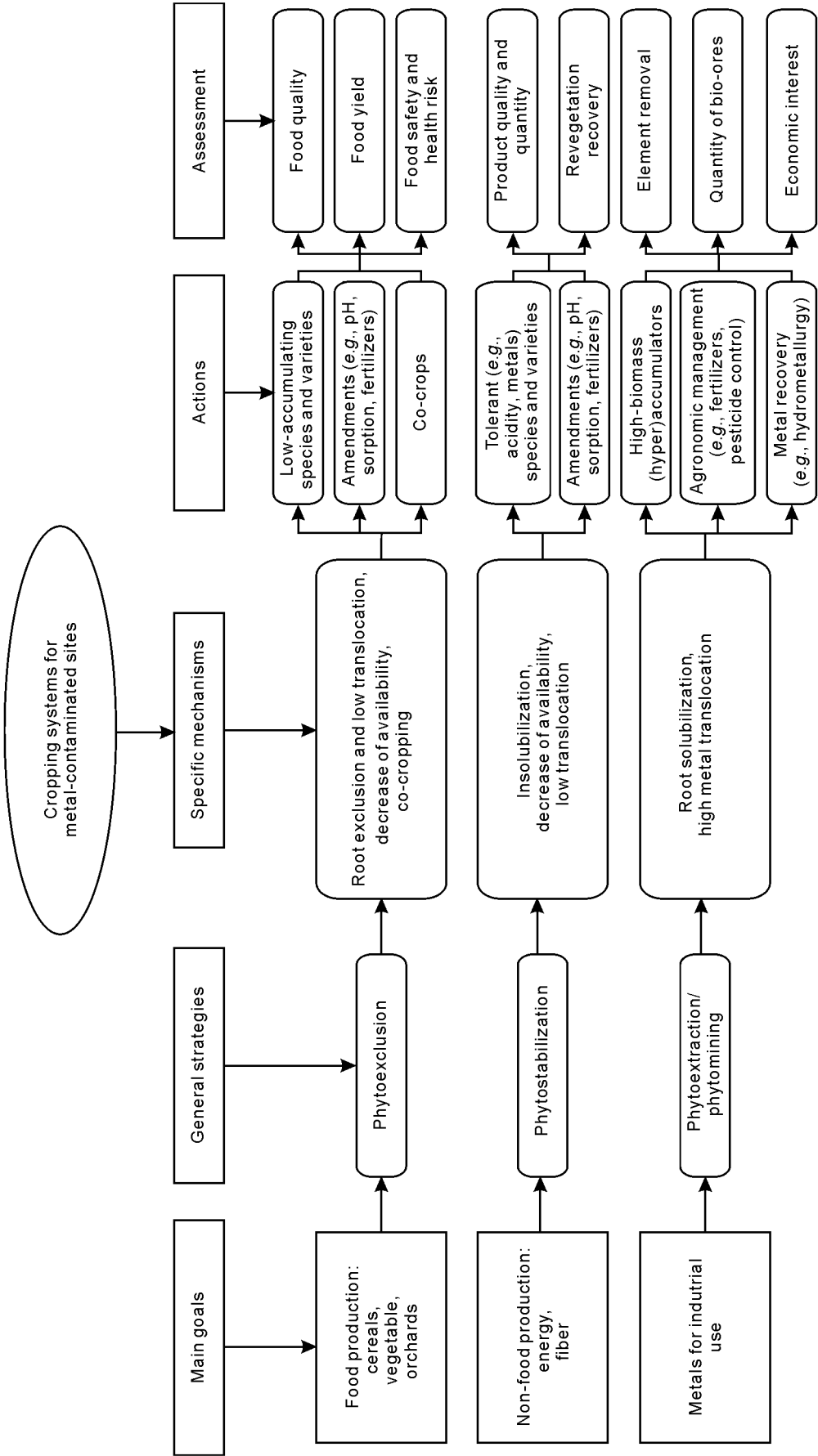


Fig. 2 Summary of designing cropping systems for metal-contaminated sites.

derstanding the mechanisms involved in metal hyperaccumulation. Further, biomass produced by phytomining is a substantial resource (*i.e.*, 2% Ni), for which a specific separation process should be developed.

Co-cropping two plant species with contrasting metal-accumulating abilities could improve phytoremediation efficiency and remediate metal-contaminated soils in a naturally enhanced way. The interaction mechanisms between the two plants deserve further investigation and the combination ratios of the two plants are warranted to be optimized.

In summary, the majority of these cropping systems are still far from maturity at present, but they have shown the potential for a safe and regular agriculture on large-scale sites contaminated with trace elements in a cost-effective manner. With appropriate combination of soil types, plant species/cultivars, and agronomic managements, toxic metals into the food chain will be restricted and the potential benefits offered by these cropping systems can be substantial in the long run.

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